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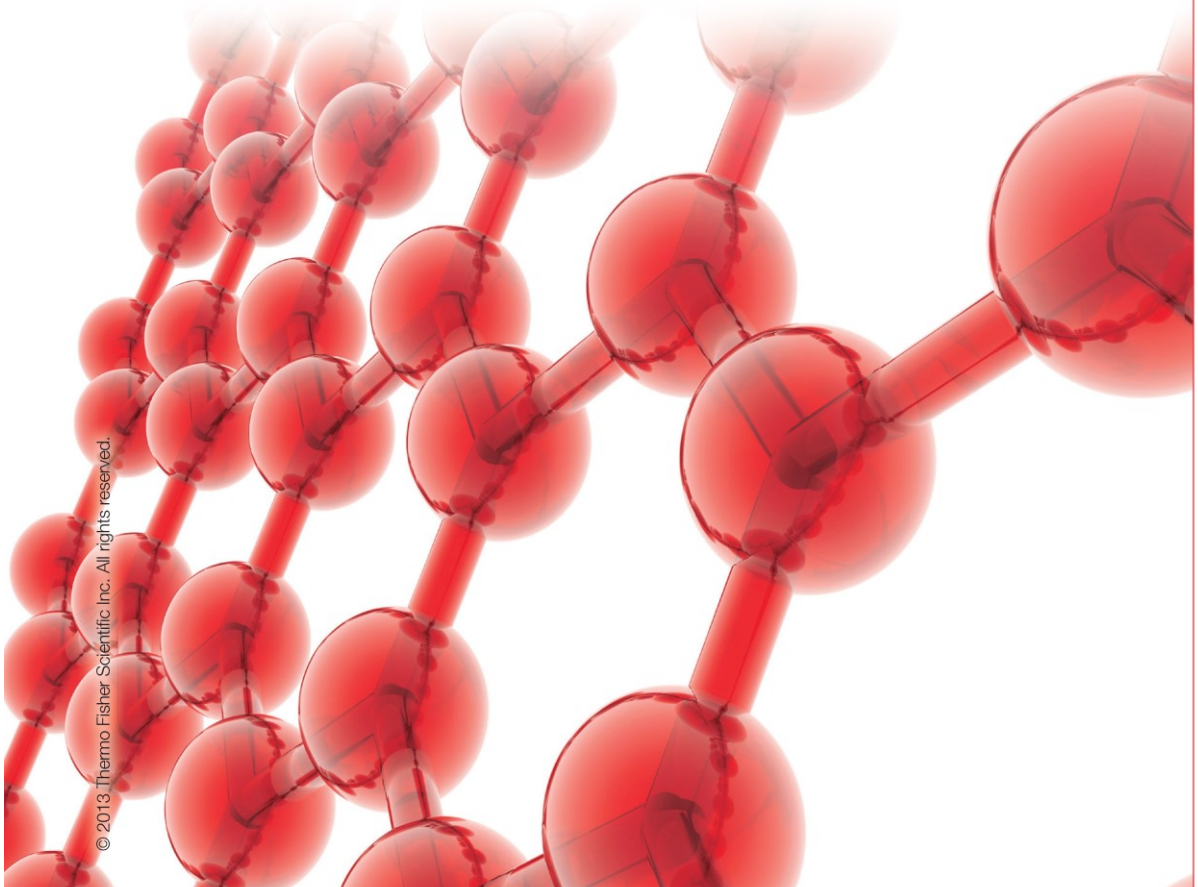

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FOREWORD

On behalf of the Organising and the International Scientific Committees we take great pleasure in welcoming you to Manchester for the 13th in-person edition of the Graphene and 2D Materials International Conference & Exhibition (Graphene2023).
Over the past 12 editions, the Graphene Conference strengthened its position as one of the main meeting points of the Graphene community Worldwide. Graphene2023 is now an established event, attracting global participant's intent on sharing, exchanging and exploring new avenues of graphene-related scientific and commercial developments.

Graphene2023 Highlights:

- Expected attendance: 650 participants in-person
- 97 Plenary, Keynote & Invited Speakers
- More than 190 posters
- Nearly 170 oral contributions
- More than 30 Exhibitors
- 13 Parallel Workshops covering the whole value chain of "Graphene and 2D Materials innovation"
- 2-days Industrial Forum in parallel to get an updated understanding of Graphene based technologies from worldwide industries.

We are also indebted to the following Scientific Institutions, Companies and Government Agencies for their help and/or financial support:
AIXTRON, Reliance Industries, Tata Steel, RIC-2D, HORIBA Scientific, ThermoFisher, UK Science & Innovation Network, KAUST, OXFORD Instruments, GDR-I Graphene & CO – HOWDI and Digital University Kerala, KSIDC & Invest Kerala.

We also would like to thank all the exhibitors, speakers and participants that join us in-person this year.

We truly hope that Graphene2023 serves as an international platform for communication between science and business.

Hope to see you again in the next edition of the Graphene and 2D Materials International Conference & Exhibition (Graphene2024).

Graphene2023 Organising Committee


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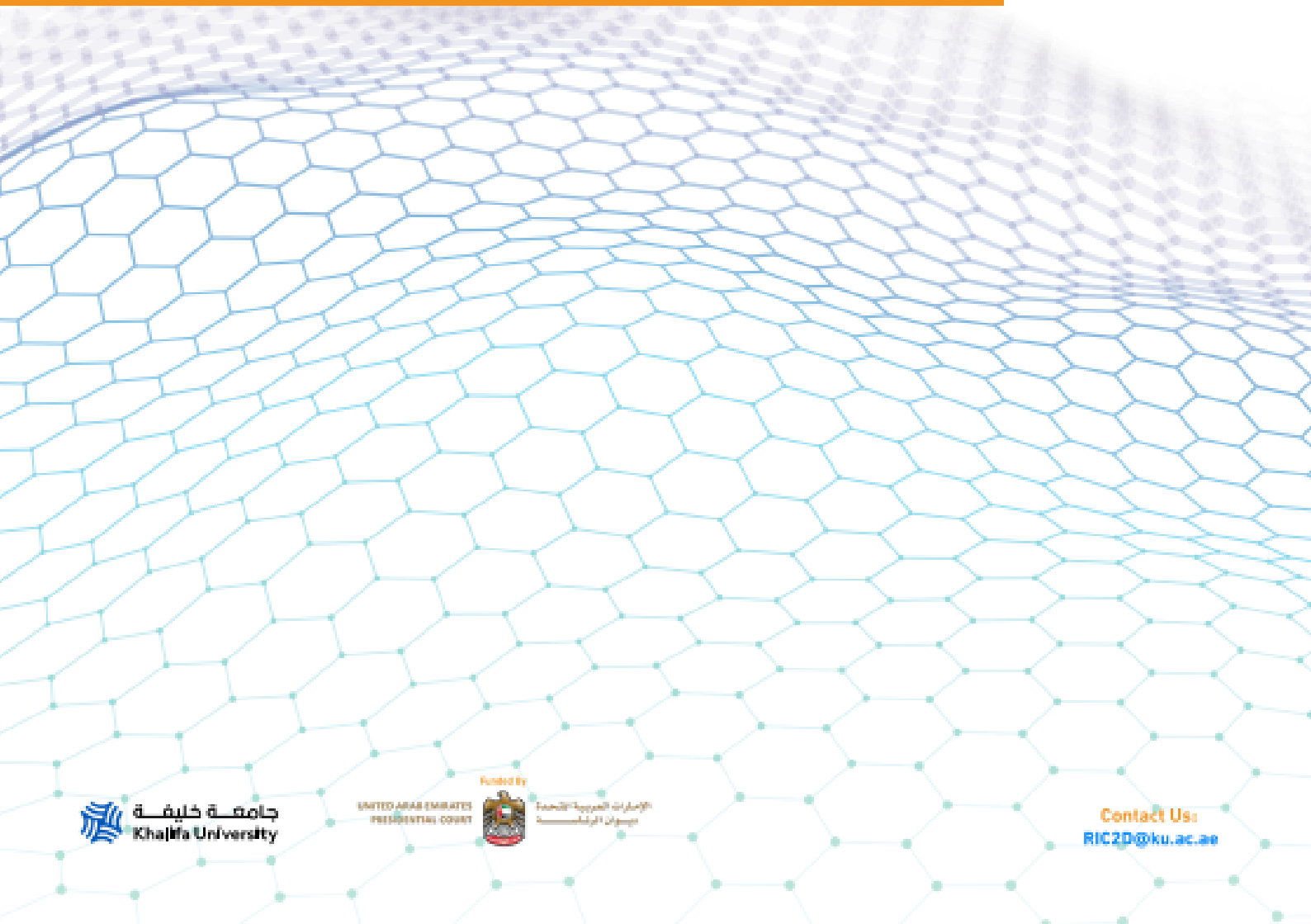

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The Research & Innovation Center for Graphene and 2D Materials (RIC-2D) hosted by Khalifa University of Science and Technology is part of a strategic investment by the Government of Abu Dhabi, United Arab Emirates to advance the scientific development and commercial deployment of technologies derived from graphene and other 2D materials. RIC-2D will serve as an integral part of an advanced materials innovation ecosystem being developed in Abu Dhabi.

Established in 2022, Research & Innovation Center for Graphene & 2D Materials will host a range of activities to support both research and technology advancement of graphene and other enhanced 2D materials in the region.

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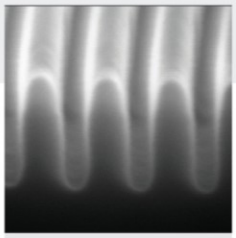

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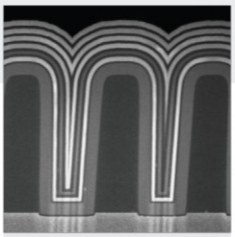

Growth of dielectrics and etching.

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Exploring 2D Empty Space

Andre Geim
University of Manchester, Manchester M13 9PL, United Kingdom

It is now possible to create angstrom-scale channels that can be viewed as if one or a few individual atomic planes are pulled out of a bulk crystal leaving behind a 2D space. I shall overview the work done in Manchester on this subject over the last several years, which covers studies of various properties of gases, liquids and ions under such an extreme confinement (for review, see [1]).

References

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Figures

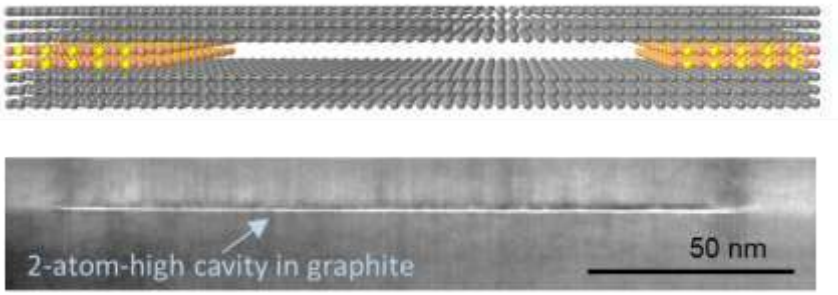

Figure 1: (Top) Artist's impression: Angstrom-scale cavities with atomically flat walls can be made from different materials and have different heights. (Bottom) Transmission electron micrograph of a 2D cavity in graphite with a nominal height of 6.7 Å. It is made by placing strips of bilayer graphene between two atomically-flat graphite crystals.

Quantum Limited Thermal and Thermoelectric Transport in Graphene

Philip Kim
Department of Physics, Harvard University, 11 Oxford Street, Cambridge MA 02138, USA
philipkim@g.harvard.edu

In low-dimensional systems, a growing number of many-body quantum phenomena have emerged from the combination of reduced dimensionality, strong interactions, and topology. Thermal and thermoelectric transport, which is sensitive to energy- and entropy-carrying degrees of freedom, provides a discriminating probe of emergent excitations in quantum materials. In this talk, I will discuss several recent developments in the measurement of thermal and thermoelectric transport in graphene-based nanostructures in the quantum limit. In the first part, we discuss electronic thermal conduction in bilayer graphene near charge neutrality as a function of external field strengths. We use nonlocal noise thermometry to probe the quantum Hall-ferromagnetic phase diagram. Here we show clear signatures of phase transitions between different broken symmetry states in strongly correlated states that appear in the quantum limit. In the second part of the talk, we will discuss thermoelectric transport in the lowest Landau level formed in disordered graphene quantum dots. Here, the combination of quantum confinement and disorder effects leads to a novel non-Fermi liquid behavior. We will discuss the implications of electrical and thermoelectric transport in this system in the context of strongly entangled quantum systems that is enabled by the strong interactions between localized states engineered in graphene quantum structures.

2D materials for the semiconductors industry

Mario Lanza
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Two-dimensional layered materials (2D-LMs) materials have outstanding physical, chemical and thermal properties that make them attractive for the fabrication of solid-state micro/nano-electronic devices and circuits. However, synthesizing high-quality 2D-LMs at the wafer scale is difficult, and integrating them in semiconductor production lines brings associated multiple challenges. Nevertheless, in the past few years substantial progress has been achieved and leading companies like TSMC, Samsung and Imec have started to work more intensively on the fabrication of devices using 2D-LMs. In this invited talk, I will present our work towards hybrid 2D/CMOS microchips, with special emphasis on those dedicated to realize memristive operations. I will show state-of-the-art performance as electronic memory, as well as other novel properties that traditional microchips don't exhibit, and that enable different innovative applications[1-10].

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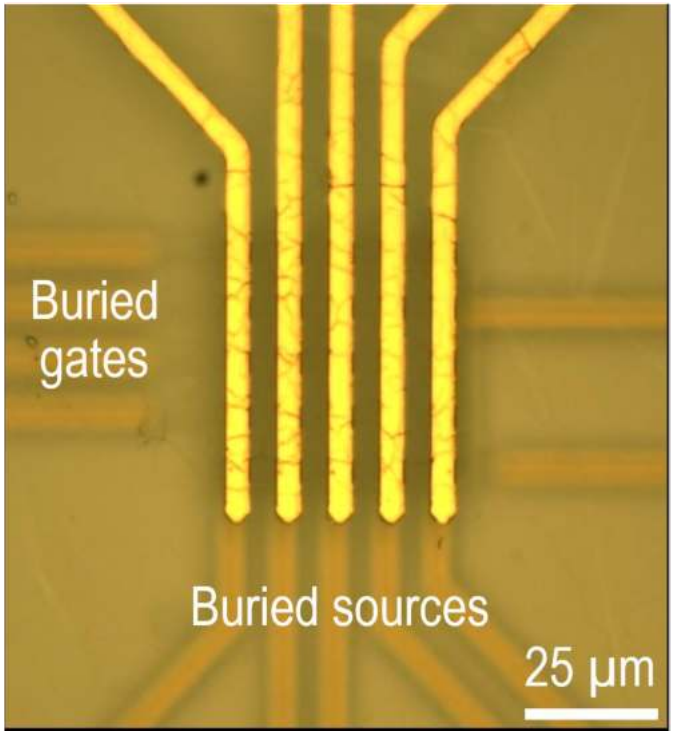

Figure 1: Optical microscope image of a hybrid 2D/CMOS microchip containing a 5×5 crossbar array of one-transistor-one-memristor cells, with h-BN memristor and 180-nm-node CMOS transistor.

van der Waals layered magnetic semiconductors

Young Hee Lee
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Ferromagnetism in van der Waals two-dimensional (2D) materials has been reported recently. Intrinsic CrI3 and CrGeTe3 semiconductors reveal ferromagnetism but the Tc is still low below 60K. In contrast, monolayer VSe2 is ferromagnetic metal with Tc above room temperature but incapable of controlling its switching via gating due to metallic nature. Moreover, the long-range ferromagnetic order in diluted metal chalcogenide semiconductors has not been demonstrated at room temperature. The key research target is to realize the long-range order ferromagnetism, Tc over room temperature, and semiconductor with gate tunability. In this talk, we introduce magnetic dopant, vanadium in semiconducting WSe2 and manifest Tc at room temperature and gate tunability at low doping concentration. We further explore different doping concentrations including highly degenerate regime and demonstrate unconventional magnetic order by random telegraph spin noises via interlayer coupling and strange metal.

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CVD Graphene: Growth Frontiers & Mass Production

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With 18 years history of graphene materials, there still exist lots of technical challenges towards graphene industry, including: 1) Low-cost mass production technology; 2) Batch peeling-transfer technology; 3) Transition from single-layer graphene component to macroscopic materials with retained intrinsic performances; 4) Dispersing technology into matrix materials; 5) Irreplaceable killer applications. Beijing Graphene Institute (BGI), established in 2018 and currently having nearly 300 employees, aims to solve these challenging issues by paying particular emphasis to mass production of high-quality CVD graphene materials and manufacturing equipments. BGI is devoted to providing the best graphene materials and related equipments to the market as well as the graphene-related R&D services to enterprises.

The current R&D emphasis on CVD graphene materials at BGI is laid on three different directions: 1) high-quality graphene films and wafers for general purpose; 2) graphene-skinned materials (GSM); 3) purpose-oriented graphene materials focusing on thermal managements, optical communications and medical applications. The star products into market are A3-size graphene films with mm grain boundaries, 4- and 6- inch single crystal graphene wafers, superclean graphene films with best thermal and electrical performances, graphene-skinned glass & fibers, graphene-skinned aluminum oxide fibers and etc. The graphene-skinned glass fibers have been used for deicing applications with extremely high electrothermal conversion efficiency up to 94%. At BGI, we are also working for the customized graphene growth depending on the special needs and requests from customers. There is actually a great space in this area, which is particularly important before graphene films and wafers find their practical application market in next ten years.

Graphene-Based Composites: From Nano to Macro Applications

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Abstract
Given its superior mechanical behaviour, combined with exceptional electrical and thermal properties, graphene is the ideal candidate for lightweight, high strength composite materials with several multi-functionalities. Over the last decade, our group has made significant progress in the design and the development of graphene-based composites, thus bringing the full nanoscopic functionality of 2D materials from nano- and microscale into the macroscopic world. These composites typically combine graphene and related materials with polymers to create components or even structures with enhanced properties, which are interesting for a wide range of applications. The talk aims to present our recent progress, opportunities and challenges in this field, including discussion on innovative production and assembly techniques, mechanical performance and other functional properties, real-world applications with examples spanning from strain sensing to EMI shielding.
An important area of our research, for instance, is the development of *ad-hoc* tailored composite architectures, in which the fine control of filler distribution within the matrix allows for full exploitation of graphene properties. In fact, we have demonstrated that the sequential alternation of continuous CVD graphene and ultra-thin polymer layers in the nanolaminate architecture results in the remarkable combination of mechanical reinforcement and multifunctionalities, such as impressive EMI shielding behaviour, thermal conductivity and barrier properties.
The development of lightweight graphene-enhanced fibre composites for use in the aerospace and automotive industry will be also discussed, as key area of focus towards the development of smart and functional components with reduced fuel consumptions and gas emissions. The incorporation of graphene nanosheets in fiber-reinforced polymers can enhance the mechanical properties - such as strength, stiffness, fracture toughness and damping – and offer other functionalities, such as fire retardancy.
We have also developed functional coatings with improved durability and corrosion resistance, and graphene-based sensors that can detect body motion and a wide range of chemicals and gases with high sensitivity and selectivity, making them useful in environmental monitoring and medical diagnostics.
Overall, our research reveals that the incorporation of graphene into various forms of composite materials can add multi-functionality and, in some cases, significantly improve their performance, making them more efficient, durable, at a cost which is not prohibiting for a number of applications. As research in this area continues to advance, it is likely that we will see the development of a wider range of applications in various industries.

Electrothermal Applications of Graphene for Consumer Electronics

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Recently, we developed transparent graphene heating modules to be applied to a new type of cookware, the "Graphene Kitchen Styler." The name came from the idea of replacing the old cooking styles based on 100-year-old coil-heater technology with novel transparent planar graphene heaters that generate IR radiation more efficiently from atom-thick, layers more robust than diamond. The same idea has been applied to a graphene-based heat radiator, simply called "G-radiator", which is a virtual fireplace that generates heat from graphene, more efficiently with less space and less energy.

The graphene heater is also important for automotive applications. The graphene heater film can be inserted between two layers of front windshield glass and uses minimum electrical energy to heat the windshields, which is more efficient than heating the whole interior of a vehicle by air conditioning for EVs. The same technology can be applied to the defrosting of RiDAR covers and digital side view mirrors as well as various industrial heating modules for semiconductor fabrication processes. These electrothermal applications of the graphene heater are branded "Thermo-Graphene."

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Figures

Figure 1: A photograph of "Graphene Radiator".

Chemical and Electrochemical Modulation of Physical Properties of MXenes

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MXenes (carbides, nitrides, oxycarbides and carbonitrides of early transition metals) are a very large family of 2D materials. They have a chemical formula of $M_{n+1}X_nT_x$, where M represents a transition metal (Ti, Mo, Nb, V, Cr, etc.), X is either carbon and/or nitrogen (n=1, 2, 3 or 4), and T_x represents surface terminations. The large variety of structures and compositions, availability of solid solutions on M and X sites, and control of surface terminations, such as O, OH, halogens, chalcogens, etc., offer a plethora of chemistries to investigate.[1] Combining their plasmonic properties with ease in aqueous processing, high electronic conductivity (over 20,000 S/cm) and excellent mechanical properties, which exceed other solution-processable 2D materials, MXenes have the characteristics necessary to develop as optical and electronic materials.[2] Inherent to their 2D structure, the charge carriers responsible for MXene's optical responses and electronic transport are very close to an external interface that has exceptional ability to undergo reversible chemical and electrochemical reactions to add or change surface terminations. By design of the MXene composition, the carrier plasma can be rendered sensitive to the resulting changes in band structure and state-filling. As a result, properties of MXenes, such as conductivity, work function, plasmon resonance; visible, IR and microwave absorption/reflection can be controlled by introducing reversible or irreversible changes in their surface chemistry. Taking into account that electrochemical charge/discharge with milliseconds frequency is possible for MXenes, electrochemical modulation of optical and electronic properties can facilitate new ways of influencing material interactions with electromagnetic waves over visible, IR, THz and even GHz wavelength ranges. Many technological advances can be enabled by these chemically responsive, conductive materials in the fields of electromagnetic interference shielding, antennas, sensors, and other devices. In this talk, I explain how optical, electronic and transport properties of MXenes can be manipulated by tuning their chemical composition. This presentation will also demonstrate electrochemical modulation of the optoelectronic properties and describe potential applications of MXenes in photonic, optoelectronic and electrochemical devices.[1,3,4]

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From functionalizing inorganic two-dimensional materials on the level of single atoms towards molecular imaging of organic two-dimensional materials

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To functionalize and image two-dimensional inorganic materials using the electron beam in a transmission electron microscope, a prerequisite is the detailed understanding of the beam electron - sample interaction is required. We derive this basic knowledge from atomically-resolved, time-dependent in-situ TEM imaging of inorganic two-dimensional (2D) transition metal dichalcogenides (TMDs) using the chromatic- and spherical-aberration-corrected low-voltage SALVE instrument operating in the voltage range between 80kV and 20kV.

We show for different single-layer TMDs that in dependence of the electron accelerating voltage and applied electron dose the defect formation can be initiated. Applying this knowledge, in-situ and ex-situ structural and chemical transformations of different freestanding TMDs and of rarely reported TMPTs (TM phosphorus tri-chalcogenides) are performed and varified by complementary ab-initio calculations.

For lateral heterostructures we show near-atomically sharp junctions with a typical extent of 3 nm for the covalently bonded $MoSe_2$-WSe_2 interface and use this knowledge to explain the considerably narrowed optical transition linewidth in the PL and Raman spectroscopy. Further we show proof-of-principle experiments in which we transfer electron-exposed TMD flakes from a TEM grid to arbitrary substrates to directly relate the results of the photoluminescence and transport measurements to their structural origin.

For vertical few-layer heterostructures we study the effect of interlayer excitons in WSe2 located in the low-loss range of the EELS spectrum as function of the twist-angle and material. In few-layer graphene heterostructure we discuss the Li crystal nucleation mechanism from in-situ studies of a miniaturized electrochemical cell, where reversibly single-crystalline bilayer graphene is lithiated and delithiated in controlled manner using an electrochemical gate confined to a device protrusion.

The knowledge gained for the study of 2D inorganic materials we apply to the study of 2D polymers and 2D metal-organic frameworks (MOFs), however, not surprisingly, functionalization and even atomically-resolved imaging is hindered due to much lower resilience of the organic material during electron irradiation. We present key strategies to achieve nevertheless higher resolution in high-resolution TEM images of imine-based 2D polymer films, which include the selection of a surprisingly low electron accelerating voltage of 120kV for achieving a resolution of 1.9A, enabling imaging the linker molecules. Moreover this resolution allowed even imaging the molecular nature of interstitial defects, which was interpreted by means of quantum mechanical calculations. Further, we study experimentally and computationally the role of different organometallic bonds and hydrogen content on electron radiation stability, using a group of four structurally similar Cu-based 2D MOFs.

We summarize our results in one sentence that 2D materials and lower-voltage atomic (molecular) resolution TEM/STEM are just made for each other.

Is the Future 2D?

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Semiconductor sales reached over $570 billion worldwide in 2022, a gigantic industry that keeps on growing with increasing demand for faster, more powerful, and smaller chips. However, as we keep scaling CMOS transistors, the silicon (Si) transistor will soon reach its physical limit, and there is a pressing need to find an alternative post-Si material to enable the continuation of Moore's Law. Furthermore, as we scale our interconnects and further constrain our metals, resistivity soars, there is a critical need to alleviate this resistance hit. As we search for this set of next generation materials, 2D materials such as Transition Metal Dichalcogenides (TMDs), at angstrom thicknesses, have been shown in academia to possess remarkable properties. Could 2D materials play a role in future electronic devices?

In this talk, I will present some of Intel's published research on 2D materials focusing on TMDs, from synthesis and characterization to innovative applications. How each year, we take a step further to attaining our vision of stacked 2D nanoribbons, while also continuously finding novel applications for 2D materials. I will demonstrate, that in Components Research at Intel, we are always looking for ways to improve future technologies and enable the continuation of Moore's Law.

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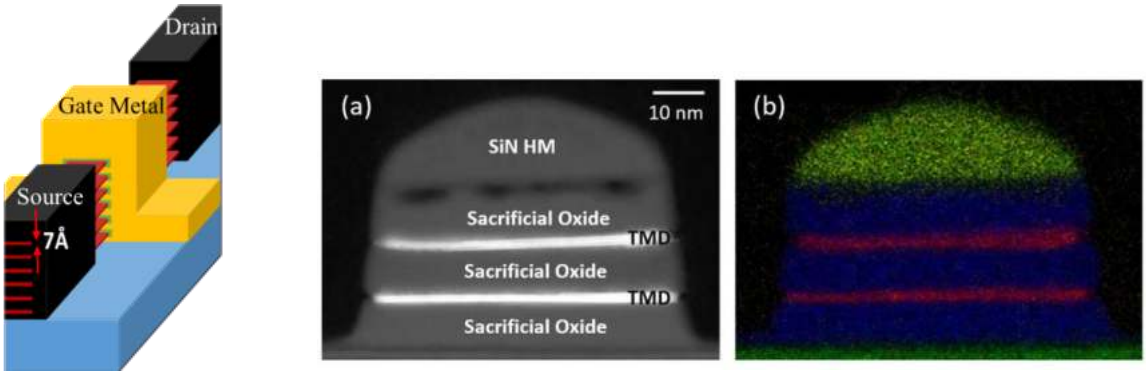

Figure 1: Schematic of Ultimate CMOS scaling with stacked 2D nano-sheets, followed by (a) TEM cross section of a two-layer TMD stacked nanoribbon structure showing 2 – 3 ML per nanoribbon, (b) TEM elemental mapping of stacked 2D nanoribbons.

Low-Thermal-Budget BEOL-Compatible Beyond-Silicon Transistor Technologies for Future Monolithic-3D Compute and Memory Applications

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If Silicon material system for massive monolithic-3D (M3D) integrated circuits is difficult, are there solutions that are beyond Si? In this talk, we discuss two low-thermal-budget approaches: Oxide Semiconductor and 2D Materials for M3D integration. By reviewing some of our recent work with IGZO-based transistors and memories, followed by our investigation of the 2D material opportunities for 3D memories, we highlight the need for new low-thermal-budget additive techniques for heterogenous multi-material integration as well as low-temperature material modification. Given the unlikelihood of "perfect materials", new system architecture-material-device co-design intervention will be essential to capitalize on the specific trade-offs of the components.

The importance of morphology in printed nanosheet networks for device applications.

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Abstract

Printed networks of graphene and other 2D materials will be important in a range of applications from electrodes to sensors to energy storage devices. For many of these applications, maximizing (or minimizing) the conductivity of such networks is important. However, the mesoscale physics of electrical transport in these networks is still poorly understood. In particular, the role of inter-sheet junctions has not been quantified. For example, achieving the ultimate performance of such printed devices requires the junction resistance to be suppressed below the resistance of the individual nanosheets themselves. However, values of these quantities remain unknown. It is however expected that the morphology of printed networks will very strongly impact the resistance of inter sheet junctions. However. in general, the morphology of printed networks is also poorly understood, because of the lack of appropriate tools to interrogate it. Here, I will demonstrate a nano-tomography technique, based on a combination of SEM &FIB, that can be used to obtain 3D images of nanosheet networks with 5 nm resolution. From such images, one can obtain a range of different parameters including pore network porosity and tortuosity, nanosheet alignment and stacking arrangement as well as nanosheet network connectivity and tortuosity. We will also use new techniques to estimate the junction resistance in nanosheet networks and link them to morphology. We have extended the 3D imaging technique to interrogate printed devices, separating different nano-layers from electrodes and measuring parameters such as interfacial roughness and identifying pin holes. Having demonstrated the morphology of nanosheet networks I will describe a number of electronic devices based on these networks. Such devices include piezo resistive sensors which require poorly connected low conductivity networks as well as printed transistors which require highly connected networks to achieve high mobility. Finally, I will show preliminary results on printed heterostacks acting as printed diodes with rectification ratios up to 10,000 that can act as sensitive photodetectors.

Figures

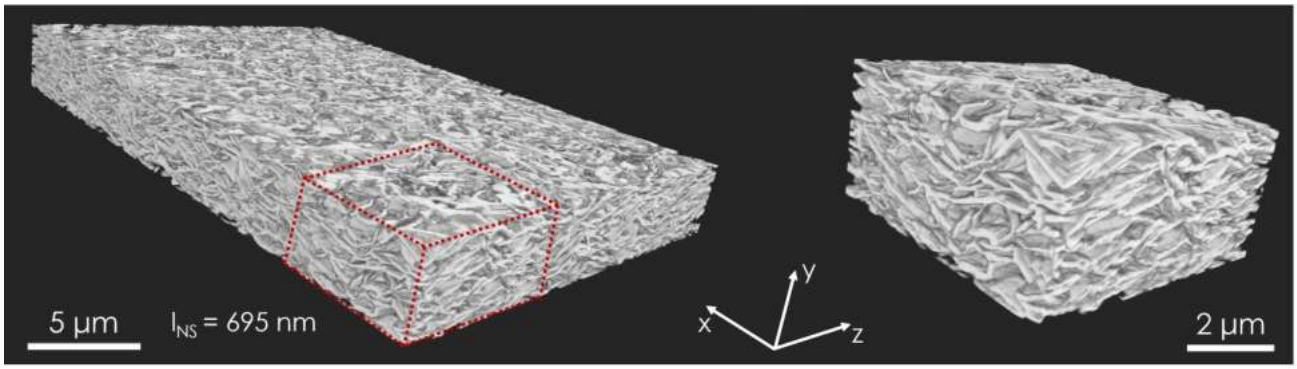

Figure 1: 3D image of a nanosheet network spray cast from LPE graphene nanosheets.

Studying Surfaces and Interfaces in Transition Metal Dichalcogenides with Advanced Transmission Electron Microscopy

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Surfaces and interfaces are key to the performance of 2D materials heterostructures including for controlling optoelectronic phenomena [1], enhancing electron interactions in moire superlattices [2], or for studing transport with nanofluidic devices [3]. Progress crucially depends on knowledge of the local atomic structure, which in many cases can only be analysed by transmission electron microscopy (TEM) techniques. In this talk I will present our scanning TEM (STEM) investigations of unusual lattice reconstruction that occurs at the interfaces in twisted transition metal dicholcogenide bilayers [4]. Complementary scanning tunnelling measurements show that such reconstruction creates strong piezoelectric textures, which can be engineered by the application of applied field in the electron microscope [5]. This talk will also illustrate how we can use 2D heterostructures to produce a new design of in-situ liquid cell for the TEM [6,7]. This approach overcomes limitations of conventional silicon nitride window membranes allows atomic resolution imaging of adatom dynamics at solid-liquid interfaces [7]. We further show how a combination of STEM imaging and spectroscopy methods can be used to probe intercalation at the nanoscale and how these structures evolve as a function of time and annealing temperature (Figure 1) [8].

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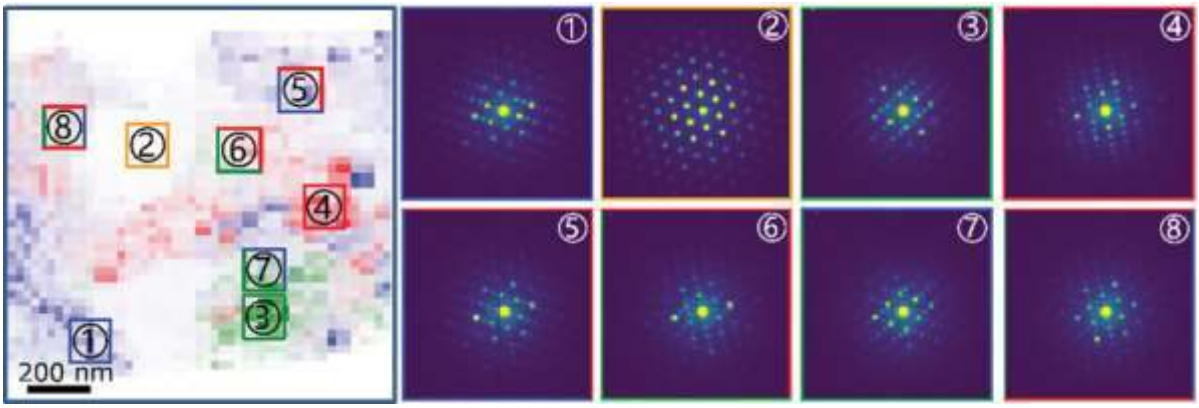

Figure 1: 4D STEM (scanning electron diffraction analysis of intercalation ordering in KMoS2 showing local disorder at the nanoscale (reproduced from [8].

Gate-Tunable Neuromorphic Electronics Enabled by 2D Materials

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The exponentially improving performance of digital computers has recently slowed due to the speed and power consumption issues resulting from the von Neumann bottleneck. In contrast, neuromorphic computing aims to circumvent these limitations by spatially co-locating logic and memory in a manner analogous to biological neuronal networks [1]. Beyond reducing power consumption, neuromorphic devices provide efficient architectures for image recognition, machine learning, and artificial intelligence [2]. This talk will explore how 2D nanoelectronic materials enable gate-tunable neuromorphic devices [3]. For example, by utilizing self-aligned, atomically thin heterojunctions, dual-gated Gaussian transistors have been realized, which show tunable anti-ambipolarity for artificial neurons, competitive learning, spiking circuits, and mixed-kernel support vector machines [4]. In addition, field-driven defect motion in polycrystalline monolayer MoS_2 enables gate-tunable memristive phenomena that serve as the basis of hybrid memristor/transistor devices (i.e., 'memtransistors' [5]) that concurrently provide logic and data storage functions [6]. The planar geometry of memtransistors further allows multiple contacts and dual gating that mimic the behavior of biological systems such as heterosynaptic responses [7]. Moreover, control over polycrystalline grain structure enhances the tunability of potentiation and depression, which enables unsupervised continuous learning in spiking neural networks [8]. Overall, this work introduces foundational circuit elements for neuromorphic computing by utilizing the unique quantum characteristics of 2D nanoelectronic materials [9].

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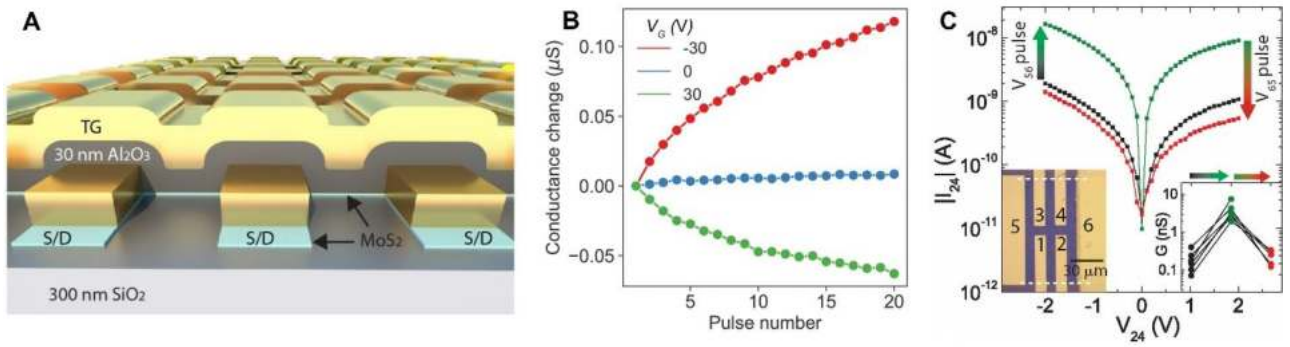

Figure 1: (A) Schematic of a dual-gated 2D memtransistor crossbar array. Adapted from Ref. 7. (B) The unipolar synaptic response of 2D memtransistors can be tuned from potentiation to depression as a function of the gate bias. Adapted from Ref. 8. (C) Multi-terminal 2D memtransistors show heterosynaptic responses between orthogonal contacts. Adapted from Ref. 5.

Field-effect transistors on 2D magnetic semiconductors

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Since the demonstration that magnetism persists in individual monolayers of van der Waals magnetic materials, transport measurements have proven to be a powerful technique to map the magnetic phase diagram as a function of magnetic field, temperature and thickness. In most cases, "vertical" transport measurements (i.e., measurements of current perpendicular to the layers) have been performed using the magnetic materials as tunnel barriers. Very limited work has focused on in-plane transport measurements using field-effect transistors, even though these devices are of extreme interest, as they allow the transport – and possibly the magnetic –properties to be tuned electrostatically with a gate. The reason for this is that the vast majority of compounds investigated in the first generation of 2D magnetic materials are semiconductors with extremely narrow bands (~100 meV), in which carriers localization effects dominate and prevent the measurement of in plane transport over any sizable distance.
In this talk, I will discus field-effect transistor measurements on different compounds having a bandwidth of 1 eV or larger, in which transport can be probed all the way down to cryogenic temperatures, deep in the magnetic phases of the 2D magnetic semiconducting materials used. I will first discuss the case of CrSBr, in which measurements reveal an astounding anisotropy of the transport properties, suggesting that the material behave as a collection of incoherently coupled 1D chains. I will then move to discuss experiments on CrPS4, in which a very large and gate-tunable magnetoconductance is observed, and in which the magnetic state itself can be controlled by the application of a gate voltage. IN this case, the analysis of the transistor characteristics as a function of temperature and magnetic field allow us to reveal the microscopic mechanism responsible for the large magnetoconductance observed.

2D $MoSi_2N_4$ family and HOPG-like graphene films

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Identifying new 2D materials and developing new synthesis methods are essentially important for the applications of 2D materials. I will first introduce a newly emerging artificial 2D layered material discovered by my group, $MoSi_2N_4$ [1], which has no natural counterparts and is grown by chemical vapor deposition [1-3]. It can be viewed as a monolayer MoN (MoN_2) sandwiched between two Si-N bilayers and exhibits semiconducting behavior (bandgap, ~1.94 eV) with a potentially high carrier mobility up to 1200 cm^2/Vs, high strength (~66 GPa), and good thermal conductivity (~200 W/mK) [1,3]. Motivated by the discovery of $MoSi_2N_4$, a large family of such structured materials with a general formula MA_2Z_4 have been predicted [4], including semiconductors, metals, magnetic half-metals, superconductors, and topological insulators. Then, I will demonstrate the synthesis and industralization of HOPG-like highly conductive graphene films for heat dissipation, which include the efficient and green synthesis of graphene oxide by water electrocatalysis [5], scalable fabrication of highly ordered and compacted graphene oxide laminates by centrifugal casting [6.7], and in particular, our recently developed defects promoted highly efficient graphitization strategy [8].

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Figures

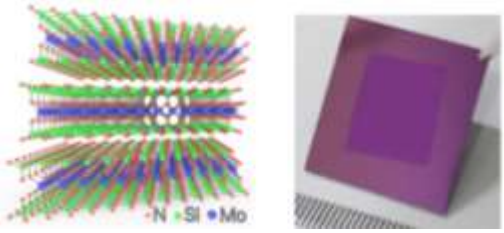

Figure 1: The structure of $MoSi_2N_4$ and a monolayer $MoSi_2N_4$ film grown by chemical vapor deposition.

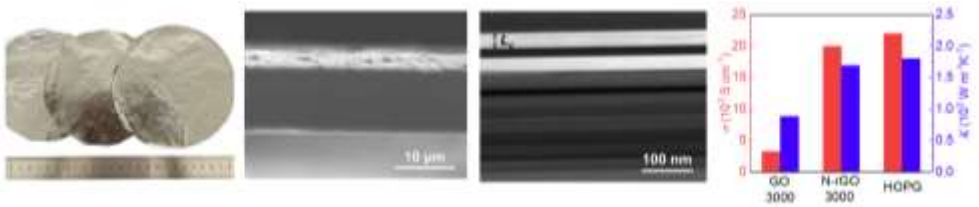

Figure 2: HOPG-like graphene films synthesized by defects promoted graphitization strategy.

Functionalization of MXene for energy storage and catalysis

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MXene are currently broadly studied for application in energy storage and conversion as well as in catalysis. Huge advantage for these applications originate from its high electrical conductivity, variability in surface chemistry and good chemical stability. The surface chemistry can be tuned by various methods using alternative chemical methods of MAX phase exfoliation or by chemical treatment of exfoliated MXene. The MXene exfoliated by hydrogen fluoride methods (like based on HF or LiF systems) led to mixed surface termination consisting from fluorine and oxygen functionalites. The oxygen functionalites like hydroxyl groups can be functionalized using triethoxysilane derivatives giving Ti-O-Si bond and can be effectively used for effective modifications of surface[1]. Schematic drawing of functionalization process is on Figure 1. Covalent bonding of Zwitterionic compounds on MXene surface can significantly improve performance of MXene in supercapacitor applications. Other strategy based on topochemical conversion by reaction of MXene with chalcogen and other elements like phosphorus or nitrogen can convert surface to chalcogen termination or even formation of composite consisting from dichalcogenide and modified MXene. These hybrid materials can be applied in supercapacitors, photodetectors as well as catalysts for electrochemical nitrogen reduction or water splitting. MXene surface chemistry offer a lot of variabilities for controlled functionalization for various applications not only for energy storage but also for optoelectronic and sensing applications.

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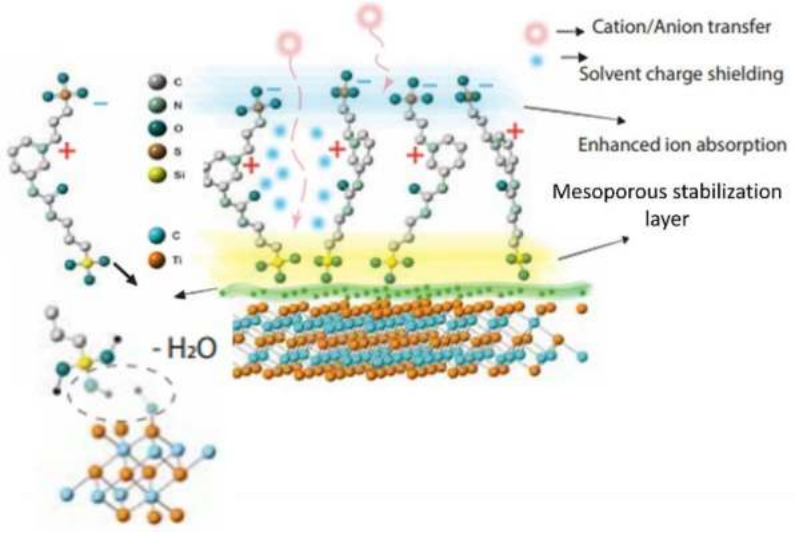

Figure 1: Schematic functionalization of MXene with Zwitterionic molecules.

Heavy quasiparticles and cascades without symmetry breaking in twisted bilayer graphene

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Twisted bilayer graphene (TBG) exhibits a plethora of electronic phases. Among the variety of correlated states, the cascades in the spectroscopic properties and in the compressibility happen in a much larger energy [1,2,3], twist angle and temperature range than other effects, pointing to a hierarchy of phenomena. Using Dynamical Mean Field Calculations (DMFT), in this work [4], we show that the spectral weight reorganization associated to the formation of local moments and heavy quasiparticles, and not a symmetry breaking process, is responsible for the cascade phenomena. Due to the fragile topology of TBG, a strong momentum differentiation is found in the incoherent spectral weight. The phenomena reproduced here include the cascade flow of spectral weight, the oscillations of the remote band energies and the asymmetric jumps of the inverse compressibility. We also address other posible measurements which may help distinguishing the phenomenology of the cascades discussed here from proposals involving symmetry breaking.

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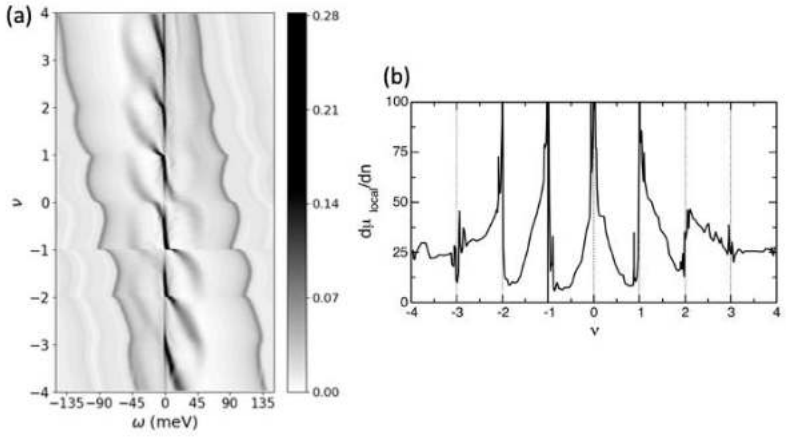

Figure 1: (a) Density of states and (b) inverse compressibility as a function of doping ν obtained in our DMFT calculations for a 1.08° twisted bilayer graphene [4].

Ladder Ferroelectricity by vdW Sliding

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We demonstrate and study a polar layered system with distinct ladder-like electric polarization steps that accumulate with each extra atomic layer. Moreover, the symmetries of these diatomic crystals translate planar shifts by one interatomic spacing to the out-of-plane switching of the structure and its polarization. I will discuss the origin of this ultimately thin interfacial polarization, the unique cumulative response at the atomic limit, the robust co-existence with in-plane conductivity, and the switching dynamics observed in our KPFM experiments and modelled by our first principle calculations.

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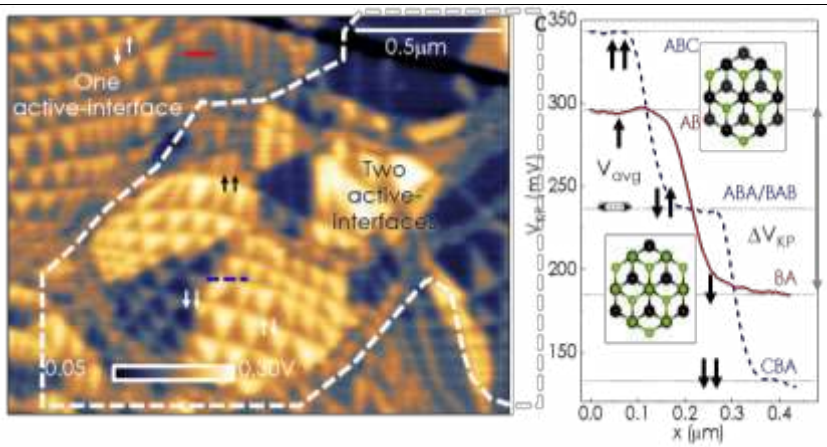

Figure 1: Electric surface potential of interfacial polarization in parallel TMD trilayers

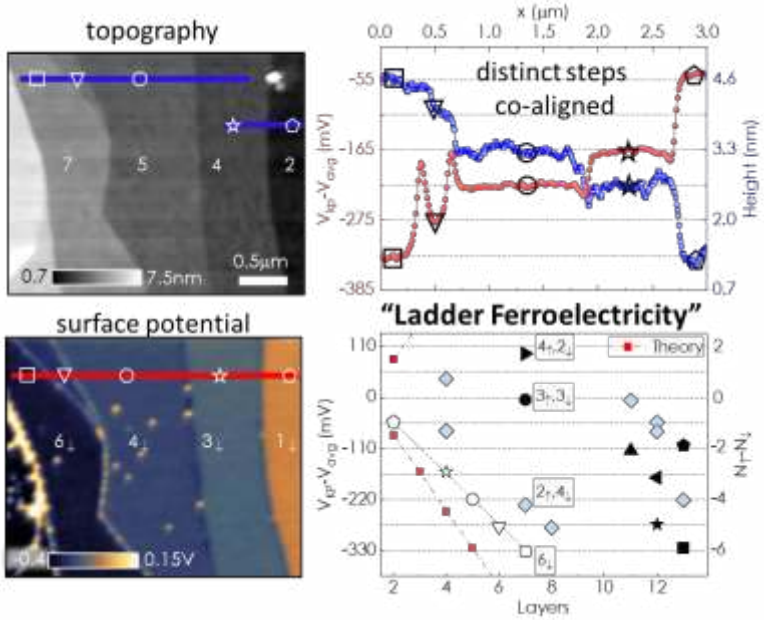

Figure 2: Distinct surface potential steps in multilayer polytypes

The Quantum Twisting Microscope

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In this talk, I will present a new type of scanning probe microscope, the Quantum Twisting Microscope (QTM), capable of performing local quantum interference measurements at a twistable interface between two quantum materials. Its working principle is based on a unique tip made of an atomically-thin two-dimensional material. This tip allows electrons to coherently tunnel into a sample at many locations at once, with quantum interference between these tunnelling events, making it a scanning electronic interferometer. With an extra twist degree of freedom, our microscope becomes a momentum-resolving local probe, providing powerful new ways to study the energy dispersions of interacting electrons. I will present various experiments performed with this microscope, demonstrating quantum interference at room temperature, probing the conductance of in-situ twisting interfaces, and imaging local energy dispersions of graphene and twisted bilayer graphene[1].

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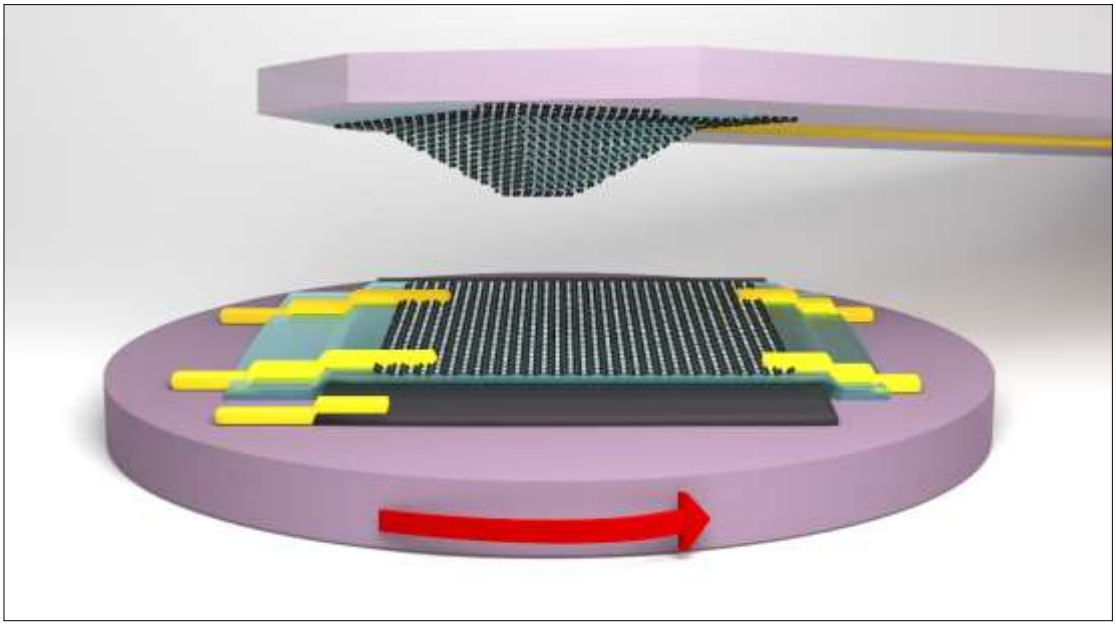

Figure 1: The Quantum Twisting Microscope.

High dimensional immune profiling of 2D materials: applications for human health

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Abstract

We depicted the "Nano-immunity-by-design" where the characterization of 2D materials is not solely based on their physical-chemical parameters but also on their immuneprofiling. [1] The immune-profiling can be revealed on its complexity by unique, informative ways: high dimensional approaches. [2,3] We exploited high-dimensional approaches, such as single-cell mass cytometry and imaging mass cytometry on graphene and other novel two dimensional materials, such as transition metal carbides/carbonitrides (MXenes). [4-6] We revealed that the amino-functionalization of graphene oxide increased its immunocompatibility. [4] Moreover, we combined graphene with AgInS2 nanocrystals, enabling its detection by single-cell mass cytometry on a large variety of primary immune cells. [5] Recently, we reported the immune modulation of specific MXenes, and their label-free detection by single-cell mass cytometry and other high dimensional approaches. [6-7] Together with our published works, I will present unpublished results on a wider variety of novel 2D materials, MXenes, MoS_2, WS_2, and bismuthene. Our results conceptualize that chemical and immnuological designs of 2D materials offer new strategies for their safe exploitation in biomedicine.

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Time for field test of Graphene technology: The Graphene-Perovskite Solar farm and beyond

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In the present talk I will discuss the design, fabrication, implementation and test of the Graphene-Perovskite Solar Farm. [1]
The Solar farm includes 9 halide perovskite solar panels realized exploiting the Graphene and Related 2D material (GRM) interface engineering.
The Solar farm was tested in Crete and permitted to evaluate the photovoltaic properties of this new generation graphene-perovskite solar cells.
The information gathered by the field test are now used to realize graphene-perovskite/silicon tandem panels. The last achievements of this tandem technology will be discussed.

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Electric polarization of van der Waals crystals and their heterostructures probed on the atomic scale

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In this talk I will review our works on the study of electric polarization of van der Waals crystals and their heterostructures, which we probe directly on the atomic scale using scanning probe microscopy [1-3]. In particular, I will discuss the case of hBN (for few-layers down to monolayer crystals), and its dielectric, piezoelectric and ferroelectric properties. I will focus on our recent experiments in which we showed that stacking hBN crystals twisted at small angles, ferroelectric-like domains arranged in triangular superlattices emerge. Our results opened up new possibilities for understanding electric polarization and designing novel 2D devices with ferroelectric properties.

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Cathodoluminescence excitation spectroscopy of 2D materials

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Fast electrons spectroscopies have had huge success for nano-optics [1]. For phase-locked excitations (e. g. surface plasmons) electron energy loss spectroscopy (EELS) is an optical extinction analogue and cathodoluminescence (CL) that of optical scattering [2]. For "incoherent" excitations, EELS also measures optical extinction for atomically thin materials [3,4,5], while CL measures spectra similar to off-resonance CL [3]. Despite clear benefits (link to structural and chemical information, atomic-scale spatial resolution and broadband excitation), electron spectroscopies have some penalties which limit applications to nano-optics: lack of resonant excitation and polarization degrees of freedom and still limited spectral resolution (EELS). In this contribution, we will discuss how temporally resolved spectroscopies can mitigate some of these issues, specifically for 2D materials, as h-BN and TMD heterostructures.

The lack of excitation energy control can be circumvented by measuring the energy lost by each electron in time coincidence EELS-CL experiments. This has been achieved using a nanosecond-resolved direct electron detector (Timepix3) [6], correlation electronics and a PMT. The information retrieved here is analogous to that of photoluminescence excitation spectroscopy (PLE), hence we name it cathodoluminescence excitation spectroscopy (CLE) [7]. With it, we explored the relative quantum efficiency of different excitation energies and decay pathways towards 4.1 eV defect photon emission in h-BN flakes [7] (Figure 1). We observe a higher quantum efficiency towards light emission of energies just above the optical band gap of h-BN. Our future experiments will aim at understanding the excitation mechanisms leading to light emission in transition metal dichalcogenide monolayers encapsulated in thin h-BN flakes [3].

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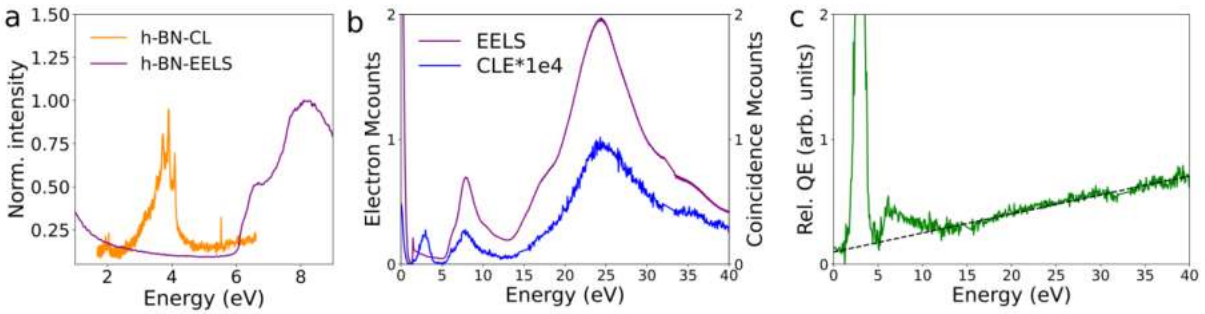

Figure 1: a Average CL an

Molecular and ionic transport in 2D nanochannels and nanopores

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The molecular transport phenomena within nanometer-scale confinements are critical for understanding basic biological and physical processes, as well as the rational design of materials for energy and water technologies. With atomically clean interfaces, sub-nanometer control of geometry, and exquisite control of surface charges and functionalization, 2D materials make an excellent model system.

In this presentation, I will discuss the transport of water and molecules across nanopores in 2D membranes, atomically smooth 2D nanochannels, and laminar graphene-based membranes. Employing a novel micro-pervaporation chamber, we could measure nanoscale transport of liquids and mixtures through individual nanopores and nanochannels with varying height, and we could detect differential fluxes of each component. Comparing different 2D materials, and different liquids and binary mixtures, we could discern the effects of geometry vs. materials properties, and the explore the coupling of molecules with the solid surfaces. Next, I will focus the mechanical coupling of the liquid with the walls of the nanochannels, leading to elastocapillary-driven collapse of channels, i.e. switching. After developing theoretical framework and exploring the switching dynamics, we used such accumulated knowledge to design an active nanofluidic components: nanoswitch and nanocapsule (Figure 1). We demonstrate that nanocapsule could reversibly seal off zepto-liter volumes of liquids – comparable to the volume encapsulated in viruses. Nanocapsules could become elements of integrated nanofluidic circuitry, and could allow us to controllably explore biochemical and biophysical processes in crowded environments.

Figures

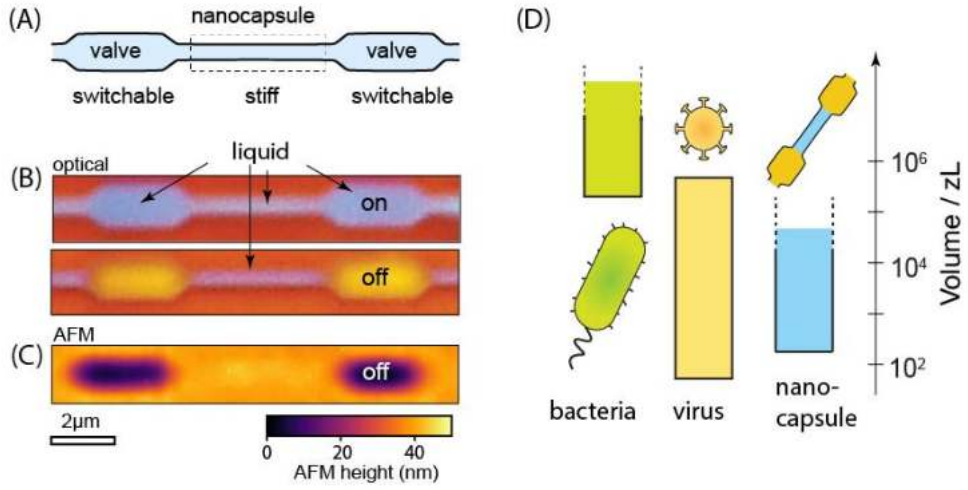

Figure 1: a) Sketch of a nanocapsule based on nanochannels in 2D materials (top view), consisting of a collapsible valve (switches), with a narrow channel section between them (nanocapsule). Elastocapillarity collapses the top walls of the valves during the drying process, capturing a zepto-litter volume of liquid in the stiff capsule. B) Optical image of a nanocapsule in open (top) and closed position with captured water (bottom). C) AFM image shows collapse of top wall of the walls. D) Comparison of volumes enclosed by different structures.

Seebeck measurements on the 2D magnet CrSBr

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The Seebeck effect describes the conversion of heat into electricity and has direct impact on future energy harvesting strategies. However, its intimate coupling to the fundamental electronic properties of a material makes the Seebeck effect also a powerful tool to investigate electronic correlations [1], thermodynamic effects [2], or (quantum)phase transitions with superior precision compared to conventional charge transport measurements. To this end, we implemented few-layer CrSBr into our recently developed cryogenic thermoelectric device architecture [3]. This allowed us to study the magnetic field and temperature dependent thermovoltage in few-layer CrSBr flakes. We observe strong variations of the thermovoltage when magnetic order is changed (PM-AFM, FM-AFM magnetic phase transitions) in the material. Furthermore, we reveal an increase of thermoelectric conversion efficiency by one order of magnitude around 40 Kelvin which we attribute to a spin-entropy change in the material driven by a spin-freezing process.

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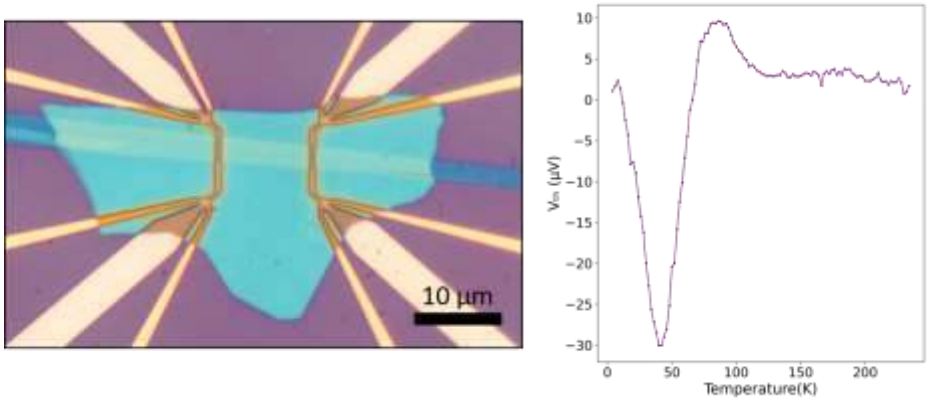

Figure 1: Left: Optical micrograph of a thermoelectric device. A few-layer CrSBr flake is stamped onto gold contacts and encapsulated by a thin hBN flake. Right: Thermovoltage as a function of temperature measured at zero magnetic field.

Engineering and probing new phases in twisted heterostructures of graphene and transition metal dichalcogenides

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Abstract

Twisted van der Waals heterostructures have enriched the phase space of two-dimensional (2D) materials by introducing a new knob to engineer Coulomb interaction, structural symmetry breaking, to electron-phonon interaction, among others. Partnering different genres of 2D materials provides further flexibility in introducing spin-orbit interaction, topological properties etc. The resulting structures are not just ideal platforms for new fundamental discoveries, but also for designing novel device architectures. In this talk, I shall cover a range of such devices, where partnering of twisted bilayers of graphene or transition metal dichalcogenides leads to new correlated phases at unconventional band filling, as well as unexpected device properties when incorporated in field effect devices. The novelty of these properties is probed with multiple experimental techniques, such as thermoelectricity and electrical transport.

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Optics with a twist: Switching with Dirac fermions in Graphene

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The dynamics of 2-D Dirac fermions in graphene, driven by the symmetries of their underlying pseudospins, can manifest in wavefunctions and current transmissions in ways that are fundamentally different from conventional optical interfaces. I will present numerical simulations and experimental demonstrations of several textbook optical effects mirror in graphene, albeit with a twist – specifically (i) trajectories set by *Snell's Law* but *with* ***negative index*** *of refraction* [1], (ii) angular transmission set by *Fresnel's equation* but *with* ***universal transmission*** at normal incidence, (iii) *Brewster angles set by electron pseudospin* rather than photon polarization [2], and (iv) polarizer-analyzers following *Malus' law* but *with half angles* [3]. These subtleties all arise because the photon like bandstructure comes with particle-antiparticle pairs, so that the corresponding band effective mass flips signs at a split-gated PN junction, even while electrons across differently gated graphene segments are unable to alter their group velocity. Symmetry dictates that the pseudospins (mixing coefficients of its frontier dimer pz orbitals) for an N-layer Bernally stacked graphene execute N rotations around the Fermi surface, with a fixed Berry phase Nπ and Chern number of N/2. This implies that the reflectivity at small incident angle θ across the PN junction resembles sinNθ for odd N and cos Nθ for even N, giving alternately perfect head-on transmission (Klein tunneling or KT) or head-on reflection (anti-Klein tunneling or AKT), with an angular sweet spot (Brewster angle). At the same time the higher angle electrons collimate with increasing voltage barrier (refractive index mismatch), so that by placing multiple junctions at an angle, the overall transmission can be quenched much like Malus' law in a polarizer-analyzer pair. The result is a gate tunable transmission gap that can be used to turn graphitic electrons off for bulk samples, both for digital [4, 5] and analog electronic applications [6].

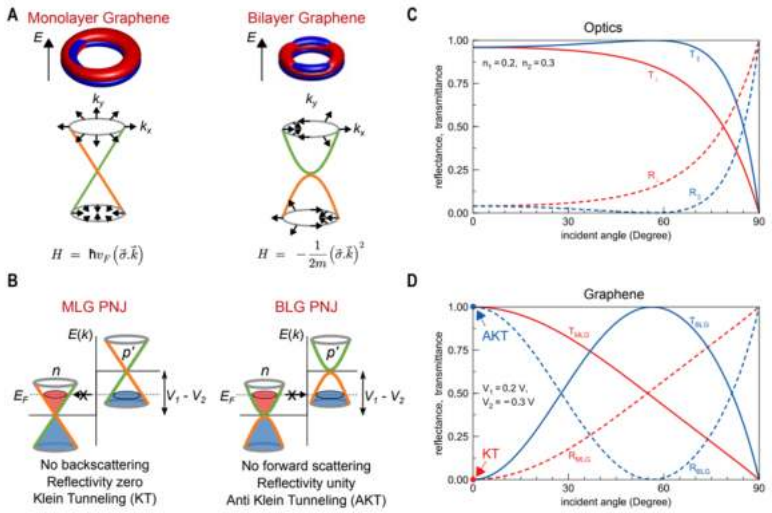

Figure 1: Electron wavefunction symmetries, their matches across a PN junction, and the resulting transmission/reflection plots, contrasting with their optical counterparts (Fresnel law).

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On-surface synthesis of atomically precise carbon nanostructures with tuneable electronic and magnetic properties

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Graphene provides an ideal platform to tune its electronic properties by rational control of its nanoscale structure. Quantum confinement effects in graphene nanoribbons for example can be exploited to tune their electronic band gap and specific edge and hetero-junction topologies can lead to localized in-gap sates showing π-electron magnetism. However, atomically precise synthesis of these graphene-derived nanostructures is the key to fully control their electronic properties.
Here, I will briefly review the concept of on-surface synthesis as a versatile tool to create nanographene materials previously inaccessible via wet-chemistry routes due to insolubility or reactivity of the final structures. Next, I will discuss the concept of localized topological states in GNRs, which can occur at their ends, hetero-junctions or edge extensions. By creating well defined periodic sequences of these topological electronic modes, one-dimensional electronic bands can be created, which are described by the Su-Schrieffer-Heeger (SSH) Hamiltonian representing the dimerized atomic chain [1]. A strategy to realize small-band gap 1D GNR and polymers using the concept of a topological phase transition in a GNR structure family as well as their experimental realization and characterization by scanning tunneling microscopy and spectroscopy of such chains will be presented [2,3]. Finally, I will discuss the magnetic properties of localized π-electron states [4,5].

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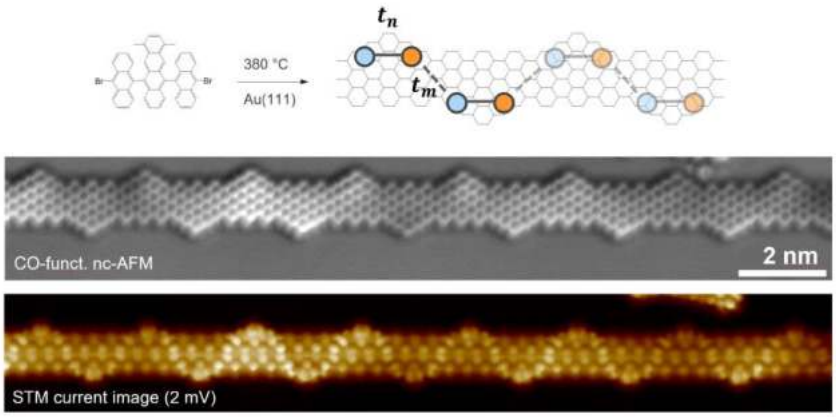

Figure 1: Synthesis pathway and STM/AFM images of an edge-extended armchair graphene nanoribbon (adapted from [1])

Hydrovoltaics

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In contrast to conventional technologies that harvest solar energy by directly converting the energy of light into electrical energy through the photovoltaic effect and kinetic energy of water by mechanical systems, hydrovoltaic effects generate electricity from the direct interaction of graphene and nanostructured materials with water, using the solar energy arriving the Earth indirectly1.

Water is not only the essence of life, but also the largest energy carrier on the earth. Water covers about 70% of the earth's surface, absorbing 70% of the solar energy arriving the earth, and in the atmosphere it can exist in liquid, gaseous and solid states. In human history, through a variety of scientific principles, such as running water driven wheel, steam locomotives, water driven generator as well as the electrokinetic effects, the potential energy or kinetic energy of water can be converted into useful mechanical motion and electrical energy according to the principles of classical mechanics and electromagnetic dynamics2. In the recent decade, hydrovoltaic effects include waving potential3, drawing potential4, evaporation-induced electric potential5 or evaporating potential6 have been found. With hydrovoltaic effects, energy from flowing, waving, dropping, condensing, as well as evaporating water can now be harvested, significantly extending our capability in harvesting environmental energy, leading to the emerging hydrovoltaic technology7 and hydrovoltaics: New ways of harvesting electricity from water8.

In the past years, intensive efforts have been devoted to hydrovoltaics with notable developments made. The power generation has been improved by several orders with incorporation of new materials and devices, putting the Hydrovoltaic Energy on the Way9.

Here, we will review the recent advances in hydrovoltaics for harvesting environmental energy10,11, serving as a potential Negative thermal emission energy technology12, and envision the future directions for hydrovoltaics, from hydrovoltaic energy, to hydrovoltaic ecology and hydrovoltaic intelligence.

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Scanning Probe Lithography for Localized 2D Material Bio-Functionalization

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Scanning Probe Lithography (SPL) techniques like dip-pen nanolithography (DPN), polymer pen lithography (PPL) and spotting with microchannel cantilevers (µCS) offer unique opportunities for highly-localized chemical surface functionalization with resolutions in the micron to even nanometer scales (Figure 1). All these techniques have unique strengths in terms of resolution, obtainable throughput and patterning speed and broad compatibility with delicate chemical and biological inks [1]. Generally, they offer mild process parameters and are capable of multiplexing (i.e. deposition of different compounds within a desired micropattern). Hence, these methods are inherently of special interest for the generation of bioactive surfaces in biomedical applications [2].
One particular subset of applications for SPL methods is the printing of tailored supported lipid membranes (SLM) by DPN with phospholipids (L-DPN). This allows for highly targeted and multiplexed deposition of SLMs e.g. to graphene / graphene oxide surfaces that show unique properties in their interaction, thus could be used for biosensing applications [3,4]. Also, complex functional proteins can be deposited into arbitrary shaped micropatterns, e.g. to enable positioning of cells into specific places on a surface [5]. The talk will give a brief overview over SPL methods and introduce some examples specifically relevant for functionalization of 2D materials with bioactive or biological materials.

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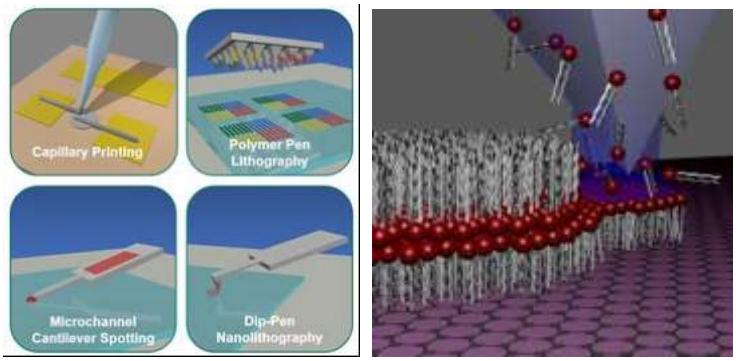

Figure 1: SPL methods (left) and a DPN tip writing a lipid membrane on graphene (right).

Synthesis, characterization, and quantum properties of ultrapure transition metal dichalcogenide crystals

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Two-dimensional transition metal dichalcogenides (TMDs) have attracted tremendous interest due to the unusual electronic and optoelectronic properties of isolated monolayers and the ability to assemble diverse monolayers into complex heterostructures. To understand the intrinsic properties of TMDs and fully realize their potential in applications and fundamental studies, high-purity materials are required. This talk will review progress in synthesis and characterization of TMD crystals using a flux synthesis technique. Using scanning tunnelling microscopy, we identify dominant charged and isovalent defects in these crystals. The flux method reduces density of both defect types by roughly one order of magnitude compared to crystals grown by chemical vapor transport. To further reduce defect density, we have implemented a two-step method [1] that minimizes possible sources of contamination. We find that the two-step method reduces density of by roughly an order of magnitude compared to the single-step flux technique. For WSe_2, we show that increasing the Se:W ratio during growth further reduces point defect density, with crystals grown at 100:1 ratio achieving charged and isovalent defect densities below 10^{10} cm^{-2} and 10^{11} cm^{-2}, respectively. STM spectroscopy and imaging, combined with conductive AFM imaging and statistical analysis, provide clues as to the nature and location of these defects. Initial temperature-dependent electrical transport measurements of monolayer WSe_2 yield record values for both room-temperature and low-temperature mobility, as well as fully developed quantum Hall states. Using these high-quality crystals, we demonstrate that WSe_2 can function as a high-quality barrier in Josephson junctions toward compact qbits.

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Figures

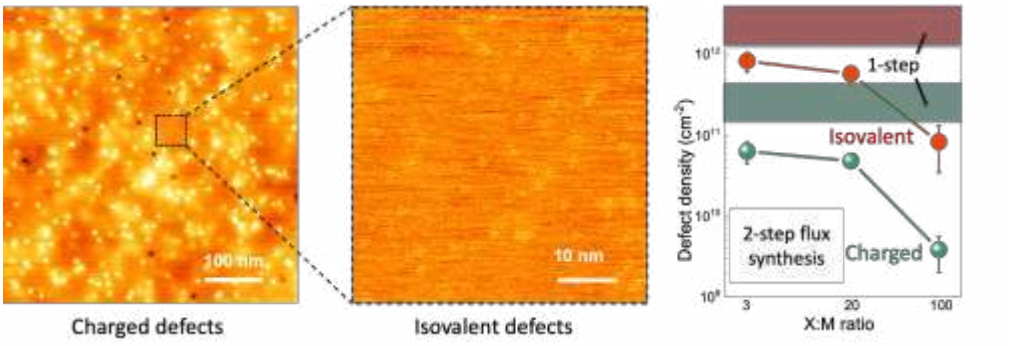

Figure 1: Defects in flux-grown TMDs. Left, large-scale STM image showing charged donor (bright) and acceptor (dark) defects. Middle, smaller-scale STM image showing isovalent defects. Right, density of both defect types as a function of selenium:tungsten ratio.

Twist-angle controlled spin-valve operations with all van der Waals magnetic tunnel junctions

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Two-dimensional magnetic materials and their heterostructures with atomically clean van der Waals interfaces provide an ideal experimental platform for investigating spintronic device operations. Twistronics, a research field that exploits the misalignment angles of van der Waals heterostructures with 2D single-crystalline layered materials, has recently gained significant attention. However, the experimental demonstration of 'spin twistronics' employing the versatility of twist angles with 2D magnets has not yet been realized. Here, we present the twist-angle dependent spin-valve operations with all van der Waals-assembled vertical magnetic tunnel junctions made of the 2D metallic ferromagnet Fe_3GeTe_2 (FGT) and the tunnel insulator hexagonal boron nitride (hBN). Our results demonstrate that the vertical spin-dependent charge transport of the FGT–hBN–FGT heterostructures is highly sensitive to the twist angles and the relative spin configurations of the 2D metallic ferromagnets. We observe a tunnelling magnetoresistance ratio up to ~400% for the FGT–hBN–FGT spin valve with a ~2° twist angle, which decreases continuously to ~100% and lower as the misalignment angle increases up to 30°. The experimental realization of twist-angle-dependent spin-valve operations presents a promising direction for expanding the spintronic device capabilities with low-dimensional van der Waals magnets.

Figures

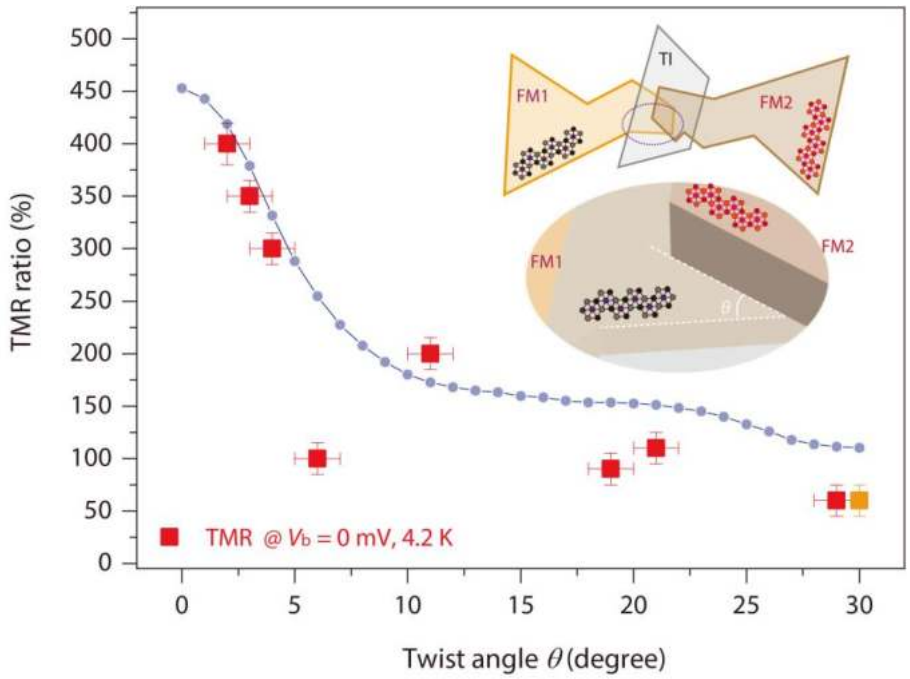

Figure 1: Twist-angle dependent TMR ratio variation from FGT–hBN–FGT magnetic tunnel junctions

Selective transport of water molecules through cation-controlled interlayer spaces in graphite and vermiculite systems

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Ion channels of living cells play a crucial role in life's day to day functioning. Understanding and mimicking their functionalities will provide a breakthrough in the membrane-based desalination, chemical separation, and dialysis technologies. Moreover, when the fluidic channel dimensions approach the sizes of ions and molecules, interesting effects such as deformation of hydration shells, steric effects, van der Waals and even quantum related effects are expected to emerge. These studies require channels of sizes in the sub-nm range which is extremely difficult to fabricate. We chose single crystals of graphite and vermiculite and modified the interlayer spaces with the help of salt intercalation. In the case of graphite, the use of aqueous KCl ions and an electric field is found to expand the interlayer spaces sufficiently large to allow the passage of water molecules. When low voltage or a concentration gradient is applied across the expanded graphite samples, it rejects > 99 % of the sea salt ions, making them ideal for desalination applications. Majority of the ions are rejected based on the steric exclusion, however a small proportion (< 1%) is transported exhibiting hydration energy dependant conductance. On the other hand, vermiculite interlayer spaces are tuned by the intercalation of cations such as K^+, Na^+, Ca^{2+} and Al^{3+}, which made them water stable. 600 nm thick Na-intercalated vermiculite membrane exhibits dye rejection efficiencies >99% with high water permeation rates. We also achieved a high salt rejection efficiency of 95% with a 1.2 µm thick membranes. The water flux and salt rejection are found to be a function of external pressure and membrane thickness. These cation-intercalated vermiculite membranes exhibit several properties related to highly confined systems such as conductance saturation at low concentrations of 10 mM, and K^+ mobility enhancement.

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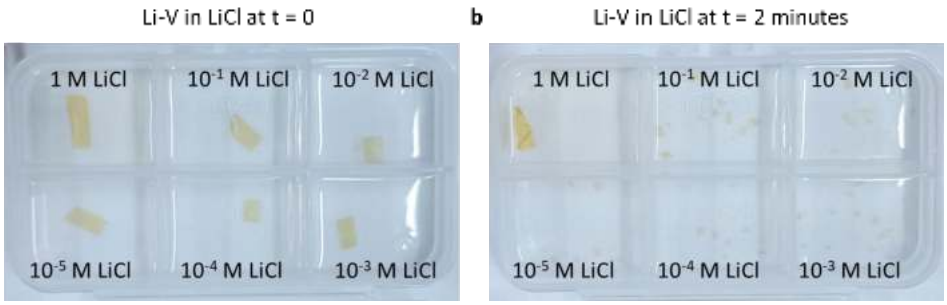

Figure 1: The stability of Li-Vermiculite membrane in aqueous LiCl solution. Li-V membrane in various concentrations of LiCl ranging from 1 M to 10^{-5} M at time (a) t = 0, and (b) t = 2 minutes.

Optical probing of topology and interactions in graphene moiré systems

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Twisted two-dimensional layers have unfolded as the ultimate frontier in quantum materials. The engineering and understanding of their electronic, magnetic and optical properties calls for novel probing techniques. We present various nanoscale infrared and terahertz optoelectronic probing schemes that shed light on the interplay between between topology and interactions. The power of photoresponse lies in its ability to diagnose a wide range of phases resulting from broken symmetries in moiré materials. This understanding is essential for the design of quantum material systems with tailored properties.

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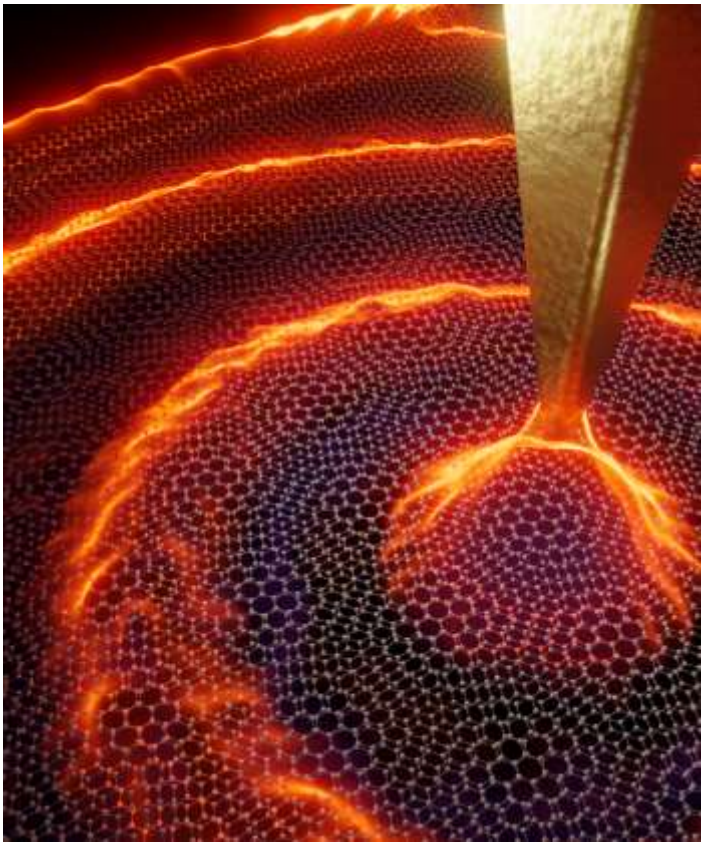

Engineering non-covalent graphene oxide complexes with biomolecules for therapeutic use

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Graphene oxide (GO) - the oxidized form of graphene - has become one the most investigated materials in the biomedical field due to its hydrophilicity and improved compatibility with biological systems. We have made a systematic bottom-up effort during the last decade to generate 'medical grade' GO suspensions. These stable water-based suspensions can be reproducibly synthesized with minimum structural, chemical, and biological impurities, are produced from graphite following a modified Hummers' method [1]. Further improvements ensure endotoxin-free [2] suspensions of single-to-few layer GO sheets of the highest chemical purity. Such 'medical grade' 2D materials have been produced in a range of different lateral dimensions [3] to assess the impact of this structural parameter on biological responses and reactivity [4]. All the above-mentioned properties turn GO into a potential platform for drug delivery, enhancing the therapeutic activity of carried molecules by increasing their bioavailability. Most of the protocols described in the literature to either chemically conjugate or non-covalently complexed drug molecules onto the GO surface are lacking in detailed molecular and atomic characterisation, that makes reproducibility very challenging. This talk will describe how to engineer a robust GO platform for biomedical applications, non-covalently complexed to active biomolecules regardless of their molecular weight, ranging from protein, peptides, and small molecules. pH, water-solubility, and dose of biomolecule are the key parameters that will determine the quality of the colloidally and chemically stable complexes. We are currently working on non-covalent GO platforms for cancer immunotherapy [5], vaccine development, neuropharmacological applications, antiviral drug, and anti-cancer therapeutics [6].

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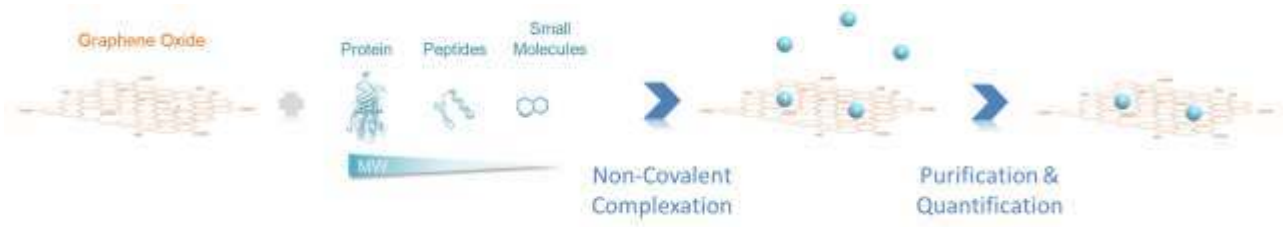

Figure 1: Schematic of engineered non-covalent 'medical grade' GO complexes for biomedical use.

Scanning Tunnelling Microscopy of Twisted Transition Metal Dichalcogenides

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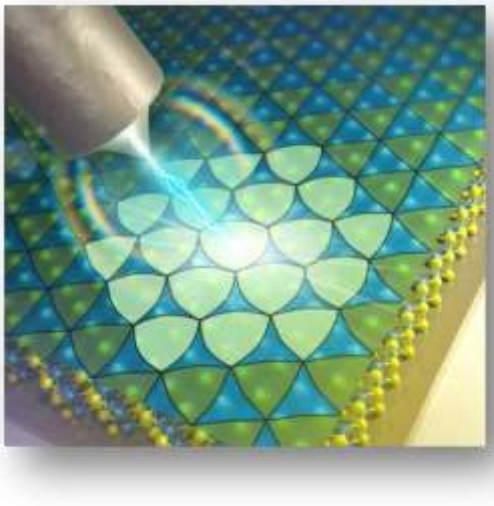
In this talk, I will focus on scanning tunnelling microscopy and spectroscopy experiments aimed at creating novel moiré structures by twisting 2D layers, including the demonstration of reversible local response of domain wall networks in ferroelectric interfaces of marginally twisted WS2 bilayers and spectroscopic evidence of flat bands in antiparallel twisted WS2 bilayers. I will also discuss our progress in realizing quantum-confined devices in WS2 monolayers including demonstration of gate-defined quantum dots, charge detectors and 1D transport.

Figures



Synthesis and processing of Xenes for functional applications

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As a major breakthrough in condensed matter physics, the isolation of graphene paved the way to the class of monoelemental two-dimensional materials known as Xenes. Currently, this category of materials comprises two generations: the first one includes elements from the IV column of the periodic table, such as silicene, germanene, and stanene, and the second generation is based on elements from adjacent columns, such as borophene, antimonene, tellurene, etc. [1].
Here, we will take as case studies the epitaxy of silicene, stanene, and their heterostructure as well as the synthesis of emerging Xenes like blue-phosphorene, and tellurene [2-3]. We will critically review the key aspects of the Xene synthesis, in terms of scalability and stability, which are fundamental for the exploitation of Xenes in nanotechnology applications. Furthermore, we will summarize the ongoing efforts to develop process schemes for the Xene handling and integration into device platforms [4]. In this respect, we will focus on the recent realization of bendable Xene membranes to prove their versatility in flexible devices, such as strain sensors and piezoresistors. Acknowledgement: funding from H2020 ERC-COG grant n. 772261 "XFab" and ERC-PoC 2022 Grant N. 101069262 "XMem".

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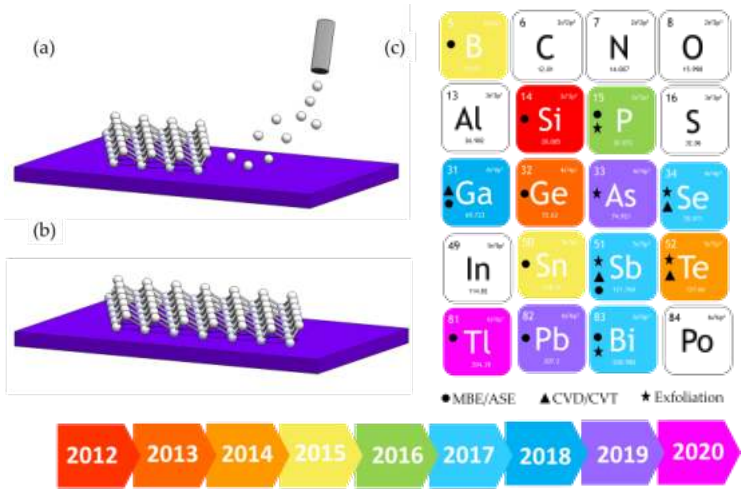

Figure 1: (a)-(b) sketches of the Xene synthesis on a substrate. (c) The periodic table of Xene discovered so far (from 2012 to 2020) and the methods (MBE-CVD-Exfoliation) used for their synthesis. Figure adapted from [1].

A Platform of 3D Printed Devices to Power Wearable Sensors

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Abstract

3D-printing a sustainable manufacturing technique with applications in different industries, from automotive to aerospace, medicine, and energy.
At small scale, 3D printing can enable the fabrication of miniaturized electrodes with free form factors and high mass loading over small footprint areas. 2D materials with outstanding electrochemical properties are suitable to serve in energy conversion and storage devices.
In this talk, I will present our work on 3D printed miniaturized electrodes in the form of supercapacitors and rechargeable batteries beyond lithium. We use earth-abundant electrode materials and water-based electrolytes or gel electrolytes to manufacture energy storage devices to meet the growing energy demand to power wearable and portable electronic devices.
I will discuss the materials challenges in formulating inks of nanomaterials and different architectural design to optimize the electrochemical performance. I will then show how these devices can power wearable sensors.

Fabrication and analysis of 2D and mixed-dimensional heterostructures from combinations of aligned stacking, e-beam patterning, ion doping and molecular sandwiching

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Aberration-corrected electron microscopy is a versatile tool not only for analyzing, but also for manipulating materials down to the level of single atoms. I will present a recent development where we combine spatially controlled modifications of 2D materials, using focused electron irradiation or electron beam induced etching, with the layer-by-layer assembly of van der Waals heterostructures [1] (Fig. 1). A new transfer and assembly process makes it possible to stack the layers under observation in an electron microscope, such that pre-patterned features can be aligned to each other. The aligned stacking of individually patterned 2D materials layers can be considered as a form of 3D printing, where each layer is only one or a few atoms thick, and features within each layer can be defined with a nm-scale resolution. Beyond the results of Ref. [1], I will also present from encapsulation of molecules between graphene layers for electron microscopy studies, new ways of doping graphene with nitrogen, and applications of such doped nanocarbons for gas adsorption.

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Figures

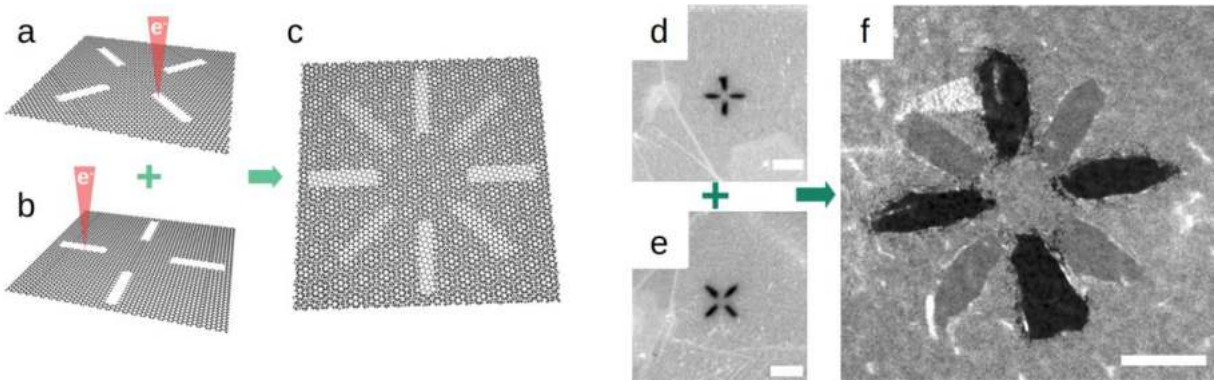

Figure 1: (a-c) Schematic of cutting a pattern into individual graphene layers (a,b) followed by aligned stacking (c). (d-f) Experimental realization, (d,e) SEM images of crosshair structures cut into graphene, (f) Dark-field TEM image showing the assembled structure, set for highlighting one of the two layers. Scale bars are (d,e) 500 nm and (f) 200 nm. Adapted from Ref. 1.

Printable Graphene-Sustainable Elastomer-based Sensors for Point-of-Care Diagnostics

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Abstract
Point-of-care diagnostic devices to monitor various healthcare indicators can not only be life-saving but seems quintessential to develop to complement the advancement in artificial intelligence and machine learning to create an IoT-rich healthcare sector. One of the major working arenas to extract useful information from a person's body is the fabrication of body-conformable small strain sensors capable of detecting muscle movement, pulse, and phonetics. Further, the rise in electronics fabrication to create an IoT-based world would also generate a lot of electronic waste. Therefore, epoxidized natural rubber (a sustainable elastomer) and graphene have been leveraged to prepare the requisite sensing device. Epoxidized natural rubber (ENR 50) and graphene interact via oxirane and vinylidene groups, thereby delivering a highly conducting ink. Thus, rheologically optimized inks are stencil printed to develop a single patch that can monitor multiple physiological parameters. Prepared sensors can detect even tiny strains (<1%) with a gauge factor (GF) of 819 and large strains of human skin stretching (<20%) with a GF of 93.5. Moreover, at only 0.49% volume fraction, the percolation threshold has been achieved. To realize the applicability of the sensor in real-time monitoring, it has been attached to an Arduino UNO board and Bluetooth module to wirelessly transfer the data to the mobile phone, thereby providing a promising route to fabricate multifunctional sensors for IoT-based healthcare devices.

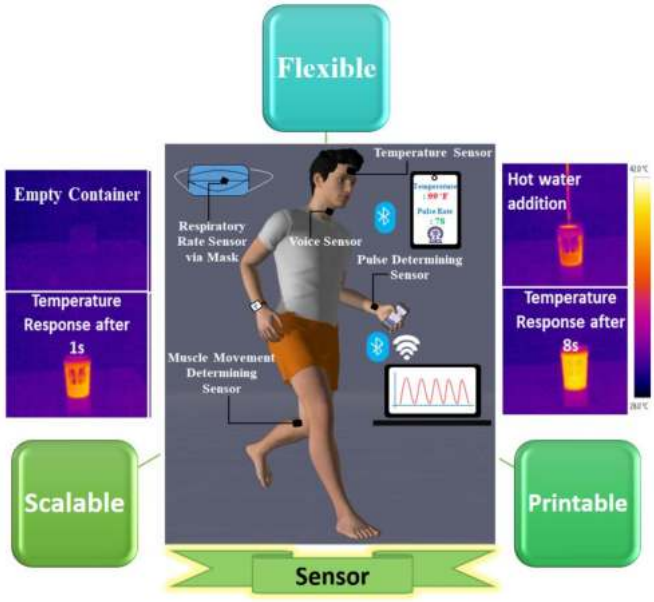

Figure 1: Printable Sensor Based on Graphene-Sustainable Elastomer and its Capability to Decipher Various Physiological Signals.

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Emergent 2D magnetism in MBE-grown van der Waals superstructures

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Two-dimensional (2D) quantum materials and their integrated superstructures host emergent phenomena associated with reduced dimensionality, modified lattice symmetry, and enhanced proximity coupling. Such 2D quantum materials and their superstructures have been mainly fabricated by a top-down approach by combining exfoliation, pick-up, and dry-transfer techniques, while a bottom-up synthesis by molecular-beam epitaxy (MBE) should provide an alternative approach, enabling growth and integration of different types of materials including hardly-cleavable, chemically-unstable, as well as thermally-metastable compounds. We have so far established a route to layer-by-layer epitaxial growth of a number of 2D materials by MBE on insulating substrates [1], and explored their transport properties in the 2D regime [2-6]. In this presentation, we will introduce our recent activities on exploration of emergent 2D magnetism in MBE-grown van der Waals superstructures [7-9]. We will in particular focus on an emergent ferromagnetic state in 2D $NbSe_2$ induced by a strong proximity coupling at a magnetic van der Waals interface with a 2D ferromagnet V_5Se_8 [8], which was proven by the anomalous-Hall effect signals unique to ferromagnetic $NbSe_2$. Owing to the strong spin-valley locking effect in 2D $NbSe_2$ associated with a characteristic Zeeman-type spin-orbit interaction, this emergent ferromagnetic state should accompany a ferrovalley state with spontaneous valley polarization as well. We will also introduce our attempts to fill a van der Waals gap of 2D $NbSe_2$ with 3*d* transition-metal ions to form an "intercalated van der Waals superstructure" [9], which should provide a unique and powerful approach to exploration of emergent 2D magnetic and topological properties.

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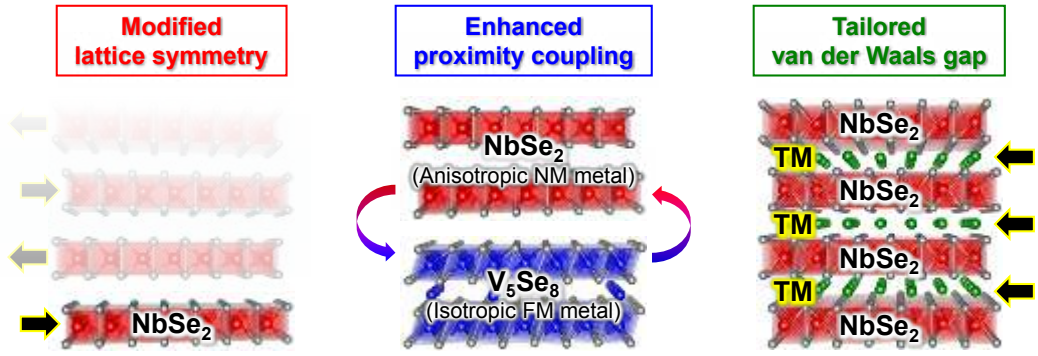

Figure 1: Exploring emergent 2D properties in MBE-grown van der Waals superstructures

Tweaking Atomic Layer Interfaces for Heterogeneous Catalysis to Energy Storage

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Our past research on atomic layer (2D) interfaces has shown that their interfaces play a seminal role in catalysis (photo/electro) while the pristine layers are inert towards the respective process.[1-4] Interfacing of 2D layers seems to be an effective method in developing atomic layers based next generation catalysts which are scalable and reproducible. This addresses one of the lacunas of doped atomic layers based catalysts, where though they are effective,[5] their controllable reproduction is in question due to the variations in the resultant structures with the differences in the synthesis conditions.[6] The interlayer rotation is found to be insignificant in the catalytic performance while the layer stacking sequence seems to be quite important.[1,7] Different types of non-covalent (van der Waals) and covalent interfaces were studied in the past using atomic layers such as graphene, hBN, MoS_2 etc. Moreover, our recent studies show that some such atomic layer platforms are also interesting in single atom catalysis where they can address the issues related to the existing single atom platforms. Atomic membranes can also play vital roles in mass transfer process, as our recent studies indicated. The talk will be focussing on some of our recent studies on atomic layer interfaces in catalysis and energy storage applications,[8] after giving a brief account of our past works on the related area.

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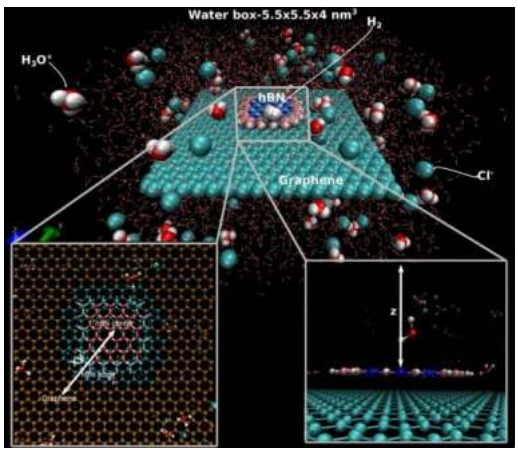

Figure 1: An MD simulation snap shot of Graphene-hBN Interface showing enhanced catalytic (hydrogen evolution reaction) efficacy of the interface.[7]

Nanofluidics in Carbon Nanotube Porins

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Abstract
Nanofluidic systems provide an opportunity to control ion and water transport on a molecular scale, which is important for applications ranging from precision separations to industrial water treatment. Living systems, which move ions and small molecules across biological membranes using protein pores, often rely on finely controlled nanoscale confinement effects to achieve efficient and exquisitely selective transport. I will show that carbon nanotube porins—pore channels formed by ultra-short carbon nanotubes assembled in a lipid membrane—can exploit similar physical principles to transport water [1], protons [2], and ions [3] with efficiency that rivals and sometimes exceeds that of biological channels. I will discuss how molecular confinement, slip flow, and the nature of the pore walls influence the mechanisms of ion diffusion, ion selectivity, electrophoretic transport, and electroosmotic coupling in these nanopores. Overall, carbon nanotube porins represent simple, versatile, and highly controlled biomimetic membrane pores that provide an ideal test bed for development of the next generation of separation technologies.

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Figures

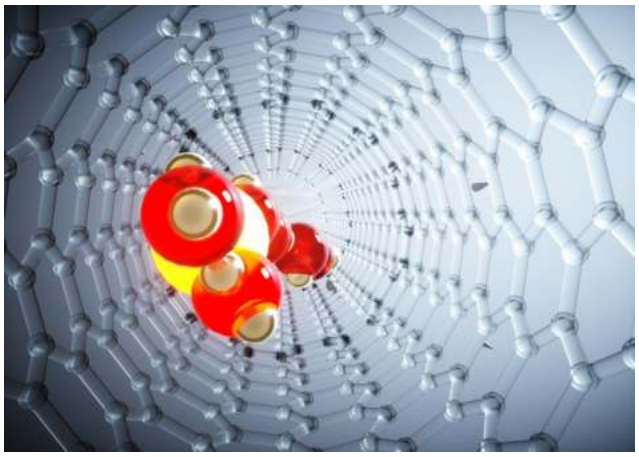

Figure 1: Artist view of an ion-water cluster inside a carbon nanotube.

Sustainable graphene manufacturing from wastes and its lightweight thermoplastic composites

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Plastic waste is a growing environmental and climate concern that threatens the ecosystem and leads to soil and water contamination. Although plastic recycling provides several benefits, the recycled plastics do not have the same performance as virgin plastic composites. Instead of traditional recycling processes, it is possible to produce high value-added carbon nanomaterials by using a rich hydrocarbon source in plastics and rubbery materials which are also primary source for graphene. At this point, upcycling is a significant concept to bring an end to the life cycle of materials and open various new application routes for nanomaterial production. The present work provides an insight into the importance of green synthesis methods in graphene nanomaterials synthesis by combining recycling and upcycling technologies. It is observed that different plastic wastes based on their aromaticity and alifaticity can lead to the formation of different dimensional graphene structures such as 2D sheets and 3D spheres. The produced graphene materials are used for the design of lightweight composite structures for automotive and plastic industry by reducing adverse environmental impacts and adopting energy-efficient manufacturing technologies. In addition, a scalable technology is developed to produce graphene nanoplatelets (GNP) from recycled carbon black obtained from the pyrolysis of waste tire by using recycling and upcycling technology. These graphenes produced from waste sources are comparably cheaper and eco-friendly than available ones produced from graphite flakes and thus can carry significant potential to be used as co-reinforcing agent together with primary reinforcements in thermoplastic composites. With this developed technology, a pilot production line with the capacity of 1 ton/month in Nanografen Co (start-up of Sabancı University) was established to initiate the industrialization of graphene in thermoplastic market. Nanografen becomes an official supplier of Renault and they developed OEM certified masterbatch to reduce glass fiber and mineral amount in polypropylene based interior and exterior parts. Consequently, this multidisciplinary work ensures significant innovation potential of graphene in the field of thermoplastic-based composites and overcomes the needs by addressing greenhouse gas emissions with sustainable designs.

DFT and QM/MM simulations of nanoconfined water between electrified gold surfaces using Non-Equillibrium Green's Functions

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Albeit water is the most common and best studied solvent, understanding its structure and properties at the surface of materials is still an open problem. Besides, important modifications of its structure and dynamics occur when the surfaces are electrified (as in electrochemical environments), and when the confinement space is nanometric. Density Functional Theory (DFT) simulations can deal with these issues, although imposing the external voltage in the simulation has proven difficult. Non-Equillibrium Greens functions (NEGF) techniques [1,2] as implemented in the SIESTA DFT package [3,4] are used here to address this problem, allowing first-principles molecular dynamics simulations of nanoconfined water in the presence of a finite voltage between the two confining surfaces. We will present proof of concept calculations of water between gold electrodes, showing the potential of our approach. We also show how to increase the size of systems (in terms of number of atoms) and the simulation time which can be addressed by these simulations by using a quantum mechanics/molecular mechanics (QM/MM) approach coupled to the NEGF method, to investigate the metal-water interaction, providing a good balance between accuracy and computational cost. We validated our results against full QM calculations and analysed the performance. This hybrid approach emerges as a viable way of studying way larger systems compared to those currently used to investigate the dynamics of electrified metal-water interfaces using first principles.

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Figures

$Au - (H_2O)_{236 \times n} - Au \qquad (n=3)$

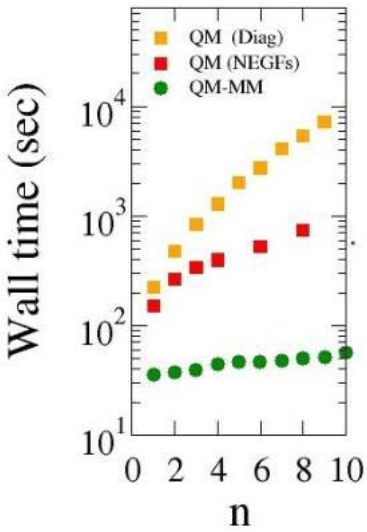

Figure 1: Top: QM/MM setup for nanoconfined water between gold electrodes. Systems containing repetitions of the 236 H_2O molecules were used for timing purposes. Right: CPU time per MD step for n=1 to n=10, showing the wall time needed per Molecular Dynamics step, for the full QM approach using straight diagonalization (yellow points), the NEGF approach (red points) and the QM/MM hybrid method (green points). The calculations were done using 384 Intel Platinum 8160 (2.1GHz) cores of MareNostrum IV at the Barcelona Supercomputing Centre.

2D Amorphous Materials and their Applications

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Bulk amorphous materials have been studied extensively and are widely used in industry, yet their detailed atomic arrangement is unknown. Unlike bulk materials, the structure of 2D or monolayer amorphous carbon (MAC) can be determined by atomic-resolution imaging [1]. Recently we have shown that such a structure can indeed be synthesised as free-standing membrane and is topologically different from its crystalline counterpart. Interest in atomically thin amorphous materials has recently surged, because they exhibit properties not observed in their crystalline counterparts. From an application point of view, they have the potential to address bottlenecks in both quantum device fabrication and industry. I will give three examples.

First, I will discuss the low temperature synthesis, by laser-assisted chemical vapour deposition of free-standing monolayer amorphous carbon/amorphous graphene. Extensive characterization by transmission electron microscopy reveals the complete absence of long-range periodicity and a threefold-coordinated structure with a wide distribution of bond lengths, bond angles, and five-, six-, seven- and eight-member rings. Electronic characterization of free-standing MAC shows that it is insulating, with resistivity values comparable to boron nitride grown by chemical vapour deposition. Also, it is surprisingly stable and deforms to a high breaking strength, without crack propagation from the point of fracture. I will discuss its potential use both as a multifunctional barrier film for applications such as magnetic media and interconnects. Next, I will discuss how such a 2D film can be synthesised as nanoporous monolithic amorphous carbon foam using Spark Plasma Sintering (SPS). I will discuss its potential for applications, e.g., as supercapacitors. II will conclude my talk by discussing the use of amorphous BN as an atomic sieve for dilute magnetic doping of 2D materials. I will demonstrate its potential for creating ferromagnetic superconductors through an usual RKKY coupling within the superconducting gap.

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Scientific and technological opportunities with emerging two-dimensional ferroelectric semiconductors

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The electronic band structure of a semiconductor crystal underpins its physical and chemical properties. Unlike traditional semiconductors with imperfect surfaces, the emergent class of van der Waals (vdW) semiconductors can be readily cleaved or exfoliated to reveal high quality surfaces ideally suited to the imaging and tuning of band structures. Here, we utilise angle-resolved photoemission spectroscopy and high magnetic fields to reveal the unique band structure of the van der Waals semiconductor In_2Se_3, a ferroelectric material of broad interest for science and technologies [1].
As in many III-VI compounds, the electronic band structure of In_2Se_3 undergoes changes in the transition from the bulk to the single layer. In many III-VI semiconductors, such as InSe, this transition is accompanied by the emergence of a ring-shaped valence band maximum referred to as an inverted Mexican hat or camelback dispersion with hole effective masses much heavier than in traditional semiconductors [2]. In contrast, we show that this unusually shaped band exists in bulk In_2Se_3 and that it can be tailored by the specific polymorph [3] and stacking (1T, 2H and 3R) of the vdW layers. The significance of this unusually shaped band and the ability to modify it are motivated by the possibility to create new forms of magnetic and charge order driven by weakly screened electron correlations. Thus, these results provide the foundation for band engineering of carrier correlations for emerging technologies. Finally, we report on ferroelectric semiconductor junctions in which the ferroelectric In_2Se_3 is embedded between two single-layer graphene electrodes. In these two-terminal devices, the ferroelectric polarization of the nanometer-thick In_2Se_3 layer modulates the transmission of electrons across the graphene/In_2Se_3 interface, leading to memristive effects that can be controlled by applied voltages and/or by light [4].

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Non-identical moiré twins in bilayer graphene

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The relative angular alignment between 2D layers of a van der Waals (vdW) heterostructure can dramatically alter its fundamental properties [1]. A striking example is the recent observation of strongly correlated states and intrinsic superconductivity in twisted bilayer graphene [2]. Another remarkable effect of angular layer alignment, predicted for certain vdW heterostructures, is the emergence of phases of matter with non-trivial topological properties, where charge carriers flow without dissipation, being protected against perturbations. In graphene aligned with boron nitride (BN), such a phase has been predicted, with topological protection linked not to the spin, as commonly observed, but rather to the valley degree of freedom.

The experimental observations of these topological valley currents [3] has been largely put in question by theorist, results of numerical simulation [4] and recent scanning SQUID results [5]. In these, the observed non-local signal has been attributed mostly to localized states on the edge of graphene. In this talk, we will show how these two pictures are not incompatible and can be re-conciliated if we take the angular layer alignment into account.

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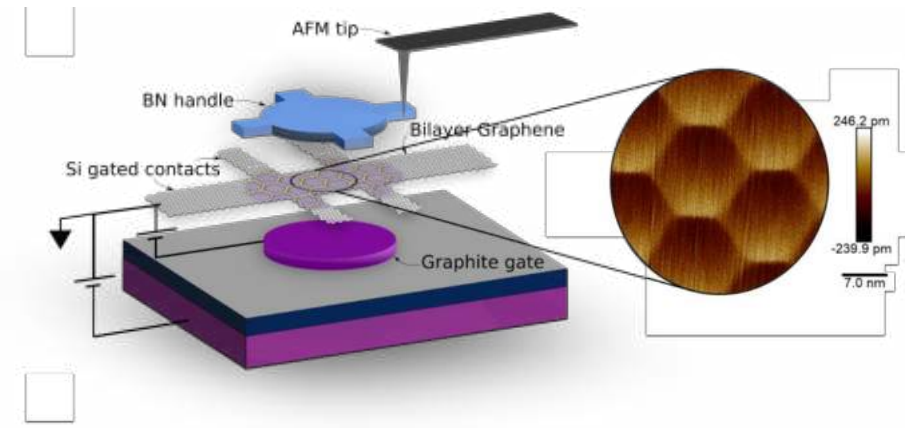

Figure: Schematics of our rotatable van der Waals heterostructure and AFM image of a bilayer graphene/BN moiré superlattice.

Advances in pressure and chemical sensing with 2D materials

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Abstract

Physical and chemical sensor bear a direct societal impact on well-being, which includes, along others, the monitoring of human's health, the quality and composition of the air we breathe, the water we drink, and the food we eat.
Physical sensors are essential components for the fabrication of devices for medical diagnosis and health monitoring, upon use of active materials with sensitivities in the low-pressure or medium-pressure range, respectively. In this framework, flexible piezoresistive pressure sensors are compatible with wearable technologies for digital healthcare, human-machine interfaces and robotics. Among active materials for pressure sensing, graphene-based materials are extremely promising because of their outstanding physical characteristics. Currently, a key challenge in pressure sensing is the sensitivity enhancement through the fine tuning of the active material's electro-mechanical properties. We have achieved this by combining chemically reduced graphene oxide (rGO) with (macro)molecular materials with controlled mechanical properties. Among these, the use of a polybenzoxazine thermoresist matrix in combination with rGO made it possible to tailor electrically conductive nanocomposites where the thermally triggered resist's polymerisation modulates the active material rigidity and consequently the piezoresistive response to pressure. [1]
In chemical sensing higher sensitivity, faster response time and fast recovery time can be achieved by using 2D materials as active components. Instead, selectivity can be achieved either through the optimization of the energy levels of the analyte with respect of those of the active material, as demonstrated via the fabrication of sensors of heavy metals[2] or polyaromatic molecules[3], or through the functionalization of the latter with supramolecular receptors ensuring a high discrimination of ions and small molecules in the sensing event.[4]
Strategies for simple integration of working devices with electrical read-outs will be presented.

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Ti3C2Tx in ophthalmic medical device interventions

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The properties of MXenes lend themselves to bioactive medical device applications. One such application is the use of MXenes within devices for the treatment of cataracts, the primary cause of preventable blindness globally. Current treatment replaces the opaque natural lens with a transparent polymer plastic device called an intraocular lens, allowing a return to visual clarity. However, complications occur including loss of accommodative focus and potential complications associated with a poor wound healing response. We have considered the use of Ti3C2Tx as a transparent, conductive, hydrophilic and flexible coating within an accommodative IOL design. Ti3C2Tx facilitated a stimuli responsive shift in accommodative power and repression of an inflammatory and fibrotic response linked to loss of device function over time. Our studies suggest a compatible device which may be further explored for propensity to moderate dysregulated wound healing to resolve, in this example, loss of accommodative focus and reduce complications leading to ophthalmic device failure. These materials may also be used in bioactive lens devices for sensor diagnostics.

Atomically Thin 2-Dimensional Materials for Electrocatalysis

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Abstract

Graphene research has sparked great interest in a wide range of 2-dimensional layered materials with varying electronic properties. Atomically thin layered transition metal dichalcogenides (TMDs) such as MoS2, WS2, MoSe2 and WSe2 have been emerging as the cutting edge in materials science and engineering, due to their interesting electronic properties.[1] These materials open up new opportunities for a variety of applications, including optoelectronics, energy conversion, and catalysis. To realize their potential device applications, it is highly desirable to achieve controllable growth of these layered nanomaterials, with tunable structure and morphology.[2-4] TMDs exhibit promising catalytic properties for hydrogen generation and several approaches including defect engineering have been shown to increase the active catalytic sites.[5,6,7] The talk will present some of our efforts on morphological and electrocatalytic studies of engineered TMD nanomaterials and their heterostructures. Some of our recent works on the controllable growth of high-quality, ultra-thin flakes of elemental 2D materials such as bilayer selenene and its heterostructures with MoS_2 monolayers will also be discussed.[8] Our work highlights the opportunities for tuning the electrocatalytic properties of atomically thin electrocatalysts based on defect engineering and surface modification.

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A Roadmap for Disruptive Applications and Heterogeneous Integration Using Two-Dimensional Materials: State-of-the-Art and Technological Challenges

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This talk will attempt to establish a roadmap for 2-Dimensional (2D) material-based Nanoelectronic technologies for beyond Si and other future/disruptive applications with a vision for the semiconductor industry to enable a universal technology platform for heterogeneous integration. The heterogeneous integration would involve integrating orthogonal capabilities such as different forms of computing (classical, neuromorphic and quantum), all forms of sensing, digital and analog memories, energy harvesting, etc. – all in a single chip using a universal technology platform. This talk will also cover the technological and fundamental challenges in pushing the 2D technology to the market, where the world stands today, and what gaps are required to be filled. Talking about the gaps, I will particularly touch base on the Metal (3D) to graphene/TMD (2D) contact engineering challenges, which has been considered as one of the most fundamental challenges towards harnessing the full potential of 2-dimensional materials. And, how the fundamental understanding of the contact's quantum chemistry resulted in unique ways to engineer it, resulting into record transistor performance. Besides, I will talk about some of the fundamental process challenges which can unintentionally perturb the 2D channel's electrical, optical and mechanical properties. In the end, I will talk about some of the reliability gaps, which are urgently required to be addressed and the fundamental understanding we have developed so far.

Unconventional Phase Transitions in Layered 2D Crystals

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In this talk, we will introduce our recent studies on anomalous phase transitions in physical properties of layered materials. First, we will show a metal-to-Mott insulating phase transition (MIT) as a function of a number of layers in 1T-$TaSe_2$ [1]. Using our newly developed first-principles calculation method for the extended Hubbard functionals [2], we reveal that the intricate competition between the strong screened Coulomb interaction and kinetic energy gain across the layers is a key to the MIT here. We show that our simulations of spectroscopic signals near the MIT agree with experiments from our collaborators very well. Second, we will present a new theory on anomalous charge density wave (CDW) transition in kagome metals of AV_3Sb_5 (A = K, Rb, Cs) [3]. Using our newly developed molecular dynamics simulation tools, we uncover asynchronous condensation processes to the charge density wave states in kagome metals. We demonstrate that the CDW forms first within each layer but their phases fluctuate across the kagome layers owing to unavoidable degeneracy in energetic costs for stacking CDWs. We will discuss several consequences and experimental implications for kagome metal physics and layered crystals based on our discoveries of condensation of preformed CDW orders. If time allowed, we also present strain-induced magnetic transitions in a new carbon 2D crystal of Biphenylene Network (BPN) [4,5].

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In 2D: Ferroelectric Switch of Electronic Topology, and the Flexo-Ferroelectricity of Actuator Motor

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The low dimensionality (of 1D nanowires [1] or 2D monolayers [2-4]) brings about two distinctly new features to ferroelectric: (I) its polarization ***P*** can be along, longitudinal (***P***$_l$) or perpendicular, transverse (***P***$_t$) with respect to the material line or plane—a strong anisotropy due to low-D, absent for 3D crystals. (II) Compliance, to bend easily into "empty-dimension", a new degree of freedom on display. We discuss examples, emerging from (I) and (II), where flipping intrinsic ***P*** switches something else—the essence of multiferroics, where electrical and magnetic polarizations are coupled (also possible in low-D, e.g. in MXene [2]). We show how ±***P***$_t$ flip switches the topological state of 2DM [3] or, in very different manifestation, can alter the shape, performing as actuator [4].

We design and explore with DFT a hetero-bilayer of realistic 2D components of matching lattice symmetry, the β-phase antimonene β-Sb (known for its strong spin−orbit coupling) and ferroelectric In_2Se_3. The ±***P*** of the In_2Se_3 induces distinctly different electronics in the bilayer: if ***P*** points "inward", the bilayer is a trivial insulator; when switched "outward", its state is nontrivial topological, $Z_2 = 1$ — inviting future multifunctional devices' applications [3].

In another striking example, the often-overlooked (blocked by the substrate) flexibility of 2DM leads to unexpected behavior of the ferroelectrics. We introduce a "ferro-flexo" coupling term $\sim P\cdot\kappa$ into the Ginzburg-Landau-Devonshire φ^4-energy, to connect the ferroelectricity and curvature, κ. The DFT and Monte-Carlo methods allow one to quantify all parameters, so to predict the spontaneous curvature κ_s and the effects on phase transition (rising Curie temperature, domain wall width, etc.) for InP, $CuInP_2S_6$ and In_2Se_3 as chemically-specific representatives. Polarization switches do bend the layer, converting electrical signal to movement as an actuator, with efficient work-cycle.

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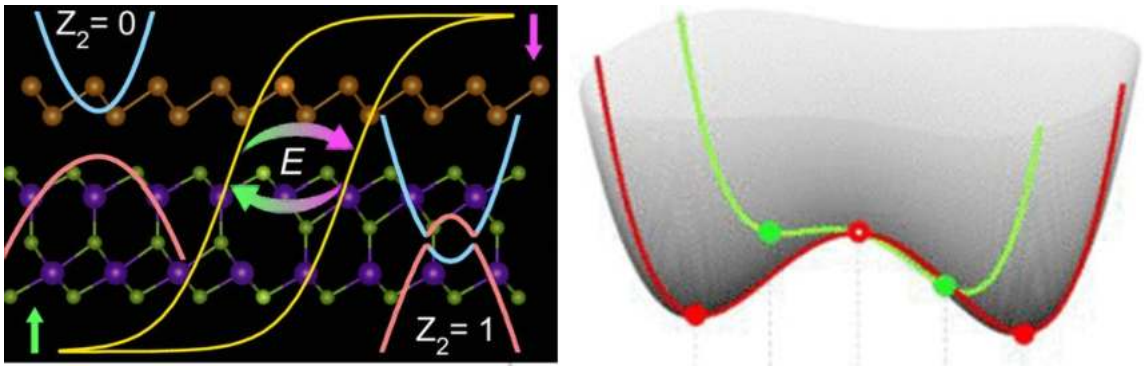

Figure 1: Left: The hysteresis of *P*, switching the topological state [3]. Right: The generalized φ^4-potential surface $U(P,\kappa)$ of a 2DM flexo-ferroelectric, flat (green) or with relaxation (red) [4].

Layertronics of valley electrons in twisted bilayers

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Long wavelength moiré pattern in van der Waals stacked 2D materials has provided a powerful tool towards designer quantum materials that can extend the exotic properties of the building blocks. For band edge carriers located at the Brillouin zone corners (valleys), the interlayer coupling features sensitive dependence on the atomic registry between the constituting layers. In twisted TMDs homobilayers, such coupling in the moiré pattern manifests itself as a location-dependent Zeeman field acting on the active layer pseudospin. Berry curvature arising from such layer pseudospin texture corresponds to a pseudo-magnetic field that realizes fluxed honeycomb superlattices with layer-sublattice locking [1,2], underlying the recently observed gate-programmable magnetic states [3] and quantum anomalous Hall effect [4]. Ultrafast dynamic modulation of the layer pseudospin texture by bias pulse or Terahertz field further introduces pseudo-electric field for spin/valley manipulation [5]. In moiré patterns distorted by non-uniform strains, the interplay of moiré interlayer coupling and strain together leads to non-Abelian Berry phase effects [6]. I will also discuss novel Hall effects of band geometric origin arising from the layer pseudospin structures in twisted homobilayers [7,8].

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Graphene and 2D materials: from cancer research to antivirals

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Abstract

Graphene and 2D materials have attracted a lot of attention in biomedical sciences since their discovery, especially in cancer research. Thanks to their extraordinary properties, we have been using them to diagnose cancer, delivery chemotherapeutics or exert anti-tumor activities through various mechanisms. The COVID-19 pandemic has shown the success of nanotechnology in combating with viruses in different applications including the development of vaccines or personal protective equipment (PPE). Among these nanotechnology-based systems, two-dimensional (2D) materials with intrinsic physiochemical properties can efficiently favor antimicrobial activity and maintain a safer environment to protect people against pathogens. During this presentation, the antiviral studies performed using 2D materials will be discussed by shedding light on how these materials can reduce the chance of infection effectively. 2D materials can be used alone or combined for the disinfection process of microbes, antiviral or antibacterial surface coatings, air filtering of medical equipment like face masks, or antimicrobial drug delivery systems. At the same time, they are promising candidates to deal with the issues of conventional antimicrobial approaches such as low efficacy and high cost. Specific attention will be also provided in order to suggest new approaches to develop and design novel antimicrobial agents using 2D materials for combating global infectious diseases in the future.

Embedding Ethics in Research Practices

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Over the past decades, an increasing number of national and international policy statements and guidelines such as the Horizon 2020 funding programme of the European Commission directed the attention of researchers toward 'Responsible Research and Innovation' (RRI). This broad movement emphasises a number of key points such as ethical conduct; transparency; social utility; anticipating impacts and assessing alternatives; public outreach; critical reflection; and stakeholder collaboration. Nanoscience and nanotechnology research programs have been exemplar for developing a 'safe, integrated and responsible strategy' including the anticipation of potential impacts and the establishment of effective dialogue with all stakeholders. However the desire for more responsible research and innovation does effectively impact the daily practices of researchers? What is the impact of RRI policy? The study of the potential safety impacts of engineered nanomaterials for both human health and the environment has been included as a necessary component of nanotechnologies.

But how nanoscientists understand RRI and how does it affect their daily practices of research and development? While nanosafety research has become a central focus with toxicological and ecotoxicological studies, other aspects such as public outreach, collaboration with stakeholders upstream in research projects, and assessing alternatives are often perceived as too far from the actual conditions to become an integral part of competitive researcher.

I will argue that the divergence in the norms and values between what is being presented as RRI and what the key characteristics of good science can be reduced when scientists are able to look beyond their laboratories and institutions and confront the real world, its messiness, complexities and uncertainties. This cannot be achieved in the practices of individual research scientists without a range of institutional changes, such as more cultural diversity in research training and recruitment , supporting and facilitating interdisciplinary knowledge and competence in academic institutions and changing the evaluation criteria of what counts as good science.

Nanomaterials and Graphene for Cancer Therapeutics: A Patient Public Involvement and Engagement Study

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We were interested in gaining a deeper understanding of patients' opinion and acceptability on the use of nanomaterials to treat cancer. 2D-Health researchers developed a patient-led questionnaire to explore patient and public views on our technology and to gain an understanding of their knowledge and acceptance of nanotherapeutics. Then the questionnaire was circulated widely through social media networks of various cancer patient groups and the captured data analysed and communicated. We aimed to educate and understand how we can better communicate our scientific research and help bridge any gaps in knowledge with patients that would potentially be users of our technologies.

The initial step was the questionnaire design between 2D-Health researcher volunteers and eight patients with a range of cancer types that were recruited to advise upon the development of the questionnaire. The questionnaire contained 45 questions aimed to capture information relating for four key areas: demographics, baseline knowledge, acceptance of immunotherapy containing nanomaterials and informed understanding/acceptance using infographics to convey information. Following ethical approval, the questionnaire was circulated online. 242 responses to the Questionnaire were received, with 147 complete and quality-validated responses used for data analysis. The respondents identified their gender, had a range of educational backgrounds and were aged from 18-70+ years old. The majority of respondents were cancer patients (65%) diagnosed with a range of malignancies and at all Stages from I-IV.

The results from this survey indicated that 26% of respondents were familiar with the term 'nanomaterials' at start of survey, with average rating of understanding 3.4 [scale 0-10]. After viewing the educational infographic this increased to 5.5 [scale 0-10] and further increased to 6.7 by the end of the Questionnaire. In response to any concerns about accepting a cancer therapy containing graphene, the majority of respondents had no concerns and were generally accepting of graphene-based treatments. This was regardless of age/gender/cancer stage/severity of side effects. Finally, after reading an infographic about how graphene is used in cancer treatment, respondents were asked to rate [scale 0-10] their understanding of how graphene could be used as a potential cancer therapeutic agent and the likelihood of them accepting graphene as a potential cancer therapeutic agent. The response was again positive with an average score of 8/10 for acceptance.

Overall, this PPI&E project highlighted the importance of direct engagement between researchers and patients to gauge patient perception when developing new technologies. The total cohort of patients we interacted with at various levels of the project were generally very accepting of novel therapeutic methods, including graphene-based immunotherapies. We will use the data to guide our future research and to design clear information for patients.

Water-based, Defect-free and Biocompatible Graphene for Biomedical Applications

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The outstanding properties of graphene make this material very attractive for biomedical applications. However, these applications require the material to be stable in water and in the medium, to be biocompatible and able to penetrate the cellular membrane. Graphene oxide is therefore the most used graphene derivative for biomedical applications.
In this talk I will present an alternative approach, based on supramolecular chemistry [1], that enables to produce defect free, highly concentrated, stable, and biocompatible graphene dispersions in water [2]. Non-covalent functionalization enables to easy tune the surface chemistry of graphene, hence allowing to produce amphoteric, anionic and cationic graphene dispersions [3-6]. Amongst them, cationic graphene dispersions show excellent stability in the medium and the highest cellular uptake, making them very attractive for a wide range of biomedical applications.

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Graphene oxide: a promising delivery platform to enhance cancer immunotherapy

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Despite cancer immunotherapies such as checkpoint inhibitors having demonstrated remarkable clinical efficacy in recent years, they remain effective in only a limited proportion of patients, with many experiencing severe inflammatory side effects. Novel approaches for activating and maintaining more targeted and effective anti-tumour immune responses are vitally needed. With properties such as large surface area and versatile functionalisation, the nanomaterial graphene oxide (GO) has been suggested as a promising therapeutic delivery platform for bespoke modification of immune responses and inflammation, to enhance the efficacy of cancer immunotherapies. We have investigated the use of GO in complex with immunostimulants such as polyinosinic:polycytidylic acid (Poly (I:C)) for activation of immune cells, and for targeted delivery of immunotherapies to enhance anti-cancer immune responses, *in vitro* and *in vivo*. We have found that appropriately functionalised GO can dramatically augment murine innate and adaptive immune cell activation and function, as well as restrict tumour growth and enhance efficacy of checkpoint inhibitor immunotherapy in murine cancer models. Our data highlight the potential of GO as a modifiable platform to increase effectiveness of cancer immunotherapies.

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2D materials-based intelligent membranes

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Permeation through nanometre-pore materials has been attracting unwavering interest due to fundamental differences in governing mechanisms at macroscopic and molecular scales, the importance of water permeation in living systems, and relevance for filtration and separation techniques. Latest advances in the fabrication of artificial channels and membranes using two-dimensional (2D) materials have enabled the prospect of understanding the nanoscale and sub-nm scale permeation behavior of water and ions extensively. In particular, several laminate membranes made up of 2D materials show unique permeation properties such as ultrafast permeation of water and molecular sieving. In my talk, I will discuss our recent results on controlling molecular transport through various 2D materials-based membranes by an external parameter and will discuss the prospect of developing next-generation intelligent membranes based on 2D materials.

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pH-dependent water permeability switching and its memory in 1T' MoS_2 membranes

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In recent years, 2D material-based membranes have attracted significant attention for their potential applications in various fields, such as water desalination, gas separation etc. So far, most of the developments have been focused on improving efficient separation processes for industrial applications. However, in the last few years, there has been some effort toward making next-generation intelligent membranes. Intelligent translocation of molecules is one of the vital roles of biological membranes. Mimicking such systems can help us develop smart membranes, which can autonomously change their permeation depending on the external environment and would play a pivotal role in intelligent technologies for tomorrow.

In this talk, I shall discuss about our recent work, where we developed MoS_2 membranes that show phase responsive transport of water molecules and prove that only 1T' phase of MoS_2 is water permeable. We demonstrate the memory effects and stimuli responsive transport through such 1T´ MoS_2 membranes by executing water and ion permeation that follow a pH-dependent hysteresis with a permeation rate that switches by a few orders of magnitude. We further illustrate the potential application of this phenomenon in autonomous wound infection monitoring and pH-dependent nanofiltration.[1]

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Figures

1D Andreev bound states along quantum Hall edges

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A supercurrent flow in a superconductor-normal metal-superconductor junction is made possible via resonances of normal charge carriers (electrons and holes): Andreev bound states, transmitting Cooper pairs in the superconductors. Engineering superconductivity in the quantum Hall regime is a promising route to create novel electronic states [1,2], but, in this regime different carriers move on opposite device sides which necessitates a tedious coupling between distant edges to achieve small supercurrents [3-5].
Here we present a new geometry where quantum Hall edge states are carried along narrow domain walls (DWs) at the centre of the device allowing localised Andreev bound states insensitive to the magnetic field [6]. At magnetic fields as high as 8T, we observe Josephson coupling with relatively large critical currents. We find superconducting interferences between domain walls and Fabry-Pérot oscillations in individual DWs, effects attributed unambiguously to a 1D nature.
Such localised Andreev states may host non-trivial excitations and could be a platform for new devices and applications, such as SQUID magnetometers, Andreev qubits, CQUIDs of fluxon devices [7].

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Magneto-exciton instability and quantum Hall breakdown in graphene

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In this talk, I will present our study of the velocity-induced breakdown of the integer quantum Hall effect (QHE) in monolayer graphene [1]. Low-bias quantum Hall transport is notoriously described in terms of single-electron physics. The situation is different at large bias where electrons can couple to large-momentum collective excitations (magneto-excitons). It was previously revealed in bilayer graphene using radiofrequency shot noise [2] that an intrinsic drift-velocity limit of QHE is the collective magneto-exciton instability, with breakdown velocities mimicking the single-particle Zener inter-Landau-level tunnelling mechanism.
Using the same technique, we demonstrate in this work that the magneto-exciton instability takes a very different form in relativistic single-layer graphene, with a doping- and magnetic field-independent breakdown velocity. Based on theoretical calculations of the excitations spectra [3], we show that this instability takes place at a phase-velocity which is solely controlled by the strength of the interactions and is determined by a value of the magneto-exciton conductivity equal to a universal fraction of the Hall conductance.

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Figures

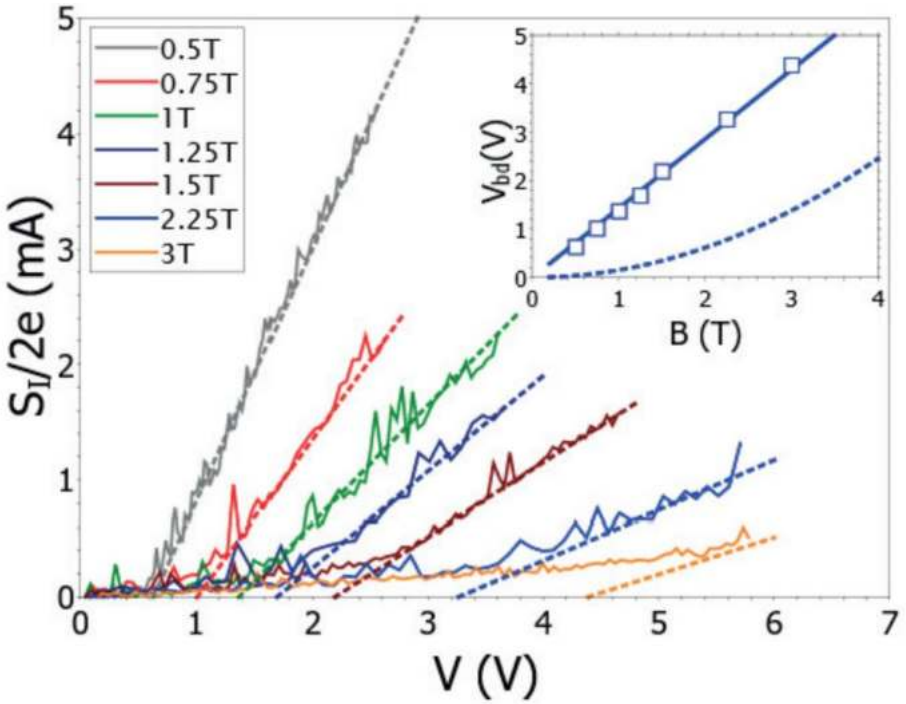

Figure 1: Breakdown shot noise as function of bias voltage, measured at a fixed charge carrier density $n=2.10^{12}$ cm^{-2} for magnetic fields between 0.5T and 3T in a graphene transistor. Breakdown is signalled by a strong increase of noise. Inset shows the linear increase of breakdown voltage with B-field (blue line corresponding to a constant breakdown velocity $v_{BD}=0.14v_F$), that strongly deviates from the Zener inter-Landau-level tunnelling (dashed line).

Near-field radiative heat transfer of hBN-encapsulated Graphene Field Effect Transistors

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Heat management is critical for the performance of most modern electronic devices. The case of Van der Waals materials is no exception, but thanks to their anisotropic layered structure, most of them host collective bulk excitations, called hyperbolic phonon polaritons (HPhP), which are efficient radiative heat carriers. [1,2]

In this talk, I will present a direct observation of the out-of-plane heat transfer in a vertical SiO_2-hBN-graphene-hBN heterostructure. The out-of-plane heat transfer is deduced from the steady-state temperature of the hot 285nm-thin SiO_2 layer in contact with the bottom hexagonal Boron Nitride, when the graphene transistor is operated under a large bias. This temperature is deduced from the characteristic incandescent peaked emission of SiO_2 in the middle-infrared. Using this technique, we show that out-of-plane heat transfer through hBN is triggered by interband tunnelling in graphene at large bias [2], and we quantify the amplitude of this effect. [3]

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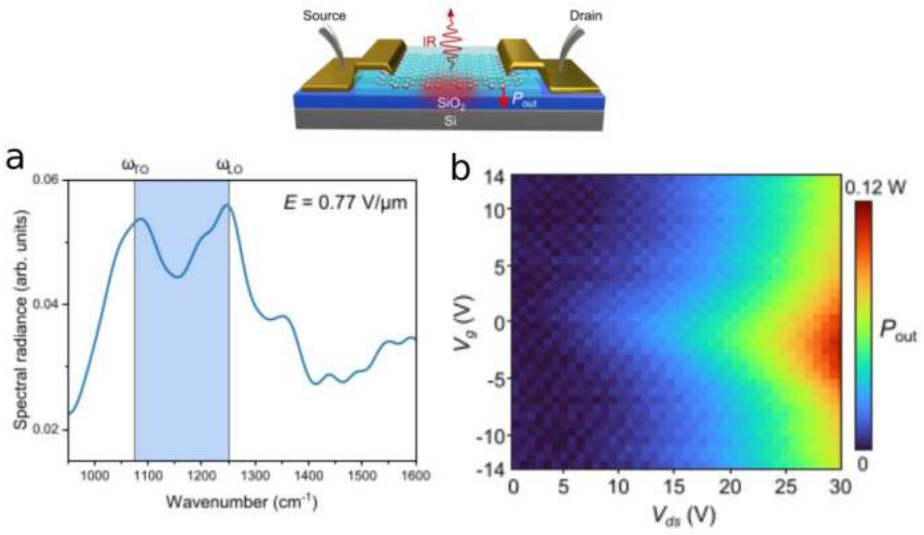

Figure 1: Depiction of the out-of-plane heat transfer in a high-mobility graphene field-effect transistor with an SiO_2 back-gate. The subsequent mid-IR emission is represented by a red wavy arrow. a) MIR spectral radiance of the transistor for an applied electric field E = 0.77 V/μm (Vg = 0 V), the blue-shaded region encloses the Reststrahlen band of SiO_2. b) Out-of-plane dissipated power as a function of electrical bias and gating, deduced from the SiO_2-temperature increase.

Heat transport in h-BN

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2D materials have new and intriguing properties, including their thermal properties. Like 3D materials, there is a wide variety of 2D materials with all ranges of thermal conductivity that can be simply assembled on top of each other [1]. Moreover, the thermal conductivity is extremely anisotropic in these materials. It has been demonstrated in the same 2D heterostructures ratios of 3 orders of magnitude between vertical and in-plane thermal conductivity, a record. Moreover, the interest from a fundamental point of view is the appearance at room temperature of a particular regime of phonon scattering, which allows to describe the heat transport as a hydrodynamic flow [2]. Thanks to these characteristics, it is interesting to study their thermal properties. Here, we focus on the thermal conductivity of suspended h-BN. It is an interesting material because it has a very high thermal conductivity but is electrically insulating. Nevertheless, it is difficult to heat it with a laser because of its low absorbance. To overcome this problem and to perform thermal mapping, we have made two heating bridges of Silicon between which a flake of hBN is suspended. One of the bridges is heated by Joule effect while the other one allows us to calibrate the heat flow through the 2D materials. Raman spectroscopy measurements allow to measure the temperature at each point of the system. We were able to extract a thermal conductivity of h-BN at different temperatures.

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Figures

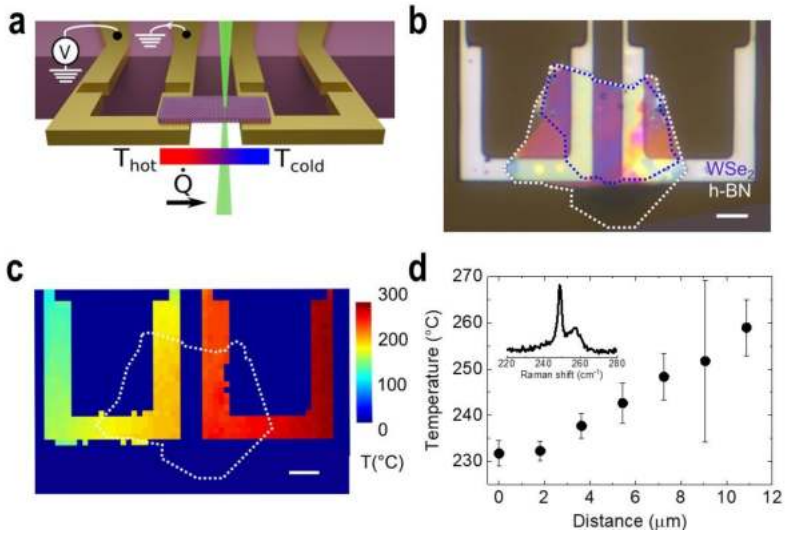

Figure 1: a) b) Schematic sketch (a) and optical image (b) of the device structure with a suspended hBN between two silicon heaters.) c) Temperature map of Si when 6 V are applied on the right electrode. d) Gradient of temperature of WSe_2 along a line from the cold electrode to the hot one (inset: Raman spectrum of WSe_2) All scales bar : 10 microns

Giant optical anisotropy in van der Waals materials

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Abstract

Materials with high optical anisotropy are of great importance in technology and science [1]. Recently, one of the largest birefringence in the visible and near-infrared intervals up to 0.8 was reported in quasi-one-dimensional crystal BaTiS3 [2]. However, anisotropic nanophotonics requires optical anisotropy of about 1.5 to fully exploit advantages of anisotropic properties [3,4]. Inspired by this challenge, we focused on two-dimensional materials and their bulk counterpart – van der Waals (vdW) materials. Our findings showed that their fundamental difference between interlayer strong covalent bonding and interlayer weak van der Waals interaction leads to unprecedented high birefringence with values exceeding 1.5 in the infrared and 3.0 in the visible spectral intervals (for example, see optical constants of MoS_2 in Figure 1). Thus, our studies enable a new field of vdW anisotropic nanophotonics.

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Figures

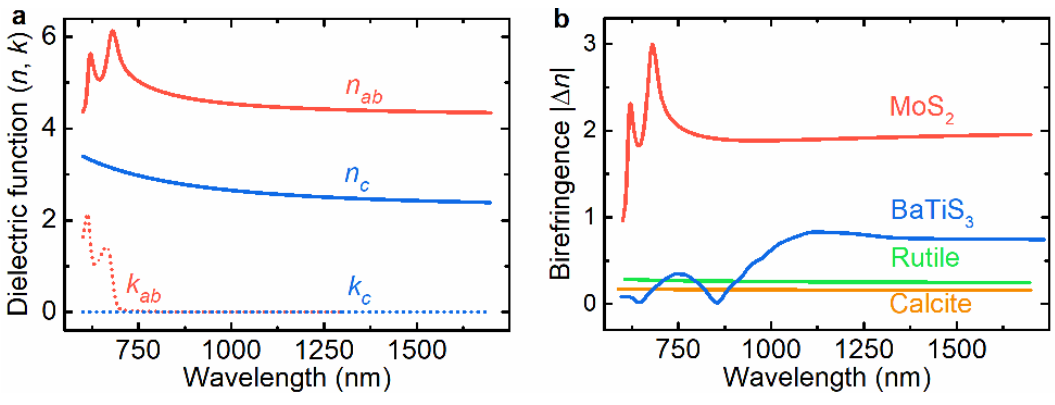

Figure 1: a. Optical constants of MoS_2. **b.** Birefringence of MoS_2 in comparison with other anisotropic materials.

Controlled alignment of supermoiré lattice in double-aligned graphene heterostructures

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Abstract: The supermoiré lattice, built by stacking two moiré patterns, provides a platform for creating flat mini-bands and studying electron correlations [1-3]. An ultimate challenge in assembling a graphene supermoiré lattice is in the deterministic control of its rotational alignment, which is made highly probabilistic due to the random nature of the edge chirality and crystal symmetry of each component layer. In this work [4], we present an experimental strategy to overcome this challenge and realize the controlled alignment of double-aligned hBN/graphene/hBN supermoiré lattice, where graphene is precisely aligned with both the top hBN and the bottom hBN. Remarkably, we find that the crystallographic edge of neighboring graphite can be used to better guide the stacking alignment, as demonstrated by the controlled production of 20 moiré samples with an accuracy better than ~0.2°. Employing this technique, we are the first to fabricate the perfect double-aligned graphene supermoiré lattice and to observe the sharp resistivity peaks at band filling of 0, -4 and -8 electrons per moiré unit cell. Finally, we extend our technique to other strongly correlated electron systems, such as low-angle twisted bilayer graphene and ABC-stacked trilayer graphene, providing a strategy for flat-band engineering in these moiré materials. This work is supported by the MOE Singapore Tier 2 Grant No. MOE-T2EP50120-0015.

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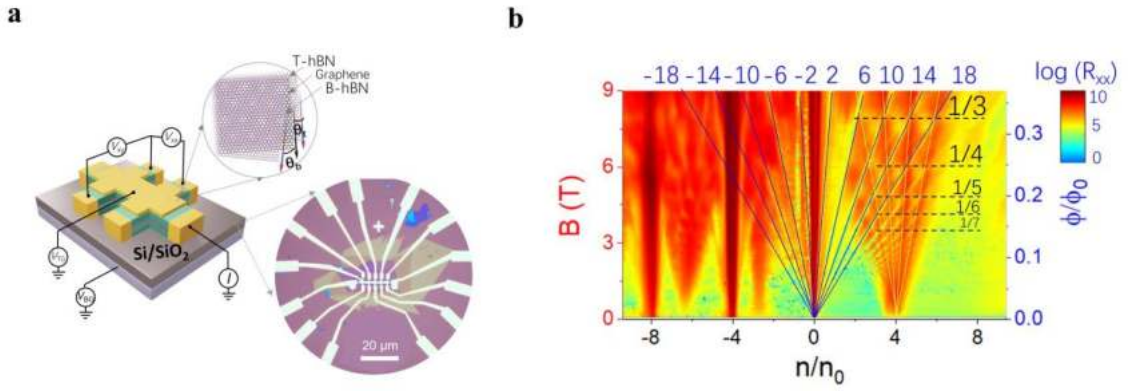

Figure 1: Double-aligned hBN/graphene/hBN heterostructure. a, Schematics of the dual-gate device with supermoiré lattice. **b**, Landau fan diagram showing the Brown-Zak (BZ) oscillations in a perfect double-aligned device.

Graphene-Integrated Steep-Slope Field-Effect Transistor

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The continuous down-scaling of MOSFETs has generated the urgent demand for steep-slope transistors with reduced sub-threshold swing (SS) and low power consumption [1-3]. In this work, single-layer graphene is integrated in the gate structure of a MoS2 channel FET which led to a subthreshold swing as low as 31 mV/dec, well below the "fundamental" limit of 60mV/dec. Exploiting the negative electronic compressibility in single-layer graphene with low density of states, negative quantum capacitance was obtained within a certain gate voltage range. Similar to "traditional" negative capacitance obtained with ferroelectric materials, the negative quantum capacitance enables internal gate voltage amplification and subthreshold swing well below 60 mV/dec [4, 5]. Theoretical simulations reveal the fundamental mechanism which explains the negative quantum capacitance observed when a single-layer graphene is encapsulated (integrated) within the gate stack. The results point to a new pathway for the development high speed and low power transistor devices.

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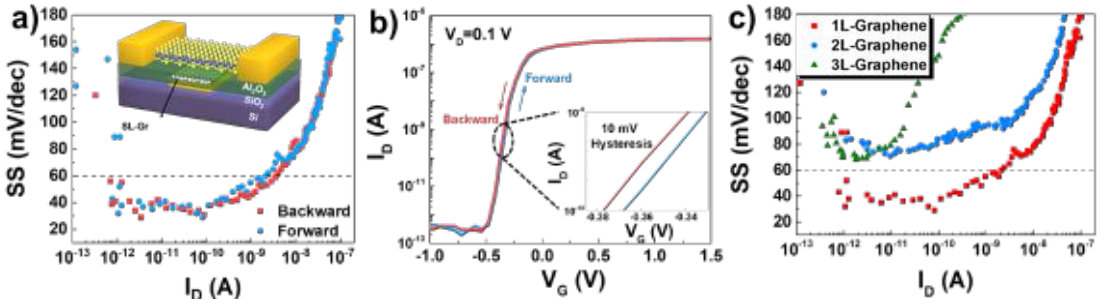

Figure 1: (a) SS values extracted from (b) ID-VG curves. (c) SS values of the device with different graphene layer thickness.

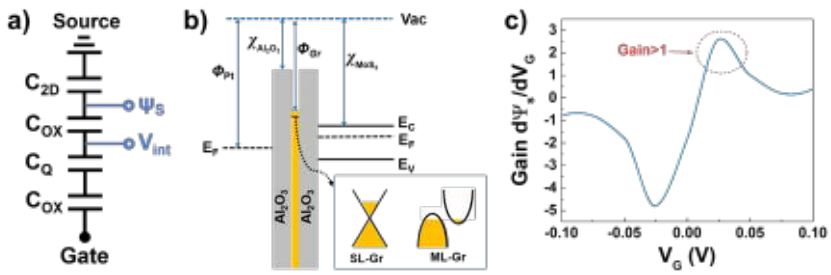

Figure 2: (a) Capacitor divider model and (b) band diagram of the gate stack. (c) Simulation results.

Controlling electrical percolation in thermoplastic-hybrid nanocomposites

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Carbonaceous nanomaterials, including graphene, carbon nanotubes (CNT) and carbon blacks (CB), are excellent fillers for electrically conductive polymer composites due to their low density, high aspect ratio and high conductivity [1,2]. However, the nature of the percolated networks formed within these nanocomposites is still poorly understood due to the complex interdependencies between the filler morphology, processing conditions and network structure. Herein, we present our latest understanding of these interdependencies based upon a detailed combined modelling and experimental approach.

~0D (carbon black), 1D (nanotubes) and 2D (graphenes) nanomaterials were used as reinforcements within polycarbonate. The electrical percolation curves were established for both singular reinforcements and hybrid systems where different combinations of reinforcements (e.g. graphene-CB, CB-CNT etc) were used. The nature of the networks were explored through transmission electron microscopy and conductive AFM (Fig.1a). The experimental work was interpreted using Monte-Carlo simulations where we have focussed on understanding the role of clustering and secondary aggregate formation (Fig.1b). Despite reports of possible synergy between different nanomaterials in the literature, we show that the percolation threshold is ultimately determined by the average aspect ratio of the fillers and, importantly, their clusters. It was found that through selection of the hybrid fillers, one can control the percolation threshold, exponent of the percolation curve and ultimate electrical conductivity within an over-arching parameter window, allowing industry to produce composite systems which are robust against differing processing conditions.
We thank the DPI, European Graphene Flagship, Morgan Adv Materials & RAEng for funding.

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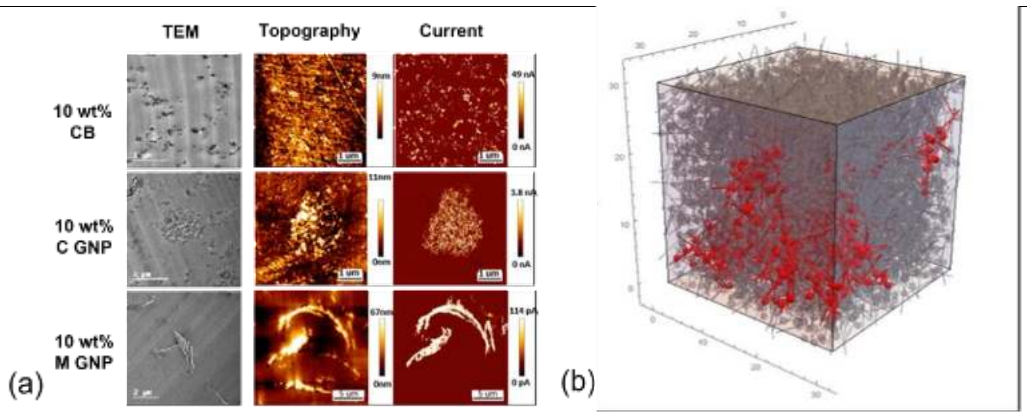

Figure 1: a) TEM image as well as topography and current mapping obtained from conductive-AFM of composites and (b) example Monte-Carlo simulation of hybrid system with the percolated pathway highlighted in red.

Negative Valley Polarization of the Intralayer Exciton via One-Step-Growth of H-type Heterobilayer WS_2/MoS_2

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Abstract

Vertical Type-Ⅱ van der Waals heterobilayers of transition metal dichalcogenides (TMDs) have attracted wide attention due to their distinctive features mostly arising from the emergence of novel electronic structures that include moiré-related phenomena.[1] Owing to strong spin-orbit coupling under noncentrosymmetric environment, TMD heterobilayers host non-equivalent +K and –K valleys of contrasting Berry curvatures, which can be optically controlled by the helicity of optical excitation.[2] The corresponding valley selection rules are well established by not only intralayer excitons but also interlayer excitons.[3] Quite intriguingly, here, we experimentally demonstrate that unusual valley switching can be achieved using the lowest-lying intralayer excitons in H-type heterobilayer WS_2/MoS_2 prepared by one-step growth. This TMD combination provides a very unique case of an ideal interlayer coupling with almost perfect lattice match, thereby also in the momentum space between +K and –K valleys in the H-type heterostructure. The underlying valley-switching mechanism can be understood by bright-to-dark conversion of initially created electrons in the valley of WS_2, followed by interlayer charge transfer to the opposite valley in MoS_2. Our suggested model is also confirmed by the absence of valley switching when the lowest-lying excitons in MoS_2 are directly generated in the heterobilayer. In contrast to the H-type case, we show that no valley switching is observed from R-type heterobilayers prepared by the same method, where interlayer charge transfer does not occur between the opposite valleys. We compare the case with the series of valley polarization data from other heterobilayer combinations obtained under different excitation energies and temperatures. Our unique case of the valley switching mechanism can be utilized for valley manipulation by controlling excitation photon energy together with the photon helicity in valleytronic devices derived from H-type TMD heterobilayers.

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Hybrid Graphene for The Development of Next Generation Cancer Screening Device and Chiral Spintronics Applications

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Abstract
A new breakthrough has been achieved in the design and characterization of spatiotemporal nano-/micro-structural arrangements that have opened up new horizons in graphene based sensor applications, including ultra-fast and on-site biopsy-decision-making for intraoperative surgical oncology, wearable-spectrometry, and spin controlled chiral molecule identification.[1,2] The spatiotemporal sensing arrangement is achieved through the use of scalable, binder-free, functionalized hybrid spin-sensitive (<↑| or <↓|) graphene-ink printed sensing layers on free-standing films made of porous, fibrous, and naturally helical cellulose based quantum sieve in a hierarchically stacked geometrical configuration (HSGC) inspired by butterfly wings. The HSGC operates according to a time-space-resolved architecture that modulates the mass-transfer rate for the separation, elution, and detection of each individual compound within a mixture of various volatile organic compound (VOCs), thus providing a mass spectrogram. The HSGC has the potential to be used in a wide range of applications, including the fast and real-time generation of a spectrogram of VOCs during liquid-biopsy, without the need for any immunochemistry-staining and complex power-hungry cryogenic machines. It can be used for wearable spectrometry that provides a spectral signature of molecular profiles emitted from the skin during various dietary conditions. Furthermore, such spin sensitive detectors could be used for chiral spintronics and quantum computing applications. The applications studied are presented schematically in Figure 1.

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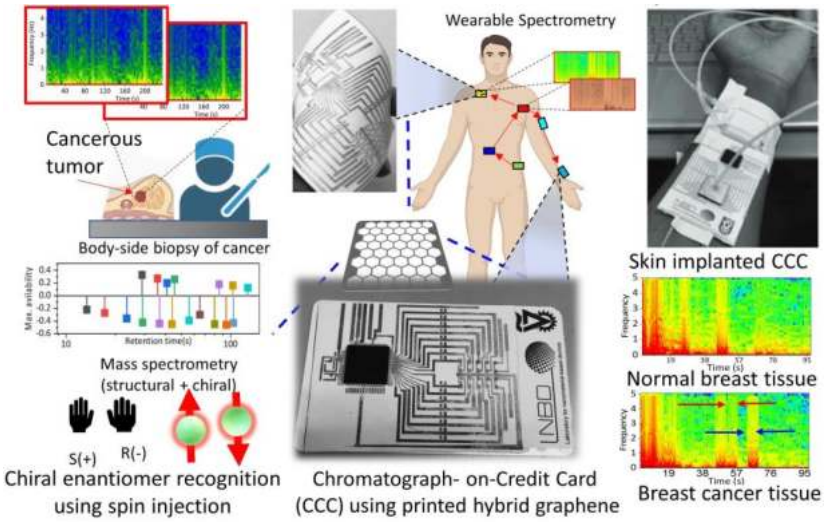

Figure 1: Hybrid Graphene based HSGC integrated on paper for cancer spectrometry, spin sensitive chiral recognition, clinical, medical, and wearable spectrometer applications.

Umklapp-Assisted Electron-Phonon Coupling Enables Ultrafast Cooling in Magic Angle Twisted Bilayer Graphene

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In rotated van der Waals heterostructures, the twist angle controls the moiré lattice constant, which in turn modifies the electron momentum and the phonon spectra. Theoretical studies predict that the moiré potential also strongly affects electron-phonon coupling. [1-4] However direct measurements of moiré-enhanced electron-phonon coupling, and a clear understanding of its origin, are lacking.
Here we reveal the occurrence of electron-phonon Umklapp scattering in twisted bilayer graphene near the magic angle. [5] Using time-resolved photovoltage measurements and a direct analysis based on Boltzmann theory, we show that it provides the main relaxation mechanism for hot carriers across a broad temperature range. By comparing twisted and non-twisted bilayer graphene, we find that a twist angle of 1° speeds up cooling by several orders of magnitude.
Our work demonstrates the ability to engineer electron-phonon coupling and the resulting cooling power in twistronic systems. These results are relevant for transport measurements, while the short carrier lifetime will enable the development of ultrafast photodetectors based on moiré materials.

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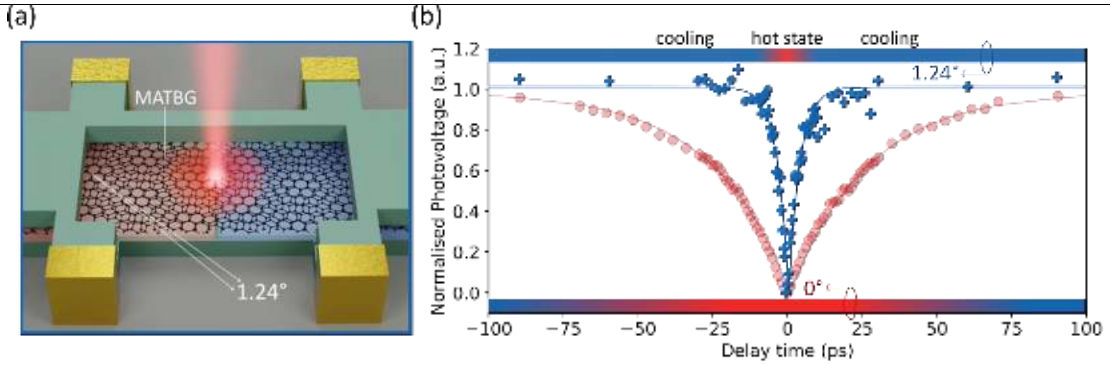

Figure 1: (a) Illustration of the hBN-encapsulated MATBG device. We generate a photovoltage by illuminating the pn-junction (red/blue regions). (b) Controlling the time delay between two ultrafast pulses reveals the hot carrier cooling dynamics. At low temperatures, these are significantly faster in the case of MATBG (1.24°) than non-twisted BLG (0°).

Charge density waves in atomically thin transition metal dichalcogenides

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Understanding complex electronic phases, such as charge density wave (CDW), superconductivity (SC), exciton condensate, and Mott insulator, in two-dimensional (2D) confined geometry offers an interesting perspective toward a comprehensive account of collective electron interactions. Transition metal dichalcogenides (TMDCs) provide an ideal platform for investigating how CDW order evolves in a true 2D limit, often accompanied by competing orders of similar temperature and energy scales. An outstanding question would be how the CDW order evolves as the quantum confinement increases and electron screening reduces with dimensionality approaching 2D and how the competing orders would affect such evolution.

Our approach to this question is to investigate the electronic structure and CDW transition using angle-resolved photoemission (ARPES), scanning tunneling microscopy and spectroscopy (STM/STS), and density functional theory (DFT) calculations on the TMDC samples grown by molecular beam epitaxy (MBE). For example, we have found that CDW order persists in single layers of 2*H*-$NbSe_2$ [1] and 2*H*-$TaSe_2$ [2] with only a moderate increase in transition temperature despite dramatic changes in the low energy electronic structures and Fermi surface topologies. In 1*T*-$TaSe_2$, a robust Mott insulating phase with unusual orbital texture is found in the single layer on top of the well-known star-of-David CDW order [3].

In this talk, I will present more recent examples of CDW orders with exotic origins. In monolayer $IrTe_2$, we have found a large-gap insulating dimer ground state, in which lattice and charge instabilities, as well as local bond formation, collectively enhance and stabilize a charge-ordered ground state [4]. In monolayer $ZrTe_2$, we have discovered a novel band- and energy-dependent folding behavior in ARPES that results in a two-step CDW, which we interpret as the formation of excitonic gas and its condensation into a final CDW state [5].

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High-Speed Optoelectronic Sampling at 1.55 µm with High-mobility Graphene

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In communication systems, radiofrequency (RF) signals are carried by a high carrier frequency. Thus, before being digitalized, the signal is first down converted to baseband using an electronic or optoelectronic sampler. However, all-electronic samplers are limited due to narrow bandwidth, nonlinearities and jitter noise that are introduced by electrical clocks [1,2], as opposed to optoelectronic samplers, with ultra-stable optical pulse trains having extremely low phase noise and jitter (generated by active mode-locked lasers). We show in this presentation improvement over state-of-the-art optoelectronic sampling at 1.55 µm using high mobility graphene (see fig.1), featuring a conversion efficiency similar to the most performant samplers (GaAs, working at 0.8 µm [3]) over a 40 GHz bandwidth with harmonic rejection below –43 dB at 20 GHz. We also show a comprehension of the harmonic rejection origin in graphene, leading to a methodology to optimize device performances.

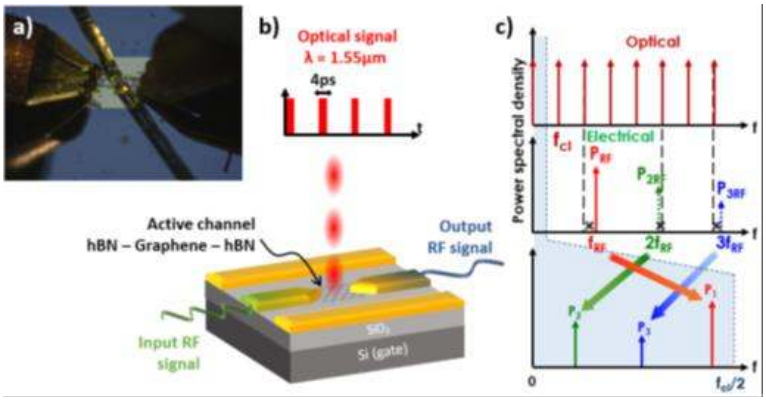

Figure 1: a) Picture of the graphene-based optoelectronic sampler. **b)** Experimental scheme: a 1.55 µm lensed fiber fed by a mode-locked laser illuminates the graphene channel embedded in a coplanar waveguide. RF probes allow both the injection of the high-frequency RF input signal along with DC channel biasing, and the measurement of the down-converted output RF signal. A Si backgate controls the graphene doping. **c)** Principle of subsampling : the optical clock signal (top panel) is mixed with the input RF signal (middle panel), generating down harmonics in baseband (bottom panel, light blue area).

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Cryogenic photovoltage nanoscopy in twisted multilayer graphene.

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Magic angle twisted bilayer graphene has been realized as a new platform to study strongly correlated quantum states of matter such as superconductors, correlated insulators and topological orders hosted by its native flat bands. Recent observations of anomalous Hall effect in MATBG revealed a non-zero Berry-curvature-induced orbital ferromagnetism and has been followed by extensive experimental and theoretical studies of its origin. Initial electronic transport studies demonstrated anomalous Hall effect at v=+3 electrons per moiré unit cell in the samples aligned with the adjacent insulating hBN layer, which breaks MATBG moiré inversion symmetry thus assisting in opening non-trivial insulating gaps with finite Chern numbers. Here we report on photovoltage scanning near-field imaging at cryogenic temperatures (10 K) of a MATBG device structurally aligned with hBN layer. We reveal a complex pattern of quasi-local photovotage fringes attributed to the second-order superlattice (supermoiré) potential modulated through the sample bulk. We assume that predominantly the photocurrent originates from PV generation mechanisms induced by the proximity of a hot spot formed on the metallic atomic force microscopy tip radiated by mid-infrared photons. Strikingly, we find a clear change of the PTE fringes' real-space orientation when the sample doping changes between the valence and conductance flat bands which may signal a nematic ordered state in a highly doping-dependent low-energy spectrum of MATBG. Our observation sheds a new light on the microscopic mechanisms of the inversion symmetry breaking on a mesoscale lengthscale.

High-performance monolayer MoS_2 field-effect transistors and photodetectors on SiO_2 gate dielectric passivated with cyclic olefin copolymer

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Trap states of the semiconductor/gate dielectric interface give rise to a pronounced subthreshold behavior in field-effect transistors (FETs) diminishing and masking intrinsic properties of 2D materials. To reduce the well-known detrimental effect of SiO_2 surface traps, this work spin-coated an ultrathin (≈5 nm) cyclic olefin copolymer (COC) layer onto the oxide and this hydrophobic layer acts as a surface passivator. The chemical resistance of COC allows to fabricate monolayer MoS_2 FETs on SiO_2 by standard cleanroom processes. This way, the interface trap density is lowered and stabilized almost fivefold, to around 5×10^{11} cm^{-2} eV^{-1}, which enables low-voltage FETs even on 300 nm thick SiO_2. In addition to this superior electrical performance, the photoresponsivity of the MoS_2 devices on passivated oxide is also enhanced by four orders of magnitude compared to nonpassivated MoS2 FETs. Under these conditions, negative photoconductivity and a photoresponsivity of 3×10^7 A W^{-1} is observed which is a new highest value for MoS_2. These findings indicate that the ultrathin COC passivation of the gate dielectric enables to probe exciting properties of the atomically thin 2D semiconductor, rather than interface trap dominated effects.

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Figures

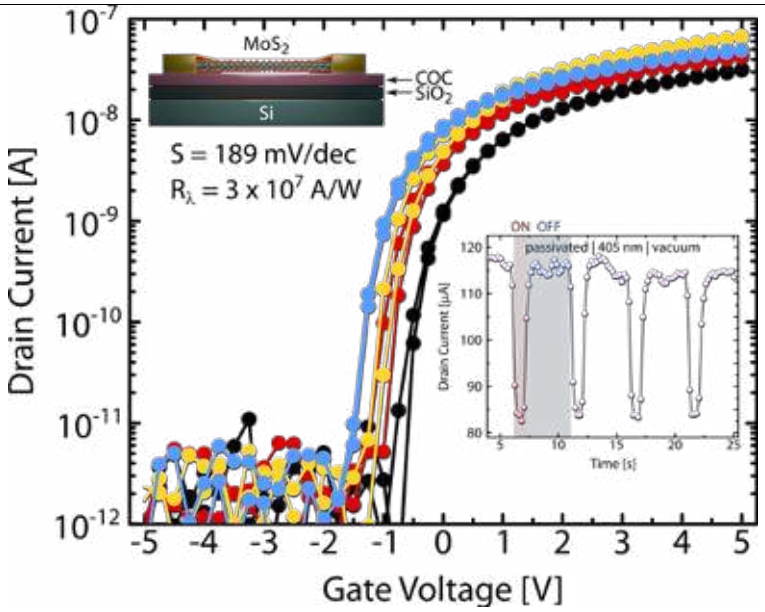

Figure 1: Performance of monolayer MoS2 based electronic and optoelectronic devices on SiO_2 gate dielectric passivated with cyclic olefin copolymer.

Highly nonlinear Mie-exciton-polaritons in monolayer semiconductors placed on WS_2 nanoantennas on a gold substrate

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Abstract: Subwavelength dielectric photonic structures exhibiting low absorption losses and a wealth of distinct Mie resonances in contrast to their plasmonic counterparts[1]. Layered transition metal dichalcogenides (TMDs) have excellent performance with a high refractive index, low losses below the bandgap, and straightforward compatibility with various substrates thanks to the van der Waals forces, thus providing a promising novel nanophotonic platform[2-4]. Here, we demonstrate that TMDs can also be used as a versatile platform for observation of the strong light-matter interaction. We observe strong coupling between a Mie resonance in 30 nm tall WS_2 nanoantennas attached to a gold substrate and excitons in a monolayer WSe_2 attached on top of the nanoantennas. Fig.1a shows an SEM image of an individual WS_2 nanoantenna. Fig. 1b shows the optical image of an array of WS_2 nanoantennas covered by a monolayer WSe_2. Fig.1c shows the dark-field scattering spectra before and after transferring monolayer WSe_2 collected for WS_2 nanoantennas with the radius increasing from 84 to 155 nm. Thanks to the high refractive index of WS_2 and additional effect of the gold substrate, Mie resonances with a bandwidth as narrow as ~90 meV are achieved. In Fig.1c and d we observe that a characteristic anti-crossing arises as the Mie mode is tuned in resonance with WSe_2 exciton, which signifies observation of the strong light-matter coupling. Crucially, our results show that the observed Rabi splitting for Mie-exciton-polaritons of ~85 meV is of the same order of magnitude as reported in plasmonic systems (~50 meV[5]), but is accompanied by lower absorption losses. The FDTD simulation of a WS_2 nanoantenna wrapped in a monolayer WSe_2 further supports our interpretation (Fig.1e). Furthermore, we observe strong nonlinearity of Mie-exciton-polariton with increasing optical power in comparison with the exciton in bare monolayer WSe_2. Our results highlight van der Waals nanophotonic structures as a versatile platform for the observation of strong light-matter coupling.

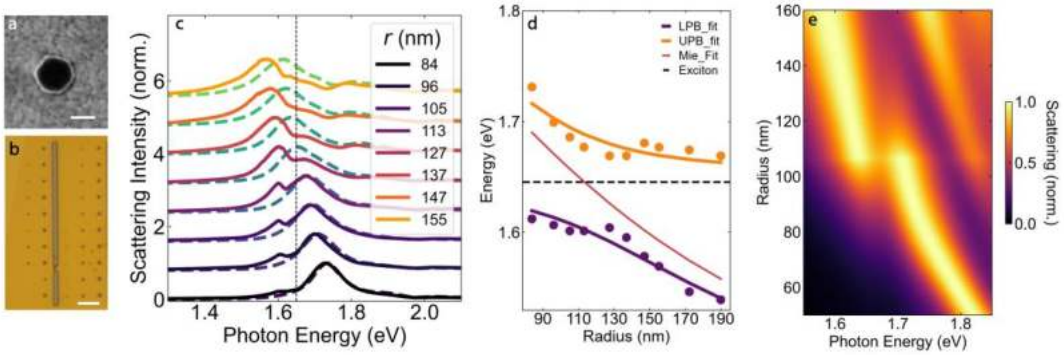

Figure 1. Mie-exciton-polariton achieved by van der Waals materials. a. SEM image of the nanoantenna. The scale bar is 200 nm. b. Optical image of bulk WS_2 nanoantenna covered by monolayer WSe_2 on a gold substrate. The white dashed line indicates the boundary of monolayer region, where the left of line is pure gold. The scale bar is 5 μm. c. Dark-field scattering spectra of WS_2 nanoantenna covered with monolayer WSe_2 (solid curve), with the comparison with nanoantennas before WSe_2 transfer (dashed curve). The vertical dashed line indicates the monolayer exciton position. d. Symbols show peak positions from Fig.c. The solid lines show the fitting obtained using the coupled-oscillator model providing the Rabi splitting of 85 meV. e. Simulated scattering cross section of nanoantenna wrapped with monolayer WSe_2 with the nanoantenna radius.

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Mixed-dimensional moiré tuning of transport properties in graphite thin films

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Moiré systems have attracted attention as a host of correlated states and non-trivial topology, as in the case of magic-angle twisted bilayer graphene. Recently, it has been shown that these moiré tuning capabilities persist in few layer graphene systems, such as in the twisted tri/tetra/penta-layer graphene, twisted mono-bilayer graphene (t1+2), and twisted double bilayer graphene (t2+2). However, it remains unclear if significant moiré modification will occur when each layer of the twisted structure is composed of more than a graphene mono- or bilayer, since additional low energy bands must be hybridized as the layer number increases towards the bulk graphitic limit. In this work, we present transport studies of graphitic thin films with a single rotated interface, primarily focusing on the case of t1+Z systems where Z≥6. We find that such systems constitute a new class of mixed-dimensional moiré materials, where localized moiré surface states interact with and modify the bulk graphitic states, even up to around Z=40 layers of graphite. Our results are generally applicable to other layered semimetals and establish mixed-dimensional moiré systems as an exciting path for future research.

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Figures

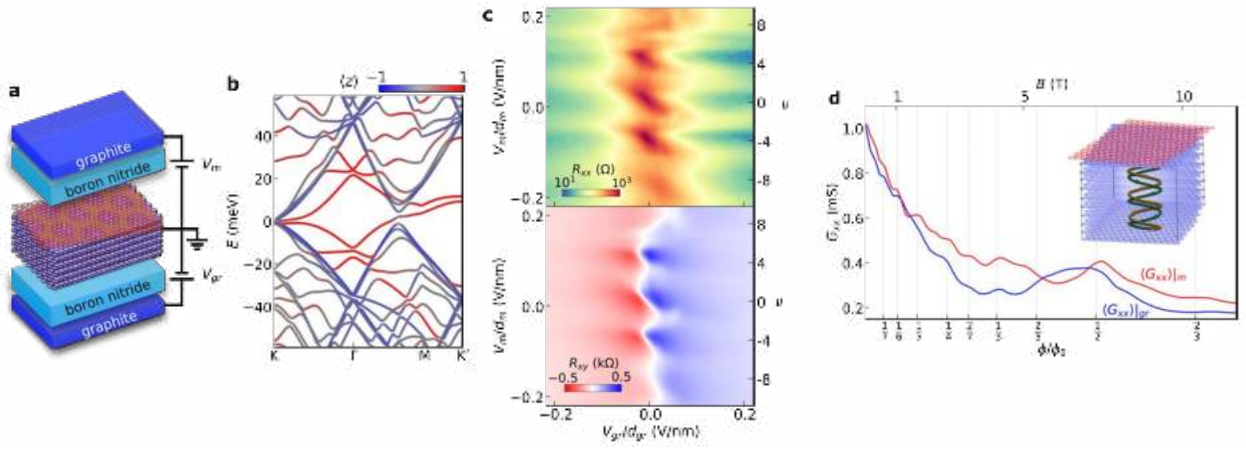

Figure 1: a) Schematic showing our t1+Z devices, V_m and V_{gr} denote gates that are closest to the moiré interface and outer graphite surface, respectively. **b)** Band structure for a t1+10 device, with states denoted by the expectation value along the z-axis, with red (blue) corresponding to the moire interface (outer graphite surface). **c)** R_{xx} and R_{xy} maps taken at B=0.5 T for a t1+10 device, with filling factor denoted on the right-hand axis. **d)** Conductance as a function of magnetic field sweeping both the graphite and moiré gates. Select values of rational flux fillings indicated, where peaks can be observed as Brown-Zak oscillations. (inset) Schematic illustrating the high field coupling of the 2D moiré states with the bulk 3D graphite states.

MXenes application in microelectronics (MXetronics)

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MXenes are a family of emerging two-dimensional (2D) materials composed of atomic-thin early transition metal carbides and nitrides. Their unique properties, such as excellent conductivity, surface plasmons, hydrophilicity, tunable surface groups and work function (theoretically in between 2 and 8 eV), and good water dispersibility without surfactant. Such abundant properties together with the large compositional variety make them attracitve to design MXene-based microelectronics with novel functionalities. Our research efforts recently are heavily focused on the application of MXenes and their derived materials in the field of integrated transistor circuits. We demonstrated that $Ti_3C_2T_x$ MXene films and V_2CT_x-derived Metal organic framework (V_2CT_x-MOF) films can be reliably processed through the industry compatible nanofabrication patterning process. Using patterned $Ti_3C_2T_x$ MXene films as source/drain/gate metal electrode, we were able to fabricate the lab-made ultrathin $Ti_3C_2T_x$ –MoS_2 circuits with a high yield and a low performance variability. We have also fabricated the first 2D MOF-MoS_2 electron-double-layer transistor (EDLTs) using patterned V_2CT_x-MOF films as the solid electrolyte. Furthermore, when replacing the traditional metal Au/Ni with a $Ti_3C_2T_x$ MXene film as the schotcky gate electrode to control the 2D electron gas (2DEG) in the GaN high-electron-mobility transistor (HEMT), we observed an incredible performance enhancement. The remarkable results obtained so far using MXenes in microelectronics suggest a bright future for MXetronics.

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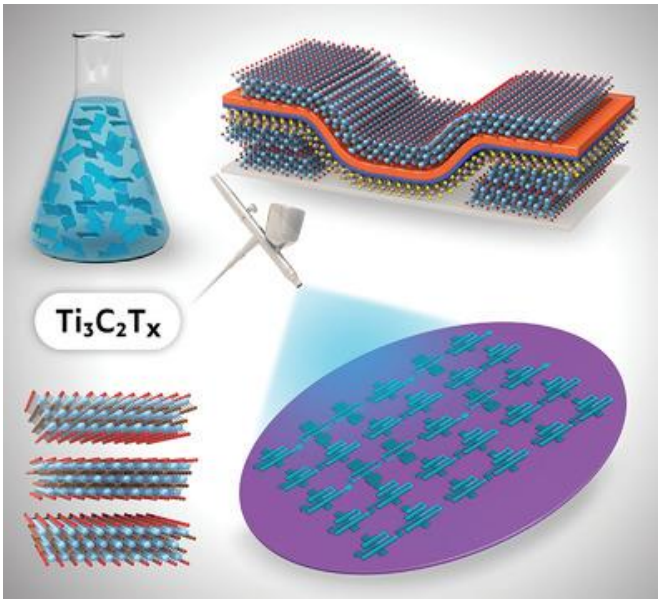

Figure 1: $Ti_3C_2T_x$ MXene in wafer-scale integrated 2D electronics

Characterization and Manipulation of Intervalley Scattering Induced by an Individual Monovacancy in Graphene

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Abstract (Century Gothic 11)

Intervalley scattering involves microscopic processes that electrons are scattered by atomic-scale defects on the nanoscale. Although central to our understanding of electronic properties of materials, direct characterization and manipulation of range and strength of the intervalley scattering induced by an individual atomic defect have so far been elusive. Using scanning tunneling microscope, we visualize and control the electronic properties especially the intervalley scattering from an individual monovacancy in graphene [1-3]. By directly imaging the affected range of monovacancy-induced intervalley scattering, we demonstrate that it is inversely proportional to the energy; i.e., it is proportional to the wavelength of massless Dirac fermions. A giant electron-hole asymmetry of the intervalley scattering is observed because the monovacancy is charged. By further charging the monovacancy, the bended electronic potential around the monovacancy softens the scattering potential, which, consequently, suppresses the intervalley scattering of the monovacancy [4].

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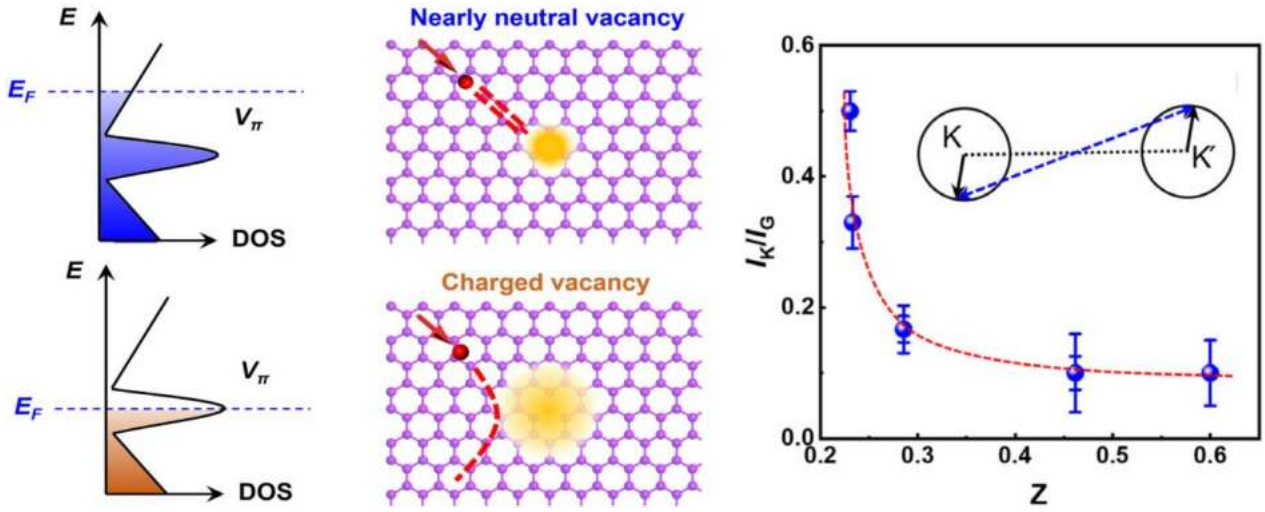

Figure 1: The suppression of the intervalley scattering induced by the charged monovacancy in graphene.

Better than Hall? Increasing sensitivity of graphene magnetic sensor based on extraordinary magnetoresistance with peculiar effects

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Since its discovery, extraordinary magnetoresistance (EMR) has drawn a lot of attention to applications such as hard drive read heads and magnetic sensors. Recently, an extremely large magnetoresistance, $MR=(R(B)-R_0)/R_0 \sim 10^7\%$ are achieved in devices of graphene encapsulated in hexagonal boron nitride with a central metal shunt [1], exceeding that achieved in state-of-the-art semiconductor devices by one order of magnitude [2]. Encapsulated graphene is the most promising material for EMR devices among all reported to date [3]. We find the room-temperature sensitivity dR/dB of encapsulated graphene EMR devices to reach 102.2 kΩ/T at ~-0.2T [5], more than 3 times and 60 times larger than the previous records achieved in encapsulated graphene [1] and unencapsulated graphene [4], respectively. This sensitivity is comparable to recently reported in encapsulated graphene Hall sensor, however measured at 4.2K [6]. This suggests that graphene EMR sensors could outperform state-of-the-art sensors in room-temperature conditions, including graphene Hall sensors. We highlight that the room-temperature sensitivity in our EMR devices can reach 13 kΩ/T at B=-6.7 mT. We also demonstrate that the sensitivity can be enhanced by reducing the charge carrier density, lowering the temperature and increasing the charge carrier mobility. EMR devices also exhibit magnetic focusing, magnetic commensurability effects, weak localization and weak antilocalization [5], which may be employed to tune or improve the magnetic field sensing performance further. The appearance of these effects associated with cyclotron orbits, quantum coherence and device geometry suggests the EMR geometry as an interesting alternative to the Hall geometry for fundamental physics studies.

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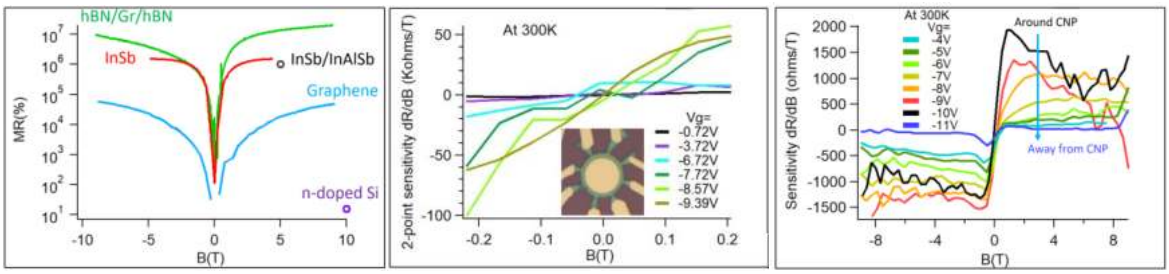

Figure 1: (a) The best magnetoresistance (MR) values reached in EMR devices made by different materials reported in literatures. (b) The largest room-temperature sensitivity dR/dB reached in encapsulated graphene EMR devices. (c) The sensitivity dR/dB of encapsulated graphene EMR devices increases with decreasing charge density (closer to charge neutral point (CNP)).

Quantum spin Hall states and topological phase transition in germanene

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The realization of the topological field-effect transistor requires an electric field-induced transition from a topological state with dissipationless conductive channels ('On') to a trivial insulator state ('Off'). Monoelemental and buckled quantum spin Hall insulators with large topological band gaps are ideal candidates to investigate topological phase transitions[1-3]. We provide compelling experimental evidence that low-buckled epitaxial germanene [4] is a quantum spin Hall insulator with a bulk gap and robust metallic edge states. The low-buckled structure of germanene allows for topological phase transitions to take place. Upon the application of a critical transversal electric field, the topological gap closes and germanene becomes a Dirac semimetal. Increasing the electric field further results in the opening of a trivial gap and the disappearance of the edge states. This reversible electric field-induced switching of the topological state, shown in Figure 1, and the sizeable gap make germanene suitable for room temperature topological field-effect transistors, which could revolutionize low-energy electronics and spintronics.

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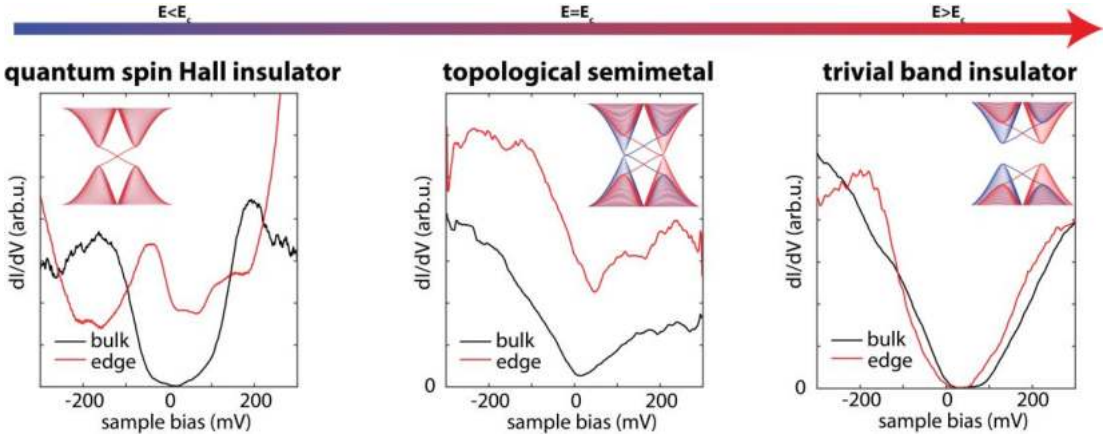

Figure 1. Scanning tunneling spectroscopy (dI(V)/dV) recorded at the bulk (black) and step edge (red) of germanene for increasing electric fields (from left to right). Left: dI(V)/dV spectra recorded at the bulk and the step edge of germanene for an electric field below the critical field to close the topological gap of germanene (E_c=1.95 V/nm). Inset: the topological insulator band structure. Middle: dI(V)/dV spectra recorded at the bulk and the step edge of germanene for an electric field of about 1.95 V/nm, showing the V-shaped density of states of germanene for this field. Inset: the topological semimetal band structure. Right: dI(V)/dV spectra recorded at the bulk and the step edge of germanene for an electric field of about 2.32 V/nm, showing the trivial gap in germanene. Inset: the trivial band insulator band structure.

Asymmetric magnetic proximity interactions in van der Waals heterostructures

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Magnetic proximity interactions (MPIs) between atomically thin semiconductors and two-dimensional magnets provide a means to manipulate spin and valley degrees of freedom in non-magnetic monolayers, without using applied magnetic fields. In such van der Waals heterostructures, MPIs originate in the nanometre-scale coupling between spin-dependent electronic wavefunctions in the two materials, and typically their overall effect is regarded as an effective magnetic field acting on the semiconductor monolayer. Here we demonstrate that MPIs in $MoSe_2/CrBr_3$ van der Waals heterostructures can in fact be markedly asymmetric [1]. Valley-resolved reflection spectroscopy reveals strikingly different energy shifts in the K and K´ valleys of the $MoSe_2$ due to ferromagnetism in the $CrBr_3$ layer. Density functional calculations indicate that valley-asymmetric MPIs depend sensitively on the spin-dependent hybridization of overlapping bands and as such are likely a general feature of hybrid van der Waals structures.

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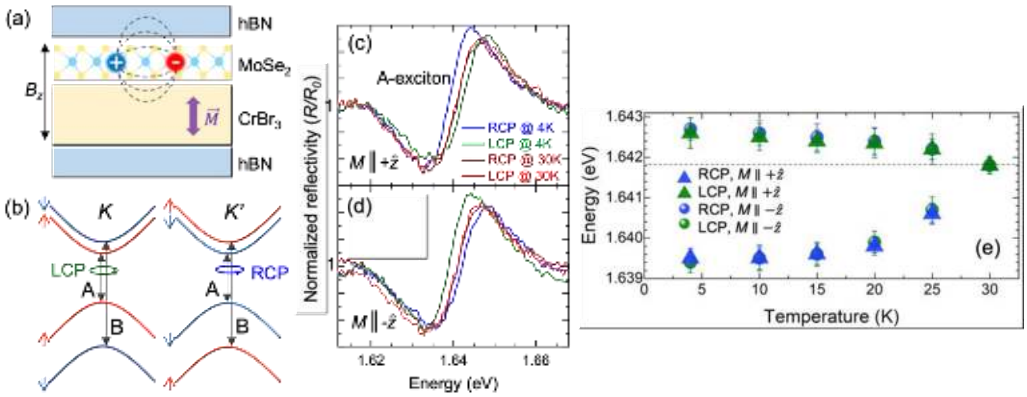

Figure 1: (a) Layer schematic of hBN/$MoSe_2$/$CrBr_3$/hBN heterostructure. The red and blue spheres depict and electron and hole pair in the $MoSe_2$, forming an exciton. (b) Band diagram depicting the relevant optical transitions of A and B excitons in $MoSe_2$. (c, d) Comparison of the MPI-induced shift of the $MoSe_2$ A exciton in the K and K´ valleys at T=4 K, as measured by the normalized reflection spectra, R/R_0, of left circularly polarized (LCP) and right circularly polarized (RCP) light (green and blue traces, respectively). The $CrBr_3$ magnetization M is oriented along $+\hat{z}$ or $-\hat{z}$. Red and orange traces show reference spectra acquired at 30 K (above T_C), where the $CrBr_3$ is unmagnetized. (e) Temperature dependence of the MPI-induced valley shifts of the A exciton, for both circular polarizations and for both $\pm\hat{z}$ $CrBr_3$ magnetizations. The horizontal line indicates the exciton resonance energy at 30 K when the $CrBr_3$ is not magnetic.

Twisted bilayer graphene in magnetic fields

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The band structure of graphene bilayers strictly depends on the twisting angle between layers. Bernal stacking (AB) generates parabolic semimetallic bands, while AA stacking keeps the Dirac linear dispersion and removes degeneracy. At very small so-called "magic angles", the large size of AA stacked regions explains the appearance of almost flat bands, which correspond to states localized in the AA regions as reported in [1].
Here, based on the same tight-binding model and with the inclusion of an orthogonal magnetic field [2], we observe the formation of dispersive Landau levels, which can be localized (in the AA or AB regions, for example) or extended depending on their velocity. These states are expected to be experimentally visible by scanning tunnelling microscopy and to determine the magnetotransport properties.
In-plane magnetic fields still affect the band structure due to the layer-dependent induced Dirac point shift. In an AA bilayer, where the Dirac points perfectly coincide, such an effect opens a small energy gap. These results smooth the path toward further investigations beyond the continuum model [3].

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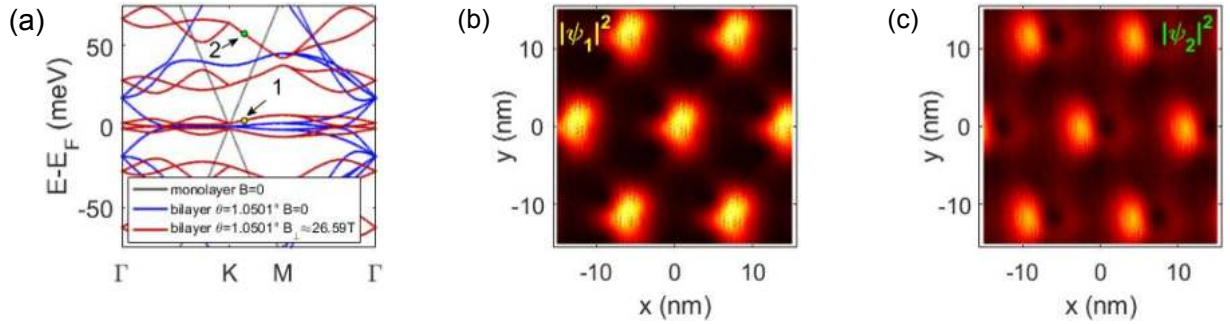

Figure 1: (a) Energy bands of monolayer and twisted graphene bilayer with a magic angle θ=1.0501° for orthogonal magnetic fields B=0 and B≈26.59 T. The almost flat low-energy bands for B=0 are typical of magic angles and confined in the AA regions [1]. (b) Squared eigenfunction at point 1 for B≈26.59 T. This state is mainly localized (lighter colour) in AA regions. (c) Same as (b) at the higher energy point 2. This state is more delocalized (with lower density, dark colour, in the AA regions).

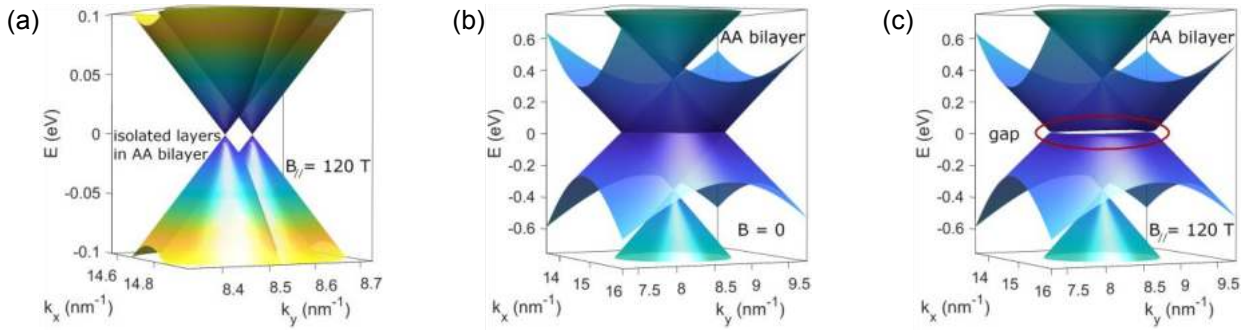

Figure 2: Energy bands around the Dirac point for an AA bilayer graphene and in the case of (a) isolated layers with in-plane magnetic field B=120 T, the Dirac point shift is proportional and orthogonal to the magnetic field; (b) coupled layers with B=0; (c) coupled layers with B=120 T, we observe the opening of a *k*-dependent gap varying up to about 33 meV and oriented in the reciprocal space according to the magnetic field direction.

Nanophotonics with van der Waals magnets

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Abstract

Layered magnetic materials have recently attracted intensive studies of their unique properties opening access to novel physics of magnetism in the low dimensional limit [1]. An attractive prospect in magnetism is achieving control of magnetic properties via coupling to light [2, 3]. To facilitate such control, photonic structures supporting resonant optical modes may be an attractive solution. Here, we fabricate nanoantennas made from thin films of high-refractive-index antiferromagnetic semiconductors $NiPS_3$, $MnPSe_3$ and $MnPS_3$ [3] and demonstrate their Mie resonances tunable by the nanoantenna size. By utilizing van der Waals forces typical for layered materials, we were able to fabricate such nanoantennas on gold substrates, where, due to high reflectivity of gold, we observed notable narrowing of Mie resonances. Nanoantennas on gold substrates were fabricated by mechanical exfoliation of thin (20-400 nm) films of $NiPS_3$, $MnPSe_3$ and $MnPS_3$, followed by electron-beam lithography, and reactive ion etching. Figure 1 shows the bright field optical image (a), the dark-field image (b), and SEM (c) of a $NiPS_3$ nanoantenna array. We observed the tuning Mie resonance by changing the antenna radius [Figure 1 (d)]. Our fabrication approach provides a way for enhancing light-matter interaction in a wide range of materials classes including magnets, dielectrics and semiconductors, as well as potentially superconductors and metals existing in the layered crystal form.

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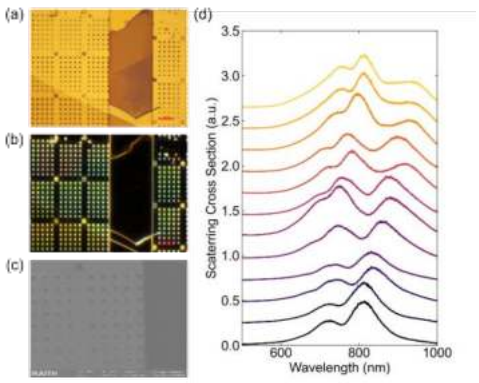

Figure 1: $NiPS_3$ magnetic nanoantenna on gold. (a) Bright field and (b) dark field optical images. (c) SEM image. (d) Dark-field scattering spectra.

Electron correlation and topology in rhombohedral graphene

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Rhombohedral stacked multilayer graphene is an ideal platform to search for correlated electron phenomena, due to its pair of flat bands touching at zero energy and further tunability by an electric field. Furthermore, its valley-dependent Berry phase at zero energy points to possible topological states when the pseudospin symmetry is broken by electron correlation. In this talk, I will first show our efforts on the optical spectroscopy study of trilayer graphene/hBN moire superlattice. We observed optical signatures of flat moire band formation and Mott insulator due to band splitting at half-filling. Then I will talk about DC transport measurements of pentalayer graphene without a moire with the hBN substrates. We observed a plethora of correlated and topological states including a ferro-valleytronic half-metal, a correlated insulating state and Chern insulator states. Our results point to a new direction of exploring electron correlation and topology in natural 2D crystals where the layer number plays a critical role

Observation of topological valley current in non-encapsulated hBN/bilayer graphene heterostructure

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The valley Hall effect (VHE) is a phenomenon in which the carriers in a material move to the opposite edges depending on their valley index. The VHE requires a non-zero Berry curvature which is present only in systems where either time-reversal symmetry or spatial inversion symmetry is broken. It is well known that aligning graphene on hexagonal boron nitride (hBN) with a near-zero twist angle breaks the symmetry of the system. For both single-layer and bilayer graphene, this method has been used to break the symmetry of the system and observe the VHE. The sub-lattice symmetry is broken for single-layer graphene, whereas for bilayer graphene, the layer symmetry is broken. In the case of single-layer graphene, the effect of configuration (i.e., whether the hBN is present on both sides or one side), as well as the orientation of both the top and bottom hBN on the VHE is studied in detail [1-2]. However, for bilayer graphene, although it has been shown theoretically that the configuration, as well as the orientation of the hBN, has an immense impact on the asymmetry of the system [3], experimental studies in this aspect are lacking. K. Endo et al. have studied encapsulated bilayer graphene where the top hBN is aligned with the bilayer graphene and observed VHE [4]. In this work, we conducted valleytronics study on a non-encapsulated hBN/bilayer graphene heterostructure where hBN is present only at the top. The hBN is aligned with the bilayer graphene to break the symmetry of the system. We observed a strong VHE signal through non-local resistance (R_{NL}) measurement (Fig. 1). We also demonstrate that the valley Hall effect can be tuned by applying an out-of-plane electric field, which changes the band gap and the Berry curvature of the bilayer graphene system. We have also performed ab initio calculations to substantiate the experimental observations [5].

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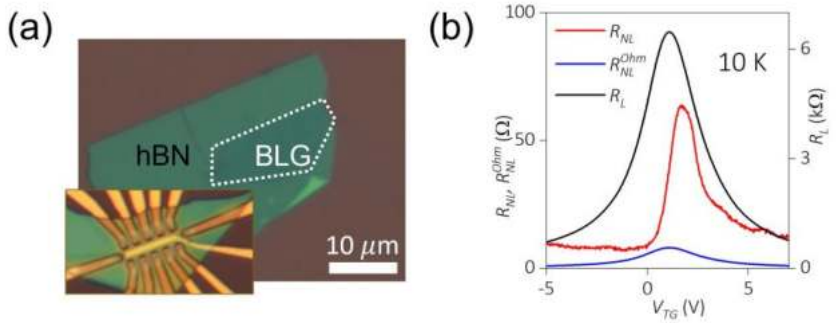

Figure 1: (a) hBN/bilayer graphene heterostructure with the edges aligned. Inset: Optical image of the final device. (b) Measured local (R_L) and non-local (R_{NL}) resistance for the heterostructure.

Interaction induced terahertz photocurrents in twisted bilayer graphene

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Magic-angle twisted bilayer graphene hosts a highly interacting electron system that exhibits phenomena bridging topology and strong correlations [1]. Polarization resolved photocurrent measurements offer a direct access to the geometry of electron wave functions including berry curvature and the quantum metric [2],[3]. In this talk we will present terahertz photocurrent measurements in twisted bilayer graphene close to the magic-angle. Our measurements reveal several fragile states not detectable by quantum transport that originate from hidden symmetries intrinsic to twisted bilayer graphene. Amongst them, we observe signatures of a gapped state at charge neutrality and Hartree interaction induced band renormalizations manifesting in the photoresponse. Our measurements demonstrate how terahertz photocurrent is a powerful tool for probing interacting electrons and their quantum geometry in flat band systems.

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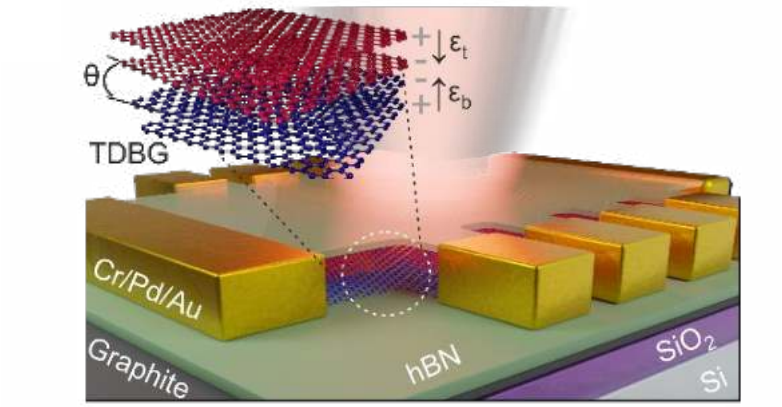

Figure 1: Schematic of photocurrent experiments in twisted graphene heterostructures

3D twistronics of mixing moiré-surface and bulk states in graphite

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Van der Waals twistronics enables designing two-dimensional (2D) electronic states by using moiré superlattices. This approach resulted in many new phenomena, including, among other, strong correlations and superconductivity in magic-angle twisted bilayer graphene[1,2], charge ordering and Wigner crystallisation in transition metal chalcogenide moiré structures[3-6], Hofstadter's butterfly spectra and Brown-Zak quantum oscillations in graphene superlattices[7-9]. In this work we show that both surface and bulk electronic states in three-dimensional (3D) graphite can be tuned by a superlattice potential occurring at the interface with crystallographically aligned hexagonal boron nitride[10]. Such alignment results in numerous Lifshitz transitions and Brown-Zak oscillations arising from near-surface states whereas, in high magnetic fields, fractal states of Hofstadter's butterfly penetrate deep into the bulk of graphite. Our work provides novel control of 3D spectra using the approach of 2D twistronics.

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Electron-Phonon Coupling in Magic-Angle Twisted-Bilayer Graphene

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Twisted graphene systems are outstanding platforms to explore correlated electron physics and superconductivity with exceptional tunability [1]. The importance of phonons in these strong correlation phenomena in twisted bilayer graphene (TBLG) at the so-called magic-angle is therefore very necessary to be clarified [2-4]. In this talk, we will present our recent study [5] using gate-dependent Raman spectroscopy and atomistic modeling to investigate the electron-phonon coupling through the C-C stretching mode (G band) linewidth in magic-angle TBLG. Our study has been performed particularly for three TBLGs at twist angles θ = 0° (Bernal stacking), ~1.1° (magic-angle) and ~7° (large angle). Overall, the value of the G band linewidth in magic-angle TBLG is shown to be much larger when compared to that of the other samples, in qualitative agreement with our calculations. In addition, the obtained results also show that a broad and p/n-asymmetric doping behavior is observed at the magic-angle, in clear contrast to the behavior observed at other angles. Simulations reproduce these experimental observations, revealing how the unique electronic structure of magic-angle TBLGs impacts the electron-phonon coupling, reflected by its effects on the G band linewidth. Our study thus points to a relationship between electron-phonon coupling and the strongly correlated phenomena in the magic-angle TBLG.

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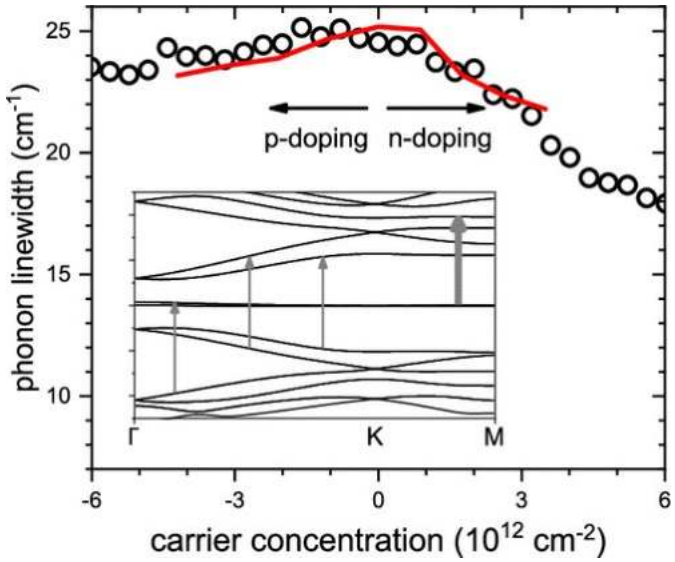

Figure 1: Gate-dependent G-band Full-Width-Half-Maximum in the magic-angle TBLG

Artificial Graphene Spin Polarized Electrode for Magnetic Tunnel Junctions

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2D materials offer the ability to expose their electronic structure to manipulations by a proximity effect. This could be harnessed to craft properties of 2D interfaces and van der Waals heterostructures in devices and quantum materials.[1] We explore the possibility to create an artificial spin polarized electrode from graphene through proximity interaction with a ferromagnetic insulator to be used in a magnetic tunnel junction (MTJ). Ferromagnetic insulator/graphene artificial electrodes were fabricated and integrated in MTJs based on spin analyzers.[2] Evidence of the emergence of spin polarization in proximitized graphene layers was observed through the occurrence of tunnel magnetoresistance. We deduced a spin dependent splitting of graphene's Dirac band structure (~15 meV) induced by the proximity effect, potentially leading to full spin polarization and opening the way to gating.[3] The extracted spin signals illustrate the potential of 2D quantum materials based on proximity effects to craft spintronics functionalities, from vertical MTJs memory cells to logic circuits.

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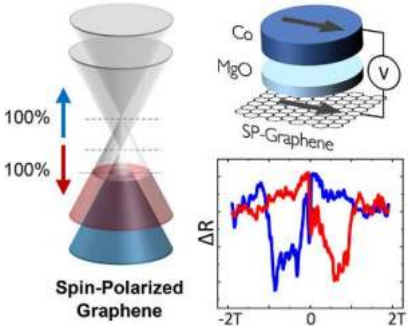

Figure 1: (Left) Exchange-induced spin splitting of the graphene Dirac cones by proximity effect. (Upper right) Device concept of magnetic tunnel junction with magnetized graphene as spin polarizer. (Lower right) Typical spin signal recorder on one of our devices

Hyper-magic manifold of Majorana flat bands in twisted Kitaev bilayers

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The booming interest in quantum technologies and neuromorphic computing is driving the search of exotic phases of matter for futuristic applications. The recent discovery of van der Waals (VdW) materials (e.g., $RuCl_3$, 1T-TaS_2) that approximate the intriguing physics of quantum spin liquids [1] has attracted a large deal of interest. The quantum spin excitations (spinons) of such materials are predicted to behave as anyons [2] and intense research efforts have been put in proving their existence to harness their power for futuristic applications. Simultaneously, the capability of controlling the twist angle between stacked VdW materials and producing moiré superlattices (MSLs) has opened the possibility of engineering quantum phases with no counterpart in Nature [3]. Compared to conventional artificial systems, MSLs emerge at much smaller (nanometre) scales. Thus, emergent artificial phases can retain their quantum nature while being still relatively easy to be probed experimentally. Here we show that twisted bilayers of (Kitaev) quantum spin liquids, can exhibit unique phases as a function of the interlayer coupling [4]. By constructing a mean-field approximation in terms of solutions of commensurate bilayers, we show that the band structure of deconfined spinons is greatly modified and exhibits a hyper-magic manifold. A series of nearly perfectly flat bands appear at energies above the lowest gap, exhibiting a very large local (spinon) density of states that could potentially be probed in STM experiments. Intriguingly, flat-band eigenstates exhibit a localization akin to wavefunctions of Kagome lattices. These results prove that a wealth of novel phases of matter are hiding in twisted bilayers of the relatively less studied strongly-correlated VdW materials.

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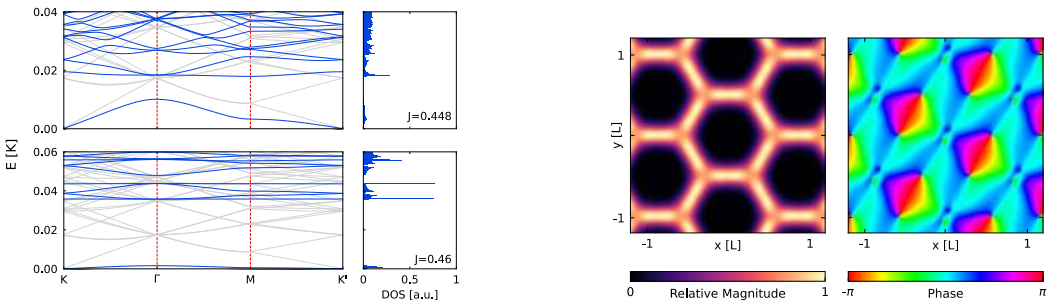

Figure 1: (left) the band structure of deconfined spinons exhibiting an emergent hyper-magic manifold of flat bands. The density of states (DOS) of spin excitations becomes extremely large at the corresponding energies, thus offering a way of identifying these elusive quasiparticles in STM. (right) Flat-band eigenstates exhibit a Kagome-like pattern which could be detected in experiments.

Electron correlation and complex magnetism in 2D Fe_nGeTe_2 (n=3-5) magnets

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Abstract
The Fe_nGeTe_2 (n=3, 4, 5) family of two-dimensional magnets has emerged as a potential candidate for spintronic applications. These magnets exhibit high temperature ferromagnetism, complex temperature dependent magnetization, structural reconstructions, skyrmionic features etc. Electron correlation, isotropic and anisotropic exchange interactions and magnetic anisotropy have enriched the overall magnetic properties, which require thorough investigations for a detailed understanding. Here, we present a systematic study of this family using first-principles electronic structure calculations with different flavors, viz. (i) standard density functional theory (DFT), (ii) static electron correlation (DFT+U) and (iii) dynamical electron correlation with dynamical mean field theory (DFT+DMFT). Moreover, complex structural aspects regarding site occupancy of Fe in Fe_5GeTe_2 [1] have been considered in connection to scanning tunnelling microscopy images. A thorough analysis of critical temperatures and spectral properties reveal [2] that DFT+DMFT is the most accurate method to correctly reproduce the experimental data on temperature dependent magnetization, valence band and angle-resolved photoemission spectroscopies respectively. Moreover, Monte Carlo simulations show peculiar magnetic structures at low temperature with site-selective canted Fe moments. The inaccurate values of structural parameters, magnetic moments and exchange interactions obtained from DFT+U make this method inapplicable for the FGT family. In summary, our study provides a comprehensive view of the electronic structure and magnetism of this important class of 2D magnets.

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Discovering emergent correlated states and quantum phase transitions in a moiré superlattice

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Abstract:
New phase of matter usually emerges when a given symmetry breaks spontaneously, which can involve charge, spin, and valley degree of freedoms. By twisting graphene multilayer to form a moiré superlattice, it leads to moiré flat band where various correlated states are developed. Here, we observe valley polarized correlated insulator [1] at half fillings in twisted double bilayer graphene (TDBG), as shown schematically in Fig.1a, resulted from an isospin competition between spin and valley flavours. The valley polarized correlated insulators are found hosting anomalous quantum oscillations (QOs) [2] with a period of 1/B (Fig.1b), and the periodicity strongly displacement field dependent: the carrier density extracted from the 1/B periodicity decreases almost linearly with D from -0.7 to -1.1V/nm. In addition to the discoveries in the valley polarization, we have observed first order phase transitions and ferromagnetism [3] in the spin polarized correlated insulator in TDBG, and have also observed many other first-order phase transitions with hysteresis when valley degree of freedom competes in finite magnetic field. Our study suggests that TDBG is an excellent platform to discover exotic phases where correlation and topology are at play.

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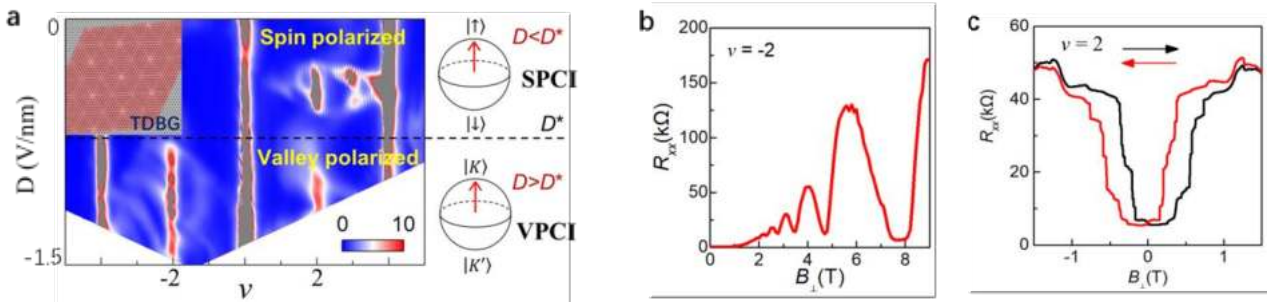

Figure 1: (**a**) A colour mapping of resistance as a function of moiré band filling (v) and displacement field (D) in TDBG, and SPCI and VPCI denote spin polarized and valley polarized correlated insulator, respectively. (**b**) Unconventional quantum oscillations in the VPCI at v=-2 (D>D*). (**c**) Evidences of the first-order phase transitions and ferromagnetism in the SPCI at v=2 (D<D*).

Theoretical and experimental characterization of sp-, sp^2- carbon 2D networks obtained via on-surface synthesis

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Network formed by sp- and sp2-hybridized carbon atoms with high degrees of π conjugation, namely graphynes and graphdiynes, are stimulating intense research efforts for their potential use in nanoelectronics, catalysis and photo-conversion, due to their high charge carrier mobility and tunable band gaps and band structures [1, 2,3]. The significant advancement in the synthesis of these materials through on-surface assembly of molecular precursors opens the way to the design of novel carbon networks, displaying different geometries and physical properties. Nevertheless, the fabrication of extended ordered 2D monolayers, their characterization and the definition of protocols for their manipulations represent challenging steps in view of their applications. We present a multidisciplinary characterization of sp-, sp^2- and hybrid sp- sp^2 2D systems obtained by on-surface synthesis of brominated molecular precursors on Au(111) surface. By combining *ab initio* calculations based on Density Functional Theory (DFT) and different experimental techniques, such as Scanning Tunneling Spectroscopy and Microscopy (STS and STM) and Raman spectroscopy we give a thorough description of nanoscale linear structures and 2D materials, as carbyne and graphdiyne, and of their interaction with the underlying metal substrate [4-8]. In particular we analyze the evolution of the structural, electronic and vibrational properties during the different stages of the formation, passing from the as deposited metallorganic network to pure sp-/ sp^2- nanostructures during annealing, and we show the effect of the substrate coupling on their semiconducting properties.

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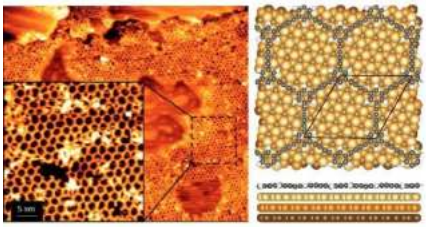

Figure 1: 2D sp-sp2 carbon network on Au(111). STM and theory model

Tungsten Oxide Mediated Quasi - van der Waals Epitaxy of WS_2 on Sapphire

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Abstract
Today's state-of-the-art semiconductor technology relies heavily on conventional epitaxy for controlling thin films and nanostructures at the atomic level, which are the building blocks for nanoelectronics, optoelectronics, sensors, etc. Four decades ago, the terms "van der Waals" (vdW) and "quasi-vdW (Q-vdW) epitaxy"[1] were coined to explain the oriented growth of vdW layers on 2D and 3D substrates, respectively. In this study, WS_2 is grown by sequentially exposing metal and chalcogen precursors in a metal-organic chemical vapor deposition (MOCVD) system[2], introducing a metal-seeding stage before growth. The ability to control the delivery of the precursor made it possible to study the formation of a continuous and apparently ordered WO_3 mono-or few-layer at the surface of a c-plane sapphire. Such an interfacial layer is shown to strongly influence the subsequent quasi-vdW epitaxial growth on sapphire: This represents an important breakthrough in this field since a new epitaxial growth mechanism is elucidated[3]. A rational design of epitaxial growth on different materials systems may be enabled by this work.

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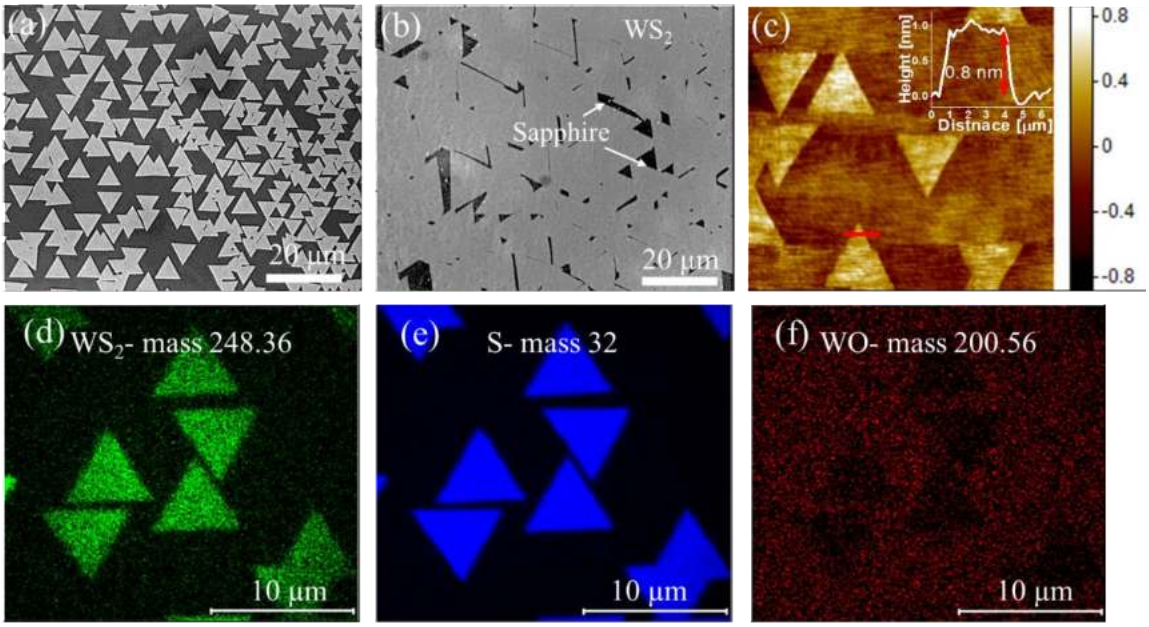

Figure 1: WS_2 epitaxial growth on sapphire (a-b)optical image; (c)AFM image;(d-f) TOF-SIMS analysis showing the domain and the WO_x interface layer between WS_2 to the sapphire.

Electrodeposition of TMDC 2D-materials on Graphene

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The development of scalable techniques to make two-dimensional transition metal dichalcogenides (2D-TMDC) material heterostructures is a major obstacle that needs to be overcome before these materials can be implemented in device technologies. Electrodeposition is an industrially compatible deposition technique that offers unique advantages in scaling 2D heterostructures. In this work, we demonstrate both the ability of electrodeposition the grow atomic layers of MoS_2 and WS_2 on patterned graphene electrodes as well as lateral growth of said 2D materials over an insulator resulting in promising electrical characteristics for photodetectors and transistors. This paves the way towards future possibilities such as electrodepositing different TMDCs to form lateral heterostructures of 2D materials and graphene, such as creating novel p-n-p or Schottky junction in a single electrodeposition experiment [1-4].

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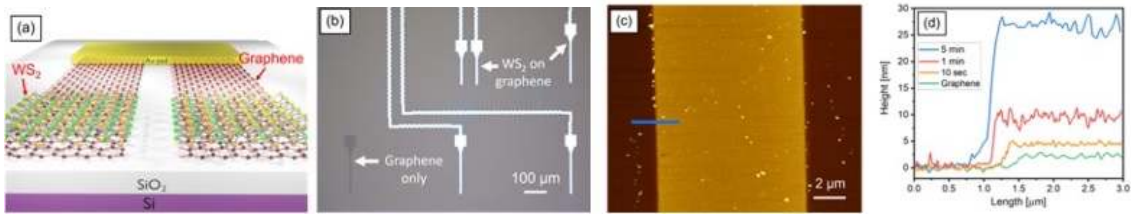

Figure 1: (a) A schematic illustration of WS_2 deposited on patterned graphene that is biased through Au contact pad. (b) An optical microscope image. (c) AFM topography images of WS_2 deposits on graphene for 1 minutes. (d) Line profiles showing total step height.

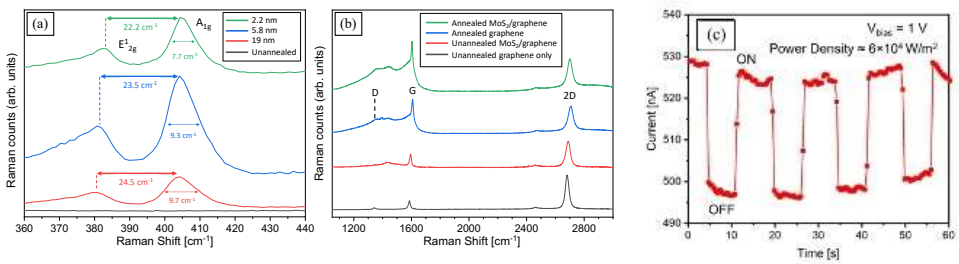

Figure 2: a) Raman spectra of MoS_2 with the separation between the A_{1g} and E^1_{2g} peaks reducing with deposition thickness. (b) Raman spectra of graphene showing the 2D peaks remain after annealing. (c) photo-illumination cycles showing induced photocurrent with a switching laser source

Crystal Phase Engineering of Silicene by Sn-modified Ag(111)

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The synthesis of silicene by direct growth on silver is characterized by the formation of multiple phases and domains, posing severe constraints on the spatial charge conduction towards a technological transfer of silicene to electronic transport devices. Here we engineer the silicene/silver interface by two schemes, namely, either through decoration by Sn atoms, forming an Ag_2Sn surface alloy, or by forming a silicene-stanene heterostructure with stanene buffering the substrate [1]. Whereas in both cases Raman spectra confirm the typical features as expected from silicene, by electron diffraction we observe that a very well-ordered single-phase 4×4 monolayer silicene is stabilized by the decorated surface, while the buffered interface exhibits a sharp √3×√3 phase at all silicon coverages. Both interfaces also stabilize the ordered growth of a √3×√3 phase in the multilayer range, featuring a single rotational domain. Theoretical ab initio models are used to investigate low-buckled silicene phases (4×4 and a competing √13×√13 one) and various √3×√3 structures, supporting the experimental findings. This study provides new and promising technology routes to manipulate the silicene structure by controlled phase selection and single-crystal silicene growth on a wafer-scale.

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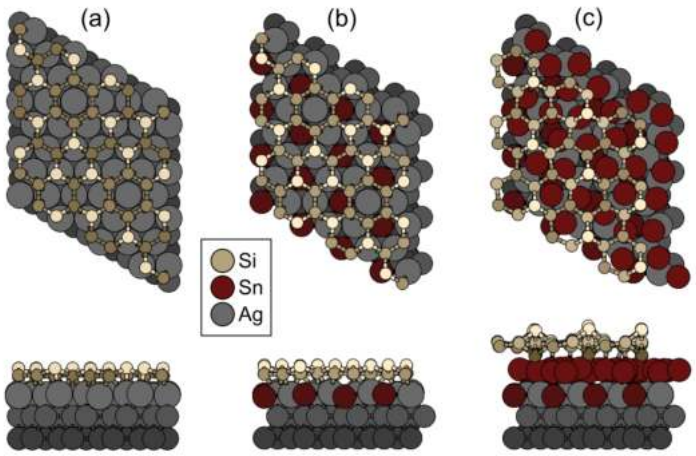

Figure 1: Structural models of silicene grown on (a) Ag(111), (b) $SnAg_2$/Ag(111), and (c) stanene/$SnAg_2$/Ag(111).

N-Heterocyclic Carbenes as Modifiers of Metal-Supported Graphene

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Abstract

N-heterocyclic carbenes (NHCs) provide a new method to functionalize graphene. We have demonstrated the NHC functionalization of graphene monolayers on Pt(111) and Ru(0001)[1]. As probed using reflectance IR spectroscopy, the grafted NHCs display thermal stabilities similar to that reported for aryl groups on graphite and graphene. Differences in the strength of the interaction between the Gr/Pt(111) and Gr/Ru(0001) systems are attributed to their respective p-doped and n-doped characters coupled with the electron-donor property of the NHC. Intercalation of oxygen between graphene and Ru(0001) leads to NHC bonding similar to that for Gr/Pt(111) consistent with the removal of the strong Gr/Ru interaction to form a quasi-freestanding p-doped layer. These observations are significant since NHCs form an extremely diverse family of compounds that may enable custom approaches to modify graphene for specific applications. The persistent character intrinsic to NHCs should facilitate functionalization from the vapor-phase while limiting co-deposition of dimerization products. For the same reason, NHCs may offer greater homogeneity of functionalization in that they are less indiscriminately reactive than aryl radicals or simple carbenes. Observations related to catalysis will be discussed.

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Secondary-ion mass spectrometry evaluation of MAX and MXene production approaches

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With recent development in secondary-ion mass spectrometry (SIMS) instrumentation complemented by establishing dedicated measurement procedures – tailored for specific samples – it is possible to push the boundaries of SIMS analyses and reach atomic depth resolution.

SIMS measurements of the MAX phase and resulting MXene samples produced using a conventional process reveal that oxygen atoms are incorporated in the carbon sites. Thus, they should be considered as early transition metal oxycarbides and not carbides as it is commonly assumed [1]. However, modification of the production procedure [2,3] may lead to the formation of pure carbide materials. The outcome can be evaluated with the SIMS technique which can unambiguously detect and identify all elements, starting with hydrogen, with atomic depth resolution, atomic layer by atomic layer. Such precision may prove invaluable for further developing MAX and MXenes and other 2D materials.

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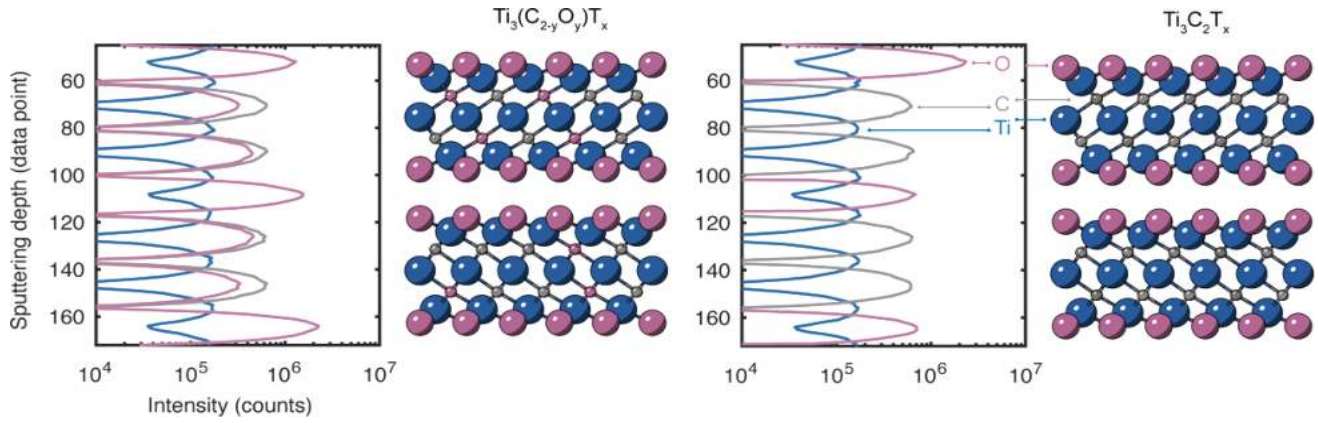

Figure 1: SIMS depth profiles of multilayer Ti3C2Tx MXene samples. Samples produced with conventional processes (left) show significant levels of oxygen in the carbon layer, qualifying them as oxycarbides. Pure carbides can be obtained with modified production procedures (right).

High Yield, Bottom-Up/Top-Down CVD Synthesis of 2D Layered Metal Selenides—A Promising Class of Materials for Applications in Electronics and Electrochemistry

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Since the excitement about graphene, a monolayer of graphite, with its 2010 Nobel Prize, there has been extensive research in the synthesis of other non-carbon few/mono-layers exhibiting a variety of bandgaps and semiconducting properties (e.g., n or p type). The main approaches to deposit few/monolayers on a substrate are: (a) bottom-up synthesis from precursors using chemical vapor deposition (CVD) or (b) top-down exfoliation (liquid or mechanical) of bulk layered material.
Here we show a combined bottom-up and top-down approach where (a) we synthesize in one step high yields of bulk layered materials by annealing a metal in the presence of a gas precursor (sublimated selenium from selenium powder) using chemical vapor deposition (CVD) and (b) we exfoliate and deposited (dropcast or Langmuir Blodgett) few/mono-layers on a substrate from a sonicated mixture of our material in a specific solvent. It is interesting to note that, besides the structure being 2D layered, the properties of the nanomaterials synthesized slightly differ from the materials with the same stoichiometry synthesized using conventional chemical methods (e.g., solvothermal).
In this talk, we will discuss the chemical synthesis, the very extensive characterizations, and the lessons we learned in making multiple metal selenides (Ag-Se, Cu-Se, W-Se, Mo-Se, etc.). We will see how we integrated these new materials into sensors, as functional coatings, and into electrochemical devices (see selected published papers below).

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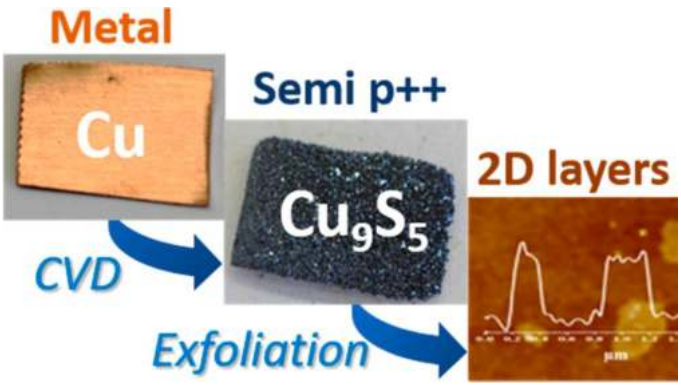

Synthesis of digenite from controlled CVD sulfurization of bulk copper foil

Nano-modulated electronic properties of borophene on Ir(111)

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Detailed insight into electronic properties of 2D materials is crucial for a complete understanding and application of these atomically-thin systems. For borophene, a polymorphic 2D sheet of boron atoms [1], it is challenging to produce large-scale uniform samples, thus hindering thorough characterization of its electronic structure. Here we present core-level and valence band characterization of borophene on Ir(111) substrate (see Fig. 1(a)). Borophene synthesis has been performed by means of segregation-enhanced chemical vapour deposition with borazine as a precursor [2,3], which enables production of large (cm-sized), single-layer borophene samples. Several spectroscopic methods were employed in order to determine the electronic properties of borophene. X-ray photoelectron spectroscopy (XPS) and scanning tunnelling spectroscopy (STS) disclosed different chemical environments of B atoms and indicate inhomogeneous Bo-Ir interaction modulated at the nanoscale. Furthermore, angle-resolved photoelectron spectroscopy (ARPES) mappings in the vicinity of the Fermi level (see Fig. 1(b)) reveal distinct electronic bands which are associated with the borophene monolayer, including signatures of electron scattering on the crystal lattice of borophene. Our data provides a comprehensive insight into the electronic structure of borophene sheets, which could expedite their utilization in future technologies.

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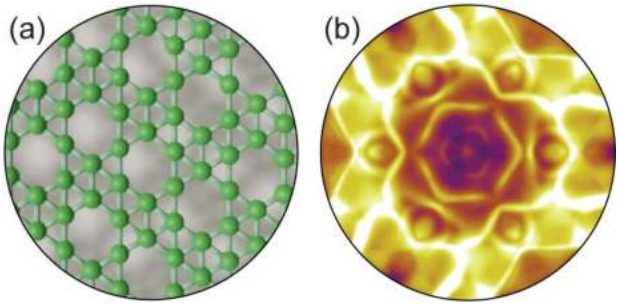

Figure 1: (a) Ball-and-stick model of borophene on Ir(111). (b) Fermi surface of borophene on Ir(111), as determined with ARPES.

Optical and structural properties of ternary $MoWS_2$ grown by atomic layer deposition

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Molybdenum disulfide (MoS_2) and tungsten disulfide (WS_2) are members of the transition metal dichalcogenide material family. Two-dimensional (2D) monolayers of MoS_2 and WS_2 are semiconductors with a direct optical band gap of ~1.9 and ~2.0 eV, respectively. The similar crystal structures and lattice constants of both materials promise full miscibility of ternary $MoWS_2$ and no strain/relaxation induced defects and phase separation issues are expected. Depending on the concentration x in $Mo_xW_{1-x}S_2$ it is possible to tune the exciton transition energy [1].

In this work, atomic layer deposition (ALD) was used for the growth of monolayer MoS_2, WS_2 as well as $(Mo,W)S_2$ in different compositions on a SiO_2/Si substrate. ALD uses a pulsed precursor supply, i.e. the S, Mo and/or W precursors are provided separately during the growth process to enable the chemical reaction between the absorbed precursors on the surface only. Raman spectroscopy and scanning transmission electron microscopy (STEM) allow to investigate the homogeneity of the distribution of Mo and W atoms in monolayers depending on the number and sequence of alternating pulses for ALD growth.

$Mo_xW_{1-x}S_2$ monolayers were grown with different concentrations in the whole composition range from x=0 to x=1. Raman and photoluminescence (PL) investigations show a concentration dependent shift of the A_{1g} mode (Figure 1a) and the recombination energy of the A and B exciton (Figure 1b), respectively. The distribution of Mo and W atoms in the $MoWS_2$ layer with different concentrations is studied by TEM. It is shown that clustering of Mo and W appears for concentrations close to binary MoS_2 and WS_2 due to the pulsed precursor scheme.

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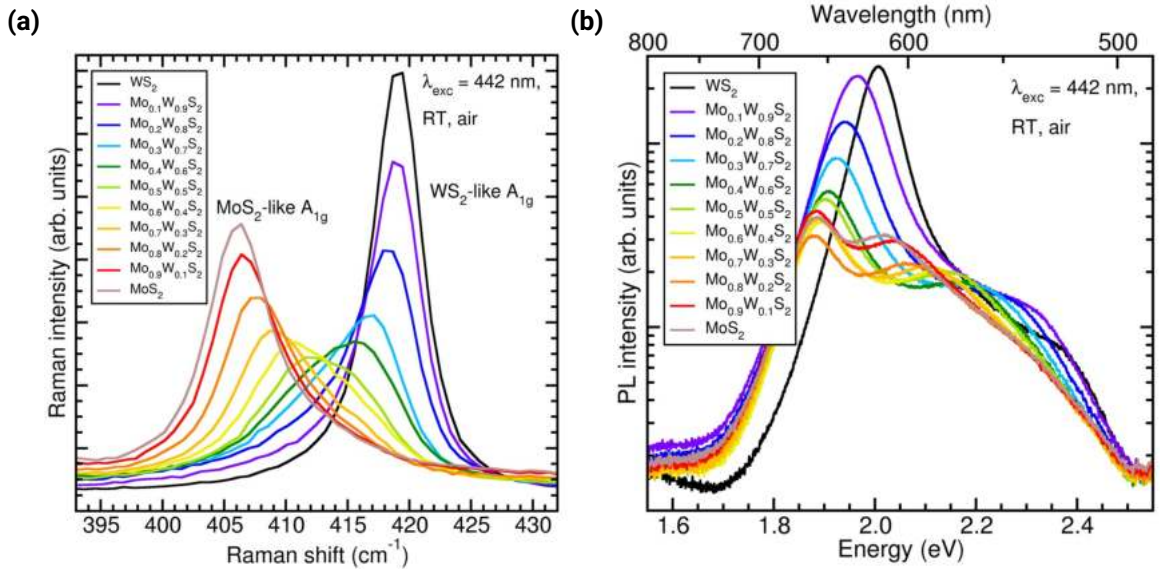

Figure 1: Raman spectra (a) and PL spectra (b) of $Mo_xW_{1-x}S_2$ monolayers with different concentrations.

Photoelectrochemical behaviour of WSe_2 nanoflakes: Structure-dependency and the effect Pt-decoration

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The two-dimensional (2D) semiconductors, the transition metal dichalcogenides, have received great interest during the last decade because of their high chemical stability and good electrocatalytic properties [1-2]. The photoelectrochemical (PEC) behaviour of 2D nanoflakes depends on their structural properties (the number of layers, the basal planes, and edges). Defect-rich in-planes and edges create surface states within the bandgap, which in turn act as recombination centres for the photogenerated charge carriers [3-4]. Pt nanoparticles (NPs) on 2D surfaces are considered either in a catalytic context for the PEC hydrogen evolution reaction (HER), or as passivating agent of defect states [3-4]. We need to understand the fundamental PEC properties of 2D semiconductors in the function of their structural properties, and the role of Pt NPs in defect healing using a microscopy-based approach with spatial resolution.

The WSe_2 nanosheets were mechanically exfoliated to get bulk, few-layered, and monolayer specimens. The WSe_2 nanoflakes had well-defined thicknesses as measured by atomic force microscopy (between ca. 0.9 nm and 250 nm) and the Pt NPs were deposited by a variable number of atomic layer deposition cycles. I will show our recently developed custom-designed PEC-microscope setup [5]. The deposited 5–50 µm sized microdroplet acts as an electrochemical cell on the chosen sample area of the 2D flake, which is illuminated by either white or monochromatic light. Then, I present the use of model reversible redox species to mimic photoelectrocatalytic processes, proving the differences between basal planes and edges. Additionally, I show a record high photocurrent and photon-to-electron conversion efficiency values for Pt-decorated WSe_2 nanoflake photocathodes applied in PEC HER. The effect of WSe_2 nanosheet thickness, as well as Pt surface loading was carefully quantified [6].

Finally, I demonstrate the effect of structural domains on the PEC performance of 2D WSe_2. i) The photocurrent losses with growing the fraction of the edges, and the parallel rise of dark currents. ii) The LPE produced WSe_2 bulk and few-layer nanoflakes in water reduction and oxidation, achieving only µA cm^{-2} current densities. This decrease in the PEC performance can be explained by the growing of defect densities, because of the number of edge sites is increased and the area is decreased of LPE prepared nanoflakes, which means the main issue in the larger-scale application of these materials [5, 7].

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The functionalization of graphene oxide as a route towards reduced graphene oxide with an increased water-dispersibility

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Abstract

The large-scale production of graphene is of great importance owing to its unique electronic, electrochemical, optical, thermal and mechanical properties that open the way to its application in a variety of research and industrial areas [1]. However, the employment of graphene on a large scale is still limited owing to its low yield of production and dispersibility both in water and in many organic solvents. Graphene oxide (GO) is the oxidized form of graphene that shows exceptional dispersibility in water due to the presence of oxygen-based functional groups in its structure, such as hydroxyls, epoxides and the less abundant carboxyl groups [2]. In the presence of reducing reagents, GO is converted to reduced graphene oxide (RGO), with the removal of a large portion of oxygen-based functional groups and the partial restoration of the extended conjugation. As in the case of graphene, the layers of RGO are prone to aggregate in water by restricting its employment in many fields, especially in the biological ones. Herein, the sustainable preparation of highly water-dispersible RGO is presented. In particular, GO has been obtained from high-purity synthetic graphite, enriched with carboxyl functional groups via the reaction with succinic anhydride, and eventually chemically reduced with L-ascorbic acid. The obtained dispersion displays unprecedented colloidal stability in water in a wide range of pH, afforded by the ionizable carboxylic groups (Figure 1). All the reaction steps and intermediates have been widely monitored through spectroscopic and morphological techniques in addition to theoretical calculations.

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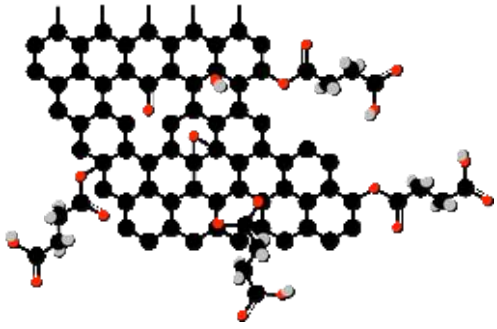

Figure 1: Proposed structure of a single layer of carboxyl-rich reduced graphene oxide

Densely and selectively functionalized graphenes for energy storage and catalysis

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Selective and dense functionalization of graphene with redox-active, bioinspired, descrete molecular species or single metal atom ions can mitigate its tendency for restacking and boost interactions with target chemicals, combined with stability in challenging environments. Such properties are pre-requisites for sustainable electrode materials for energy storage and catalysis. By leveraging the susceptibility of fluorographene to nucleophiles, advanced and tailored graphene derivatives can be obtained for targeted applications. Graphene acid (GA), cyanographene (G-CN), superdoped graphene are indicative examples. GA bears carboxyls which are strong metal-coordination sites[1] and handles to immobilize aminoacids for development of catalysts for fuel production.[2] As Li-ion battery anode, GA reveals high redox capacity stemming from its carboxyl groups, and high conductivity.[3] The nitrile groups of G-CN mediate electronic communication between the graphene and metal ions, affording mixed valence Cu(I)-Cu(II) undercoordinated catalytic centres, enabling the effective production of pharmaceutical synthons via cooperative single-atom catalysis.[4] Nitrogen superdoping affords dense conductive electrodes with superior elelctroactivity and energy density in supercapacitors.[5] Such graphene derivatives lay the ground for the development of the next generation materials for energy storage, and catalysis but also for sorption, environmental monitoring and biomedical applications (Fig. 1).

Acknowledgements: All co-authors are acknowledged for their key contribution in all related articles. We thank the support from ERDF/ESF project "Nano4Future" (No. CZ.02.1.01/0.0/0.0/16_019/0000754), from GACR (EXPRO No19-27454X).

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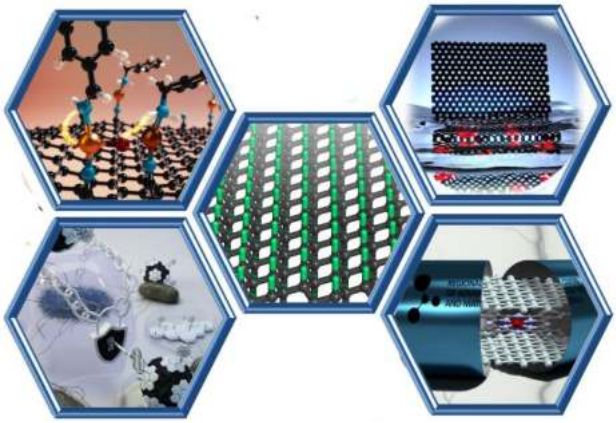

Figure 1: Fluorographene chemistry offers densely and selectively functionalized conductive materials for energy storage and catalysis.

Improved electrical transport properties of CVD graphene by sulfur doping

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Graphene from the metal catalyzed chemical vapor deposition, CVD, methodology opened the way for the large area applications of this material as transparent electrodes for substituting conductive oxides. However, CVD graphene layers are polycrystalline and their structure at atomic level is well far to be free of defects, which affect graphene properties. Atomic scale defects act as scattering centers and lead to a loss of carrier mobility. On the other side, structural disorder at grain boundaries provides additional resistance in series that affect the material conductivity.

Graphene chemical functionalization has been demonstrated to be an effective way for improving graphene conductivity mainly by increasing carrier concentration in the material [1, 2]. The present study reports the healing effects of sulfur doping on the electrical transport properties of single layer CVD-graphene. Single layer of graphene on Corning-glass and Si/SiO_2 substrates are treated by a post-growth thermal sulfurization process operating at 250°C. Combined, XPS and Raman analysis reveals the covalent attachment of sulfur atoms in graphene carbon lattice without creating new C-sp^3 defects. Measurements of transport properties show a strong improvement of material conductivity that is related to an increased mobility as revealed by Hall measurements. The sulfur-chemistry leading to the graphene defects healing, including the creation of disulfur bridges at grain boundaries, and their effect on carrier mobility are discussed.

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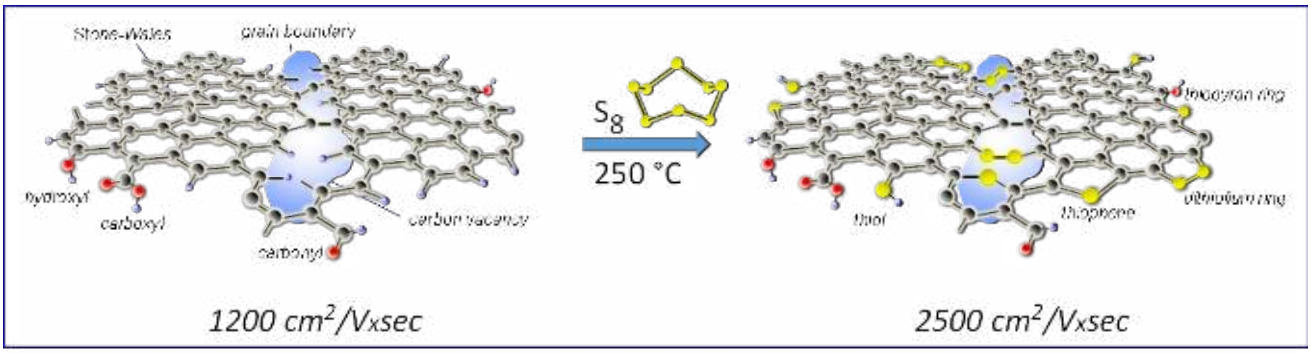

Figure 1: . Schematic representation of CVD graphene as prepared and after thermal sulfurization process. The yellow spheres represent sulfur atoms. The different C–S bonding configurations inserted into the graphene network are evidenced as well as the typical carbon vacancies, grain boundaries and the Stone–Wales defects.

2-dimensional Material Inks for Additive Electronics Manufacturing of Planar and Conformal Optoelectronics

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Recent advances in the synthesis of 2-dimensional (2D) materials-based inks has increased the design space for additive electronics manufacturing for sensors, 2D-silicon integrated photonics, energy harvesting and storage, and more. In particular, the Advanced Nanomaterials and Manufacturing Laboratory at Boise State University has undertaken several projects to help overcome obstacles facing the integration of 2D materials in applications for energy, healthcare, and water. This talk will highlight results of several ongoing studies on the development of 2D and layered materials inks for materials jetting platforms such as inkjet printing (IJP), aerosol jet printing (AJP), and plasma jet printing (PJP). These tools are promising technologies for direct deposition of functional 2D and layered nanomaterials. Such printers have significant advantages over standard microfabrication techniques, including low cost, noncontact printing, rapid prototyping, and compatibility with roll-to-roll production of electronic devices and sensors on flexible substrates. However, formulating stable inks which can meet the various rheological requirements of these various platforms can be quite challenging. Here we present recent developments in 2D and layered nanomaterial inks including black phosphorus [1], transition metal dichalcogenide alloys, Bi_2Te_3 thermoelectric [2], and MXene [3] based inks for a variety of additive electronics manufacturing applications. Such multifunctional material inks highlight a new dimension for research on next-generation printing of electronic devices such as low-cost sensors, energy conversion and storage devices, and microscale electronics.

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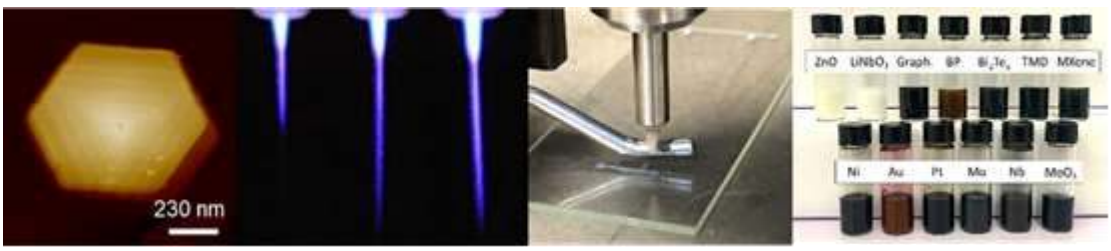

Figure 1: Examples of inks and printers for direct writing of electronics. Left to right shows thermoelectric flakes, plasma jet printing, aerosol jet printing, and library of developed inks.

Graphene decorated with uniform nanoholes via an electrochemical route for energy storage applications

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Graphene nanosheets decorated with nanometer-sized holes are of interest in different technological applications, such as electrochemical energy storage or molecular separation [1]. They are usually obtained by selective etching of the highly oxidized domains of graphene oxides derived from traditional (e.g., Hummers) methods. However, control over the size uniformity of the nanoholes can be difficult due to the spatial randomness and high connectivity of the oxidized domains. Here, we propose a new route to obtain graphene decorated with uniform nanoholes by using oxidized graphene nanosheets prepared by an electrochemical exfoliation/oxidation approach [2,3]. This oxidized graphene is shown to have larger and better interconnected aromatic domains (higher electrical conductivity) as well as smaller and more labile oxidized domains than those of its counterpart obtained by traditional oxidation routes (standard graphene oxide). As a result, selective removal of the oxidized domains by H_2O_2 treatment led to holey graphene nanosheets with smaller and more uniform nanoholes (typically ~4-6 nm) than those attained using standard graphene oxide (Figure 1). When used as an electrode material for electrochemical charge storage, the electrochemically derived holey nanosheets exhibited a higher capacity and energy density than those of their counterpart prepared by the traditional oxidation route.

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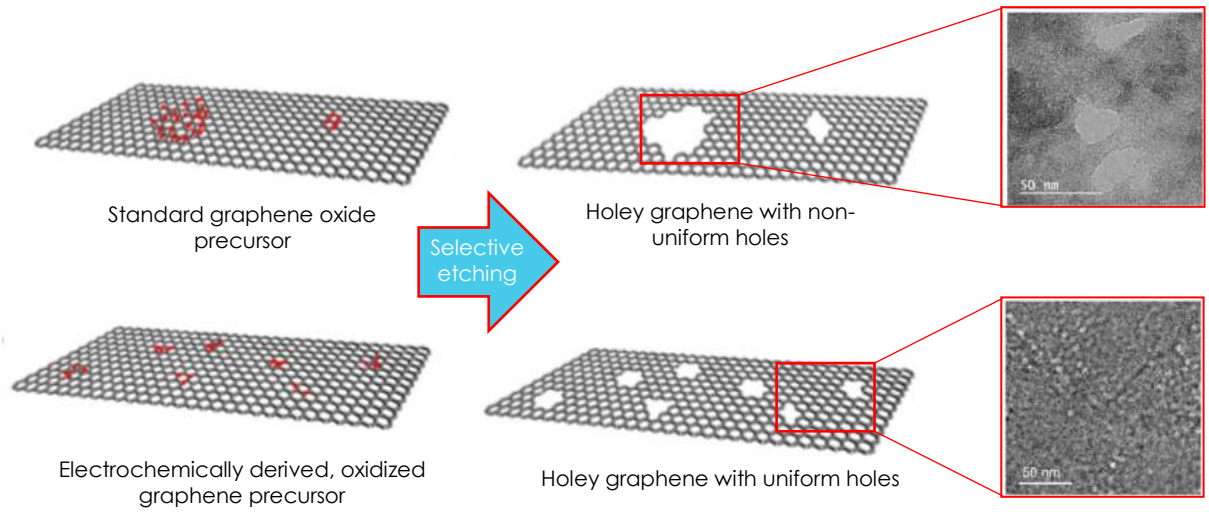

Figure 1. Schematic of the preparation of holey graphene from structurally/chemically different oxidized graphene precursors and TEM images of the corresponding holey nanosheets

Understanding Flake Size Separation during Centrifugation

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Liquid phase exfoliation is a powerful tool for the preparation of 2D nanomaterial inks and dispersions that can be used for a wide range of applications. [1] While such dispersions have enormous potential because of their flexibility, their polydispersity is an intrinsic drawback. [2] Centrifugation has been widely applied to separate flakes of different sizes while maintaining the easily processable dispersion; however, the centrifuge conditions required are still poorly understood and trial and error is usually required to optimise the desired flake size. [3] We have developed a model, balancing the complex shapes and sizes of nanoflakes with the need for suitable ensemble parameters, to predict the behaviour of dispersions under centrifuge conditions. Assuming regular rhombus flakes and using previously measured surfactant binding, the density and friction of the nanosheets can be expressed. [4] This theoretical model has been successfully applied to transition metal dichalcogenides (Figure 1). With this model it should be possible to identify the exact centrifuge conditions required for different sized nanoflakes and simplify the purification required from polydisperse samples commonly prepared from liquid phase exfoliation.

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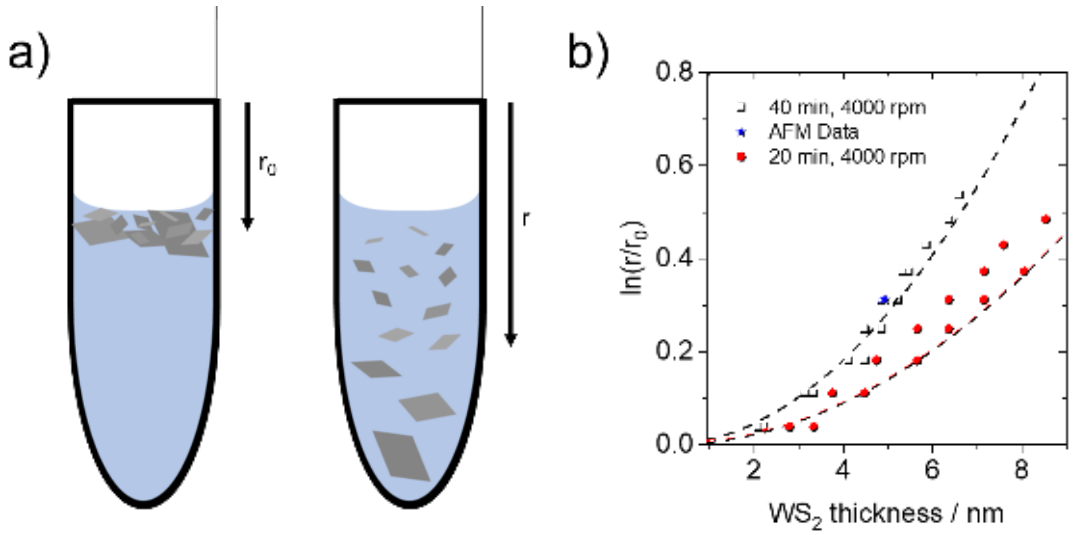

Figure 1: a) Schematic of band sedimentation process to separate nanoflakes according to size. b) Measured distribution of tungsten disulphide flake sizes down centrifuge tube matching the theoretically predicted distribution (dashed lines).

CVD synthesis of few-layered h-BC_2N and optical applications

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Arranging carbon, boron, and nitrogen atoms in a sp^2 network can give rise to tailored electronic properties from insulators (*h*-BN) to metals (graphene)[1,2]. For semiconductor applications, the construction of a ternary structure (*h*-$B_xC_yN_z$) is highly desirable, but its uniform and large-area synthesis has remained a great hurdle. This challenge has been attempted by a fchemical vapor deposition method with a single molecular precursor, N-tri-methyl borazine where boron, carbon, and nitrogen atoms are covalently bonded, onto Ni catalysts in conjunction with the quenching method after the synthesis. The atomic structure closely resembles *h*-BC_2N as presented by XPS and nanometer resolution EELS mapping, and the photoluminescence and electroluminescence observed from the *h*-BC_2N film were in agreement, proving its well-established bandgap of 2.15 eV [3]. As optical application, the utilization of *h*-BC_2N film for 2D light emitting diodes was demonstrated. Though films might have impurities such as small *h*-BN fragments and *h*-$B_xC_yN_z$ other than *h*-BC_2N phase, we believe that this work provide a starting point of controlling the ternary BCN compounds that retain sp^2 hybridized chemical bonds.

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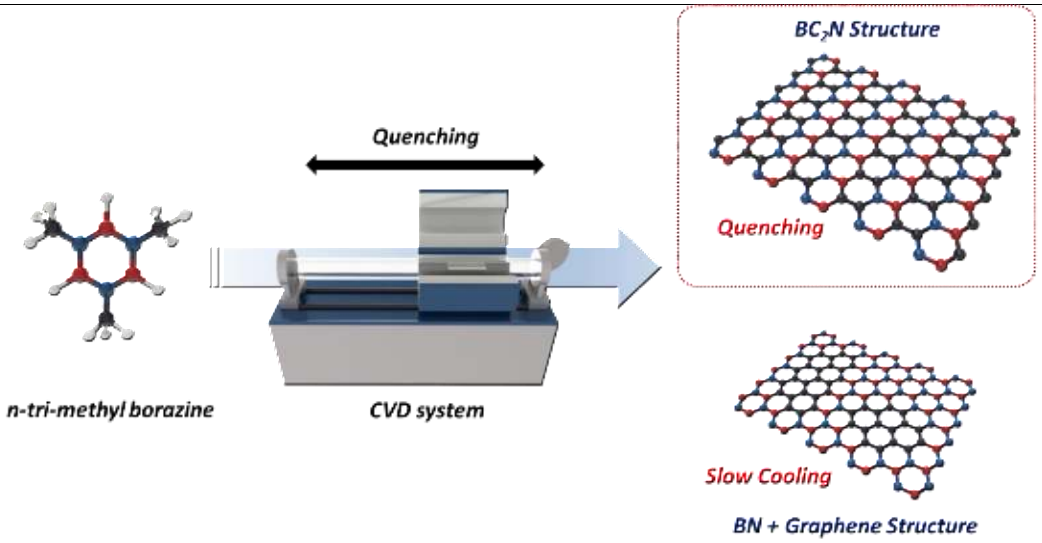

Figure 1: Schematic diagram of *h*-BC_2N synthesis.

Defect Engineering of Au@MoS_2 Nanostructures for Conventional and Plasmon-Enhanced Hydrogen Evolution Reaction

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2D materials such as MoS_2 has received a lot of attention for key electrochemical processes such as the hydrogen evolution reaction (HER). A lot of efforts have been dedicated to increase the HER activity of MoS_2 basal plane by phase and defects engineering. Core@shell systems such as Au@MoS_2 nanostructures (Fig. 1a) present an enhancement of their HER properties with respect to MoS_2 due to charge transfer effects [1]. In this work, we show that the structural and HER properties of Au@MoS_2 structures can be further enhanced by performing a thermal treatment under reducing conditions. In addition of the extensive catalytic tests performed in acidic conditions, the samples were investigated by combining ex-situ and in-situ aberration-corrected STEM under controlled atmosphere as well as Raman and XPS spectroscopies. We show that the thermal treatments lead to a decrease of the number of external MoS_2 layers as function of the temperature (Fig. 1b). In particular, 56% of the Au@MoS_2 structures annealed at 800°C under H_2 atmosphere present an incomplete MoS_2 layer. This sample shows the highest HER performances with an overpotential of 203 mV vs RHE at 10 mA/cm^2 (Fig. 1c) and a Tafel slope of 55 mV/dec which is similar to the value reported for edges sites in MoS_2 [2]. Measurements of the electrochemical double-layer capacitance also shows that it presents the highest number of active sites which is consistent with the greater number of incomplete shell layers observed by TEM. We also explored the plasmonic-assisted HER performances of these nanostructures under LED illumination (Fig. 2) and observed an enhancement by about 10% of the current density, depending on the wavelength used. All these results on the tailoring of the microstructural and HER properties of Au@MoS_2 nanostructures will be discussed in details [3].

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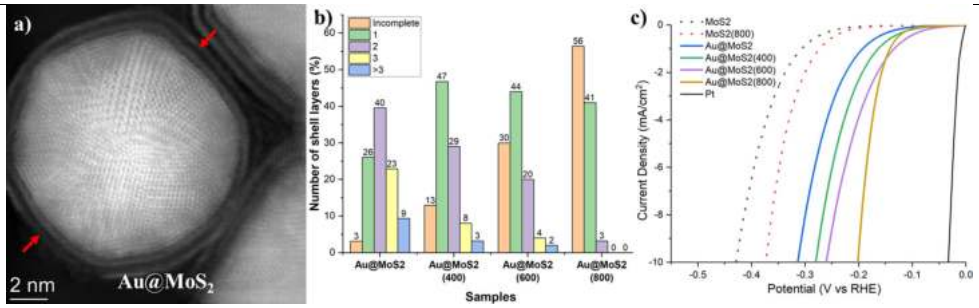

Figure 1: Aberration-corrected STEM image of the initial Au@MoS_2 sample, b) Evolution of the number of shell layers c) LSV curves obtained in 0.5 M H_2SO_4 at 10 mV·s^{-1}.

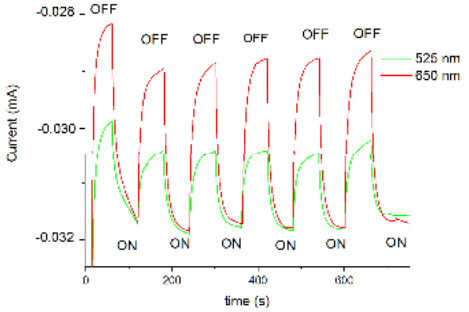

Figure 2: Evolution of the current under illumination at a fixed potential of -0.4 V.

The True Amphipathic Nature of Pristine Graphene Flakes

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Commercialization of graphene is slower than anticipated. Graphene due to their extraordinary electronic, thermal and mechanical properties, holds great promise for applications ranging from optoelectronic, through environmental to biomedical technologies. However, the conflicting reports about its chemical character [1,2,3,4,5] hinder potential applications. For many potential large scale processing routes to efficiently manufacture and commercialize graphene based devices, composites, coatings, membranes or inks it is essential to understand the fundamental colloidal properties of pristine graphene flakes (GF).

Our recent studies into colloidal properties of pristine GF [6] revealed why and how they can be used as emulsion stabilizers without using any additional surfactants. The rigorous quantum-mechanical, molecular dynamics and Monte Carlo calculations supported by wet-chemistry testing, optical and electron microscopies, Raman spectroscopy and thermogravimetric analysis, explained the physico-chemical mechanism governing their amphiphatic nature. In contrast to commonly used graphene oxide flakes, pristine graphene flakes possess well-defined hydrophobic and hydrophilic regions in the basal plane and edges, respectively. These properties allow small flakes to be utilized as stabilizers with an amphipathic strength that depends on the edge-to-surface ratio. The interactions between flakes can be also controlled by varying the flake thickness and the oil-to-water ratio. Our findings reconcile all previous results on the chemical nature of graphene flakes. In addition, it is predicted that graphene flakes can be efficiently used as a new generation surfactants that is active under high pressure, high temperature, and in saline solutions, greatly enhancing the efficiency and functionality of applications based on this material. The direct applications of GF stabilized emulsions will be also discussed [7].

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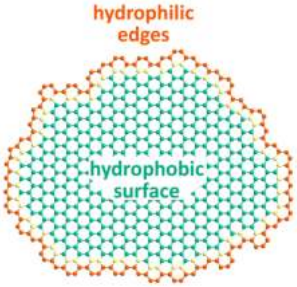

Graphene nanoarchitectures: from fundamentals to applications

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On-surface reactions, via programmed interactions of molecular building blocks, has recently emerged as a promising route to synthesize atomically precise materials from the 'bottom-up'. This approach ensures exquisite atomic-scale control of the structural and chemical functionalization, allowing to design a vast number of carbon-based nanoarchitectures not available by traditional solution chemistry nor with the 'top-down' methodologies.

In this talk, I will discuss our recent results to synthetize atomically precise nanoporous graphene [1], graphene nanoribbons and their chemical functionalization and how to organize them into atomically-sharp heterojunctions [2-4], and the molecular bridge engineering (fig.1) for tuning quantum electronic transport and anisotropy in nanoporous graphene [5].

At the end of the day, this talk will demonstrate the full path to synthetize a semiconducting graphene material with a bandgap similar to that of silicon, its atomic-scale characterization, and its implementation in a three-terminal electronic device, as well, its implementation as atomically-thin membrane for gas filtration and integrated into photonic biosensors.

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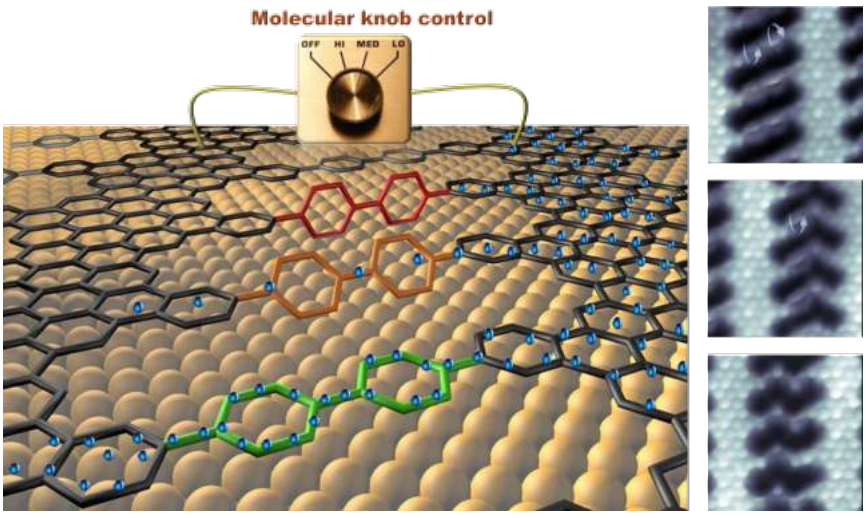

Figure 1: Concept sketch of molecular bridge engineering for tuning quantum electronic transport and anisotropy in nanoporous graphene, and (right) STM images displaying its experimental realization.

Preparation and application of graphene glass fiber

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Graphene glass fiber (GGF) is a new advanced graphene composite material developed through chemical vapor deposition (CVD) method with graphene covering on the surface of glass fiber. Glass fiber is a commercial lightweight structural material with high mechanical strength and flexibility, and has been widely used as a reinforcing material in aircraft, automobiles, etc. Considering the intrinsic excellent infrared properties of graphene and glass fiber, a dual-infrared-emitter design was reasonably constructed in GGF.[1] Dual-emitter GGF followed the law of gray-body radiation, showing high infrared radiation capability. Meanwhile, its infrared radiation can be effectively modulated through the band structure engineering of graphene.[2] Graphitic nitrogen doping can regulate the infrared emissivity of GGF from 0.96 to 0.68 under the premise of keeping high solar absorption. GGF showed promising potentials targeting the high-performance photothermal conversion for electric-energy-free crude oil collection.

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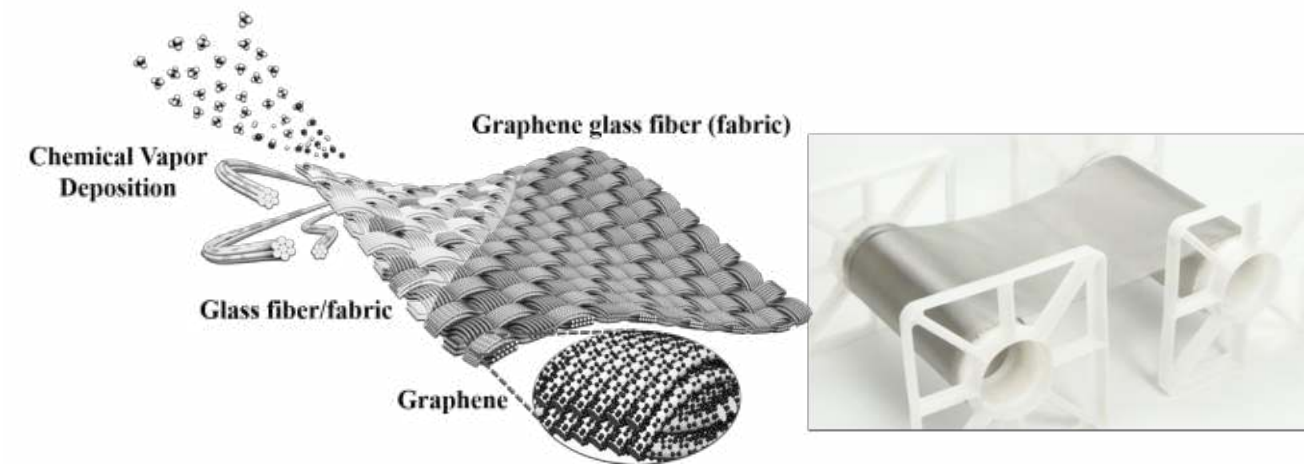

Figure 1: Graphene glass fiber (fabric)prepared by chemical vapor deposition strategy

Highly Efficient Gold Extraction by Graphene

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Abstract

Gold is the most valuable element in e-waste. Currently, only less than 20% of e-waste has currently been recycled primarily for a lack of technologies with sufficient efficiency and economic viability to recover valuable elements within it[1-3], Therefore, developing materials capable of extracting gold from complex sources, especially electronic waste (e-waste), can turn the e-waste recycling challenge into a profitable business. Here we report an exceptionally high gold extraction capacity of chemically reduced graphene oxide (rGO), reaching >1.8 g/g when extracting gold from its 10 ppm solution at 25 °C (Fig.1), one order of magnitude higher than other reported gold adsorbents, and an ability to extract gold from ppt concentrations. During extraction, rGO reduces >95% gold ions to metallic gold, avoiding elution and precipitation necessary in post-adsorption processing (Fig. 1b). Moreover, this reductive adsorption of gold by rGO is found different from (predominately) electrostatic adsorption of other metal ions, hence, exploiting the protonation process of rGO, a precise gold extraction without adsorption of the other 14 elements normally present in e-waste is achieved. Finally, by assembling rGO nanosheets into a membrane, we have developed a rGO membrane-based continuous process that a 1 m^2 rGO membrane is capable of recycling gold from ~22,000 L of ~100 ppb gold solution. Our findings show a promising venue for addressing global e-waste challenges and gold scarcity.

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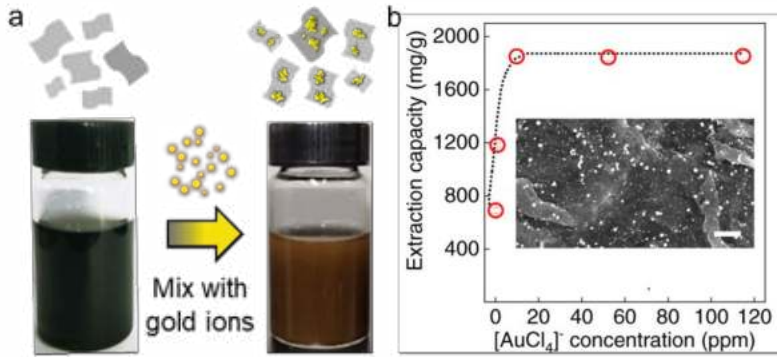

Figure 1: Gold extraction by rGO. (a) Schematic of the extraction process using rGO. After mixed with gold ion, rGO suspension gradually changed its color from black to brown. (b) Extraction capacity as a function of gold concentration after 24 h. Inset is a Scanning electron microscopy image of reduced gold nanoparticles adsorbed on rGO. Scale bar is 500 nm.

Graphene Epitaxy: from Misorientation-Free to Misorientation-Engineered Graphene Films

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Abstract (Century Gothic 11)
Graphene has garnered widespread interest and confer remarkable potential for next-generation technological applications, which relies on the controllable preparation of high-quality graphene films. Chemical vapor deposition (CVD) is considered the most promising method, and great progress has been achieved over the last decade. Currently, this field is being pushed to new heights that pursuit structure control (e.g. orientation, layer, stacking order, contamination, doping, etc.) and low-cost production (e.g. increasing the production capacity and growth rate)[1,2]. In this talk, I will introduce our recent works on controlled growth of high-quality graphene films via CVD approach, especially on controlling the crystallographic orientation of graphene. By designing and preparing single-crystal Cu(111) foils, we have opportunities in realizing the epitaxial growth of large-area single-crystal misorientation-free graphene film[3]. We designed and constructed a pilot-scale CVD system suitable for producing A3-size graphene films, which works well and output high-quality graphene films with high capacity. In another hand, we explore the possibility on controlling the layer number and stacking order, which is motivated by the emerging twistronics. Here I will present our state-of-the-art hetero-site nucleation method for growing twisted bilayer graphene (tBLG)[4]. Gas-flow perturbation and switching of the graphene edge termination play crucial roles in triggering the formation of interlayer twist. The growth mechanism is carefully investigated by using an isotope-labelling technique, and the as-obtained tBLGs show high crystalline quality (high carrier mobility of 68,000 $cm^2 V^{-1} s^{-1}$ at room temperature). We also established a slip-line-guided growth principle to explain and predict the crystal orientation distribution of graphene on a variety of metal facets, further enabling the controllable synthesis of single-crystal graphene and grain boundary engineering of bi-crystal graphene on designed metal facets[5], which opens a new avenue for manipulating the crystal orientations, grain boundary structures, and even twisted angles of bilayer 2D materials in a bottom-up manner

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How permeable is the impermeable graphene?

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Abstract

It is widely believed that, despite being one-atom thick, graphene and other defect-free 2D crystals are completely impermeable to all gases and liquids. This statement has extensively been justified in theory. In addition, the lowest detection limit (that is, highest sensitivity) achieved in the past experiments supporting graphene's impermeability was about 10^5–10^6 helium atoms s^{-1} for micrometer-scale membranes, or 10^{17}–10^{18} atoms m^{-2} s^{-1}.
In this talk, I will present my recent research[1-3] on the topic "How permeable is the impermeable graphene?" Using monocrystalline container made from atomically flat graphite, which is tightly sealed with graphene, our team have achieved measurements that put the permeation limit through 2D materials at 8–9 orders of magnitude lower than previously, such that we would discern (but did not observe) just a few helium atoms per hour crossing micrometer-size membranes. This detection limit is also valid for all other gases tested (neon, nitrogen, oxygen, argon, krypton and xenon), except for hydrogen. Hydrogen shows noticeable permeation, even though its molecule is larger than helium and should experience a higher energy barrier. This observation is attributed to a two-stage process that involves dissociation of molecular hydrogen at catalytically active graphene ripples, as shown by experiments[3], followed by adsorbed atoms flipping to the other side of the graphene sheet with a relatively low activation energy of about 1 electronvolt, a value close to that previously reported for proton transport[4,5].
The described device can identify only a few gas molecules per hour piercing a micrometer size membrane. This remarkable sensitivity allows detection of subtle transport phenomena that were not possible to observe previously, as exemplified by the "anomalous" hydrogen-gas permeation through defect-free graphene as discussed above. On the other hand, to move a step forward and make the generally "impermeable" graphene not only "permeable" but also highly "selective" toward gas molecules with a tiny difference in size, our team have developed a controllable perforation technique, which involves a short-time exposure of the graphene membrane to a low-energy electron beam. Using the same monocrystalline containers, we are able to study gas transport through the created individual graphene pores with an effective diameter of only 2 angstroms, or about one missing carbon ring. Helium and hydrogen permeate easily through these pores whereas larger molecules such as xenon and methane are blocked. Permeating gases experience activation barriers that increase quadratically with the kinetic diameter, and the transport process crucially involves surface adsorption.
The presented research would be important not only for fundamental understanding of the newly emerged physics and chemistry regarding molecular transport under atomic scale confinement, but also for developing new technologies for sustainable applications in energy and environment (for example, highly sensitive molecular detection and sensing techniques, inexpensive and nonmetallic graphene-based catalysts, and angstroporous 2D membranes for filtration and separation).

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A General Method to Disperse Solid Carbon Materials without Passivating Agents or Surface Modifications

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Graphite was treated with concentrated ammonium hydroxide for some days at room temperature, washed and exfoliated in water [1]. The resultant aqueous dispersion is metastable and has lyophobic nature. This graphite dispersion has concentration next to 25 mg L^{-1} and particles with lengths and widths of hundreds of nanometers and mean thickness between 10 and 40 nm typical of exfoliated graphite. The aqueous dispersion does not have any passivating agents, but remains dispersed for at least 6 months due to the mean zeta potential of -45 mV that provides electrostatic repulsion between dispersed particles. No functional groups or nitrogen doping were detected on graphite after the treatment with ammonium hydroxide, and the treated material was washed twice before sonication to remove most hydroxide, therefore neither functional groups, nitrogen atoms, nor hydroxide excess are responsible for the experimental zeta potential [1]. The contact of ammonia with pristine graphite promotes redox reactions and ammonia transfers electrons to graphite, which raises the Fermi level of the solid material and generates the negative zeta potential, consequently, such electron transfer is responsible for the colloidal stability. We also have extended this method to other carbon-based solids (sp^2). A carbon material that has gained attention is activated carbon (AC), not only because of the adsorptive properties highly explored in many applications, but because it is an edible conductor [2] and can be used to prepare edible electronic devices. AC was treated with ammonium hydroxide, washed and sonicated in water, resulting in an aqueous AC dispersion with concentration of about 75 mg L^{-1}, dispersed aggregates with sizes between about 60 and 500 nm, mean size of 220 nm, and mean zeta potential of -40 mV. No surface modification was detected, which is consistent with the hydrophobic behaviour and previous results for graphite. This method proved to be efficient for different carbon materials and has provided colloidal stability for long periods without passivating agents or functional groups, highlighting its great potential in the context of greener carbon-based dispersions.

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Process optimization of Joule heating CVD of graphene on meter-scale copper foil

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Abstract

Chemical vapor deposition (CVD) is currently the most promising method for producing large-area, high-quality graphene films [1]. Roll-to-roll CVD synthesis has been attempted to minimize the time in heating and cooling the CVD chamber [2-4]. A Joule heating-based synthesis method that uses high current flows through a copper foil and resistively heats the foil up to a prescribed temperature, has the advantage of low power consumption and small thermal mass [4]. However, this method has not been extensively researched due to difficulties in achieving uniform temperature distribution across the copper foil. To address this issue, we propose a U-shaped copper foil configuration that enhances temperature uniformity. The experimental design method was used to establish the optimal graphene synthesis conditions, and graphene was continuously synthesized while transporting a 2-meter-long copper foil. Electron microscopy and sheet resistance measurement confirmed the high-quality of the synthesized graphene.

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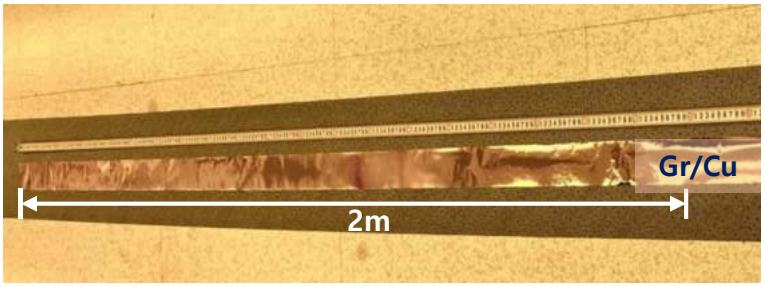

Figure 1: Schematic of Joule heating-based roll-to-roll graphene synthesis equipment

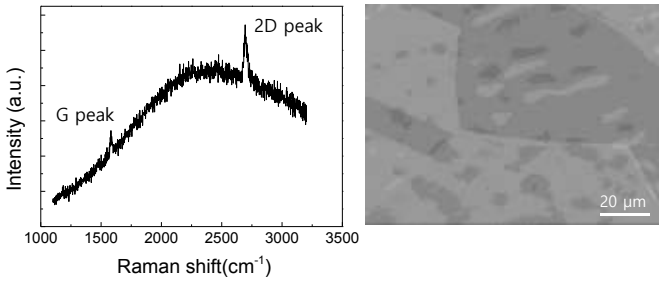

Figure 2: Raman spectra and SEM image of graphene/Cu foil

Two-dimensional material-enabled encapsulation for perovskite solar cells and modules

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Perovskite solar cells (PSCs) have emerged as a high-efficiency photovoltaic technology, but their instability has posed challenges to their commercialization. Recent stability assessments have been conducted on perovskite solar farms[1], but reliable accelerated aging tests on large-area cells remain scarce. To achieve a Levelized Cost of Energy (LCoE) comparable to commercial silicon photovoltaics, perovskite solar modules (PSMs) are expected to provide stable output for at least 20 years in outdoor conditions, while withstanding thermomechanical stresses caused by temperature fluctuations. In this study, we present an innovative industrially compatible encapsulation process by laminating a strain-free two-dimensional (2D) material-based encapsulant adhesives onto PSC/PSMs. The incorporation of 2D hexagonal boron nitride (h-BN) flakes, produced by liquid-phase exfoliation of their bulk counterpart[2], into the polymeric matrix is beneficial for the barrier and thermal management characteristics of the encapsulant[3]. The as-produced encapsulated PSCs and PSMs withstood multifaceted accelerated aging tests, including ISOS-D1 (shelf life storage under ambient conditions), ISOS-D2 (85°C, >1000 h), ISOS-L1 (light soaking, >1000 h), as well as customized thermal shock (200 cycles with abrupt temperature changes between +85°C and -40°C) and customized humidity freeze tests (10 cycles with abrupt temperature changes between +85°C and -40°C and including a water immersion step before device freezing), retaining more than 80% of their initial efficiency. Our results represent a significant progress towards the realization of long-term stable PSMs by utilizing industrially compatible laminable advanced composite encapsulants enabled by 2D materials.

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Figures

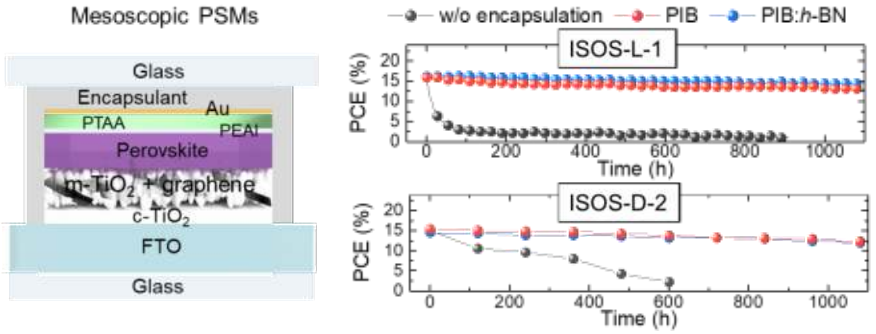

Figure 1: Schematic of the cell layout (active area = 1 cm^2), in which the non-compact layers of the device are fully covered by the encapsulant.

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Polymorphic Structural Phase Transformations in 2D Materials

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Abstract

Two-dimensional (2D) materials and their associated compositional polytypes, which are materials with identical chemical compositions that decorate different crystal structures, offer unique properties with potential uses in various emerging and hypothesized technological devices. Structural phase transformations provide a means for controlling the properties of single-layer 2D materials in a reversible manner through various methods such as post-synthesis modification using strain engineering or strong light-matter interactions. Exploring the polytype phase space can aid in identifying new potential candidates for structural phase transitions. Here, we utilized the C2DB [1] to identify the feasibility of structural phase transformations for 1,555 dynamically stable compositional polytypes. Out of these polytypes, we focus on 200 2D material compositions that possess an E_{hull} < 200 meV/atom and have more than one compositional polytype, which were analyzed to assess the feasibility of each composition's thermodynamic viability for polymorphic structural phase transformation. Finally, we highlight experimentally synthesized 2D compositions.

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Figures

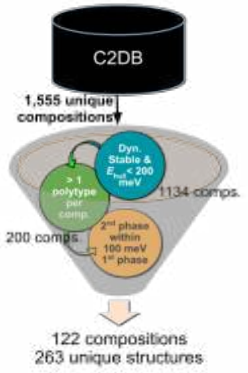

Figure 1: Selection criteria for filtering materials from the C2DB.

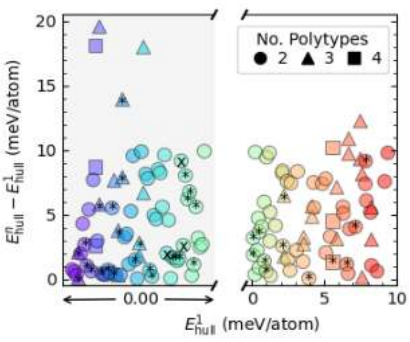

Figure 2: The relationship between the most stable primary polytype and higher energy polytypes for a given composition. The y-axis represents the difference between the higher energy and primary polytypes' E_{hull}. The x-axis indicates the E_{hull} of the primary phase. Each unique composition with E_{hull} = 0.0 is shifted horizontally to avoid overlapping data points. Marker color denotes the chemical composition, the * and x indicate 2D materials with a corresponding bulk parent structure in the ICSD/COD or has been synthesized in monolayer form, respectively, while marker shape indicates the total number of polytypes for each composition.

Bosonic Delocalization of Dipolar Moiré Excitons

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In superlattices of twisted semiconductor monolayers, tunable moiré potentials emerge, trapping excitons into periodic arrays. In particular, spatially separated interlayer excitons are subject to a deep potential landscape and they exhibit a permanent dipole providing a unique opportunity to study interacting bosonic lattices. Recent experiments have demonstrated density-dependent transport properties of moiré excitons [1], which could play a key role for technological applications. However, the intriguing interplay between exciton-exciton interactions and moiré trapping has not been well understood yet. In this work [2], we develop a microscopic theory of interacting excitons in external potentials allowing us to tackle this highly challenging problem. We find that interactions between moiré excitons lead to a delocalization (Figure 1) at intermediate densities and we show how this transition can be tuned via twist angle and temperature. The delocalization is accompanied by a modification of optical moiré resonances, which gradually merge into a single free exciton peak. The predicted density-tunability of the supercell hopping can be utilised to control the energy transport in moiré materials.

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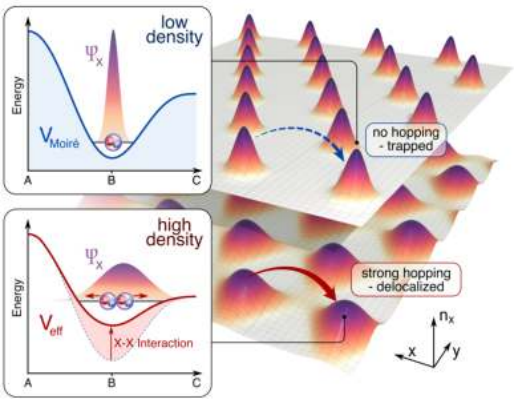

Figure 1: The moiré potential captures excitons within its minima creating arrays of localized excitons. For large densities the inter-excitonic repulsion gives rise to a decreased effective potential and a change of the exciton wave function with far-reaching consequences for optical properties and exciton transport.

Orbital origin of hidden spin textures in centrosymmetric PtSe2 monolayer and their proximity applications

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Harnessing the quantum degrees of freedom has become an essential paradigm for sustainable technological development. In 2D materials, the combination of spin-orbit coupling and reduced crystalline symmetries gives rise to the Rashba-Eddelstein which enables electrical control of the spin degree of freedom of electrons. It is a common belief that global inversion asymmetry is required to the existence of such effect. However, there is experimental data has confirmed this prediction and evinced the existence of opposite helical spin textures on the atomic planes of centrosymmetric 1T PtSe2 [1,2]. The 1T family of TMDs has not received as much attention as the other TMD polytypes and their topological aspects and properties are just being explored. Recent works from orbitronics -the orbital angular momentum analogue of spintronics- have inquired into their topological properties and demonstrated that the orbital angular momentum transport in these systems coexists with a higher-order topological phase [3]. In this work, we leveraged first-principles calculations and tight-binding models extracted from these, with symmetry analyses and large-scale transport simulations to demonstrate that the electrostatic origin of helical layer-localised spin and orbital textures and demonstrate their overlooked applicability for proximity effects and present an electrical probe for it.

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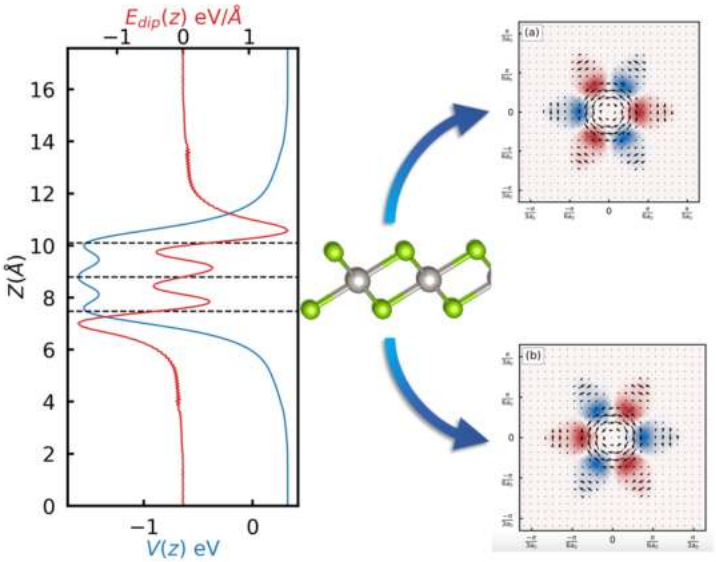

Figure 1: Real-space representation of the dipolar electric field in PtSe2 monolayers and schematic depiction of layer-projected spin textures.

Spin-filtering in 2D based magnetic tunnel junctions

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In this talk we first discuss an emerging physical picture of spin-filtering in multilayered graphene-ferromagnet systems supported by *ab initio* calculations and we compare it with experimental data. This picture involves spin filtering effects of arising from (i) graphene-FM hybridization, (ii) graphene k-point selection at the interface and (iii) "graphite" bulk band structure purification. These effects are shown to be either cooperating or competing. These results on graphene-FM systems are then compared predictions regarding other 2D based magnetic tunnel junctions (MTJs). Overall this study unveils paths to better harness the potential of 2D based MTJs.

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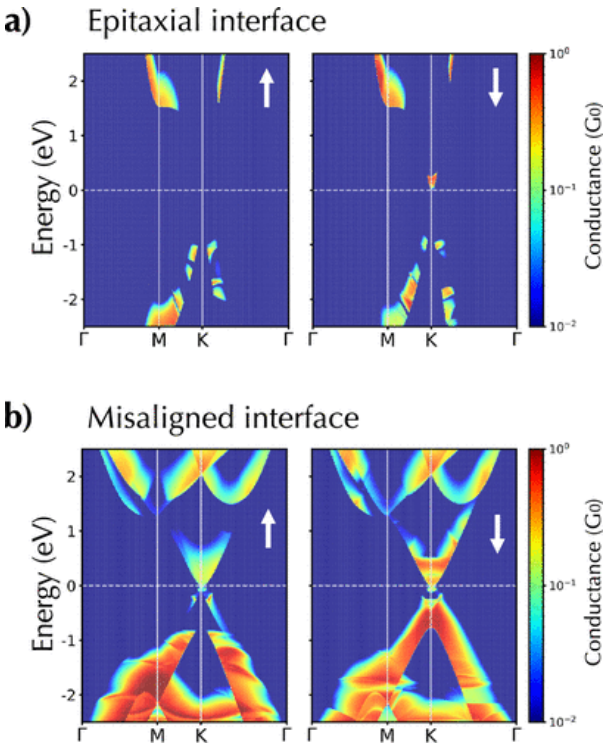

Electronic transmission across (a) the epitaxial and (b) the misaligned Ni/MLGr interfaces. Transmission coefficients are depicted in units of the quantum of conductance (G_0) along the conventional high symmetry k-path in the plane parallel to the interface. Left and right panels correspond respectively to majority and minority spin carriers. It is observed that the epitaxial case provides a highly asymmetrical spin-dependent transport channel, while a more balanced (but hence less spin polarized) spin transport is achieved in the misaligned case.

Domain wall network model for control of interfacial ferroelectricity in twisted bilayers of transition metal dichalcogenides

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Twistronic heterostructures are van der Waals materials for which mesoscale moiré periodicity considerably modifies electron and structural properties. Recently, a new phenomenon, interfacial ferroelectricity [1,2], has been discovered in slightly twisted homobilayers of semiconducting transition metal dichalcogenides (TMD) with parallel orientation of unit cells in constituent layers. Origin of the effect is related with interlayer charge transfer in domains formed by relaxation of moiré pattern. Thanks to out-of-plane direction of spontaneous polarisations in shape of domains can be modified applying external electric field in field effect transistor geometry. In my talk I formulate an analytical domain wall network model [3], which allows one to efficiently follow evolution of domain structure under external electric field. In particular, I show that for perfect (C_3-symmetric) relaxed moiré superlattice our model admits two regimes set by threshold electric field, determined by the model parameters. For electric field below the threshold, each domain wall in the network behaves like spring with clamped ends at the network nodes and bends to increase area of domains with favourable direction of polarisation. At the threshold field, the bending finishes by touching conditions for each pair of domain wall coming to the same node of the network. For post-threshold electric fields evolution of domain wall network represents a universal scaling of threshold structure with scaling parameter given by ratio of applied field to the threshold one. Finally, I demonstrate extension of the model on the case of irregular domains emerging in real twistronic heterostructures because of transfer-induced inhomogeneous strain [4].

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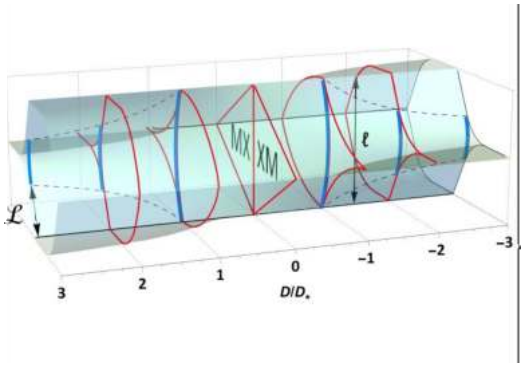

Figure 1: Variation of reconstructed moiré supercell in marginally twisted TMD homobilayers under out-of-plane displacement field D in units of threshold field D_*.

Tailoring Mechanical and Thermal Properties of Graphene Origami

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Abstract: Origami approach, an art of folding paper, is used to generate three-dimensional complex structures for extraordinary properties. However, the generation of graphene origami is still challenging. Here, using molecular dynamics simulations and density functional theory calculations, we show that patten-based hydrogenation (Figure 1a) can be employed to generate a wide range of complex graphene origami. Our results show that graphene Miura origami (Figures 1b and 1c) can show excellent properties, such as super compressibility and stretchability, negative Poisson's ratio behaviour (Figure 1b), and highly tunable coefficient of negative thermal expansion (Figure 1d). The super compressibility and stretchability and negative Poisson's ratio behaviors is due to the Miura origami geometry (extrinsic property). On the other hand, the negative coefficient of thermal expansion is due to inseparable combination of the pattern-based hydrogenation, Miura origami geometry, and large out-of-plane thermal fluctuations (intrinsic property of graphene). This study also opens opportunities to obtain other multi-functional materials by combination of hydrogenation, intrinsic properties of graphene, and three-dimensional geometry.

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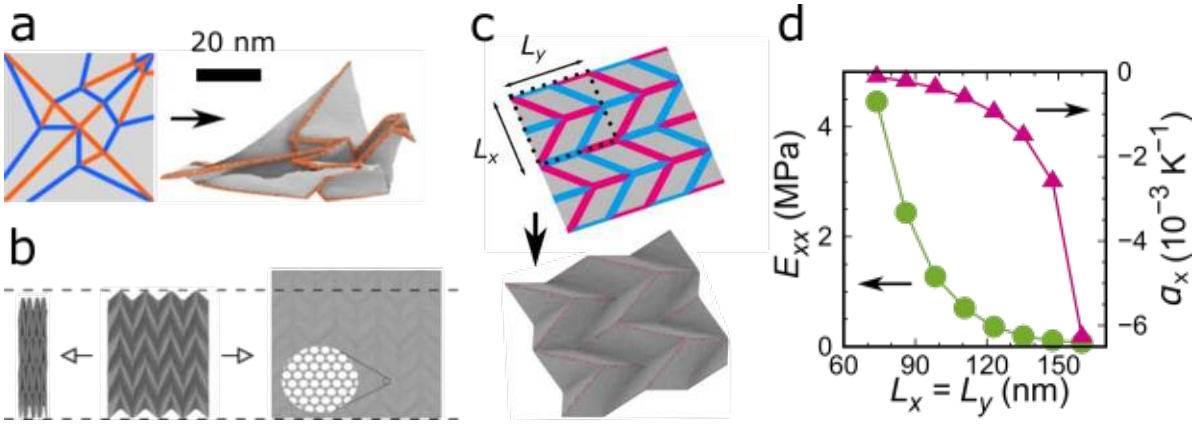

Figure 1. (a) Patten-based hydrogenation for the formation of a graphene flapping bird origami. Hydrogenation is conducted on the top of graphene in the areas coloured in red and on the bottom of graphene on the area coloured in blue. (b) Super compressibility and stretchability and negative Poisson's ratio behaviour of a graphene Miura origami structure. (c) The pattern-based hydrogenation of a Miura graphene origami. (d) Coefficient of thermal expansion α_x and Young's modulus E_{xx} as functions of $L_x = L_y$ defined in (c).

Band alignment and interlayer hybridisation in transition metal dichalcogenide/hexagonal boron nitride heterostructures

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In van der Waals heterostructures, the relative alignment of bands between layers, and the resulting band hybridisation, are key factors in determining a range of electronic properties. This work[1] examines these effects for heterostructures of transition metal dichalcogenides (TMDs) and hexagonal boron nitride (hBN), an ubiquitous combination given the role of hBN as an encapsulating material. We compare results of linear-scaling density functional theory (DFT) calculations using large low-strain supercells with experimental angle-resolved photoemission spectroscopy (ARPES) results. We explore the hybridisation between the valence states of the TMD and hBN layers, and show that it introduces avoided crossings between the TMD and hBN bands, with umklapp processes opening 'ghost'[2] avoided crossings in individual bands. Comparison between DFT and ARPES spectra for the $MoSe_2$/hBN heterostructure (Fig. 1) shows that the valence bands of $MoSe_2$ and hBN are significantly further separated in energy in experiment as compared to DFT. We then show that a novel scissor operator can be applied to the hBN valence states in the DFT calculations, to correct the band alignment and enable quantitative comparison to ARPES, explaining 'ghost' avoided crossings and other features in the ARPES spectra.

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Figures

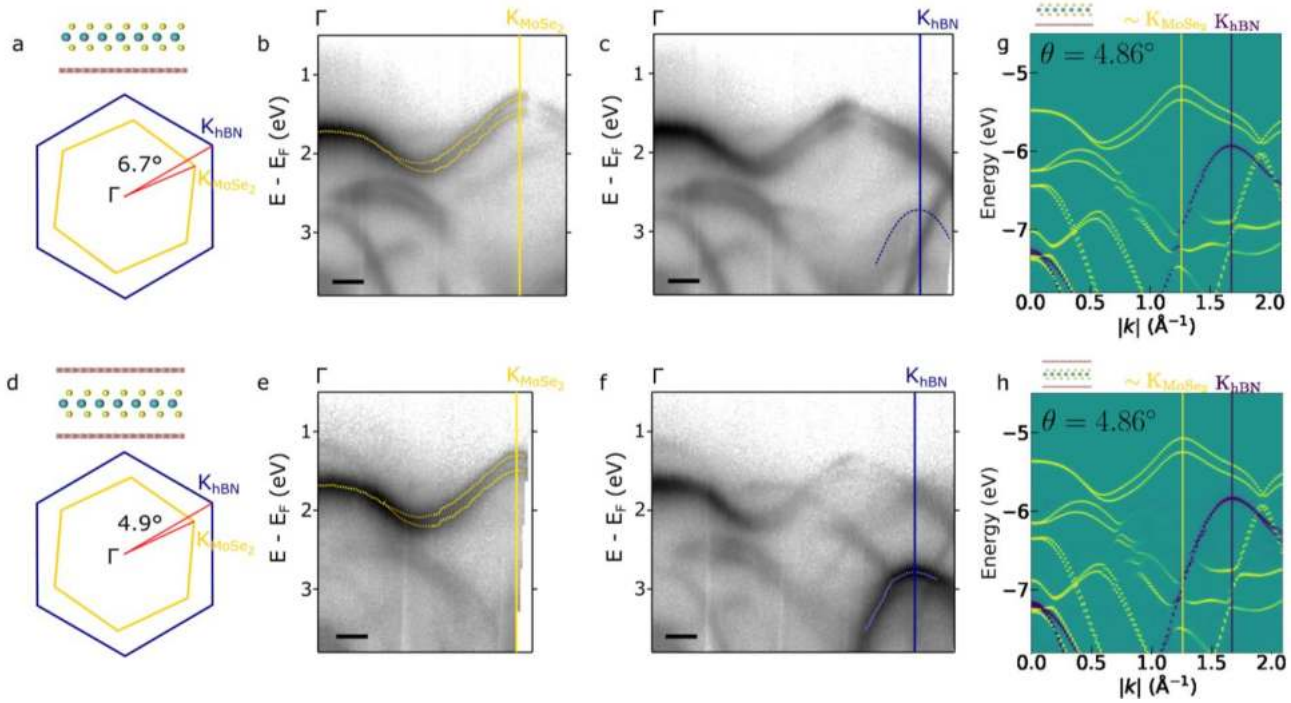

Figure 1: Comparison of ARPES and unfolded DFT spectra (with no scissor correction applied at this stage) for $MoSe_2$/hBN heterostructures. (a) Schematics of $MoSe_2$/hBN heterostructure and of $MoSe_2$ and hBN Brillouin zones. (b) and (c) slices from ARPES spectra along K ($MoSe_2$) and K (hBN) directions, respectively. (c)-(f) as (a)-(c), but for a region of the heterostructure with hBN also on top of the $MoSe_2$. These are shown with LS-DFT spectra for $MoSe_2$ nearly aligned with hBN, for monolayer hBN on one side (g) and both sides (h) of $MoSe_2$.

Theory of plasmon-magnon coupling in 2D honeycomb magnets

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Two-dimensional (2D) materials have been investigated for almost two decades [1] and have been the source of truly spectacular discoveries [2]. Magnetic 2D materials, in particular, have been recently discovered [3,4] and are currently representing a very active and interesting research field[5]. In particular, two-dimensional honeycomb ferromagnets offer the unprecedented opportunity to study interactions between collective modes that in standard bulk ferromagnets do not cross paths. Indeed, when doped with free carriers, they also host the typical gapless plasmonic mode of 2D itinerant electron/hole systems. Moreover, they display an optical spin-wave branch which disperses weakly near the Brillouin zone centre. The plasmon branch, eventually, meets the optical spin-wave branch at a certain energy and momentum, paving the way for interactions between the charge and spin sector. In this talk we present a microscopic theory of such plasmon-magnon interactions, which is based on a double random phase approximation[6]. We show that plasmon-magnon interactions do not require spin-orbit coupling to exist, and they naturally arise from the exchange interaction between the itinerant carrier and the localized magnetic moments. We will also discuss on the possibility to unveil this physics in recently isolated 2D honeycomb magnets such as Cr2Ge2Te6[7].

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Probing minibands in gated graphene superlattice by magnetic focusing

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Graphene is considered an excellent material for the creation of electronic devices with ballistic transport occurring in presence of external electrostatic potential. That makes it advantageous as a base for superlattices induces by gating. The resulting periodic modulation of on-site energy leads to the reconstruction of the band structure and formation of mini bands and higher-order Dirac cones. Gated superlattices are particularly interesting thanks to tunable modulation strength and flexibility in defining the geometry. At low magnetic field, semiclassically, fermions follow cyclotron trajectories that reflect the Fermi contours, and can be probed by transverse magnetic focusing. This technique has been used for investigating band structures in moire superlattices [2, 3]. We perform a theoretical study of the magnetic focusing in 2D rectangular gated superlattices, and analyze the relation between the observed magnetotransport spectra and the miniband structure. Our investigations of the reconstructed band structure via magnetotransport calculations pave the way to band structure engineerging through periodic gating of graphene.

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Controlling charge density order in 2H-TaSe2 using a van Hove singularity

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The understanding and manipulation of correlated states of matter like superconductivity or ferromagnetism are amongst the principal challenges in physics. From magnetic phases, high-temperature to topological Kagome superconductors and magic-angle twisted bilayer graphene, the correlated states often appear alongside a high density of electron states induced by van Hove singularities (vHs) [1-5]. Here, we report on the interplay between a vHs and a charge density wave (CDW) state in 2HTaSe$_2$. We use angle-resolved photoemission spectroscopy to investigate changes in the Fermi surface of this material under surface doping with potassium. At high doping, we observe modifications which imply the disappearance of the (3×3) CDW and formation of a different correlated state. Using a tight-binding-based approach as well as an effective model, we explain our observations as a consequence of coupling between the single-particle Lifshitz transition, during which the Fermi level passes through a vHs, and the charge density order. The high electronic density of states associated with the vHs induces a change in the periodicity of the CDW from the known (3×3) to a new (2×2) superlattice [6]. Our observation of the (2×2) phase validates a prediction from almost 50 years ago: we present the first spectral evidence of saddle-point nesting-driven CDW in transition metal dichalcogenides as originally proposed [1]. Moreover, the tunability of our system opens a new avenue to explore the interrelationships between CDW, van Hove singularities and superconductivity.

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Piezoelectricity in Magnetic 2D Materials

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Piezoelectric materials convert mechanical energy to electrical energy, and vice versa. A wide range of electromechanical devices relies on piezoelectric-based energy conversion. Being intrinsically nano-size, flexible two-dimensional (2D) piezoelectric materials can miniaturize these electromechanical devices e.g. nanoscale sensors, energy harvesters, and actuators. However, a large piezo-response is desired for any practical applications. Recently, based on Density Functional Theory (DFT) calculations, we have predicted that ferroelectric/multiferroelectric MOX_2 (M=Ti, V and X = F, Cl, Br) monolayers possess large in-plane stress (e_{11}) and strain (d_{11}) piezoelectric coefficients[1]. For example, $TiOBr_2$ monolayer has approximately one order of magnitude larger in-plane piezo-response ($e_{11} = 28.793\times10^{-10}$ C/m and d_{11} = 37.758 pm/V) than the widely studied piezoelectric 1H-MoS_2 monolayer. Furthermore, MOX_2 monolayers exhibit large d_{11} coefficient ranging from 29.028 pm/V to 37.758 pm/V, significantly higher than the d_{11} or d_{33} of traditional 3D piezoelectrics such as *w*-AlN (d_{33} = 5.1 pm/V) and *α*-quartz (d_{11} = 2.3 pm/V)[1]. MOX_2 monolayers possess a large d_{11} because of their low in-plane elastic constants and large e_{11}. Large Born effective charges (Z_{ij}) and atomic displacement in response to an applied strain ensure a large e_{11}. Note that multifunctional spintronic devices can utilize coupling between piezoelectricity and magnetism in 2D materials. However, piezoelectricity requires a non-centrosymmetric structure with an electronic band gap, whereas magnetism demands broken time-reversal symmetry. Most of the well-known 2D piezoelectric materials, e.g., 1H-MoS_2 monolayer, are not magnetic. Being intrinsically magnetic, semiconducting 1H-$LaBr_2$, 1H-VS_2 and VOX_2 monolayers can combine magnetism and piezoelectricity. We show the possibility of opening a new way of controlling piezoelectricity by changing the magnetic order such as changing antiferromagnetic to ferromagnetic, or vice versa. For example, a change in magnetic order can enhance (reduce) the piezo-response of 1H-$LaBr_2$ (1H-VS_2)[2].

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Predicting magnetic edge behaviour in graphene using neural networks

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A large number of proposed spintronic devices are predicated on the formation of local magnetic moments near the edges of graphene flakes and ribbons [1], and recent experimental progress allows high-precision edges to be engineered [2].
Simulations play a key role in both interpreting experimental measurements and confirming the presence of desired magnetic behaviour.
However, computational costs prevent the simulation of large-scale disorders that can occur in experiment and could quench the desired behaviour.
We have developed a machine-learning approach which removes this computational bottleneck [3].
I will discuss its performance on a range of geometries, and show how spin currents in graphene nanoribbons unexpectedly survive in the presence of long-ranged edge roughness [4].

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Figures

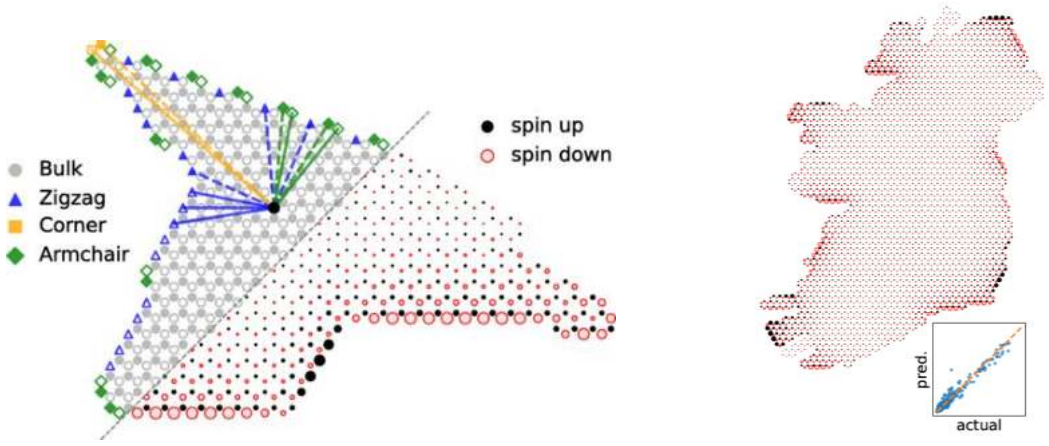

Figure 1: (Left:) Breakdown of sites in a graphene flake by sublattice, edge type and moment magnitude. (Right:) Neural network predictions of moments on an unseen geometry.

Computational Screening for Sustainable Two-dimensional Ultra-wide Bandgap Materials

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Abstract

Presently used ultra-wide bandgap semiconductors comprise predominantly bulk materials, but their large size, high power and high cost is an obstacle to meeting the future requirements of high-performance (opto)electronic devices [1]. Ultra-wide bandgap two-dimensional materials serve as a promising solution to meet these needs. While the rise of materials databases specific to two-dimensional materials provides us with many options to choose from, various considerations, such as the difficulty of synthesizing these materials and the fact that certain elements are under serious supply threat in the near future while some others are environmentally harmful [2, 3], complicate how we choose new candidate materials for such applications. We devised a strategy to screen for sustainable, easily exfoliable and stable candidate two-dimensional materials from the 2DMatPedia database [4] for various (opto)electronic applications. We assessed the screened candidate materials for their performance in specific (opto)electronic device applications using density functional theory (DFT) and related first-principles methods. The properties computed include the HSE06 band alignments, optB88 static dielectric constants and GW-BSE optical spectra, besides some properties from transport simulations. These calculations inform us of the potential for these candidate materials to be used as transistor materials, specifically gate dielectrics and channel materials, as well as for (polarization-sensitive) ultraviolet photodetection.

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Exciton optics and dynamics in organic and organic/TMD heterostructures

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Organic semiconductors are exciting candidates for photovoltaic and biosensor technologies. They are cheap, flexible, relatively easy to fabricate and posses excellent light-matter coupling, crucial for a host of application. Currently however there are limitations preventing these materials reaching the level of efficiency of competing devices. These materials possess low mobilities and cannot be easily mechanically modified by strain. These limitations can be overcome by interfacing organic crystals with transition metal dichalcogenides (TMDs). In this joint theory-experiment work, we study the exciton landscape in organic molecular crystals as well as organic/TMD heterostructures, with a particular emphasis on their optical response. We demonstrate both theoretically and experimentally that the low-temperature photoluminescence (PL) is dominated by the formation of interlayer excitons, with the electron and hole located on the TMD and molecule layer, respectively [1]. We find that additional sidebands emerge as a result of the phonon-mediated indirect recombination of excitons. Furthermore, we study the exciton landscape within an organic crystal layer focusing on tetracene and pentacene crystals, We find a unique polarisation- and temperature-dependence of absorption and PL spectra, stemming from the Davydov-split excitons [2]. We describe the exciton dynamics in these crystals, where the flatness of the exciton bands gives rise to phonon-bottlenecks in the exciton relaxation, where momentum-indirect excitons offer crucial relaxation channels. We then turn our attention to energy transfer processes in organic/TMD heterostructures, which is governed by the Förster interaction, and explore the resulting signatures in the differential absorption and PL spectra. Our joint theoretical-experimental work unveils the behaviour of excitons in organic TMD heterostructures, and sheds light on the optical and dynamical response of these materials, which is imperative for understanding and designing future device architectures.

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Figures

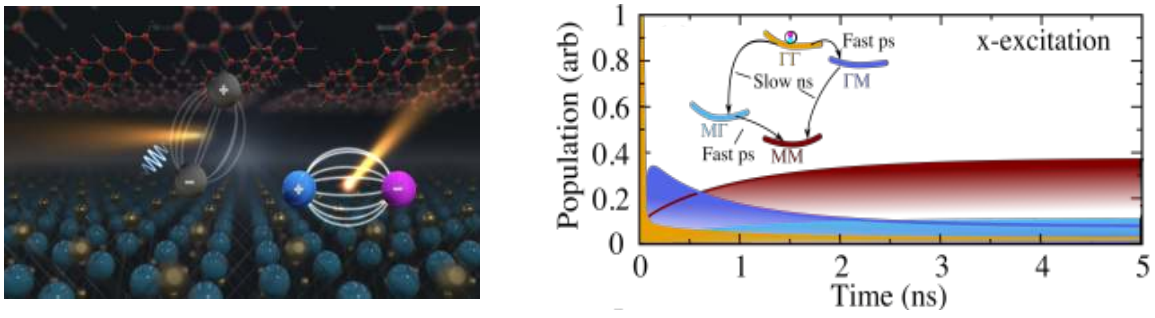

Figure 1: (Left) Artist's illustration of inter- & intralayer exciton landscape in an organic/TMD heterostructure. (Right) Exciton relaxation dynamics in an organic semiconductor.

Charge and spin transport in 2D Rashba system

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Abstract
The microelectronics revolution is driven by the motto 'the smaller the better', as it aims to miniaturize electronic components and improve performances. Nowadays silicon transistors are approaching the scaling limit. Recently, the MoS_2 transistor with an atomically thin channel and a gate length of sub-1-nm has been fabricated successfully [1], illustrating the potential of 2D materials for applications in electronic devices. In addition to the electron, spin is another degree of freedom to manipulate the logic operations, and the spintronic devices have high-density, low-power, and non-volatile advantages [2]. Using *ab initio* calculations, we demonstrate the superior charge and spin transport properties in 2D buckled III-V semiconductors. Due to the broken inversion symmetry, Rashba splitting is revealed in the conduction bands, where high electron mobilities over 1400 $cm^2/(V{\cdot}s)$ are predicted [Fig. 1(a)]. This can be explained by the negligible scattering on the single Γ valley [Fig. 1(c)]. In contrast, the hole mobility [Fig. 1(b)] is lower due to the strong scattering on multiple K valleys [Fig. 1(d)]. Fig. 2(a) presents the spin Hall conductivity (SHC), and the universal SHC are identified in Rashba systems. More significantly, via heavy hole doping, the semiconductors are turned to be metallic systems, where efficient spin-charge conversions are discovered [Fig. 2(d)], which can reduce the writing power in spin-orbit torque devices. This work highlights the promising application for 2D Rashba systems in electronic and spintronic devices.

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Figures

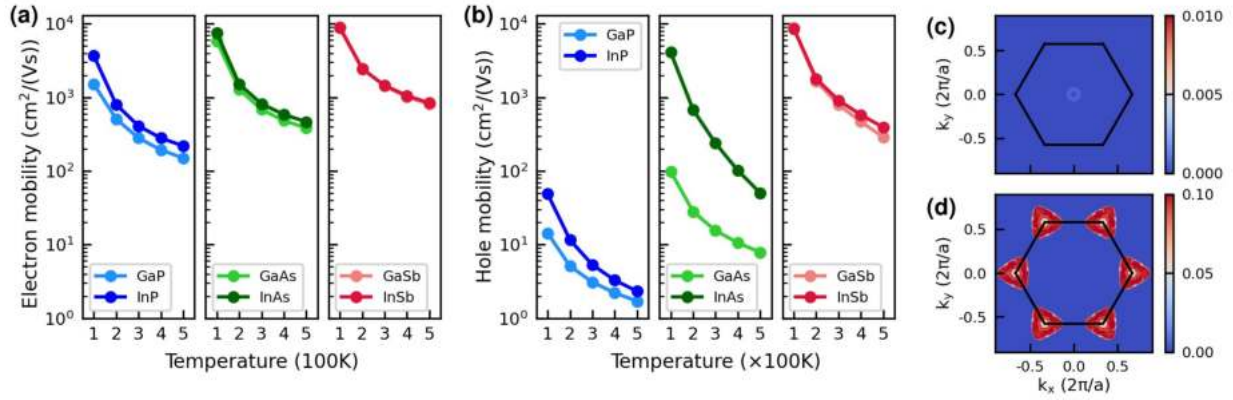

Figure 1: (a)-(b) Temperature-dependent electron and hole mobilities of 6 III-V monolayers. (c)-(d) **k**-resolved scattering rate of InSb electron transport and GaP hole transport, respectively.

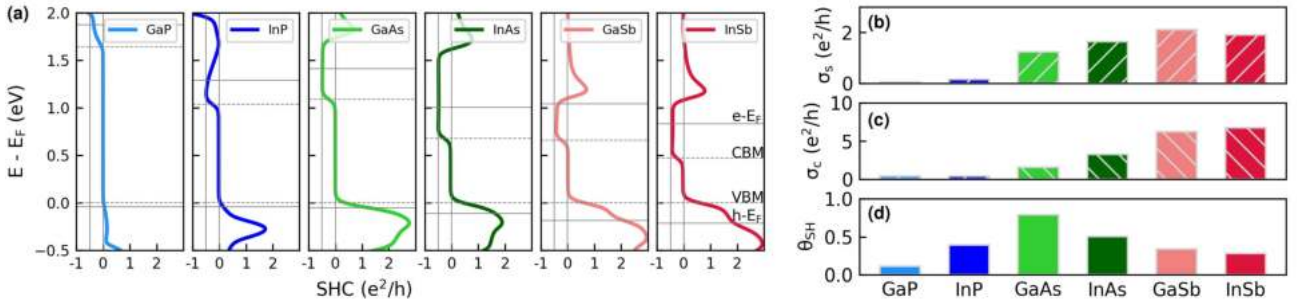

Figure 2: (a) Spin Hall conductivities of 6 III-V monolayers. The dashed lines denote the conduction band minimum and valence band maximum of intrinsic semiconductors, and the solid lines indicate the position of Fermi energy after electron doping and hole doping, respectively. (b)-(d) The spin Hall conductivity, charge conductivity, and spin-charge-conversion efficiency after hole doping of $2{\times}10^{13}$ cm^{-2}.

Solution Processing of Low-dimensional Materials and Applications

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Low-dimensional Materials like graphene, transition metal dichalcogenides, h-BN, transition metal oxides and layered double hydroxides possess the potential for applications across various fields. Liquid phase exfoliation of these layered crystals enables the solution processing of dispersions of mono- and few-layers, and provides a scalable viable alternative to other physical and chemical routes. The quality of the dispersions and their applicability are dependent on the exfoliation and stabilization of the exfoliated material by the solvent, often chosen on the basis of Hansen solubility parameters (HSP). In this work, the factors at play in liquid phase exfoliation besides HSP are explored via various experimental methods, in order to further enhance the versatility of the process by providing a deeper insight. By considering molecular aspects of the solvents, highly concentrated nanosheet dispersions were obtained in a low boiling point solvent. I will also discuss about experimental determination of the HSP of layered materials. I will be concluding my talk by discussing some of our recent efforts in exfoliating non-layered materials and applications of the 2D dispersions in various fields like flexible electronics, energy storage/conversion devices and electrochemical biosensors.

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Opto-evoked neurotransmitter release detection with graphene aptasensor multitransistor arrays

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Neural interfaces, bridging brain physiology and external electronic devices, allow brain function monitoring for neurochemistry research or clinical purposes. Despite recent advances in monitoring electrical brain activity, measuring chemical neurotransmission remains a significant challenge. We recently developed a platform for robust and ultrasensitive detection of dopamine [1], an essential neurotransmitter that underlies several brain disorders, based on graphene multitransistor arrays (gMTAs) functionalized with a selective DNA aptamer. Not only did we achieve the lowest limit-of-detection ever reported (1 aM), but we could also detect dopamine with great sensitivity in complex samples such as artificial cerebral spinal fluid and brain homogenate, including in a mouse model of Parkinson's Disease. Herein, we present a novel nanobioelectronic neural interface based on gMTAs that allows monitoring of neurotransmitter opto-evoked release in ex vivo brain slices of transgenic mice. Optimizing our gMTAs' fabrication process [1,2], we developed an interface with higher sensor density that accommodates mice's brain slices and allows optogenetic modulation through integrated micro-LEDs. The platform presented in this work can lead the way to novel neurotransmitter sensors suitable for real-world academic and pre-clinical pharmaceutical research and clinical diagnosis.

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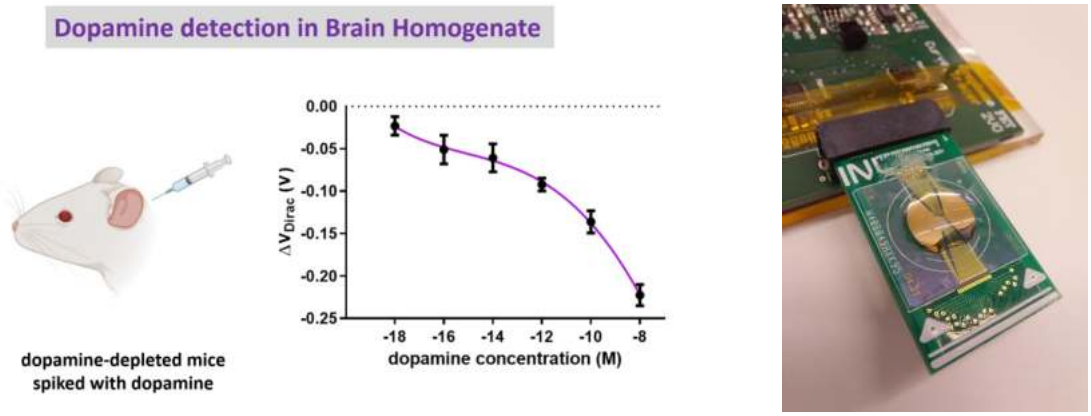

Figure 1: Dopamine detection in brain homogenate with gMTAs (left). Graphene nanobiosensing platform for opto-evoked neurotransmitter detection (right).

Graphene-Device Operated in DC Transistor and AC Electrochemical Modes for DNA-sensing and Molecular Characterization

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Abstract
Graphene, a carbon-atomic monolayer, due to its unique electrical and electronic properties, such as its ambipolar transport and ballistic electron dynamics, is considered the background material for developing different technological applications,[1] in particular biosensing. Thus, graphene-based setups and experimental approaches that allow fast and simple access to its quantum properties are required to advance in the design of high-performance devices. In this regard, non-destructive methodologies based on electrical perturbations offer the possibility of accessing graphene's electronic structure (V-shaped DOS) in mild experimental conditions and using this response in applications.[2] In this line, we present a device comprising 20 graphene functionalized channels per chip (L= 25 µm and W= 75 µm) that can be operated in two modes: 3-terminal DC transistor and 2-terminal AC electrochemical setup. Both setups resolved the graphene electronic structure, used as a transduction signal to detect a single-stranded DNA-biomarker (tDNA). In DC transistor mode, the device detected tDNA down to the attomolar range (~10 aM) by following the charge neutrality point voltage in the transfer curves (Fig.1a). In the AC electrochemical mode, the signal was obtained from impedance spectroscopy by monitoring the minimum of the graphene quantum capacitance as a function of a superimposed DC bias potential,[3] with a limit-of-detection of 1 aM (Fig.1b). Another exciting application for the electrochemical mode was tested, which reveals the electronic structure of π-conjugated molecules[4] stacked over graphene through the quantum capacitive response of this latter (Fig.1c).

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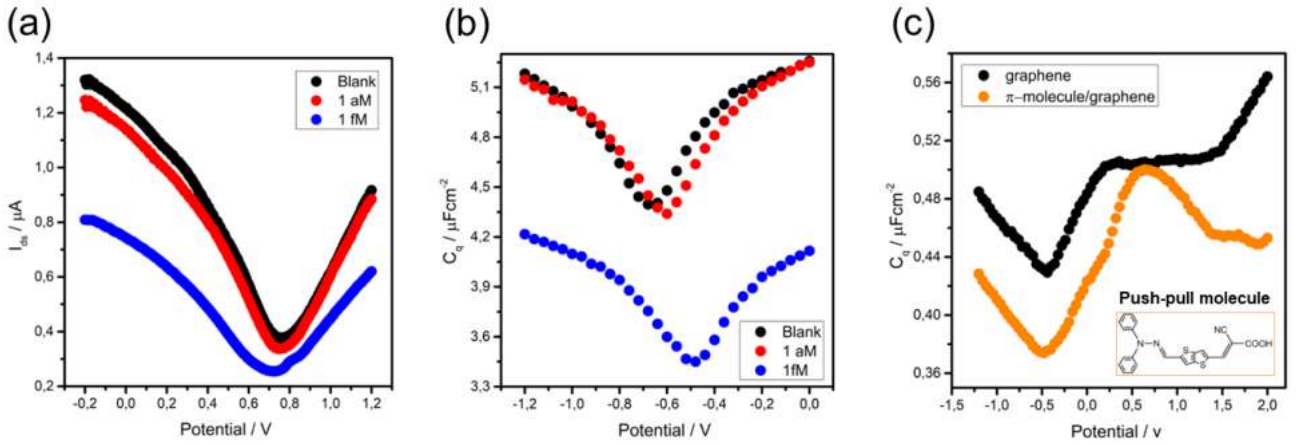

Figure 1: (a)Transfer curves obtained in DC transistor mode, (b) quantum capacitance response in functions of the electric potential resolved in AC electrochemical mode for two concentrations of target-DNA in PBS. (c) The electronic structure effect of the push-pull π-conjugated molecule appears as a modulation of graphene quantum capacitance trace.

Pulmonary toxicity of boron nitride nanomaterials: a comparison between two-dimensional sheets and nanotubes of similar composition and purity

Cyrill Bussy

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Background: Boron nitride (BN) nanomaterials have recently drawn a lot of interest in the material community owing to interesting properties. Nevertheless, their biosafety has yet to be confirmed in vivo. Herein, we investigated the biological impact and clearance of two-dimensional hexagonal boron nitride nanosheets (hBN) and boron nitride nanotubes (BNNT) of similar purity in mouse lungs.
Methods: Mice were exposed by single pharyngeal aspiration to 30 µg of either hBN or BNNTs. At days 1, 7, and 28, bronchoalveolar lavage (BAL) fluids and lungs were collected. The pulmonary adverse effects were evaluated (immune response, histopathology, tissue remodelling, genotoxicity) and put in perspective with materials' accumulation, distribution and clearance from the lungs.
Results: hBN did not cause any significant immune response or lung damages in the exposed mice, despite the presence of materials confirmed by Raman spectroscopy. Moreover, hBN nanosheets were found in alveolar phagocytes, resulting in an efficient clearance from the lungs over time. Conversely, BNNTs caused a strong and chronic inflammatory response, characterized by a sustained inflammation up to 28 days after exposure, as well as the activation of both the innate and adaptive immunity. These responses could be ascribed to the poor clearance from the lungs and the high aspect ratio of BNNTs. Additionally, we observed granulomatous structures as well as fibrosis. However, we did not observe significant DNA damages after performing global lung analysis. Further analysis is ongoing to check if significant DNA damages could occur in the inflammatory areas of the lung.
Conclusion: Despite a similar chemical composition and purity, we demonstrate the safer toxicological profile of BN nanosheets in comparison to BN nanotubes. We also reveal the strong similarities in the lung response to BNNTs and multi-walled carbon nanotubes (MWCNTs), highlighting that the high aspect ratio is a major driver of pulmonary response to nanomaterials.

Wearable Sensors for Breath Monitoring made with Water-based Hexagonal Boron Nitride Inks via Supramolecular Functionalization

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Smart wearable humidity sensors are attracting strong attention as they enable to monitor important physiological information in real time, such as pulse oximetry, and enable activity tracking and air quality assessment [1]. Two dimensional (2D) materials, especially graphene oxide (GO), have triggered strong research interest for humidity sensing due to their tuneable surface chemistry, high surface area, ultrathin thickness, processability in water and easy integration onto flexible substrates [2]. However, large hysteresis, low sensitivity and strong cross-sensitivity issues limit the use of GO for practical wearable applications [3], where continuous monitoring is needed. Herein, we demonstrate a wearable and wireless impedance-based humidity sensor made with functionalized hexagonal boron nitride (h-BN) nanosheets [4], which shows enhanced sensitivity (>10^{10} Ohms/%RH from 5% to 100% RH) and fast response (0.1 ms) [5]. We finally show that the sensor is able to record in real time the subtlest changes of respiratory signals associated with different daily activities as well as various symptoms of flu, without any direct contact with the individual [5].

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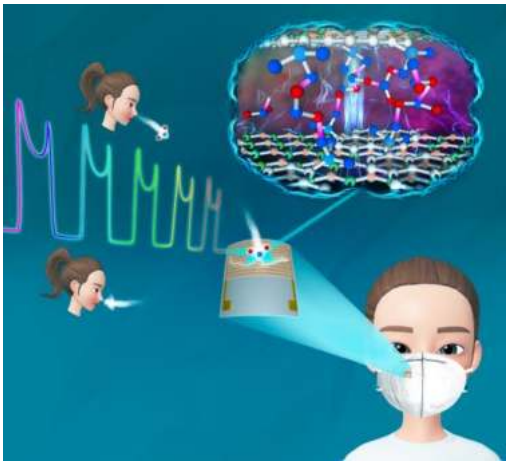

Graphical abstract

Graphene and WS_2 interactions with neutrophils and MSCs for nerve injury regeneration

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The employment of graphene and 2D materials in tissue engineering has been recently exploited for the repair and regeneration of nerve tissue. Among the possible applications, these relatively new materials display a great potential as peripheral neural interface, especially thanks to their unique combinations of electrical, optical and tribological properties [1-5].
However, the use of these innovative materials has raised questions about their interaction with immune cells, including neutrophils, whose immune response is known to influence the regenerative outcome [6], and mesenchymal stem cells (MSCs), a novel therapeutic avenue for peripheral nerve regeneration [7].
In this work we investigated graphene and WS_2 influence on neutrophils and MSCs. We tested WS_2 on sapphire and different CVD graphene, namely graphene on sapphire, graphene on SiC, both as-grown and H-intercalated, graphene grown on copper and transferred on glass, to assess the effect of the substrate and growth technique.
We first characterized neutrophil activation and discussed how material properties influenced the NETs production and their adhesion to the substrates. Furthermore, planar graphene resistance to NETs-induced degradation was carefully investigated and compared with the results reported for graphene oxide [8]. Ultimately, 2D materials cytocompatibility for MSCs are tested to estimate cell viability, morphology and mitochondrial health.
Overall, our results are aimed at understanding the interface between 2D material and some of the players involved in nerve injury, a critical point for regenerative medicine.

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Figures

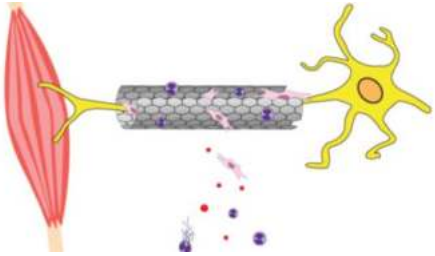

Figure 1: Graphene conduit for nerve regeneration

TAILORED CHEMICAL DESIGNS FOR GRAPHENE FETS IN BIOSENSING

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Graphene field effect transistors (GFETs) have gained prominence in the field of biosensing due to their high sensitivity and selectivity, low detection limit and ability to function *in vivo*.[1] This is mainly due to the high carrier mobility of graphene and its added properties, such as biocompatibility, transparency, and flexibility. Thus, GFETs are able to detect various biomolecules, including proteins, DNA, and small molecules, with high specificity and sensitivity in a variety of media. The mandatory functionalization of graphene with different receptors or biorecognition elements has been achieved using various chemical approaches, (*i.e.*, covalent binding, non-covalent binding, and electrostatic adsorption). However, the challenge of finding the best immobilization strategy for the receptor remains, as not all graphene chemistry strategies can be easily adapted to transistor modification.
By controlling graphene functionalization and tuning the device design, we have developed diverse GFET microarrays to detect small molecules, such as neurotransmitters and air pollutants; and viral proteins with an extremely low limit of detections.[2-3] These results could establish the basis for a new category of analytical platforms based on well-defined graphene modification. Such platforms would have the potential to detect a wide range of pathogens and biomarkers even before their isolation. This capability could prove useful in health and environmental monitoring, as well as in fighting future pandemics.

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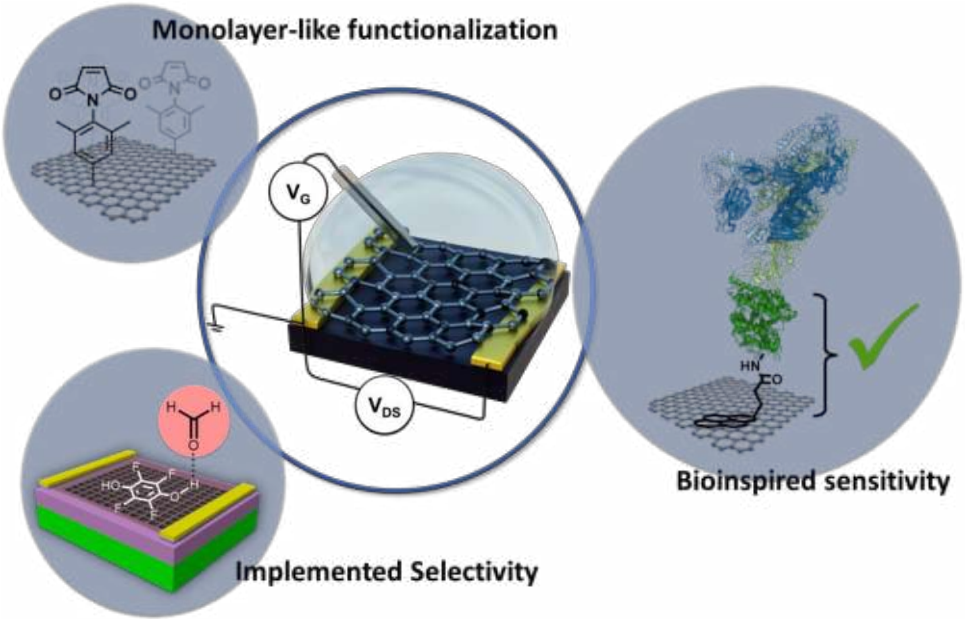

Figure 1: Schematic representation of diverse designs used in GFET sensors.

Graphene nanoelectronics meets neurofluidics for versatile labs on chip

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New methods and new technology are currently required to interrogate neuronal cells by many means and at multi-scale, in-vivo and within model neural networks in-vitro. In particular, to understand how neural circuits operate, we need access to activity of large numbers of neurons at the same time, and record their activity at the single cell level regarding the lot of information which relies at the level of synapses and ion channels. In that race, Graphene offers an ideal platform for recording and culturing neural networks, regarding its exceptional neuronal affinity and the presence of readily accessible surface charges which give the unprecedented possibility to realize a direct coupling with cells to detect ion fluxes at the nano[1,2] and mesoscale.[3,4] Here, we report on a novel and versatile approach that combines array of graphene field effect transistors (GFET) and microfluidic platforms for culturing and sensing neurons in designable network architecture.[5] The fluidic microchannels, somatic and synaptic chambers enable to define the neuron network topology, while the graphene devices provide localized, highly sensitive and optically transparent sensing sites. The efficient cell-sensor alignment obtained by the microfluidic circuit enables to reach the highest reported signal-to-noise ratio for single-units detection with GFETs, revealing additional information that remain hidden from recordings when using conventional microelectrode arrays (MEAs). Thus, the combination of graphene sensors and microfluidic circuits leverages the advantages of two state-of-the-art technologies for highly efficient sensing of model neural networks. Being fully transparent and therefore compatible with optogenetic tools and high-resolution microscopy, this novel platform could provide a versatile lab-on-chip for diagnosis and treatment of tomorrow, and open avenues of investigation for studying topological neuron network and living matter in general.

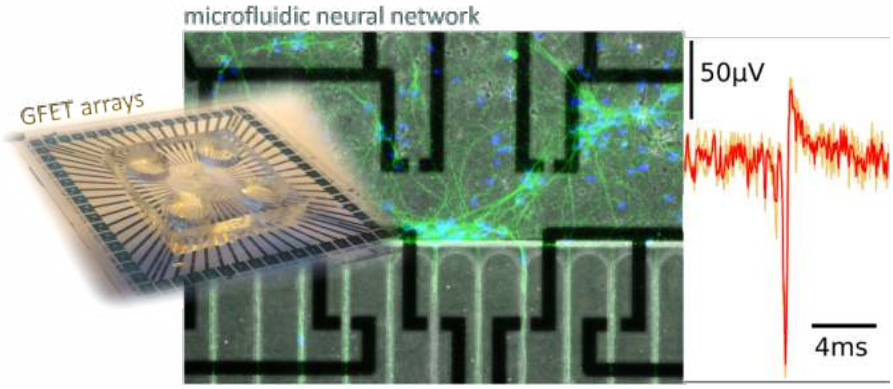

Figure 1: Neuron-gated GFET arrays with microfluidic circuits (left) allows for highly efficient extra-cellular detection of action potential (right). The graphene sensing site being optically transparent, cells can be observed in real-time during the culture (several weeks) providing multiple way to follow both structural and functional changes within model neural network (center).

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Conductive railways on graphene wrinkles

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Since its discovery, lot of effort has been devoted to harness the exotic properties of graphene with the quest of nanoscale engineering at the forefront. Graphene growth by Chemical Vapor Deposition (CVD) takes place mainly on top of metal catalytic substrates, but due to the high temperatures and the mismatch in thermal expansion coefficients of graphene and the growth substrate, out-of-plane deformations in the form of wrinkles are unavoidable [1]. Furthermore, in order to exploit graphene, it needs to be transferred onto other arbitrary substrates by the means of wet and dry methods, which induces further wrinkling. Wrinkles are commonly treated as defects which degrade the overall properties of graphene, hence routes for smoothening these structural glitches are under continuous investigation [2]. Nonetheless, the inevitable presence of wrinkles has offered many interesting pathways to be explored [3].
In the present work, we visualize for the first time the spatial distribution of local conductivity in monolayer graphene and its wrinkle network arising naturally from synthesis and subsequent transfer. Atomic force microscopy (AFM) tapping current measurements reveal that the conductivity of wrinkled graphene, on top of the wrinkle structure, is up to 2 *orders of magnitude* higher relative to that of flat graphene. Furthermore, we discuss the impact of the substrate on the electronic properties of graphene and electronic transport through wrinkles. Computations within the framework of density functional theory and theoretical treatment suggest that the observed contrasts of local conductivity qualitatively correlate with the off-plane electric susceptibility differences between wrinkled and flat graphene monolayers. The findings in this work unravel the implications of graphene wrinkles as electrical conduits, while establishing that we are still at infancy in grasping the possibilities of multifaced nanoscale engineering.

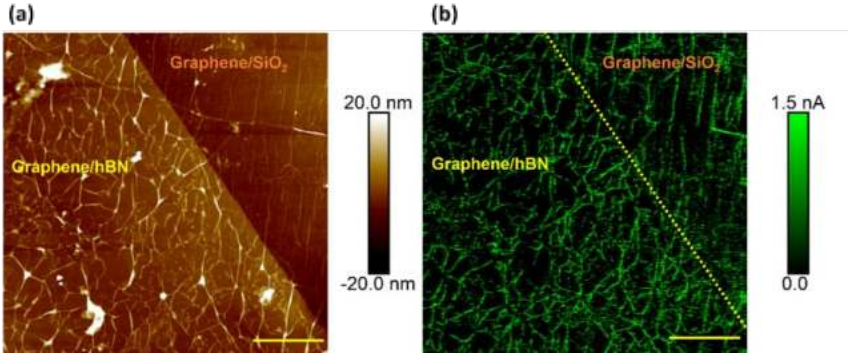

Figure: (a) Topography of CVD graphene transferred on top exfoliated 10-layer hBN through wet transfer. (b) Current mapping of CVD graphene on 10-layer hBN and SiO_2/Si areas. Dashed yellow line denotes the edge of the graphene/hBN heterostructure. Scale bars are 2 μm.

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Structural Characterisation of Liquid Phase Exfoliated Transition Metal Dichalcogenides – An Electron Microscopy Study

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Transition Metal Dichalcogenides (TMDs) have risen to popularity due to their range of bandgaps and variety of applications [1]. Platinum diselenide ($PtSe_2$), in particular, has shown many promising characteristics such as high room-temperature mobility, strong layer-dependent band structures and high stability in the air [3,4] presenting opportunities for applications in high-speed sensors and opto-electronic devices [5]. In pursuing novel materials, in-depth characterization is vital and defect studies need to be well established when considering future device applications. In this regard, electron microscopy has shone through as an essential tool in thorough structural and chemical characterisation. In this study $PtSe_2$ was exfoliated through sonic probe Liquid Phase Exfoliation (LPE). Few layer flakes were achieved and subsequently characterized. Flake morphology, point defects and stacking sequences were investigated using various electron microscopy techniques. Transmission electron microscopy (TEM) and Selected Area Electron Diffraction (SAED) was performed using a FEI Titan 80-300 Thermo Fisher Scientific to observe crystal orientation and morphology. Scanning Transmission electron microscopy (STEM) images were recorded with a high angle annular dark-field (HAADF) detector on a Nion Ultra STEM, for atomic resolution structural characterization and point defect analysis. Energy Dispersive X-Ray Spectroscopy (EDS) was used for chemical analysis while Electron Energy Loss Spectroscopy (EELS) allowed for the estimation of the monolayer bandgap.

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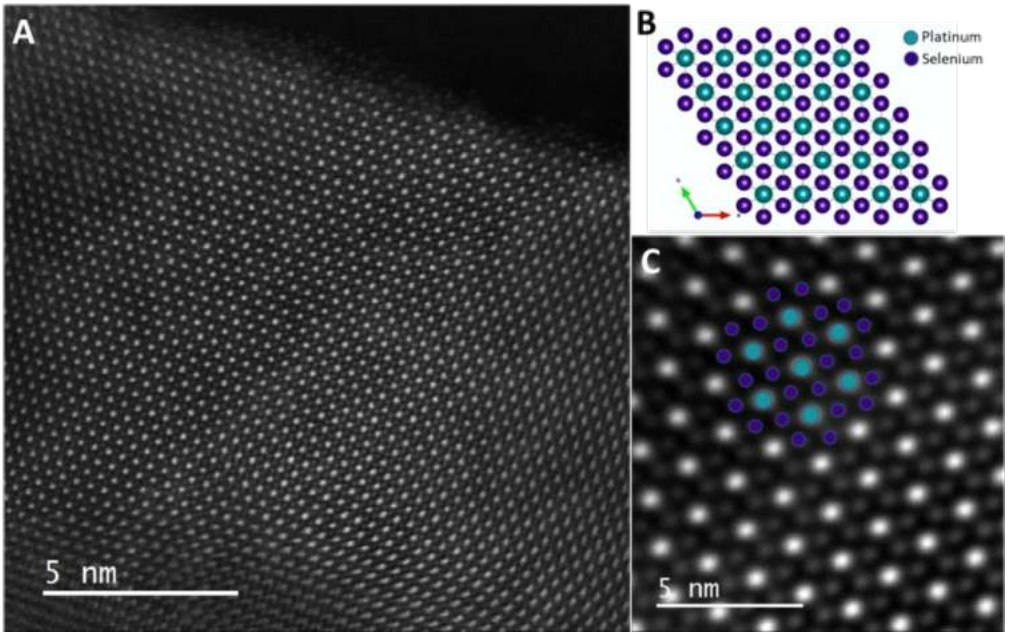

Figure 1: (a) Atomic resolution HAADF image of LPE $PtSe_2$ (b) Model of $PtSe_2$ preferential 1T structure (c) Z-contrast showing clear 1T structure of $PtSe_2$.

Ellipsometry Study of Hexagonal Boron Nitride Grown on CMOS-compatible Substrates via CVD

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Hexagonal boron nitride (hBN) has a range of promising applications, including deep ultraviolet optoelectronics and passivation layers for high-mobility graphene. For integration into Si technology, growth of hBN thin films directly on CMOS-compatible substrates, such as silicon, germanium or dielectrics, is desirable. In particular, germanium has proven suitable for chemical vapor deposition (CVD) growth of high-quality 2D materials [1], due to its catalytic activity. In fact, low pressure CVD growth of polycrystalline hBN monolayers on Ge using ammonia borane as the precursor has been reported previously.[2] We grow few-layer hBN thin films on epitaxial Ge(001)/Si substrates via CVD, using borazine as a single-source precursor.[3] Characterization of the grown films is of utmost importance for growth studies, but can be quite challenging in the case of 2D materials. Transmission electron microscopy (TEM) analysis provides comprehensive insight into the crystalline structure and revealed a film thickness of 1-5 nm, dependent on growth time and borazine partial pressure, with an interlayer distance of 3.35 Å (Fig. 1a). However, TEM analysis is very time consuming and higher throughput methods are needed. Here, we focus on multiangle spectroscopic ellipsometry (SE) in the DUV-VIS range to assess thickness and optical constants of hBN. First SE investigations show that hBN films grown on Ge(001)/Si can be detected starting at ~2 nm thickness. The obtained ellipsometry spectra are fitted using a Tauc-Lorentz oscillator model and the refractive index n and extinction coefficient k are extracted, see Fig. 1b-c. Noticeably, the extinction coefficient near the fundamental absorption edge (>5 eV) indicates a good quality material with a bandgap close to 6 eV. The refractive index at 633 nm (1.96 eV) is in the range of 1.7-2.5, depending on the specifics of the samples.

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Figures

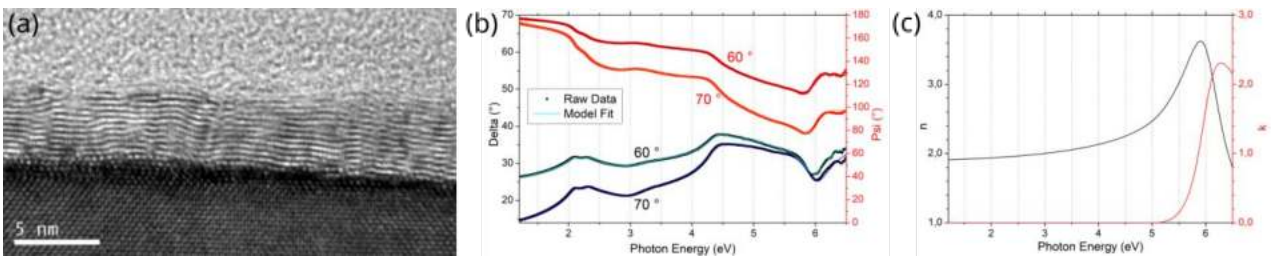

Figure 1: (a) TEM image of hBN film grown on Ge(001)/Si, (b) fitted ellipsometry spectra and c) determined optical constants n and k.

The influence of sample preparation on XPS quantification of oxygen-functionalised graphene nanoplatelets

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Industry using graphene-related two-dimensional materials (GR2Ms) need an accurate understanding of their material properties to be able to develop innovative new products. X-ray photoelectron spectroscopy (XPS) is widely used for characterising the chemistry of GR2Ms, however the careful preparation of the sample for analysis is important in obtaining representative quantifications. [1,2] We report an investigation by three laboratories showing that the preparation method for oxygen-functionalised graphene nanoplatelet (GNP) powders has a significant effect on the homogeneous-equivalent elemental composition measured in XPS. We show that pressing GNP powders onto adhesive tapes, into recesses, or into solid pellets results in inconsistencies in the XPS quantification. The measured O/C ratio from GNP pellets depends upon the die pressure used to form them and the morphology of the GNPs themselves. We recommend that powder samples of GR2Ms are pelletised prior to XPS analysis to improve repeatability and reproducibility of measurements. [3]

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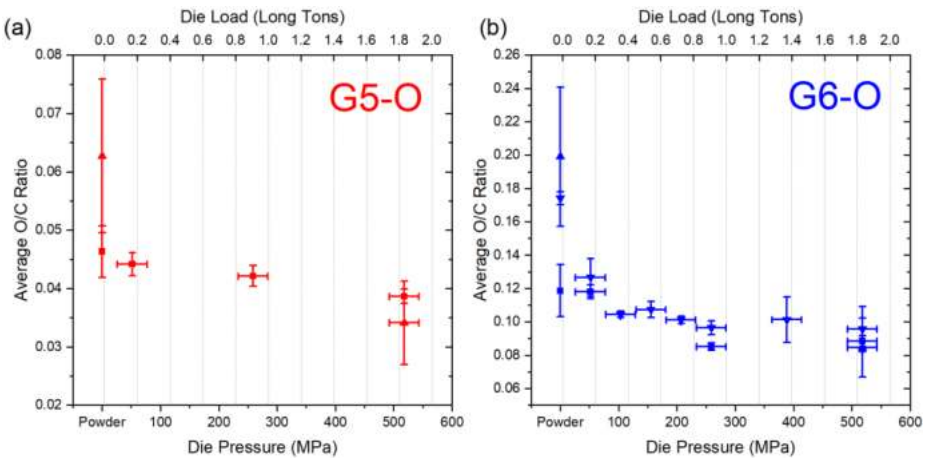

Figure 1: Influence of pellet formation pressure on the quantification of oxygen functionalised graphene. The measured O/C ratio decreases with increasing pellet formation pressure.

Mechanical behaviour of graphene reinforced polypropylene composites

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Abstract

Polypropylene (PP), as one of the most important thermoplastics in the world, has been widespread used in plastic packing, plastic parts for machinery and equipment, plastic furniture and even fibres and textiles [1]. However, many innovate materials require enhanced strength as well as multifunctional properties. In this sense, the utilisation of graphene opens a new perspective for polymer nanocomposites, revealing extensive research and development in the last two decades. Besides, adding graphene as a reinforcing agent in polymer matrices has improved the overall performance and properties of such composites, revealing promising applications in wide range of fields such as, electronics, biomedical aids, membranes, mechanical structures, among others [2]. This work reports some advancements and challenges of graphene reinforced polypropylene composites related to their mechanical properties, such as tensile strength, elastic modulus, and impact resistance. Also, scanning electron microscopy (SEM) and RAMAN hyperspectral maps are analysed to give a glimpse of the dispersion of the particles. The RAMAN and SEM images were then used to explain the observed mechanical behaviour of graphene-based composites.

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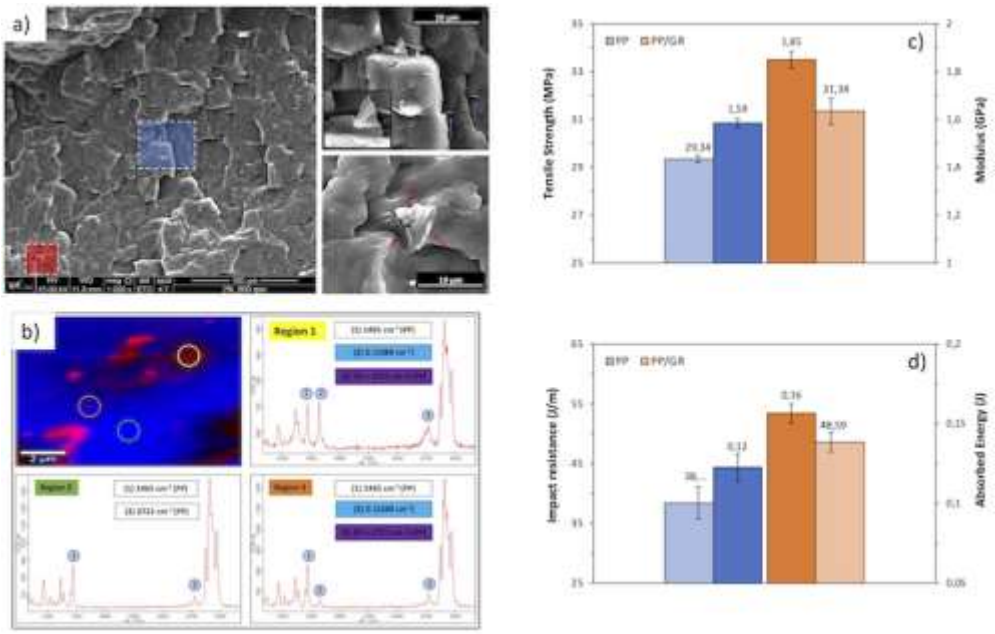

Figure 1: (a) SEM; (b) RAMAN spectroscopy; (c)Tensile strength and elastic modulus, and (d) Impact resistance and absorbed energy of graphene reinforced polypropylene composites.

Defect Implementation in 2D vdW Materials: Highly Correlated Fermion States and Autonomous Experimentation

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Defects within two-dimensional materials, such as transition metal dichalcogenides, enable a path to engineer new material functionality beyond conventional doping schemes. Chalcogen vacancies (0D) and mirror twin boundaries (1D) can be created at controllable densities to provide reactive sites for subsequent functionalization and the visualization of exotic phenomena such as the presence of quantum fluids in WS_2 [1]. The investigation of these systems with cross-correlative measurements that combine nano angle-resolved photoemission spectroscopy and scanning probe microscopy will be presented. Additionally, the usage of convolutional neural networks and Gaussian processes enables the direction of autonomous experimentation without the need of a human operator [2]. The implementation of a machine-driven workflow for hyperspectral tunnelling spectroscopy will also be discussed.

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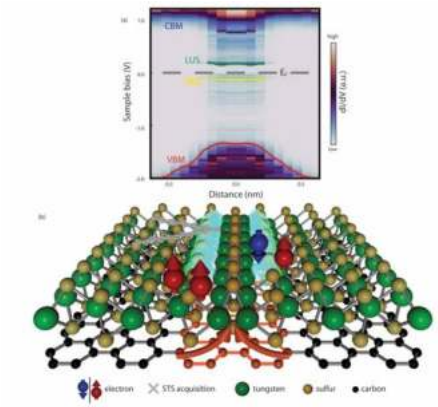

Figure 1: Band gap renormalization over a 1D defect within WS_2 realized at the atomic scale.

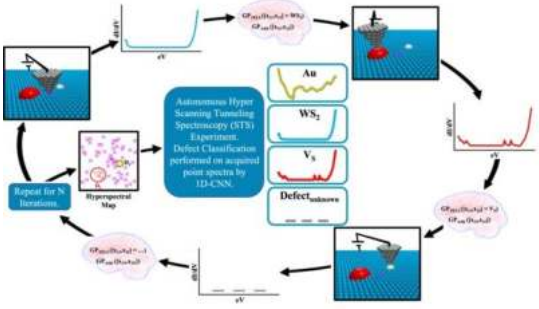

Figure 2: Hyperspectral data collection using a machine-driven workflow across a substrate accessible with a scanning tunnelling microscopy.

Multiferroic rotated CrI_3 bilayers with a three-dimensional intrinsic electric dipole

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Abstract

Magnetic bilayers can have their magnetoelectric multiferroic properties enhanced by the removal of a center of inversion through a relative rotation. Working with a prototypical CrI_3 bilayer, we have increased its intrinsic out-of-plane polarization by an order of magnitude through this procedure [1]. Surprisingly, we are also observing a robust and even larger in-plane polarization—similar to that observed in group-IV monochalcogenide monolayers [2]—which has not been reported to date. We also lay out a process to calculate the magnetoelectric tensor. Those results speak of the versatility of 2D bilayer magnets to become viable materials for novel and engineered magnetoelectric couplings.

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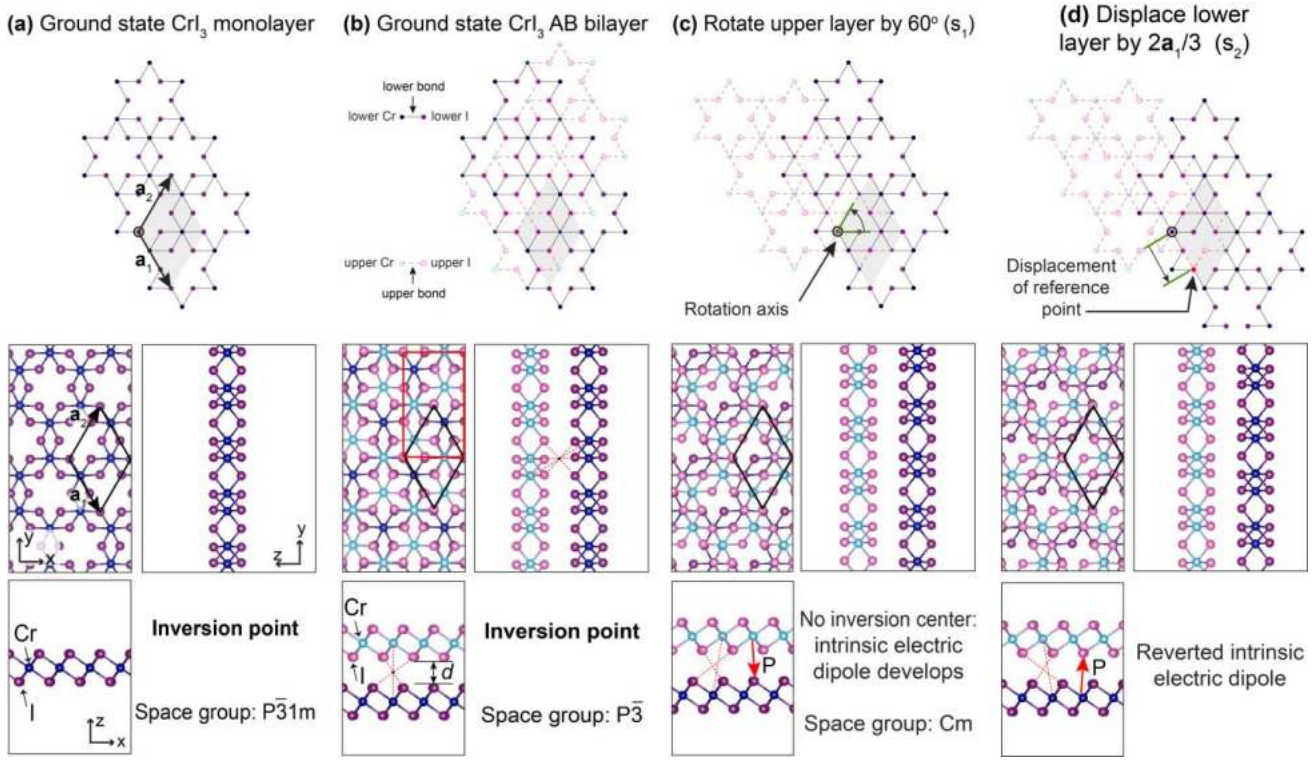

Figure 1: Process to enhance the magnetoelectric coupling on a CrI_3 bilayer.

Dynamic Ferroelectric Transistor-based Reservoir Computing for Spatiotemporal Information Processing

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Abstract

Reservoir computing (RC) architecture mimics the human brain and is a fundamentally preferred method to process dynamic systems that evolves with time.[1] However, generating rich reservoir states using two-terminal devices remains challenging, which hinders its hardware implementation.[2] Herein, we demonstrate the 1D array of ferroelectric field-effect transistors (Fe-FET) based on the 2D semiconductor a-In_2Se_3, which shows a volatile memory effect for realizing various RC systems. The ferroelectricity in this material is confirmed by PFM measurement, as shown in Figures 1 a and b. Using the read-after-write model, the dynamic polarization model sufficiently investigates the fading effect in a-In_2Se_3 (Figure 1c). Pattern recognition and waveform classification tasks are carried out with excellent training and testing accuracy to verify the ability of Ferro-RC systems (Figure 1e). Furthermore, time-series real-life chaotic systems, *e.g.*, Earth's weather, can be accurately forecasted using our Ferro-RC based on the Jena climate dataset recorded in one year. A remarkable determination coefficient (R^2) of 0.9983 and normalized root mean square error (NRMSE) of 8.3×10^{-3} are achieved using a minimized readout network. Demonstrating integrated memory and computation opens a route for realizing a compact RC hardware system.

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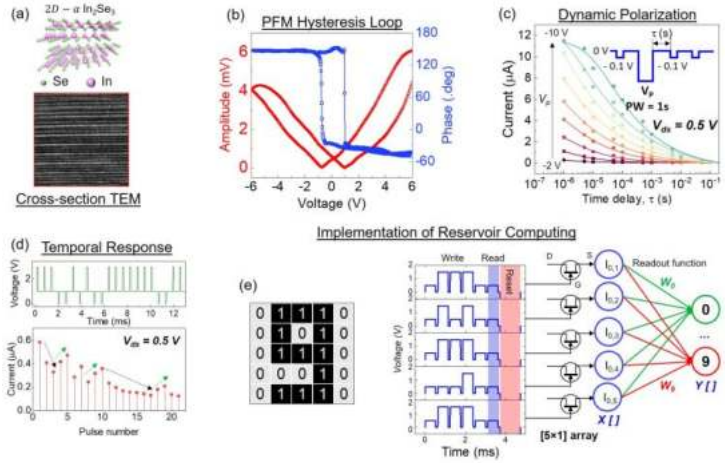

Figure 1: (a) Single crystalline Ferroelectric Semiconducting a (*R3m*) - In_2Se_3 and the layered structure indicated by HR-TEM image (b) Equivalent PFM hysteresis loops of phase and amplitude (c) Retention loss of drain current indicated by the read-after-write scheme with various V_p = -2 V to -10 V (d) Response current of dynamic a-In_2Se_3 Fe-FET on discrete V_g pulse stream (e) Measurement setup of Pattern Recognition, an example of the general machine learning task.

Rhombohedral graphite with a twin boundary defect: Flat bands, ferroelectricity, and spectroscopic signatures

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Abstract
Materials featuring flat bands, like twisted bilayer graphene or rhombohedral graphite, often exhibit rich correlated physics, due to their high electron density at the Fermi level. In this presentation, I will analyse one such material: rhombohedral graphite with a twin boundary. In this material, two rhombohedral graphite films are stacked on top of each other with a different stacking orientation, leaving an ABA-stacked trilayer buried inside the structure [Fig.1 (a)]. Its band structure features two pairs of nearly flat bands localised at the surfaces and the twin boundary [Fig.1 (b)]. I present an effective model to describe these bands [1] and demonstrate how this class of materials can host ferroelectricity due to the lack of inversion symmetry [2]. Finally, I will discuss several spectroscopic techniques, such as optical absorption or Raman scattering, can be used to characterise these materials, with emphasis on the smallest member of this family: ABCB tetralayer graphene [3].

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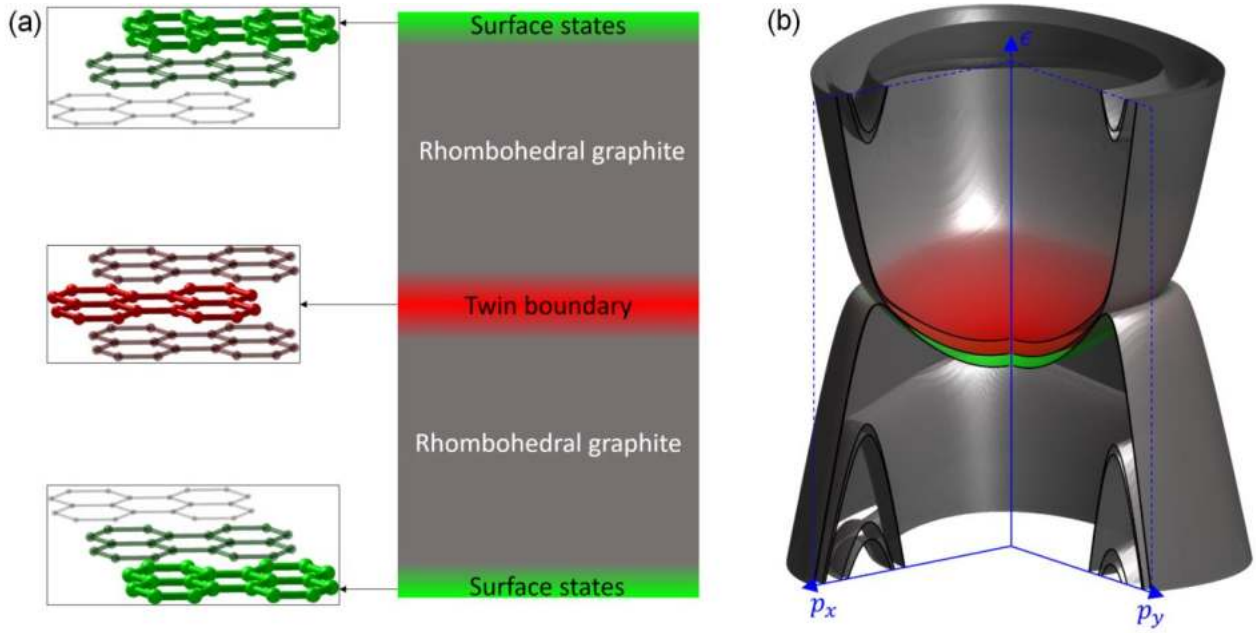

Figure 1: (a) Rhobohedral graphite with a twin boundary. The top and bottom insets present the characteristic ABC-stacking of rhombohedral graphite, while the middle inset presents the only ABA-stacking order in the film. (b) Band structure of thombohedral graphite with a twin boundary, coloured according to the localisation of the wavefunction. The red bands, localised inside the film, could induce correlated phases protected from environment.

Mapping tunnelling currents across switchable ferroelectric domains in parallel stacked layered materials with atomic force microscopy

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Controlling the twist angle between flakes of layered materials leads to interfacial ferroelectricity in the case of parallel stacked hexagonal boron nitride (hBN) [1-3]. We provide an account of the fabrication of such parallel stacked hBN samples using a home-built transfer setup [4] and the mapping of ferroelectric domains using various scanning probe techniques. We then discuss different ways in which such domains may be mapped and manipulated both electrically and mechanically [3,5]. We utilise conductive AFM (C-AFM) (where a tunnelling current is measured across a bilayer of parallel stacked hBN) to systematically study the variation of domain morphology versus mechanical setpoint and tip-sample bias. We show electrostatically induced switching of layer registry, enabling elevated relative areal coverage of AB domains versus BA domains depending upon the magnitude and direction of the applied bias. By performing C-AFM on parallel stacked hBN bilayers on graphene, we simultaneously map and manipulate the superlattice morphology to gain insights into both the mechanics of domain switching and tunnelling mechanisms under the influence of interfacial ferroelectricity. Such measurements are the nanoscale analogue of memristive tunnel diodes formed from parallel stacked hBN, which offer functionalities desirable for 'more than Moore' electronic devices.

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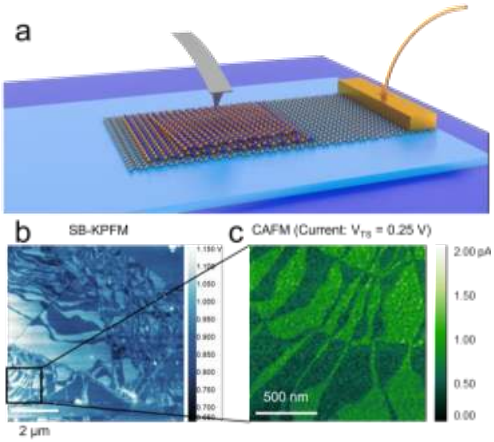

Figure 1: Parallel stacked hBN may be formed by breaking and restacking exfoliated layers mechanically (a). Once formed, these structures may be characterised using KPFM to map the morphology of ferroelectric domains (b). For sufficiently thin layers (~0.6 nm), tunnelling current may be mapped using CAFM.

Sliding ferroelectricity in bulk misfit layer compound $(PbS)_{1.18}VS_2$

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The recent theoretical prediction [1] and experimental confirmation of sliding ferroelectricity has significantly expanded the group of two-dimensional (2D) ferroelectrics. Due to the weak van der Waals interactions in layered van der Waals multilayers, an out-of-plane polarization can be created in many of those systems via in-plane interlayer sliding of a layer and thereby breaking the inversion symmetry.

Although the most common bulk phase of 'perfect' TMD crystals is the centrosymmetric 2H-form, the presence of sliding ferroelectricty should not be limited to manually stacked 2D few-layer crystals. As was recently shown for the amphidynamic crystal (15-crown-5)Cd_3Cl_6 [4], sliding ferroelectricity can also be observed in bulk crystals, as long as there is no inversion symmetry between the layers in van der Waals materials.

Here, the sliding ferroelectric properties of bulk $(PbS)_{1.18}VS_2$ misfit layer compound (MLC) crystals have been investigated. MLCs are thermodynamically stable, bulk, materials with a natural superlattice, consisting of the alternating stacking of two different 2D layers, here PbS and VS_2. The superlattice's formation and stability are still under debate, but it is suggested that charge transfer between the individual layers creating a strong electrostatic bond might stabilize these compounds

Using single crystal X-ray diffraction and a combination of imaging techniques, the sliding ferroelectric properties of $(PbS)_{1.18}VS_2$ were explored. The interaction between the two subsystems is derived from the presence of satellite reflections in the diffraction pattern of the composite. We find that the subtle interaction between the two subsystems causes the presence of twins, where two of the majority twins have a twist angle below one degree, the the necessary condition for sliding ferroelectricity The presence of ferroelectric domains, with a triangular shape and size from tens of nanometers to tens of micrometres, and their surface electrical potential from the induced sliding ferroelectricity can be observed using scanning electron microscopy, photoemission electron microscopy, imaging x-ray photoelectron spectroscopy and scanning probe microscopy imaging.

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Ultrahigh permeance metal coated graphene membranes for efficient gas separation applications

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Graphene is considered as the ideal membrane due to its atomic thickness. Indeed, million times higher permeance values compared to the traditional membranes were realized using porous graphene. However, the selectivity values of these membranes were rather low, in the limits of Knudsen selectivity. In order to achieve high selectivity on the graphene membranes, large number of pores below 3 nm are required to operate in the molecular sieving regime without sacrificing the permeance. However, this is extremely challenging and currently existing pore generation techniques result either in high permeance-low selectivity or vice-versa. Herein to overcome this challenge we have developed novel adsorptive membrane approach, in which one of the gases interact with the adsorbent surface and retained while the non-interacting one passes through the membrane. By doing so, we achieved complete separation of helium and hydrogen mixture, which is not possible using conventional approaches. Moreover, using graphene as a membrane support allowed to obtain permeance values in the rage of 10^7 GPU (1GPU = 3.35×10^{-10} mol s^{-1} m^{-2} Pa^{-1}).
In another approach, we developed controlled pore tuning method to create large number of pores below 3 nm. We deposited gold layer in stepwise manner and studied molecular transport properties. Initially, pristine graphene showed Knudsen selectivity, however, upon deposition of few nm of gold layer, the gas transport started to shift to surface diffusion where hydrogen was favored over other gases. Eventually, deposition of more gold led to the molecular sieving and we achieved record high H_2/CO_2 selectivity of 31.3 at H_2 permeance of 2.23×10^5 GPU.

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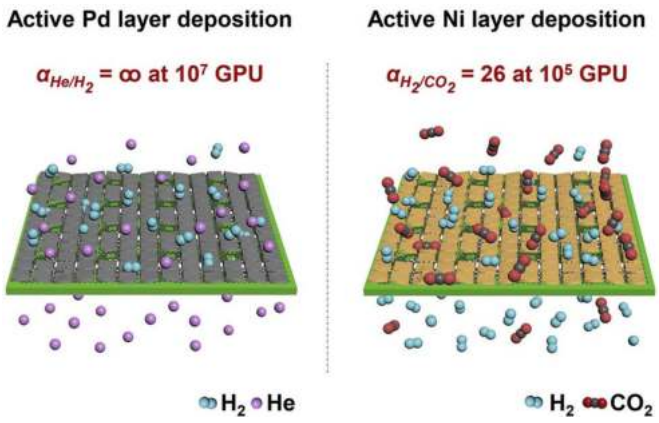

Figure 1: Schematic representation of Pd-coated for He/H_2 (left) and Ni-coated membrane for H_2/CO_2 (right) separation.

Flame-resistant cellular graphene aerogels and sensors

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Abstract

Protecting materials from fire damage is vital to many industrial and life safety applications. Although a handful of inherently flame-retardant materials exist, they are often expensive, brittle, or do not have suitable physical properties for the desired applications. Here we investigate structural and chemical changes of 2D graphene and 3D cellular graphene aerogels when exposed to different flames in air. We show that the arrangement of graphene flakes in the material strongly influences the flammability and combustion rate of graphene [1]. We demonstrate that free-standing graphene layers assembled into a 3D cellular structure exhibit 1000 °C higher flame resistance than 2D graphene on a substrate. The cellular graphene aerogels resist flames at a temperature of 1500 °C for a minute without degrading their structure or properties. Our findings reveal the exceptional fire-retardant and self-extinguishing properties of the cellular graphene aerogels, which can be used to for protecting materials from fire. Moreover, we demonstrate the use of the elastic graphene aerogels in tactile sensors [2,3]. We show the graphene aerogel sensors are fast and can operate over a very broad range of stress and strain both in compression and tension independently of the temperature.

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Figures

Figure 1: Flame-resistance testing of a cellular graphene aerogel using a propane flame.

Ink-jet Printed Graphene-Silicon Schottky Diodes

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Abstract

Integration of graphene (Gr) in silicon-based technology is of crucial importance for enabling the next generation electronics, photonics and sensors [1]. Although numerous works have reported devices based on Gr-Si junctions, the integration process relies on the use of high quality Gr produced by Chemical Vapour Deposition (CVD), making the fabrication steps expensive, time consuming and by limiting the large scale devices' reproducibility.

In this work we show that inkjet-printing enables simple and scalable integration of Gr into Si-technology [2]. We developed a simple fabrication procedure, based on the mechanical or chemical etching of the SiO_2 layer from a standard Si wafer, followed by the inkjet printing of water-based printable Gr inks [3] on the exposed area, leading to Schottky diodes with excellent rectifying behaviour and figures of merit, comparable to those produced with CVD graphene. We fully characterized the devices and applied several theoretical models achieving deep understanding of the underlying physics of the devices. We also investigated the optical response of the diodes by demonstrating a spatially selective photodetector.

Our results demonstrate that inkjet printing is a cost-effective and scalable method, which is also compatible with back-end-of-line fabrication processes for the integration of graphene in the modern Si-technology.

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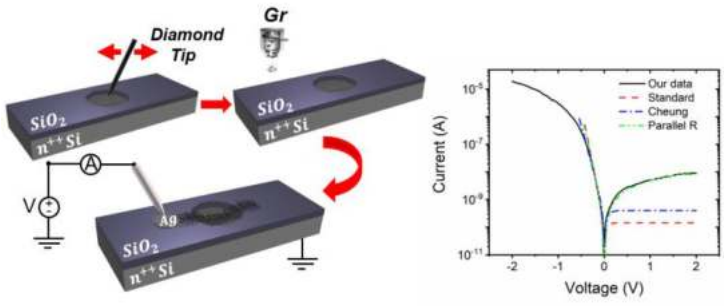

Figure 1: Schematic of the fabrication process and I-V characteristic of the printed Gr-Si diode

LIQUID PHASE INTERCALATION OF NIOBIUM FLUORIDE INTO GRAPHITE FILMS INCREASES ELECTRICAL CONDUCTIVITY UP TO 27 MS/m

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The intercalation of metal fluorides in graphite is well-known by vapor phase [1]. For most metal fluorides, the presence of gaseous fluorine is even required for intercalation [2]. Nakajima et al. report an electrical conductivity of 12.5 MS/m for niobium fluoride graphite intercalation compounds [3]. We show a liquid phase approach for intercalation of niobium fluoride by dissolving it in fluorosulfuric acid. Niobium fluoride intercalates into graphite films resulting in a color change of the film into deep blue. The measurement of the electrical conductivity of the deep blue samples reaches up to 27 MS/m.

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Highly Sensitive Graphene Nanoplatelets Strain Sensor for Measuring Impact Loading in Infrastructures

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Abstract: Recently, graphene-based composites have been subjected to extensive research for their potential use as strain sensors to monitor the health of infrastructure [1, 2]. The purpose of this study was to develop strain sensors using nonwoven fabrics coated with graphene nanoplatelets (GNPs). The polyester nonwoven fabric was coated with graphene nanoplatelets using a standard laminator and laminating pouches. An electron microscope (TESCAN MIRA3 FEG-SEM) was used to characterize the morphologies of the graphene coated surfaces. Figure1(a) illustrates the morphology and characterizations of the graphene coated nonwoven fabric. The graphene coated strain sensor was mounted onto a built house stretching equipment using a customized linear step motor controlled by Arduino UNO. Under 3% strain with 0.5 % strain-step, a 2450 Source Measure Unit (SMU) instrument was used to measure the relative resistance changes during stretching-relaxation of the strain sensor, as depicted in Figure1(b). To demonstrate the static response of the strain sensor under a series of loading and unloading tests, a metal weight (150 g) hung and remained stationary onto one end of a plastic ruler holding the strain sensor, the other end of the ruler was fixed. The resistance changes during the loading cycle of the strain sensor were shown in Figure1(c). The results indicated that the trained strain sensor showed excellent sensitivity and stability with average gauge factor of 6.33 under an applied strain of 3%. The prepared strain sensor demonstrated a good static response during loading cycles, which enabled it to be used for the monitoring of infrastructure health.

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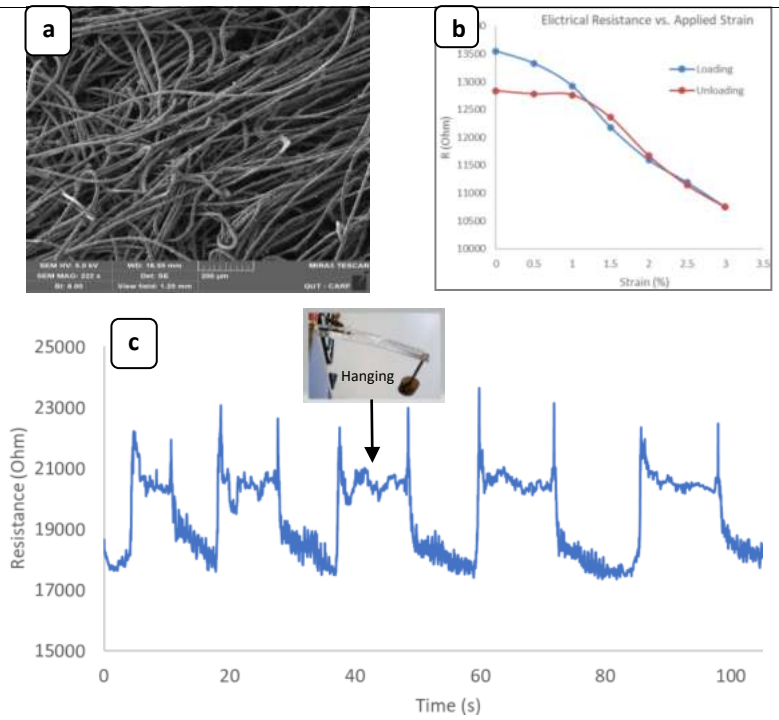

Figure 1: (a) SEM image of the graphene coated nonwoven fabric, **(b)** Electrical Resistance vs. Applied Strain **(c)** the static response of the strain sensor under strain

Hierarchical Graphene-Based Aerogel Catalysts and Sorbents

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Directed self-assembly of graphene derivatives into structured aerogels and foams is explored as route to create unique, porous catalyst systems with highly-tailored materials characteristics, including hierarchical porosity, mechanical durability, and electro-thermal responsiveness. Here, template-based assembly approaches are explored to produce sponge-like nanocarbon aerogels with a wide range of well-controlled hierarchical microstructures. Different gas-phase and wet-chemical methodologies are developed to enable uniform and structure-preserving aerogel functionalisation with catalyst nanoparticles (anionic clays, mixed metal oxides, precious metals).[1,2] Advanced X-ray micro-CT and FIB-SEM-EDX techniques are employed to characterise the aerogels' three-dimensional microstructure and surface chemistry. Embedding nanoparticles within the aerogels is shown to provide remarkable improvements in functional performance (activity, selectivity, kinetics, recyclability) across a range of chemical applications, including high-pressure CO2 capture,[1] fine-chemical catalysis,[2] and fuel desulfurisation.[3] Beyond this boost in functional nanoparticle performance, graphene-derived aerogels also provide valuable additional functionality. For example, the electrical conductivity of the 3D-interconnected graphene network can be utilised for energy-efficient flash Joule-heating.[4] Ultrafast and ultrahot resistive aerogel heating (>2000°C) is exploited for highly controlled nano-catalyst synthesis and rapid thermal catalyst recycling. Nanoparticle-decorated aerogels are also explored as flow-through catalysts within chemical flow processes, an area of increasing interest due to substantial benefits in process control and sustainability. Specifically, the performance of aerogel catalysts in chemical flow reactions and potential for controlling the chemical reaction profile through graphene aerogel microstructure are explored.

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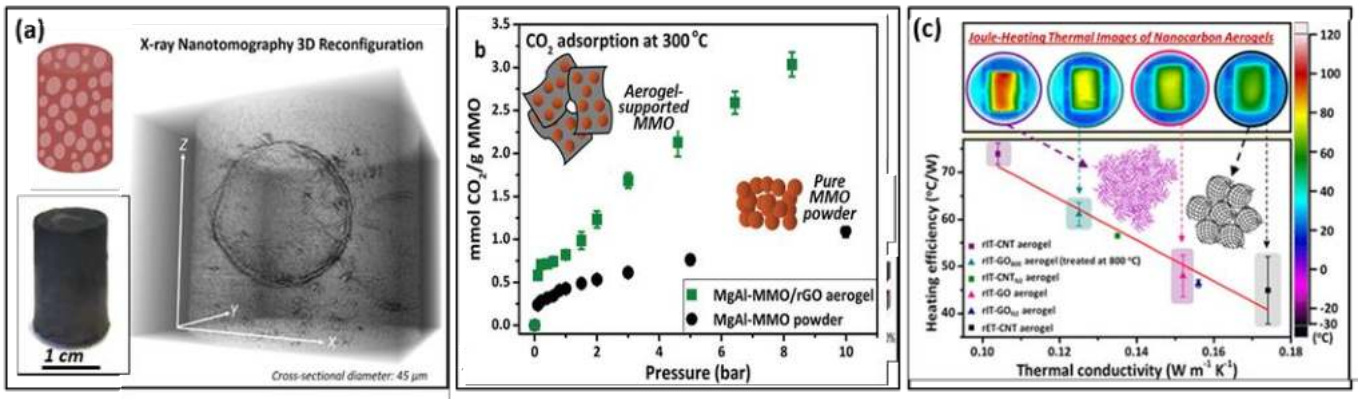

Figure 1: (a) X-ray nanotomography of an emulsion-templated nanocarbon aerogel catalyst; (b) High-pressure CO_2 capture performance of rGO aerogels functionalised with mixed-metal-oxide (MMO) nanoparticles; (c) Thermo-electric structure-property relationship study of different nanocarbon aerogels.

GNPs/epoxy nanocomposites as conductive adhesives for Out-of-Autoclave in-situ Carbon Fibres Reinforced Polymers repair

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Carbon fibre reinforced polymers (CFRPs) have been widely used in the aerospace and automotive industries over the last decades due to their light weight and high tensile strength [1]. However, there are important issues associated with replacing or recycling damaged CFRPs. Indeed, the methods most commonly used to dispose waste CFRPs, i.e., landfill and incineration, involve high cost and energy consumption [2]. In addition, even though some efforts are currently being made on developing chemical and thermal methods for recycling them, they are quite aggressive and very often lead to irreparable damage to the carbon fibres [2]. Hence, strategies to efficiently repair damaged CFRPs at a low cost are in great demand [3]. Epoxy adhesives have been used for joining composite components because uncured epoxy wets well the adherend surface, promoting a strong interface and hence ensuring a good mechanical performance after curing [4]. Recently, researchers are adding micro/nano fillers into the adhesive epoxy matrix to improve the mechanical performance of the bonded joints [5]. However, the curing of these epoxy adhesives is normally performed in an autoclave, which use is labour-intensive and involve both high energy consumption and high cost. Thus, there is a desire to develop out-of-autoclave (OoA) strategies to repair CFRPs. Herein, we report the use of GNPs/thermoset nanocomposite mixtures as conductive adhesives for an in-situ out-of-autoclave (OoA) repair of CFRPs through Joule heat curing of the electrically conductive network of GNPs flakes embedded in the epoxy matrix. Due to the relevance of the bondline thickness for the targeted application, the electrical and Joule heating properties of the graphene/epoxy nanocomposite systems both as bulk and films were investigated as model systems and related to their microstructure. Establishing the structure/property relationship of the model systems allowed most promising GNPs loadings to be taken forward as conductive adhesives for in-situ OoA CFRPs repair. The influence of the adhesive's GNPs loading on the heating rate and distribution of the Joule heat generated during the repair process was investigated to optimize the process. The mechanical properties of the CFRPs repaired by Joule heat were compared with those found for the CFRPs repaired in an oven. The failure mechanism of the joints was also studied to gain insight on new directions to improve this OoA repair method.

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Thermally conductive h-BN/polymer composites for textiles thermal management

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Abstract

Textile materials with high thermal conductivity are ideal for personal cooling by accelerating the heat dissipation between the human body and hot atmosphere via thermal conduction. Current works on advanced textiles for thermal management adopted thermally reinforced composites to form nanofibers using carbon-based or boron-based nanofillers, achieving an in-plane and out-of-plane thermal conductivities (κ) of 13.1 W m^{-1} K^{-1} and 1.9 W m^{-1} K^{-1}, respectively (1). However, the complex fabrication and the insufficient thermal conductivity of nanofiber composites impede their use in applications such as sportswear which requires more efficient transfer ability of heat flux to surroundings. Hexagonal boron nitride (h-BN) is a promising candidate as thermal conductive filler with an in-plane thermal conductivity value of up to 370 W m^{-1} K^{-1} (2). It can be exfoliated into single layer form achieving a value up to 751 W m^{-1} K^{-1} (3). In this work, we demonstrate a thermally conductive composite embedded with exfoliated h-BN, achieving κ ~ 21.7 W m^{-1} K^{-1}, (figure 1) about 5-fold higher than bulk h-BN embedded composites (κ ~ 4.5 W m^{-1} K^{-1}). Exfoliated h-BN were produced via probe sonication of bulk h-BN with carboxymethyl cellulose (CMC) in water and subsequently purified via centrifugation. Atomic Force Microscopy characterisation revealed the average thickness of the exfoliated h-BN flakes is around 6 nm. The h-BN/CMC aqueous composite is then drop-casted onto a non-woven fabric and dried in the air to prepare a thermally conducting textile. Comparative temperature measurements at the thermal equilibrium between the h-BN/CMC coated textile and the uncoated textile shows that the coated textile reaches a higher temperature than the uncoated textile by 1 ℃, indicating that the exfoliated h-BN-assisted composite improved the heat amount dissipated from the heater to the environment. The cooling effect of the exfoliated h-BN-assisted composite textile is calculated to be 5.5% greater than the commercial textile under natural air convection (4, 5), which displays a better thermal management capacity of the textile for applications in active thermal management clothing for sportswear, aerospace, or heavy-duty industries.

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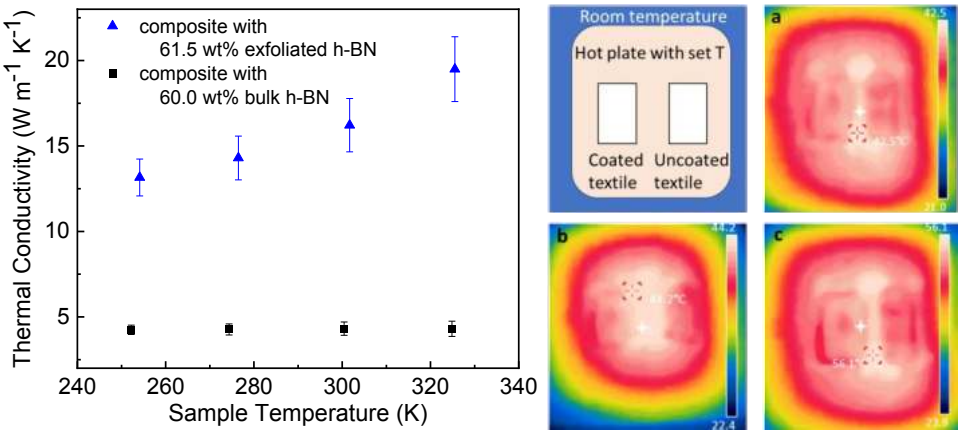

Figure 1: Left: Thermal conductivity of the exfoliated and bulk h-BN assisted composites; Right: Comparative temperature measurements between the coated textile and the uncoated textile.

The Role of Filler Aspect Ratio in the Reinforcement of Polymers with Graphene and 2D Materials

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The mechanisms of reinforcement of an epoxy resin by the addition of graphene nanoplatelets (GNPs) has been studied in detail. It is found that the addition of GNPs increases both the stiffness and fracture toughness of the epoxy resin. The dependence of the flexural modulus upon the volume fraction of the GNPs has been modelled using a combination of the rule of mixtures and shear lag analysis [1] and it is shown that the reinforcement is controlled principally by the aspect ratio (length/thickness) of the GNPs. The dependence of the fracture energy upon the GNP volume fraction has also been modelled assuming failure takes place through the debonding of the GNP particles followed by their pull-out as shown in Figure 1. This behaviour is similar to that found for the toughening of elastomers by 2D materials [2]. It is again shown that the aspect ratio of the GNPs is a vital parameter in controlling the level of toughening. It is found that the mechanical behaviour can be modelled using a similar value of GNP aspect ratio to model both the flexural stiffness and fracture behaviour, demonstrating the importance of this parameter in controlling the mechanical properties of GNP/epoxy resin nanocomposites. The application of the above analysis to our understanding of the reinforcement of a wide range of polymers by 2D materials will be discussed in detail.

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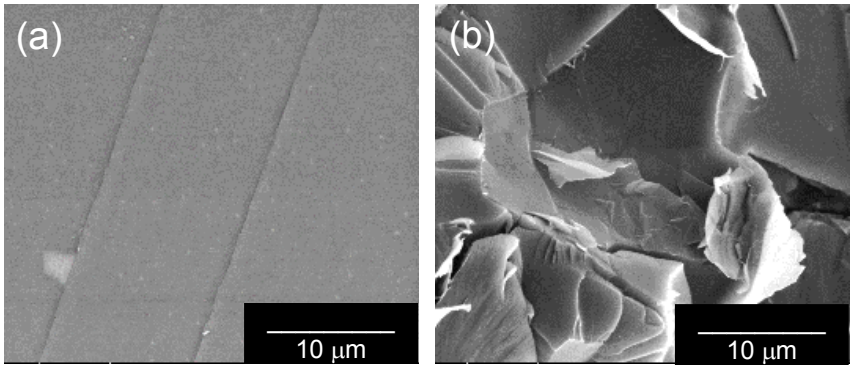

Figure 1: SEM micrographs of the fracture surfaces of (a) Pure epoxy and a nanocomposites with (b) 1 wt% GNPs, showing pulled-out GNPs and cavities.

Multifunctional graphene films for aerospace composites

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Abstract

Since its discovery in 2004, graphene has marked its presence in several sectors such as biomedical, sports, electronics, energy harvesting and storage, and transportation. Transportation, in particular aerospace, is third largest sector after electronics and energy. Among its major applications, graphene is largely implemented as a composite reinforcement, thermal management in electronic components to name a few. Nevertheless, there are several other application areas where graphene has potential to replace metallic counter parts. For instance, metal meshes or foils are used to dissipate electrical and thermal energies in the event of lightning strike [1]. Similarly, inflatable or electro-thermal devices are used to remove accumulated ice (de-icing) or to prevent ice accumulation (anti-icing) in sub-zero temperatures [1]. Both of these scenarios deal with external surface of an aerospace vehicle such as an aircraft. In this case, graphene layer with its excellent electrical and thermal properties [2] can be implemented to replace metallic parts and eliminate maintenance, galvanic corrosion and weight issues. In this work, graphene films have been proposed as highly conductive skins to protect against lightning strike. Graphene films with high concentration of nanofillers have been realized by solution blending, evaporation of solvent and film deposition followed by a calendaring process. The final films of around 200 µm thickness were achieved that are equivalent to an epoxy resin impregnated carbon fiber ply (pre-preg). The as-prepared graphene films were laminated onto baseline pre-pregs and co-cured in a single cycle, at recommended temperature and pressure. The graphene film laminated CFRP composites have been studied for their morphological, mechanical and electrical properties. It has been demonstrated that graphene films with high electrical and thermal properties can be prepared and implemented onto CFRP panels. In future, these graphene films laminated CFRP composites will be tested under lab scale lightning strike events.

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2D materials based antifouling membranes for ultrafiltration

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Abstract
Inherently hydrophilic surfaces exhibit supreme ability to bind water molecules via hydrogen bonding and provide a steric repulsive barrier to adsorption of organic and protein contaminants[1]. These surfaces offers formidable solutions to improve fouling resistance, hence actively pursued in the case of ultrafiltration membranes. Improved wetting is typically achieved by either chemical modification or by controlling the hierarchical structures of surfaces to manipulate the surface energy[2]. Here we report a paradigm shift approach where the wetting properties of vermiculite laminates are controlled by the hydrated cations on the surface and in the interlamellar space. A superior wetting transition from superhydrophilic to hydrophobic is demonstrated simply by exchanging the cations. The hydrophilicity is observed to decrease with cation hydration free-energy for common cations, whilst the lithium-exchanged vermiculite laminate is found to be a mere exception with its anomalous hydrated structure at the vermiculite surface leading to a superhydrophilic surface. By exploiting this unprecedented wetting control, superior microfiltration membranes are engineered via coating a thin layer of superhydrophilic lithium exchanged vermiculite on polymeric microfiltration membranes to demonstrate their excellent fouling resistant operation, and thus, we address one of the major challenges in membrane-based separation technology.

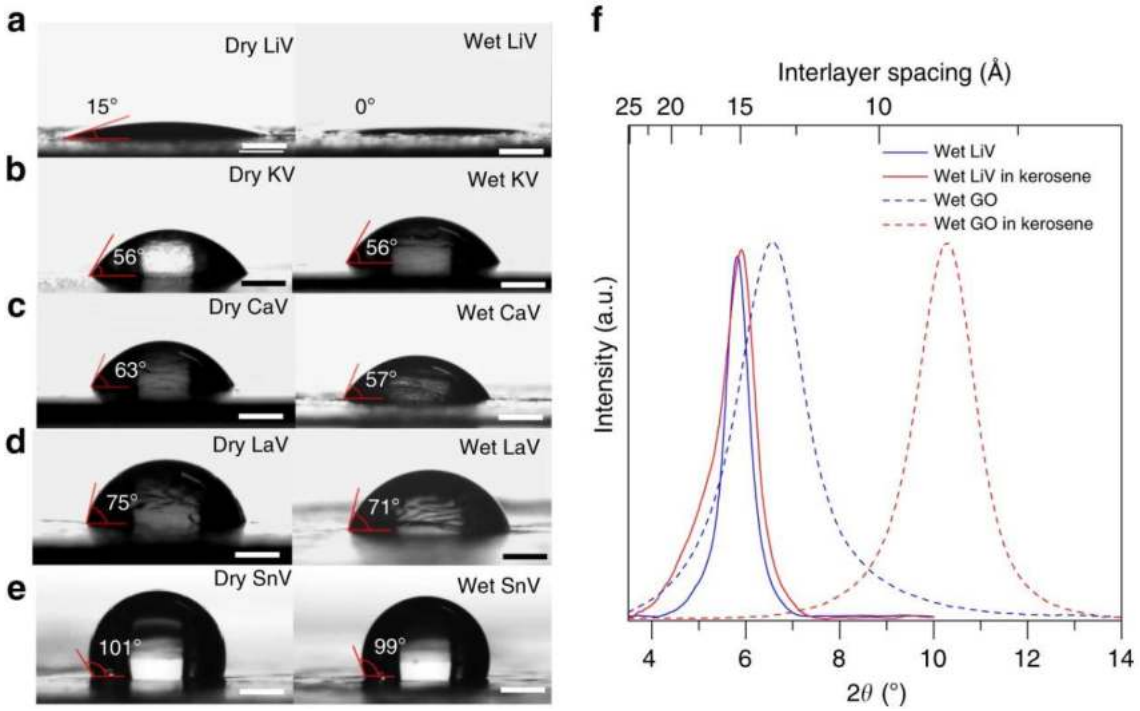

Figure 1: Wetting properties of vermiculite laminates: (a–e) Water contact angle of lithium vermiculite (LiV), potassium vermiculite (KV), calcium vermiculite (CaV), lanthanum vermiculite (LaV), and tin vermiculite (SnV) -laminates in dry and wet states. Scale bar, 750 µm f) XRD from wet LiV laminate before and after immersing in kerosene for a week along with XRD spectra of a reference GO membrane tested at identical condition.

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Molecular simulations of the wettability of Graphene and Graphite

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Abstract (Century Gothic 11)

The water wettability of carbon surfaces, such as graphene and graphite, has been extensively discussed over the past few years due to its broad range of applications. [1] Experimentally however it is difficult to untangle the effect that impurities, surface defects and number of layers have on the wetting properties. Here using molecular dynamics we report the wetting behavior of graphitic surfaces–water interfaces through the calculation of the local stress tensor based on the Irving–Kirkwood–Noll theory. We compare the interfacial properties of fully wetted and partially wetted graphene and graphite and observe that the wettability is the result of a fine balance between van der Waals (vdW), electrostatic interactions and hydrogen bonds structure.[2] We then explore how 2D confinement changes graphite wettability and the possible consequences that such changes have on water flux properties. [3]

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Figure

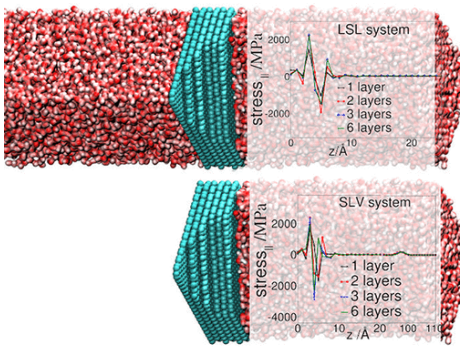

Figure 1: Fully wetted and partially wetted graphite slabs. Corresponding surface tension plots as a function of the number of graphene layers.

Wien effect and photo-accelerated interfacial water dissociation across proton permeable graphene electrodes

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The phenomenon known as the Wien effect was previously observed using high-voltage electrolysis cells that produced fields of about 10^7 V m^{-1}. The observation of the Wien effect for the common case of water dissociation has remained elusive. Here we study the dissociation of interfacial water adjacent to proton-permeable graphene electrodes and observe strong acceleration of the reaction in fields reaching above 10^8 V m^{-1}. The observed exponential increase in proton currents is in quantitative agreement with Onsager's theory. The use of graphene as a membrane allows for measuring the proton currents arising exclusively from the dissociation of interfacial water. Illumination of the graphene under visible light produced an order-of-magnitude acceleration of the interfacial water dissociation reaction. The found photo effect is attributed to the combination of graphene's perfect selectivity with respect to protons, which prevents proton−hydroxide recombination, and to proton transport acceleration by the Wien effect. Our findings provide fundamental insights into ion dynamics near atomically thin proton-selective interfaces.

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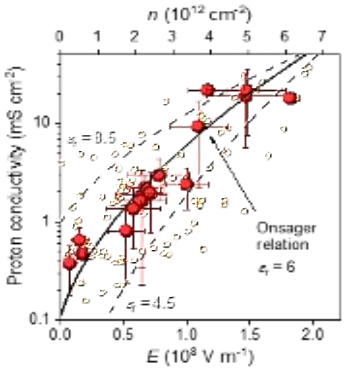

Figure 1: *Proton conductivity through graphene electrode* where *n* is the carrier density. Bottom x-axis, *is Electric field strength* on graphene electrode. Solid curve, best fit of Onsager model to data. Dotted curves, Onsager model for different *dielectric constant*.

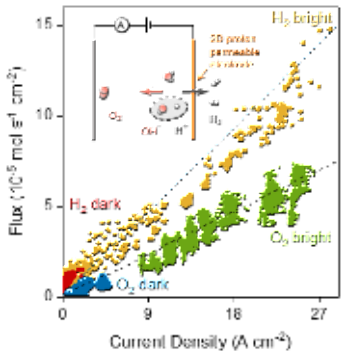

Figure 2: Faradaic efficiency measurements. Hydrogen and oxygen fluxes as a function of I under dark and bright conditions.

2D Nanoconfined Fluidic Channels and its Application

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Abstract

Nanoconfined electrical double layers have attracted significant attention due to their potential benefits in environmental and biomedical applications by facilitating ion-selective mass transport. [1] Our research explores the ion selectivity of nanoconfined fluidic channels under chemical potentials using interplanar nanochannels created by stacking two-dimensional (2D) nanosheets such as metal carbide and nitride (MXene) or graphene oxides in an ordered manner. [2-5] The overlapped electrical double layers between neighbouring 2D sheets allow for selective ion transport while maintaining a consistent interlayer distance. Our investigations demonstrate the potential of subnanometer-scale channels derived from a lamellar structure for ion-exchange membranes, salinity-gradient energy harvesting, and sensory transduction. Specifically, MXene-based membranes have exceptional salinity-gradient energy harvesting capabilities, achieving an output power density of up to 54 $W \cdot m^{-2}$ by regulating surface charges and ionic mobility. [3] We have also proposed a new type of lamellar membrane constructed by holey 2D nanosheets, which exhibits simultaneous enhancement in permeability and ion selectivity beyond their inherent trade-off. [4] The perforated nanopores on the plane lower the energy barrier for cation passage, thereby boosting preferential ion diffusion across the membrane. Additionally, we have demonstrated how MXene-based ion conducting channels can be utilized for a photothermal sensory transduction system, which converts light-driven thermochemical potential to active ion transport.[5]

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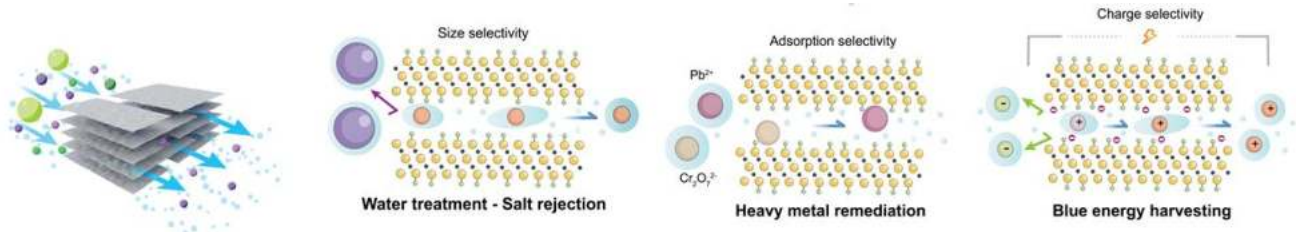

Figure 1: 2D Nanoconfined fluidic channels and its applications for selective separation [1]

2D water: How does it flow?

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Two-dimensional (2D) water, confined by atomically flat layered materials, may transit into various ordered phases even at room temperature [1]. However, the transport of such water is still not well understood. The problem is that conventional hydrodynamic approach in terms of the slip length requires a well-developed out-of-plane flow velocity profile absent in the truly 2D limit. Here, we consider the Navier-Stokes equation in the 2D limit assuming no vorticity (rot **v** = 0) but a certain compressibility (div **v** ≠ 0), where **v** is the flow velocity [2]. The first and second viscosity coefficients deduced from the viscous stress tensor then acquire the physical meanings of the effective interfacial and dilatational viscosities, respectively. At the same time, we perform molecular dynamic (MD) simulations and fit the resulting flow velocity using the effective viscosity coefficients. **Figure 1** demonstrates how the effective viscosity coefficients are influenced by the channel's material and height. The result opens an interesting opportunity to obtain various nanofluids out of the same water molecules just by using alternate materials to fabricate the 2D channels.

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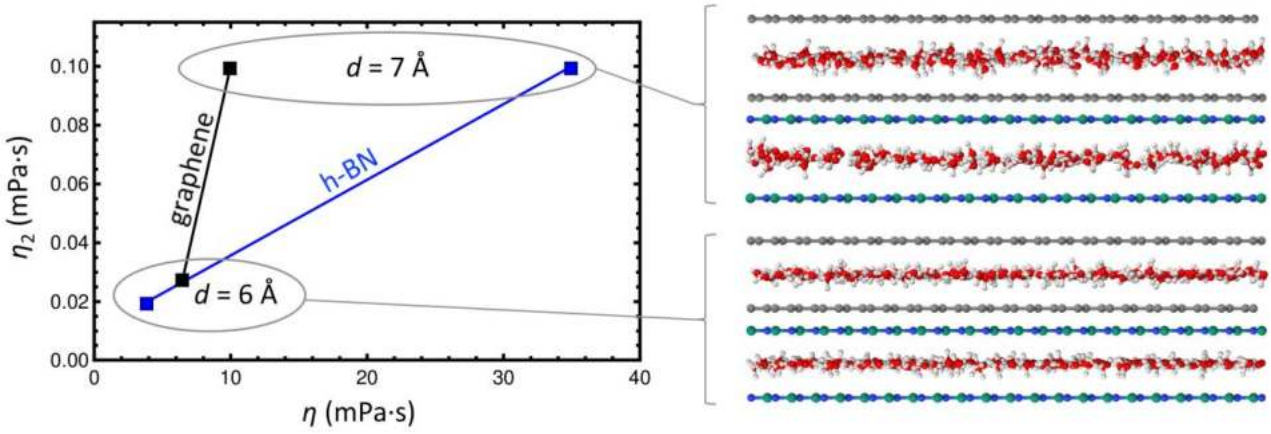

Figure 1: Changing the channel's height and material strongly influences the effective viscosity coefficients obtained by fitting our MD simulations. The snapshots on the right show that while all oxygen atoms are nearly aligned in one plane in narrow channels (6 Angstrom), they acquire an out-of-plane staggering pattern in wider channels (7 Angstrom) leading to stronger interactions with the channel walls. The water layer remains 2D in all cases (monolayer). The staggering pattern is more pronounced in the h-BN channel, which is reflected in the higher viscosity coefficients. Here, O, H, C, B and N atoms are represented in red, white, grey, green and blue, respectively.

Ultraconfined two-dimensional water in gypsum

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Abstract

Water plays an important role in a variety of processes, such as electrochemical reactions, energy storage, and biological processes, to name but a few. Most of the unusual properties and phenomena, such as abnormal dielectric constant [1] and unusual phase transitions [2, 3], are related to the extensive hydrogen bonding network in water, which is strongly altered under confinement.
In this study, we investigated the vibration of two-dimensional (2D) water sheets confined in a natural layered mineral, gypsum ($CaSO_4 \cdot 2H_2O$). Water O-H stretching modes are found to be localized on two types of O-H dipoles due to the two disparate hydrogen bonding strengths, resulting in two distinct vibration frequencies, which enabled us to study the dielectric behaviour of these two confined O-H dipoles independently. A distinct vibrational anisotropy is observed for the two local modes, indicating the orientation of O-H dipoles in such 2D confinement. By analysing the vibration frequency under different confining geometry of O-H dipoles, the dielectric polarization of nanoconfined water can be revealed. A concise model was constructed to describe the vibration and polarization of water under confinement.

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Enhanced edge-nonlinear response and photoresponse in MoS_2 nanoribbons

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This work will present our findings on the growth of highly crystalline MoS_2 nanoribbons and multilayer triangular crystals with controlled stacking orientation. The synthesis method relies on the reaction between ultra-thin films of MoO_3 grown by Pulsed Laser Deposition and NaF in a sulfur-rich environment [1]. The MoS_2 nanoribbons can reach up to 10 µm in length and 500 nm in width, resulting in a high aspect ratio. Atomic Force Microscopy (AFM) and Tip-enhanced photoluminescence (TEPL) spectroscopy reveal that the MoS_2 nanoribbons feature single-layer edges with blue-shifted exciton emission, forming a versatile single-multilayer homojunction. Multiphoton microscopy reveals a significant optical second harmonic generation (SHG) from the single-layer edges of the nanoribbons. We will discuss these findings based on the non-centrosymmetric single-layer edge and/or symmetry breaking at the surface. Moreover, we report on a high-performance single-nanoribbon MoS_2 photodetector with a remarkable responsivity of 7.59×10^2 A/W, superior to previously reported nanoribbon photodetectors.

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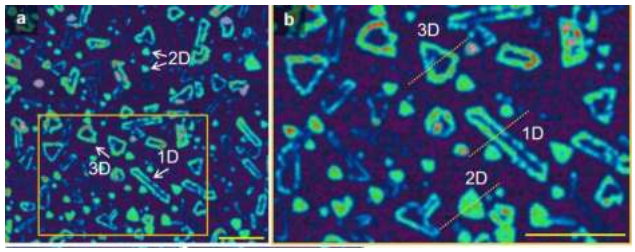

Figure 1: (a) SHG map of the MoS_2 nanostructures and (b) zoom over a selected area with edge-enhanced SHG.

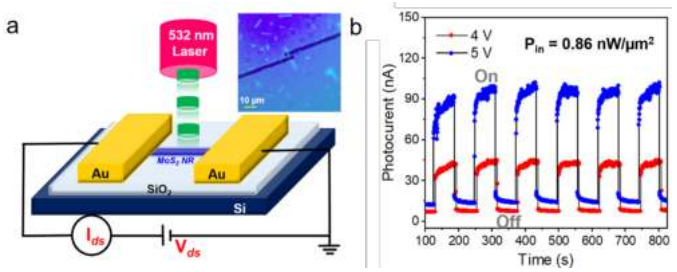

Figure 2: (a) Schematic of the MoS_2 nanoribbon device under 532 nm laser illumination and (b) ON/OFF photo response of the nanoribbon.

Acoustic THz graphene plasmons and mid-infrared polaritonic nanoresonators at low temperatures

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Polaritons are coupled excitations of incident light with charged particles (plasmons polaritons) or lattice vibrations (phonon polaritons). In particular, the polaritonic properties of two-dimensional (2D) materials become extreme in several aspects, such as light confinement and long lifetimes. Hence, they lead to a myriad of applications for gas and molecular sensors, enhanced photodetectors and quantum technologies.

In this work, we show an efficient platform to launch acoustic THz graphene plasmons by using an antenna that defines the plasmonic cavity.[1] These plasmons are measured via far field THz photocurrent spectroscopy. We test several devices that show peaks in the photovoltage, which correspond to peaks in absorption due to graphene acoustic plasmons present at the same gate voltage/Fermi level doping. We measure from room temperature down to 5 K, where the resonant peaks vanish at high temperatures, while at low temperatures become prominent. We observe that these resonances are shifted and additional features appear when changing local gate and back gate voltages. The photoresponse and resonant peaks drop significantly when polarizing the incident light perpendicular to the antenna's main axis. Moreover, we measure at different incident frequencies (from 1 to 4 THz) to further tune these resonances The experimental results show good agreement with the numerical calculations and dispersion relation of the acoustic THz graphene plasmons.

Then, we show a novel concept of 2D polaritonic nanoresonators that consist of merging into one single platform the polaritonic material and the detector as shown in Figure 1b.[2,3] We obtain a highly compact device since we get rid of the need for an external detector for performing infrared spectroscopy. We geometrically and electrically tune these nanoresonators to change their spectral photoresponse. Due to this, we can identify different interactions such as the hybridization between graphene plasmons with the HPPs and modification of the HPPs waveguide modes due to the graphene doping. We investigate the photoresponse of these 2D polaritonic nanoresonators as a function of the temperature, reaching values down to 30 K.[3] The devices show higher values of photocurrent due to the interplay of the contributions of the photothermoelectric effect and longer lifetimes of the polaritonic resonators. We determine that the Q factor values increase significantly at these lower temperatures by reaching values up to 300 and the results are supported by theoretical simulations.

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Figures

a) b)

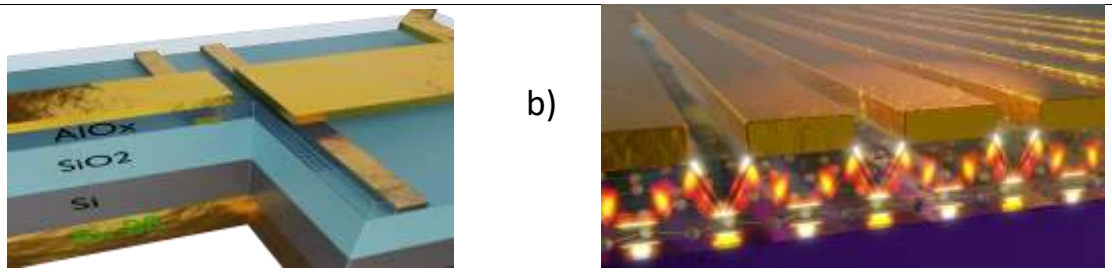

Figure 1: a) Cross-section view of the graphene plasmonic THz photodetector. **b)** Schematic of the 2D polaritonic nanoresonator that shows the field intensity of the propagating mode.

Integration of optoelectronic response in 2D materials into Si photonics

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By using optical platforms instead of metallic interconnects, photonic devices can achieve both high speed and low power consumption suitable for next generation information processing. Although the state-of-the-art silicon (Si) photonic chips are outstanding optical platforms for light propagation, it requires external active optical components such as light sources and photodetectors. A potential solution comes in the form of atomically thin two-dimensional (2D) materials. Their remarkable optoelectronic properties are widely tunable by doping, strain, and external fields, owing to their atomic thickness and unique characteristics. Moreover, their two-dimensional planar structure is suitable for integration into a planar photonic platform.
In this talk, I will discuss my current endeavors of novel photonics and optoelectronics functions using 2D materials integrated Si photonic, including light generation/detection and phase modulations[1,2]. I will discuss the challenges and opportunities of integrating 2D materials with Si photonic devices, and present experimental results that demonstrate their potential for enhancing device performance. Furthermore, I will demonstrate how these advancements can be leveraged to enable novel information processing techniques that can outperform conventional computers.

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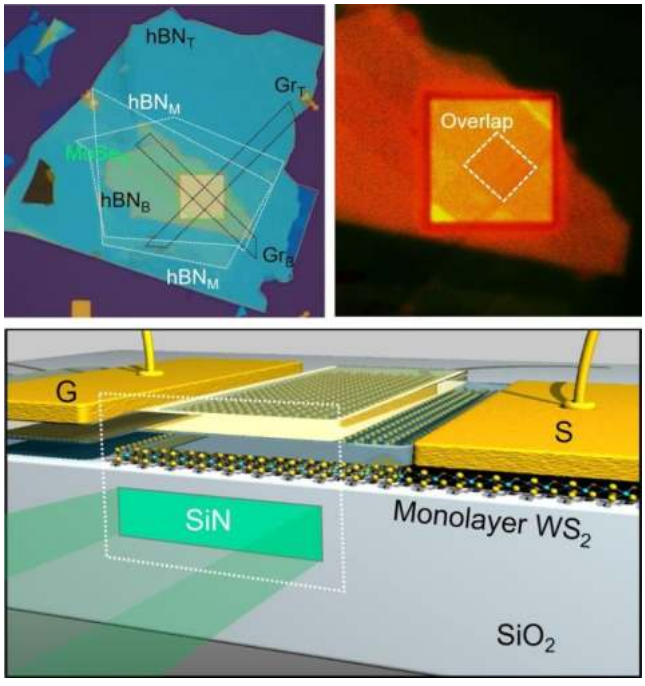

Figure 1: 2D materials-based optoelectronic devices and those integration into Si photonics platform. Light emitting device using MoSe2 (top) and WS2-SiN hybrid photonics for low-loss modulation (bottom).

Propagating Plexcitons in 2D Semiconductors Launched by Surface Acoustic Waves

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In this work, we theoretically demonstrate the strong coupling between excitons in 2D semiconductors and surface plasmons in a thin metal film forming exciton-plasmon polaritons (or plexcitons)that can be optically accessed by means of a surface acoustic wave (SAW). The strain field of the SAW creates a dynamic diffraction grating, providing the momentum match for the surface plasmons, whereas the piezoelectric field that could dissociate the excitons is cancelled out by the metal. This is exemplified for monolayer MoS_2 and mono- and few-layer black phosphorus on top of a thin silver layer on a $LiNbO_3$ piezoelectric substrate, providing Rabi splitting of 100-150 meV [1]. Thus, we demonstrate that SAWs are powerful tools to modulate the optical properties of supported 2D semiconductors by means of the high-frequency localized deformations tailored by the acoustic transducers, that can serve as electrically switchable launchers of propagating plexcitons suitable for active high-speed nanophotonic applications.

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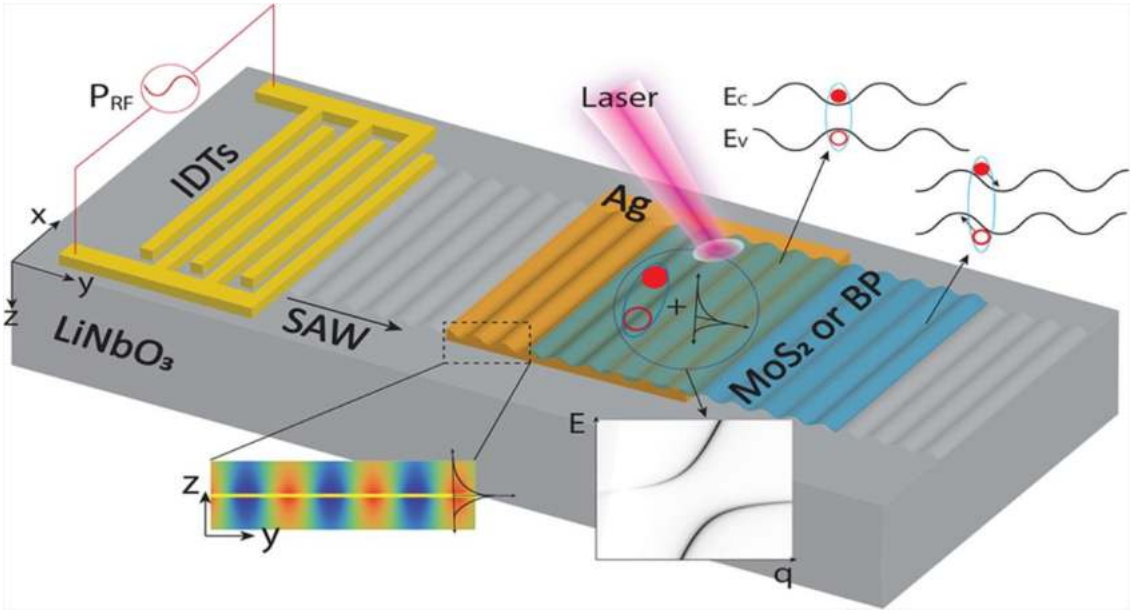

Figure 1: Schematic of the proposed plexciton launcher where a SAW generated by interdigitated transducers (IDTs) placed on top of a piezoelectric substrate leads to the coupling between metal plasmons and excitons in 2D semiconductors by means of the dynamic surface rippling and bandgap modulation.

Elementary excitations of quantum emitters in highly defective hexagonal Boron Nitride

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The recent discovery of bright and tunable quantum emitters (QEs) in hexagonal Boron Nitride (hBN) stable up to room temperature enables the realization of novel and scalable quantum photonic platforms [1,2]. While it is well-accepted that SPEs in hBN stem from defects [3], key details on their origin, electronic levels, and orbital involvements are still unknown. Here we interface, for the first time, measurements with resonant inelastic X-ray scattering (RIXS) and photoluminescence (PL) on pristine and plasma-treated hBN with high density of defects. RIXS measurements performed at the π* antibonding orbitals of Nitrogen uncover a fundamental excitation E_0 at 285 meV that generates harmonics with energy $E_n=nE_0$ (n=1,2, 3,...) ranging from the mid-IR through the UV (Fig.1a,b). These fundamental harmonics are observed only in highly defective samples and not in pristine samples. PL spectroscopy at low temperature indicates that highly defective samples host several QEs (Fig.1c). We analyse the QE emission pattern with a model that accounts for donor-acceptor-pair (DAP) recombination process. In the spectral range covered by our PL measurements, the DAP fit shows the presence of four transitions with energy overlapping the one of the harmonics with n=5,6,7,8 (Fig.1d). The correlation between the harmonics and the DAP transitions indicates that most of the QE observe in hBN have a common origin that can be ascribed to the N π* orbitals. Our interpretation generalizes QEs in the IR, visible, and UV range through a single energy scale explaining the stability and robustness of SPEs in hBN. Moreover, due to the orbital sensitivity of RIXS, we can underscore the association of QEs in hBN with the π* antibonding orbitals.

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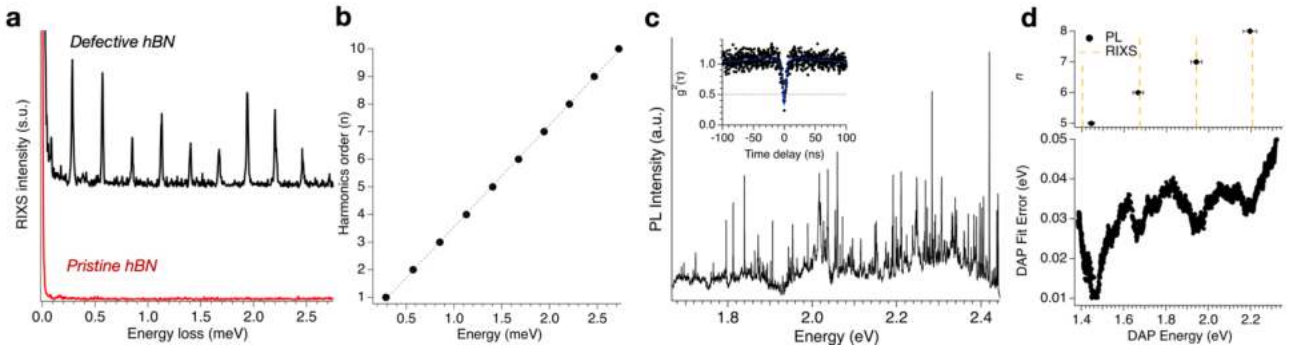

Figure 1: a – RIXS spectra taken at the N K pre-edge (π*) of defective (black curve) and pristine (red curve) hBN. **b** – Energy of the harmonics extracted from **a**. **c** – PL spectrum of defective hBN at T=5 K. Several sharp peaks indicate the presence of many QEs. The inset show an example of second-order correlation function ($g^2(\tau)$) measured on an individual line. Antibunching below 0.5 confirms the single-photon emission. **d** – Results of the DAP fit of the energy of the QEs measured with PL experiments. Four DAP transitions result from the fit as their energy minimize the fit error. The top panel shows the comparison between the energy of the DAP transitions and RIXS harmonics for n=5,6,7,8.

Terahertz Spin-Light Coupling in Proximitized Dirac Materials

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The atomically thin nature of two-dimensional (2D) van der Waals materials makes them highly susceptible to the influence of their neighbours, thereby enabling the design of their electronic band structure by proximity phenomena. We investigate spin-light coupling phenomena in van der Waals heterostructures consisting of two-dimensional (2D) Dirac crystals proximitized by a magnetic layer. A strong magnetic proximity effect results in the modification of the 2D electron spectrum with the appearance of different spin-splitting terms and electron's equilibrium spin polarization [1]. Here, we reveal a remarkable terahertz (THz) spin-light interaction in 2D Dirac materials that arises from proximity effects of magnetic and spin-orbital character. We demonstrate theoretically that the electric dipole spin resonance (EDSR) of Dirac electrons displays distinctive features in the THz range, upon emerging spin-pseudospin proximity terms in the Hamiltonian. To capture the effect of fast pseudospin dynamics on the electron spin, we develop a mean-field theory and complement it with a quantum-mechanical treatment
. As a specific example, we investigate the THz response of a single graphene layer proximitized by a magnetic substrate, using realistic parameters. Our analysis demonstrates a strong enhancement of the THz-light absorption with the increase of the spin-pseudospin coupling, pointing towards promising prospects for THz detection and efficient generation and control of spins in spin-based quantum devices.

The derived features of THz spin-light coupling suggest that EDSR could be a powerful experimental probe in the studies of proximitized vdW layers and to elucidate spin-dependent phenomena. The EDSR framework allows one to quantify proximity induced spin splittings and gives a direct access to spin-relaxation mechanisms. The derived description of coupled spin-pseudospin dynamics could be implemented in graphene quantum dots and nanoflakes, the basic elements realizing qubits for quantum computing in THz range.

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Adaptive Photocurrent Generation in ReS_2-2D Te Heterostructure

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Semiconducting layered materials (SLMs) have attracted much attention due to their versatile potential applications in photodetectors, phototransistors, and light-emitting diodes [1,2]. The devices based on these materials can be far more adaptable when they are hybridized in heterostructures [3,4]. Here, we report a heterostructure based on n-type rhenium disulfide (ReS_2) and p-type tellurene (2D Te). The light response of the heterostructure device was elucidated by the magnitude, phase (flow direction), and position of the light-induced current. Diverse photocurrent generation mechanisms were discovered at the interface of ReS_2-2D Te hybrid structure, including the photovoltaic effect (PV), photothermoelectric effect (PTE), and the hybrid of PV and PTE, rather than the expected PV only, which has been unprecedented for LSMs-based phototransistors so far. Furthermore, adaptive photocurrent can be achieved by controlling back-gate bias. These results are expected to contribute to the fundamental understanding of photocurrent generation in the heterojunction and develop new concepts of optical sensors with a designed detection mechanism.

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Figure

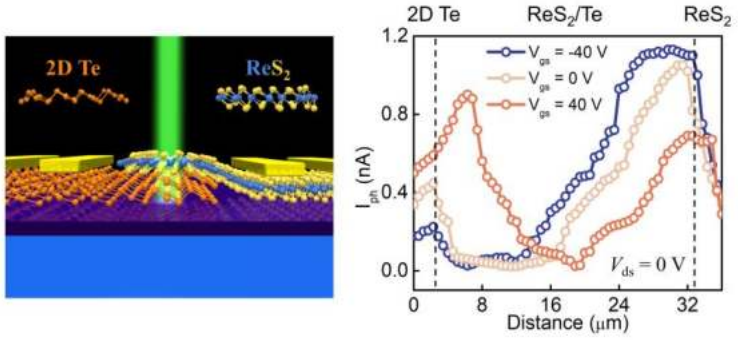

Figure 1: Adaptive photocurrent generation in ReS_2-2D Te Heterostructure at different back-gate biases.

Engineering versatile graphene oxide-based nanoplatforms for immunomodulation

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Abstract

The efficiency of current immunotherapeutic approaches is limited due to variable response rates resulting from the heterogeneity of immune mechanisms from patient to patient [1]. To overcome this issue, engineered nanoscale-based immunomodulation platforms able to improve specificity and durability of immunotherapy have been recently introduced [2]. Among their unique advantages are improved targeting to specific tissues followed by an enhanced cellular uptake, higher loading capacity as well as possible synergistic action between the immunomodulation platform and the immunotherapy [3]. In this frame, 2D materials characterized by a large available surface area are considered as suitable nanoplatforms for increased loading of biologically-active molecules. Graphene oxide (GO) nanosheets provide additionally the advantages of high colloidal stability and dispersibility in biological fluids, along with a biocompatibility and biodegradability profile that renders it an excellent carrier for medical use [4,5]. In the present study, GO was complexed non-covalently with a small immunomodulatory molecule (a synthetic TLR7/8 agonist) resulting in colloidally and chemically stable nanoplatforms (**Figure 1**). A simple and highly reproducible protocol for the fabrication of the thin and nanoscale GO-based drug complex will be presented. Moreover, the successful complexation has been demonstrated through a series of physicochemical characterizations along with *in vitro* biological studies demonstrating its immunomodulatory activity. The results of this study allow the future *in vivo* development of the present nanoplatform, and more generally encourage the use of non-covalent graphene-based nanoconstructs in biomedicine.

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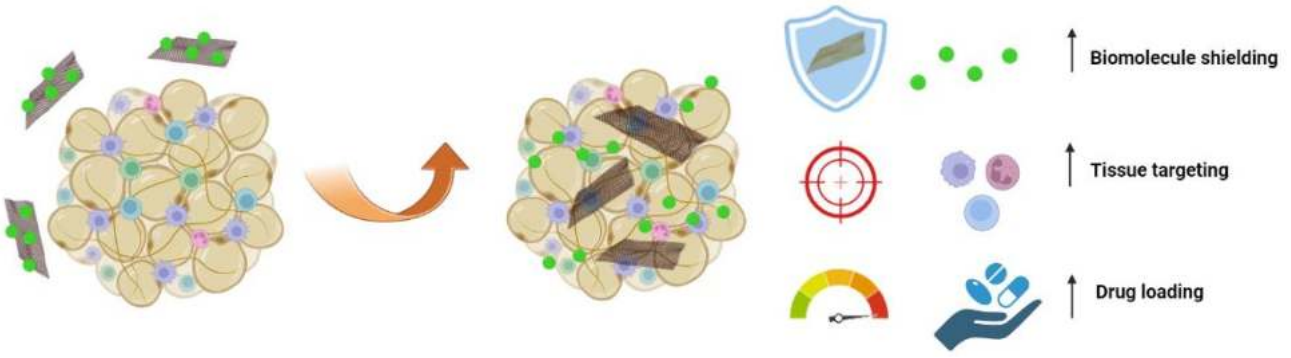

Figure 1: Schematic depiction of a GO-based immunotherapeutic platform.

Mechanically-flexible, graphene-based, microelectrodes for simultaneous recording and electrical stimulation of deep brain microstructures: an acute *in vivo* study

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ABSTRACT:
Parkinson's disease (PD) is a neurological motor disorder that negatively impacts the quality of life of its patients. Deep brain stimulation (DBS) is a well-established therapy used to alleviate PD symptoms. However, standard DBS systems consist of stimulating electrodes also known in the field as 'leads' that are mechanically rigid since metal is used in their construction. The rigid nature of these electrodes can result in excessive glial scarring, lead displacement, or fracture, limiting the longevity of the DBS system. In addition, standard stimulating leads are bulky (⌀ > 1 mm, larger than the brain structures they target) which not only lead to significant tissue damage, but also precludes capturing single unit activity. The ability to record from single units can help surgeons quickly localize deep brain structures and achieve accurate electrode placement required for efficacious DBS. Thus, to overcome the above limitations of current DBS systems, we have developed a mechanically-flexible, 8-channel, graphene-based microelectrode (⌀ < 1 mm) called Egnite where each channel can either be used for stimulating or recording. Sprague Dawley rats received an intracranial injection of either 6-hydroxydopamine (30 ug/4ul) or vehicle into their right Medial Forebrain Bundle. Four weeks post-surgery, the rats were anesthestized with an i.p. injection of urethane (1.2 g/kg) and subsequently underwent a burr hole procedure over the subthalamic nucleus (STN). The Egnite was lowered into STN using a microdriver at an insertion speed of 3 µm/s. The Egnite was able to electrographically map the STN i.e. channels within the STN recorded fast spiking activity (10-30 spikes/s) while those outside showed a few number of spikes. Once the STN was reached, a DBS protocol consisting of 75 µA biphasic pulses with duration of 100 µs/phase was applied at 100 Hz for 1 min. This was preceded and ensued by a 2 min period of recording to capture STN activity pre and post-stimulation respectively. The Egnite was able to reliably localize the STN and delivered DBS that suppressed the excessive firing of the STN neurons which is thought to underlie the motor symptoms of PD. In conclusion, we demonstrate that the Egnite reliably localized and delivered modulatory DBS to the STN.

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Graphene oxide ovalbumin nano-complexes in liquid formulation for immunomodulation

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Abstract

Graphene oxide (GO) is an ideal water dispersible and mechanically flexible, drug carrier nanomaterial, with a specific surface area of ≈ 740 m_2 g^{-1} [1], with chemically heterogeneous planar surface for functionalisation and tunable diffusion via controlled lateral size. *In vivo* biodistribution studies of bare GO [2, 3] have indicated that upon intravenous (i.v.) injection in mice the material is accumulated in the spleen, which has important immunological functions. Furthermore, recent studies have indicated that GO in complex with ovalbumin (OVA) can offer immunomodulatory activity *in vitro* and *in vivo* [4].

In order to assess the GO potential as a nano-carrier for drug delivery to the spleen we formulated and characterised pharmaceutical dispersions of GO in complex with ovalbumin (OVA). Then, we studied the *in vivo* biodistribution of the GO:OVA complexes, by dynamic whole-body imaging (μSPECT:CT), histology and Raman mapping. OVA exhibited affinity for the GO nanosheets, with AFM demonstrating that GO upon complexation with OVA protein was still dispersed as individual flakes of single to few-layer thickness. Raman indicated that the GO chemical structure was not alterered. The pH strictly influenced the dispersion stability, suggesting that adsorbed OVA prevented aggregation of the complexes by means of electro-steric stabilization. Remarkably, administration of GO:OVA complexes *in vivo* by i.v. injection showed clear positive Raman signal in the spleen, indicating a great potential of GO nanosheets as platforms for immunotherapeutic applications.

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Figures

Figure 1: Graphene oxide is a promising nanocarrier for the delivery of proteins to the spleen for immunomodulation applications.

Induction of Neural Stem Cell Differentiation in 3D Bioprinted Scaffolds Containing 2D Materials

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Abstract:

Studies on neural scaffolds generated by bioprinting of cells and conductive nanomaterials are very limited and available nanotechnology-based approaches do not provide enough nerve regeneration and a complete functional recovery. Due to its extraordinary features including immunomodulatory properties, high electrical conductivity, chemical stability and mechanical properties, graphene based 2D materials attract great interest in regenerative medicine [1]. In order to propose a novel strategy to achieve a better functional recovery during neural tissue regeneration, for the first time in literature, simultaneous bioprinting of adipose derived stem cells or neural stem cells together with different 2D materials was performed. Following 3D bioprinting, the structure of these scaffolds, cell viability, differentiation status, mechanical and electrical properties were determined in vitro [2]. Scaffolds were also transplanted in vivo in order to further prove the regeneration capacity following nerve damage. 3D bioprinted and 2D material containinig scaffols enhanced the differentiation status of stem cells without affecting cell viability and proliferation. Electrical stimulation further improved this material based effect on the differentiation capacity of stem cells. Action potential measurements showed that functional neurons can be obtained in these scaffolds. In vivo transplantation resulted in improved neural tissue regeneration in rats. Overall, results suggested that 3D bioprinting of stem cells and 2D nanomaterials is a potential approach in neural tissue regeneration [3].

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New Graphene Pharmaceutical Formulations for Phototherapy of Skin Cancer: *in vitro* validation and *ex-vivo* human skin permeation studies

Artur M. Pinto

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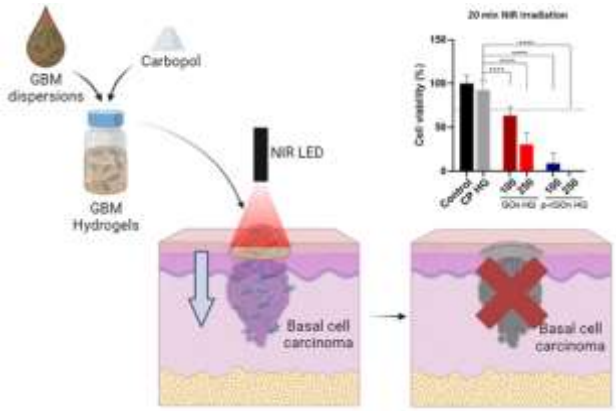

Basal cell carcinoma (BCC) is the most common form of human cancer, and treatment usually involves surgery. However, several non-invasive strategies such as photothermal therapy (PTT) have been explored. Graphene-based materials (GBMs) are good candidates to act as photothermal agents since they can absorb near-infrared (NIR) light energy that can induce hyperthermia, leading to tumour cells apoptosis [1]. In this study, we proposed the use of nanographene oxide (GOn) and partially-reduced graphene oxide (p-rGOn) as platforms for photothermal therapy of BCC. GO was produced through the modified Hummers method, and further ultrasonicated to obtain GOn. GOn was then photo-reduced to p-rGOn, a new water stable material recently developed at our laboratory using an innovative industrially scalable process. Both materials were incorporated in carbopol hydrogels (HG) to produce pharmaceutical formulations that could be administered to patients skin, and characterized physical-chemically. GBMs were obtained with mean lateral dimensions of 216 ±77 nm (GOn) and 206 ±107 nm (p-rGOn). GOn and p-rGOn HG showed zeta potential values of -49.2 ±3.4 and -50.0 ±3.3 mV, respectively. After 30 min irradiation with a near-infrared photothermal therapy lamp source (15.70 mW cm^{-2}), GOn HG increased temperature to 45.7 °C, while p-rGOn HG reached 48.2 °C. GBM HG (250 µg mL^{-1}) have been shown not to affect human skin fibroblasts (HFF-1) morphology or viability. Therefore, GBM HG photothermal effect was tested towards a human squamous carcinoma cell line (A-431). After 20 min irradiation, p-rGOn HG (250 µg mL^{-1}) completely eradicated cancer cells, confirmed through cell viability and immunocytochemistry studies. GBM HG *ex vivo* human skin permeability (from a healthy patient, S. João Hospital, Porto) was evaluated using a Franz cell system. Materials were capable of permeating across skin in a time-dependent manner. After 6 h of skin contact, 71.7 % or 83.9 % of GOn or p-rGOn, respectively, reached the receptor compartment, which means that a tumour could be topically infiltrated with the materials. This is the 1st pharmaceutical formulation ever reported to deliver graphene through skin for cancer therapy. *In vivo* studies are ongoing.

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Exploitation of the 2D graphene oxide biomolecule corona in secretome-based cancer biomarker discovery

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Nanotechnology has emerged as a promising tool for cancer biomarker discovery. Nanoparticles (NPs) undergo rapid modification once they come into contact with the biological milieu to form a "biomolecule corona" due to their interfacial reactivity [1]. Analysis of the biomolecule corona by mass spectrometry-based proteomics has shown an enhanced discovery of previously unidentified low-abundant proteins and has attracted significant interest as a promising technology in cancer biomarker discovery [2].

Graphene oxide is a two-dimensional (2D) nanomaterial with a distinctively large surface area and high surface reactivity [3]. In this study, the biomolecule corona formed around graphene oxide nanosheets is exploited to provide an in-depth analysis of the secretome and identify unique proteomic signatures of different cancer cell lines.

The secretome of lung cancer (A549), glioblastoma (GL261 and U251) and cervical cancer (HeLa) cell lines was obtained by harvesting the conditioned media of the cultured cancer cells. The collected secretome was incubated with graphene oxide nanosheets to form the biomolecule corona. Using the 2-step NanoOmics purification protocol [4], the graphene oxide biomolecule corona was isolated via a combination of size exclusion chromatography and membrane ultrafiltration. Proteomic mass spectrometry analysis of the isolated biomolecule corona showed a significant increase in the number of identified proteins in the corona-processed secretome of all the cancer cell lines when compared to the unprocessed secretome samples. Significant enrichment of low-abundance secreted protein was observed due to the corona-processing. Ultimately, the graphene oxide corona-processing protocol enhanced the discovery of uniquely secreted proteins from different cancer cell lines.

In the future, we plan to utilize the graphene oxide biomolecule corona platform to correlate the cancer secretome proteomic fingerprints with proteomic analysis of plasma samples obtained from cancer patients, with the ultimate goal to identify highly-specific blood biomarkers for cancer early detection.

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Graphene-based surface plasmon resonance biosensors for multi-parametric analysis of living cells

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White blood cells (leukocytes) are a key component of the immune system. Changes in their composition and biological activity are important markers of health status. Studies of multiple activity parameters, e.g. densities of various receptors, would allow earlier and more accurate disease diagnosis. Due its high sensitivity, graphene is used as a component in different types of optical and electrical biosensors [1,2]. Among existing biosensing techniques for cell analysis, surface plasmon resonance (SPR) is very promising because it combines high sensitivity with label-free and real-time read-out. The integration of graphene in SPR-based sensors may further increase sensor performance. To test this hypothesis, we transferred large-area CVD-grown graphene onto SPR biosensor chips. Novel synthetic peptides were chemically attached to graphene via pyrene-based linkers. These high affinity peptides can specifically interact with receptors on the cell surface (Figure 1, left). Living model cells with controlled expression of specific receptors were used to study the peptide-cell interaction on graphene-coated SPR chips. We observed a strong signal upon binding of cells with receptors on peptides (Figure 1, right). Control measurements with cells without receptors showed much lower signal due to non-specific binding. Preliminary data also suggests that graphene reduces non-specific binding compared to standard gold-coated SPR chips. Unlike conventional cell analysis methods, our technology is label-free and allows analysis of biological cell activity with high temporal resolution.

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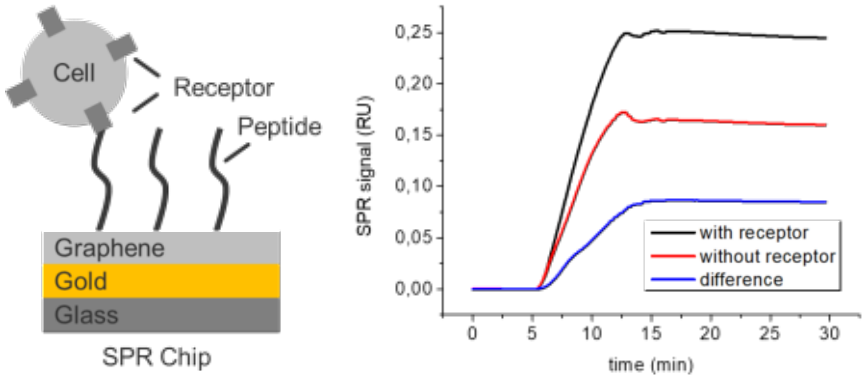

Figure 1: Binding of cells on biosensors with graphene and peptide coating. Left: Schematic sensor setup. Right: SPR signal as a function of time. Here, the binding of cells with and without receptors was compared. The difference between the two measurement curves gives the proportion of specific binding to peptides. The cell concentration was 275,000/ml in both cases.

TMD Engineering of 2D-Magnetic Tunnel Junctions – From Barriers to Electrodes

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Spin-based electronics has already revolutionized data storage and readout technologies. Nowadays it targets a variety of new architectures like embedded MRAMs, spin logics or neuromorphic computing, which makes it one of the most promising post-CMOS approaches. Meanwhile, 2D materials and their combination in heterostructures have opened novel exciting opportunities in terms of functionalities and performances for spintronics devices. The broad family of 2D materials offers many possibilities to engineer the properties of layered stacks and devices in particular via interfacial exchange and proximity effects. One very attractive topic is the field of MTJs based on 2D materials (2D-MTJs).[1] Being able to control the physical behaviour of the MTJ is of crucial interest, in order to improve their functionality and efficiency. From major influence herby are electrodes and the tunnel barrier, in terms of interfaces and band alignments. Recent work utilised 2D materials to enhance barriers and their performance. Graphene has proved its strong potential as a barrier for MTJs with evidence for spin-filtering through band structure or strong hybridization effects (i.e. spinterface) achieving a record spin polarization of up to -98%. In parallel, advances within the broad Transition Metal Dichalcogenides family of 2D semiconductors and 2D ferromagnets have opened new possibilities to tailor spintronics properties further. As an example, we will show how TMDs could be integrated into a hybrid spin-valves 2D-MTJs and show layer-dependent spin filtering effects. We can show that the spin polarisation can be reversed depending on the number of layers. The layer thickness largely influences the band structure and thus allows control over the open spin channels for vertical electron transport. We will also discuss how to reach one step further with the large scale integration of these materials into tailored 2D heterostructures. For this we developed 2D ferromagnets based on $Fe_{3+x}GeTe_2$ which can act as a spin source. We will show that they can be grown in large scale using Pulsed laser deposition (PLD) and reach curie Temperatures (T_c) above room temperature (RT) while being integrated with other TMDs. We will highlight how these PLD grown ferromagnetic 2D layers could further reinforce the 2D materials family's potential for 2D-MTJs and how they open the way for the design of in-situ full 2D MTJ fabricated devices with artificial properties. [2,3]

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Investigating ballistic transport via 1D graphene/FM spin injectors

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Despite its great promise for spintronics, experimental values for spin transport parameters extracted from graphene are currently significantly below theoretical predictions. Our group recently reported encouraging results from a state-of-the-art device architecture, constituting a fully encapsulated single layer graphene channel with one-dimensional (1D) ferromagnetic (FM) contacts, in which we observed record high charge mobility for a spintronic device, and long-range spin transport [1]. Full encapsulation of the channel in hexagonal boron nitride preserves the quality of the graphene, giving rise to the high charge mobilities we observed, while 1D contacts mitigate the high levels of doping associated with tunnel contacts [2, 3]. Here, we explore how the nanoscale geometry and potential profile of the graphene/FM interface places transport across it in the ballistic regime (Fig. 1a). This allows for achieving sizeable contact resistance without the need for tunnel barriers, as well as for realising quantum transport phenomena such as quantized conductance across the 1D graphene/FM junction, previously unexplored in such a device. We have focussed on ballistic transport through these junctions, at low temperature. Bias spectroscopy measurements demonstrate quantized conductance through the junction, displaying 1D-subbands at fractions of the conductance quantum, G0 - indicating a transmission factor $T \approx 0.25$ (Fig. 1b). Furthermore, application of an out-of-plane magnetic field leads to better defined quantization, due to a transition into the quantum Hall regime [4]. Finally, we investigate the effect of quantized conductance at our contacts, on spin transport through the channel of our devices. Quantized conductance arising from nanoscale 1D FM contacts, in the absence of a fabricated graphene nanoconstriction, is a previously unreported result and demonstrates a path for the development of ballistic spintronics.

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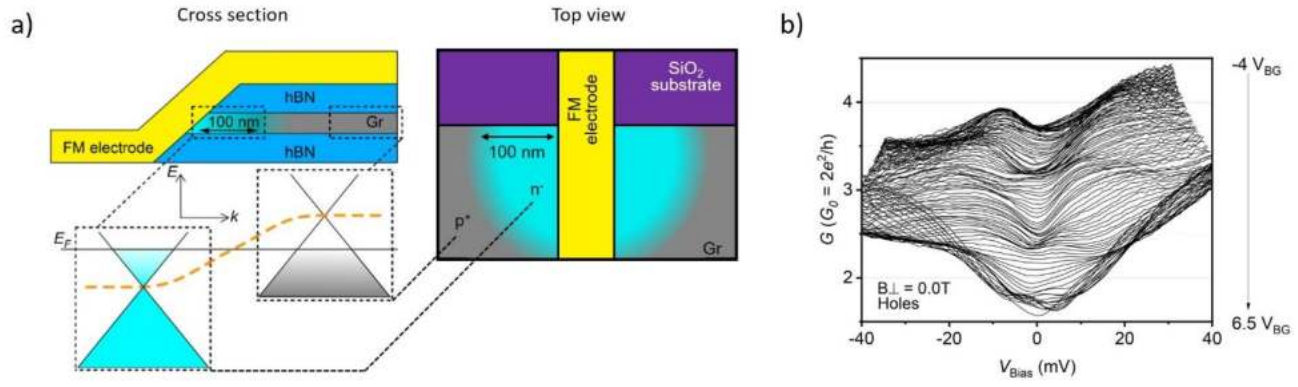

Figure 1: (a) 2D cross section and top view of the 1D contact architecture. Figure adapted from [1]. (b) Bias spectroscopy conductance measurement of a 1D contact, for hole transport ($V_{BG} > V_D$) without an applied magnetic field

Local atomic stacking and symmetry in twisted graphene trilayers

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Moiré superlattices formed from twisting trilayers of graphene are an ideal model for studying electronic correlation, and offer several advantages over bilayer analogues, including more robust and tunable superconductivity and a wide range of twist angles associated with flat band formation. [1,2] Atomic reconstruction, which strongly impacts the electronic structure of twisted graphene structures, [3] has been suggested to play a major role in the relative versatility of superconductivity in trilayers. Despite this, atomic reconstruction has only been probed using indirect measurements or those only applicable to exposed samples. [4] Here, we exploit an inteferometric 4D-STEM approach to image a wide range of trilayer graphene structures. Our results unveil a considerably different model for moiré lattice relaxation in trilayers than that proposed from previous measurements, informing a thorough understanding of how reconstruction modulates the atomic stacking symmetries crucial for establishing superconductivity and other correlated phases in twisted graphene trilayers. [5]

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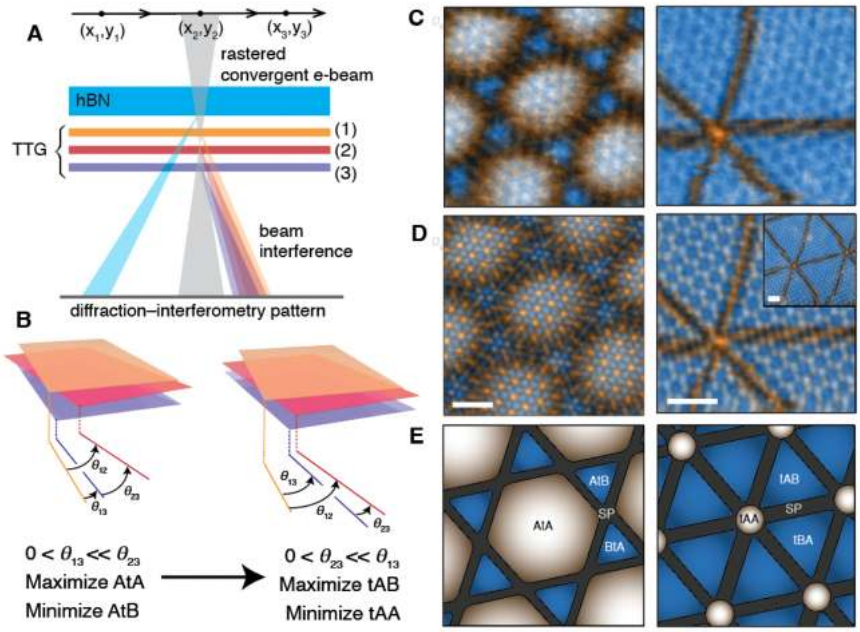

Figure 1: (A) Schematic of the 4D Scanning Transmission Electron Microscopy (STEM) approach, wherein beam interference is used to extract stacking order. (B) Schematics of layer alignment in TTLG. (C) Maps of local atomic stacking from the larger moiré pattern only.(D) Local atomic stacking obtained from considering all three graphene layers. (E) Qualitative schematic illustrating the atomic reconstruction observed.

Long distance all electrical magnon transport in the van der Waals antiferromagnet $CrPS_4$

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Magnon spintronics studies the transport of spin currents through an insulating and magnetically ordered material using magnons [1]. Electrically and thermally induced magnons can be transported in insulating ferromagnetic[2] and antiferromagnetic materials [3]. In particular, antiferromagnetic materials have interesting properties for future spintronic applications: they possess no net magnetic moment and are therefore robust against magnetic perturbations and have ultrafast dynamics. We demonstrate for the first time all electrical long-distance magnon spin transport in the electrically insulating antiferromagnet chromium thiophosphate (CrPS4) with perpendicular magnetic anisotropy. The spin currents are injected electrically, via the spin Hall effect and detected via the inverse spin Hall effect using the nonlocal geometry (see Fig. 1) as described in Ref.[2]. We monitor the non-local resistance as a function of an in-plane magnetic field up to 8 Tesla. We observe a non-local resistance over distances up to at least a micron below the Neel temperature (T_N = 38 Kelvin) close to magnetic field strengths that saturate the sublattice magnetizations [4]. These results herald the potential of 2D van der Waals magnets for scalable magnonic circuits.

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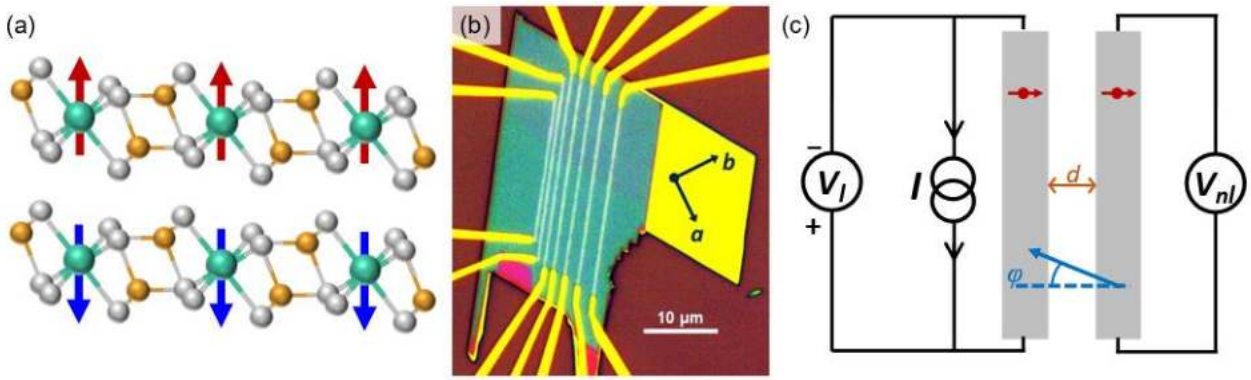

Figure 1: a) Bilayer of CrPS4, arrows representing magnetic moments, b) optical image of device with Pt contacts on CrPS4 flake, c) Circuitry of non-local measurements.

Correlation effects on topological end-states in finite-size armchair graphene nanoribbons

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Finite-size armchair graphene nanoribbons are expected to host topological states that are spatially located at the two zigzag ends of the ribbons (see fig.1). The number of topological states is predicted by topological invariant defined for periodic ribbons and depends on the number of carbon atoms in the ribbons' width. These classifications on the ribbons' width rely most often on topological invariant evaluation based on tight-binding Hamiltonian models, i.e. with no interaction between electrons [1, 2, 3]. One can include interaction on top of a tight-binding models by inserting new terms in the model Hamiltonian and adopting the Hubbard Hamiltonian. This model is a well-known model to describe graphene and graphene nanofragments but is often treated in a mean-field approximation. In this approximation, each electron interacts with a mean-field resulting from all the electrons. No direct two-electron interactions are accounted for and the correlation is neglected. Treating the Hubbard model with the exact interaction terms and the total interaction for a system with more than a few tens of electrons is currently intractable from a numerical point of view. In-between approximations – restoring a part of the correlation but numerically tractable – are thus important. In this study, we investigated in more details the so-called GW approximation [4, 5, 6]. In particular, we investigated how this correlation impacts the topological end-states in graphene nanoribbons and show how their energies or spatial properties are affected. We also compare our results to published experimental data, showing a greater agreement when correlation effects are included.

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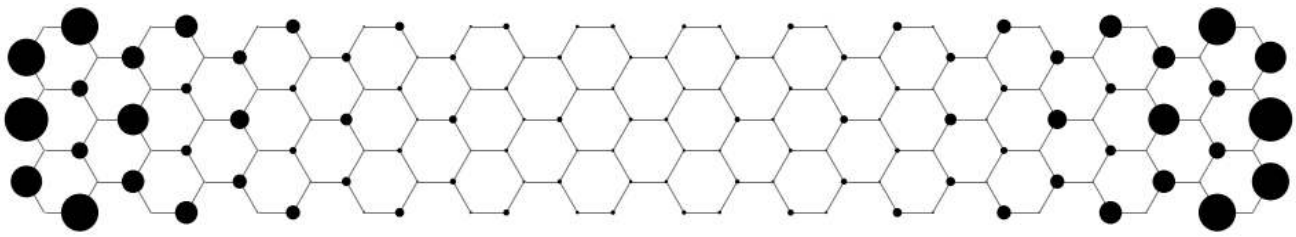

Figure 1: Electronic density of a topological end-states in a 7-atom width armchair graphene nanoribbon. The size of the dots is proportional to the mean densities of electrons.

Room-temperature proximity-induced Anomalous Hall Effect in Graphene Coupled to A Ferromagnetic EuO

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Abstract

Graphene has emerged as a potential candidate in exploring the exciting field of spintronic as it can transport spin information with high efficiency. It has a long spin diffusion length at room temperature and can control the degree of freedom of spin through the spin relaxation mechanism. When kept in proximity to a ferromagnetic material like EuO a large spin polarization has been predicted [1] opening the way to various interesting phenomenon and new spintronic device concepts [2]. In this work we describe the experimental efforts which includes the growth of magnetic Europium Oxide magnetic thin film by an original Molecular Beam Epitaxy method, and their structural and magnetic characterizations. Finally, we present transport measurements revealing Anomalous Hall effect in Graphene in proximity with EuO thin film. Two types of magnetic order are observed where a transition occurs from a ferromagnetic (FM) behaviour to a superparamagnetic (SPM) like behaviour when increasing the temperature. Surprisingly, a large SPM signal survives up to room temperature which could make possible new spintronic applications in graphene.

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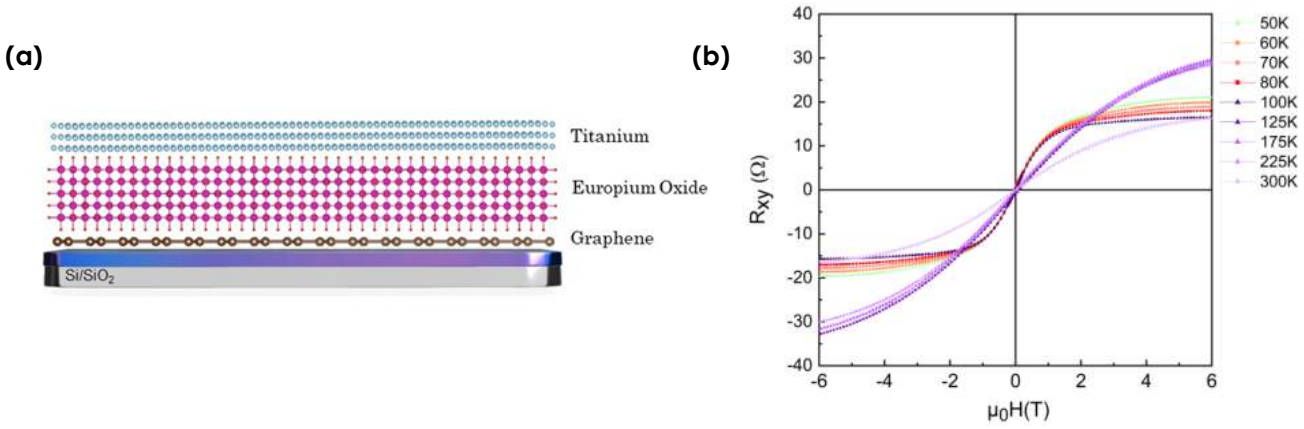

Figure: (a) Sketch of Graphene/EuO heterostructure. **(b)** Magneto-transport measurements – $R_{xy}(H)$ curves at different temperatures.

Ballistic transport in graphene on WSe_2

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Abstract

Proximity-induced spin-orbit coupling (SOC) was shown to be strong in graphene on transition metal dichalcogenides (TMDCs), and a band splitting induced by both Rashba and valley-Zeeman SOC terms is expected in such a system [1]. Here we employ the transverse magnetic focusing (TMF) technique [2] to study the effects of enhanced SOC in monolayer graphene on WSe_2, including the SOC-driven band splitting and electron dynamics, where the ballistic motion of electrons was used, instead of the spin relaxation due to electron scattering in most previous studies. We clearly observed a splitting in the first focusing peak whose evolution in carrier density and magnetic field can be well fitted theoretically by an overall SOC strength of ~13 meV, while no splitting the second focusing peak indicates an interband scattering at the sample edge [3]. Temperature dependence analysis further shows the possible suppression of the electron-electron scattering in the system. We will further discuss potential impact of this study especially in utilizing ballistic transport effects in graphene systems for spintronic applications.

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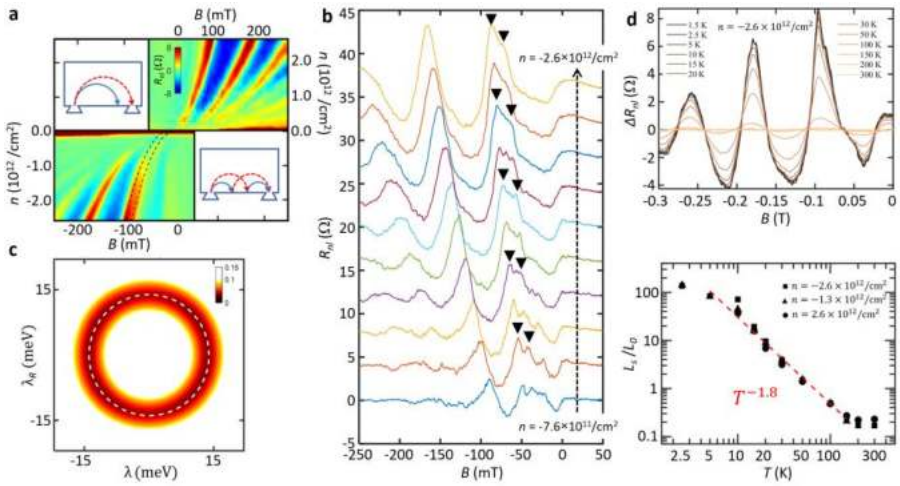

Figure 1: TMF study on monolayer graphene on WSe_2. **a**, Color-scale map of TMF signal measured at 1.5 K. The broken lines show the theoretically calculated focusing peaks when the overall SOC strength is 13.9 meV. Inset: carrier trajectories for the first and second focusing peaks (top left and bottom right, respectively). **b**, 1D cuts of the data shown in **a**. The black down-triangles mark some of the two split peaks for guidance. **c**, The color-scale map of the average difference of spin-split peak positions derived from theory and experiment, as a function of valley-Zeeman SOC (λ) and Rashba SOC strength (λ_R). **d**, Temperature dependence of the TMF spectra at n=-2.6×10^{12} cm^{-2} and the relative scattering lengths as a function of temperature plotted in a log scale, which follows the $T^{-1.8}$ dependence, indicated by the dashed red line.

Correlated phases near the van Hove singularity in trigonally warped bilayer graphene

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Diverging density of states can lead to correlated phases in low dimensional systems. This includes the graphene family that hosts electric-field controlled Lifshitz transitions and concomitant van Hove singularities in the density of states. Here, we present the observation of experimental signatures consistent with various interaction-driven phases in naturally occurring AB bilayer graphene including the fractional metals of Stoner type [1]. More prominently, we have found competing nontrivial insulating and metallic phases that exhibit intriguing temperature dependences and nonlinear I-V characteristics at zero magnetic field [1]. Evidencing interacting physics in this simple and reproducible system offers a fertile ground for exploring intricating many-body phases.

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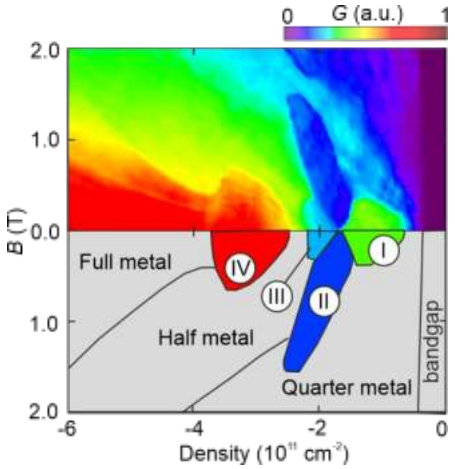

Figure 1: Conductance map as a function of charge carrier density and magnetic field B at a large electric displacement field measured in encapsulated bilayer graphene at a temperature of 10 mK. Several correlated phases of Stoner (quarter metal and half metal) and non-Stoner type (phases I-IV) appear.

Spin injection control in High quality Graphene 1D-contact systems

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To deliver spintronics applications, spins need to undergo controlled injection, transport, and detection. Progress on high-quality graphene structures [1] opened the route towards ballistic channels and spin coherence phenomena in graphene. Employing a 1D contact architecture [2], fully hBN-encapsulated graphene is contacted via electrodes by means of its edge (Fig. 1a). Our devices yield exceptionally clean spin-transport channels, with minimal doped regions within ~100 nm near the contacts, by virtue of the metallic contacts touching the graphene channel only at its edge between the encapsulating hBN layers (Fig. 1b). We report spin transport at low temperature with mobilities up to ~ 130 000 $\mathrm{cm}^2/\mathrm{Vs}$, spin relaxation lengths of ~ 18 $\mu\mathrm{m}$, and mean a free path of ~ 1 $\mu\mathrm{m}$. Thanks to the 1D contact resistance, R_c, ranging from $3 - 15\ \mathrm{k}\Omega$, and a smaller spin resistance of the channel, R_s, we observe different values of the R_c/R_s ratio, and the corresponding efficiency of spin injection, at different carrier densities which leads to the tunability of the measured spin signal (Fig. 1c) [4]. Finally, we demonstrate the introduction of an out-of-plane spin component onto graphene. In the geometry of our device the electrode climbs up the hBN-Gr-hBN heterostructure acquiring a partially vertical magnetization. As a result, we simultaneously observe both spin precession and spin-valve phenomena implying spins possess components both inside the plane of the device and perpendicular to it (Fig 1d), permitting alternative anisotropy studies to those currently in use [3]. Together, these observations demonstrate a system where ballistic spin injection, tunable spin signal and effective oblique spin injection is possible by exploiting the device architecture.

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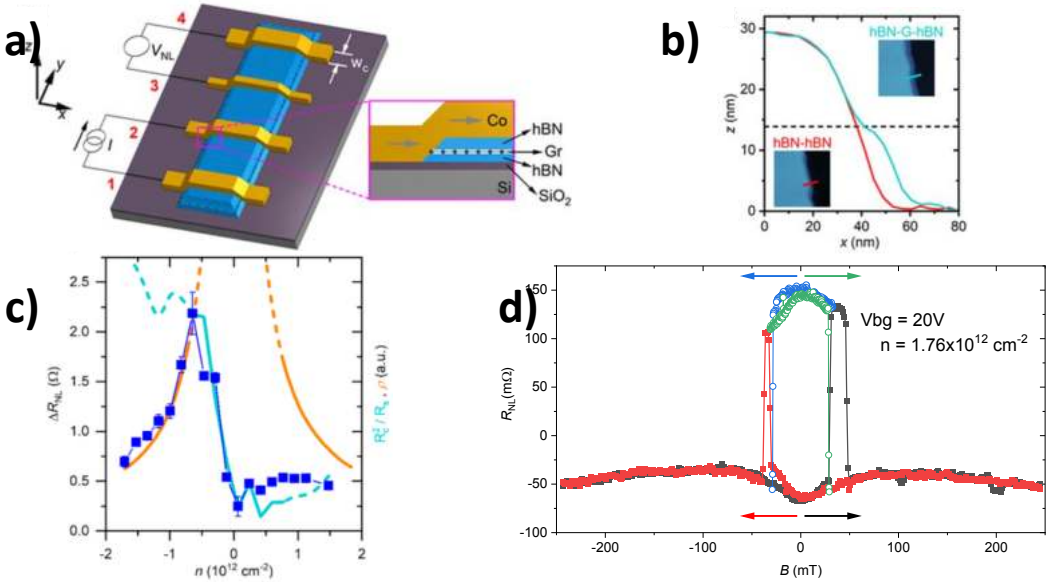
Figure 1: (a) Schematic of our device with a typical non-local connection showing a cross section of



the 1D contact. (b) AFM profile of the edges of a device comparing hBN-only vs hBN-encapsulated graphene. (c) Spin signal ΔR_{NL} as a function of charge carrier density, compared to trends in resistivity(orange) and the contact-to-spin-resistance ratio (cyan). (d) Spin-valve measurement showing a Hanle-like baseline revealing presence of spin components in-plane and out-of-plane.

Spin transport in CrXY monolayers: multiscale computational study

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Two-dimensional magnetic materials show a high potential for spintronic devices [1] thanks to intriguing phenomena at their interfaces. Their experimental investigation, while being actively performed, poses many challenges and numerical simulations can be of great help by pin-pointing high-gain materials and guiding the experimental research [2]. In this study we use a complete set of computational techniques to assess the suitability of CrXY [X, Y ∈ {S, Se, Te}] monolayers for spintronics, focusing on spin-momentum locking, highly relevant for spin manipulation by an electric current. We start from *ab initio* calculations and create tight-binding models [3] further used to compute the exchange parameters, Curie temperature and anomalous Hall conductivity. We find spin-momentum locking of complex forms which is shown to come from higher-order terms in the in-plane momentum expansion of the spin-orbit Hamiltonian [4]. Along with the calculated structural properties and magnetic anisotropy, this allows us to draw important conclusions about the possible use of CrXY monolayers in spintronic devices.

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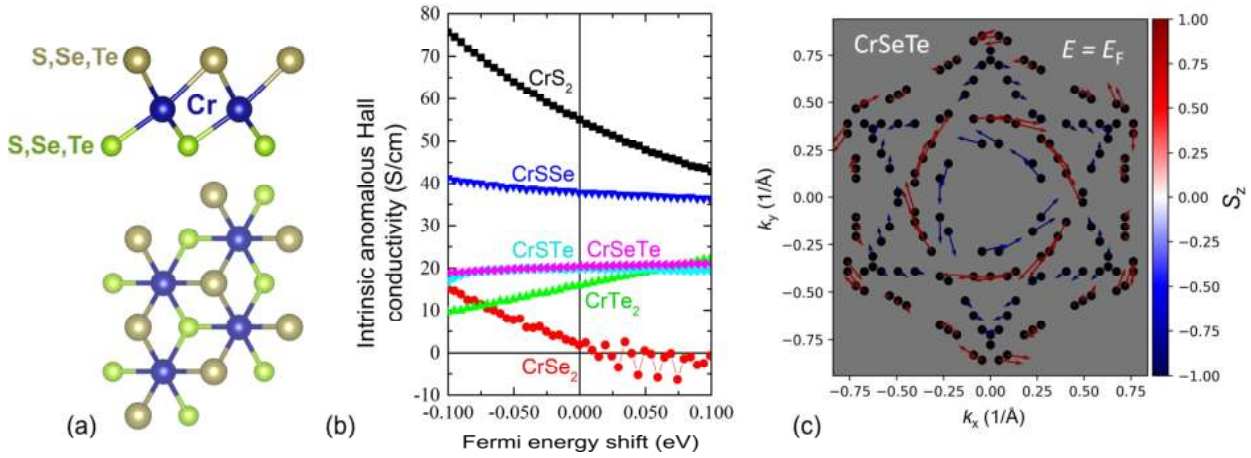

Figure 1: *Ab initio* calculations of CrXY [X, Y ∈ {S, Se, Te}] monolayers in the 1T phase. (a) Crystal structure of the monolayer 1T phase. (b) Calculated intrinsic anomalous Hall conductivity as a function of Fermi energy shift for the 6 materials. (c) Spin texture at the Fermi surface of a monolayer CrSeTe as a function of the in-plane wave vectors. The asymmetrical structure leads to an effective out-of-plane electric field and resulting Rashba-like spin-momentum locking.

A Graphene Nanoplatelet-Polydopamine Molecularly Imprinted Biosensor for Ultratrace Creatinine Detection

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Abstract

Accurate and reliable analysis of creatinine is clinically important for the early detection and monitoring of patients with kidney disease. [1] We report a novel graphene nanoplatelet (GNP)/polydopamine (PDA)-molecularly imprinted polymer (MIP) biosensor for the ultra-trace detection of creatinine in a range of body fluids. Dopamine hydrochloride (DA) monomers were polymerized using a simple one-pot method to form a thin PDA-MIP layer on the surface of GNP with high density of creatinine recognition sites. This novel surface-MIP strategy resulted in a record low limit-of-detection (LOD) of 2×10^{-2} pg/ml with a wide dynamic detection range between 1×10^{-1}-1×10^{9} pg/ml. The practical application of this GNP/PDA-MIP biosensor has been tested by measuring creatinine in human serum, urine, and peritoneal dialysis (PD) fluids. The average recovery rate was 93.7–109.2% with relative standard deviation (RSD) below 4.1% compared to measurements made using standard clinical laboratory methods. Our GNP/PDA-MIP biosensor holds high promise for further development as a rapid, accurate, point-of-care diagnostic platform for detecting and monitoring patients with kidney disease. [2]

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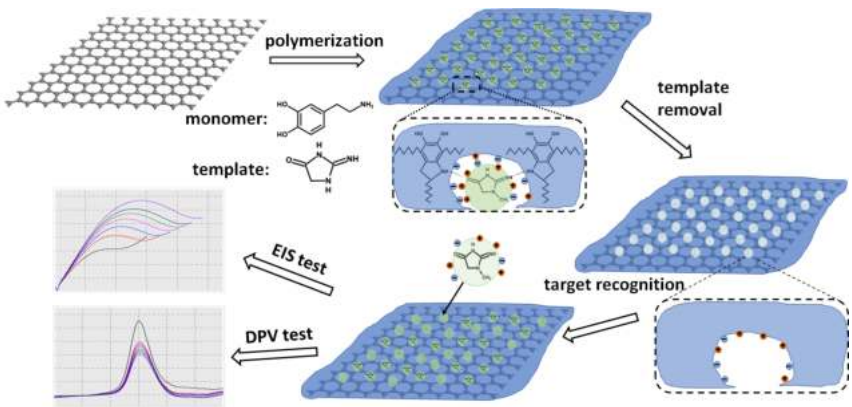

Figure 1: Schematic representation for the fabrication and application process of GNP/PDA-MIP.

Sensitive Piezoresistive sensors made of Graphene-based 3D ordered porous structures for wearable electronics

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Recent years have shown significant progress in developing stretchable electrodes, which remain the fundamental building block in flexible and wearable electronic devices [1]. The excellent properties of graphene have made it the best candidate for the next generation of flexible electronics applications. Wearable sensors require several properties such as high flexibility, stretchability, lightweight, and inexpensive fabrication, and they have to be suitable for integration with electrical components. One solution is the combination of nanostructures that act as sensors and polymers that guarantee the flexibility of the device. Different methods have been proposed in the literature to achieve these essential properties [2]. However, all techniques develop a flexible sensor that is completely enclosed by the polymer, which reduces the effect of external stimuli, limiting the graphene sensitivity. In order to improve the sensitivity of the piezoresistive system, here we present a device in which the 3D polymer skeleton is covered by graphene layers grown by the CVD method, as shown in figure 1a.
We investigated the electrical and mechanical properties of the graphene/polymer 3D structures by measuring their electrical resistance variation as a function of compressive and tensile strain. In this presentation, the optimised methodology to produce these materials will be presented and the results obtained discussed.
The realization of a piezoelectric sensor with an exposed graphene surface would be exploitable for several applications, such as biosensing, where the device will also be sensitive to external analytes.

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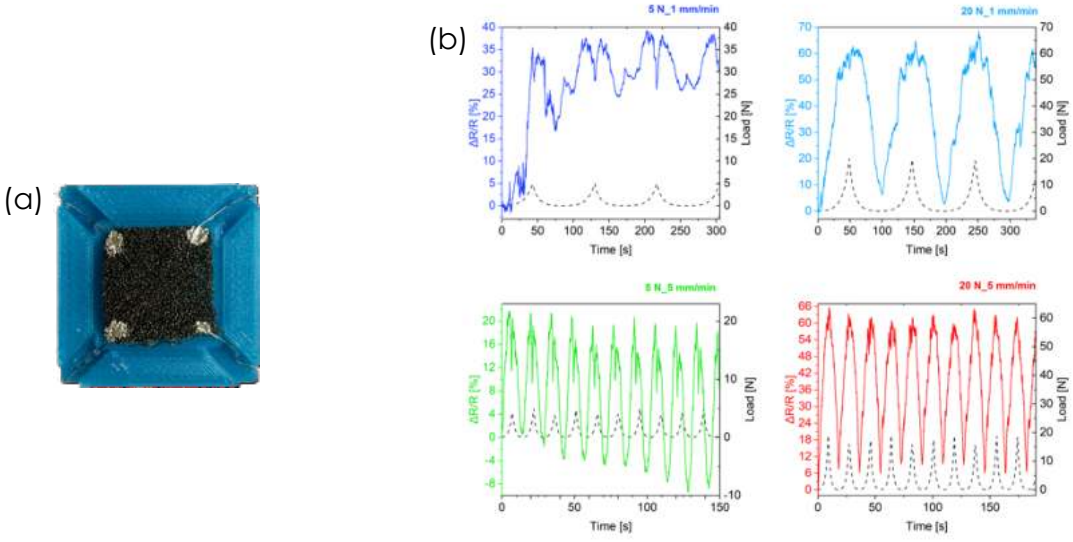

Figure 1: (a) 3D Graphene/PDMS foam contacted with silver paste. (b) Electrical-resistance variation of the composite as a function of compressive strain

Impact of Carbon and Hydrogen Doping on Stability and Mechanical Properties of Amorphous Boron Nitride

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Abstract

Amorphous boron nitride (aBN) has been revealed as an ultralow dielectric constant material (κ<2), with strong thermal stability and mechanical properties, making this material suited for next generation interconnects technologies [1, 2]. The uniqueness of amorphous materials derives from the inherent imperfect structure nature, which can be controlled at fabrication level, represents the key ingredient for specific target applications. In this respect, new fabrication strategies to modify the structural properties and a systematic theoretical characterization of the impact of the structural properties on thermal stability and mechanical properties are urgent. In this talk, a theoretical investigation of thermal and mechanical properties of aBN as a function of varying external parameters such as temperature, quenching rate, presence of unwanted or dopant atoms using molecular dynamic simulations will be presented. Using machine learning technique, we ensure the reliability of calculations of properties by first describing the atomic interactions more accurately, introducing two Gaussian Approximation Potentials [3] (for aBN:C and aBN:H) which are trained on a large dataset of atomic structures which generated ab-initio calculations [4,5]. We found that then incorporation of both C and H atoms causes a significant change in atomic environment of aBN, which is strongly reflected in the resulting the thermal stability and mechanical properties of the compounds [4]. We will also discuss the antioxidation properties of aBN.

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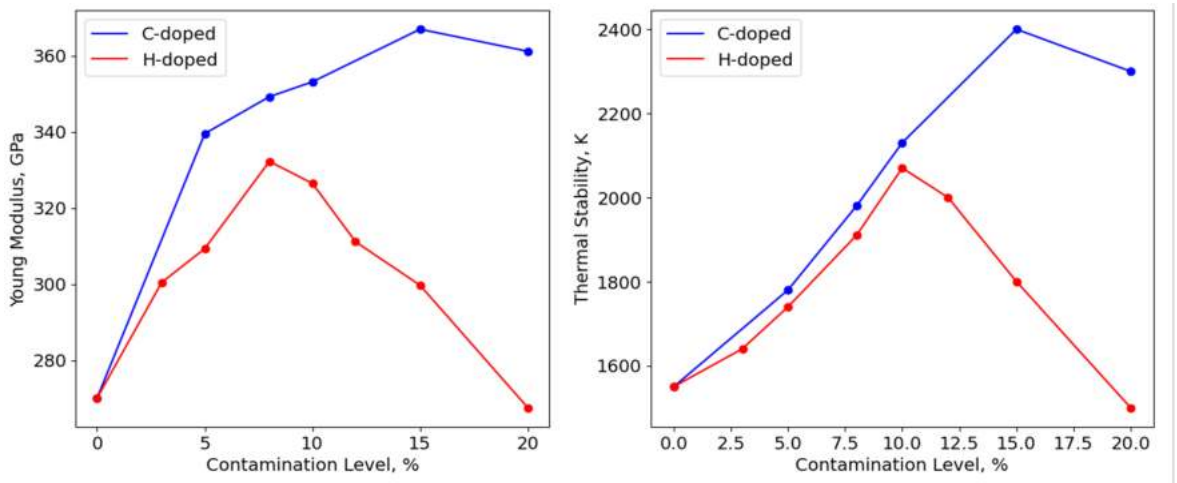

Figure 1: Young's modulus and thermal stability of C- and H-doped amorphous boron nitride.

Acknowledgment

This project has been supported by Samsung Advanced Institute of Technology and is conducted under the REDI Program, a project that has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement no. 101034328. ICN2 acknowledges the Grant PCI2021-122092-2A funded by MCIN/AEI/10.13039/501100011033 and by the "European Union NextGenerationEU/PRTR". Simulations were performed at the Center for Nanoscale Materials, a U.S. Department of Energy Office of Science User Facility, supported by the U.S. DOE, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357. Additional computational support was received from the King Abdullah University of Science and Technology-KAUST (Supercomputer Shaheen II Cray XC40) and Texas Advanced Computing Center (TACC) at The University of Texas at Austin.

A Spin-Coatable Dry Transfer Method of 2D Materials

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In recent years, researchers have tried to develop various methods to synthesize high-quality large-scale graphene and 2D materials using chemical vapor deposition (CVD) processes.[1] However, transferring and patterning steps during the fabrication of the 2D materials often degrade the electrical performances, because, for example, a conventional transfer method using poly (methyl methacrylate) (PMMA) leaves PMMA residue on the surface of the 2D materials, leading to undesirable doping effect. Moreover, acetone used to remove PMMA damages organic layers.[2] Thermal release tapes (TRT) also cause considerable contamination. Thus, it is needed to develop a new transfer technology to solve the above-mentioned problems.[3]

Here we report a novel transfer method based on the use of pressure sensitive adhesive (PSA) that can be simply spin-coated on the sample, and completely removed by peeling-off after the transfer, which is expected to allow the use of soluble layers for various 2D device fabrication processes. We confirmed that the sheet resistance of graphene has not been altered after transfer thanks to the good adhesion between graphene and PSA layers. In addition, the transparent graphene FETs transferred by PSA shows high charge carrier mobility compared to the FETs transferred by PMMA or thermal release tapes (TRT), implying that the graphene surface is ultra-clean without residues after the removal of PSA. TMDCs including WS_2 and MoS_2 were successfully transferred on a lens for smart glasses applications by PSA with minimized contamination. In addition, using PSA enables simpler patterning process eliminating the need for using photo or e-beam lithography. Thus, we believe that the current method is applicable to various flexible and wearable applications that have been limited by wet transfer conditions for more practical and larger scale device fabrications of 2D materials.

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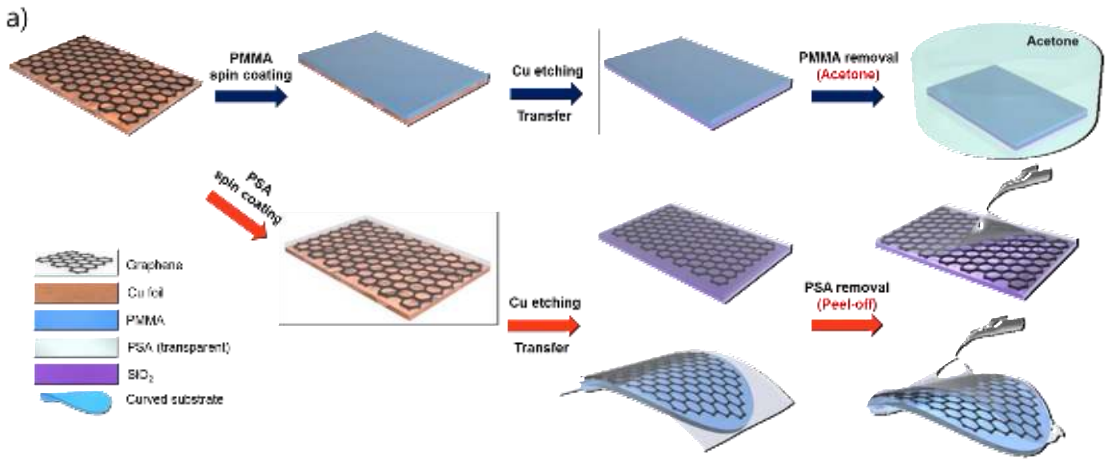

Figure 1: Comparison between PMMA spin coating method and PSA spin coating method

Structural properties of twisted transition metal dichalcogenides heterobilayers

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Abstract

Introducing an interlayer twist angle in bilayers of two-dimensional (2D) graphene resulted in new and unexpected physics [1]. Despite the absence of strong interlayer interactions, the twist angle and the stacking domain formation due to relaxation, determine the properties of these twisted bilayers. Also 2D crystals beyond graphene, such as transition metal dichalcogenides (TMDCs), show enhanced electronic correlations that are affected by twisting [2]. Furthermore, due to different lattice constants, heterobilayers result in moiré structures even without twist. Yet, the atomic structure of twisted TMDC heterobilayers is not explored thoroughly due to the large computational cost resulting from the moiré cells which typically include many thousands of atoms. Therefore, here, we systematically explore the structure of group 6 TMDC heterobilayers consisting of MoS_2, WS_2, $MoSe_2$ and WSe_2 monolayers as function of the interlayer twist angle. We find for all systems significant lattice reconstruction, involving in- and out-of-plane relaxations, which strongly depends on the twist angle. We can categorize the results in two principal cases: at large twist angle, the two constituting layers exhibit little adjustment; at small twist angle, the high-energy stacking regions shrink, while low-energy ones expand, forming multiple domains with matching lattice constants which are separated by boundaries. In particular, the two constituting layers show significant asymmetrical corrugation. We reveal that the lattice reconstruction results from the competition between strain energy cost and van der Waals energy gain. These superstructures with domains of different local stacking suggest intriguing electronic properties intriguing electronic properties of these systems.

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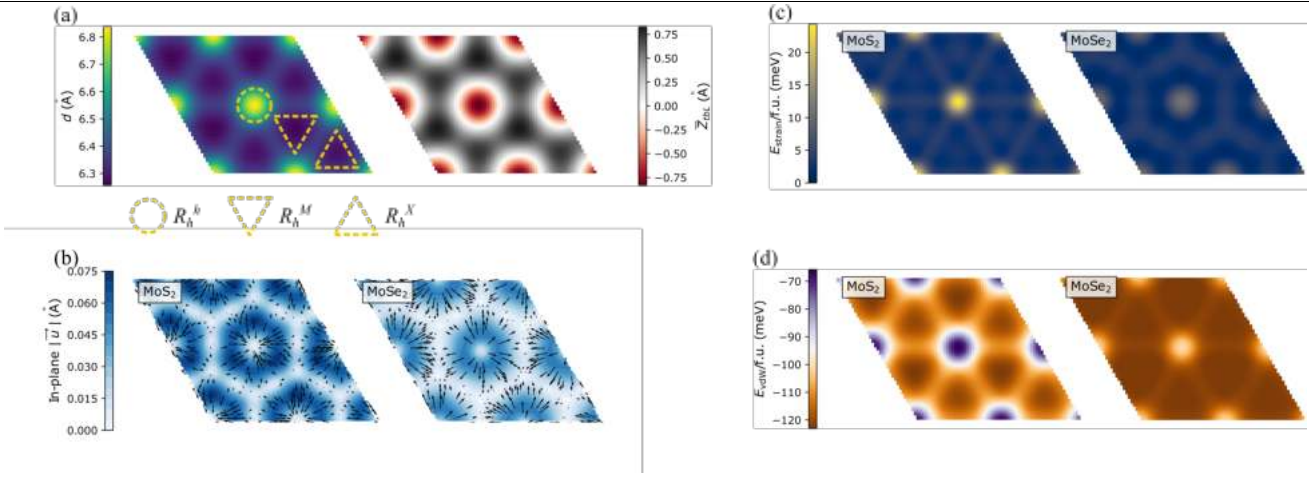

Figure 1: In $MoS_2/MoSe_2$ heterobilayers at θ = 0°, distribution of (a) Interlayer distance *d*, average out-of-plane displacement $\overline{z}_{tbL}$, and (b) in-plane displacement of metal atoms in each constituting layer. (c) Calculated strain energy and (d) vdW energy of all atoms in each constituting layer.

out-of-plane displacement $\bar{Z}_{tbL'}$ and (b) in-plane displacement of metal atoms in each constituting layer. (c) Calculated strain energy and (d) vdW energy of all atoms in each constituting layer.

Electrochemical Functionalization of 2H-Phase MoS_2 nanosheets towards Enhanced Catalytic Applications

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Abstract

We describe a novel electrochemical strategy for the surface functionalization of 2H-phase MoS_2 nanosheets, which affords the grafting of different molecular groups (e.g., acetic acid groups) derived from organoiodides [1]. Upon cathodic treatment of a pre-expanded MoS_2 crystal in an organoiodide containing electrolyte, water-dispersible nanosheets with a derivatization degree of ~0.10 molecular groups per surface sulfur atom were obtained, expanding the scope of covalent molecular functionalization of two-dimensional MoS_2, typically restricted to the metastable 1T phase [2]. Functionalization is shown to be driven by the external supply of electrons to the MoS_2 nanosheets and to be controlled by the presence of S vacancies in the 2H-MoS_2 lattice, where the molecular groups can bind. The functionalized nanosheets were tested as a catalyst for the reduction of nitroarenes and organic dyes with $NaBH_4$, which is relevant in environmental remediation and chemical synthesis [3]. These derivatized 2H-MoS_2 nanosheets showed a remarkably enhanced catalytic activity compared to that of non-functionalized 1T- and 2H-phase MoS_2 nanosheets as well as non-noble metal catalysts previously reported for this application, and retained a good catalytic activity even at realistic reactant concentrations. The 2H-phase MoS_2 catalyst could also be immobilized on a polymeric scaffold in order to facilitate its manipulation and reutilization for several catalytic cycles. Analysis of the reduction kinetics revealed a correlation between the reaction order and the net electric charge of the organic substrates.

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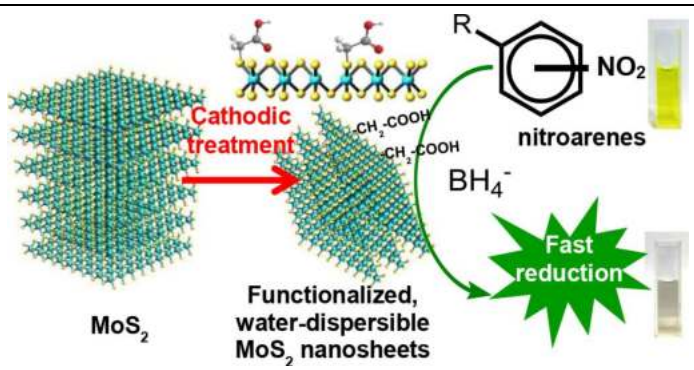

Figure 1: Scheme of the electrochemical cathodic strategy used to obtain derivatized 2H-MoS_2 dispersions and its application to the reduction reaction of several nitroarenes and organic dyes.

Synthesis of Hydrogen and Fluorine Passivated Diamanes from Graphene on Silicon Carbide

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The miniaturisation of devices has become more challenging due to material and thermal constraints. In order to overcome these limitations, synthesis of diamane (atomically thin diamond-like structures) has attracted interest to develop 2D electronics and spintronics. Recently, diamane was synthesised from bilayer graphene [1] using fluorine from XeF_2 ("F-diamane") on CuNi(111). While this represents significant progress, the CuNi(111) substrate makes it impossible to probe electronic properties of the material and build devices. Synthesis of hydrogen passivated diamane ("H-diamane") [2] has similarly been demonstrated on gold TEM grids. Building upon our previous work in the production of bi- and mono-layer graphene on semiconducting SiC substrates [3][4], we work to passivate graphene on 4H-SiC(0001) using XeF_2. Fluorination has thus far resulted in C-F ratios of between 16 and 25, an example of which can be seen in figure 1B. We also present results on H-diamane synthesis, obtained by exposing graphene on SiC to atomic hydrogen. Following successful synthesis, material-level electrical properties and device-level operational capabilities will be explored.

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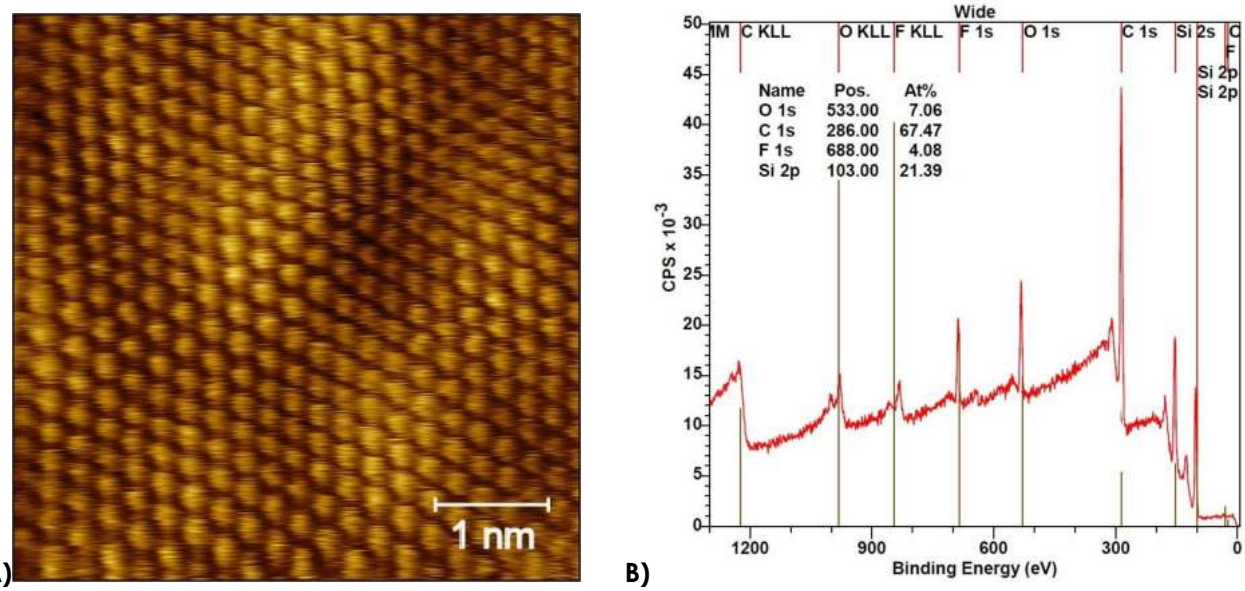

Figure 1: A: Atomic resolution preliminary unfiltered STM image of fluorinated graphene showing preserved graphene-like structure. B: Wide XPS spectrum of the fluorinated sample, showing the presence of fluorine and proportion of fluorine and carbon.

2D Xenes for Straintronics

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Abstract
The tremendous success of graphene has initiated a paradigm shift towards the expedition of various properties of graphene-like 2D materials, referred to as Xenes. The rapid miniaturization of silicon devices and beneficial electro-mechanical properties of silicene in the field of flexible electronics [1] have paved the way for the xenes in nano electro-mechanical systems (NEMS). Based on the results obtained for silicene [2], we extend this investigation to inspect the utility of straintronics for other xenes in nanoscale regime using ab-initio density functional theory and quantum transport approach based on Landauer formalism. Effect of strain on K-point is seen in the band structure for silicene, germanene and stanene. The directional piezoresistances have been calculated as per the critical strain limit, and the relevant gauge factors are compared. They sinusoidal dependence on the transport angle akin to silicene and graphene [3]. Another application like conductance modulation has been explored, which is quantized in nature. The strained tight binding parameters of phosphorene have been evinced, which is the first of its kind. Based on the above results, we propose a voluminous model for the monolayer Xenes keeping the perspective of straintronics and its applications.

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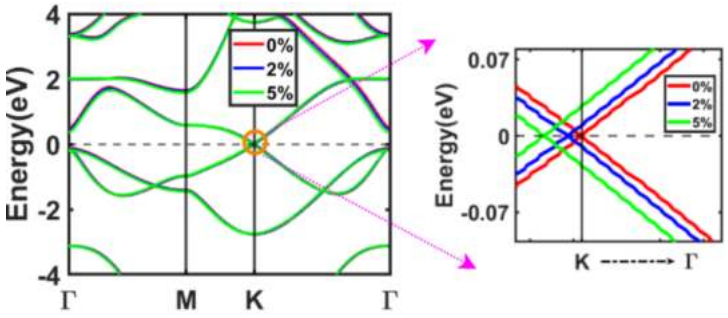

Figure 1: Band structure of stanene, showing movement of K-point with AC strain applied.

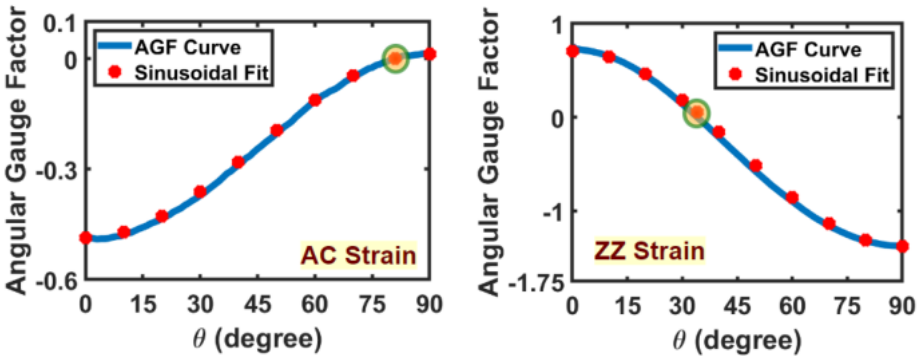

Figure 2: Variation of Gauge factor and it's sinusoidal fit for silicene against transport angle.

PHOTO-FERROELECTRIC ALL-VAN DER WALLS NEUROMORPHIC DEVICE

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2D ferroelectric materials have attracted interest since their introduction to the van der Waals (vdW) family.[1] The vdW interfacial coupling capabilities ease their implementation into complex hybrid architectures challenging with standard thin film technology.[2] Here, I will demonstrate the non-volatile electrical and optical control of the ferroelectric polarization in all-vdW ferroelectric/semiconductor heterostructures.[3] The wavelength-dependent study unveils ferroelectric polarization control and decouples the mechanisms driven by photogenerated carriers for each material. The vdW Ferroelectric Field-effect transistors show On/Off ratios exceeding 10^7, large hysteresis memory windows, and multiple remanent states, sorting them as good artificial synapse candidates. Following, long-term potentiation/depression, and spike rate-dependent plasticity are shown using electrical control. Moreover, the synaptic functionalities were complemented by the unique dual optical and electrical control, enabling optically stimulated and optically assisted synaptic devices. We benchmark our device with a simulated artificial neural network and achieve an excellent accuracy level of 91%, close to the ideal synaptic case (96%). The combination of the Photo-Ferroelectric functionalities and the shown synaptic characteristics put all-VdW ferroelectric/semiconductor heterostructures on the roadmap for novel computing architectures.

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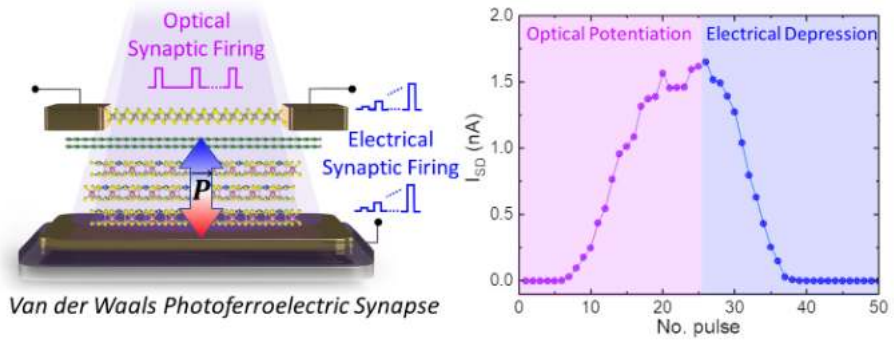

Figure 1: Left: schematic of the device.
Right: Light potentiation/Electrical depression Synaptic Plasticity.
(Adapted from [3])

Structure-Property Relationship in Amorphous Monolayer Carbon

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Abstract

Correlating atomic configurations—specifically, degree of disorder (DOD)—of an amorphous solid with properties is a long-standing riddle in materials science and condensed matter physics, owing to difficulties in determining precise atomic positions in 3D structures. To this end, 2D systems provide insight to the puzzle by allowing straightforward imaging of all atoms[1,2]. Direct imaging of amorphous monolayer carbon (AMC) grown by laser-assisted depositions has resolved atomic configurations, supporting the modern crystallite view of vitreous solids over random network theory[3]. Nevertheless, a causal link between atomic-scale structures and macroscopic properties remains elusive. Here we report facile tuning of DOD and electrical conductivity in AMC films by varying growth temperatures[4]. Specifically, the pyrolysis threshold temperature is the key to growing variable-range-hopping conductive AMC with medium-range order (MRO), whereas increasing the temperature by 25 °C results in AMC losing MRO and becoming electrically insulating, with an increase in sheet resistance of 10^9 times. Beyond visualizing highly distorted nanocrystallites embedded in a continuous random network, atomic-resolution electron microscopy shows the absence/presence of MRO and temperature-dependent densities of nanocrystallites, two order parameters proposed to fully describe DOD. Numerical calculations establish the conductivity diagram as a function of these two parameters, directly linking microstructures to electrical properties. Our work represents an important step towards understanding the structure–property relationship of amorphous materials at the fundamental level and paves the way to electronic devices using 2D amorphous materials.

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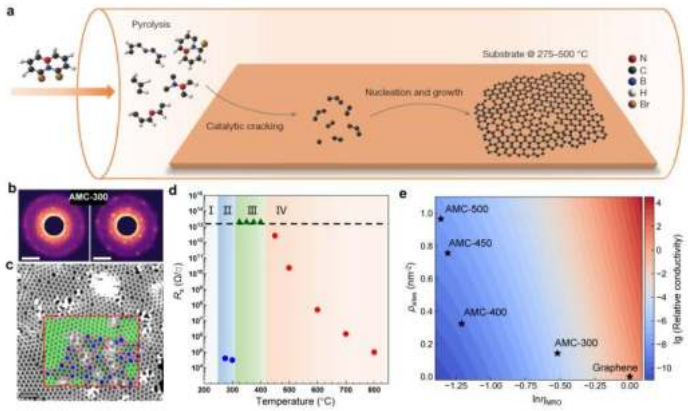

Figure 1: Structure-property relationship in AMC.

Effects of disorder in the electronic properties of monolayers and nanoribbons MoS_2

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Comprehensive research on electronic and spintronic properties of graphene and MoS_2 has been the focus of scientific attention for several years and still is [1,2]. An important issue, however, is the presence of defects that can influence these properties [3,4]. In the case of MoS_2, experiments have demonstrated that edges (1D defect) can host local magnetic moments [5]. However, the computational cost of the ab-initio DFT calculations for experimentally relevant system size is a downside. In this work, we first show that sulfur vacancies in monolayer MoS_2 induce gap states in the electronic band-structures. As a second contribution, we have investigated theoretically the magnetic properties for several nanometer long MoS_2 nanoribbons with zigzag edges using fine-tuned parameters in a tight-binding (TB)-Hubbard Hamiltonian. We could successfully reproduce the metallic state induced by the edges, compute large-scale nanoribbons and predict the spin domain-wall energy as well as study the effect of edge disorders on the magnetic properties [6]. Besides the full TB parametrization of the nanoribbon, we also described the band crossing the Fermi level with a one-dimensional linear chain model, allowing us to easily study ferromagnetic and anti-ferromagnetic configurations and giving us a useful way to study the energy cost for switching spins on various spots and scales. This model can be useful to study the stability and the properties of real size nanoribbons presenting spin defects and their applications.

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Study of $PtSe_2$ synthesis by molecular beam epitaxy for high frequency optoelectronics

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$PtSe_2$ is a 2D material with high intrinsic qualities suitable for high frequency IR optoelectronics [1], especially for photodetection at the 1.55µm telecom wavelength. We investigated the synthesis of $PtSe_2$ thin films on sapphire substrates by molecular beam epitaxy. In particular, we studied the impact of a post-growth annealing for various Se fluxes on the full width at half maximum (FWHM) of the $PtSe_2$ E_g Raman peak (Fig.1): a small E_g FWHM value means a high crystallinity [2], essential for good (opto)electronic performances. We also grown $PtSe_2$ on vicinal sapphire(0001) surfaces and demonstrated an improvement of film crystallinity and electronic mobility. We characterized the films using grazing incidence X-ray diffraction (GIXRD) and transmission electron microscopy (TEM). To fabricate optoelectronic devices, we synthesized a 7.5nm-thick $PtSe_2$ film on a 2 inches sapphire substrate and coplanar waveguides integrating a 4x4 µm $PtSe_2$ channel were then realized. The channel was illuminated with a 1.55µm laser beam modulated in intensity at frequencies varying between 2 and 67 GHz. Our $PtSe_2$ photodetector exhibits a record 3dB bandwidth of 60GHz (Fig. 2). Moreover, we demonstrated a 30GHz bandwidth optoelectronic mixer, comparable to those obtained with graphene [3]. These results confirm that $PtSe_2$ is a highly promising material for high frequency optoelectronics.

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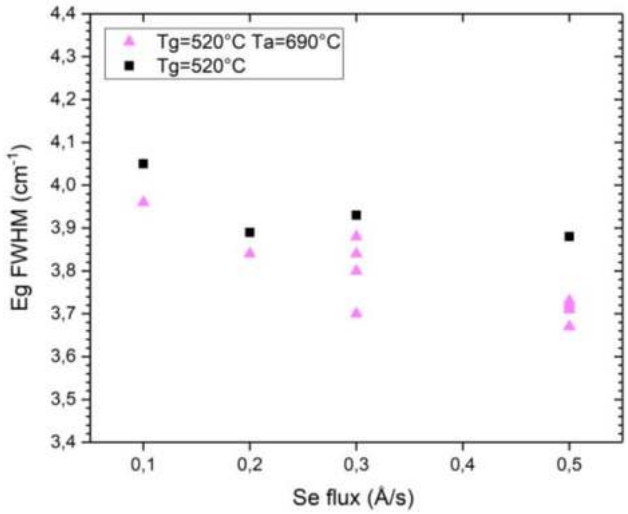

Figure 1: FWHM of Eg RAMAN peak of $PtSe_2$ films grown at Tg=520°C with a post-growth annealing at Ta=690°C (purple triangles) or without anneal step (black squares), under different Se fluxes.

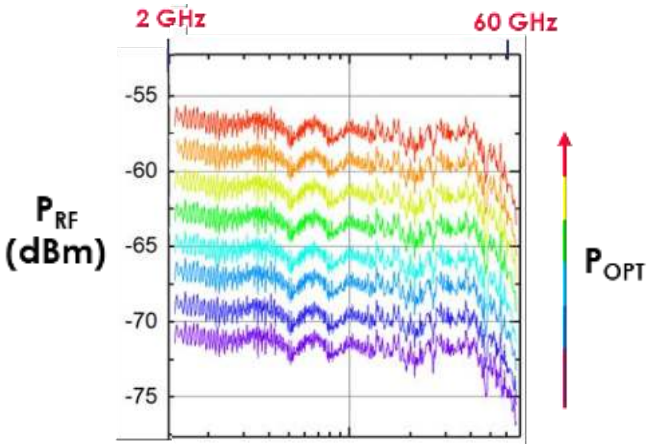

Figure 2: High frequency 1.55µm photodetection with a 7.5nm-thick $PtSe_2$ channel inserted in a coplanar waveguide. A 60GHz bandwidth photodetector is demonstrated.

Ultrafast phonon-driven charge transfer in van der Waals heterostructures

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Transition metal dichalcogenides (TMDs) are characterized by enhanced light-matter and Coulomb interactions leading to a rich energy landscape of tightly bound excitons. Stacking TMD monolayers into van der Waals heterostructures enriches the scenario even more, introducing spatially separated interlayer states and adding another exciton species with long lifetimes and an out-of-plane dipole moment.
Recent experiments demonstrated an ultrafast charge transfer in TMD heterostructures. However, the nature of the charge transfer process has remained elusive. Based on a microscopic and material-realistic exciton theory, we reveal that phonon-mediated scattering via strongly hybridized intervalley excitons governs the charge transfer process that occurs on a sub-100fs timescale. We track the time-, momentum-, and energy-resolved relaxation dynamics of optically excited excitons and determine the temperature- and stacking-dependent charge transfer time for different TMD bilayers [1]. In a very recent joint experiment-theory study, the predicted two-step charge transfer process has been compared with time-resolved ARPES measurements (performed in the group of Stefan Matthias, Georg-August-University Göttingen) [2] showing an excellent agreement.
The provided insights present a significant step for a microscopic understanding of the technologically important charge transfer process in van der Waals heterostructures.

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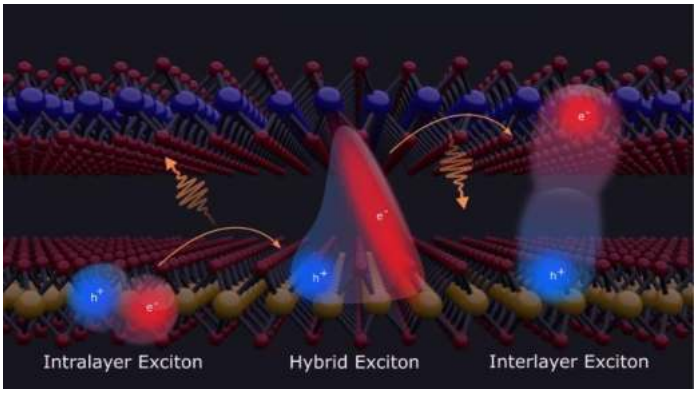

Figure 1: Sketch of the charge transfer process. Starting from an exciton localized in the bottom layer, phonon-mediated scattering to an hybrid exciton state (where e.g. the electron lives in both layers) allows for the transfer of the charge (here electron) to the upper layer resulting in a spatially separated interlayer exciton state.

First-principles modelling of graphene-atomic cluster interactions

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Abstract

The 2D nature of graphene renders its electronic structure extremely sensitive to proximity-induced interactions with e.g. adatoms, atomic clusters, or other 2D materials, to induce desirable properties. In this work, we employed size-selected few-atom transition metal clusters (Au, Cu) to provide a unique atom-by-atom control over the induced physico-chemical properties in graphene. Both Au and Cu clusters have similar geometries in gas phase calculations, but their interaction with graphene is different. While the former adsorb with binding energies of about -1 eV, indicating (weak) chemisorption, the latter are physisorbed with binding energies of a few hundred meV. Cluster size-dependent effects are evidenced by band structure calculations. Clusters with an odd number of atoms dope graphene, due to charge transfer interaction. Clusters with even-numbered atoms open an energy gap of ~20 meV (Cu_n) and ~40 meV (Au_n) at the Fermi level [1].
The adsorbed clusters induce spin-orbit (SO) coupling effects in graphene, which can be evidenced by the spin splitting of the band structure. Overall, cluster adsorption enhances the SO strength of graphene by three orders of magnitude, but the relative spin-splitting values depend on the atomic composition of the cluster. The average spin splitting for Cu_n clusters is in the range of 1-5 meV, whereas Au_n cluster adsorption causes a relatively larger band splitting of 2-9.5 meV [2]. Apart from SO-induced band splitting, these coinage metal clusters are also capable of imprinting cluster-size specific spin textures to graphene's Dirac cone. We observe a hedgehog-type spin texture for clusters with an even number of atoms. For odd-sized clusters, a canted Zeeman-type spin texture is observed, with the spins oriented along a particular direction, due to the local magnetic moment of the cluster.
Graphene/clusters-based devices can also be used to probe the catalytic and chemical reaction kinetics of the clusters. We performed simulations of the desorption kinetics of O_2 adsorbed on the Au_3/graphene system and compared them with experimental results [3]. The computed energy barrier is in agreement with the experiments. The catalytic properties of the Cu_4/graphene system for carbon reduction reaction are also explored and compared with Cu_3X/graphene (X= Ni, Mn, Fe, Co) systems. The results suggest that the limiting potential of the reaction can be tuned by varying the binding energy of the clusters with graphene, which in turn depends on the cluster size and composition.

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Fully Printed 2D Material-based Heterostructures: from Memristic to Hysteresis Effects

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Realization of high density and reliable memristors is very important for the next generation information storage devices[1]. Solution processed 2-Dimensional Materials (2DMs) are very attractive for use in resistive-switching memories as they are compatible with low cost device fabrication processes and easily enable their integration into recyclable and flexible substrates such as paper.
In this work we investigated hysteresis and memristic effects of a fully inkjet printed 3-layers heterostructures made of water-based 2DM inks[2] and silver (used as bottom electrode only). Clear and reproducible resistive-switching effects were observed in the Ag/MoS_2/Gr heterostructure, providing low SET/RESET voltage, short switching times (within 0.1 s) and high resistance switching ratios (10^3-10^5).[3] By measuring the electrical output at different temperatures and by including CVD graphene as interface layer in the heterostructure, we found that the memristor resistance switching is attributed to the migration of Ag ions leading to conductive filaments as well as defects at the interface. Indeed, the use of CVD graphene not only enables to quench any memristic effect, but also eliminates any hysteresis, strongly improving device reproducibility.[3]

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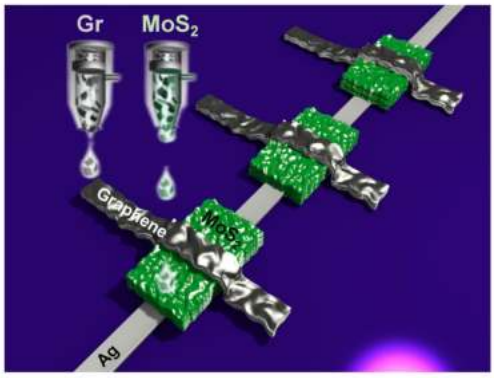

Schematic of fully printed Ag/MoS_2/Gr memristors.

Laser-assisted synthesis of porous graphene-like structures for electrochemical energy storage

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3D graphene-based porous networks have proven to be excellent candidates for electrochemical energy storage as they combine a considerable specific surface area with good electronic conductivity and light weight. Currently, most synthesis protocols rely on wet-chemistry or high temperature processes, often requiring the use of harmful reagents or inert atmosphere. An alternative synthesis method involves the use of laser sources. Laser-induced graphitization relies on the high energy of the laser pulses which can decompose a carbon-based precursor and form a porous 3D graphene-like network. Laser-based approaches have been recently employed by our group for the "dry" preparation of turbostratic graphene-based structures by decomposing a diverse group of precursors including biomass [1], phenol-based resins [2] and polymers [3]. The simultaneous irradiation of two different precursors can provide graphene-/nanoparticles hybrids with enhanced functionalities [4]. The use of industrial-type laser sources operating at ambient conditions testify towards a "green" process, able to provide electrode materials. The process is compatible with additive manufacturing, providing the flexibility to directly fabricate patterned electrodes onto a desired substrate. Such a material synthesis approach enables the fabrication of flexible or even stretchable electronic components, which are compatible with smart textiles. Here, we will present recent results on the production of porous graphene-like structures and nanohybrids arising from the decomposition of various precursors. Physicochemical characterization revealed that the materials exhibit high sp^2/sp^3 and C/O ratios. Also, the graphene-like structures are highly crystalline and demonstrate increased interlayer spacing, i.e. turbostratic arrangement. The graphene-like materials were evaluated as supercapacitor electrodes.

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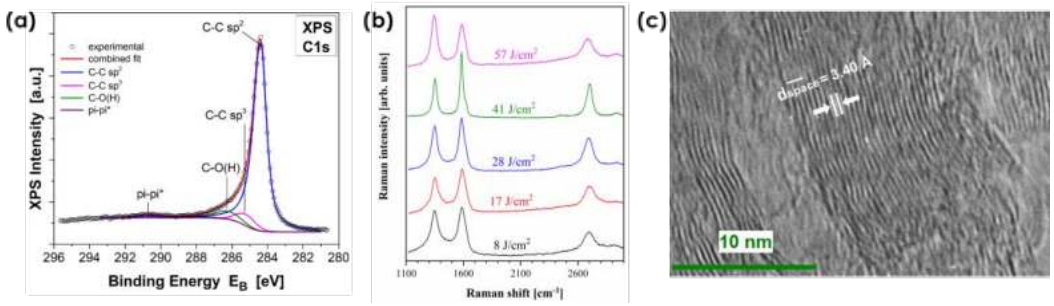

Figure 1: (a) C1s XP spectrum of laser-induced graphene, (b) Raman spectra dependence on laser fluence, and (c) TEM image of laser-induced graphene.

High-field 1/f noise in hBN-encapsulated graphene transistors

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In this talk, I will present our study of the 1/f noise in high-mobility hBN-encapsulated graphene transistors under high bias [1]. Flicker (1/f) noise, albeit ubiquitous in condensed matter devices, still lacks an understanding of its intrinsic origin. Being important for modern quantum technologies [2], this understanding becomes a crucial issue, and graphene appears as a model material in this respect thanks to its high quality and versatility. Previous studies of flicker noise in graphene [3] have concentrated on the low-bias regime, analysed with the conventional Hooge formula [4].
We examine 1/f noise in a large series of high-quality devices over an extended bias range including the non-linear intraband velocity saturation regime as well as the graphene-specific Zener interband regime. Based on extensive transport, low-frequency noise and microwave noise-thermometry analysis, we report on a velocity flicker scaling, and we introduce a phenomenological model in the spirit of the quantum-coherent bremsstrahlung interpretation of flicker noise [5]. Our model introduces a modified Hooge formula that accounts for non-linear regimes, and it supports a light-matter coupling interpretation of electronic flicker noise.

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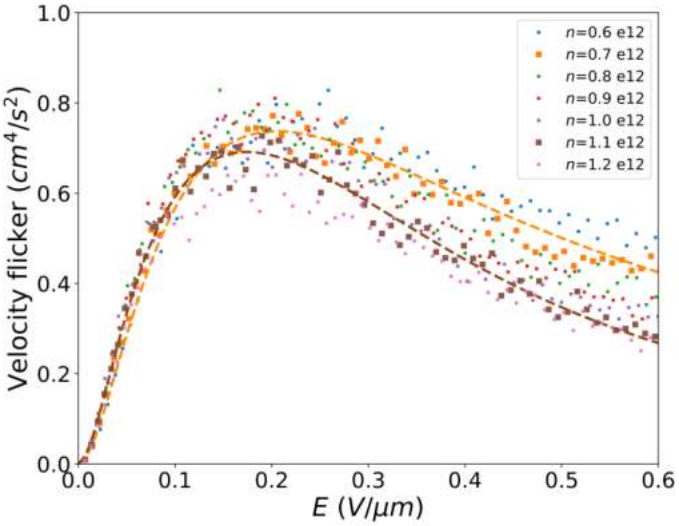

Figure: Velocity flicker noise measured in a high-mobility graphene transistor, as function of bias electric field for different dopings, demonstrating a quasi-scaling. The noise level presents a bell-shape with respect to bias, that is accounted for by a modified Hooge formula (dashed lines) in the non-linear velocity saturation regime.

Optical Absorption Spectroscopy of Exfoliated Few Layer $PtSe_2$

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Two-dimensional Transition Metal Dichalcogenides are promising candidates for optoelectronics thanks to their large electronic mobility and strong light-matter coupling. Among them, PtSe2 thin films are of particular interest because of their outstanding bandgap tunability starting from 1.2 eV for one monolayer and vanishing as the number of layers increases [1]. This property makes them especially suitable for wide-range near-infrared to visible ultrafast photodetectors and telecom optoelectronics. PtSe2 thin films are manufactured using several growth methods, where material purity is sought to achieve ultimate device performances [2-4]. However, intrinsic $PtSe_2$ optical absorption mechanisms remain elusive, as an apparent optical bandgap (of about 0.5 eV for thick samples) can be observed regardless of the semiconductor or semimetallic nature of the film. In this talk, I will present an extensive 0.8 – 3.0 eV optical absorption study performed on superior quality gold-assisted mechanically exfoliated $PtSe_2$ flakes [5]. A comparison to DFT calculations allows us to shed light on the intrinsic in-gap absorption mechanism responsible for a strong absorption tail (figure 1) and the creation of an effective optical bandgap.

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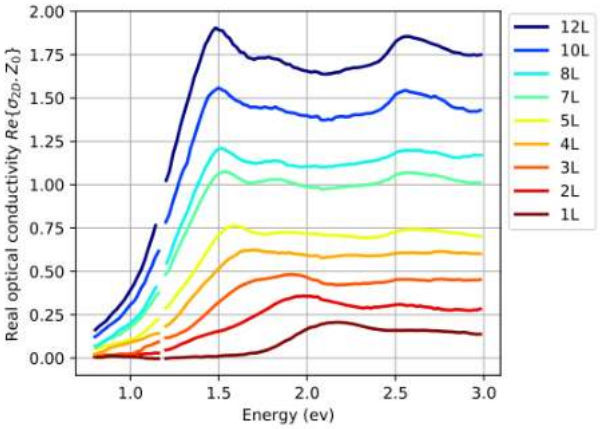

Figure 1: Optical spectroscopy of monolayer to quasi-bulk 12-layers exfoliated PtSe2. The high-energy plateau of the 2D real sheet conductivity $Re\{\sigma_{2D}.Z_0\}$ (with Z_0 the impedance of vacuum) scales with the number of layers. While 2D confinement explains the redshift of the peaks position with the thickness, a non-trivial and intrinsic mechanism is responsible for the slowly decaying low-energy absorption tail.

Cataloguing $MoSi_2N_4$ and WSi_2N_4 van der Waals Heterostructures: An Exceptional Material Platform for Excitonic Solar Cell Applications

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Two-dimensional (2D) materials van der Waals heterostructures (vdWHs) provides a revolutionary route towards high-performance solar energy conversion devices beyond the conventional silicon-based *pn* junction solar cells.[1,2] Despite tremendous research progress accomplished in recent years, the searches of vdWHs with exceptional excitonic solar cell conversion efficiency and optical properties remain an open theoretical and experimental quest.[3,4] Here we show that the vdWH family composed of $MoSi_2N_4$ and WSi_2N_4 monolayers provides a compelling material platform for developing high-performance ultrathin excitonic solar cells and photonics devices. Using first-principle calculations, we construct and classify 51 types of $MoSi_2N_4$ and WSi_2N_4-based [$(Mo,W)Si_2N_4$] vdWHs composed of various metallic, semimetallic, semiconducting, insulating and topological 2D materials. Intriguingly, $MoSi_2N_4$/(InSe, WSe_2) are identified as Type-II vdWHs with exceptional excitonic solar cell power conversion efficiency reaching well over 20%, which are competitive to state-of-art silicon solar cells. The $(Mo,W)Si_2N_4$ vdWH family exhibits strong optical absorption in both the visible and UV regimes. Exceedingly large peak UV absorptions over 40%, approaching the maximum absorption limit of a free-standing 2D material, can be achieved in $(Mo,W)Si_2N_4/a_2$-$(Mo,W)Ge_2P_4$ vdWHs. Our findings unravel the enormous potential of $(Mo,W)Si_2N_4$ vdWHs in designing ultimately compact excitonic solar cell device technology.

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Figures

Figure 1: $(Mo,W)Si_2N_4$-based vdWHs as promising candidates for high efficiency excitonic solar cells

Photolithography fabrication of CVD 2H-$MoTe_2$ field effect transistors

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Transition metal dichalcogenides (TMD) materials are generating significant interest for "post silicon" electronic applications. Among the various TMDs investigated to date, two-dimensional (2D) transition metal ditellurides are promising materials owing to their narrow bandgap energy (1.1–1.2 eV). They are also compatible with bulk silicon and have inherent superior electrical transport [1].

Interestingly, when attempting to fabricate FET devices from 2D-$MoTe_2$, the fragility of thin films poses a significant fabrication challenge [2]. In numerous studies, the general approach for producing 2D-FETs uses top-down thin-film exfoliation techniques and situates them on patterned substrates. Although it achieves auspicious device performance, the random positioning of the $MoTe_2$ film with uncontrollable thickness renders it unsuitable for manufacturing [3]. More suitable and manageable manufacturing methods are required.

In our research, FET devices are fabricated and characterised directly on CVD-grown $MoTe_2$ films [2] using photolithography. This permits the precise design of FET device geometries with controlled thickness and other characterisation structures such as TLMs, Figure 1 depicts TLMs and FET structures fabricated from CVD $MoTe_2$. The characterisation process involves using scanning electron microscopy (SEM), atomic force microscopy (AFM), and Raman spectroscopy to detect any alterations in the properties of the film. Concurrently, electrical measurements were conducted on the produced TLM and FET structures. Before and after each step of the fabrication process, no change in the physical and chemical properties of $MoTe_2$ thin films was observed.

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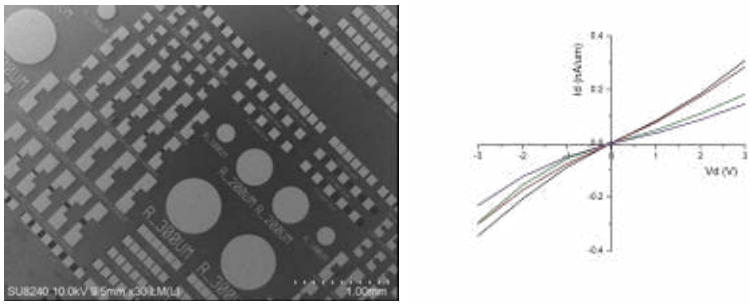

Figure 1: 5nm CVD $MoTe_2$ -FET SEM image and electrical measurement result

Graphene reinforced thermoplastic polymer composites for the transport sector

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The transport sector is facing different challenges: on the one hand, the climate crise imposes the reduction of the carbon footprint of transportation and on the other hand the users ask for greater safety and comfort[1]. The implementation of graphene-based polymers composites (GPCs) into structural elements of aerial and terrestrial transportation is devoted to tackle both challenges. Thanks to their high strength and stiffness,[2,3] the use of GPCs allows for a physical reduction of the structural elements, and consequently a decrease of the weight. GPCs show also high resistance to wear,[2] ultimately extending the lifespan of the components based on pristine polymers. Furthermore, compared to bare polymers, GPCs have demonstrated improved dumping capacities, lower flammability, electrical and thermal conductivity, which may result in a safer and more comfortable travel experience for the users.[4]
In this view, we designed and produced graphene-loaded thermoplastic polymer filaments and pellets for their use in additive manufacturing and injection moulding. Polyamide12 (PA12), polypropylene (PP) and polyether ether ketone (PEEK) composites containing up to 25% wt. of few-layer graphene (FLG)[5], as well as additional fillers such as carbon black (CB), carbon fibres and few-layer h-BN flakes, were produced by melt-compounding and extrusion. The optimized GPCs have shown an improvement in elastic modulus and strength up to ~130% and ~50%, respectively (PA12 containing 14% of FLG+CB) compared to the pristine polymers. Furthermore, the designed GPCs exhibited electrostatic discharge properties (electrical conductivity in the 0.1-1 S/m range) and improved thermal conductivity (up to 2-fold increase compared to pristine counterpart). Therefore, the exploitation of GPCs filaments will produce lighter structural components with added multifunctionalities.

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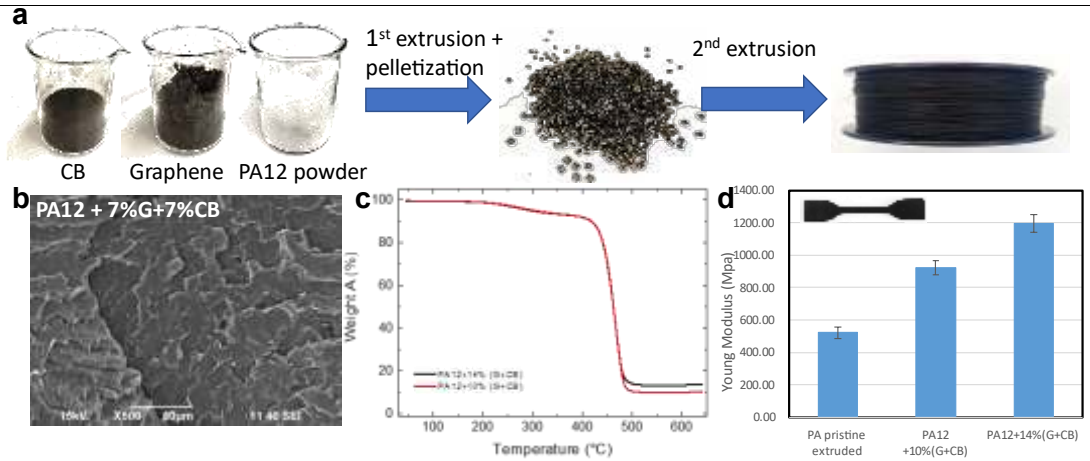

Figure 1: (a) Scheme of the GPCs filament production. (b) Scanning electron microscopy image of PA12+14% (FLG+CB). (c) thermogravimetric analysis of PA12+10% and 14% (FLG+CB) (d) flexural modulus of pristine PA12 and of the composite PA12+10% and 14% (FLG+CB).

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Full wafer-scale characterization method for 2D materials: on the way to 300 mm integration

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Uniform large-scale growth is one of the most important challenges for 2D integration in microelectronics devices. Recently, teams proved their capability to process 200 mm or 300 mm wafers of MoS_2 or WS_2 [1,2]. However, in most cases, only local characterizations are performed and data given are not representative of the whole wafer quality. Full size-wafer characterization is in that sense a key point to monitor the quality, homogeneity and defectivity for the growth and integration processes for large-scale applications.
This study is based on large scale characterization of MoS_2 grown on 200 mm Si/SiO_2 wafer in clean room [3]. Using wafer-scale tools, the whole wafer is characterized by Raman spectroscopy, photoluminescence, optical reflectance, defectivity and haze (surface scanning). These large-scale results are corroborated with small-area surface e.g. AFM, SEM, XRD…, to better understand them and to determine the most relevant parameters. With the help of software developed for wafer-scale tools and using data analysis, mapping can be performed for each of these five large-scale methods previously cited. The Figure 1 gives an example of one wafer characterized with four different large-scale methods. It exhibits that the quality is lower on the left edge with more defects in this area. The peak position in photoluminescence is linked to more defects as seen on defects analysis. This has been confirmed by SEM and AFM images. Thus, a non-destructive high-speed method, allowing mapping of 2D material at wafer scale in clean room, has been developed for 200 mm MoS_2 wafers and can be easily scaled up to 300 mm wafers.

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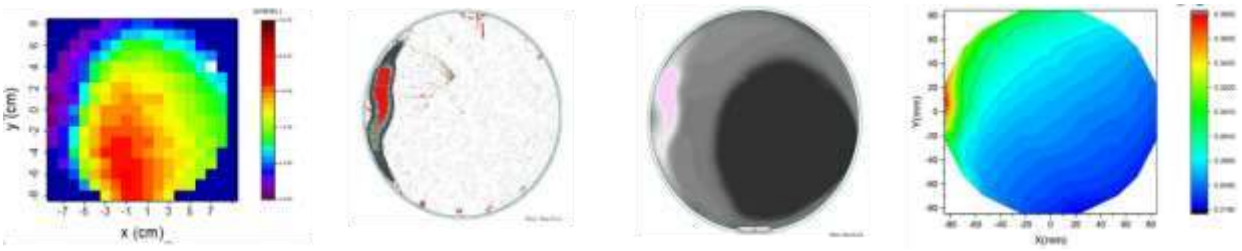

Figure 1: From left to right: mapping of MoS_2 200 mm wafers by photoluminescence (peak position), defectivity, haze and optical reflectance at 249 nm

Scaling the production of high-quality graphene devices for sensing applications

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Abstract

Thanks to its unique electronic properties, graphene has great potential to produce ground-breaking devices for a wide variety of industries. However, a key challenge remains: to translate the required low defect level high mobility graphene from lab device demonstrators to production scale. Paragraf has realised this by being the first company in the world to produce graphene using our proprietary and commercially scalable growth method directly onto target substrates, using standard semiconductor manufacturing tools. [1] Paragraf covers all aspects of production, from the growth of graphene to processing into final devices, which has enabled the demonstration of industry-ready magnetic- and bio-sensing products for a range of applications.

We have demonstrated graphene magnetic hall-effect sensors with a sensitivity significantly beyond silicon [Fig1], enabling applications in automotive industries such as positional, rotational, and current sensing. They offer high resolution, instantaneous response, wide measurement range, low power consumption, and ease of use. Further details can be found in Dr Lok Yi Lee's poster presentation "Improved graphene-based Hall effect sensors through control of defect levels in graphene".

Graphene biosensors enable sensitive, real-time measurement of biomolecules directly from samples without enrichment or amplification, reducing both time to result and cost without sacrificing accuracy. The recent acquisition of Cardea Bio-Inc, now Paragraf USA, enables the use of mass-produced, transfer-free monolayer graphene to manufacture the state-of-the art graphene-based biosensor products developed by the US team over the last ten years [2].

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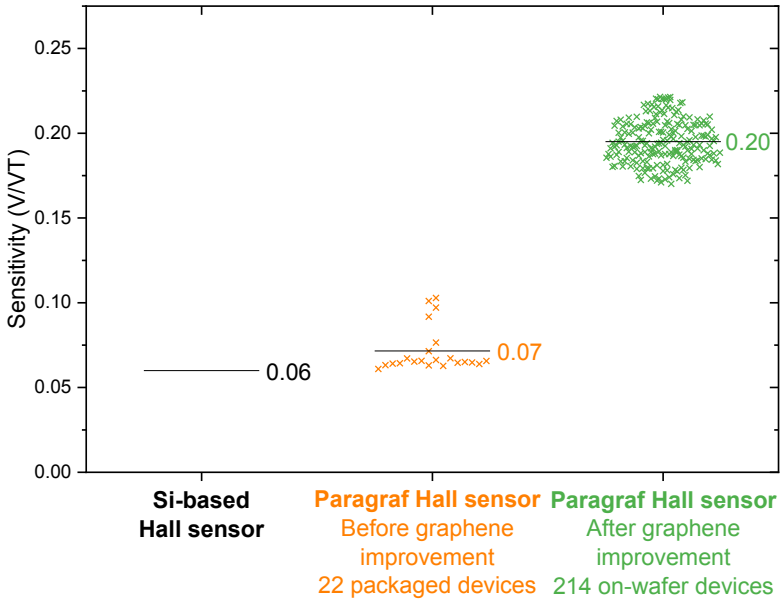

Figure 1: Recent advancements in Paragraf Hall Sensor performance

A universal approach for formulation of additive-free 2D material-based inks for room-temperature printing of electronics

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Abstract

We here introduce a new approach for the formulation of 2D materials into printable or coatable inks for the fabrication of functional devices [1]. In a traditional ink formulation, additives are introduced in large concentrations to address processing challenges, but they drastically degrade the electronic properties of the materials. For the removal of the additives, a high-temperature post-deposition treatment can be used, but this complicates the fabrication process and limits the choice of materials (i.e., no heatsensitive materials). The unique properties of 2D materials offer the possibility to formulate additive-free inks in which the roles of the additives are taken over by van der Waals (vdW) interactions. The approach is universal and is demonstrated with a number of 2D materials. In this new class of inks, solvents are dispersed within the interconnected network of 2D materials, increasing the possible choice of solvents over traditional inks where dispersibility-related issues limit the selection. Furthermore, flow behavior of the inks and mechanical properties of the resultant films are mainly controlled by the inter-flake vdW attractions and can be largely controlled via concentration and choice of solvent. The structure of the vdW inks, their rheological properties, and film-formation behavior are discussed in detail. A method for large-scale production of inks for all major high-throughput printing and coating is introduced.

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Figure 1: Aggregated graphene nanosheets are processed into a homogeneous gel that can be used for formulation of additive-free functional inks.

Crack- and contamination-free transfer of graphene films

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Abstract
Owing to its fascinating properties, graphene has exhibited great potentials in the applications of high-performance electronics, flexible devices and encapsulation. The availability of graphene on a wide range of substrates forms the basis for large-area applications, such as graphene integration with silicon-based technologies, which requires graphene on silicon with outperforming carrier mobilities. In this regard, although currently, large-area graphene films have been successfully produced based on chemical vapor deposition methods, with outperforming quality and uniformity. Graphene films were only produced on limited archetypal substrates, such as metal foils. Reliable after-growth transfer techniques, that do not produce cracks and contamination are critical for layering 2D materials onto arbitrary substrates for further applications. During the transfer of graphene, with atomically thin and highly flexible nature, polymer-based transfer medium is usually introduced for avoiding the formation of cracks and wrinkles, which, however, introduces unavoidable contamination on graphene surface. In addition, traditional transfer routes, including etching and bubble-based delamination, involve the aqueous solution-based reactions or processing, which would induce the water-related p-doping in graphene and improve the complexity in the design of industrial equipments for batch transfer.

Here, in our group, based on the structural design of transfer medium and careful modulation of interfacial forces, we have achieved the crack, contamination and wrinkle-free transfer of graphene films over large area: (1) we achieved the mechanical delamination of graphene both from the Cu surfaces and from the polymer surface, based on the uniform oxidation of Cu substrates and the controllable tuning of the interaction between graphene and polymer, and all the transfer process were conducted without using water or organic solvent; (2) we showed that, by incorporating oxhydryl groups-containing volatile molecules, the supporting films can be deformed under heat to achieve a controllable conformal contact, enabling the large-area transfer of graphene wafers without cracks, contamination, and wrinkles onto silicon wafers, and the ultraclean surfaces provide the carrier mobilities of transferred graphene up to 14,000 $cm^2\ V^{-1}\ s^{-1}$ at room temperature; (3) we achieved the preparation of high-performance water barrier films based on layer-by-layer transfer of A3-sized graphene films, and the water vapor transmission rate of double-layer graphene can be as low as 5×10^{-3} $g/(m^2 \cdot d)$, which is one order of magnitude lower than previously reported values; (4) we designed and built the transfer systems for the batch transfer of graphene films, including the automatic spin-coater, delaminator and laminator

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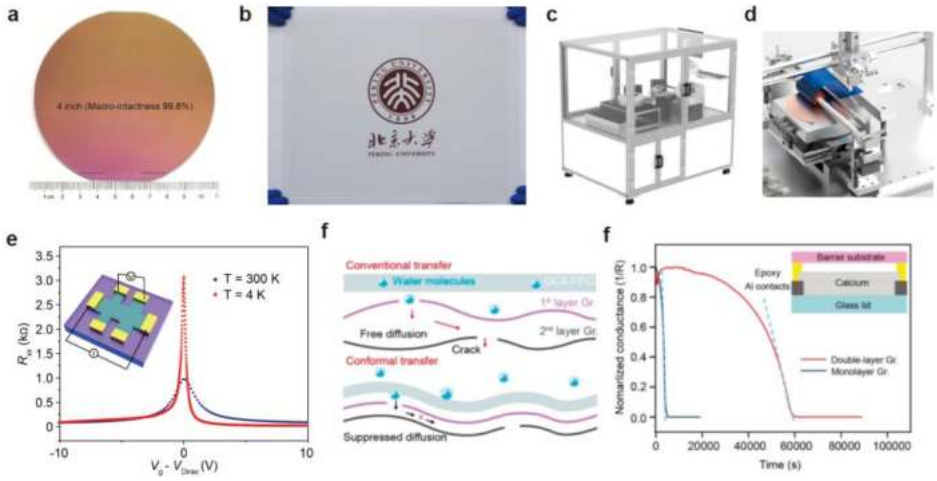

Figure 1: The crack, contamination and wrinkle-free transfer of large-area graphene films and the related applications.

Growing transition metal dichalcogenides into industrial reactors, achievements and remaining challenges after processing more than thousands 300mm wafers

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Abstract
Intensive studies are ongoing to grow Transition metal dichalcogenides (TMDC) such as WS2, MoS2, WSe2 as 2D channel material for building competitive and reliable ultra-scaled logic devices. Several years ago, imec has adapted 200 and 300 mm cross flow epitaxial reactors to growth TMDC [1,2], either on template like sapphire or directly on amorphous gate or sacrificial dielectrics, using metal organic chemical vapor deposition (MOCVD), a technique compatible with industrial reactors.
In this presentation, we report on some of the learnings obtained after processing more than 1000 runs on each reactor. After describing the basic MOCVD processes used to control the tool and process along this development period, we share on safety and update on studies carried out to improve the tool and process stability and variability [3]. We illustrate the maturity of the process and material with a selection of results obtained on lab devices and 300 mm fab lots [4,5,6], and identify some remaining tool/process challenges to grow high quality TMDC in industrial tools.

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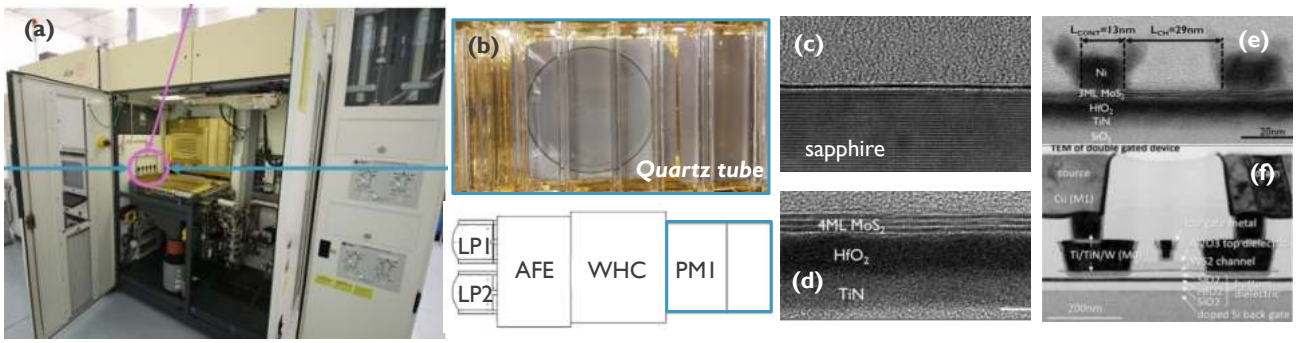

Figure 1: (a) tool mainframe, (b) cross-flow reactor and layout, growth on (c) either sapphire aiming a subsequent transfer to the final substrate or (d) directly on amorphous dielectric, either the gate dielectric or a sacrificial material to pursue the integration on same substrate. Cross section of lab (e) and fab (f) devices.

Mass Synthesis and Application of Graphene Flower and Graphene-Related Products

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We, in Incubation Alliance, Inc., have been working on the development of a method for mass synthesis of graphene and other graphene-related products as raw materials for industrial uses. We have developed a method using HIP (Hot Isostatic Pressing) for the manufacture of Graphene Flower® with high purity and quality[1]; a method for producing Graphene-coated carbon fiber cloth[2]; an in-house molding method for manufacturing a highly-oriented graphite block, Graphene Flower® BL[3]; a method for producing porous graphene[4] and neutron strengthened graphene material.[5]

In our patented method for producing Graphene Flower® using HIP, it is possible to produce a bulk material in which high-purity few-layer graphene is three-dimensionally densely grown without using any substrate and catalyst. It is also possible to control the number of graphene layers and the dimension or size of our graphene. Using the same technology, graphene-coated carbon fibers having several layers of graphene densely grown on all surfaces of woven and non-woven carbon fibers are produced. This can be used as materials of electrodes for field emission, biofuel cells, supercapacitors and fuel cells, for both research and development purposes.

Graphene Flower® BL, which is a highly oriented graphite block of 100% carbon that utilizes the Van der Waals force of graphene as a binder. It has high thermal conductivity, electrical conductivity and sliding properties. Using our in-house developed molding technology, it is possible to manufacture blocks with a thickness of 100 mm or more. And by multi-wire slicing, sliced graphene parts with arbitrary thickness of 0.2 mm or more can be practically used as heat dissipation and heat transfer materials for IT equipment, medical equipment, next-generation energy furnaces and others.

Recently, by optimizing the dimension and shape of graphene flowers and graphene seeds, we have demonstrated the coherent scattering of neutrons by graphene for the first time in the world. [5] In this presentation, we will introduce these graphene and graphene-related products and their practical applications.

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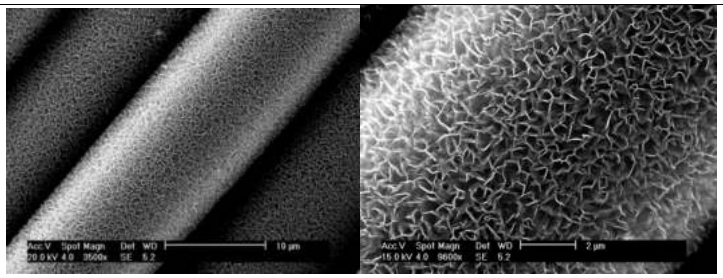

Figure 1: Graphene coated carbon fiber(SEM)

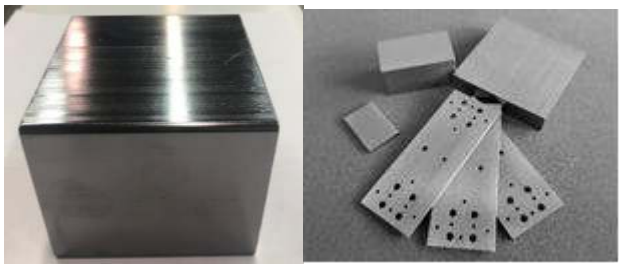

Figure 2: Graphene heat dissipation products

Liquid graphene dispersions: formulation of a multipurpose additive

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Graphene, a monolayer of graphite, is a hexagonal lattice of carbon. It is a universal material that lends itself to a broad array of development, from energy storage and microelectronics to sensing or biomedicine. As graphene is presenting outstanding mechanical, electrical and thermal properties [1], a lot of scientific research is channelled on its production. Some techniques such as the adhesive-tape method or the chemical vapour deposition produce graphene of good quality but are not suitable for scaling-up to an industrial level. For this purpose, numerous research is focusing on the development of liquid formulations of graphene.

Thanks to an innovative process, Carbon Waters has achieved a major breakthrough in the field of advanced materials with the development of an industrializable process. We develop and provide ready-to-use graphene-based additives compatible with a wide range of materials and most common industrial processes, for a vast array of applications. We are fully involved in developing a more sustainable and safer industrial environment, with projects on nontoxic protective coatings, mechanical reinforcement, thermal resistance, and battery management. As a solution provider and a partner, we are continuously developing and testing new formulations based on customer's request. We support our clients throughout their entire project by understanding their expectations and proposing concrete solutions with fast implementation that are compatible with their process and cost objectives.

The CWEP (Carbon Waters Exfoliation Process) allows us to obtain liquid graphene dispersions of a very high quality, that are stable and "ready to use". The process – which generates a low carbon footprint and is recycling friendly - is based on the mechano-chemical exfoliation of graphite [2]. The first step is the preparation of a graphite intercalation compound (GIC), obtained through the reaction of alkali metals with graphite. In the second phase, the GIC is mixed with an organic solvent and exfoliated in solution to convert the graphite from several thousand layers to a few-layer graphene. A solution called "graphenide" is then obtained. The third step is the oxidation of the graphenide solution to produce organic graphene dispersions, which can be used directly for solvent-based applications (polymers-paints) or transferred into water to produce a very stable aqueous graphene dispersion, after solvent evaporation. Thanks to the continuous development of our process, the maximum concentration of graphene dispersions that we could reach so far is 1 $g.L^{-1}$.

The advantage of our process is the production of thin and high-quality graphene, leading to outstanding performance. Besides, the dispersions obtained do not contain any surfactant, commonly used to stabilize graphene. In order to strengthen its unique manufacturing process, Carbon Waters is currently filing two patents that focus on the adaptation of the process at the industrial scale.

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Nano-sized graphene material development for neutron intensity enhancement below cold neutrons

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Abstract
Slow neutrons, such as cold neutrons, are important non-destructive probes not only for basic physics but also for the structural genomics advancements in the life sciences and the battery technology advancements needed for the transition to a hydrogen society. Neutron-based science is also known as high-neutron-intensity-dependent science. In generally, cold neutrons are generated by cooling neutrons produced by nuclear reactions, such as spallation reactions based on accelerators, and fission reactions in nuclear reactors. However, it is not easy to produce slow neutrons, such as ultra-cold neutrons or very-cold neutrons, below thermal equilibrium using cooling method. A new unique method focusing on nanosized particle aggregation has been proposed to increase neutron intensity in that energy region [1]. The method is based on intensity enhancement by multiple coherent scatterings with nanosized particle aggregation. The aggregation of nanosized particles matches the wavelength of below cold neutrons, causing a similar effect to coherent scattering, so-called Bragg scattering, leading to neutron intensity enhancement by several orders of magnitude. Nanodiamonds [2–4] and magnesium hydride [5] have recently been studied numerically and experimentally. The major challenge with nanodiamonds in practical applications is the molding method.
Another carbon structure, graphene is focused on to find a solution to this problem.
It is hypothesized that nanosized graphene could aid coherent neutron scattering under particle size conditions similar to nanodiamonds. Moreover, it might be possible to use it in high neutron radiation conditions due to graphene's strong sp2 bonds.
In this paper, we report the potential of nanosized graphene as a reflector material below cold neutrons, together with experimental results.

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The Roadmap to Applications of Graphene & Related Materials in UAE

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A priority area of Abu Dhabi's economic vision 2030 is Infrastructure Development and Environment Sustainability for robust economic development [1]. This aligns with the strategic UAE initiative to achieve net zero GHG emissions by 2050 [2]. Currently, the value of ongoing infrastructure development projects in the UAE stands at ~$90 billion, which is considerably higher than most of the developed nations when compared per capita [3]. A substantial part of these projects includes Research & Development (R&D). The per capita R&D spending of UAE is among the highest in the world [4]. Some of the prominent priority sectors for future development in the UAE are Renewable & Clean Energy, Transportation, healthcare, water, and space, among others [1]. The Research & Innovation Center for Graphene and 2D Materials (RIC-2D) is complementing Abu Dhabi's vision of transforming into a knowledge-based economy by spearheading research and commercialization projects on graphene and related materials (GRMs). Unique and combination of properties of GRMs could make them key enablers for most if not all aspects of the current application areas of interest in the UAE as broadly mentioned before [5]. RIC-2D has started low (<3) and high TRL (>5) research in four thematic areas: 1) Water, 2) Energy, 3) Lightweight materials and 4) construction. Research work on these thematic areas is being conducted locally and with a network of global institutions among which University of Manchester is a key partner. Application of 2D materials for water are currently focused on desalination and water purification by incorporating 2D materials as selective sensors, membrane additives, active anti-fouling agents, and as corrosion protective coatings among others. Other innovative ideas with GRMs which are also explored include atmospheric water generation [6] and cloud seeding [7]. Green and blue hydrogen generation technology is being developed under the energy thematic area, in addition to other energy storage and generation technologies. For example, UAE is one of the largest sulphur producers in the world [8]. Therefore, it is appealing to build technologies around GRM for sulphur-based batteries. Similarly, applications of GRM-enhanced, stronger, lighter, and smart composite materials and concrete are research areas currently pursued in UAE. Additionally, RIC-2D will eventually also explore the application of 2D materials in quantum information system and products, which cannot be obtained with current materials technologies.

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2D-Exerimental Pilot line: progress in 2D-materials integration in industry relevant environment.

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A decade ago, the kick-off was given to the Graphene flagship initiative [1] by the European Union which enabled the exploration of a broad application zone for graphene and related materials. In time, based upon the many success-stories, the wording started to shift from "intriguing science" to "unique applications". Among those fields: sensors, photonics, spintronics and electronics [2-3] are named. While each of these applications, require specific material and process requirements, many commonalities are found as well. In October 2020, the 2D-Experimental Pilot Line project has been launched with the aim to establishing an ecosystem for integrated 2D materials in the semiconductor industry [4]. In this project, tool manufactures, chemical and material providers and research institutes join forces to provide for new capabilities and prototyping offerings to universities, SME and companies.

A key challenge addressed, to obtain high performance active layers, is the deposition process which consists of the combination of a 2D-material growth and (semi)-automated transfer process [5]. Moreover, the combination of interface control and careful process selection is key for outstanding device performance and wafer-uniformity [6]. Further highlights of pathfinding activities in enabling process transfer from lab to fab [7] will be addressed in this presentation.

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Towards large-area growth of 2D materials by Direct Liquid Injection Chemical Vapor Deposition (DLI-CVD)

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The isolation of graphene has sparked considerable interest from both academic and industrial players around vast new possibilities offered by 2D materials. This class of materials, as their name suggests, are formed of a stacking of 2D layers weakly-bonded to one another, which properties tend to dramatically change when their thickness is reduced to a monolayer [1]. These effects have been investigated extensively and while they show great promise, industrial scaling remains challenging [2]. In this talk, the Chemical Vapor Deposition (CVD) growth of monolayer graphene, molybdenum and tungsten disulfide (MoS_2 and WS_2) and boron nitride (BN) will be tackled with large-scale introduction in mind. Emphasis will be put on the degree of control and detection needed and displayed in Annealsys machines to ensure batch-to-batch reproducibility. In particular, catalyst-assisted CVD and SiC sublimation growth for graphene, Direct Liquid Injection MOCVD (DLI-MOCVD) and DLI-ALD of MoS_2 and WS_2 monolayer, as well as CVD of hexagonal and amorphous boron nitride (hBN and aBN) will be presented. Direct liquid injection (DLI) is a type of precursor delivery that uses liquids as delivery agent [3]. This way, a large variety of liquid and solid precursors can be used in CVD while remaining safely stored at ambient temperature (Figure 1). Furthermore, the feeding rate can be precisely monitored and controlled using fast injectors allowing for very precise tuning of vapor stoichiometry in the reaction chamber.

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Figures

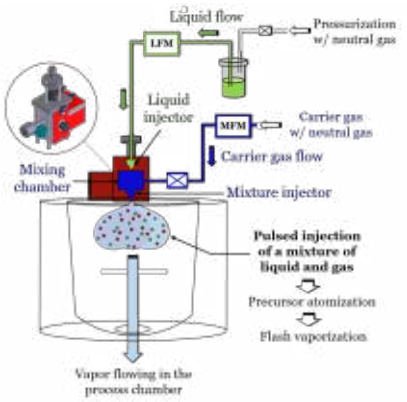

Figure 1: Schematic representation of KEMSTREAM's two-stage DLI system

TERS and TEPL Imaging for 2D Materials Research

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This presentation focuses on the application of Tip-Enhanced Raman Spectroscopy (TERS) in the research of 2D materials.

Starting with a brief overview of the historical development of TERS instrumentation at HORIBA Scientific, we will showcase the technical innovations over the past 15 years. These breakthroughs have enabled TERS to be widely utilized in various applications today.

We will then demonstrate the usefulness of TERS and TEPL (Tip-Enhanced Photoluminescence) in the investigation of 2D materials, including graphene, 2D semiconductors, and MXenes. Through specific examples, we will showcase how TERS provides valuable insights into the structural and chemical properties of these materials at the nanoscale. Additionally, we will present our latest developments, such as environmental control solutions for measurements in liquid and electrochemical environments.

Finally, we will briefly discuss the range of solutions offered by HORIBA Scientific for graphene and other 2D materials research, emphasizing their significance in academic and industrial applications.

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Stakeholder approach for graphene applications

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Abstract

It has been two decades since the first isolation of graphene. Since then, numerous scientific discoveries have been made regarding its extraordinary properties. The development of synthesis methodologies and related applications has also been remarkable, raising high expectations that graphene will revolutionize many industries [1]. As scientific knowledge is accumulated, engineering issues that apply this knowledge to actual products are garnering more attention. Thus, successful commercialization of graphene-based products would require tight collaboration among academia, industry, and potential customers [2].

MCK Tech was established in 2017 to develop and commercialize graphene product. it became a research-based spin-off company of the Center for Advanced Metamaterials (CAMM) in 2019. CAMM, which was launched in 2014 as a center for Global Frontiers Projects supported by the Ministry of Science and ICT, is geared towards developing core technologies in controlling wave energies by incorporating creative artificial structures of sub-wavelength sizes. We are collaborating to develop graphene metamaterials that can control electromagnetic waves using large area CVD graphene.

At MCK Tech, we work closely with our customers. We believe that listening to their needs is crucial since they are more interested in what the material can deliver rather than what the material is. Therefore, we provide customized graphene considering customer requirements such as form factor (roll/sheet), electrical properties, and chemical properties.

During this talk, I will present some of our applications, including graphene metamaterials, as well as sensors for monitoring sodium/potassium excretion in urine [3], among others.

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Figures

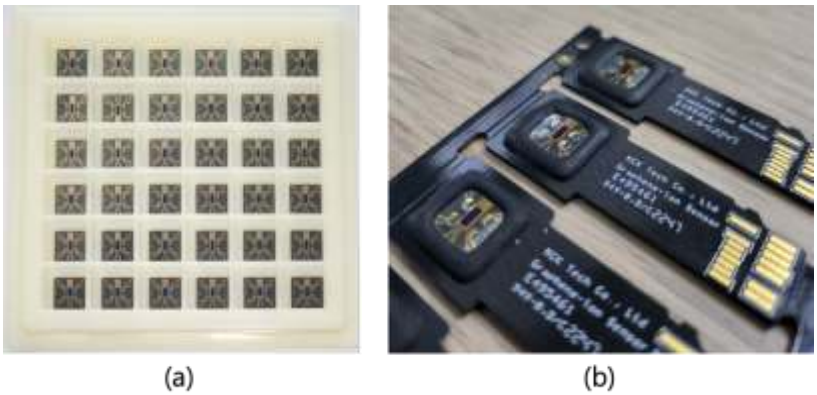

Figure 1: (a) Graphene FET (G-FET) (b) Packaged G-FET

Graphene Analysis Using Coincident XPS- Raman

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Advanced materials present ever increasing challenges to the analytical scientist. Composite materials built from nanostructures or ultra-thin films, often with complex chemistries present, are now required in a broad range of applications, and achieving full characterization is rarely managed using only one analysis method. To maintain confidence in the results from the utilization of several different methods, it is advantageous to be able to perform experiments on the same platform. Ideally, this should be without having to move the sample, removing the need for additional registration or processing to ensure that the data is being collected from the same position.

For surface analysis, it has been common for many years to incorporate related analysis techniques onto the same instrument. For example, X-ray photoelectron spectroscopy (XPS) systems which provide elemental and chemical information from the top ~10nm of samples are commonly equipped with UV light sources to facilitate investigation of additional properties of materials via ultra-violet photoelectron spectroscopy (UPS). The ion source that is typically used for sample cleaning and depth profiling can also be used for low energy ion scattering (LEIS or ISS), providing more surface sensitive elemental composition information than can be delivered from XPS alone. The addition of a focused electron source enables Auger electron spectroscopy (AES) which provides surface sensitive composition at higher spatial resolution than XPS can offer. With the exception of ISS, all these routine additional analysis techniques are electron spectroscopy based, and offer similar information.

We have recently integrated further instrumentation onto a standard XPS system. The integrated system has combined a Raman spectrometer with a micro-focused, monochromated XPS system. The focal points are aligned such that data can be acquired from the same point simultaneously, and that the sizes of the analysis areas are comparable in size. Chemical modifications of the material can be easily determined and quantified with XPS. Raman offers a fast way of determining the quality and conformity of the material, and direct compound identification. The greater depth of field of the Raman spectrometer also offers bulk information to complement the surface sensitive XPS data.

In this presentation we will discuss the strengths of this combined, in-situ approach to surface analysis, illustrated with examples from a range of applications.

Reproducible measurements of the lateral size and thickness of few-layer graphene flakes using SEM and AFM

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Graphene nanoplatelets (GNPs), typically produced as powders or in liquid dispersions are already starting to find commercial application via small-to-medium enterprises (SMEs) to multi-national corporations, for a large range of application areas. There are currently over 100 commercial 'graphene' producers worldwide, including leading producers in Europe, with an 'on paper' offering of materials with vastly different properties and types. However, the activity of many suppliers (and buyers) is hindered due to materials properties being unknown or poorly characterised, often consisting of graphite rather than GNPs or having large batch-to-batch variations. Real world products and applications suffer as a result. Thus, validated and disseminated measurement methods of GNPs are a key industry requirement. To this end, an international interlaboratory comparison (ILC) to determine the lateral flake size distribution of graphene nanoplatelets (GNPs) using scanning electron microscopy (SEM), and to then correlate this distribution to measurements of lateral flake size and thickness using atomic force microscopy (AFM), has been undertaken [1]. The outcomes of this ILC will directly input in the future revision of the international standard for measuring the structure of GNPs - ISO/TS 21356-1 [2,3], providing an insight on method variability and objective data to build robust and widely applicable measurement procedures across different instrument models and laboratories.

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Figures

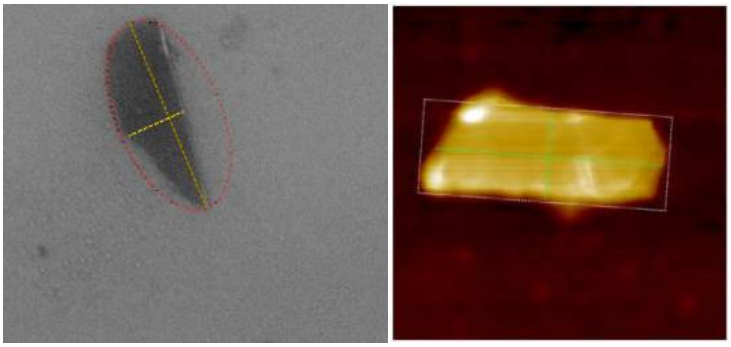

Figure 1: An example of high-resolution images, SEM (left) and topographic AFM (right) of individual particles showing the measurement methodology [1].

Graphene-Realization of large commercial scale applications

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Abstract: Emerging advanced material Graphene is showing exponential growth in the past decades with immense business opportunities globally. Multiple superlative properties [1] packed in a single material which is being adapted by majority of commercial applications. In this present work we are reporting a use-inspired research of graphene performances in a large-scale commercial application such as polymer packaging, sports wears, home textiles and infrastructure applications. We are successfully able to produce in a large-scale.

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Enabling a world of enhanced perception

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Qurv develops wide-spectrum image sensor technologies and integrated solutions to enable next-gen computer vision applications, addressing the expanding needs of an autonomous and intelligent new world. Qurv's technology leverages current CMOS scalable manufacturing and quantum materials to unlock new levels of performance, reliability and function in XR devices, service robots and automotive.

Qurv image sensors based on CMOS compatible quantum materials such as 2D materials and colloidal quantum dots are sensitive to visible (Vis), near-infrared (NIR) and short-wave infrared (SWIR) light. [1,2] Sensitivity in the range beyond silicon CMOS imager sensitivity (>1 µm wavelength) provides for dramatically reduced sunlight interference and eye safe LIDAR operation. Furthermore, sensitivity in the SWIR range can enable true night vision, all weather vision and molecular vision.

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Figures

Figure 1: . *The accessible electromagnetic spectrum with different imaging technologies and their respective spectral range. The icons illustrate the advantages for different types of use cases enabled by the different ranges of the spectrum.*

Growth of TMD Nanoribbons for Quantum Electronics

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The dependence of single atomic layer materials band structure on their width and internal strain offers a new dimension for realization of quantum electronic and optoelectronic devices. Therefore, developing facile methods for their controllable synthesis is of central importance. We will present surfactant-mediated growth of single atomic layer of transition metal dichalcogenides (TMD) nanoribbons from nanoparticles of Ni-Me-Na-S (M=Mo, Se, S2 and Se) via vapor-liquid-solid mechanism. The width of the precipitated nanoribbons (7-100nm) correlated with the size of the seed nanoparticle. We observed width-dependent Coulomb blockade oscillation observed in the transfer characteristics of the nanoribbon's with width less than 20nm at temperatures up to 60 K. Moreover, remarkable flexibility of grown nanoribbons and resilience to the high strain allows to realize clean quantum emission associated with deformation induced in the electronic states. The method provides basic synthesis route for atomic scale width control quantum nanoribbons of metal dichalcogenides for potential applications in the quantum electronics.

Graphene Flagship summary and the future CSA

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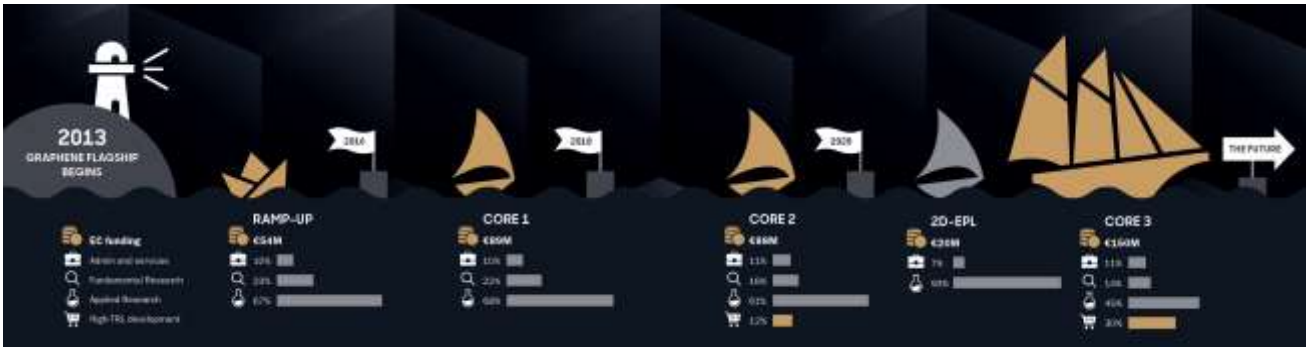
Abstract

The formation of the business development function, creation of spearheads and active collaboration between work packages and external stakeholders have all had a profound effect on the results and success of the project. Given the 106 products created, 17 spin-off companies launched and the increase of industrial partners in the project from 30% in 2013 to 47% in 2022, our commercialisation efforts are on the right track.
In October 2023, the Core 3 project will end, and the Graphene Flagship will kick off a new initiative under Horizon Europe, the European Commission's financial instrument from 2021–2027. The 2D Experimental Pilot Line project will continue to be funded through Horizon 2020 for another year. With this transition, the structure of the Graphene Flagship initiative will also change. Rather than a single project, the initiative will be organised into separately funded Research and Innovation Actions and Innovation Actions (RIA/IAs) connected by a Coordination and Support Action (CSA).
To date six RIAs and one IA have been selected across composites, photonics, energy and biomedical areas.
A brief summary of the Graphene Flagship and the future RIAs/IAs and CSA will be presented.

Figures



Accurate Measurement of Size of Graphene Oxide Flakes by Scanning Electron Microscopy (SEM)

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Accurate characterisation of the morphology (size) and chemistry of graphene-related 2D materials (GR2M) is key in understanding their extraordinary functionalities [1]. Hence, not only the tailoring of these functionalities aiming at applications of increased-performance becomes possible, but also the correlation of the physico-chemical properties with the understanding of the potential toxicity eventually enables a safe and sustainable development of the GR2M for new applications [2,3]. Whilst AFM and Raman Spectroscopy are recommended to measure the thickness of GO flakes, Scanning Electron Microscopy (SEM) is the most suited method to assess their lateral size, which varies between tens of µm down to below 100 nm [4-6]. In this paper, procedures for the accurate determination of lateral size of graphene oxide (GO) flakes by SEM are presented. The prerequisite for accurate flake size analysis is the proper sample preparation, i.e. deposition of ideally isolated flakes on a substrate, with the flakes being unfolded, non-overlapped, parallel with the substrate, and having a high coverage density. Examples of optimum image caption conditions and image analysis procedures will be presented. The size descriptors and their measurement are described in the context of the corresponding analysis approach: i) length and width of the flakes with a quick and rough, but robust procedure, and ii) exact contouring of the flakes as part of a highly accurate, but more time-consuming measurement approach. The possibility of application of automated image analysis is discussed as the alternative to the manual flakes analysis.

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Figures

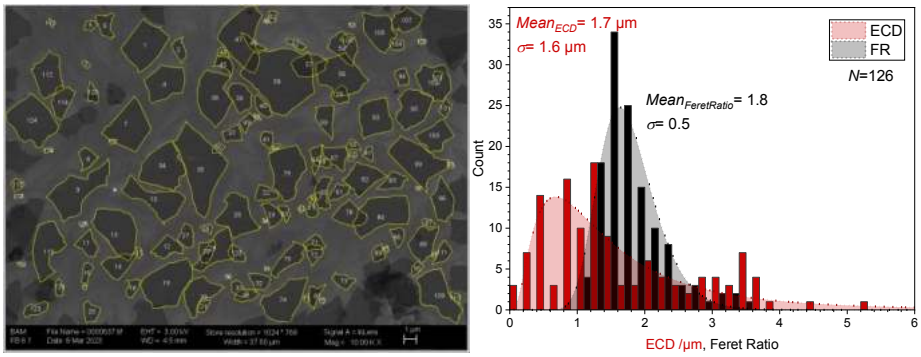

Figure 1: SEM image after manual contouring of 126 graphene oxide flakes including their size measurement with ECD (equivalent circle diameter) and Feret ratio as selected size descriptors.

Boron Nitride Materials for Next-Generation Interconnect Technologies

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Abstract

With the current explosion in data, high-performance computing and the advent of new technologies such as super-large artificial intelligence (AI), the demand for high-performance processors for computation and high-capacity memory for data storage is rapidly increasing. Also, with continued device scaling, the interconnect for fast and energy-efficient signal processing between processors and memory have increasingly become the technological bottleneck significantly impacting the performance and reliability of electronic devices. The performance of the interconnect is basically dictated by the resistance and capacitance (RC) signal delay, which in turn, is related to the metal wire resistance, dielectric layer capacitance, and interconnect dimensions. Over the years, key interconnect components such as metal wires, diffusion barrier/liners have evolved with material and structural innovations to match the performance requirements of next-generation devices. However, the low-k interlayer dielectric materials have had difficulties with materials innovation since the early 2000's, struggling with limits to permittivity scaling and required material requirements until the recent discovery of amorphous Boron Nitride (a-BN) which demonstrated mechanically and electrically robust films with ultralow κ values <2.0.

In this talk, the evolution of the development of low-k materials and the recent progress of a-BN ultralow-k (ULK) dielectric materials are presented. The materials design of ultralow-k materials and the outlook on the prospects of ULK dielectric applications in next-generation semiconductor devices are discussed.

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GrapheNovation – Commercialising High Value Nanotechnology Solutions to the Market

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Malaysia's first National Graphene Action Plan (NGAP) on Graphene commercialization was formed in 2014 spearheaded by NanoMalaysia Berhad through 11th Malaysia Plan. Considering the technology and market landscape of Malaysia manufacturing, the key areas of graphene commercialization was identified to be lithium-ion batteries, rubber additives, plastic additives, conductive inks and nanofluids.

Among the achievement through this programme includes deployment of graphene based nanofluids for cooling system which improves energy saving up to 29%, sago starch based graphene additives for oil drilling fluid loss which increases the life span of drill bit about 75% or 40% cheaper, reclaimed tyres improved with graphene to increase set compression and tensile strength by 10% together with 30 other products.

Aligning to the global fourth Industrial Revolution (4IR), graphene commercialization has also taken a pivotal paradigm shift. GrapheNovation is a programme under NanoMalaysia Berhad which focusses on enhancing Malaysian nanotechnology ecosystem using Graphene in Internet of Nano-Things (IoNT). Four main pillars for GrapheNovation includes energy storage, energy generation, sensor technology and advanced technology. These key areas are integral component of 10-10 Malaysian Science, Technology, Innovation and Economy Framework (MySTIE) and Malaysian Advanced Material Technology Roadmap, Industry4WRD.

As a summary, since the start of NanoMalaysia's commercialisation programmes in 2016, to-date more than 70 technologies, products and solutions has been developed with various industries resulting in more than 50 project Intellectual Properties developed for value creation; with more than 30,000 potential high value job opportunities and more than RM34 billion in potential GNI contribution over the next 5 years.

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Performance, challenges, and reliability of 300mm FAB integrated 2D TMDCs based devices

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2D TMDCs have demonstrated to be rightful contenders as the material of choice for high performance logic devices. However, most of these demonstrations have been done, up to now, relying on lab-base integrated devices. In this talk I will start by covering the performance of devices and circuits integrated in a 300mm FAB [1]. Then I will proceed to put the spotlights on some of the challenges such an integration might represent for the industry. I will then follow with the solutions that we have found for some of them such as top contacting integration on 300mm scale [2] and 300mm FAB integrated devices using a 300mm compatible transfer of 2D materials [3].

I will then proceed to highlight the importance of the reliability of such devices and a first order analysis of the BTI impact on 2D TMDCs based 300mm FAB integrated devices [4] and the effect of the gate-stack oxide charge trapping on the performance of such devices [5].

Finally, I will motivate and challenge the community to commit to tackle further obstacles foreseen on the integration in a reliable and reproducible way of this materials on novel high performing devices architectures [6].

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Enabling Reproducibility and Robust Measurement: International Interlaboratory Comparisons for Graphene Standardisation

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With graphene now being used in real-world products, industry needs internationally standardised procedures more than ever, to determine the material properties of graphene and related 2D materials (GR2Ms), so that different materials produced by different manufacturers in different countries can be reliably compared. This work has been ongoing within international standardisation bodies for several years, with international standards recently being published [1].
However, to ensure accurate and reproducible standards are produced, measurement protocols must be verified through international interlaboratory comparisons (ILCs), testing the same method for the same one material across many laboratories around the world.
To introduce this session of the Industry Forum, focussed on the metrology research undertaken within the EU-funded ISO-G-SCoPe project and through the Versailles Project on Advanced Materials and Standards (VAMAS), the current landscape of graphene standards within ISO and IEC will be discussed, as well as the overall strategy of addressing the measurement of different types of material properties of GR2Ms and how VAMAS ILCs can be used to verify the methods that are being standardised. How these studies reveal the sources of uncertainty, which allow us to improve the precision or accuracy of a measurement and quantify the associated uncertainty, will be discussed and a recent example detailed for Raman spectroscopy of graphene produced using chemical vapour deposition [2]. The impact of this work on industry demonstrates how metrology is an important part of providing different companies the confidence in both their products and materials produced around the globe.

References

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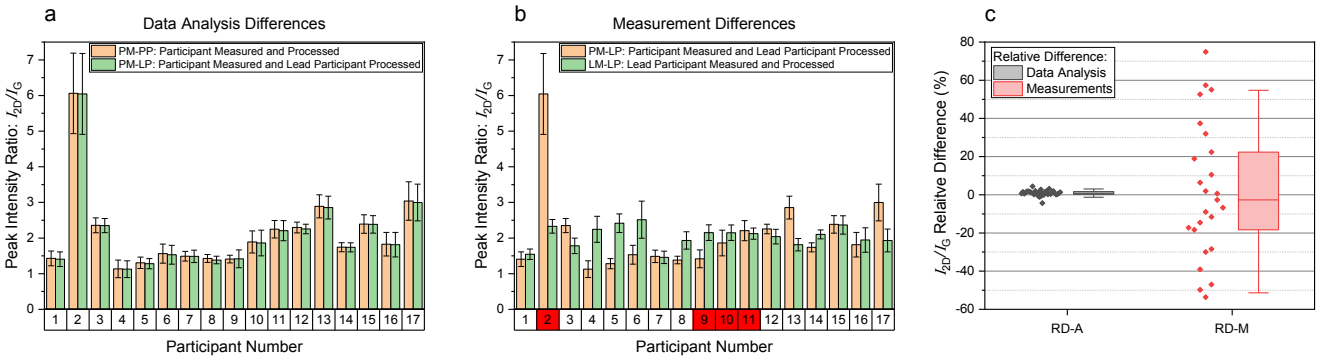

Figure 1: Example results from an international interlaboratory study to determine the source of uncertainties in the measurement of graphene using Raman spectroscopy [2].

Interlaboratory comparison on the quantification of the number of layers of graphene by Raman spectroscopy

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As of today, in Europe alone there are more than a hundred commercial producers of products advertised as "graphene". Still, these materials are often uncharacterised and present great variability even on a batch-to-batch basis, and they are often ascribable to graphite. Considering the widespread impact that graphene is predicted to have on so many industry and research areas, the need for standard characterisation procedures is high; however, these cannot be developed without proven and reproducible methods, established by international efforts and validated through interlaboratory comparisons.
VAMAS (Versailles Project on Advanced Materials and Standards [1]) promotes international efforts on measurement standardisation and harmonisation; the work presented here is a VAMAS project (project 11) under technical working area (TWA) 41 — Graphene and Related 2D Materials — concerning an interlaboratory comparison on the use of Raman microspectroscopy to characterise two types of graphene-related materials: flakes originating from commercially available electrochemically exfoliated powder containing few-layer graphene; and samples of mechanically exfoliated graphene from highly ordered pyrolytic graphite, both deposited on SiO_2/Si substrates. By using a standard operating procedure for measurements using Raman spectroscopy and its parameters [2], including instrumental calibration [3], and making use of a micro-patterned substrate for unambiguous sample location and measurement, the typical variability in results obtained by different users and laboratories across the world will be evaluated. The objectives of the study are the validation of a standard method for the determination of the number of layers in few-layer graphene, determining the uncertainties associated with measurement and data analysis, and to steer input into a future revision of ISO/TS 21356-1 "Structural Characterization of Graphene" [4].

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Reliable chemical characterization of industrial graphene related materials

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International standards describing reliable protocols will facilitate the commercialization of graphene and related 2D materials [1]. One physico-chemical key property next to flake size and thickness is the chemical composition of the material. Therefore, an ISO standard is under development with X-ray photoelectron spectroscopy having a prominent role [2]. With its information depth of around 10 nm which is the similar length scale as the thickness as of particles of 2D materials consisting of a few monolayer XPS seems to be highly suitable for this purpose. Different sample preparation methods like pressing the powders onto adhesive tapes, into recesses, or into solid pellets result in inconsistencies in the quantification. For the validation of the quantification with XPS an interlaboratory comparison was initiated under the auspice of the "Versailles Project on Advanced Materials and Standards" (VAMAS). First results confirm that the sample preparation method (pellet vs. powder) influences the quantification results clearly. Considering this effect, a good agreement of the results from the different participants were observed (see Figure 1). Similar results were observed for raw, N- and F-functionalized graphene.

References

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Figure

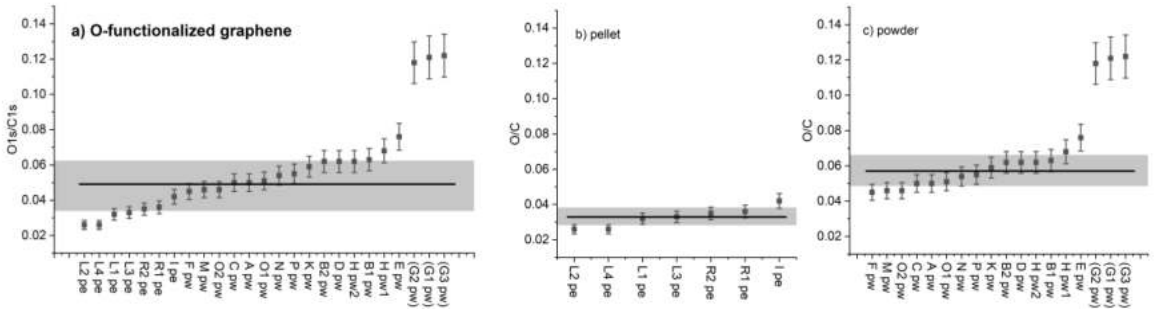

Figure 1: O/C ratios found with XPS from different laboratories for O-functionalized graphene.

Graphene CMOS integration for sensors and imagers

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Abstract

The complementary metal-oxide semiconductor (CMOS) readout integration of the graphene devices enables several key application areas including biosensor and gas sensor arrays[1] as well as imaging arrays[2]. For biosensing the reliability of analysis is becoming increasingly important as point-of-care diagnostics are transitioning from single analyte detection towards multiplexed multianalyte detection. Multianalyte detection benefits greatly from CMOS integrated sensing solutions, offering unique opportunities with multiplexed sensing arrays, integrated readout electronics and sensor array miniaturization. The development of CMOS integration compatible graphene field-effect transistor (GFET) based biosensing has been rapid during the last few years, both in terms of the fabrication scale-up and functionalization towards biorecognition from real sample matrices. The next steps in industrialization relate to improving reliability and require increased statistics. Regarding functionalization and referencing towards truly quantitative sensors and on-chip bioassays, improved statistics require sensor arrays with reduced variability in functionalization. Such multiplexed bioassays, whether based on graphene or on other sensitive nanomaterials, are among the most promising technologies for label-free electrical biosensing. As an important step towards that, we report wafer-scale fabrication of CMOS integrated graphene FET arrays (Figure 1a) with high yield and uniformity, designed especially for biosensing applications. We demonstrate the operation of the sensing platform array with 512 GFETs in simultaneous detection for sodium chloride concentration series (Figure 1b). This platform offers a truly statistical approach on graphene FET based biosensing and further to quantitative and multi-analyte sensing. The technique can also be applied to other fields relying on functionalized GFETs, such as gas or chemical sensing or infrared imaging. For these we also demonstrate the wafer-scale fabrication of the GFET devices on CMOS readout wafers and the multi-project wafer run possibilities in the 2D-EPL.

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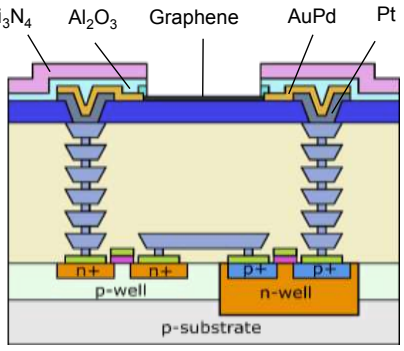
Figures

a)



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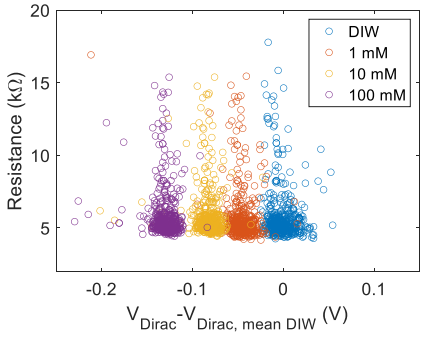

Figure 1: a) A schematic illustration of the post-processed GFET on CMOS readout b) The resistance values as a function of V_{Dirac}-$V_{Dirac,\ mean\ DIW}$ for the 512 GFETs.

2DM-Industry Consortium to Boost the industrial landscape in layered materials

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The two-Materials Industry Consortium (2DM-IC) aims to catalyze economic growth and bolster the competitiveness of the European 2DM industry by promoting the broad-based implementation of 2DM technology across various EU industrial sectors. We draw inspiration from the groundbreaking work of the Graphene Flagship project, which brought significant innovation to the research sphere of 2DM, yet left much potential untapped in the industrial application of these materials.

To bridge this gap between research and industry, we propose the establishment of a business-driven association, aiming to cultivate an industrial framework for 2DM in Europe. This consortium will bring together large enterprises, Small and Medium Enterprises (SMEs), start-ups, and investors currently operating within the EU's 2DM sector. Through a collective drive towards an innovation-focused agenda and high Technology Readiness Level (TRL) research and development, we aim to stimulate the much-needed industrial growth in the 2DM sector.

We further advocate for increased public and private investments in the 2DM sector to support this innovative growth. Our ultimate objective is to enable EU companies to expedite their deployment within the EU market, thereby directly stimulating the expansion of the field and contributing to the overall benefit of the EU economy and society. Through these concerted efforts, we intend to enhance the EU's 2DM industry's landscape, fostering an environment of innovation, competitiveness, and economic prosperity.

Graphene Research and Commercialisation (From the Lab to the Marketplace)

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Abstract

Graphene@Manchester works to **accelerate** the **research** and **commercialisation** of graphene and other 2D materials, by directly supporting startups, spin-outs, SMEs and MNEs, with **advice**, **expertise,** and access to **world class facilities and capabilities.** The GEIC is home to technological excellence, both in our advanced facilities and in the expertise of our application managers, engineers and technicians in the field of graphene and wider 2D materials.
The GEIC is home to a wide range of **highly flexible laboratory spaces**, from dedicated and shared partner labs to application-specific spaces (eg. membranes, energy, coatings, thin-film technologies, composites, construction technologies & characterisation) and an industrial-sized 'high bay' lab for large equipment and process scale-up.
More than 150 companies to date from all over the world have chosen to partner with The University of Manchester to make their graphene and 2DMatereials projects a reality. We continue to receive many enquiries from large and small companies to work with us and we want to see how we might collaborate to take those ideas and interests into projects and then products and applications. We now offer a number of tailored partnerships for organisations that wish to have either a short or a deeper and more prolonged engagement than traditional short-term contracts might provide.

Projects can be delivered in a number of ways, typically starting with feasibility studies or ideation workshops and long term using our membership models.

Figures

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