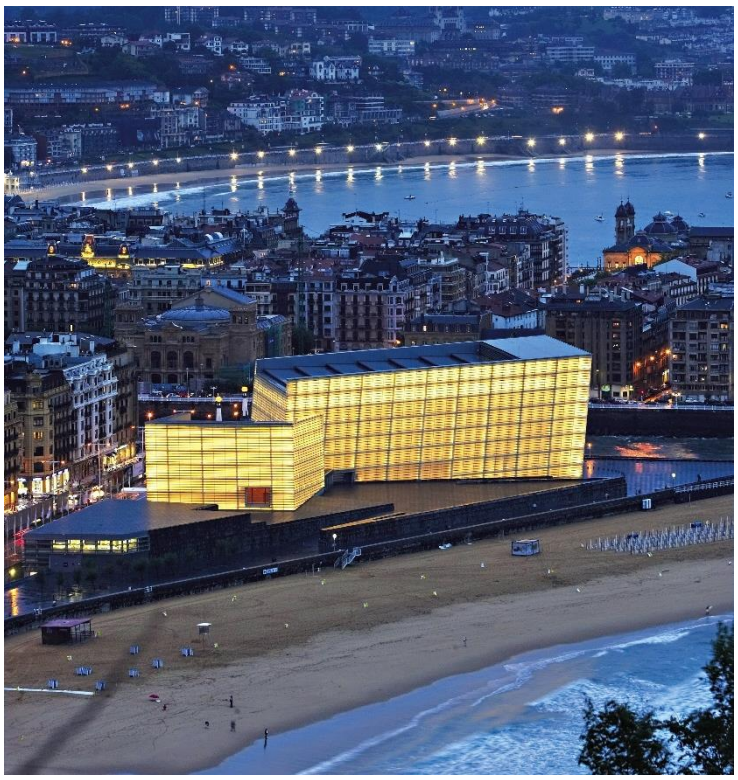
Graphene Flagship EU-US Workshop
Graphene and related 2D materials

Kursaal Congress Centre, San Sebastian, Spain
13-14 September 2018



Workshop Report

Workshop chairs: Vladimir Fal'ko (Manchester University, UK), Joshua Robinson (Pennsylvania State University, US), Frank Koppens (ICFO, Spain)

Content

Overview

The 4th Graphene Flagship – US NSF Workshop was held at the Kursaal congress centre, San Sebastian (Spain), during 13-14 September 2018. For the first time, the workshop was organised as a parallel session of the Graphene Week 2018 conference and gathered over 150 attendees.

The workshop provided an open technical forum that brought together leading researchers from the US and Europe to discuss cutting-edge research in 2D materials, their heterostructures and devices based on such materials. The aim was to facilitate scientific exchanges and discuss needs and mechanisms for future collaborations. This was a follow up to the third "US-EU Workshop on 2D Layered Materials and Devices" held on 23-25 October 2017 in Arlington, Virginia (USA).

Co-organized by the European Graphene Flagship project and the National Science Foundation (NSF), the workshop provided a ground for discussing the common challenges and opportunities in area of graphene and 2D materials. The aim of the workshop was to learn about each other's work and to further the strong tradition of collaboration between the US and European Union.

The meeting was co-organised by US and European researchers and was co-chaired by:

- **Professor Joshua Robinson (Pennsylvania State University, US)**
- **Professor Vladimir Fal'ko (Manchester University, UK)**

The workshop gathered **18 speakers** (8 from the United States and 10 from Europe), coming mainly from academic institutions. For details about the scientific content of the presentations, see the *Speakers and Abstracts* section.

This report summarises the main conclusion of the discussions and the envisaged ways for collaborations.

Common challenges and opportunities for collaboration

Graphene-research in EU is at the transition between academia and industry as more and more activities are industry-driven. There are about 30 products on the market derived from the Flagship and more are to come in the coming years.

Graphene Flagship is starting the preparation of the Core 3 project and the possibility of collaborations with the US may come up, therefore this should be closely followed in the future.

Quantum research

Quantum research can be one common topic for collaborations between EU and US. Quantum science is one of the US priorities and the National Quantum Initiative Act will start in 2019 and last for 10 years. In Europe, Quantum Flagship will be kicked-off at the end of November 2018 and the Graphene Flagship cannot duplicate research that will be performed within the new flagship, therefore Graphene Flagship will reduce its quantum activities. Quantum sensing, information and communication are targeted in the Quantum Flagship; however, quantum engineering is not included in the Quantum Flagship activities, not even in the fundamental activities. Therefore, there will be a lot of possibility available for EU-US collaborations in quantum activities.

Access to infrastructure

Facilitating access to infrastructure dedicated to 2D materials synthesis across the Atlantic would be highly beneficial for researchers as it would allow the best use of available resources and access to complementary expertise and facilities. Unique facilities and centres of competence for the investigation of 2D materials existing in the US or EU have been named as relevant for enhancing transatlantic collaborations.

There are also potential collaborative opportunities with the NSF 2D Materials Innovation Platform based on the 2D Crystal Consortium at Pennsylvania State University (PSU). This is a service centre and so there is need for writing a proposal in order to have access to the facility. At the moment, the facility is open at national level, but it may be envisaged to have it open to international collaborations.

CVD crystal growth facilities are also attracting students from EU in order to be trained.

Standardisation

Standardisation of materials produced by different techniques (and suppliers) was identified as a common challenge which is important to solve as to advance towards the industrialisation of graphene and other 2D materials-based products and technologies. Collaboration would be very important as to set common and accepted standards across the Atlantic and even worldwide. Supporting industrial innovation with measurement is therefore an area where there are real opportunities to foster collaborations between the Flagships' efforts in standardization and the US National Institute of Standards and Technology (NIST). A list of materials that currently are being growth can be shared with the EU partners.

Collaboration grants

The mobility program for researchers supported by the Graphene Flagship is a very good opportunity to enhance collaboration and provide opportunities for European and US researchers; however, the program is not very utilised. NSF provides also grants for US researchers and there has already been a visitor from Texas coming to perform research in Europe.

Future collaborations and next steps

At the end of the meeting, there was clear interest to continue the series of workshops by organising the next workshop again in US in 2019. The format of this year workshop was discussed, and it was concluded that it is a good opportunity to have the workshop embedded in the Graphene Week conference as it benefits young researchers/students who can this way attend the high-level scientific presentations, and it is also a gain of time for the EU and US delegates. This format definitely increases the visibility and impact of the workshop. In two-year time (2020), the EU-US workshop could be again held jointly with the Graphene Week. The format and time may be changed in order to allow more US delegates and conference participants to attend.

There was consensus regarding the next US-EU workshop (2019):

• The workshop should be held jointly with a conference or another event taking place in the US. One envisaged option will be to hold the workshop in spring 2019 (mid-May) jointly with another workshop on graphene that will be open to the EU delegates. Practical arrangements should start soon, and the format of the workshop needs to be discussed.

- Visit of facilities will be also considered for the next year edition.

Programme

Time	13 September	14 September
10:00		**Joshua Robinson** Discovering, Creating, and Exploring Novel Atomically-Thin Materials and Heterostructures
10:15		**Marco Polini** Linear and non-linear hydrodynamic flow in graphened
10:30		**Roger Lake** One dimensional van der Waals materials
10:45		**Alexander Tartakovskii** Resonantly hybridized excitons in moiré superlattices in van der Waals heterostructures
11:00-11:30	**Coffee break**	**Coffee break**
11:30		**Alessandro Tredicucci** On-chip strain engineering platform for two-dimensional materials
11:45		**Eric Vogel** Two-dimensional Materials for Vertical Heterostructures
12:00		**Robert Wallace** Contact interfaces and transition metal dielectrics
12:15		**Peide Ye** 1D van der Waals Nanomaterials: Selenium and Tellurium
12:30		**James Hwang** Wafer-scale fabrication of MoS2 and PtSe2 MOSFETs
12:45		**Eric Pop** Electrical, thermal, and unconventional applications of 2D materials
13:15-15:30	**Lunch**	**Lunch**
15:30	**Andrea Ferrari** Light scattering and emission from layered materials	**Discussion on collaborations** **Moderators: Eric Pop, Joshua Robinson and Vladimir Fal'ko**
15:45	**Vladimir Fal'ko** Infrared and THz optics of atomically thin films of transition-metal dichalcogenides and post-transition metal chalcogenides	
16:00	**Marco Romagnoli** Graphene use in photonics for telecommunications	
16:15	**Susan Fullerton Shirey** Using ions to control transport in two-dimensional materials	
16:30-17:00	**Coffee break**	**Farewell Reception** *Topa Sukalderia* *Calle Aguirre Miramón 7 - Donostia* *+34 943 569 143*
17:00	**Thomas Mueller** Second harmonic generation in strained 2D semiconductors	
17:15	**Roman Gorbachev** Fabrication of the next generation of devices	

17:30	**Giulio Cerullo** Electron cooling and carrier dynamics	*http://topasukalderia.com/en/*
17:45	**Jurgen Smet** Ultrafast diffusion and superdense ordering of lithium in bilayer graphene	

Participants

1. Andrea Ferrari, Cambridge University, **United Kingdom**
2. Marco Romagnoli, CNIT, **Italy**
3. Susan Fullerton, University of Pittsburgh, **United States of America**
4. Thomas Mueller, TU Wien, **Germany**
5. Roman Gorbachev, Manchester, **United Kingdom**
6. Giulio Cerullo, Politecnico de Milano, **Italy**
7. Jurgen Smet, MPI Stuttgart, **Germany**
8. Joshua Robinson, Pennsylvania State University, **United States of America**
9. Marco Polini, ITT, **Italy**
10. Roger Lake, University of California Riverside, **United States of America**
11. Alexander Tartakovskii, Sheffield, **United Kingdom**
12. Alessandro Tredicucci, University of Pisa, **Italy**
13. Eric Vogel, Georgia Institute of Technology, **United States of America**
14. Robert Wallace, University of Texas at Dallas, **United States of America**
15. Peide Ye, Purdue University, **United States of America**
16. James Hwang, Lehigh University, **United States of America**
17. Vladimir Fal'ko The Manchester University, **United Kingdom**
18. Eric Pop, Stanford University, **United States of America**
19. Joan Redwing, Pennsylvania State University, **United States of America**
20. Angela Height Walker, **United States of America**
21. Jari Kinaret, Chalmers University, **Sweden**
22. Frank Koppens, ICFO, **Spain**
23. Ana-Maria Ciubotaru, ESF, **France**
24. Ana Carolina Selvati, ESF, **France**

Speakers and abstracts

Graphene use in photonics for telecommunications

Marco Romagnoli
CNIT – Photonics Networks and Technologies National Laboratory, via Moruzzi 1, 56124 Pisa, Italy

Abstract:

Graphene is a single atomic layer of carbon atoms with outstanding electrical and optical properties. From the optical point of view graphene is a material that can be operated both in electro-absorption and in the electro-refraction regime. These two regimes depend carrier doping of the material. In the electro-refractive regime there is a change of index of refraction with minimum contribution of the absorption. This indicates that graphene can be a phase modulator. In terms of electrical properties graphene shows an electrical mobility dependent on carrier density largely superior than Si, InP and InGaAs and this property may influence the transparency in high doping regime. The combination of efficient electro-refractive effect and possible low absorption due to the high carrier mobility makes graphene a good candidate for good figure of merit phase modulators of potential importance for optical communications.

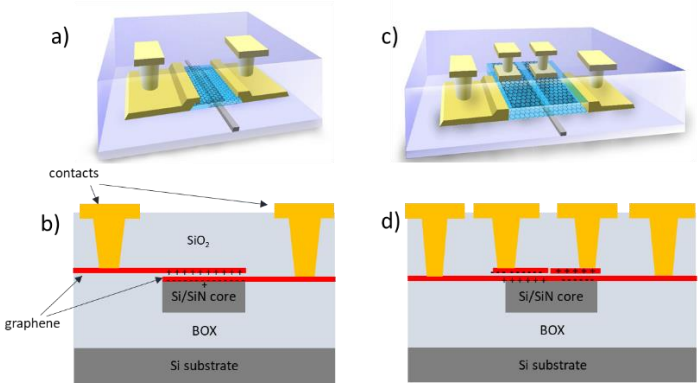


Figure. 3D image and cross section of a) – b) graphene double layer modulator on silicon waveguide and, c) – d) split gate photodetector based on photothermal effect

In the electro-absorption regime graphene shows always simultaneous occurrence of absorption and instantaneous phase change, chirp. The chirp plays a role in fiber transmission for short distances as for instance short interconnects commonly used in data centers.

Another unique behaviour of graphene is observed in photodetection. Being a gapless material the photodetection is wavelength independent, ultraphast photodetection has been observed either in the bolometric configuration and in the photothermal effect (PTE) case. PTE effect is attractive because it can be optimized for both high detection speed (>100 GHz) and efficiency. The PTE effect involves hot carriers that reach temperatures sufficient to generate a voltage through the Seebeck effect. These electrons and holes generate a local photovoltage via the Seebeck effect which can be amplified by a voltage amplifier rather than a transimpedance amplifier. This property has advantages in terms of low dark current. In addition, direct voltage detection may overcome issues with the dark current of photovoltaic schemes with bias. On-chip integrated photodetectors with Si photonics have been reported, typically based on metal–graphene–metal structures evanescently coupled to Si waveguides. In these photodetectors, the guided mode enables longer interaction between SLG and the optical waveguide compared with free- space illumination. A PTE graphene photodetector integrated on a Si waveguide with 0.36 A/W responsivity and 40 GHz bandwidth was also demonstrated by placing drain and source asymmetrically with respect to the waveguide core and using a gate to maximize the Seebeck coefficient of the graphene. These results are very promising for next generation datacom and telecom applications.

Reference:

[1] M. Romagnoli et al, Graphene based integrated photonics for next generation datacom and telecom, Nat. Rev. Mat., vol. 3, no. 18, pp. 392 - 414 (2018)

Using ions to control transport in two-dimensional materials

Ke Xu[1], Jierui Liang[1], Md Mahbubul Islam[2], David Guzman[2], Alan C. Seabaugh[3], Alejandro Strachan[2], **Susan Fullerton-Shirey***[1]

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Abstract:

Electric double layer (EDL) gating is a popular tool for exploring transport in two-dimensional (2D) materials. Mobile anions and cations drift through an electrolyte by field-effect to the interface of a semiconducting channel where image charges are induced (Fig. 1a). The approach is powerful because large sheet carrier densities (~1013 - 1014 cm-2) [1] and reconfigurable doping (p-type, n-type, p-n junctions) [2] can be achieved, revealing exciting new physics (e.g., spin-polarization [3] current induced electroluminescence [4], and superconductivity [5].) Although the EDL gating approach provides exceptional gate control and reconfigurability, multiple limitations of commonly used electrolytes prevent their transition from a tool for studying basic transport properties to an active device component. One limitation relates to the physical properties of the electrolytes – ionic liquids and soft polymers are often used which have poor mechanical properties and are deposited as thick films (i.e., many microns). Another limitation relates to speed – for some electronic device applications, EDL formation/dissolution must occur on pico- to nanosecond timescales. We are addressing the drawbacks to the physical properties of commonly used electrolytes by developing a solid-state, monolayer electrolyte – a single molecular layer of an ionically conductive material that can be deposited as an ordered array on 2D crystals [6] (Fig. 1b). Reversible ionic doping on graphene, MoS2 and WSe2 field-effect transistors (FETs) has been demonstrated using the monolayer electrolyte. We are also addressing speed by measuring and modeling the EDL dynamics in a commonly used solid polymer electrolyte based on polyethylene oxide (PEO). From the experiments, we show that the field strength can be increased by applying voltage pulses to the system; these pulses are long enough to drive EDL formation, but short enough to avoid electrochemistry. From the modeling, we learn that EDL dynamics can occur on fast (pico- to nanosecond) timescales if the field strength is sufficiently high (V/nm). EDL formation dynamics from both modeling and experiment show that the timescale for EDL formation can be tuned by 109 by adjusting the field strength by 103 (Fig. 1c).

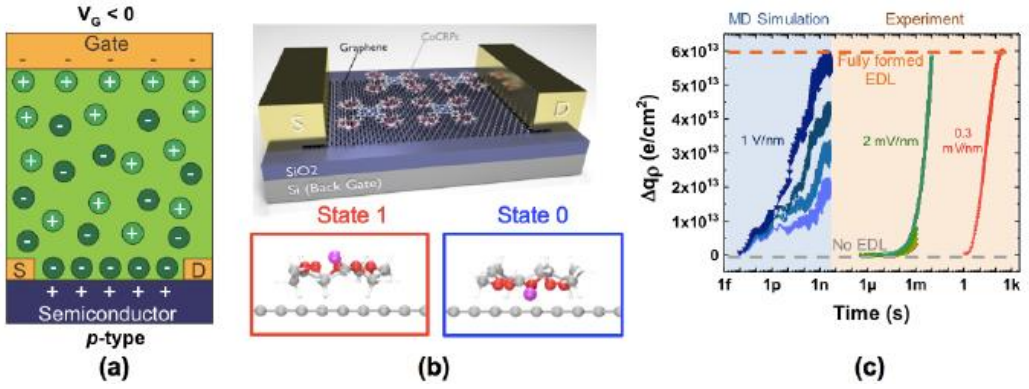

Figure 1: (a) Schematic of electric double layer (EDL)-gated FET. Negative topgate bias (VG) drives anions to the surface of the semiconductor inducing p-type doping. (b) Schematic of monolayer electrolyte doping a graphene FET, and the two possible states. (c) Sheet density versus time showing the time-dependence of EDL formation in response to three different electric field strengths: 1

V/nm in MD simulation, 2 mV/nm pulsed voltage (experiment) 0.3 mV/nm continuous (experiment).

References:

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[2] H. Xu et al., ACS Nano 2015, 9 (5), 4900.
[3] H.T. Yuan, et al., Nature Phys. 2013, 9 (9), 563-569.
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[5] W. Shi, et al., Scientific Reports 2015, 5. [6] K. Xu et al., ACS Nano 2017, 11, 5453.

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Second harmonic generation in strained 2D semiconductors

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Abstract:

Strain engineering is widely used in material science to tune the (opto-)electronic properties of materials and enhance the performance of devices. Two-dimensional atomic crystals are a versatile playground to study the influence of strain, as they can sustain very large deformations without breaking. Various optical techniques have been employed to probe strain in two-dimensional materials, including Raman and photoluminescence spectroscopy. Here we demonstrate that optical second harmonic generation constitutes an even more powerful technique, as it allows extraction of the full strain tensor with a spatial resolution below the optical diffraction limit [1]. Our method is based on the strain-induced modification of the nonlinear susceptibility tensor due to a photoelastic effect. Using a three-point bending technique, we determine the photoelastic tensor elements of four different transition metal dichalcogenide monolayers: MoS2, MoSe2, WS2 and WSe2. Once identified, these parameters allow us to spatially image the two-dimensional strain field in an inhomogeneously strained sample.

References:

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Gate tuning of the ultrafast carrier dynamics in single-layer graphene

E.A.A. Pogna[1], O. Balci[2], M. Guizzardi[1], A. Tomadin[3], G. Soavi[2], M. Polini[3], A.C. Ferrari[2] and **G. Cerullo**[1]

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Abstract:

The ultrafast carrier dynamics in single-layer graphene (SLG) is of key importance for its applications to optoelectronic devices, such as photodetectors, saturable absorbers and modulators [1]. Absorption of a photon of energy $\hbar\omega$ in SLG promotes an electron from an energy -$\hbar\omega$/2 in the valence band to an energy $\hbar\omega$/2 in the conduction band. This strongly non-equilibrium distribution thermalizes by electron-electron scattering (τ≈10-20 fs) to a hot Fermi-Dirac (FD) distribution, which in turn equilibrates with the cold lattice via interaction with the strongly-coupled optical phonons (SCOPs, τ ≈200-300 fs) followed by defect-mediated scattering with acoustic phonons (τ≈1-2 ps).

In this work we study the doping level dependence of carrier relaxation dynamics in SLG. We perform femtosecond pump-probe spectroscopy on a large area CVD SLG sample (p-doped, E_F = -0.2 eV) gated with an ionic liquid, according to the configuration shown in Fig. 1(a). This device allows changing EF from -0.8 to +0.5 eV with a comparatively low gate voltage (V_g =± 1.2 eV). Figure 1(b) shows the differential transmission (ΔT/T) signal at zero pump-probe delay as a function of Vg, following excitation at ω_{pu} = 0.8 eV for various probe photon energies ω_{pr}. We observe dramatic changes of the amplitude, sign and dynamics of the signal with V_g. For V_g = 0, we measure for all probe photon energies a positive ΔT/T, due to Pauli blocking of the interband absorption. For negative values of V_g the sample becomes increasingly p-doped until, for $|E_F| > \hbar\omega_{pr}/2$, the signal becomes a photoinduced absorption (ΔT/T <0). This change in sign can be understood considering that, for such values of EF, the absorption of unpumped graphene is blocked as there are no available electrons in the valence band; the pump creates a hot FD distribution thus

making some valence band electrons available for absorption by the probe. The cooling dynamics also strongly depends on V_g, with the SCOP channel blocked when $|E_F|$ approaches $\hbar\omega_{pu}/2$. These results have important implications for the application of SLG to non-equilibrium optoelectronic devices.

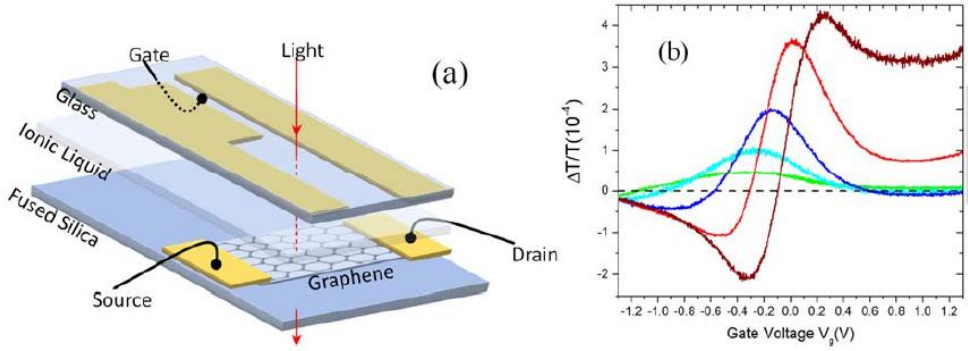

Figure 1: (a) scheme of the SLG device gated with ionic liquid; (b) ΔT/T dynamics at zero pump-probe delay following excitation at ω_{pu} = 0.8 eV, for probe photon energies ωpr = 0.75 eV (brown), 0.88 eV (red), 1 eV (blue), 1.13 eV (light blue) and 1.24 eV (green).

References:

[1] F. Bonaccorso et al., Nat. Photon. 4, 611–622 (2010).

Ultrafast diffusion and superdense ordering of lithium in bilayer graphene

J. Smet[1], M. Kühne[1], S. Fecher[1], F. Paolucci[1], J. Popovic[1], P.M. Ostrovsky[1], D. Samuelis[1], J. Maier[1], F. Börrnert[2], J. Biskupek[2], U. Kaiser[2], M. Ghorbani-Asl[3], A.V. Krasheninnikov[3]

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[2]Materialwissenschaftliche Elektronenmikroskopie, Universität Ulm, D-89081 Ulm, Germany
[3]Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden Rossendorf, D-01328 Dresden, Germany

Abstract:

Solids that simultaneously conduct electrons and ions are key elements for the mass transfer and storage required in battery electrodes. Single-phase materials with a high electronic and high ionic conductivity at room temperature are hard to come by, and therefore multiphase systems with separate ion and electron channels have been put forward instead. Here we report on bilayer graphene as a single-phase mixed conductor and demonstrate the diffusion of lithium faster than in graphite and even surpassing the diffusion of sodium chloride in liquid water [1]. To measure diffusion of lithium, we have developed an on-chip electro-chemical cell architecture in which the redox reaction, that forces lithium intercalation, is localized at a protrusion of the device only. This geometry offers a well-defined one-dimensional diffusion front so the 2D material can be operated as a true single-phase mixed conductor. It also leaves the bilayer unperturbed from the electrolyte during operation. Time-dependent Hall measurements across spatially displaced Hall probes enable to monitor the in-plane diffusion kinetics within the single van der Waals gap. The intercalation is reversible.

The device concept with a perimetrical galvanic cell is transferrable to other 2D materials and the immediate accessibility of the surface offers the possibility of deploying local probe and techniques to study the local kinetics and ordering of an intercalate otherwise hidden underneath the electrolyte. Here, we have chosen to perform transmission electron microscopy [2] even though probing light elements such as lithium ions and carbon atoms is severely hampered by their low scattering cross section for impinging electrons and their susceptibility to knock-on damage. Working at low acceleration voltage becomes crucial but demands the use of spherical and chromatic aberration correctors to maintain true atomic resolution. We achieve true atomic resolution and contrary to expectation, observe the formation of a high density, multi-layered crystalline phase of lithium in between the graphene sheets. The associated storage capacity exceeds by far the densest configuration realized in bulk graphitic carbon under similar conditions.

References

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[2] M. Kühne, F. Börrnert, S. Fecher, M. Ghorbani-Asl, J. Biskupek, D. Samuelis, A. Krasehninnikov, U. Kaiser, J.H. Smet, Reversible superdense ordering of lithium between two graphene sheets, submitted.

Discovering, Creating, and Exploring Novel Atomically-Thin Materials and Heterostructures

Joshua A. Robinson[1]

[1] Department of Materials Science & Engineering; The Center for 2D and Layered Materials; The Center for Atomically Thin Multifunctional Coatings; and The 2D Crystal Consortium; The Pennsylvania State University, University Park, PA 16802

Abstract:

The last decade has seen an exponential growth in the science and technology of two-dimensional materials. Beyond graphene, there is a huge variety of layered materials that range in properties from insulating to superconducting. Furthermore, heterogeneous stacking of 2D materials also allows for additional "dimensionality" for band structure engineering. In this talk, I will discuss recent breakthroughs in two-dimensional atomic layer synthesis and properties [1-7], including novel 2D heterostructures [1,3,4,5] and realization of unique 2D allotropes of 3D materials (e.g. 2D-GaN and Ga2O3) [6]. Our recent works demonstrate that the properties and doping of 2D materials, especially synthetic 2D materials, are extremely sensitive to the substrate choice. I will discuss substrate impact on 2D layer growth and properties, doping of 2D materials, selective area synthesis of 2D materials, and 2D nitrides beyond hBN. Our work and the work of our collaborators has led to a better understanding of how substrate not only impacts 2D crystal quality, but also doping efficiency in 2D materials, and stabilization of nitrides at their quantum limit.

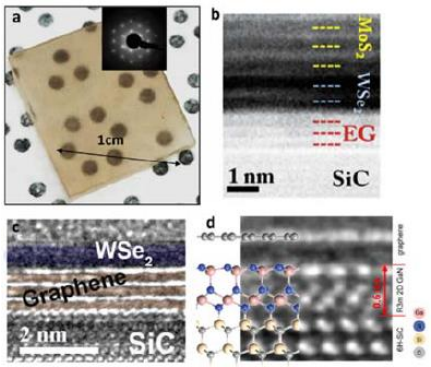

Fig.1: Examples of (a) wafer-scale WSe2, (b,c) 2D heterostructures, and (d) 2D-Gallium Nitride.

References:

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[3] Deconvoluting the Photonic and Electronic Response of 2D Materials: The Case of MoS2, Scientific Reports 7, Article number: 16938

[4] Properties of synthetic epitaxial graphene/molybdenum disulfide lateral heterostructures, Carbon (125), Pages 551-556

[5] Resonant Tunnel Diodes Built from Atomically Thin Materials; Nat. Commun. 6:7311, doi:10.1038/ncomms8311 (2015)

[6] Two-dimensional gallium nitride realized via graphene encapsulation; Nature Materials (1166–1171)

[7] Recent Advances in Two-Dimensional Materials Beyond Graphene; ACS Nano, 2015, 9 (12), pp 11509–11539

One Dimensional van der Waals Materials

Roger K. Lake [1], Bishwajit Debnath[1], Sinisa Coh[1], Tina T. Salguero [2], Alexander A. Balandin[1]

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Abstract:

While a massive world-wide effort has been directed at two-dimensional (2D) layered materials over the last 10 years, one-dimensional (1D) bundled materials have received considerably less attention. Similar to the 2D layered materials with covalently bonded layers separated by van der Waals (vdW) gaps, the 1D materials consist of covalently bonded one-dimensional wires with van der Waals gaps between the wires. Do to their lower dimensionality, these 1D van der Waals materials are more susceptible to instabilities such as charge density wave (CDW) transitions and metal-insulator transitions that can be exploited for switching or storage. The CDW phase also exhibits the sliding-CDW transition triggered by small electric fields. Data mining has identified 487 1D materials with bandgaps ranging from metallic to 6 eV [1]. Because of their single-crystalline structure and self-passivated side-walls, nanowires (NWs) of the 1D vdW crystal TaSe3 were demonstrated to have a more than an order of-magnitude higher breakdown current density than Cu [2]. Thus, the 1D vdW materials can provide both functional devices and interconnects. The same transition metal (M) chalcogenide (X) material systems that result in the MX2 2D-layered materials also form the MX3 1D-bundled materials. Similar to the 2D materials, the number of polymorphs can be large. An illustrative example is NbS3. It was previously known to crystallize in four forms: types I, II, III, and a high-pressure phase. Very recently, two new stable polymorphs of NbS3 designated as phases IV and phase V were demonstrated [3]. The positions of the Nb atoms established by X-ray diffraction of the phase IV polymorph show dimerization (a CDW) along the chain direction. In the phase V polymorph, the Nb atoms are equally spaced. Density functional theory calculations of these two new phases have been carried out by our group. At the PBE level of theory, the dimerized phase I and IV structures show an energy gap at the Fermi level of ~0.2 eV, whereas the phase V structure, that lacks dimerization, is metallic with bands crossing the Fermi level. The calculated energy-momentum relation of phases I, IV, and V are shown in Fig. 1 along with the TEM images showing the atomic spacings. This demonstrates how small changes in the arrangement of the atoms have a dramatic effect on the electronic properties.

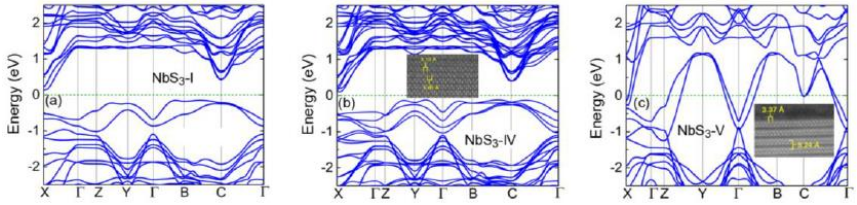

Fig. 1. Calculated energy momentum relations for NbS3 phases I (a), IV (b) and V (c). TEM images are inset in (b) and (c) that show the atomic distances [3]. The dimerization is apparent in NbS3-IV.

This talk will describe our theoretical work analysing the energetics, electronic structures, vibrational modes, and energy level alignments of a family of these metallic, semiconducting, and strongly correlated 1D materials.

Acknowledgements: This work was supported in part by the National Science Foundation under Award EFRI-1433395. This work used the Extreme Science and Engineering Discovery Environment (XSEDE) which is supported by National Science Foundation Grant No. ACI-1548562 and allocation ID TG-DMR130081.

References:

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Resonantly hybridized excitons in moiré superlattices in van der Waals heterostructures

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Abstract:

Atomically-thin layers of two-dimensional materials can be assembled in vertical stacks held together by relatively weak van der Waals forces, allowing for coupling between monolayer crystals with incommensurate lattices and arbitrary mutual rotation [1]. A profound consequence of using these new degrees of freedom is the emergence of an overarching periodicity in the local atomic registry of the constituent crystal structures, known as a moiré superlattice. Its presence in graphene/hexagonal boron nitride structures led to observation of the electronic minibands resulting in the Hofstadter butterfly spectra [2], whereas its effect enhanced by the interlayer resonant conditions in twisted graphene bilayers has been shown to produce van Hove singularities [3], recently culminating in the discovery of the intriguing superconductor-insulator transition at magic twist angles [4]. Here, we demonstrate that in semiconducting heterostructures built of incommensurate $MoSe_2$ and WS_2 monolayers excitonic bands can hybridize, which results in the resonant enhancement of the moiré superlattice effects. $MoSe_2$ and WS_2 are specifically chosen for the near degeneracy of their conduction band edges to promote the hybridization of intra- and interlayer excitons, which manifests in a pronounced exciton energy shift as a periodic function of the interlayer rotation angle. This occurs as hybridized excitons (hX) are formed by holes residing in $MoSe_2$ bound to a twist-dependent superposition of electron states in the adjacent monolayers. For heterostructures with almost aligned pairs of monolayer crystals, the resonant mixing of the electron states leads to amplified effects of the heterostructure's geometrical moiré pattern on the hX dispersion. Our findings pave the way to creation of the new states of matter, the topological excitons [5], and underpin novel strategies for band-structure engineering in semiconductor devices based on van der Waals heterostructures [6].

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On-chip strain engineering platform for two-dimensional materials

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Abstract:

The control of strain in two-dimensional materials opens exciting perspectives for the engineering of their electronic properties1,2. The investigation of custom strain profiles, however, requires a high design flexibility, which has been hardly achieved so far by available experimental techniques. Furthermore, practical implementations in actual devices need to be compatible with the employed electronic or photonic technologies and architectures. Here, we address a novel method to induce arbitrary strain profiles in graphene and other 2D materials that is scalable to nanometric dimensions, can be realized in any geometry directly on-chip in real devices, and even allows many strain erasing/redesign cycles. For the purpose we use polymeric micrometric artificial muscles (MAMs) that contract in a controllable and reversible way under an electronic stimulus. As a proof of concept here we exploit the local mechanical shrinkage of poly-methyl-methacrylate (PMMA) under high-dose electron-beam irradiation. The pulling geometry and strength are defined by electron-beam nano-patterning, which allows the implementation of a vast range of strain profiles (Fig. 1a). Moreover, the induced strain can be released by heating the sample and later restored by re-irradiating the polymer. Implementation of inhomogeneous uniaxial strain and out-of-plane deformations are demonstrated in graphene, and studied by Raman, scanning-electron and atomic-force microscopy (Fig.1b)3, as well as in WS2 where we observe the strain directly in the photoluminescence (Fig. 1c). The flexibility of our experimental approach opens new opportunities for the investigation of strain and nanomechanics in two-dimensional materials.

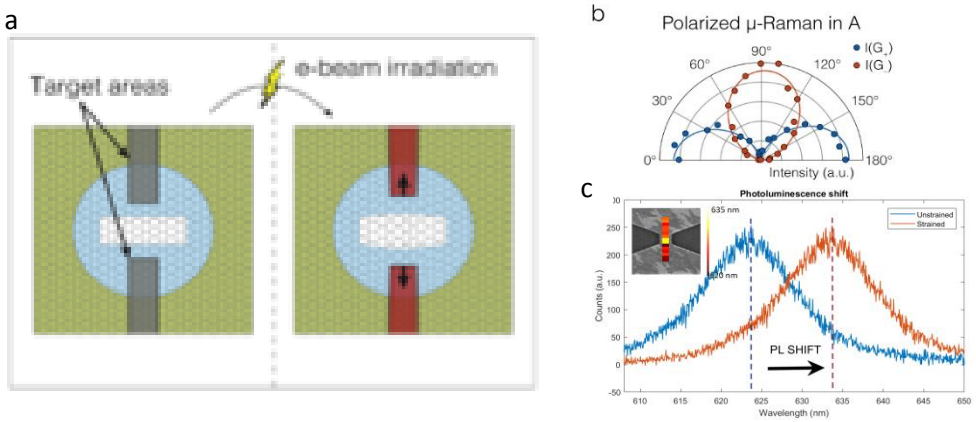

Fig. 1. (a) Example of device architecture involving two MAMs. Uniaxial strain in the ntral region is expected. (b) Local uniaxial stretching induced on graphene using two MAMs and demonstrated with polarized micro-Raman spectroscopy. (c) Photoluminescence spectrum and map (inset) of WS2 uniaxially strained in between two MAMs.

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Two-dimensional Materials for Vertical Heterostructures

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Abstract:

Vertical heterostructures have received significant attention recently due to their potential device application including memory select, resonant tunnelling devices, and steep-subthreshold transistors. Two-dimensional materials such as transition metal dichalcogenides (TMDs) offer several unique advantages: they are ideal quantum wells with no additional sub-bands; the lack of covalent bonding between layers allows for stacking of monolayers with a wider variety of properties and can potentially lead to monolayer thickness control and improved reproducibility; and devices fabricated with two-dimensional materials can potentially be synthesized on or transferred to arbitrary substrates. In this work, the opportunities and challenges associated with two-dimensional material heterostructure devices are described using simulation and experiment.

The current-voltage characteristics of TMD heterostructures have been simulated using an energy-space formulation of the Bardeen Transfer Hamiltonian [1-3]. Like-band tunnelling (conduction to conduction, valence to valence) will be shown to potentially result in negative differential resistance with theoretical peak-to-valley ratio exceeding that of conventional III-V heterostructures. Opposite band tunnelling (valence to conduction) will be shown to potentially result in steep-subthreshold behaviour. The impact of materials properties, band structure and non-idealities such as defects will be shown. Even for the case of highly defective TMDs, the results show that Schottky barrier limited injection can result in current-voltage characteristics suitable for memory select applications.

While synthesis methods have been developed for large-area uniform TMDs (MoS2, WSe2) [4-5], many of these techniques require synthesis temperatures of 800°C or higher to enable low defect density. As a result of this thermal budget, direct synthesis on substrates of interest (e.g. the interconnect levels of a CMOS process) is not possible. Furthermore, intermixing of synthesized heterostructures can occur at these temperatures. Low-temperature, plasma assisted, cyclic synthesis of TMDs will be described including the impact of key process parameters on resulting structure and the characterization of associated devices [6-7].

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Contact Interfaces and Transition Metal Dichalcogenides

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Abstract:

Contacts to transition metal dichalcogenides (TMDs) present several challenges in the context of realizing optimum device performance. [1-3] In the case of traditional metal contacts, the TMD surface preparation method and the resultant condition plays a prominent role. [4] Recently, the impact of the deposition ambient on contact resistance [5,6] and the associated metal/TMD interfacial chemistry has been explored. [7-10] It is found that deposition under conditions where the residual gas pressure is controlled to minimize spurious surface reactions (viz. ultrahigh vacuum (UHV): Pdep ≤ 10-8 mbar) can result in a lower contact resistance for some metal/TMD systems. Comparing in-situ and ex-situ deposition in conjunction with surface/interface analysis of such interfaces reveals substantial differences in the interfacial chemical properties relative to contacts deposited under HV (Pdep ≤ 10-6 mbar) conditions more typical in device fabrication. These properties can be correlated to the interfacial band alignment and associated barriers. We will present our studies of the surface preparation and interfacial analysis of contact metals with WSe2 where substantial differences in the interfacial chemistry are noted depending upon the deposition vacuum conditions and subsequent anneals (vacuum vs. forming gas), resulting in improved contact resistance and transport.

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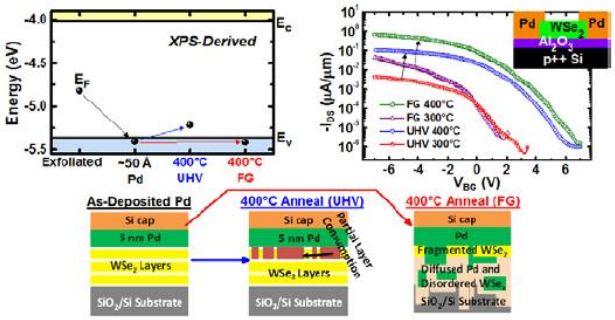

Fig. 1. Impact of post-metal anneals on Fermi level control, transport and the associated interfaces for Pd/WSe2 ohmic hole contacts. After [11].

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1D van der Waals Nanomaterials: Selenium and Tellurium

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Abstract:

Selenium and Tellurium are two special elemental materials which are 1D helical atomic structures and formed by van der Waals force between helical atomic chains. In this talk, we will report on the fundamental studies of these two new nano-materials at atomic scale in terms of their electrical, optical, thermal and mechanical properties. [1,2] The helical atomic structure offers strong anisotropic properties of these 1D van der Waals materials. The band-structures of the materials themselves also offer some excellent materials properties such as highest Seebeck coefficient for selenium and high carrier mobility of 700 cm_2/Vs for tellurium with demonstrated field-effect transistor drain current exceeding 1 A/mm. [3] The work is in close collaborations with Prof. Wenzhuo Wu at Purdue University.

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Wafer-scale Fabrication of MoS2 and PtSe2 MOSFETs

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Abstract:

To date, most 2D devices were individually crafted by e-beam lithography on flakes exfoliated from bulk crystals, making them unsuitable for practical applications. To overcome the challenge, the back-end-of-line (BEOL) process of a state-of-the-art BiCMOS foundry was used to batch-process thousands of individually probable RF MOSFETs on each chip of a 200-mm Si wafer. The BEOL process allowed not only submicron gate to be buried flat in the field oxide, but also high-quality gate oxide to be deposited before it is coated with MoS2 or PtSe2. The wafer-scale fabrication resulted in uniform device characteristics with nearly 100% yield.

The wafer-scale fabrication started with the transfer of a 25 mm × 15 mm sheet of monolayer MoS2 grown by chemical vapor deposition (CVD) on SiO2-coated Si at 760 °C [1]. Large-scale study of thousands of the resulting MOSFETs showed that the yield was mainly impacted by the poor adhesion of the transferred MoS2 during wet etching of the Al2O3 gate oxide. This shows the limitation of transferred 2D materials, whether tiny flakes or large sheets.

Since MoS2 could not be grown below 450 °C, the thermal budget of typical BEOL processes, spin coating of liquid-exfoliated MoS2 was subsequently used in wafer-scale fabrication [2]. This resulted in nearly 100% yield for the MOSFETs, and their cut-off frequencies were found to be on the order of 100 MHz for the first time for liquid-exfoliated MoS2. This shows the potential of liquid-exfoliated MoS2, despite being inferior to bulk or CVD MoS2.

To further improve the MOSFET performance, thinner channel should be used. However, spin-coated MoS2 cannot be thinner than 10 nm without becoming discontinuous. Therefore, after spin coating, the channel between the source and drain contacts was selectively etched [3]. The channel recess was found to improve the current on/off ratio by three orders of magnitude without impacting the contact resistance. However, the contact resistance, on the order of 1000 Ω·μm, was still too high for practical applications, which is a common problem for most 2D materials.

To further reduce the contact resistance, channel recess was used on PtSe2 directly selenized at 400 °C from sputtered Pt [4]. Taking advantage of thickness-dependent transition, PtSe2 was kept thick (semimetallic) under the source and drain but made thin (semiconducting) under the gate. This resulted in less than 100 Ω·μm contact resistance with shorter than 0.1 μm transfer length, without impacting the on/off ratio. Note that when the gate length (critical dimension) of a MOSFET is scaled down, its channel thickness, contact resistance, and contact length need to be scaled as well. The wafer-scale fabrication resulted in uniform device characteristics, so that average vs. best results were reported, as well as RF vs. DC characteristics. For example, for MOSFETs fabricated on 12-nm-thick PtSe2, the contact resistance was 80 ± 3 Ω·μm, the forward-current cut-off frequency was 42 ± 5 MHz, and the maximum frequency of oscillation was 180 ± 30 MHz. These results confirmed the application potential of PtSe2 for future generation thin-film transistors.

The above examples proved the feasibility of 2D materials for wafer-scale fabrication, so that they can be used in thin-film transistors in the near term and ultra-thin-body high-speed transistors in the long run.

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Infrared and THz optics of atomically thin films of transition-metal dichalcogenides and post-transition metal chalcogenides

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Abstract:

Transition-metal dichalcogenides (TMD), MoS2, MoSe2, WS2 and WSe2 and post-transition metal chalcogenides (PTMC), InSe and GaSe are materials where a strong covalent bonding of atoms inside individual layers coexists with a weak van der Waals coupling of the consecutive layer of the bulk crystal. Such peculiar bonding makes it possible to isolate mechanically, or to grow epitaxially atomically thin films of such compounds with a precise number of atomic layers. At the same time, van der Waals nature of these crystals coexists with strong hybridizations of – separately – conduction and valence band orbitals in the consecutive monolayers in the film, making the band structure of few-layer atomically thin TMDs and PTMCs sensitive to the number of layers in them [1,2]. In particular, few-layer films acquire multiple subbands in their electronic spectra, with a strong coupling of inter-subband transitions of carriers (electrons in n-doped and holes in p-doped materials) with out-of-plane polarised photons. Here, we show that, when n- or p-doped, few-layer films of TMDs and PTMCs become absorbers and emitters of infrared (IR) and THz light [1,2]. Our density functional theory modelling and a specially designed hybrid k·p theory for the monolayers of these materials, combined with the tight-binding model description of the interlayer hopping (HkpTB), predicts that optical activity of few-layer films of these two classes of compounds densely covers the range from IR (1.5 micron) for bilayer films to THz for the films with 3-10 layers. The predicted spectral characteristics are confirmed [3] by SNOM studies of atomically thin films of WSe2. In a way, these thin films are analogous to quantum wells in conventional semiconductors, and, by choosing the number of layers, and/or n- or p-doping in one of TMD and PTMC compounds, one can tune such inter-subband transition energy to the desirable application range, offering a new way how 2D materials can be harnessed for developing new technologies.

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