Graphene Canada

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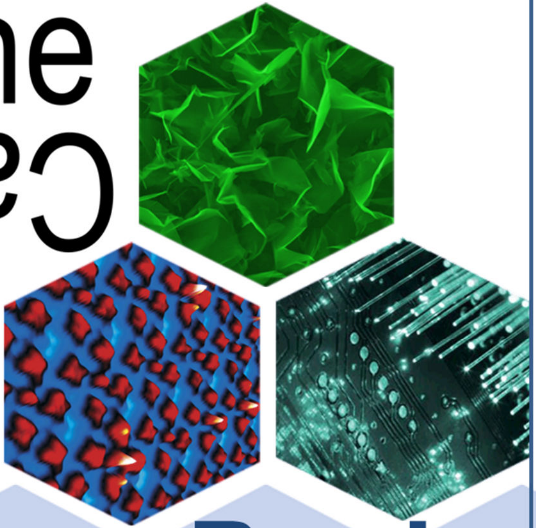

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Foreword

On behalf of the Organising Committee we take great pleasure in welcoming you to Montreal for the second edition of the Graphene Canada International Conference & Exhibition.

A plenary session with internationally renowned speakers, an industrial forum, extensive thematic workshops in parallel and a significant exhibition featuring current and future Graphene developments will be highlighted at the event.

Graphene Canada 2016 will bring together, from a global perspective, scientists, researchers, end-users, industry, policy makers and investors in an environment of cooperation and sharing towards the challenges of Graphene commercialization.

We are indebted to the following Institutions for their help and/or financial support:
Centre National de la Recherche Scientifique/CEMES, GDRI: Graphene-Nanotubes, Grafoid Inc., IIT Graphene Labs, McGill University, Nanoscale Horizons, Phantoms Foundation, Texas Instruments, The Catalan Institute of Nanoscience and Nanotechnology (ICN2), Tourisme Montréal, Université Catholique de Louvain and Université de Montreal.

We would also like to thank the following companies for their participation as exhibitors: Abalonyx AS, Go Foundation, Grafoid Inc., Group Nanoxplore Inc., PLASMIONIQUE Inc, Raymor/NanoIntegris, and SURAGUS GmbH.

We also would like to thank all the speakers and attendees that joined us this year.

We truly hope that Graphene Canada 2016 serves as a platform for communication between science and business.

Hope to see you again in the next edition of Graphene Canada to be held in 2017.

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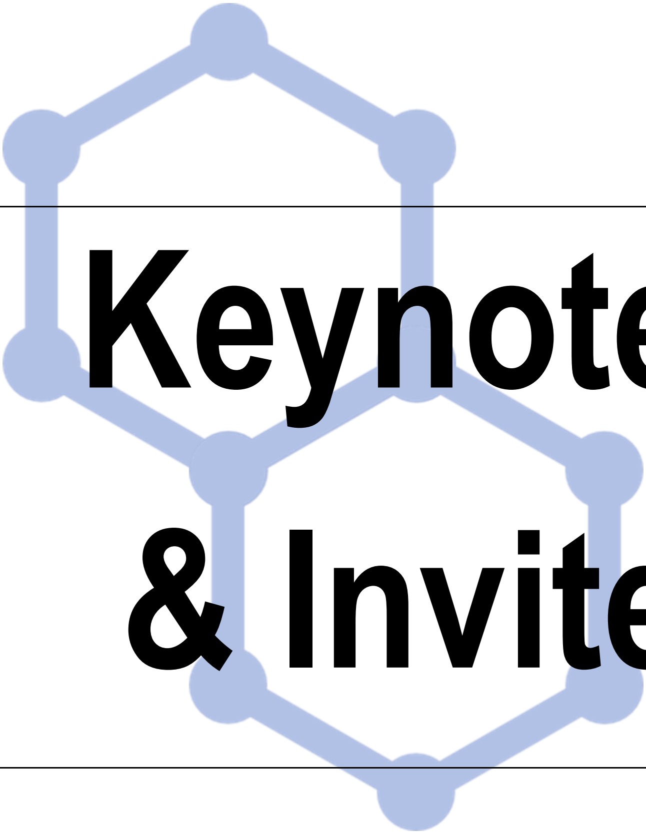

Keynotes & Invited

Terrance Barkan

The Graphene Council
USA

2016 Global Graphene Survey Results - What is the current and future state of the graphene industry?

More than 400 graphene experts world-wide were asked about the current and future state of the global graphene market. Join this session to learn industry insights from the global survey as well as industry intelligence reports and an update on the progress towards graphene standards from The Graphene Council, the worlds largest community for graphene professionals.

We explore which industries, products and applications are most likely to be impacted by graphene development and which of these are the most likely candidates for successful commercialization. We also look at what graphene industry experts expect from national governments regarding support for the commercialization of this amazing material.

This presentation will also provide a short update on the efforts to develop a global standard for graphene by the international standards bodies like ISO / ANSI/ IEC and others.

Francesco Bonaccorso

Istituto Italiano di Tecnologia, Graphene Labs,
Via Morego 30, 16163 Genova, Italy

francesco.bonaccorso@iit.it

Large scale production of 2D-crystals-based inks and their application for energy and (opto)electronics

Graphene and other two dimensional (2D) crystals thanks to their excellent and complementary material properties, have the opportunity to improve the performance of existing devices or enable new ones [1-6] that are also environmentally friendly [7]. Moreover, the possibility to assembly such 2D crystals in vertical heterostructures will provide a rich toolset for the creation of new materials [1,2,8], enabling the realisation of next-generation devices.

However, one of the key requirement for applications such as flexible (opto)electronics and energy storage and conversion is the development of industrially scalable production processes [2,8], which provides a balance between ease of fabrication and final material quality with on-demand tuneable properties.

Liquid-phase exfoliation [2,8,9] is offering a simple and cost-effective pathway to fabricate various 2D crystal-based (opto)electronic and energy devices, presenting huge integration flexibility compared to conventional methods. Here, I will show our scaling up approach for the solution processing of graphene and other 2D crystal based on wet-jet milling of layered materials. Moreover, I will present an overview of graphene and other 2D crystals for flexible and printed (opto)electronic [11] and energy [12-15] applications, throughout the fabrication of large area electrodes [12] to devices integration [10-15].

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V. Bouchiat

Z Han, F. Veliev, A. Allain, A. Bourrier, H. Arjmandi-Tash, A. Briançon, C. Delacour

Département Nanosciences, Institut Néel, CNRS-Grenoble, France

vincent.bouchiat@neel.cnrs.fr

Active hybrid devices based on physisorbed elements on Graphene: from tunable superconducting transitions to neural network bio-applications

Graphene provides an ideal 2D gas of Dirac Fermions, which is directly exposed to the environment. Therefore it provides an ideal platform on which to tune, via application of an electrostatic gate, the electronic coupling between ordered adsorbates deposited on its surface via the control of density and sign of charge carriers. This situation is particularly interesting when the network of adsorbates are actively interacting between each other and can induce some long range electronic order within the underlying graphene substrate, such as superconducting correlations [1]. To demonstrate this concept, we have measured arrays of superconducting clusters physisorbed on Graphene capable to induce via the proximity effect a gate-tunable superconducting transition. We have experimentally studied the case of macroscopic graphene decorated with an array of superconducting tin clusters [2], which induce via percolation of proximity effect a global but tunable 2D superconducting state. By adjusting the graphene disorder and its charge carrier density on one side , the geometrical order, cluster size and density of the superconducting dot network on the other side, the superconducting state can exhibit very different behaviors, allowing to test different regimes and quantum phase transition from a granular superconductor to either metallic or insulating states, leading to a bosonic-type gate-controlled quantum phase transition [3]. I will show recent experimental results involving three sets of triangular arrays sparsely distributed on graphene, in which superconductivity is destroyed for a critical gate value that we attribute to the effect of quantum fluctuations of the phase giving rise to an intermediate metallic state [4].

Finally I will give another example of organized elements on graphene involving the interaction of in-situ grown neurons onto graphene [5], showing that electrical activity of neural cells can be probed with sub-cell accuracy.

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Figures

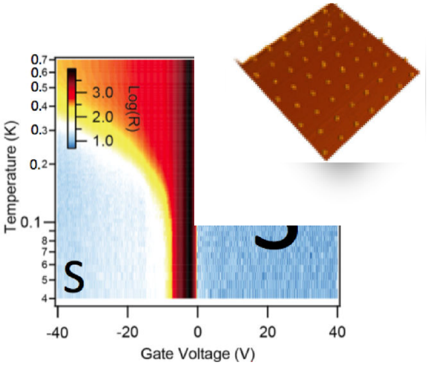

Figure 1: Low temperature phase diagram of superconducting transition triangular array of Tin dots decorating graphene inset AFM micrograph of the corresponding sample (scan size 10 microns).

Jean-Christophe Charlier

University of Louvain, Institute of Condensed Matter and Nanosciences, Belgium

jean-christophe.charlier@uclouvain.be

Ab initio modeling of novel 2D materials

Borophene, a recently synthesized two-dimensional monolayer of boron atoms, is expected to exhibit anisotropic metallic character with relatively high electronic velocities [1]. At the same time, very low optical conductivities in the infrared-visible light region have been reported. Based on its promising electronic transport properties and a priori high transparency, borophene could become a genuine LEGO piece in the 2D materials assembling game. Such early suggested properties demands for an in depth investigation of borophene electronic structure. Moreover, borophene is naturally degraded in ambient conditions and it is therefore important to assess the mechanisms and the effects of oxidation on borophene layers. Optical and electronic properties of pristine and oxidized borophene have been investigated using first-principles techniques [2]. Optical response of the oxidized layer is found to be strongly modified suggesting that optical measurements can serve as an efficient probe for borophene surface contamination.

Two-dimensional conjugated polymers exhibit electronic structures analogous to that of graphene with the peculiarity of π–π^* bands which are fully symmetric and isolated. Realistic 2D conjugated polymer networks with a structural disorder such as monomer vacancies (unavoidable during bottom-up synthesis) are investigated using both *ab initio* and *tight-binding* techniques [3] in order to check their suitability for electronic applications. As expected, long mean free paths and high mobilities are predicted for low defect densities. At low temperatures and for high defect densities, strong localization phenomena originating from quantum interferences of multiple scattering paths are observed in the close vicinity of the Dirac energy region while the absence of localization effects is predicted away from this region suggesting a sharp mobility transition [3].

Electronic structure and transport properties of N-doped graphene with a single sublattice preference are investigated using both first-principles techniques and a real-space Kubo-Greenwood approach [4]. Such a breaking of the sublattice symmetry leads to the appearance of a true band gap in graphene electronic spectrum even for a random distribution of the N dopants. In addition, a natural spatial separation of both types of charge carriers at the band edge is observed, leading to a highly asymmetric electronic transport. For such N-doped graphene systems, the carrier at the conduction band edge present outstanding transport properties including long mean free paths, high mobilities and conductivities. Such a transport behavior can be explained by a non-diffusive regime (quasiballistic transport behavior at the conduction band edge), and originates from a low scattering rate. The presence of a true band gap along with the persistence of carriers traveling in an unperturbed sublattice suggest the use of such N-doped graphene in G-FET applications, where a high I_{ON}/I_{OFF} ratio is expected. The present ab initio simulations should encourage more investigation and specific transport measurements on N-doped graphene samples where such an unbalanced sublattice doping is observed.

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Karl Coleman

Founder & Chief Scientific Officer, Applied Graphene Materials plc, UK
Professor, Department of Chemistry, University of Durham, UK

k.s.coleman@durham.ac.uk

Graphene Production and the Importance of Dispersion in Graphene Applications

Graphene has generated much interest owing to its high surface area and exceptional electrical, thermal and mechanical properties. Here I will discuss the synthesis of graphene using a simple, convenient and scalable 'bottom-up' process. I will talk about how Applied Graphene Materials works in partnership with its customers to provide dispersion and product integration expertise, to deliver solutions for a wide range of applications. During the talk I will highlight the importance of graphene dispersions in delivering the true commercial value of graphene which lies in the ability to transfer its intrinsic properties into other materials.

Applied Graphene Materials works in partnership with its customers across many different sectors with the know-how to deliver advanced material solutions for many applications, including: advanced composites and polymers; coatings; functional fluids including oils and lubricants; supercapacitors and batteries; thermal management solutions; barriers and impermeable films; display materials and packaging; and inks and 3D printed materials. I will discuss some recent case studies where graphene has been shown to offer significant benefits in the application area targeted.

Links:
Prof. Karl Coleman Home Page
www.dur.ac.uk/chemistry/staff/profile/?id=1329
Applied Graphene Materials
www.appliedgraphenematerials.com

Antonio Correia

Phantoms Foundation,
Calle Alfonso Gomez 17, 28037 Madrid, Spain

antonio@phantomsnet.net

“Graphene and 2D Materials” EUREKA Cluster: Fostering European Competitiveness

The “Graphene & 2D Materials” EUREKA cluster is defined as a complementary enabling and accelerator instrument in the European scene, fully piloted by industries to further take graphene from the mature research developed at academic laboratories into the European society in the space of 5 years, boosting economic growth, jobs creation and international leadership and investment attractiveness. This cluster will help Europe having a more dominant position in graphene patenting, will deploy the proper winning industrial strategies to gain worldwide competitiveness, and will ensure that for all promising industrial sectors of technology innovation, a fully integrated EU-value chain is established, integrating into consortia the relevant actors from low to high Technology Readiness Levels (TRL). The cluster will clarify the differentiating potential in all sectors where EU-industries is strong and could further gain in competitiveness, and will develop proper incentives towards the achievement of EU-leadership in the fields of graphene commercialization and graphene-driven technology improvement. The cluster will elaborate and foster industrially-driven innovation strategies, that will take advantage of the existing excellent science and transnational platforms in Europe (national networks, Graphene-Flagship, etc.), and will focus on solving current challenges which are limiting the time to market and business growth of graphene-related EU companies.

Gui-Ping Dai[1,2,3]
K.Vinodgopal[1], Marvin H. Wu[1]

[1]Department of Chemistry, North Carolina Central University, Durham, USA
[2]Institute for Advanced Study, Nanchang University, Nanchang 330031, China
[3]Chaowei Power Ltd., Changxing 313100, China

Triangle-Shaped Graphene Domains by LP-CVD and Update of Graphene Application in Motive Power Battery

The initial nucleation and growth dynamics of graphene play a critical role in determining the final quality of CVD graphene films, and it is highly desirable to synthesize large area high quality single crystalline graphene films [1, 2]. The shape and structure of individual graphene domains greatly influences its property and directly determines the grain boundary in polycrystalline graphene films. Thus, the tailoring and direct observation of the domain shape structure are very important for understanding the growth mechanism as well as to maximize single-crystalline graphene's inherent outstanding properties for future applications. I will report the growth of “triangle” graphene domains on Cu foils during the early stage of CVD operated under low pressure.

With the development of scientific and technological innovation over the past decades, graphene based products is growing rapidly around the world. Today lead-acid batteries are the most widely used rechargeable systems and still share about 65% of the rechargeable batteries market. Although lead-acid systems were investigated and developed over 150 years, research continues to enhance their performance in terms of rate capability, stability, cycle life and durability [3]. Graphene is envisaged to enhance the performance of these batteries as the most efficient materials in terms of high intrinsic electrical conductivity, extremely lightweight, chemical inert, and flexible with a large surface area. The presence of graphene in the electrodes improves the electrical conductivity between the active mass particles through preventing thickening and the growth of large $PbSO_4$ particles [4]. This improvement is naturally attributed to the formation of a stable conductive active mass matrix that enables the delivery and distribution of current to all the active material homogeneously. By enabling a uniform current distribution, and subsequently well distributed electrochemical redox reactions throughout the electrode matrix, arrested the formation of too large $PbSO_4$ particles. The addition of graphene is supposed to improve both the mechanical stability and electrical integrity of the electrodes and to induce uniform changes in the active mass during the complicated conversion reactions during cycling. As the largest producer of motive power battery in the world, Chaowei has a potential need of graphene with 1000 tons/year. In this talk, I will present latest research of graphene commercially application in motive power battery, especially E-bike market.

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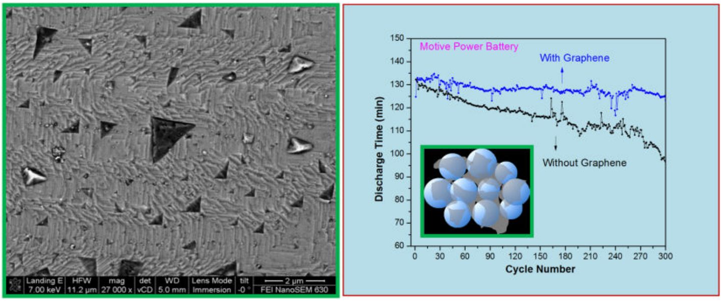

Figure 1: Typical SEM image of "triangle" graphene domains (Left) and cycle life of battery (Right).

Marc M. Dignam
Riley McGouran
Department of Physics, Engineering Physics and Astronomy Queen's University, Kingston, ON, Canada

Third Harmonic Terahertz Generation in Monolayer and Bilayer Graphene

It has been predicted that due to the linear dispersion of monolayer graphene near the Dirac points, there should be strong third harmonic generation in doped graphene at terahertz (THz) frequencies [1]. Despite this prediction, the only experimental evidence of third harmonic generation at THz frequencies has been in a 45 layer sample [2]. Our recent theoretical work has predicted that there is a different and potentially stronger way to generate the third harmonic in monolayer graphene, which relies on the strong interplay between interband and intraband carrier dynamics at THz frequencies [3]. This interplay arises due to the zero-bandgap, high mobility, controllable Fermi energy and strong intra- and interband absorption. As all of these are also features in bilayer graphene, it should also exhibit a strong third harmonic.

In this talk, I will review our formalism and present the results of our calculations of third harmonic generation in undoped monolayer and bilayer graphene as a function of field strength and THz frequency. Our model employs nonperturbative dynamic equations for the electron density matrix within the length gauge, with the inclusion of both interband and intraband carrier dynamics. We find that the emitted third harmonic field in undoped, unbiased, suspended bilayer graphene at low temperature is 53% of the reflected fundamental field for an incident single-cycle pulse at 2 THz with an amplitude of 2.5 kV/cm. Surprisingly, this is slightly larger than obtained for monolayer graphene [3]. I will finally examine third harmonic generation in *biased* bilayer graphene, for biases such that the bandgap opened is on the order of the THz photon energy.

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Ahmed Elmarakbi

Automotive Composites, University of Sunderland, UK

Hierarchical Graphene-based Polymer Composites for Automotive Applications: Challenges in the Way Forward

The automotive industry is widely viewed as being the industry in which the greatest volume of advanced composite materials will be used in the future to produce light vehicles. Nowadays, several advanced materials are widely used in automotive industry. Because of its multifunctional properties and promising applications, many expectations in composite materials are related to Graphene. However, no application of graphene-based materials is currently marketed in the automotive sector. Therefore, research activities are under development for study the potentiality of these systems and all the value's chain of automotive needs to be involved in this effort. One of most challenge aim is the economic impact of the innovative structures on the vehicle market, all the value's chain have to address their effort to get as low as possible the final cost of the innovative products.

The present initiative provides a summary overview on Graphene Related materials (GRM) for automotive applications and investigates efficient ways to integrate Graphene as polymer reinforcements within composite materials for energy-efficient and safe vehicles (EESVs). An approach that starts from the nano-scale through the Graphene elaboration by experiments to meso/macro-scale by continuum mechanics modelling is discussed with respect to some limiting factors in terms of the large scale production, the interfacial behaviour, the amount of wrinkling and network structure. Finally, a strategy for modelling such a composite is elaborated in the framework of the Graphene Flagship to well understand such limitations for a full applicability of Graphene. It is anticipated that this initiative will advance innovative lightweight graphene composites and their related modelling, designing, manufacturing, and joining capabilities suitable for automotive industry which requires unique levels of affordability, mechanical performance, green environmental impact and energy efficiency. This leads to complete understanding of the new graphene composites and their applicability in high-volume production scenarios.

Keywords: Automotive applications, Composite modelling and design, Energy efficient and safe vehicles, Graphene, Graphene composites

Giovanni Fanchini

Physics & Astronomy, University of Western Ontario, 1151 Richmond St, London
ON N6A 3K7, Canada

gfanchin@uwo.ca

Graphene thin films and their interaction with metallic nanoparticles: Materials, characterization and applications

In this presentation, we will review our recent work on large-area graphene thin films, collections of single-layer, few-layer, and multilayer graphene platelets assembled on a variety of substrates by cost-effective and widely scalable solution-processing methods. We will focus on the fact that their excellent optical electronic and thermal properties can be further enhanced through deposition of metallic nanoparticles (Me-NP) on their surfaces. Me-NP deposition on graphene can be achieved by a variety of methods, including thermal evaporation, sputtering, or organometallic precursors [1-3]. Metals strongly interacting with graphene form chemical bonds and carbides, but weakly interacting metals result in doping, without substantially altering the electronic band structure of graphene [4].

One important motivation to investigate graphene decorated with Me-NPs is to achieve precise doping of this material at the nanoscale, which is critical for several applications. Although large metallic thin films on graphene are known to move the Fermi level above or below the Dirac point [4], little has been done to study these effects when large-area graphene thin films are contacted with particles of nanometer diameters at varying concentrations, and to investigate the nature of such phenomena. We will present in detail some recent results [1] on the local effect of copper nanoparticles (Cu-NPs) on the Fermi energy of graphene domains in large-area graphene thin films, with theoretical calculations corroborating our Kelvin-probe force microscopy experiments to demonstrate that the Fermi level shifts in the presence of Cu-NPs, which breaks the electron-hole symmetry due to weak Van der Waals interactions between the graphene backbone and Cu, even in the absence of chemical bonding and charge transfer. This is in contrast to previous predictions for large and flat metallic contacts [1].

In the last part of our presentation we will show how deposition parameters control the nucleation of Cu-NPs on graphene thin films, in a way that, under specific growth situations close to thermodynamic equilibrium, periodic superlattices of Cu-NPs on large-area graphene thin films can be obtained.Theoretical calculations suggest a lowest formation energy for Cu-nanoparticle arrays aligning along armchair crystallographic directions, indicating that their self-assembly is energetically more favorable. Applications of periodic superlattices of Cu-NPs on graphene will be discussed in conjunction with scanning near field optical measurements. Their behavior as evanescent waveguides will be demonstrated [2].

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Tobin Filleter

Department of Mechanical & Industrial Engineering, University of Toronto
5 King's College Rd, Toronto, ON, M5S 3G8, Canada
filleter@mie.utoronto.ca
www.mie.utoronto.ca/labs/nmml/index.html

Mechanical Behavior of Graphene Oxide

Graphene oxide (GO) is a 2D nanostructure used as a building block for a class of materials with hierarchical features that possess vastly differing characteristic dimensions and properties. GO has been applied as a good candidate for paper-like materials, robust fibers, membranes, and reinforcement elements in composites. The use of GO in these materials has been motivated in large part by the exceptional mechanical properties of individual graphene and GO monolayers. Macroscopic papers and fibers synthesized using GO, however, typically exhibit strengths on the order of 10-100's of MPa's. This talk will explore recent studies on the mechanical behavior of GO based materials across multiple length scales that yield insights into the size-scale mechanical phenomenon exhibited by GO materials. This will include the strength and fracture of behavior of GO monolayer building blocks [1] and intermediate multiple layered GO nanosheets [2], as well as the interfacial shear properties of GO multilayers [3].

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F. Fossard[1]
E. Gaufres[1], A. Favron[2], N.Tang[3], V. Gosselin[2], M. Côté[2], R. Martel[3] and A. Loiseau[1]

[1]Laboratoire d'Etude des Microstructures, UMR 104 CNRS-Onera, Châtillon, France
[2]Regroupement Québécois sur les Matériaux de Pointe (RQMP) and Département de physique, Université de Montréal, Montréal, Canada
[3]RQMP and Département de chimie, Université de Montréal, Montréal, Canada

Towards 2D and 1D heterostructures for optoelectronics

The field of two-dimensional materials has been gradually enriched by new structures such as hexagonal Boron Nitride (hBN) dichalcogenides (e.g MoS_2), Black Phosphorus… These nearly zero thickness crystals exhibit fundamental properties that, when strongly interacting in layered heterostructures, can offer new paradigms for photonics, electronics or magnetism.

In the first part of this talk, we focus on Black phosphorus and present some spectroscopic insights of exfoliated layers of this material [1] that highlight the potential of P(black) based Van der Waals heterostructures. First we discuss the quantum oxidation mechanism responsible for the high reactivity of P(black) on air based on combined Raman and TEM-EELS (Electron Energy Loss Spectroscopy) measurements and how this mechanism can be used for preparing suitable samples. Then we show that Electron Energy Loss Spectroscopy (EELS) in the 1eV-50eV range, coupled with *ab initio* calculations, provides a unique approach to the anisotropic dielectric response of P(Black) at the nanoscale. Indeed, thanks to its angular resolution, our EELS set-up implemented in a TEM –STEM machine is capable to probe excitonic effects, optical transitions and plasmons dispersions as a function of the q momentum, for selected crystallographic orientations in the Brillouin zone of different symmetries. Finally we apply this spectroscopy on other 2D materials involved in heterostructures such as hBN and MoS_2.

The quantum confinement phenomena and interlayer effects that are now scrutinized in 2D materials and their heterostructure also fully make sense for 1D hybrids based on SWCNT or BNNT. As an illustration of this consistency, we will show in the second part of the talk, that the control of the 1D aggregation of active molecules inside nanotube, observed by Raman and EELS hyperspectral imaging [2], enables the observation of original and specific optical properties [3]. These confinement effects represent a great interest in various applicative fields such as multispectral bio-detection, multiplexing and super-dyes.

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Ray Gibbs

Chief Executive officer
Haydale Graphene Industries Plc
Great Britain

Establishing near term commercial applications for Graphene

The potential for graphene and other nano particles to significantly enhance material properties has been well documented. However, as carbon is inert and does not mix well with other materials it needs to be homogeneously dispersed and bonded into the host material in order to realise its potential. There are hundreds of "graphene's" in the market today and all display different characteristics. Knowing which one works best and applying a sympathetic surface treatment for a specific application is the Haydale expertise. This functionalisation treatment plus the years of processing know how sets Haydale apart in the ability to commercialise nano materials.

For industry to adopt these new materials we need to demonstrate a repeatable and cost effective functionalisation process that is scalable and can be tailored to the specific requirement. One of the challenges industry faces is that there is currently no standard definition of graphene which means that R&D and general procurement departments may not know exactly what it is they are buying nor be able to replicate test results with a subsequent batch of material. Industry requires a standardisation of materials to remove this uncertainty.

Further, in order to persuade industry to adopt the new materials we need to be able to incorporate the functionalised materials into existing production facilities thus avoiding the need for replacement of existing capital equipment. Ray Gibbs will examine these challenges and explain how Haydale has established a consistent supply chain of the nanomaterials and are addressing the commercial adoption challenges through the establishment of Centres of Excellence in strategic locations across the globe.

Pedro Gómez-Romero
Deepak P. Dubal
Catalan Institute of Nanoscience and Nanotechnology, ICN2 (CSIC-BIST),
Campus UAB, 08193 Bellaterra (Barcelona), Spain
pedro.gomez@icn2.cat

Graphene Electroactive Nanofluids for Energy Storage

Energy storage has never been so thriving as it is today, boosted by the need of a new and sustainable energy model. At the same time, graphene is witnessing an explosive growth both in fundamental research and applied developments, among which, energy storage is one of the most frequently considered for this unique material.

In our research group (NEO-Energy) we have developed several lines of work concerning graphene for energy storage. In particular, we have developed supercapacitors based on graphene and on their hybrid materials with polyoxometalates[1-2] and other molecular species as well as with extended phases.[3]

But, in addition to conventional solid-electrode batteries and supercapacitors, we have recently developed an alternative way to store electrical energy with solid electroactive species dispersed in electrolyte liquids. These electroactive nanofluids[4-5] can be used in flow cells broadening the potential applications of conventional Redox Flow Batteries.

In this conference we will present some of our most recent results along this line, in particular how graphene can be put to work for energy storage in a new way by making use of graphene nanofluids, which show a remarkably fast energy storage capability.[6]

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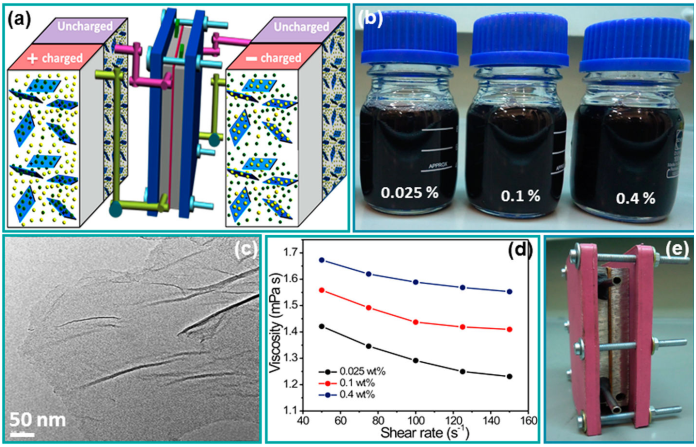
(a)
Uncharged
+ charged
Uncharged
− charged
(b)
0.025 %
0.1 %
0.4 %
(c)
50 nm
(d)
Viscosity (mPa s)
1.1
1.2
1.3
1.4
1.5
1.6
1.7
Shear rate (s⁻¹)
40
60
80
100
120
140
160
0.025 wt%
0.1 wt%
0.4 wt%
(e)

Pawel Hawrylak
I. Ozfidan, P. Potasz, A.D. Guclu, O. Voznyy, M.Korkusinski, M. Grabowski,A. Wojs

Advanced Research Complex, Department of Physics, University of Ottawa,
25 Templeton Str,Ottawa ON CanadaK1N 6N5

Photonics, electronics and spintronics with graphene quantum dots

We describe here our recent theoretical work aiming at design of graphene nanostructures capable of realizing the three functionalities of a quantum circuit: electronics, photonics and spintronics, in a single material and at the nanoscale[1]. Integration of these different functionalities using graphene quantum dots may enable Carbononics - a unified approach to Information and Communication Technologies. The design tools include size, shape, type of edge, sublattice symmetry, topology, number of layers and carrier density in graphene quantum dots[1-9]. In particular, sublattice engineering allows design of magnetic moments tunable with voltage and light, size engineering leads to optical gaps from THz to UV while shape engineering leads to a degenerate exciton spectrum allowing for the generation of entangled photon pairs via XX-X cascade[7,8]. Geometry and e-e interactions allow for the integration of topologically protected states of matter into carbononics[9,10]. Comparison with experiment on colloidal graphene quantum dots[6-8] as well as other competing material systems [10,11] will be given.

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Mark C. Hersam

Department of Materials Science and Engineering, Northwestern University
2220 Campus Drive, Evanston, IL 60208-3108, USA
m-hersam@northwestern.edu
www.hersam-group.northwestern.edu

Solution-Processed Graphene and Related 2D Nanomaterial Inks

Two-dimensional nanomaterials have emerged as promising candidates for next-generation electronics and optoelectronics [1], but advances in scalable nanomanufacturing are required to exploit this potential in real-world technology. This talk will explore methods for improving the uniformity of solution-processed graphene and related two-dimensional nanomaterials with an eye toward realizing dispersions and inks that can be deposited into large-area thin-films [2]. In particular, density gradient ultracentrifugation allows the solution-based isolation of graphene [3], boron nitride [4], montmorillonite [5], and transition metal dichalcogenides (e.g., MoS_2, WS_2, $MoSe_2$, and WSe_2) [6] with homogeneous thickness down to the atomically thin limit. Similarly, two-dimensional black phosphorus is isolated in organic solvents [7] or deoxygenated aqueous surfactant solutions [8] with the resulting phosphorene nanosheets showing field-effect transistor mobilities and on/off ratios that are comparable to micromechanically exfoliated flakes. By adding cellulosic polymer stabilizers to these dispersions, the rheological properties can be tuned by orders of magnitude, thereby enabling two-dimensional nanomaterial inks that are compatible with a range of additive manufacturing methods including inkjet [9], gravure [10], screen [11], and 3D printing [12]. The resulting printed two-dimensional nanomaterial structures show promise in several applications including photodiodes [13], anti-ambipolar transistors [14], gate-tunable memristors [15], and heterojunction photovoltaics [16].

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James Hone

Columbia University, USA

Graphene devices and phenomena in the ultraclean limit

Two-dimensional materials offer a wide range of outstanding properties but are highly sensitive to disorder from the environment. We have developed a 'van der Waals transfer' technique to encapsulate graphene within crystalline h-BN with nearly perfect interfaces, and an 'edge contact' technique to achieve electrical contact to the encapsulated channel.[1] This provides graphene channels with ballistic transport over tens of microns at low T, offering a platform to explore a wide variety of phenomena and devices. This talk will summarize recent results in three areas: novel 'Hofstader Butterfly' physics [2]; demonstration of negative refraction at graphene p-n junctions [3]; and tunable interactions between in double-layer heterostructures of bilayer graphene that result in formation of an excitonic superfluid phase [4,5]

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Kuan-Tsae Huang

AzTrong Inc.
5F, #5, Technology Road, Hsinchu Science Park, 300, Hsinchu, Taiwan

Kthuang8@gmail.com

Graphene Li-Ion Battery – How it will shape the Electrical Vehicles of tomorrow

Lithium-ion batteries make a significant contribution to green transportation, including electric vehicles, hybrid vehicles, and plug-in hybrid vehicles, because of their outstanding performance [1–3]. The high price of lithium-ion batteries is one of limitations for their widely application. In order to reduce the economic costs of lithium-ion batteries, it is critical to improve the battery performance and cycle life, which are mostly affected by many factors such as operation temperature, current and battery configuration. The use of graphene and graphene oxide for both anode and cathode materials can increase the battery capacity and lifespan of a battery. We will present the potential of graphene battery and its impact on electrical vehicles of the future.

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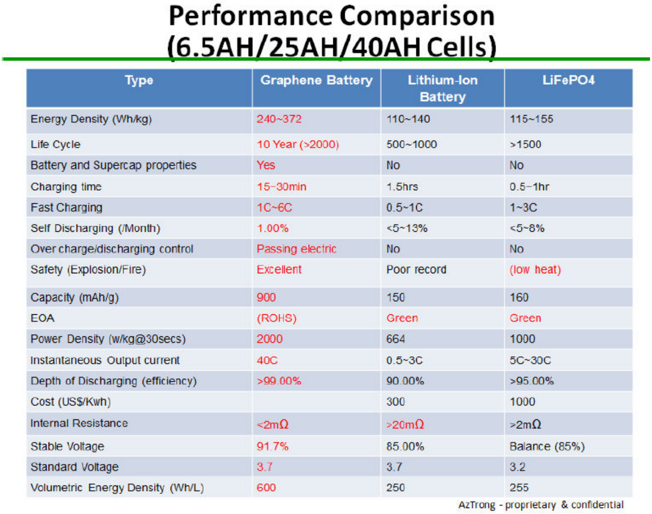
Figures



Performance Comparison (6.5AH/25AH/40AH Cells)

Type	Graphene Battery	Lithium-Ion Battery	LiFePO4
Energy Density (Wh/kg)	240~372	110~140	115~155
Life Cycle	10 Year (>2000)	500~1000	>1500
Battery and Supercap properties	Yes	No	No
Charging time	15~30min	1.5hrs	0.5~1hr
Fast Charging	1C~6C	0.5~1C	1~3C
Self Discharging (/Month)	1.00%	<5~13%	<5~8%
Over charge/discharging control	Passing electric	No	No
Safety (Explosion/Fire)	Excellent	Poor record	(low heat)
Capacity (mAh/g)	900	150	160
EOA	(ROHS)	Green	Green
Power Density (w/kg@30secs)	2000	664	1000
Instantaneous Output current	40C	0.5~3C	5C~30C
Depth of Discharging (efficiency)	>99.00%	90.00%	>95.00%
Cost (US$/Kwh)		300	1000
Internal Resistance	<2mΩ	>20mΩ	>2mΩ
Stable Voltage	91.7%	85.00%	Balance (85%)
Standard Voltage	3.7	3.7	3.2
Volumetric Energy Density (Wh/L)	600	250	255

AzTrong - proprietary & confidential

Yong-Hoon Kim

Graduate School of Energy, Environment, Water, and Sustainability (EEWS), Korea Advanced Institute of Science and Technology (KAIST), Daejeon 305-701, Korea

y.h.kim@kaist.ac.kr

Computational Study of Graphene-based Interfaces for the Realization of Next-generation Energy and Electronic Devices

In this talk, I will present our recent research effort in the characterization of graphene-based interfaces for the realization of next-generation energy and electronic devices in collaboration with experimentalists. First, I will discuss the mechanism of high electrocatalytic activity of Se edge-doped graphene nanoplatelets for iodine reduction reaction (IRR) in dye-sensitized solar cells (DSSCs) [1]. The mechanisms of IRR in carbon-based DSSC counter-electrodes are clarified based on density functional theory (DFT) and non-equilibrium Green's function calculations, together with the electrochemical kinetics analysis. Next, I will describe a DFT study of the graphene/WSe_2 "barristor" device that shows an exceptional low-temperature characteristic [2]. Combined with the experimental characterization of defect states and device modeling, self-interaction-corrected DFT calculations reveal a novel charge transport mechanism originating from trap-assisted tunneling rather than the typical thermionic emission process. If time permits, I will close the talk by mentioning the DFT study of high-performance Li battery realized with TiO_2 anatase nanoparticles adsorbed on graphene, in which we provide the atomistic picture of the "domino cascade" Li ion insertion process [3].

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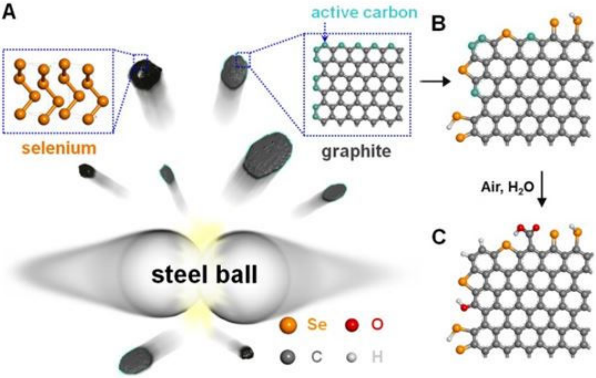
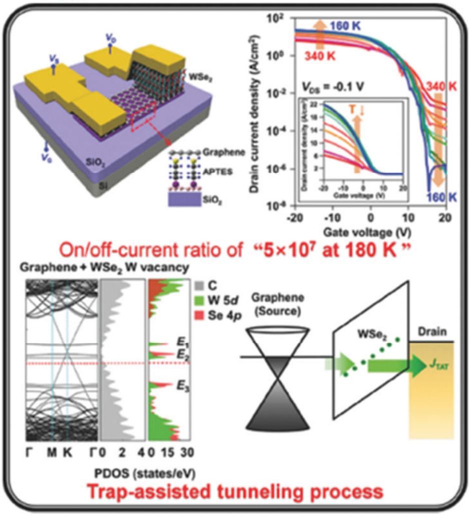
Figures


A
selenium
active carbon
graphite
B
Air, H_2O
C
steel ball
Se
O
C
H


V_D
V_S
WSe₂
V_G
SiO₂
Si
Graphene
APTES
SiO₂
10^2
10^0
10^{-2}
10^{-4}
10^{-6}
10^{-8}
160 K
340 K
V_{DS} = -0.1 V
340 K
160 K
T ↓
Drain current density (A/cm²)
Gate voltage (V)
-20
-10
0
10
20
On/off-current ratio of "5×10⁷ at 180 K"
Graphene + WSe₂ W vacancy
C
W 5d
Se 4p
E_1
E_2
E_3
Γ M K Γ 0 2 4 0 15 30
PDOS (states/eV)
Graphene (Source)
WSe₂
Drain
J_{TAT}
Trap-assisted tunneling process

François Léonard

Sandia National Laboratories, Livermore, CA, USA

Optoelectronics in 1D and 2D materials

Low-dimensional materials such as graphene, transition metal dichalcogenides, semiconducting nanowires, and carbon nanotubes, have attracted attention for applications in electronics and photonics, as well as for the wealth of new scientific phenomena that arise at reduced dimensionality. In this presentation I will discuss our experimental and theoretical work to develop a fundamental understanding of optoelectronic phenomena in such systems, and to exploit these nanomaterials for photodetection applications. For example, we use spatially-resolved photocurrent measurements combined with other techniques (e.g. thermoelectric measurements, Raman thermography) to identify the origin of the photocurrent in nanowires and 2D materials. We reveal a new regime of operation in MoS_2, $MoSe_2$, and their alloys, where the photocurrent depends superlinearly on light intensity [1]. In the case of GaN/AlGaN nanowires, we combine optoelectronic and thermoelectric measurements to reveal the coexistence of photogating and photothermoelectric effects [2]. Finally, I will discuss the realization of broadband infrared and terahertz photodetectors using macroscopically aligned carbon nanotubes [3-6].

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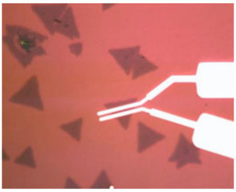
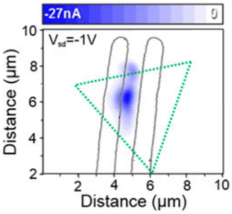
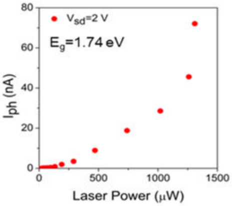
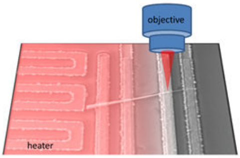
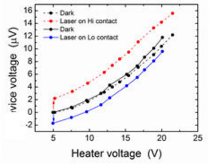
Figures

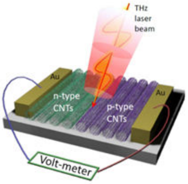


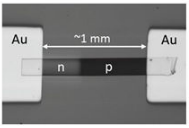

-27nA
0
V_{sd}=-1V
Distance (μm)
10
8
6
4
2
2
4
6
8
10
Distance (μm)

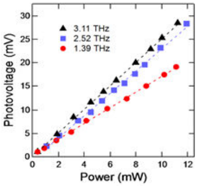

V_{sd}=2 V
E_g=1.74 eV
I_{ph} (nA)
80
60
40
20
0
0
500
1000
1500
Laser Power (μW)


objective
heater


Dark
Laser on Hi contact
Dark
Laser on Lo contact
vice voltage (μV)
16
14
12
10
8
6
4
2
0
-2
5
10
15
20
25
Heater voltage (V)


THz
laser
beam
Au
n-type
CNTs
p-type
CNTs
Au
Volt-meter


Au
~1 mm
Au
n
p


3.11 THz
2.52 THz
1.39 THz
Photovoltage (mV)
30
25
20
15
10
5
0
0
2
4
6
8
10
12
Power (mW)

Xiao Li[1]
Hongwei Zhu[2]

[1]School of Mechanical Engineering, Southeast University, Jiangning District, Nanjing, China
[2]School of Materials Science & Engineering, Tsinghua University, Haidian District, Beijing, China
liyixiaofeng@126.com

Graphene based hybrid structures for solar-driven water splitting

Developing clean and renewable energy has been considered as one of the most effective ways to address the energy and environmental related crisis. One of the most powerful sustainable energy, solar energy, can be utilized through solar–driven water splitting, which contains two energy conversion units. One is the photovoltaic solar cell, which converts light energy to electric energy for power supply. The other is the water electrolysis unit, which converts electric energy to hydrogen energy by using highly efficient and stable catalysts. Graphene and its hybrid nanocomposites, with their excellent optical, electrical and mechanical properties, can be applied in this system, to further improve the photoelectric conversion efficiency, and decrease the electrical consumption in the water electrolysis module.

Herein, to develop the solar–driven water splitting system, first, a hybrid graphene/Si–solid electrolyte solar cell is fabricated by combining graphene with solid electrolyte, achieving a conversion efficiency to be 11%, acting as the photoelectric conversion unit in this system. Then, water electrolysis catalysts are prepared by simply depositing transition metal (Co, Ni) based selenides on the conductive graphene substrate, which largely reduce the noble metal consumption in hydrogen production, realizing the conversion of electric energy to hydrogen energy at low cost. By connecting the solar cell and water electrolysis unit in tandem, a solar–driven water splitting system is constructed, with the highest solar-to-hydrogen conversion efficiency to be 6.65%.

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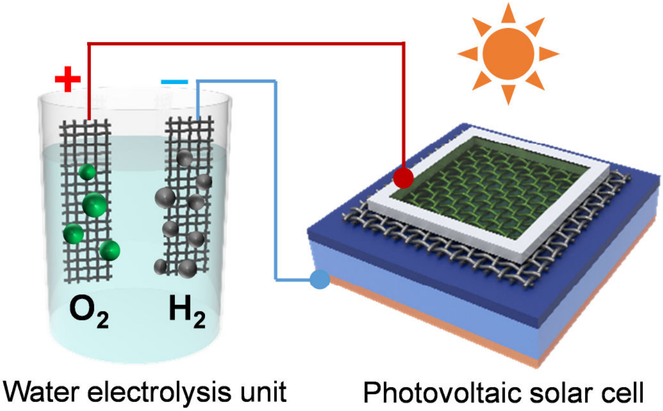
Figures



Vincent Meunier

Department of Physics, Applied Physics, and Astronomy Rensselaer Polytechnic Institute, Troy, NY 12180, USA

Modeling vertical and lateral electron transport across heterostructures of 1D and 2D materials

I will present recent developments made in the understanding of charge transport across heterostructures between nanostructured materials, including one-dimensional and two-dimensional graphene- and transition metal dichalcogenide- based systems.

The first part of the presentation will focus on electron transport taking place along on-dimensional heterostructures [1]. I will show how controlled chemical doping on one side of the junction can lead to physical properties that are well suited for photovoltaics and photocatalysis device development. In addition, it will be demonstrated that the appropriate choice of monomers used in the bottom-up assembly of these materials can yield tunable properties.

The second part of the talk will be devoted to a description of the role of collective motion in the ultrafast charge transfer in van der Waals (vdW) heterostructures. [2] Using time-dependent density functional theory molecular dynamics, it was found that the collective motion of excitons at the interface of the vdW heterostructures leads to plasma oscillations associated with optical excitation. Application to the experimentally realized MoS_2/WS_2 heterostructure yields good agreement with experiment, indicating near complete charge transfer within a timescale of 100 fs.

Finally, I will briefly present recent theoretical developments in the understanding of electronic band gap renormalization induced by substrate polarization effects, taking adsorbate size into account.

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Jean-Luc Meunier
Ulrich Legrand, Norma Mendoza-Gonzalez, Dimitrios Berk

Chemical Engineering, McGill University,3610 University St, Montreal (Qc), Canada
jean-luc.meunier@mcgill.ca

A thermal plasma route for the generation and functionalization of highly crystalline graphene structures and stable nanofluids

Thermal plasma (TP) reactors are used extensively for the generation of particles having specific compositions or phase structures. Nanoparticles (NPs) are also being generated using precursors that are either in the gas phase, in liquid solutions or even sometimes in the solid phase. More difficult is the controlled homogeneous nucleation of pure nanomaterials (chemically and structurally), and controlled nucleation of non-isotropic structures such as graphene. This talk will concentrate on achieving the 2-D structure of graphene in a TP system enabling extremely high purity and crystallinity of the material produced. The very high temperature homogeneous nucleation window (~4000-5000 K) involved in the TP processing route enables a bottom-up approach, provided one can control the 2-D structural evolution of the NP nucleated. Modeling of the energy/fluid fields as well as the graphene particle nucleation/growth fields indicate the critical clusters of carbon set the thickness (number of atomic layers) of the graphene, while the residence time in the growth field correlates with the sheet side lengths (on the order of 100nm x100nm, with on average 10 atomic layers in thickness; namely forming graphene nanoflakes (GNF)). An understanding of the physical processes enabling the 2D growth provides separate control parameters for playing on the number of layers and sheet scale lengths. The crystallinity parameters for these GNF from TEM and Raman spectroscopy are exceptional in comparison to regular top-down synthesis approaches. The graphene NPs most often require some chemical functionalization for specific applications, and again a TP can provide the active species for functionalization scenarios forming primary bonds between the functional group and graphene. The high temperature plasma forms a unique environment allowing purity from the simple precursors, unrivaled crystallinity from the extreme temperatures of nucleation and growth, and in situ flexibility for tuning of the functionalities directly inside the synthesis reactor through the use of the colder downstream recombining plasma.

This talk will describe the road for a controlled and pure graphene nucleation, followed in the same reactor by nitrogen, oxygen and iron functionalization of the graphene structure. The aimed applications here are for catalytic activity, in particular for a non-noble metal catalyst in fuel cells, and in the generation of graphene-based nanofluids that show full stability over time and high temperatures without the use of surfactants.

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Figures

Figure 1: Highly crystalline graphene nanoflakes (GNF) generated in a thermal plasma environment.

Jean-François Morin

Département de chimie et Centre de Recherche sur les Matériaux Avancés (CERMA), Université Laval, 1045 Ave de la Médecine, Québec, Canada G1V 0A6

Jean-francois.morin@chm.ulaval.ca
http://labnanoorg.chm.ulaval.ca/en

Photochemical Bottom-up Synthesis of Nanographenes and Graphene Nanoribbons

Nanographenes (NGs) and graphene nanoribbons (GNRs) are very promising materials for electronic applications such as field-effect transistors, solar cells and biosensors. However, NGs and GNRs produced by physical methods suffer from poor solubility in common solvents, which make difficult their deposition on large substrates as required for the production of the next generation of electronic devices. Moreover, physical methods do not allow control over size, shape and functionalization of GNRs, leading to undefined electronic and physical properties. Methods to prepare such materials using the solution-phase bottom-up approach have been proposed. Yet, no synthetic method provides clean aromatization step and the resulting materials can possess significant amount of structural defects. In this presentation, we will show a new bottom-up synthetic strategy for the preparation of soluble, well-defined NGs and GNRs with controllable width and physical properties.[1] Our strategy is based on the straightforward preparation of different chlorine-containing oligo- and polyphenylene precursors, followed by an intramolecular cyclodehydrochlorination (CDHC) reaction to form the graphenic structure. The synthesis and characterization of several NGs and GNRs will be presented.

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Oussama Moutanabbir[1]
Matthieu Fortin-Deschênes[1], Olga Waller[1], Amanda Hébert[1],
Pierre Levesque[2], Richard Martel[2]

[1]Department of Engineering Physics, École Polytechnique de Montréal, C. P. 6079, Succursale Centre-Ville, Montréal, Canada
[2]Département de Chimie, Université de Montréal, 2900 boulevard Edouard Montpetit, Montréal, Canada
oussama.moutanabbir@polymtl.ca

Stability and Epitaxial Growth of Group V 2D Materials: Theory and *in situ* Observations

Over the last few years, the existence and stability of several novel non-zero bandgap 2D materials have been theoretically predicted thus paving the way to developing a new class of graphene-like materials and heterostructures with superior functionalities. In this regard, group V 2D materials have been attracting a great deal of attention due to their unique physical and chemical properties such as a thickness dependent band gap in the visible to infrared range and anisotropic transport properties [1,2]. Up to date, 2D black phosphorus (2D-bP) and 2D antimony (2D-Sb) have been obtained by mechanical and liquid phase exfoliation from bulk crystals [3,4]. The exfoliated layers can be transferred and used to conduct fundamental studies and implement new device concepts. However, the large scale synthesis of these materials still remains a formidable task that is a crucial step toward scalable technologies. The epitaxial growth is by far the most promising method to synthesize these emerging 2D materials especially for mainstream electronic, optoelectronic and photonic applications. With this perspective, in this talk we present our recent studies of the epitaxial growth of group V 2D materials and real time investigations of their stability. Low-energy electron microscopy (LEEM) supported by *ab initio* calculations and kinetic Monte Carlo simulations are employed to elucidate the growth of 2D-(bP, As, Sb) materials, their thermal behavior and interaction with the substrate. In particular, we will discuss the mechanisms and dynamics of the thermal decomposition of 2D-bP [5] as well as the electronic properties and substrate-layer interaction of all three 2D materials on relevant growth substrates. Finally, we present *in situ* LEEM investigations of the growth of 2D-Sb and 2D-bP and discuss possible paths to achieve their epitaxial growth.

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Soroush Nazarpour

Group NanoXplore Inc., 25 boulevard Montpellier, Saint-Laurent, Canada

nazarpour@nanoxplore.ca

The wonderful world of Graphene - From lab to market

Graphene is a truly a “wonder” material with many promising attributes. Its outstanding thermal conductivity promises a revolution in thermal transfer and cooling industries. Its superb electrical conductivity and electron mobility makes it a natural as a filler in plastics to improve EMI and anti-electrostatic performance. Its solid performance during wear and friction tests paves the way for future heavy-duty greases and lubricants. The addition of mechanical strength, flexibility and light weight – all of these attributes together make graphene the wonder material of the century.

While all these attributes show great potential, bringing graphene from lab to market has been challenging. Such a high aspect ratio material requires special handling and processing to ensure reliability and consistent quality. Difficulties in scaling exfoliation techniques and poor dispersibility have hindered the rapid commercialization of graphene and resulted in high costs not supportable by industry business cases. Today graphene is on the verge of widespread adoption in multiple industries, but as of yet no killer applications have been identified. This talk will introduce a systematic approach on new application development using graphene, where industrial concerns are truly addressed, while identifying high potential areas where further research is essential.

Daniel Neumaier

AMO GmbH, Otto-Blumenthal-Strasse 25, Aachen, Germany

neumaier@amo.de

Graphene based electro-optical modulators and phase shifters

Graphene is a promising material for realizing high-performance on-chip electro-optical modulators because of graphene's excellent optical properties, which are controllable by electric gates, and the possibility to integrate graphene based modulators on silicon and other dielectric waveguides.

In this talk I will discuss our recent results on graphene based absorption modulators, phase modulators and thermo-electric phase shifters [1-3]. A key issue for all these devices is the residual insertion loss added by the light absorption of graphene. This parameter is essentially depending on the mobility μ and doping level n in graphene, with higher values for both μ and n providing less absorption. In addition contaminations introduced during device fabrication play an important role on the overall insertion loss of the device. Therefore techniques for maintaining the intrinsically high carrier mobility in graphene during device fabrication and for achieving high, stable and reproducible doping levels in graphene need to be developed in order to realize competitive electro-optical modulators.

Finally, I will benchmark the current results with competing technologies and give an outlook on which performance can be expected for graphene based electro-optical modulators and phase shifters.

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Figures

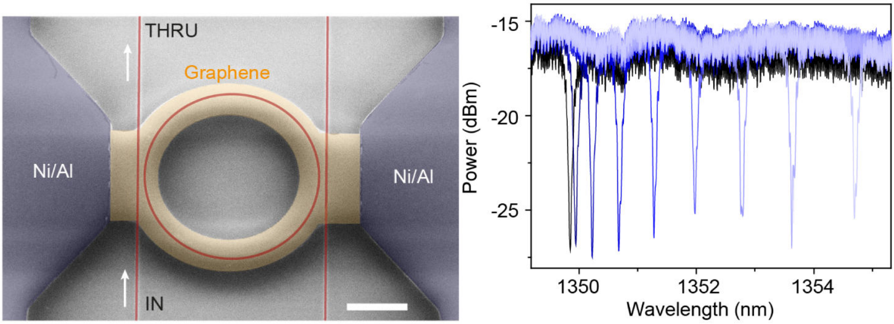

Figure 1: Left side: False color SEM image of a silicon micro-ring resonator having a graphene based thermos-electric phase shifter on top. Right side: One optical resonance of the ring resonator for different heating power levels in the graphene based heater. With increasing heating power the resonance shifts to longer wavelengths.

Emanuele Orgiu[1,2]

[1] Institut de Science et d'Ingénierie Supramoléculaires, Université de Strasbourg, Strasbourg, France
[2] INRS-EMT & Université du Québec, Montréal (QC), CA

orgiu@unistra.fr
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Supramolecular Approaches to Charge Transport Physics in Hybrid van der Waals Heterostructures

Graphene and the two-dimensional (2D) van der Waals semiconductors represent the thinnest, air stable semiconducting materials known. Their unique optical, electronic and mechanical properties hold great potential for harnessing them as key components in novel applications for electronics and optoelectronics. However, the charge transport behavior in such semiconductors is more susceptible to external surroundings (e.g. gaseous adsorbates from air and trapped charges in substrates) and their electronic performance is generally different than the corresponding bulk materials due to the fact that surface and bulk coincide when going from 3D down to 2D. Interestingly, one can take advantage of the latter property by using ordered supramolecular layers in order to tune charge transport and optical properties of such 2D materials. Hence, the electrical properties of graphene can be strongly influenced by the presence of physisorbed molecules, which induce charge transfer and doping[1]. In this context, supramolecular chemistry makes it possible to precisely tune the doping effect via the formation of ordered self-assembled monolayers (SAMs) of molecules embedding different functional groups[2].

In my presentation, I will give an example of a more general physical scenario: the charge transport and the doping on graphene can be tuned through ad-hoc engineering of the supramolecular assemblies physisorbed on its surface. A very fine photoreactivity process allows to achieve molecules featuring the same assembly motif on the surface but different terminal groups which contribute to doping the graphene in a novel way.

This approach will give an easy example of all the potential hidden in van der Waals heterostructures composed of supramolecular lattices physisorbed onto graphene.

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Stephan Roche

Catalan Institute of Nanoscience Nanotechnology, Campus UAB, Bellaterra, Spain
ICREA, Institució Catalana de Recerca i Estudis Avancats, Spain

stephan.roche@icn2.cat

Could Graphene and 2D Materials revolutionize Spintronics? Current challenges & opportunities

Graphene has been heralded as the ideal material to achieve long spin propagation and further control the spin degree of freedom, in the quest of advancing non-charge-based information processing and computing, and for creating a new generation of active (CMOS compatible) spintronic devices together with non-volatile low energy MRAM memories. Many technological roadblocks and lack of fundamental knowledge are however limiting today´s progress. Indeed, despite ultralow intrinsic and Rashba spin-orbit couplings (SOC) in clean graphene (μeV range), measured spin lifetimes remain in the range of several nanoseconds. This is orders of magnitude shorter than initially predicted, but already enough to envision disruptive non-charge-based room-temperature applications [1]. Besides, the physics of graphene "*can be enriched and manipulated*" by harvesting the large amount of possibilities of proximity effects with magnetic insulators, strong SOC materials, topological insulators, etc. One challenge is to endow a sizable spin-to-current conversion efficiency by enhancing spin-orbit interaction (say up to meV). Claims have been made that very large spin Hall angles (figure of merit for spin Hall effect-SHE) could be generated by using chemical functionalization with hydrogen or Au/Cu ad-atoms, or interfacing graphene with WS_2 substrate [2]. Those results are however fiercely questioned [3].

In this talk, I will discuss spin transport in graphene-based materials, accounting for the effect of substrate, impurities, and ad-atoms and using a fully quantum derivation of spin dynamics and calculation of multiterminal quantum resistance, going beyond the usual semi-classical theory and conventional approximations made in the literature. The role of pseudospin in driving spin dephasing and relaxation will be unveiled in the ultraclean limit for which electron-hole puddles and micron eV spin-orbit interaction are determining the fundamentals of spin lifetimes, giving foundations to experimental features [4]. Second, I will discuss the impact of chemical functionalization (fluorine, gold and thallium ad-atoms) and proximity effects with other 2D materials in generating spin-dependent phenomena such as spin filters, quantum spin Hall and tunable spin Hall effects in graphene.

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Robert Rölver

Robert Bosch GmbH
Corporate Research, Germany

Graphene for MEMS sensor products: Chances and challenges

Graphene, the 2D material with very promising mechanical and electrical properties has now reached a level of maturity where an industrial engagement is necessary on the way to commercialization. Bosch the worldwide largest manufacturer of MEMS sensing devices is now a part of the growing number of companies and startups looking into graphene applications and large scale fabrication routes for graphene synthesis.

In terms of wafer based fabrication there is still a need for improvement of the commonly used graphene synthesis process based on layer transfer techniques including wet processing. An overview of possible methods and current progress in this field will be covered in our presentation.

In terms of applications Bosch corporate research has started first collaborations with the academia to evaluate the potential of graphene for sensor applications. Here, Bosch currently focuses on magnetic sensing. The two-dimensional material graphene exhibiting substantially higher charge carrier mobility seems to be predestined for use in magnetic field sensing. Neglecting all extrinsic sources of disorder, the phonon scattering limited mobility of graphene can surpass 100,000 cm^2/Vs at room temperature at a reasonably low charge carrier density of 10^{12} cm^{-2}, exceeding all other common material candidates for Hall sensing.

Shintaro Sato[1,2]

[1] Fujitsu Laboratories Ltd, 10-1 Morinosato-Wakamiya, Atsugi, 243-0197, Japan
[2] Fujitsu Limited, 10-1 Morinosato-Wakamiya, Atsugi, 243-0197, Japan

sato.shintaro@jp.fujitsu.com

Application of graphene to electronic devices

Nanocarbon materials, such as graphene and carbon nanotubes, have excellent electrical, thermal, and mechanical properties and are thus expected to be used for future electronics devices. We have been working on synthesis of nanocarbon materials and their application to transistors and interconnects for years. In this talk, we first describe our efforts to grow nanocarbon materials, which include graphene growth over a 300-mm wafer by chemical vapor deposition (CVD), and synthesis of an interesting carbon structure consisting of multilayer graphene combined with the upper ends of vertically aligned multiwall carbon nanotubes on a substrate [1]. Bottom-up synthesis of graphene nanoribbons (GNRs) on Cu twin crystals (Fig. 1) and step edges is also explained [2,3]. CVD-synthesis of multilayer hexagonal boron nitride (hBN), which can be good substrate and passivation film for graphene devices, is also described.

As for transistor applications, we briefly review theoretical and experimental studies on transport properties of GNR-channel transistors first. We then explain one of our approaches to obtain a high current on-off ratio of graphene channel transistors. In fact, we used a graphene channel irradiated by helium ions, demonstrating a sizable transport gap and an on-off ratio of about two orders of magnitude at room temperature [4]. We then describe a double-gate graphene transistor and its applications, which includes a binary digital phase modulator making use of ambipolar properties of graphene [5].

We also explain the application of graphene to interconnects. We actually fabricated GNR interconnects with widths as narrow as 8 nm using electron beam lithography (Fig. 2) [6]. The GNR interconnects were heavily doped by intercalation of $FeCl_3$ molecules and demonstrated resistivity and reliability better than those of Cu with similar dimensions.

Acknowledgements

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Figures

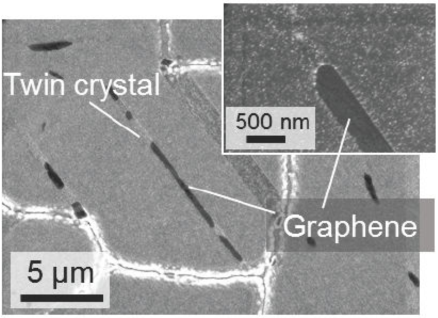

Figure 1: Graphene ribbons formed on Cu twin crystal regions

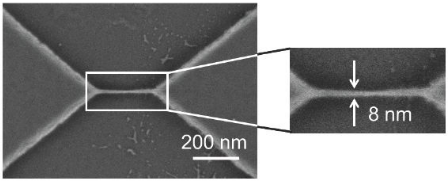

Figure 2: Graphene naroribbon interconnect with a width of 8 nm

Mauricio Terrones

Department of Physics, Department of Chemistry, Department of Materials Science
and Engineering and Center for 2-Dimensional & Layered Materials. The
Pennsylvania State University, University Park, Pennsylvania 16802, USA &
Institute of Carbon Science and Technology, Shinshu University, Japan

mut11@psu.edu

Defect Engineering in 2-Dimensional Materials: Graphene, Doped-Graphene and Beyond

This talk will first provide an overview of different defects in 2-Dimensional materials including graphene and Chalcogenides. In particular, we will focus on: 1) defining the dimensionalities and atomic structures of defects; 2) pathways to generating structural defects during and after synthesis and, 3) the effects of having defects on the physico-chemical properties and applications. We will then discuss the synthesis of large-area, high-quality monolayers of nitrogen-, boron- and silicon-doped graphene sheets on Cu foils using ambient-pressure chemical vapor deposition (AP-CVD). Scanning tunneling microscopy (STM) and spectroscopy (STS) demonstrate that defects in doped graphenes arrange in different geometrical configurations exhibiting different electronic and chemical properties. Interestingly, these doped layers can be used as efficient molecular sensors and electronic devices. In this context, Graphene enhanced Raman spectroscopy will be introduced and it will explained that by doping graphene with Nitrogen, the Fermi level (EF) of graphene shifts, and if this shift aligns with the lower unoccupied molecular orbital (LUMO) of a molecule, charge transfer would be enhanced, thus significantly amplifying the molecule's vibrational Raman modes. Concentrations as low as 10-11 mol/L of different dye molecules can be detected using GERS. Finally, we will discuss the controlled synthesis and assembly of chalcogenide monolayers on different substrates. The electronic performance of monolayers of MoS_2, WS_2 and hetero-systems operating under flexural strain will also be presented. Our findings demonstrates that it is now possible to use chalcogenide layers for the fabrication of flexible electronic devices, however, defect control is required to tailor their performance.

Keywords: graphene, dopant, defects

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Florina Truica-Marasescu[1]
Mary Gallerneault[2], Cameron Giglio[2], Osayuki Osazuwa[2],
Aristides Docoslis[2]

[1] ALCERECO Inc., Grafoid Global Technology Centre,
945 Princess Street, Kingston, Ontario K7L 0E9
[2] Department of Chemical Engineering, Queen's University, Kingston, ON K7M 8Y2

florina.truica@alcereco.com

Solvent effects control of the wettability behaviour of graphene thin films

Graphene is naturally hydrophobic, which makes it an excellent candidate for producing super-hydrophobic surfaces. Two factors play important roles for the functionality and the wetting behaviour of super-hydrophobic surfaces: the chemical composition of the material, and the geometric structure of the surfaces [1]. However, as it has a low contact angle (CA) with water droplets, the hydrophobicity of graphene is not enough to exhibit the strong water repellency needed for some practical opportunities, such as fluid transport, self-cleaning, environmental remediation, and energy applications.[2] Two approaches are generally used to improve the wetting behavior of graphene surfaces: (i) creation of surface micro / nano structures by mechanical means, or (ii) the grafting of suitable chemical groups / polymers (e.g. chemical functionalization with F-containing groups) [2,3]. In this work, a different approach was used, namely exploiting solvent-particle interactions in graphene dispersions in order to control surface morphology (and thus wetting behaviour) of the films deposited onto Si wafers. The impact of solvent-graphene platelet interactions on the morphology of the suspended particles, and on the morphology of the resulting graphene films was investigated using appropriate characterization methods, such as: Dynamic Light Scattering (DLS), CA measurements, and Scanning Electron Microscopy (SEM). The dispersant polarity, and the total surface coverage of substrates were studied to develop an understanding of each parameter on the tunability of graphene-based thin films. The study made evident that the physico-chemical properties of the graphene films vary significantly depending upon the treatment of the precursor material, the cleaning/preparation of the substrate surface, and the deposition method.

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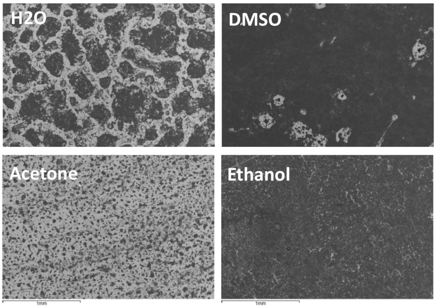

Figure 1: SEM images of graphene films prepared under identical spraying conditions on Si wafers by using dispersions with different solvents, such as water, dimethyl sulfoxide, acetone and ethanol

Helge Weman

Norwegian University of Science and Technology, NO-7491 Trondheim, Norway
CrayoNano AS, NO-7052 Trondheim, Norway

helge.weman@ntnu.no

Epitaxial growth of semiconductor nanostructures on graphene: A solution for UV LEDs

We have recently developed a generic atomic model which describes the epitaxial growth of semiconductor nanostructures on graphene that is applicable to all conventional semiconductor materials [1, 2], as shown in Fig. 1. The model was first verified by cross-sectional transmission electron microscopy studies of GaAs nanowires that grow epitaxially and dislocation-free on graphene, in spite of a lattice mismatch of 6.3 % [1]. Recently we have also shown the vertical growth of dislocation-free GaN nanowires on graphene mediated by nanometer-sized AlGaN nucleation islands [3]. The epitaxial growth of semiconductor nanostructures on graphene is very appealing for device applications since graphene can function not only as a replacement of the semiconductor substrate but in addition as a transparent and flexible electrode for e.g. solar cells and LEDs.

For deep ultraviolet AlGaN based LEDs in huge need for various disinfection and sterilization purposes, the concept offers a real advantage over present thin film based technology. Such thin film UV LEDs are today very expensive and inefficient due to the lack of a good transparent electrode (ITO is absorbing in deep UV), the high dislocation density in the active thin film layers, low light extraction efficiency, and the use of very expensive semiconductor substrates (e.g. AlN). The spinoff company CrayoNano are now developing LEDs based on the selective-area growth of AlGaN nanostructures on mask-patterned graphene, which potentially can overcome all these problems, as will be further discussed in my talk.

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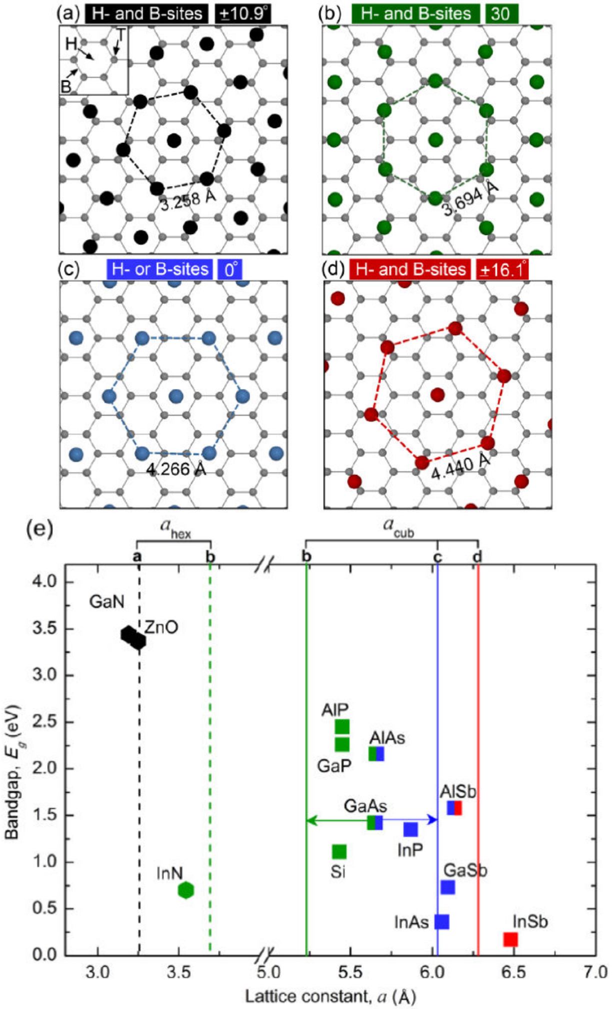

Figure 1: (a-d) Relative orientation and arrangement when semiconductor atoms are adsorbed on H- and/or B-sites. (e) Generic model describing the semiconductor bandgaps vs. lattice constants together with lattice constants for the lattice-matched atom arrangements on graphene [1].

Rune Wendelbo
Stephanie H. Santos

Abalonyx AS, Forskningsveien 1, 0373-Oslo, Norway

rw@abalonyx.no

Graphene and graphene derivatives for energy applications

Graphene has several unique properties potentially useful for a range of energy related applications. Established and potential applications include batteries and supercapacitors, solar cells, gas sensors, gas storage, nuclear industry and applications in high voltage transfer cables. These will be reviewed and illustrated with examples from recent reports.

Batteries is a high growth market where graphene's electric and thermal conductivity obviously is useful, but more interesting is graphene's potential as an agent to stabilize certain meta-stable phases, e.g. in sulfur based cathodes for post-LIB-batteries, pursued by a range of battery companies. Graphene based super capacitors have already been commercialized. For SCs, the organization of the graphene particles is important. The material must be densely packed to reduce the volumetric capacity but still having a sufficiently open structure to allow fast migration of ions. Applications for photovoltaics are less obvious, due to the black color of graphene and its lack of band-gap in its pure form. However, doping of graphene has been shown to create band-gap, and compounding with other electronic materials such as zink oxide also shows potential. In fuel cells, graphene may play a role as support for catalysts and in proton exchange membranes in DMFCs.

The use of GO to soak up heavy radionuclides from water has been reported and verified1. This is certainly one of the applications to be seen commercialized in this decade, easily requiring production of hundreds of tons of GO per year. On the other hand, applications as components in sensors, although potentially large in numbers will be small in mass due to the very limited amount used in each sensor. When it comes to energy storage, GO being an insulationg material does not have an obvious role. However, GO can easily be converted to reduced graphene oxide (rGO), effectively competing with other graphene powders. Effectively, rGO has the advantage over other graphene powders in that GO is easy to work with. rGO has a high density of defects compared to more well-crystalline graphene types, but this can also represent an advantage in that holes present in the sheets allowing for diffusion of e.g. Li-ions.

For most energy related applications, the useful form of graphene is powder. Abalonyx is active in production of GO and rGO as well as in the development of GO and rGO optimized for several applications. Abalonyx' sister company Graphene Batteries is engaged in application of rGO and pillared rGO for certain battery and supercapacitor applications with promising results.

The final, and not least important parameter for large scale use of graphene family materials is price. Graphene powders produced in plasma are expensive and the process not very scalable. Delamination of graphite in organic solvents has similar challenges. Present estimates indicate that the price of GO can come down to well below 40 USD/Kg and rGO to just under 100 USD/Kg – price levels that can be tolerated by many industries if compensated by real improvements in performance of the GO/rGO-containing products.

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Aiping Yu

Department of Chemical Engineering, University of Waterloo, Canada

Advanced Li-Ion Hybrid Supercapacitors Based on 3D Graphene- Foam Composites

Currently one of the most promising techniques is to use 2-dimensional (2D) graphene sheets to form three-dimensional (3D) networks known as graphene sponge. Graphene sponges are synonymous with the following terminology: foams, aerogels, hydrogels and 3D porous networks. Graphene sponges exhibit high surface area and highly accessible pore volume. They also display high strength and conductivity with low densities. Dr. Aiping Yu's team at the University of Waterloo has developed a few technologies to fabricate graphene foam and composite to make electrodes of Li ion supercapacitor. The presentation will cover the main achievements of the group in this field.

Hong Guo

Department of Physics, McGill University, Montreal, Canada

Calculating impurity-limited carrier mobility of 2D materials

Realistic nanoelectronic devices inevitably have some disorder which affect device operation. Unintentional impurities locate at unpredictable positions in the material and predicted physical property should be averaged over the impurity configurations. One may generate many impurity configurations, calculate each of them and average over the results. Unfortunately such a brute force approach is too costly to be practical for first principles simulations. To solve this problem, we have developed a theory of nonequilibrium coherent potential approximation (NECPA) that is integrated with the density functional theory (DFT) and Keldysh nonequilibrium Green's function (NEGF), in which the disorder configuration average is carried out analytically thereby only a single first principles NEGF-DFT computation is necessary to obtain the disorder averaged transport properties. Using NECPA, we have calculated disorder limited carrier mobility of 2D materials including graphene and monolayer black phosphorus.

Acknowledgement: the work is in collaboration with Saeed Bohloul, Zi Wang, Eric Zhu, Lei Liu, Kirk Bevan, Michael Zhang and Kui Gong.

LATE

Max Lemme

University of Siegen, Germany

Graphene Device and Process Integration – Challenges ahead!

Graphene research has matured from fundamental investigations of material properties to very specific demonstrations of devices and their potential for applications. In this talk, I will present several promising areas, where the exceptional properties of graphene may be exploited. Two examples are graphene-based hot electron transistors [1], [2] and graphene-membrane-based nanoelectromechanical systems [3], [4]. However, proper device and process integration remains challenging and currently prevents commercialization. Several such open issues will be discussed in detail [5], [6].

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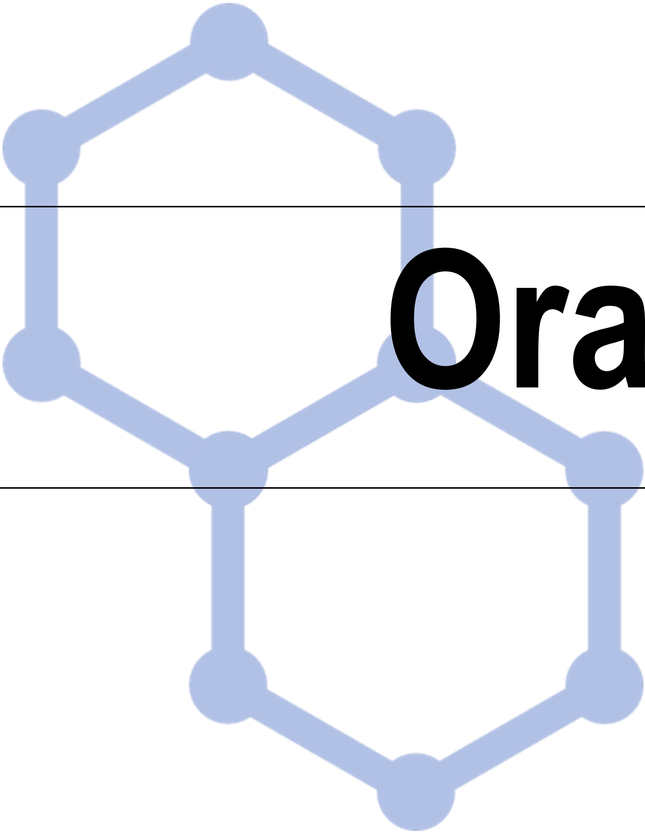

Orals

Arash Akbari-Sharbaf[1]
Michael G. Cottam [2]

[1] Département de Physique, Université de Sherbrooke, Québec, Canada
[2] Department of Physics and Astronomy, University of Western Ontario, Ontario, Canada

arash.akbari.sharbaf@usherbrooke.ca

Two-point Green's function correlations mediated by localized electronic edge modes in zigzag graphene nanoribbons

The unusual nature of the localized electronic edge modes in zigzag graphene nanoribbons (ZZ-GNRs) have been the subject of intense investigation since its inception above 20 years ago by Fujita *et al.* [1]. Nearly all theoretical investigations related to edge modes in ZZ-GNRs have been carried out numerically or semi-numerically. Here we will present the first comprehensive analytical investigation of the localized edge modes in ZZ-GNRs demonstrating that two possible distinct type of localized modes with strikingly different properties can exist in the system [2]. The first type of localized edge mode (or "zero" mode) is a topologically induced mode arising from the bipartite honeycomb lattice structure of graphene and is always present at zigzag edges. The second type of edge mode is only present at zigzag edges with a modified hopping parameter between edges sites distinct from the hopping between interior sites. In particular we demonstrate that correlations mediated by the zero mode between two atomic sites on the same edge is comparable to correlations between two atomic sites on opposite edges and independent of the width of the ribbon in leading order and in the absence of zero-mode damping effects. We also demonstrate the strength of correlations are strongly dependent on the sublattice labels of any two atomic points being considered. On the other hand correlations mediated by the "modified hopping" edge mode is independent of the sublattice labels and the two edges decouple when the width of the ribbon is greater than the attenuation length of the modified edge mode. Analytical expressions and numerical results are presented for the dependence of the mode frequencies on the ribbon width and on wave number parallel to the ZZ edges.

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JB Boland
Damien Hanlon, Jonathan Coleman
Trinity College Dublin, Dublin 2, Ireland

bolandjb@tcd.ie

Liquid-phase Exfoliation of Germanium Monosulfide

Since the introduction of graphene in 2004 there has been widespread research of two-dimensional materials in the area of nanoscience. Primary focus was initially placed on graphene, however many other nanomaterials have shown promise, such as boron nitride (BN) and transition metal dichalcogenides (TMDs) to name but a few.

Hanlon, D., *et al.* have produced nanosheets of black phosphorus (BP); a new layered material which shows great promise for optoelectronics [1]. Here we show the extension of the exfoliation process described to germanium monosulfide (GeS), a novel two-dimensional semiconductor material. The GeS monolayer is isoelectronic to that of black phosphorus.

We demonstrate that high-quality, few-layer GeS nanosheets, with controllable size can be produced in large quantities through the use of liquid phase exfoliation under ambient conditions in solvents such as N-methyl pyrrolidone and 2-propanol. Also shown is testing of these nanosheets for potential applications in battery electrodes.

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Figures

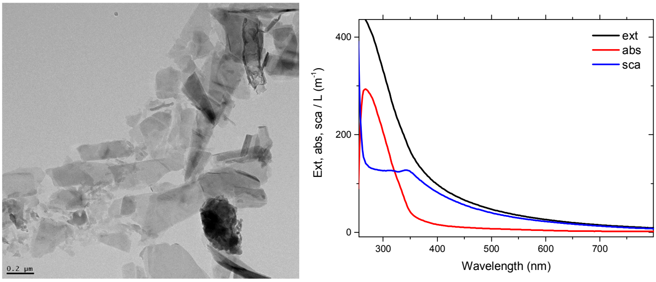

Figure 1: A) TEM image of GeS nanosheets produced from Liquid phase exfoliation, B) Ultra-Violet Visible spectra showing absorption, extinction and scattering data.

I.V. Bondarev[1]
D.Drosdoff[1], A.Widom[2], R.Podgornik[3,4] and L.M.Woods[5]

[1] North Carolina Central University, Durham, NC 27707, USA
[2] Northeastern University, Boston, MA 02115, USA
[3] Jozef Stefan Institute, Ljubljana, SI-1000, Slovenia
[4] University of Massachusetts-Amherst, MA 01003, USA
[5] University of South Florida, Tampa, FL 33620, USA

ibondarev@nccu.edu

Monopolar charge fluctuation induced forces in 2D nanostructures

Fluctuation-induced interactions have widespread applications in materials, micro- and nanoscaled devices. The much-studied Casimir and van der Waals interactions are due to vacuum electromagnetic mode fluctuations captured via the dielectric response properties of the objects.[1-4] Such forces are universal, and they are especially prominent at micron distances and below. Fluctuations of many other observables are also possible, which may give rise to different interactions.[5] Fluctuations originating from charge disorder on neutral slabs, in particular, are shown to give an additional contribution to the net interacting force while completely masking the typical Casimir–van der Waals interaction.[6]

Here, we investigate monopolar charge fluctuation induced forces in 2D nanostructures.[7] We present a general theory utilizing the capacitance concept and distinguishing between thermal and quantum mechanical effects through characteristic dependences on distance, temperature, and other factors. Graphene-based systems are taken as templates for which the theory is applied. Here, the fundamental difference of the charge-induced fluctuation forces from the typical Casimir force lies in their origin. While the Casimir force is due to the electromagnetic fluctuation excitations associated with the dielectric response of each plate, the charge-induced effect is due to monopolar charge fluctuations between the plates transferred through a connecting wire, or a metallic substrate (see Figure). Since in many cases nanostructures are characterized by a reduced Casimir force, the nanocapacitors we study offer the possibility of finding regimes where the charge-induced fluctuation interaction can be dominant. Our results are strong evidence that there are different types of dispersive interactions that occur in solid state devices, in particular when lower dimensional materials such as graphene are used.

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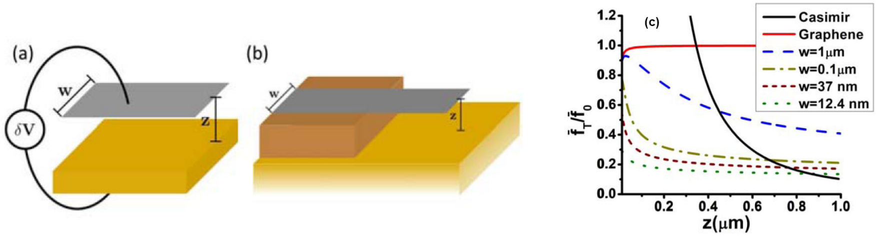

Figure 1: (a), (b) Metal sheet and carbon nanoribbon systems that exhibit monopolar charge fluctuation forces. (c) Distance (z-) dependence of the charge fluctuations thermal force normalized by $f_0 = -k_B T/(2Az)$ for graphene nanoribbons of varying width. The area A of all ribbon structures is chosen to be 1 μm^2. The graphene-metal Casimir force at room temperature is also shown.

R. Campos[1]
J. Borme[1], G. Machado Jr[1], M.F. Cerqueira[1,2], P. Alpuim[1,2]

[1] INL - International Iberian Nanotechnology Laboratory, 4715-330, Braga, Portugal
[2] Department of Physics, University of Minho, 4710-057, Braga, Portugal

rui.campos@inl.int

In-situ growth and functionalization of graphene for biosensing applications

Although in recent years we have witnessed staggering achievements in both fundamental and applied research in graphene, applications still lack the pervasiveness that this material unique properties anticipated. One of the problems with the establishment of a graphene technology comes from the required transfer step, from the native metal catalyst onto the final substrate, normally using a temporary polymeric substrate. As a consequence, a non-uniform, wrinkled, contaminated with debris sheet of graphene results, which no longer exhibits the record-breaking properties of the pristine 2D material. Moreover, the transfer step takes time, requires patience and persistence, and, crucially, it is largely operator dependent, hence very prone to errors.

Graphene extreme sensitivity to electric charges and fields in its vicinity, together with its high chemical stability, provide an ideal transducing platform for biosensing applications. Biosensing requires that the active surface is highly reproducible, which can be achieved by *in-situ* growth of the graphene layer. Here we show that graphene directly grown on copper and molybdenum surfaces, provides the required type of surface, in a transfer free process that can be used for biosensor development. Moreover, the metal catalyst is patterned previously to the graphene CVD step, which implies that, by design, graphene will grow selectively only on the substrate regions where it is required. This highly simplifies the graphene patterning process, which translates into cheaper devices and higher graphene quality, since the fabrication process becomes faster and the damage inflicted to graphene by lithographic processes is minimized.

Biosensing with graphene requires surface functionalization for specific analyte detection. We used as linker a pyrene derivative (1-pyrenebutyric acid N-hydroxysuccinimide ester, PBSE), a molecule that provides π-π interactions with graphene on the pyrene end, and has a ester group at the opposite end, which reacts with the amine group of the sensing element. Nucleic acids allow the development of biosensors with high specificity and affinity and its electrochemical characterisation is commonly performed taking advantage of the well-known thiol-gold interaction. In the present work, a 29 nucleotides long methylene blue labelled DNA strand (self-complementary on its ends to form a becon) is immobilised on the electrode surface and after electrochemical characterisation of the film, hybridisation is performed with a fully complementary strand (cDNA). Surface coverage, rates of electron transfer and biosensor performance are determined from cyclic, square-wave and differential pulse voltammetry and compared with the commonly used Au based biosensors. Both macro and microelectrodes will be fabricated and the results compared. The sensor performance will be compared with that of electrolyte-gated graphene transistors whose channel is functionalized in the same manner as described above.

Figures

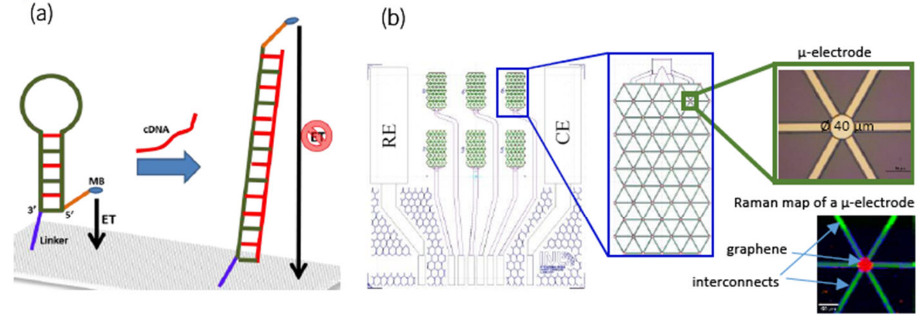

Figure 1: a) MB-labelled DNA strands immobilized on graphene surface b) Graphene **µ**-electrode arrays

Dhan Cardinal[1]

Carl-Bernard Charpin[2], Saman Choubak[1], Pierre Levesque[1], Patrick Desjardins[1], Richard Martel[2]

[1] Département de Génie Physique and Regroupement Québécois sur les Matériaux de Pointe, École Polytechnique de Montréal, Montréal, Canada
[2] Département de Chimie and Regroupement Québécois sur les Matériaux de Pointe, Université de Montréal, Montréal, Canada

Dhan.Cardinal@Polymtl.ca

Formation of Graphene Bilayers in CVD Growth

Understanding the fundamental aspects of graphene chemical vapor deposition (CVD) on copper is a key towards the mass production of this carbon allotrope. One way in which this understanding can manifest itself is through controlled growths of graphene layer, that is, time to completion, defect density and morphology [1,2,3]. Layer thickness trigger to a large extent the graphene properties and the presences of bilayer islands may be desirable or detrimental upon selected applications and nowadays needs to be controlled. Bilayers islands appear with no clear-cut explanation in sight and even those who specifically study graphene bilayer growth often do not provide a mechanism for this phenomenon [4,5,6,7]. We have shown in previous studies that at trace level, oxidizing impurity can totally govern the growth kinetics. In this study, by exercising a strict control over gas impurity in the furnace, we grow in a regime where impurities effects are minimized and no longer limits the growth. This gain of control over the growth allowed us to identify that bilayers formation, to a large extent, occur during the cool down phase. A mechanism is proposed for these inverted wedding cake bilayers invoking the thermal expansion coefficient mismatch between copper and graphene, causing a delamination and buckling of the later. By controlling the cooling parameters, we gained control over the presence or not of bilayer islands and to some extent their coverage and density. By getting rid of “cooling down” bilayers, we were able to identify a second category of bilayer with different origins. Interestingly while the “cooling down” bilayer appears to be random the second category is commensurate. While understanding bilayer growth is interesting from a fundamental point of view, this controlled growth also enables the development of graphene-bilayer related technologies, which are particularly enticing for their intrinsic bandgap.

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H. L. Chang[1,2]
C. T. Chang[2]

[1] Department of Material Science and Engineering of National Chiao Tung University
[2] Research and Technology Development Department, 2PJT,
Samsung Electronics South Korea

Selective Nano- Structured Material Growth in Nanoelectronic Device

Catalytic-assisted CNTs are integrated into trenches and holes under CH_4/H_2 gases by microwave plasma chemical vapor deposition, the trench and hole are used to fabricate for gate electrodes and interconnections, respectively. Results indicate the orientation of grown CNTs is dominated by pattern geometry. The growth models of Fe and $CoSi_2$ and application for nanoelectronics are purposed.

The integration of CNTs into Si-based metal-oxide-semiconductor field effect transistor (MOSFET) or new nanoelectronicos remains a challenge in the fields of transistors and interconnections. CNTs are accepted as candidates for use in molecular electronics to overcome the physical limitation of current Si transistors and Cu interconnections [1-2]. The feasibility of realizing this vision depends on direct approaches to selective depositions in the trenches or holes of Si wafers. Bundles of CNTs in the trenches and holes can provide sufficient current density in the form of channels and conductors, respectively. This work systematically elucidates the synthesis of CNTs by microwave plasma CVD (MPCVD). In this investigation, Fe catalyst and $CoSi_2$ film employed frequently as gate electrodes and a contact material in Si microelectronics are applied. The selective growth of CNTs in trench/hole approaches is also examined. The morphology and nanostructures of CNTs are characterized. The field emission characteristics of CNTs deposited in the trenches and holes are investigated to determine electronic performance. CNTs were synthesized on patterned Si wafers with trench arrays and holes by MPCVD system with CH_4+H_2 source gases. Two catalytic films were prepared on patterned Si wafers before CNTs synthesis: (1) an Fe film (20 Å) and (2) a Co film (75 Å). Then two-step rapid thermal annealing (RTP) was conducted at 600 ℃ for 60s and at 760 ℃ for 20s, under N_2 ambient. Si wafer with patterned $CoSi_2$ holes with a $CoSi_2$ catalyst film was formed by rapid thermal processing (RTP) and the self-alignment process. Figure 1 presents the AES depth profiles Co film on Si substrate with rapid thermal annealing (RTP) indicating the chemical composition of $CoSi_2$.The oxide film was prepared by plasma-enhanced chemical deposition (PECVD), and plasma dry etching and a wet stripping were adopted to remove the un-reacted metal film. The schematic diagrams of CNTs formation on trench and hole are shown in Fig. 2.

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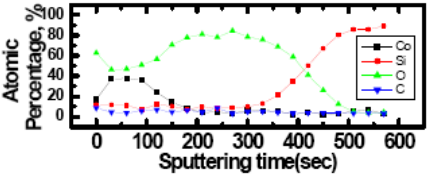

Figure 1: AES depth profile Co film/Si substrate after RTP

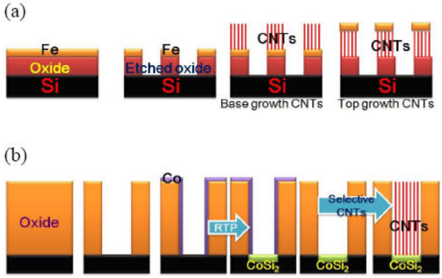

Figure 2: Process flows of forming CNTs on (a) Fe trench arrays, oxide film deposition ->Fe film deposition ->drying etching - >CNTs growth (b) $CoSi_2$ holes, oxide film deposition ->drying etching ->Co film deposition ->RTP ($CoSi_2$ formation)-> unreacted metal removal -> CNTs growth

M. T. Dau[1,2]

C. Vergnaud[1,2], A. Marty[1,2], F. Rortais[1,2], C. Beigné[1,2], G. Renaud[1,3], V. Guigoz[1,4], O. Renault[1,4], P. Mallet[1,5], J.-Y. Veuillen[1,5], C. Alvarez[1,3], H. Okuno[1,3], L. Huder[1,6], V. Renard[1,6], C. Chapelier[1,6], E. Bellet-Almaric[1,2], P. Pochet[1,3] and M. Jamet[1,2]

[1] Université Grenoble Alpes, F-38000 Grenoble, France
[2] INAC-SPINTEC, CEA/CNRS, F-38000 Grenoble, France
[3] INAC-MEM, CEA, F-38000 Grenoble, France
[4] CEA, LETI, Minatec campus, F-38054 Grenoble, France
[5] CNRS, Institut NEEL, F-38000 Grenoble, France
[6] INAC-PHELIQS, CEA, F-38000 Grenoble, France

minhtuan.dau@cea.fr

Van der Waals epitaxy of monolayer $MoSe_2$ on sapphire and on epitaxial graphene/SiC

Two-dimensional materials based on transition metal dichalcogenides (TMDs) have gained increasing attention because of their fascinating features in electronic and optical properties. Achievement of layered TMDs with large lateral dimensions is a pre-requisite for an easy implementation into devices and for the exploration of novel physics. Molecular beam epitaxy is a viable approach for synthetizing large-scale TMDs thin films down to one monolayer with high uniformity and purity. In addition, as TMDs bulk exhibits out-of-plane van der Waals-type interactions between sandwiched layers, a van der Waals interaction between as-grown films with substrates is expected when growing TMDs films on inert substrates. Therefore, using MBE approach allows to study impact of such short-range force on the growth dynamics of TMDs on different substrates [1].

We report the growth and characterizations of large-scale monolayer TMDs $MoSe_2$ on sapphire substrates and fashionable 2D heterostructure composed of one monolayer $MoSe_2$ on graphene/SiC substrates. The TMDs epilayers were fabricated by two-step growth process which consists of depositing TMDs layers at 500°C - 550°C followed by a post-annealing around 700°C. We point out that the growth rate and the substrate temperature of the first step play a key role in the growth dynamics of the TMDs. The post-annealing allows to improve the crystalline quality and to smooth the layers. We have performed several techniques in order to characterize the structure of TMDs layers such as RHEED, (synchrotron) X-ray diffraction, TEM, AFM. Firgures 1a, b show the in-plane scans of the monolayers $MoSe_2$ on sapphire and graphene, respectively. One can see that the $MoSe_2$ layer grows up without any preferential orientation with respect to the sapphire substrate, while it is clearly commensurately epitaxial on the graphene/SiC. Indeed, the phi-scan confirms the polycrystalline character of the layer on sapphire that means that the layer is composed of small multi-domains. For 2D heterostructure, we did not find any significant off-angle variants stemming from the free rotation of the layer regarding the graphene layer. The homogeneity over centimeter-sized surface of the $MoSe_2$ layers was confirmed by Raman spectroscopy. The XPS and TEM measurements reveal possible chemical interactions between sapphire substrate and deposited species that likely introduce random nucleation sites, thus, resulting in the polycrystalline growth. Powerful techniques such as STM–STS performed at 4K and k-resolved PEEM, a complementing ARUPS, were used to probe the atomic-scale structure and electronic properties of the heterostructure. The interplay between proximity effect and resulting electronic structure, which enables to shed light on the nature of van der Waals interaction within vertical 2D heterostructure, will be discussed in the talk.

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Figures

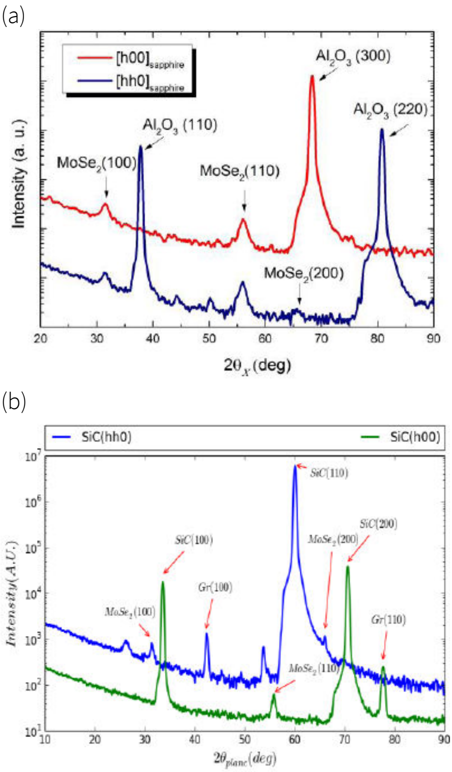

Figure 1: (a) XRD of 1ML $MoSe_2$ on sapphire (b) XRD of 1 ML $MoSe_2$ on graphene/SiC

Kyle DiCamillo
Makarand Paranjape

Georgetown University, 37th and O Streets NW,
Washington, D.C., U.S.A

kd598@georgetown.edu

Automated Mechanical Exfoliation of Few Layer MoS_2 and $MoTe_2$

Two-dimensional materials are of great interest due to their unique electronic and optical properties. Graphene has received much attention for its excellent electron mobility and other unique properties. Its use as a gas sensing material has also been tested due to its excellent surface to volume ratio [1]. Unfortunately, graphene lacks the semiconducting properties that gas sensing layers (such as indium tin oxide) traditionally possess [2]. Molybdenum disulfide (MoS_2) is another two dimensional material that is analogous to graphene in its lattice structure and is made of stacked layers of molybdenum sandwiched between layers of bonded sulfur. However, bulk MoS_2 has an indirect band gap of 1.2 eV while monolayer MoS_2 has a direct band gap of 1.8 eV. Molybdenum ditelluride ($MoTe_2$) is a similar material to MoS_2, made of tellurium layers, taking sulfur’s place, and sharing the lattice structure of MoS_2 and graphene. $MoTe_2$ has an indirect band gap of 0.86 eV in bulk and a direct band gap of 1.8 eV as a monolayer, reinforcing its similarity to MoS_2. These two materials have potential for use in applications such as gas sensing [2, 3] where a semiconducting film is more useful than a material with no band gap. One of the challenges of using two dimensional films in devices such as gas sensors is the difficulty of mass producing these materials at an acceptable quality and performance level. The methods of producing few layer films of MoS_2 or $MoTe_2$ currently include mechanical exfoliation and chemical vapor deposition (CVD) or other growth techniques. Mechanical exfoliation stands out not only for not being a chemically driven growth technique but also for its ability to produce pristine films of two dimensional materials [4]. However, mechanical exfoliation is hard to reproduce and produces small sample footprints. A programmable machine would be useful for adding consistency to the technique of mechanical exfoliation. A rheometer is a tool which can apply stresses to soft solid materials and gather data about their mechanical properties. This makes it a candidate to reproduce the pressing and peeling of flake-covered pieces of adhesive that is normally done by hand to achieve mechanical exfoliation. We show that automating the mechanical exfoliation using a rheometer adds greatly to the reproducibility of this fabrication technique. It also establishes machine parameters that can be varied to optimize and standardize a mechanical exfoliation recipe for that machine.

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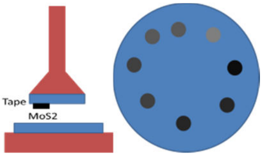

Figure 1: The rheometer presses together and peels apart pieces of tape while the rheometer rotates between each exfoliation. This results in progressively thinner sheets of material down to only a few molecular layers.

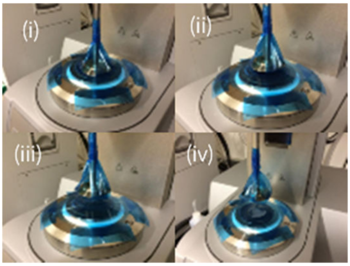

Figure 2: Rheometer tool and stage as the two pieces of tape are brought into contact and then peeled apart.

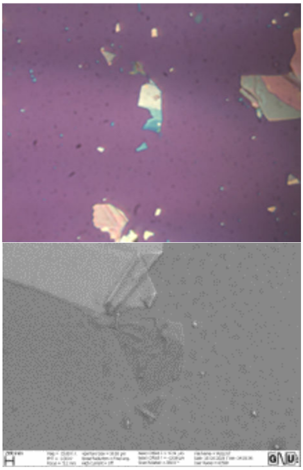

Figure 3: Optical and SEM images of a monolayers MoS_2 flake.

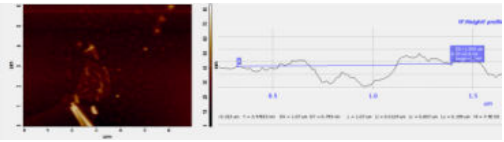

Figure 4: AFM image of monolayer MoS_2 flake an height profile of monolayer MoS_2 flake.

A. El Fatimy[1]
Peize Han[1], R.L. Myers-Ward[2], A. K. Boyd[2], K. M. Daniels[2], A. B Sushkov[3], D. Drew[3], D.K. Gaskill[2] and P. Barbara[1]

[1]Georgetown University, Department of Physics, Washington DC, USA
[2]U.S. Naval Research Laboratory, Washington, DC, USA
[3]Department of Physics, University of Maryland, Maryland, USA

a.elfatimy@gmail.com
ae497@georgetown.edu

Highly sensitive hot electron bolometer based on graphene quantum dots

We studied graphene quantum dots patterned from epitaxial graphene on SiC and from chemical vapor deposition of Graphene Films on SiO_2/Si with a resistance strongly dependent on temperature. The combination of weak electron-phonon coupling and small electronic heat capacity in graphene makes these quantum dots ideal hot-electron bolometers. We characterized the response of these quantum dot bolometers to THz radiation as a function of dot size, with sizes ranging from 30 to 700 nm and from 2.5 to 80 K temperature range. We show that quantum dots exhibit a variation of resistance with temperature higher than 430 MΩ/K below 6 K, leading to electrical responsivities for absorbed THz power greater than 1×10^{10} V/W. The power dependence of the responsivity was also investigated. The high responsivity, the potential for operation above 80 K and process scalability demonstrate that graphene quantum dot THz detectors hold great promise for practical applications.

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Figures

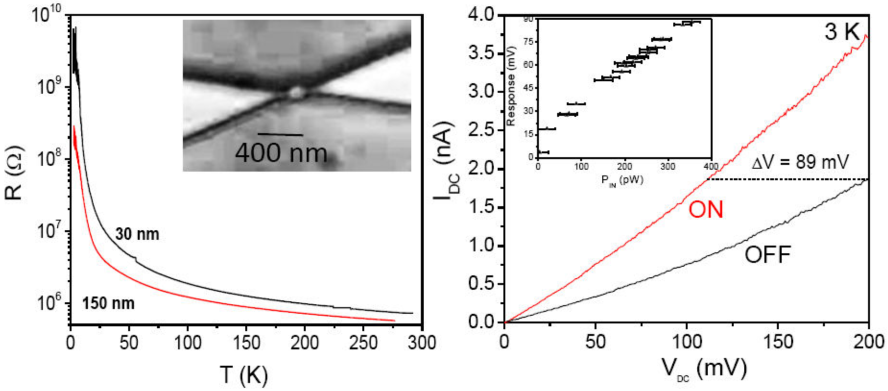

Figure 1: Temperature and THz power dependence of graphene quantum dots. a, Resistance vs. temperature for two quantum dots with different diameter at V_{DC} = 5mV. **b**, Current-voltage characteristic with (red) and without (black) of 0.15 THz radiation and the response as function of THz power absorbed for a 200-nm dot at 3K.

Randolph E. Elmquist

Yanfei Yang*, Guangjun Cheng, Patrick Mende, Irene G. Calizo, Randall M. Feenstra, Chiashain Chuang, Chieh-Wen Liu, Chieh-I Liu, George R. Jones, Angela R. Hight Walker

National Institute of Standards and Technology, USA

* yanfei.yang@nist.gov

Epitaxial graphene homogeneity and quantum Hall effect in millimeter-scale devices

Fully quantized magnetotransport is observed in two 5.6 mm × 5.6 mm epitaxial graphene (EG) devices, grown on the Si-face of hexagonal SiC(0001) by constrained Si sublimation at 1900 °C [1]. In our sample A, the quantized Hall resistance ($R_{xy} = h/2e^2$) is maintained to five parts in 10^9 up to $I_{xx} \approx 0.72$ mA at T = 3.1 K and 9 T. This result exceeds the highest QHE critical currents reported in graphene (0.5 mA) or GaAs heterostructures (0.6 mA) and is ascribed to remarkable uniformity of the EG layer. In our samples, adsorbed polar molecules act as a gate, and it is possible to vary the carrier concentration through this effect [2]. AC transport measurements were made on sample B at seven levels of carrier density, as shown in Fig. 1. Despite the presence of inhomogeneous layer number in some small areas of sample B, the measured mobility is $\mu \approx 43\ 700$ cm^{2}V^{-1}s^{-1} for $n_0 \approx 0.5 \times 10^{10}$ cm^{-2}, which is the highest reported mobility for any graphene sample of millimeter-scale dimensions. For comparison, similar characteristics of μ and n have been reported in gated, high-quality EG devices of < 5 μm width [3], but in those devices the mobility was seen to decrease for values of n below $\approx 10^{11}$ cm^{-2}. This decrease is absent in the sample B, allowing transport with high mobility at very low carrier densities. Pristine exfoliated graphene on h-BN exhibits mobility about twice what we measure, and typically the maximum mobility occurs at low carrier density [4]. We explain the high level of transport mobility and critical current using various microscopy measurements of the graphene layer and substrate, including Raman mapping that reveals low and uniform strain over much of the surface of both devices. Our results show that disorder in EG produced by the SiC substrate does not result in reduced mobility at low carrier density if the density of scattering sources is relatively low, and that more homogeneous strain in monolayer EG may reduce the level of carrier density fluctuations.

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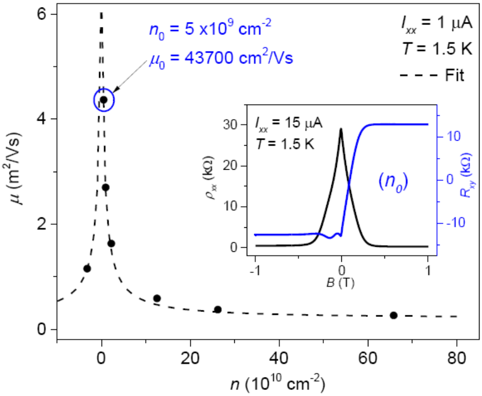

Ibrahim Fakih[1]
Farzaneh Mahvash[1,2], Mohamed Siaj[2], Thomas Szkopek[1*]

[1]McGill University, Montreal, Quebec, Canada
[2]Université du Québec à Montréal, Montreal, Quebec, Canada

ibrahim.fakih@mail.mcgill.ca
*thomas.szkopek@mcgill.ca

Improved ion sensing graphene field effect transistors using ultra-thin parylene encapsulation

Graphene field effect transistors (FETs) are an attractive candidate for sensing applications because of their high charge carrier mobility and the ideal coupling between graphene charge carriers and surface potential. However, depositing a selective layer on the graphene for these sensing applications can degrade graphene's electrical properties, increase hysteresis, and present a challenge for maintaining its stability and performance. Here, we protect the graphene by encapsulating it with an ultrathin layer (~ 4nm) of parylene, a hydrophobic polymer, and then deposit a ~ 3nm aluminum oxide sensing layer to characterize large are graphene ion sensitive field effect transistors (ISFETs). Not only do we demonstrate pH sensitivities approaching aluminum oxide's limit of ~ 48 mV/pH [1] over pH 2.5 through pH 7.5, but we also observe significant improvements in field effect mobilities of ~ 7000 $cm^2V^{-1}s^{-1}$ and in transconductance with limited hysteresis compared to previous work [2]. The observed improvements due to the graphene encapsulation has resulted in the substantial improvement in the minimum detection limit of our ISFETs to 5 mpH/ rtHz, advantageous to highly sensitive applications.

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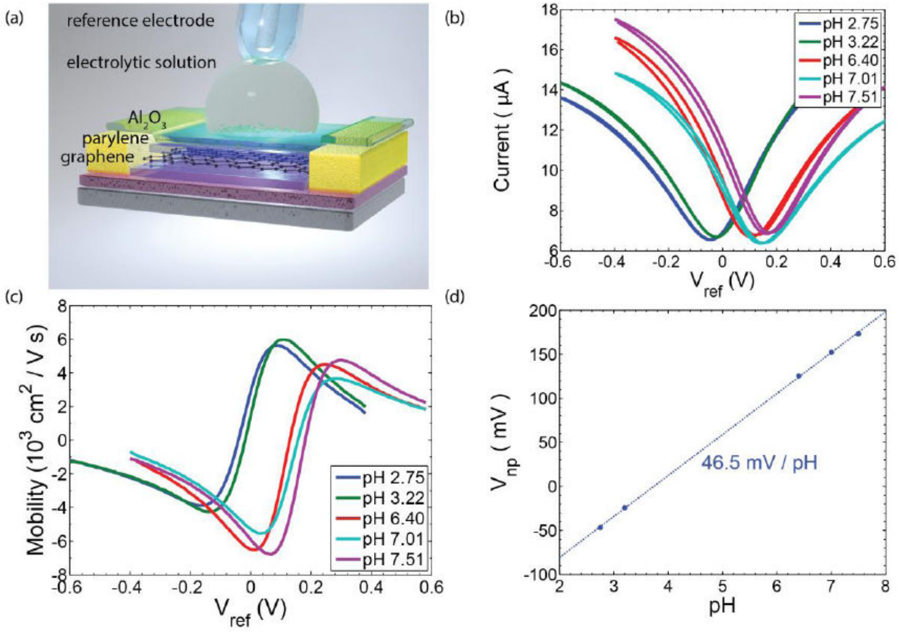

Figure 1: (a) Schematic of a graphene based ISFET encapsulated with parylene with Al_2O_3 as a sensing layer. (b) The current flowing through the graphene channel while sweeping the reference electrode voltage V_{ref} in both directions with different pH buffer solutions. (c) The field effect mobility of the carriers versus the reference electrode voltage V_{ref} for different pH buffer solutions. (d) The neutrality point potential of the minimum conductance V_{np} for the different pH buffer solutions; a 46.5 mV/pH sensitivity is observed.

Mauro Ferreira

Trinity College Dublin, Ireland

The wagon-wheel effect in graphene-based materials: Can apparently innocuous oscillations impact so many physical properties?

The key signature of any symmetry-breaking perturbation that arises due to the presence of impurities and/or defects is the appearance of spatial fluctuations in quantities like the local density of states (LDOS) and the carrier density, both of which oscillate away from the location of the perturbation. However, in graphene these oscillations are hidden by their commensurability with the lattice parameter of graphene, analogously to the optical illusion known as the wagon-wheel effect. Surprisingly, the absence of such oscillations can have a dramatic effect in a number of electronic properties of graphene and its derivative materials, as shown in this talk. For a start, these simple oscillations, or lack thereof, can explain the sublattice asymmetry seen in impurity-doped graphene where dopants prefer to bind to carbon atoms occupying one of the two sublattices of graphene even though both are absolutely equivalent. Another manifestation of the wagon-wheel effect is that the bonding symmetry with which a dopant binds to graphene may determine how good a scatterer this dopant is. As a result, certain impurities tend to be completely invisible causing no scattering whatsoever, with direct implications for the designing of efficient sensors. Finally, in addition to explaining the aforementioned physical properties, we argue that the wagon-wheel effect can also be explored to improve the accuracy of electronic structure calculations in graphene-related materials.

D. Gagnon
F. Fillion-Gourdeau, J. Dumont, C. Lefebvre, and S. MacLean

Université du Québec, INRS-Énergie, Matériaux et Télécommunications, Varennes, Québec, Canada
Institute for Quantum Computing, University of Waterloo, Waterloo, Ontario, Canada
denis.gagnon@emt.inrs.ca

Analogy between graphene light-matter interactions and strong field quantum electrodynamics

We theoretically describe the use of graphene as a "QED simulator" following the formal analogy between quantum electrodynamics (QED) and the dynamics of low-energy excitations in graphene [1]. More specifically, time-dependent electron-hole pair production in graphene can be used to simulate QED pair production from vacuum either in the multiphoton or Schwinger regime (also known as tunneling regime). We concentrate on the case where the system is driven out of equilibrium by the application of a linearly polarized or circularly polarized laser excitation. For applied fields of magnitude $E_0 = 10^7$ V/m, the multiphoton regime corresponds to optical frequencies and the tunneling regime corresponds to terahertz frequencies. The resulting electron momentum densities are computed via a split-operator method for the numerical solution of the time-dependent Dirac equation [2]. For a linearly polarized excitation and a single optical cycle applied on the graphene sample, our results can be intuitively explained in terms of Landau-Zener-Stueckelberg interferometry and the Kibble-Zurek mechanism [3]. This highlights the unparalleled contribution of this 2D material to fundamental physics studies. Graphene in a strong magnetic field is also discussed in the context of electron-hole pair creation [4].

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P. Godignon[1]
A. Ballestar[2,3], A. García-García[1,2], L. Serrano[2,3], J. M. de Teresa[3,4], M. R. Ibarra[3]

[1] CNM-IMB-CSIC, Campus UAB, Bellaterra, 08193 Barcelona, Spain
[2] GPNT - Graphene Nanotech, S.L., Miguel Villanueva 3, 26001 Logroño, Spain
[3] INA, LMA, Universidad de Zaragoza, Mariano Esquillor, 50018 Zaragoza, Spain
[4] ICMA, Universidad de Zaragoza, 50009 Zaragoza, Spain

philippe.godignon@cnm.es

High Electron Mobility and Low-Field Quantum Hall Effect in Graphene grown on SiC substrates ready for back gating

It seems that we are entering the era of two dimensional materials, that ever since they have been isolated showed amazing potential for developing new devices based on their extraordinary properties. When it comes to industrialization and commercialization, two main issues have to be faced: utilization of reproducible and affordable standardized manufacturing processes and high quality of the final product. For that purpose, we fabricate graphene by selective sublimation of Silicon Carbide (SiC), using an optimized protocol developed in our company. Our typical graphene layers grown on Si face SiC present mobilities in the range of 3000-4000 $cm^2V^{-1}s^{-1}$ at room temperature and up to almost 40000 at low temperature. Furthermore, pursuing the inclusion of graphene in the field of electronics, we produced a novel graphene product grown on top of pre-processed SiC substrates with a buried conducting layer at a depth of 300 nm, formed by ion implantation. By these means, we can provide a substrate where graphene can be easily back gated by biasing the semiconductor buried layer. Our quality inspections, in terms of Raman investigations, revealed that we obtained SiC substrates covered with more than 75% of graphene of superior quality. Moreover, measurements performed on Hall bar devices fabricated along the SiC terraces corroborated those results. That is: mobilities of the electrons as high as 8000 $cm^2V^{-1}s^{-1}$ at low temperature and 2800 $cm^2V^{-1}s^{-1}$ at RT, as well as, low magnetic field plateaux on the Quantum Hall Effect have been observed. Even if these performances are pretty good in this first generation material, the pre-processing of the semiconductor substrate is still impacting the graphene performances and we are working to improve the SiC processing to reach the electrical values obtained on graphene grown on virgin SiC substrates. All those premises make our graphene an ideal candidate to overcome the difficulties in bringing its potential to the desired industrial scale.

Figures

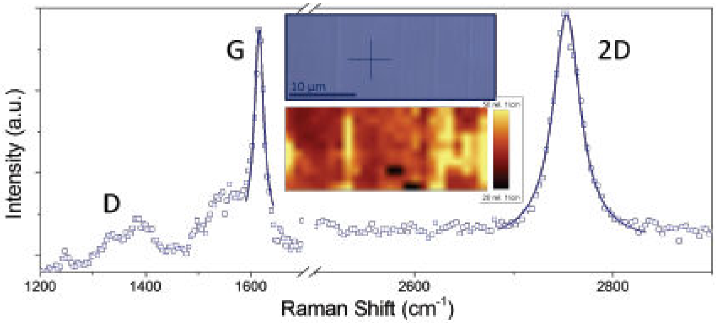

Figure 1: Raman spectra measured at the position indicated by the blue crossin the optical image at the inset. The lower inset figure consists on a maping of the FWHM of the 2D peak, in which only the yellow dots indicate positions where single layer graphene is not found, while all the rest corresponds to the presence of high quality monolayer graphene, as it can be inferred from the color scale to the right.

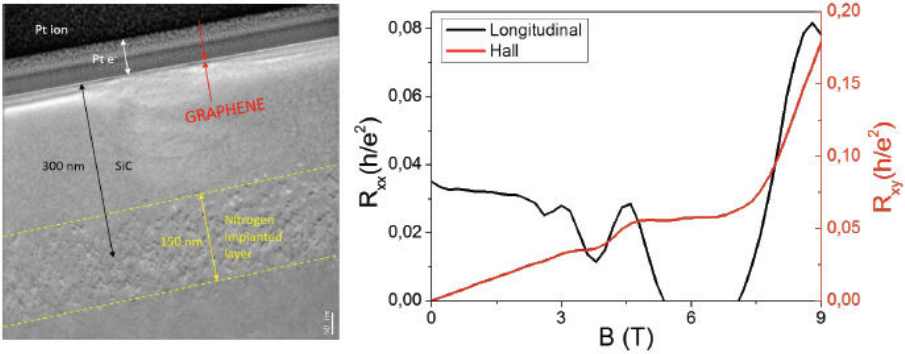

Figure 2: On the left hand side, a HRTEM cross sectional image of a studied sample. The single graphene layer, as well as the conducting buried layer can be observed. On the right side, the results obtained at 2 K for both longitudinal and Hall resistance on the mentioned device.

Damien Hanlon
Claudia Backes, Andrew Harvey, Jonathan Coleman
Trinity College Dublin, Dublin 2, Ireland
hanlonda@tcd.ie

Characterisation of Liquid-Exfoliated Black Phosphorus Nanosheets

Over the last number of years there has been significant interest in two-dimensional materials in the area of nanoscience. Initially research has been focused on the study of graphene, however; more recently focus has expanded to other 2D nanomaterials such as; BN, transition metal dichalcogenides (TMDs) and transition metal oxides (TMOs). Black phosphorus (BP) is a new layered material which has generated significant interest over the last year. It is only the second elemental 2D material to be studied after graphene. BP is a layered crystal in its bulk form, consisting of atomically thin layers of phosphorus atoms stacked on top of each other via van der Waals interactions, similar to the way graphene stacks to form graphite [1, 2]. However; in contrast to other layered 2D nanomaterials which display direct bandgaps in the monolayer form, BP displays a direct bandgap in the mono-, few-layer and bulk forms. This makes BP highly attractive for electronics and optoelectronics [3].

Accordingly there have been numerous reports producing BP nanosheets via mechanical cleavage. However; to produce large quantities of material a scalable method is required. Therefore, BP crystal has been subjected to liquid phase exfoliation in cyclohexyl pyrrolidone (CHP), a common organic solvent. This process produces high quality nanosheets with lateral dimensions up to 3μm. These nanosheets unlike graphene and some other layered materials are unstable under ambient conditions [4]. This poses a significant problem for processing and fabrication of devices using BP. Herein a spectroscopic method is presented to track the degradation of the nanosheets in solution produced using liquid phase exfoliation. A stability metric is also presented, which can be used to determine the amount of reacted nanosheets in a given sample. It was found that the instability of the nanosheets predominantly occurs via a disproportionation reaction at the edges. Interestingly the stability of the nanosheets varies across different solvents where amide solvents show the greatest stability. Samples prepared using appropriate solvents can slow down the reaction which reduces the degradation to ~8% after 3 days.

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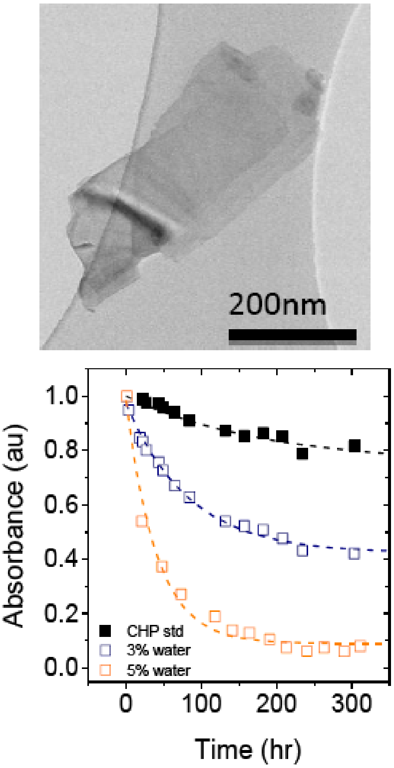

Figure 1: A) TEM image of nanosheets produced from Liquid phase exfoliation, B) Decreasing nanosheet concentration with varying water contents as a function of time.

Lin He

Center for Advanced Quantum Studies, Department of Physics, Beijing Normal University,
Beijing, 100875, People's Republic of China

helin@bnu.edu.cn

Direct imaging of topological edge states at a bilayer graphene domain wall

Looking for systems where topological edge states persist in the absence of external magnetic fields boosts rapid developments in condensed matter physics in the past few years. The AB-BA domain wall in gapped graphene bilayers is a rare naked structure that is believed to host one-dimensional topological electronic states. Here we show, for the first time, direct imaging of its topological edge states by using scanning tunneling microscope (STM) [1-3]. The simultaneously obtained atomic-resolution images of the domain wall provide us unprecedented opportunities to measure the spatially-varying edge states within it. The one-dimensional conducting channels are observed to be mainly located around the two edges of the domain wall, which is reproduced quite well by our theoretical calculations. Our experiment further demonstrates that the one-dimensional topological states are quite robust even in the presence of high magnetic fields, raising hopes of graphene-based electronics with ultra-low dissipation.

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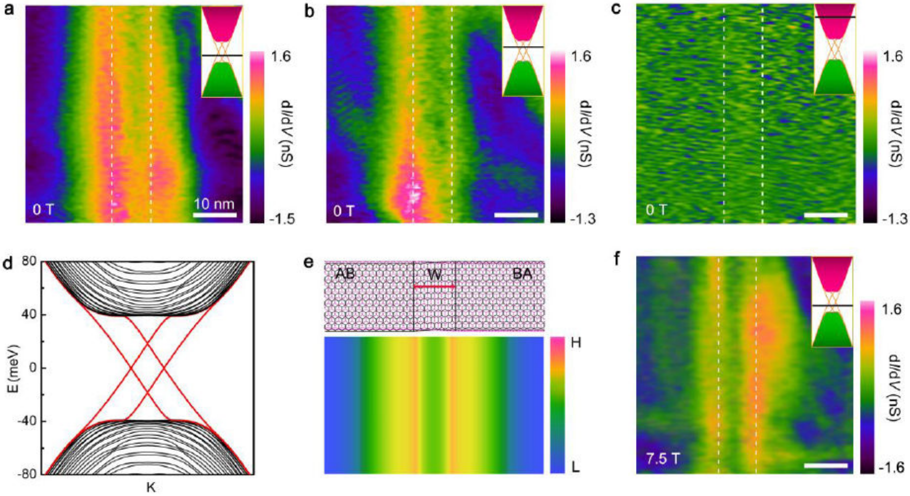
a
0 T
10 nm
1.6
dI/dV (nS)
-1.5
b
0 T
1.6
dI/dV (nS)
-1.3
c
0 T
1.6
dI/dV (nS)
-1.3
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40
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-80
E (meV)
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e
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BA
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dI/dV (nS)
-1.6

S. Hernandez Santos
Rune Wendelbo*
Abalonyx AS, Forskningsveien 1, 0314 Olso, Norway
shs@abalonyx.no
* rw@abalonyx.no

Graphene Oxide for Removal of Heavy Metals from Tunnel Wash Water

The potential of graphene-based materials like graphene oxide (GO) and reduced graphene oxide (rGO) as efficient adsorbents for contaminants such as radionuclides, cationic/anionic dyes and heavy metals has been studied in the recent years with encouraging results (Yang, 2010), (Zhao, 2011), (Ramesha, 2011), (Romanchuk, 2013). In the present work we evaluate GO as scavenger for heavy metals from tunnel wash water. Norway has approximately 800 km of tunnel construction along its territory (Torp, 2013); these tunnels are subjected to regular maintenance that includes washing episodes. Tunnel wash water can contain particulate material, soap, polycyclic aromatic hydrocarbons (PAH), oil components and heavy metals such as zinc (Zn), lead (Pb), copper (Cu) and cadmium (Cd) among others (Meland, 2012).

In order to test the performance of GO as an adsorbent for heavy metals present in tunnel wash water, three experimental series were conducted. Series 1 and 2 were done in tunnel wash water from a half-wash episode in December 2015 at the Nordby tunnel in Akershus province in Norway. The water sample was characterized and results showed measurable concentrations of Zn (650 µg/L), Cu (60 µg/L), Pb (8.3 µg/L), PAH and oil components. Series 3 was done in deionized water containing 5 ppm of Zn, Cu, Pb and Cd. Samples from the three series were treated with GO (0.6 g/L) and an inorganic flocculant* developed by our company in order to enhance separation. Graphene oxide was prepared by modified Hummers method (Hummers, 1958). The precipitate was separated by centrifugation and the supernatant was analysed by inductively coupled plasma mass spectrometry for elemental analysis. Results in terms of relative adsorption efficiency are shown in Table 1.

Efficiencies for Cu and Zn in series 1 and 2 are high, however values for Zn are higher than the ones for Cu, suggesting that mechanisms such as cation competition and concentration effects may influence the adsorption of these metals onto GO when present in a multicomponent system. Results from series 3 show very high adsorption efficiency for all the heavy metals tested, suggesting that the presence of PAH and oil components in series 1 and 2, may negatively affect the adsorption of Zn and Cu and potentially other heavy metals onto GO. The pH value in all series is near neutral value, this suggest that the removal of these metals may not be attributed to formation of precipitating species. These results represent a step forward to understanding the potential of GO in the water treatment field, where management of rest-cations and large pH adjustments are still challenges to overcome.

Note: **Flocculant is pending approval for a patent.*

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Series	pH	Heavy Metal and Relative Adsorption Efficiency (%)
1	7	Cu (71.7%), Zn (94.3%), Pb (> 75.9%)[1]
2	7.9	Cu (88.4%), Zn (94.2%), Pb (> 75.9%)[1]
3	7.9	Cu (99.9%), Zn (99.3%), Pb (99.9%), Cd (99.9%)

Table 1: Results of experimental series 1,2 and 3; pH value and relative adsorption efficiency are shown.

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[1] Concentration values for Pb (not shown) after treatment were under the LOQ (2.0 µg/l), thus it is possible to say that the adsorption efficiency for Pb is at least 75.9% in both series 1 and 2.

Kaiwen Hu[1]
Xingyi Xie[1,2] ,Marta Cerruti[1] and Thomas Szkopek[3]

[1] Department of Mining and Materials Engineering, McGill University, Montreal, Canada.
[2] College of Polymer Science and Engineering, State Key Laboratory of Polymer Materials and Engineering, Sichuan University, Chengdu, Sichuan, China.
[3] Department of Electrical and Computer Engineering, McGill University, Montreal, Canada
Kaiwen.hu@mail.mcgill.ca

Preparation of ultra-light, "shell-less" graphene aerogel via simple pre-reduction method

Reduced graphene oxide (RGO) gels are emerging three dimensional graphene macroscopic assemblies of potential use in many applications including energy storage, pollutant adsorption and gas sensing [1]. Conventional RGO gels are often prepared via solution based gelation involving the reduction of graphene oxide (GO) through chemical or solvothermal reduction. Often, the gel is formed with a shell structure enveloping the interior porous bulk [2]. The conventional gel also shows high volume shrinkage and is thus mechanically rigid, demonstrating severe plastic deformation upon compression. This makes them less attractive to applications such as reusable oil sponges, energy absorbing materials and electromechanical sensors.

In this work, we present a simple pre-reduction technique prior to hydrothermal gelation of GO that allows us to prepare RGO aerogels with ultra-low density and excellent mechanical elasticity. The pre-reduction step also weakens the surface anchoring of GO at interfaces resulting an open structure with the porous bulk structure immediately accessible from the surface [2]. These new features make the pre-reduced RGO aerogel more attractive to the aforementioned applications.

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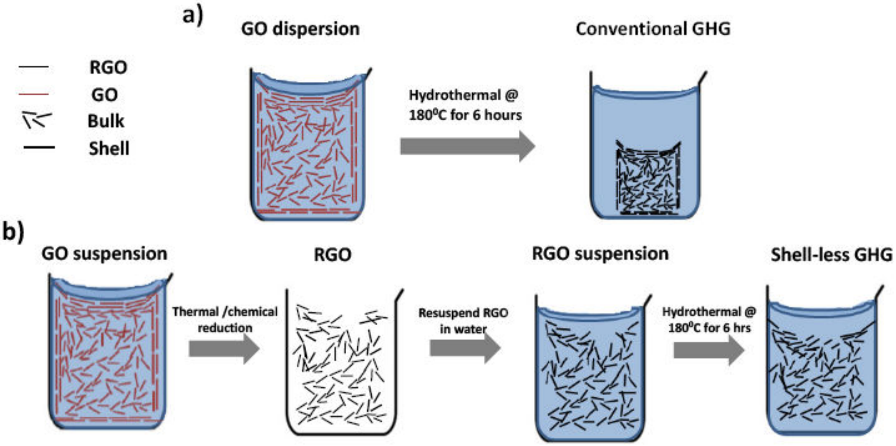
Figures



Scheme 1: Graphene hydrogel (GHG) formation under hydrothermal conditions: a) conventional GHG with shell; b) GHG by thermal or chemical pre-reduction prior to hydrothermal treatment.

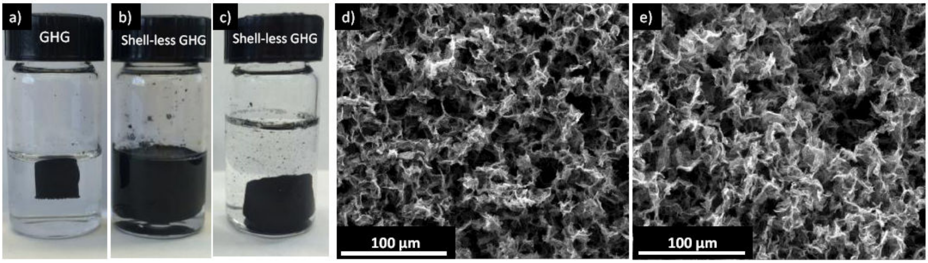


Figure 1: Optical and SEM images of conventional and shell-less GHG. a) Photograph of conventional GHG, b) shell-less GHG prepared via thermal pre-reduction and c) shell-less GHG prepared via chemical pre-reduction using vitamin C. SEM images of the exterior (d) and interior (e) structure of shell-less GHG in b) with similar microstructure.

Toshio Hyodo[1]
Y. Fukaya[2], I. Mochizuki[1], K. Wada[3]

[1] Institute of Materials Structure Science, KEK, Tsukuba 305-0801, Japan
[2] Advanced Science Research Center, JAEA, Tokai 319-195, Japan
[3]Takasaki Advanced Radiation Research Institute, QST, Takasaki 370-1292, Japan
toshio.hyodo@kek.jp

Application of Total-reflection high-energy positron diffraction (TRHEPD) to the analysis of the graphene structures

Total-reflection high-energy positron diffraction (TRHEPD) [1, 2], is the positron (the antiparticle of the electron) counterpart of reflection high-energy electron diffraction (RHEED). Its surface sensitivity originates from the inelastic scatterings, as is the case for the electron diffraction such as low energy electron diffraction (LEED) and reflection high-energy electron diffraction (RHEED), i.e., once a particle undergoes an elastic scattering it loses coherence to contribute the diffraction pattern. In addition to this, TRHEPD has a reason which renders the the method exceedingly surface sensitive, i.e., the electrostatic potential inside every solid is positive and thus the surface is repulsive for a positron. A fast positron incident on a surface with a glancing angle smaller than a certain critical angle is totally reflected. In this condition, the diffraction pattern is determined only by the atoms on the topmost surface [3]. It is also possible to get information on the immediate subsurface by increasing the glancing angle across the critical angle. In this condition the positron is refracted toward the surface in contrast to the electron which is refracted off the surface and penetrates deeper. Thus TRHEPD is best suited for the the determination of the atomic arrangement of the atomic layer materials such as graphene.

A TRHEPD station with a brightness-enhanced intense positron beam is now in operation at the Slow Positron Facility, KEK, where positrons are created via pair creation from the Bremsstrahlung of 50 MeV electron accelerated by an electron linac [4].

We report here recent application of this TRHEPD station to the structure of graphene on a Cu(111) and a Co(0001) surfaces. It was confirmed that the graphene has no buckling and the distance between graphene and the substrate was determined [5]. In a study of silicene on Ag(111), amount of buckling and the distance between the substrate and the bottom silicon atoms were determined [6]. TRHEPD has lso been applied to a more complex surface structure of rutile-TiO2(110)-(1×2) surface [7].

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Figures

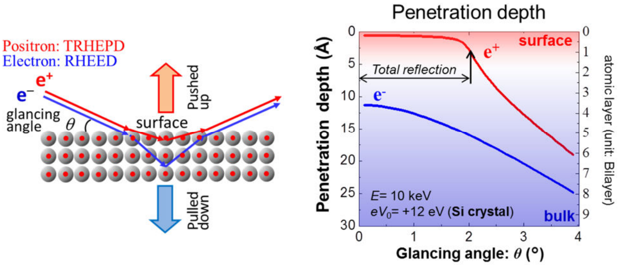

Figure 1: Because of the positive electrostatic potential of every material, the positron is refracted toward the surface, while the electron is refracted off the surface. When the incident glancing angle is smaller than the critical angle, the positron is totally reflected and thus the diffraction pattern is determined solely determin by the arrangement of the topmost atoms.

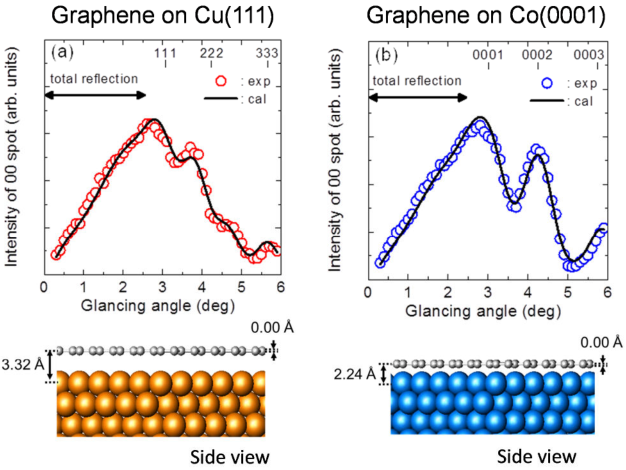

Figure 2: The TRHEPD 00-spot rocking curves for the graphene on Cu(111) and Co(0001) and the obtained structures – amount of bucking and the graphene-substrate distance.

Frédéric Joucken[1]

Yann Tison[2], Patrick Le Fèvre[3], Antonio Tejeda[3], Amina Taleb-Ibrahimi[3], Edward Conrad[4], Vincent Repain[2], Cyril Chacon[2], Amandine Bellec[2], Yann Girard[2], Sylvie Rousset[2], Jacques Ghijsen[1], Robert Sporken[1], Hakim Amara[5], François Ducastelle[5] & Jérôme Lagoute[2]

[1] University of Namur, Namur, Belgium
[2] MPQ lab, Université Paris Diderot – Paris 7, France
[3] Soleil synchrotron, Gif-sur-Yvette, France
[4] GeorgiaTech, Atlanta, USA
[5] CNRS/ONERA, Châtillon, France

frederic.joucken@gmail.com

Correspondence between STM and photoemission results on N-doped graphene

We present a combination of STM and ARPES results obtained on nitrogen-doped graphene. The pristine graphene samples were grown on 4H-SiC(000-1) using the confinement controlled sublimation method [1] and were subsequently doped by exposing them to a nitrogen radical flux produced by RF plasma source.

The electronic doping level was first measured by ARPES at the Cassiopée beamline of the Soleil synchrotron (cf. fig. 1). The same samples were then investigated by LT-STM to evaluate their nitrogen concentration (cf. fig. 1) as well as the electronic doping on a nanometer scale. Core-level photoemission experiments were also performed on the samples.

The combination of these data with tight-binding calculations allowed us to evidence several important effects. First, the number of charge carriers brought by each dopant does not appear to be constant, which, as tight binding results support, questions the reliability of the rigid band model for graphene. Second, the N 1s energy level corresponding to nitrogen in the 'graphitic' configuration depends on the level of nitrogen concentration. Connections between this dependence and the departure from the tight-binding model can be made. Third, the measure of the electronic doping by STS and another technique (ARPES) permits to demonstrate unambiguously the need for renormalization of the STS spectra and strongly supports the interpretation of the pseudogap observed in the tunneling spectra as the absence of an inelastic tunneling channel at low bias [2].

These results, as well as previous related results, are discussed in refs [3] & [4].

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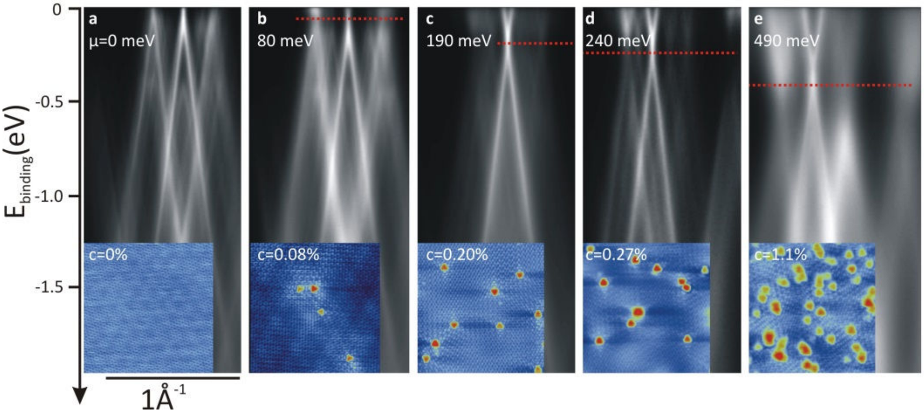

Figure 1: ARPES and typical STM images acquired on samples with different nitrogen concentration. The electronic doping levels (μ) as well as the nitrogen concentrations (c) are given on each image. N dopants appear as red protrusions on the STM images

Catherine Journet
Yangdi Li, Sheng Yuan, Vincent Garnier, Arnaud Brioude, Philippe Steyer, Bérangère Toury

Laboratoire Matériaux Ingénierie et Science, UMR CNRS 5510, INSA de Lyon
Laboratoire des Multimatériaux et Interfaces, CNRS, UMR 5615, Université Lyon 1
Université de Lyon, F-69622, Villeurbanne, France
catherine.journet@univ-lyon1.fr

Large and pure h-BN nanosheets obtained by a unique combination of PDCs route with SPS process

Since graphene's properties are strongly linked to the substrate on which it is deposited, it is clear that the promising future development of practical graphene devices will necessarily go through the development of insulating substrates on which graphene can be deposited without changing its intrinsic properties. One of the most suitable substrates appears to be the hexagonal variety of boron nitride (h-BN, also called “white graphite”), which is isostructural and isoelectronic of graphene, with a lattice matching that of graphene. As a consequence, the development of a novel source for highly crystallized h-BN crystals, suitable for a further exfoliation, is a prime scientific issue. This presentation proposes a promising approach to synthesize pure and well-crystallized h-BN flakes, which can be easily exfoliated into Boron Nitride NanoSheets (BNNSs). This new accessible production process represents a relevant alternative source of supply in response to the increasing need of high quality BNNSs. The synthesis strategy to prepare pure h-BN is based on a unique combination of the Polymer Derived Ceramics (PDCs) route [1] with the Spark Plasma Sintering (SPS) process [2]. Through a multi-scale chemical and structural investigation, it is clearly shown that obtained flakes are large (fig.1), defect-free and well-crystallized, which are key-characteristics for a subsequent exfoliation into relevant BNNSs.

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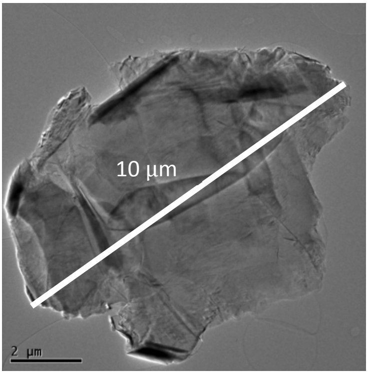

Figure 1: TEM bright field micrograph attesting the large dimension of h-BN flake resulting from the PDCs/SPS dual process

Jasneet Kaur[1]
Alfredo Maria Gravagnuolo[2], Pasquale Maddalena[1], Carlo Altucci[1], Paola Giardina[2] and Felice Gesuele[1]

[1] Department of Physics, University of Naples "Federico II", Naples, Italy
[2] Department of Chemical Sciences, University of Naples "Federico II", Naples, Italy
jasneet.physics@gmail.com

Green route for production and biofunctionalization of transition metal dichalcogenides based nanosheets

Recently, two dimensional (2D) materials including transition metal dichalcogenides (TMDs),such as MoS_2 and WS_2 have triggered the enthusiasm of the research community due to their extraordinary chemical, electronic and optical properties. Moreover, the wide spectrum of their applications in nanoelectronics, nanophotonics, biomedical and sensing devices, has also motivated research on the production of high quality 2D TMDs. Literature reports suggest that these layered materials are produced by various techniques such as mechanical and chemical exfoliation, including liquid phase exfoliation [1], being the most versatile technique for the production of large-scale, few-layered and functionalized 2D TMD based nanosheets.

Herein, we report that during liquid exfoliation of MoS_2 and WS_2 layered crystallites are sonicated in presence of green solvents i.e., water and ethanol and a fungal hydrophobin Vmh_2 [2] is added to retain stability and biological functionalization. Since, Vmh2 protein has unique physicochemical properties and superior hydrophobicity, the addition of this hydrophobin results in stable dispersions of few/monolayered nanosheets, after few steps of controlled centrifugation. Furthermore, controlled centrifugation enables the selection of bio-functionalized few-layered nanosheets of MoS_2 and WS_2, which are investigated by UV-VIS, Raman spectroscopy and atomic force microscopy. The stability of these dispersions are studied by zeta potential measurements, which also confirm the bio-functionalization of nanosheets. Thus, we have introduced an eco-friendly and economic strategy to produce large scale and biofunctionalized TMD based nanosheets.

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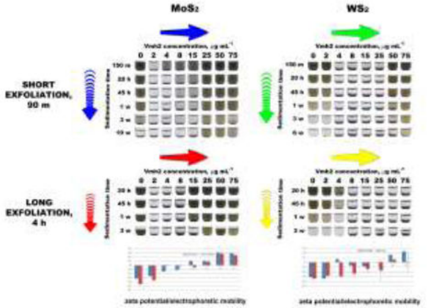

Figure 1: Stability of exfoliated samples in function of time and hydrophobin content

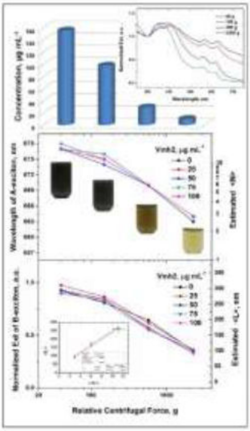

Figure 2: Extinction spectra of MoS_2 liquid dispersions

Neerav Kharche
Vincent Meunier
Department of Physics, Applied Physics, and Astronomy, Rensselaer Polytechnic Institute,
Troy, New York 12180, United States
neerav.kharche@gmail.com

Substrate Screening Induced Renormalization of Excited-States in 2D Materials

Two-dimensional (2D) materials offer an emerging platform for exploring novel electronic phenomena in reduced dimensionality systems. Owing to their atomic scale thickness, the excitation energy levels in 2D materials are strongly renormalized due to the screening by the surrounding environment such as the substrate. This effect is expected to have strong impact when the 2D materials are integrated into functional devices. Accounting for such long-range screening requires methods beyond density functional theory (DFT), such as the GW approach. Large scale GW calculations including the substrate are not feasible using the presently available computational tools.

We have developed an integrated first-principles approach combining density functional theory (DFT), the GW approximation, and a semiclassical image-charge model to compute the electronic band gaps in planar 2D systems in weak interaction with the surrounding environment, including long-range screening effects, in a computationally tractable manner [1]. This methodology is general for any planar 2D system and we apply it here to the specific case of graphene nanoribbons (GNRs) since accurate experimental data exist [2,3].

We find that the band gap of substrate-supported GNRs is reduced by several tenths of an eV compared to their isolated counterparts, with a width-and orientation-dependent renormalization. The predicted band gaps are in good agreement with the range of available experimental data on substrate-supported armchair GNRs (Figure 1). Our results indicate that the band gaps in GNRs can be tuned by controlling the electronic screening at the interface and suggest the interesting possibility of using spatially varying dielectric environment to engineer the screening effects and induce band offsets into 2D materials without any chemical modification.

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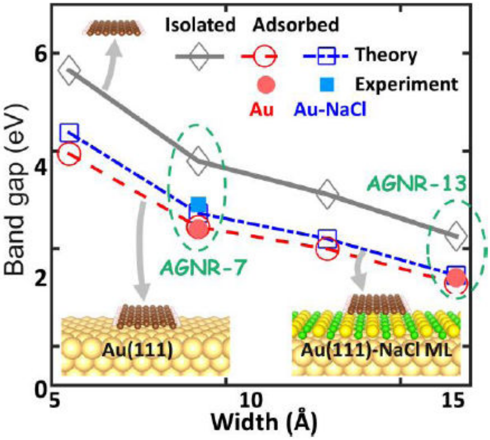

Figure 1: Quasiparticle band gap of the isolated and substrate-supported armchair graphene nanoribbons (AGNRs) as a function of their width. Energy levels in the substrate supported AGNRs are strongly renormalized by the substrate induced screening effect resulting in the reduction of the band gap.

Han Seul Kim
Yong-Hoon Kim*
Graduate School of EEWS, KAIST, 291 Daehak-ro, Yuseong-gu,
Daejeon 305-701, Republic of Korea
*y.h.kim@kaist.ac.kr

Development of a non-equilibrium electronic structure calculation method based on local constrained density functional theory: Applications to vertically-stacked 2D heterostructures

We report on the development of a novel first-principles electronic structure calculation method for the junction systems subjected to non-equilibrium conditions. Within the scheme, non-equilibrium situations induced by external bias voltages are described within the local constrained density functional theory formalism. We will discuss our solutions to the several technical difficulties in describing complicated nanointerfaces placed under bias voltages. As applicaton examples, we consider vertically-stacked van der waals heterostructure based on two-dimensional layered materials. By explicitly extracting the changes in energy level alignment and charge transfer characteristics in response to the source-drain and gate biases, we provide atomistic guidelines for the design of several novel nano-devices.

Sun Jun Kim
Seongchan Jun*

School of mechanical Engineering, Yonsei University, 50 Yonsei-ro,
Seodaemun-Gu, Seoul 120-749, Republic of Korea

*scj@yonsei.ac.kr

Radio frequency transmission characteristics of graphene under low temperature

Graphene, a two dimensional honeycomb lattice of carbon atoms with 0.335 nm thickness, has attracted great attention because of its good electric properties, flexibility and stability [1, 2]. Especially, its superior properties show remarkable potential for radio frequency (RF) applications [3]. Although there were many electronic device researches, these studies based on frequency has not been much progressed in.

In this study, we prepared ground-signal-ground (GSG) devices of graphene interconnectors for RF transmission with two-port measurements in the high frequency range from 2 to 10 GHz. From the investigation of RF transmission, we demonstrated the electrical characteristics according to temperature from 100K to 300K. Furthermore, using telegrapher's equations with equivalent circuit model, parameters such as impedance, resistance (R), inductance (L), shunt conductance (G), and shunt capacitance (C) were extracted from the RF transmission results for demonstrating effect of thermal energy. The result of this study is expected to suggest the achievability of identifying a graphene-based RF transmission device with temperature.

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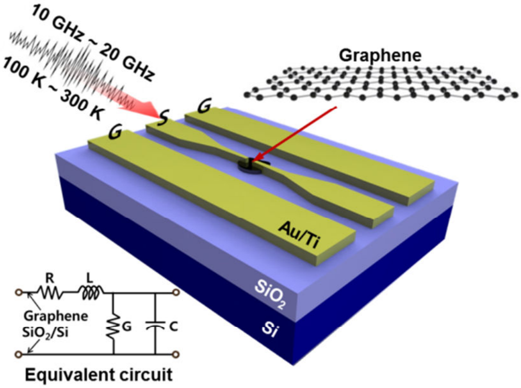
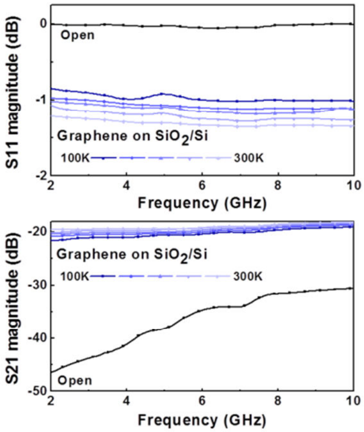

10 GHz ~ 20 GHz
100 K ~ 300 K
Graphene
G
S
G
Au/Ti
SiO2
Si
R
L
Graphene
SiO2/Si
G
C
Equivalent circuit


S11 magnitude (dB)
0
-1
-2
Open
Graphene on SiO2/Si
100K
300K
2
4
6
8
10
Frequency (GHz)
S21 magnitude (dB)
-20
-30
-40
-50
Graphene on SiO2/Si
100K
300K
Open
2
4
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8
10
Frequency (GHz)

Marcus Klein
Martina Griessbach, Richard Kupke
SURAGUS GmbH, Maria-Reiche-Str, 01109 Dresden, Germany
info@suragus.com

Electrical Characterization of Large Area Graphene Layers

The current advances in the research and manufacturing of large area graphene layers are promising towards the introduction of this exciting material in display industry and other electrical and optical applications. New production technologies in the fabrication of flexible displays, touch screens or printed electronics apply graphene layers on non-metal substrates and bring new challenges to the re-quired metrology. Traditional measurement concepts of layer thickness, sheet resistance, optical trans-parency and layer uniformity are difficult to apply to graphene and are often harmful to the product layer. New non-contact sensor concepts are required to adapt to the challenges and even the foreseea-ble inline production of large area graphene. Dedicated non-contact measurement sensors are a pioneering method to leverage these issues in a large variety of applications, while significantly lowering the costs of development and process setup. Transferred and printed graphene layers can be characterized with high accuracy in a huge measure-ment range and even right during production. Large area graphene mappings are applied for process optimization and for efficient quality control. Examples of doped, defected and excellent Graphene are presented as quality images and implications for manufacturer are explained.

L.Kouchachvili

CanmetENERGY, Natural Resources Canada
1 Haanel Dr. Ottawa, ON,K1A 1M1, Canada

lia.kouchachvili@canada.ca

Ag/Biochar Composite for Supercapacitor Electrodes

The development of new electrical energy storage devices and the improving of existing ones e.g. batteries and supercapacitors (SC) has become extremely important due to increased application of these devices in transportation and in renewable energy storage. Among them, SC are more capable of generating higher power density than are batteries, and are gaining particular interest. They are used for recovering brake energy in electrical vehicles and locomotives and for enhancing the energy efficiency of instruments requiring a peak power source. They can eliminate idling and consequently reduce fuel consumption, emission of harmful pollutants and maintenance cost and prolong engine life. However, SC store significantly less energy than batteries. One way to increase the energy storage capacity of SC without compromising their cost is to enhance the performance of electrodes. Low-cost and high-performance materials for the electrodes have been extensively investigated. The most utilised material for SC electrodes is carbon (from different sources and in different forms). Canada is rich in biomass, which is one of the potential sources of carbon for these applications. Biochar (BC) from woody residue has unique pore morphology. Its potential as a carbon source for the SC electrodes has been reported in many recent studies [1-5]. It has been observed that the physical and electrochemical properties of the BC highly depend on the source of biomass, its carbonization conditions and any subsequent activation methods. One of the key criteria for utilizing activated BC (a-BC) as an electrode material is its electrical conductivity. In this work, a potential method has been proposed for the impregnation of Ag ions into the a-BC to prepare a composite material with high electrical conductivity.

The results obtained from electrochemical measurements showed that electrodes composed of Ag/(a-BC) have higher electrical conductivity and better performance compared to a-BC based electrodes (Fig.1). The highest specific capacitance of 475 F g^{-1} in aqueous 3M $LiN((SO_2CF_3)_2)$ electrolyte was achieved which is almost 42% higher than the previously reported value (335 F g^{-1}) for electrodes composed with of a-BC [1].

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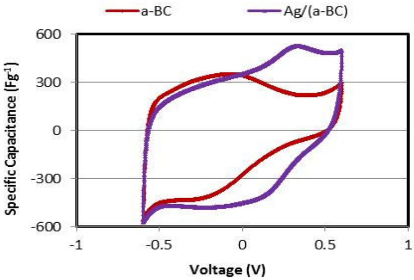

Figure 1: CV curves at 5mV s-1 scan rate

Jens Kroeger
Nicholas Larouche, Jefford Humes

Raymor Industries
3765 La Vérendrye, Boisbriand, QC, J7H 1R8, Canada

Continuous gas-phase plasma process to produce few-layer crumpled graphene

Raymor is industrializing a continuous gas-phase plasma process to produce few-layer crumpled graphene sheets at commercial scale (few hundred grams per hour) using a 60 kW plasma torch. Process optimization has allowed us to produce a material having an outstanding high crystallinity and purity.

Raymor's patent protected plasma production process is continuous, robust, and low-cost compared to other manufacturing methods. The material presents low oxygen content (1-2%) and ppm metal impurity levels, suitable for battery applications.

The turbostratic layering and 'crinkled' 3-D morphology of this graphene is unique, and provides superior dispersability in a variety of solvents. Efforts toward process stability, product quality and product standardization will be reviewed. Characterization of the material is routinely done using Raman spectroscopy and nitrogen adsorption isotherm (surface area): plasma-grown graphene presents a G/D ratio up to 4 at 514 nm excitation wavelength along with a surface area greater than 400 m^2/g. Advantages of utilizing this material in energy storage and conductive ink applications will be discussed.

E. Lago
P.S. Toth, V. Pellegrini, and F. Bonaccorso
Graphene Labs - Istituto Italiano di Tecnologia, Via Morego 30, 16163, Genoa, Italy
emanuele.lago@iit.it

Effects of 2D crystals addition on the physical properties of polycarbonate-based composite

Two-dimensional (2D) crystals-based applications are gaining increasing attention in the past few years [1]. The current bottleneck hindering the full exploitation of the 2D crystal properties relies on their production strategies. In this context, liquid phase exfoliation (LPE) of layered materials [2,3] is the most promising approach for the scalable production of 2D crystals. In fact, by LPE it is possible to realize dispersions and inks, to be deposited on target substrates, as well as polymer composites [4]. In particular, 2D-crystals-based composites have shown improvements in mechanical, electrical and thermal properties compared to pristine polymer matrices [1,4]. Unfortunately, the impact of both 2D filler nature and morphology, i.e. lateral size and thickness, on the composite properties is still poorly understood, thus an in-depth study is urgently needed for further optimization.

Herein, we select polycarbonate (PC) as polymer for the production of both graphene- and hexagonal boron nitride (hBN)-doped composites, prepared by solvent blending technique. The 2D crystals addition to the polymeric matrix is evaluated by investigating the mechanical properties of the composites.

Exfoliation of bulk layered material is carried out by ultra-sonication [5]. The solvent used for graphene is N-Methyl-2-pyrrolidone, whereas for h-BN we used a novel water/surfactant solution. 2D-based crystals are then sorted by lateral size and thickness by using ultra-centrifugation [6-8]. The obtained graphene flakes of average lateral size of ~300 nm are obtained by ultra-centrifugation at 17000g; a cascade centrifugation [9] was developed in order to separate hBN flakes in two different dispersions with average lateral sizes of 200 and 400 nm, respectively. After the ultra-centrifugation, a solvent exchange process is used for the re-dispersion of the graphene and hBN flakes in 1,3-dioxolane, the same solvent used for the dissolution of PC pellets. 2D crystals and PC dispersions are mixed by means of ultra-sonication allowing a thorough mixing of flakes and PC polymer chains. The dispersions are then pelletized by pouring water, a non-solvent for the PC, which precipitates the composite materials. Finally, the pellets of composite are hot-pressed to form ~100 µm thick composite films.

Graphene/PC and hBN/PC composites are characterized morphologically and structurally using atomic force and scanning electron microscopies, and Raman spectroscopy. Static and dynamic mechanical measurements are carried out to investigate the effect of low loading (≤1%) of the two different 2D crystal fillers in PC matrix. We found an improvement of more than 25% in Young's modulus with respect to the neat polymer at 1 wt.% and 0.5 wt% of loading of graphene and hBN, respectively. Thus, hBN flakes provided the same improvement of graphene flakes with only half of the loading, highlighting how the differences in structure and morphology of 2D-crystals-based fillers play a key role in nanocomposite science and technology.

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A. Latgé[1]
R. Carrillo-Bastos[2], C. León[1], D. Faria[3], E. Y. Andrei[4], and N. Sandler[5]

[1] Instituto de Física, Univ. Federal Fluminense, Av. Litorânea sn, Niterói-Rio de Janeiro, Brazil
[2] Faculdad de Ciencias, Universidad Autónoma de Baja California, Ensenada, Baja California, México
[3] Instituto Politécnico, Univ. do Estado do Rio de Janeiro, Nova Friburgo, Rio de Janeiro, Brazil
[4] Department of Physics and Astronomy, Rutgers University, Piscataway, New Jersey 08855, USA
[5] Department of Physics and Astronomy, Ohio University, Athens, Ohio, USA
andrea.latge@gmail.com

Fold assisted transport in graphene systems

Deformations in graphene are central elements in the novel field of “straintronics”. Various strain geometries have been to produce specific properties but their experimental realization has been limited. Because folds occur naturally in graphene samples, or could be engineered with appropriate substrates, we study their effects on graphene transport properties [1]. We show the existence of an enhanced local density of states (LDOS) along the fold that originates from localization of higher energy states, and provides extra conductance channels at lower energies. In addition to exhibit sublattice symmetry breaking, these states are valley polarized, with quasi-ballistic properties in smooth disorder potentials. We confirmed that these results persist in the presence of strong edge disorder, making folds viable electronic waveguides. These findings could be tested in currently available experimental settings.

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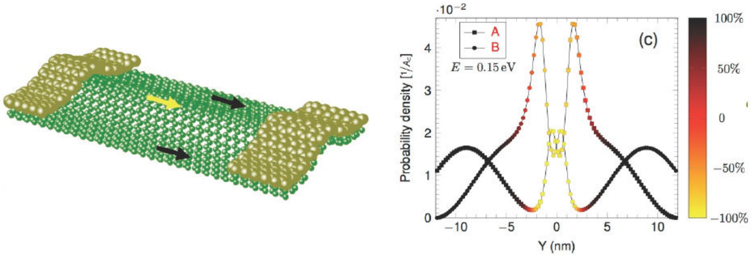
Figures



Figures1: (a) Schematic representation of a graphene nanoribbon with a Gaussian fold deformation. (b) Total LDOS for states with same velocity near both K and K’ valleys. Color scale represents percentual contribution from each valley; yellow refers to states near K and black to states near K’. Notice that arrows in (a) represent valley polarized current along the ribbon, with colors defined as in (b).

Pierre L. Levesque[1]

Saman Choubak[2], Carl-Bernard Charpin[1], Dhan Cardinal[2], Philippe Gagnon[2], Minh Nguyen[1], Patrick Desjardins[2], Richard Martel[1]

[1] Regroupement Québécois sur les Matériaux de Pointe (RQMP) and Département de Chimie, Université de Montréal, C.P. 6128, Succursale Centre-Ville, Montréal, Québec H3C 3J7, Canada
[2] Regroupement Québécois sur les Matériaux de Pointe (RQMP) and Département de Génie Physique, Polytechnique Montréal, Montréal, Succursale Centre-ville, Montréal, Québec, Canada
p.levesque@umontreal.ca

A new knob in graphene CVD growth

Several milestones have been achieved for the chemical vapor deposition (CVD) of graphene films on copper foils since it was first reported back in 2009 and the technology is now mature enough so companies are developing tools for mass productions. Still, continuous monolayer coverage of graphene on copper requires surprisingly long growth times and several inconsistencies remains to be elucidated in order to scale the production and reach the constancy and uniformity of the resulting material as required by the industry.

Our recent research revealed oxidizing impurities as a dominant factor governing the growth. For instance, oxygen impurities and not hydrogen, are responsible for graphene etching on copper and hydrogen have the protecting role of counter balancing oxidizing impurities reactions. When the level of oxidizing impurities are minimized, continuous layer of graphene can be grown in the sole presence of methane [1, 2]. The kinetics of the reactions have been addressed and revealed a competitive action between precursor oxidation and carbon growth during graphene formation in the CVD reactor. Based on our experimental evidences and the kinetic model developed, a criterion is found for the O_2/H_2 partial pressure ratio setting a limit between impurities limited growth and methane adsorption-dissociation limited growth. Most of the work to date, even using ultra-high purity (UHP) gases, are in the impurities limited growth regime. In this presentation, the adsorption-dissociation limited growth regime is explored in a series of growth experiments where the level of oxidizing impurities is strictly controlled. With this new control knob, the growth can be speeded up by a factor of x50 compared to recipes under UHP conditions without affecting graphene quality. The purified regime also allowed us to identify an origin of bilayer islands and gave us some insight on the copper surface quality effects on the resulting graphene morphology.

Studies in purified conditions allow the necessary fundamental knowledge to be gained in order to tailor the graphene growth and gives the consistency required in a manufacturing context. In addition, the growth method shown in this report is straightforward and very simple to implement in industrial manufacturing processes making graphene sheets mass production economically feasible.

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Wenwen Liu
Aiping Yu*, Jingde Li, Lathankan Rasenthiram, Kun Feng, Yangshuai Liu, Lucas Lim, Gregory Lui, Ricky Tjandra, Gordon Chiu

Department of Chemical Engineering, Waterloo Institute for Nanotechnology, University of Waterloo, Waterloo, Ontario N2L3G1, Canada
*aipingyu@uwaterloo.ca

Construction of novel high-performance Li-ion hybrid supercapacitors based on 3D graphene composites

Li-ion hybrid capacitors (LIHCs) as a novel and promising energy storage unit, consisting of a Li-ion battery type anode and an electrochemical double layer capacitance (EDLC) type cathode, has attracted enormous research attentions [1-3]. However, it remains a significant challenge to build the high-performance LIHCs due to most battery-type anodes with the sluggish kinetics of Li-ion storage and low specific capacitance of common capacitor-type cathodes [4, 5]. Thus, a search of alternative high-performance battery type anode materials and capacitor-type cathode materials, able to deliver higher energies and power densities in organic electrolyte is highly required. Herein, a novel high-performance LIHCs is constructed by combining a Li-ion battery type anode (3D graphene/MoO_3 nanocomposite) with a capacitor type cathode (3D graphene/PANI derived carbon material). Such a subtle design endow LIHCs with a wide voltage range of (3.8 V), a high energy density of 128.3 Wh·kg^{-1}, and an ultra-long cycle life up to 3000 cycles with 87% capacity retention.

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Figures

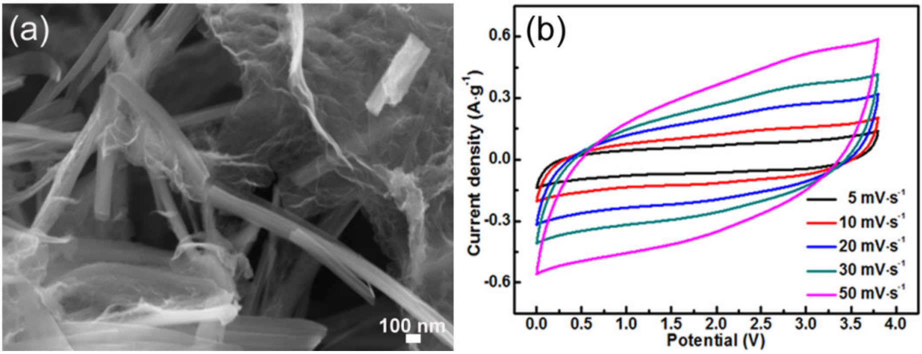

Figure 1: (a) SEM image of 3D graphene/MoO_3 nanocomposite, (b) CV test of LIHSs device based on 3D MoO_3/GNSs and 3D PANI/GNSs derived carbon at various scan rates.

Yangshuai Liu
Lathankan Rasenthiram, Aiping Yu*

Department of Chemical Engineering, University of Waterloo
200 University Avenue West, Waterloo, Ontario, N2L 3G1, Canada
*aipingyu@uwaterloo.ca

Fabrication of Graphene-Based Composites for Wearable Supercapacitors

Graphene fiber (GF) has arisen as a widely used material in both academia and industry due to its mechanical flexibility. GF has significant value because it permits the integration of rich functionalities and high performances. Many efforts were made to apply graphene fibers (GFs) to wearable energy storage devices such as supercapacitors. However, the utilization of 2-D graphene sheets presents difficulties in assembling GF with strong mechanical strength. Moreover, current methods of fabricating supercapacitor device by twisting two individual GF together could easily result in short circuit or high solution resistance. Thus a more integrated GF supercapacitor that does not compromise electrochemical performance is strongly required [1-3]. Herein we propose a novel all-in-one GF supercapacitor device based on spun graphene fibers. The proposed GF supercapacitor has strong mechanical strength and improved capacitive performance due to the utilization of giant graphene sheets and introduction of Ppy. This novel method also overcomes the high solution resistance of the conventional closely-packed GO sheets. Therefore the proposed novel all-in-one GF method holds great technological promise as an improved mean for constructing super capacitor devices.

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Gregory Lopinski
Timothy Knight, Zygmunt Jakubek, Chad Sweeting, Dusan Vobornik, Stephen Wei, Shan Zou
Measurement Science and Standards, National Research Council,
100 Sussex Dr., Ottawa, Canada
gregory.lopinski@nrc-cnrc.gc.ca

Characterization of solution processable graphene

NRC's Nanocarbon Metrology project aims to develop characterization methods and protocols for the assessment of single wall carbon nanotubes and graphene. As these materials move from the laboratory to production and into applications, there is a need for improved characterization methods and development of standard protocols to ensure material quality. Variable quality of commercially available materials and lack of proper methods and protocols to evaluate these materials is hampering the development of applications.

Many applications (i.e. printable electronics, batteries, graphene-polymer composites,…) require graphene in solution. In this case the graphene is in the form of flakes dispersed in organic or aqueous solvents. Solution processable routes to graphene generally fall into two categories- direct exfoliation of graphite or oxidation to graphene oxide (GO) followed by reduction to graphene. Properties of films made from these dispersions are dependent on both the quality of the individual flakes and how they are assembled into films. Here we will present results on evaluating the structure, morphology and electronic properties of reduced GO and graphene films derived from various protocols using starting materials from a variety of commercial sources. Size of individual flakes in solution have been measured by dynamic light scattering and compared with atomic force microscopy (AFM) measurements after deposition on a substrate. AFM height measurements have been used to determine flake thickness and monitor the thermal reduction of GO to graphene. Scanning tunneling microscopy has been used to observe the atomic scale structure of the flakes. AFM and Raman spectroscopy have been used to characterize the morphology and uniformity of continuous films assembled from these flakes. These observations are correlated with measurements of the conductivity and work function. The structure and electronic properties of films derived from thermal and/or chemical reduction of GO are compared with those based on dispersions of directly exfoliated graphene flakes.

Farzaneh Mahvash[1,2]
Thomas Szkopek[1], Mohamed Siaj[2]

[1] McGill University, 3480 University Street, Montréal, Quebec, Canada
[2] Université du Québec à Montréal, 2101 Jeanne-Mance Street, Montréal, Quebec, Canada
farzaneh.mahvash@mail.mcgill.ca

Charge transport and corrosion inhibiting properties of monolayer hexagonal boron nitride grown by chemical vapor deposition

Hexagonal boron nitride (h-BN), also known as white graphite, is a wide bandgap material that has found use as an appealing insulating dielectric layer in ultra-high mobility graphene devices, 2-dimensional heterostructures and tunneling devices [1–4]. We report the chemical vapor deposition (CVD) growth and characterization of monolayer h-BN. The growth was performed in a tube furnace on Cu foils using an ammonia borane (NH_3-BH_3) precursor. Scanning electron microscopy was used to study the morphology of the as-grown films and optimize growth conditions to yield high coverage of monolayer h-BN. Chemical analysis was performed by electron energy loss spectroscopy and X-ray photoelectron spectroscopy. The hexagonal crystal structure was investigated by electron diffraction of suspended films. Raman spectroscopy of h-BN transferred to SiO_2/Si substrates reveals a prominent Stokes shift at 1366-1370 cm^{-1}. The optical properties of our h-BN films were probed by cathodoluminescence and UV-Vis absorption spectroscopy. In-plane electron transport studies were performed on h-BN monolayer transferred to SiO_2/Si substrates using a variety of electrode geometries. Ni electrodes were used to provide electrical contact. We have observed quadratic scaling of current with voltage (Figure 1), consistent with space-charge limited transport with a mobility of up to ~ 0.01 cm^2/Vs [5]. Our observation of in-plane charge transport suggests that h-BN can function as a semiconductor with appropriately chosen contact electrodes, as is the case with h-BN nanotubes.

Moreover, the suitability of monolayer h-BN for inhibiting corrosion has been investigated. We report the quantitative measurements of monolayer h-BN as a Cu corrosion inhibitor by use of cyclic voltammetry (Figure 2) and Tafel analysis. We have found that CVD grown h-BN reduces the Cu corrosion rate by one order of magnitude compared to bare Cu, suggesting that this ultrathin layer can be employed as an atomically thin corrosion-inhibition coating.

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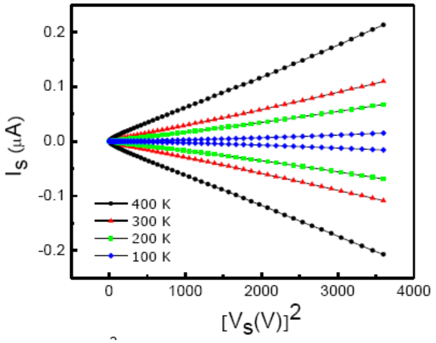

Figure 1: I_S-V_S^2 at various temperatures in vacuum (2×10^{-6} Torr) for a 8 μm channel length monolayer h-BN device.

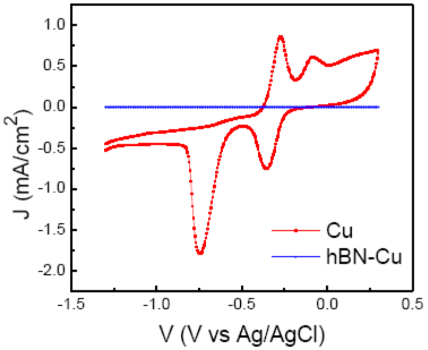

Figure 2: Cyclic voltammetry measurements for a 0.07 cm^2 area bare Cu (red) and h-BN-Cu (blue) in a 0.1 M NaOH solution.

Sana Mzali
Alberto Montanaro, Odile Bezencenet, Jean-Paul Mazellier, Bruno Dlubak, Marie-Blandine Martin, Pierre Seneor, Pierre Legagneux
Thales Research & Technology, 1 Avenue Augustin Fresnel, 91767 Palaiseau, France
sana.mzali@polytechnique.edu

Statistical study of graphene field effect transistors (GFETs) performance

Graphene has gained increasing attention over the last decade, due to its outstanding properties closely linked to its 2D material nature [1]. In particular, the potential of graphene for optoelectronic applications is currently being extensively explored because of its ultra-high carrier mobility and absorption from the far infrared to the ultraviolet [2,3]. However, to construct high-quality optoelectronic devices, it is necessary to accurately control graphene doping and to operate highly stable devices.

For that purpose, a statistical study was performed on more than 500 unsorted graphene field effect transistors (GFETs). The graphene films were grown on copper by chemical vapor deposition (CVD) and transferred on 300 nm SiO_2/Si substrates. The fabrication process includes, as a first step, the deposition of a protection layer (thin oxidized aluminum layer) after graphene transfer. Then, after device fabrication, we have deposited metal oxides layers, namely passivation layers, by atomic layer deposition, to passivate our CVD graphene based transistors.

We have statistically demonstrated the impact of each layer on our GFETs performance by comparing 3 different fabrication processes: without any protection/passivation layers, with only the protection layer and with both protection/passivation layers (Fig. 1). While we do not observe any conductance minimum with unprotected and unpassivated devices, 75% of the protected/passivated GFETs and 58% of only protected devices exhibit a conductance minimum for a gate voltage (V_{min}) below 50 V (Fig. 2).

Even more remarkable, we found out that 40% of protected and passivated devices exhibit a V_{min} below 10 V. However, only 3% of protected devices display a conductance minimum in this small window. We also achieved successful hysteresis free DC characteristics on about one-quarter of our protected/passivated GFETs.

The analysis of all these statistical results highlights the importance of the use of both protection and passivation layers to fabricate low-doped graphene devices with minimum hysteresis and long-term stability.

This work was funded through the European projects Grafol and Graphene Flagship.

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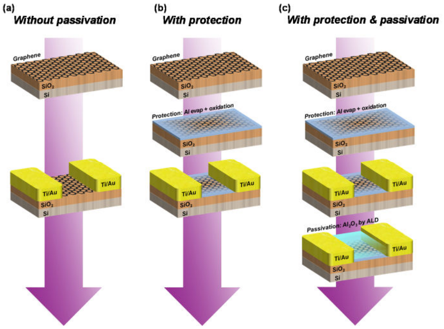

Figure 1: Process Flows of graphene devices fabrication on 300 nm SiO_2/Si substrates without any protection/passivation layers (a), with protection process (b) and with protection/passivation process (c).

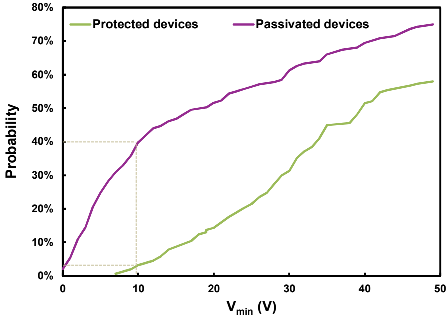

Figure 2: Cumulative distribution curves of working GFETs with protection layer (green curve) and with protection/passivation layers (purple curve) as a function of V_{min} (gate voltage at minimum conductance).

Jae Young Park
Juyeong Oh, Seong Chan Jun*
School of Mechanical Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-gu, Seoul, Korea
*scj@yonsei.ac.kr

Radio-frequency transmission of few-layer WSe_2 nanosheet Coplanar waveguide

For a decade, many researches regarding graphene, which has metallic two-dimensional structure, have been explored. However, there are a lot of limits in the field of nanoscale electronics because of its zero bandgap. Transition Metal Dichalcogenides (TMDc) has been spotlighted as a next-generation nanomaterial due to its existence of bandgap and its good electrical and mechanical characteristics [1]. Even though TMDc materials shows various electronic properties [2-3], there is no demonstration with high frequency transmission yet.

In this research, We confirmed WSe_2, one of TMDc can be a good candidate for an radio-frequency interconnector. To fabricate the device, WSe_2 nanosheet was mechanically exfoliated on highly resistive Si/SiO_2 substrate. Also, gold pattern was deposited for using ground-signal-ground (GSG) probe method. We demonstrate high frequency characteristics of WSe_2 in range of 0.5–40 GHz. We measured Scattering parameters, and extracted equivalent properties including impedance, resistance to understand the transport conductivity of WSe_2. The results of impedance and resistance within the frequency region through WSe_2 show negligible degradation. Those results show that WSe_2 has high potential for transmitting signals at gigahertz ranges although it has large bandgap.

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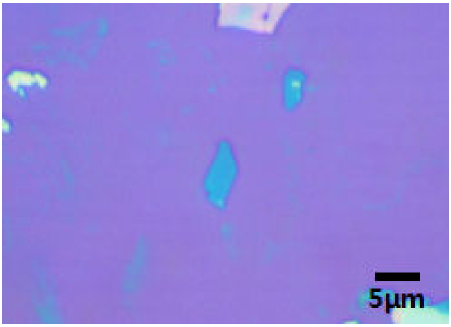
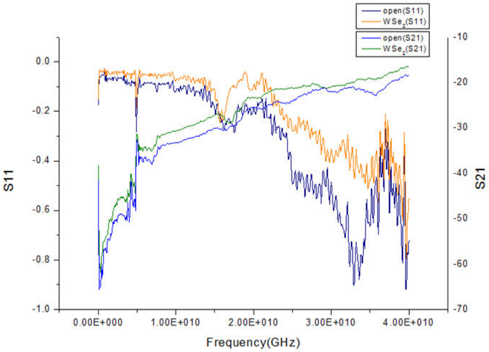

5µm


open(S11)
WSe$_2$(S11)
open(S21)
WSe$_2$(S21)
S11
0.0
-0.2
-0.4
-0.6
-0.8
-1.0
S21
-10
-20
-30
-40
-50
-60
-70
0.00E+000
1.00E+010
2.00E+010
3.00E+010
4.00E+010
Frequency(GHz)

P. Pochet[1,2]
B. McGuigan[3] , A. V. Tyurnina [1,4], H. T. Johnson[3], J. Dijon [1,4] and H. Okuno[1,2]

[1] Univ. Grenoble Alpes, Grenoble France
[2] CEA INAC 38054 Cedex 9,Grenoble France
[3] MechSE, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States
[4] CEA LITEN DTNM, 38054 Cedex 9,Grenoble France
Pascal.Pochet@cea.fr

Dislocations in 2D: blessing or curse?

Dislocations in 2D systems were first observed several decades ago in the famous bubble raft experiment [1]. Currently, 2D materials like graphene, h-BN, and transition metal dichalcogenides offer a rich variety of problems for theory, with some key potential applications. Indeed dislocations are always present in materials and are usually undesirable because they can alter the electronic properties of the target material. A classical way to approach dislocations in 3D heterostructures is found in semiconductor science, where the presence of misfit dislocations can be avoided using the critical thickness framework introduce by Matthews and Blakeslee [2]. Here we recast this classic framework to consider interface misfit dislocation formation in 2D in-plane heterostructures [3]. We consider graphene-h-BN interfaces with various dislocation core reconstructions. This makes it possible to reveal a design space where defect-free heterostructures can be grown (see figure below). In other cases, dislocations might also be desirable, as they are building blocks for strengthening of materials through pile-ups that form grain boundaries. We will use the related framework of recrystallization to propose a way to tailor the properties of graphene. Here, using a modified hot-filament-assisted Chemical Vapor Deposition setup, which does not require any control of crystal nucleation and orientation. We propose an original growth mechanism [4] that includes a stage of structural evolution from nanocrystalline to microcrystalline graphene film opening a new route to control the electrical properties of large-scale uniform graphene film.

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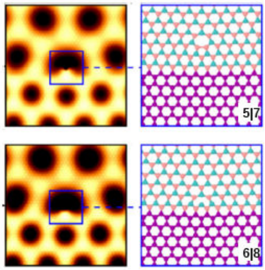
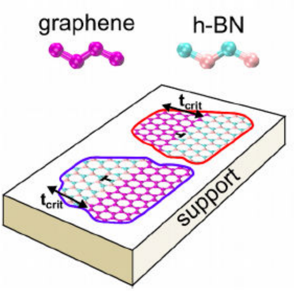
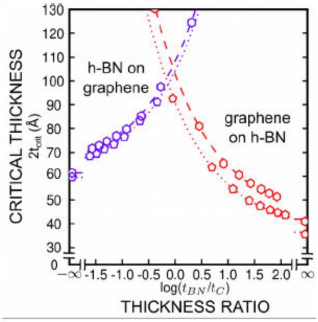

5|7
6|8


graphene
h-BN
t_{crit}
t_{crit}
support


CRITICAL THICKNESS
$2t_{crit}$ (Å)
130
120
110
100
90
80
70
60
50
40
30
0
h-BN on graphene
graphene on h-BN
$-\infty$
-1.5
-1.0
-0.5
0.0
0.5
1.0
1.5
2.0
∞
$\log(t_{BN}/t_C)$
THICKNESS RATIO

Francesco Presel[1]
Eduardo Hernandez[2], Paolo Lacovig[3], Silvano Lizzit[3], Dario Alfè[4], Alessandro Baraldi[1,3]

[1] University of Trieste, Via Valerio 2, Trieste, Italy
[2] Instituto de Ciencia de Materiales de Madrid,Campus de Cantoblanco (UAM), Madrid, Spain
[3] Elettra - Sincrotrone Trieste, S.S. 14 km 163.5, Trieste, Italy
[4] University College London, Gower Street, London, UK
francesco.presel@phd.units.it

Interaction of small molecules with epitaxial graphene

A full understanding of the nature and dynamics of interactions between graphene (GR) and gas molecules is very important for technological applications of GR: in fact, the electronic properties of GR – such as its doping – can be modified even by a very small quantity of adsorbates, including most gasses commonly found in the atmosphere [1], potentially reducing the efficiency of GR-based devices.

This interaction is dominated by van der Waals forces, weaker than chemical bonds, yet characterized by a a wider range of interaction: therefore, an accurate determination of the interaction between GR and gas molecules can not neglect the substrate over which GR is supported [2,3]. Since GR often forms moiré structures on metallic surfaces (containing hundreds of atoms per unit cell), numeric computations of such systems are very demanding, usually based on Density Functional Theory (DFT) implementations of van der Waals forces, which however have often provided contradictory results [4,5].

In order to benchmark these calculations and improve their reliability, we have performed an experimental study of an ideal system, consisting of CO molecules adsorbed on GR supported on Ir(111), combined with DFT numerical calculations on the same system. This experimental work combines complementary state-of-the-art experimental techniques such as NEXAFS, LEED and time-resolved high energy resolution core level photoelectron spectroscopy (TR-XPS) to obtain quantitative information on the adsorption geometry and energy of CO on GR and compare them to theoretical predictions. In particular, we have found that CO molecules form commensurate long-range-ordered structures on the GR moiré cell (Fig. 1a), and they lie with their axis almost parallel to the surface. From TR-XPS spectra we were able to obtain accurate desorption curves for a full CO layer as well as for coverages as low as few percent of Monolayer (Fig. 1b), from which we calculated the desorption energy with an accuracy of few meV. These results are in good agreement with theoretical calculations (Fig. 1c).

To understand the role of the substrate on CO adsorption on GR, we modified its electronic structure by intercalating cobalt below GR; moreover, we repeated the theoretical calculations after removing the substrate. Both new systems showed a variation in CO adsorption energy of tens of meV, i.e. around 10% of the adsorption energy, showing that this quantity increases for increasing GR-substrate interaction (Fig. 1b).

In this experiment, therefore, we have acquired very high quality experimental data and used them to verify the accuracy of DFT calculations implementing van der Waals forces. Moreover, this experiment shows that the contribution of the substrate on the van der Waals interactions of GR with adsorbates is significant and must be taken into account to obtain accurate results from computations.

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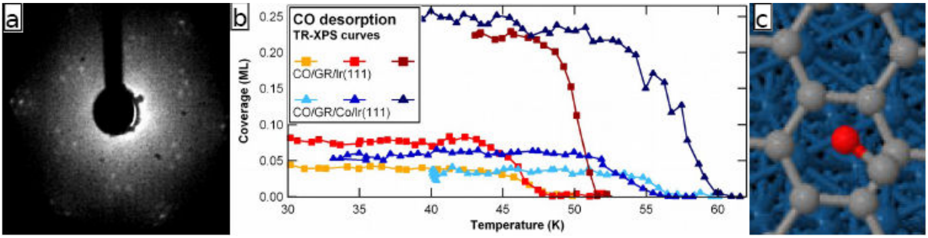

Figure 1: a) LEED pattern of CO/GR/Ir(111); b) Desorption curves of CO from GR/Ir(111) and GR/Co/Ir(111) for different initial coverages; c) Calculated minimum energy configuration of CO/GR/Ir(111).

Antonio Rossi[1,2]

Holger Büch[1], Carmine Di Rienzo[1,2], Vaidotas Miseikis[1], Domenica Convertino[1,2], Ameer Al-Temimy[1], Valerio Voliani[1], Mauro Gemmi[1], Vincenzo Piazza[1] and Camilla Coletti[1,3]

[1] Center for Nanotechnology Innovation @NEST, Istituto Italiano di Tecnologia, Pisa, Italy
[2] NEST-Scuola Normale Superiore, Pisa, Italy
[3] Graphene Labs, Istituto Italiano di Tecnologia, Genova, Italy

antonio.rossi@sns.it

Scalable synthesis of WS_2 on graphene and h-BN: an all-2D platform for light-matter transduction

By exhibiting a measurable bandgap and exotic valley physics, atomically-thick tungsten disulfide (WS_2) offers exciting prospects for optoelectronic applications. The synthesis of continuous WS2 films on other two-dimensional (2D) materials would greatly facilitate the implementation of novel all-2D photoactive devices [1-3]. We have demonstrated the scalable growth of WS_2 on graphene and hexagonal boron nitride (h-BN) via a chemical vapor deposition (CVD) approach. Spectroscopic and microscopic analysis reveal that the film is bilayer-thick, with local monolayer inclusions. Photoluminescence measurements show a remarkable conservation of polarization at room temperature peaking 74% (Fig. 1a-b) for the entire WS_2 film. Furthermore, we present a scalable approach for the design of photoconductive and photoemitting patterns. By adopting epitaxial graphene on SiC as growth substrate, one can define in a bottom-up fashion photoemitting and photoconducting ribbons (Fig. 1c). The scalable synthesis and design on 2D substrates of WS_2 films with outstanding optical properties is instrumental in the development of novel all-2D quantum optoelectronic and valleytronic devices.

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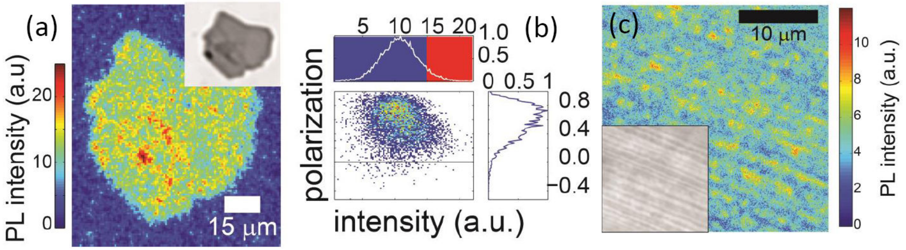

Figure 1: (a) PL intensity map of a selected h-BN flake (also shown in the optical micrograph in the inset) (b) PL polarization vs. PL intensity 2D histogram. The polarization peaks at ~74%. (c) PL intensity map taken from the area shown in the inset. The emission in stronger where WS_2 is on top of buffer layer regions.

Mohamed Siaj
Ricardo Izquierdo and Patrick P. Brisebois
Department of Chemistry, UQAM, Montreal, Quebec, Canada
siaj.mohamed@uqam.ca

New Insights into Graphene Oxide Chemistry

Graphene oxide is regarded as a major precursor for graphene-based materials. The development of graphene oxide based derivatives with new functionalities requires a thorough understanding of its chemical reactivity, especially for canonical synthetic methods such as the Diels–Alder cycloaddition. For the first part of the talk, we will show that the Diels–Alder reaction has been successfully extended with graphene oxide as a source of dienes by using maleic anhydride as a dienophile, thereby outlining the presence of the cis diene present in the graphene oxide framework. We found that this reaction could provide fundamental information for understanding the exact structure and chemical nature of graphene oxide. On the basis of high-resolution ^{13}C-SS NMR spectra, we will show evidence for the formation of new sp^3 carbon centers covalently bonded to graphene oxide following hydrolysis of the reaction product. We used DFT calculations to show that the presence of a cis dihydroxyl and C vacancy on the surface of graphene oxide are promoting the reaction with significant negative reaction enthalpies [1]. For the second part of the talk, a gram-scale synthesis of graphene oxide-based metal oxide nanocomposites with tunable fraction of reduced carbon will be discussed. Graphene oxide-based metal oxide nanoparticles hybrid materials with tunable fraction of reduced carbon were synthetized via a simple one-step reaction in aqueous solution at room temperature using water soluble metal halides such as stannous chloride, ferrous chloride, zinc chloride, aluminum chloride and magnesium chloride as a cheap, abundant, nontoxic and green source of reductants. The obtained graphene oxide-based materials have a more important fraction of reduced carbon when metallic ions with higher standard reduction potential such as Sn^{2+} (E_0=-0.14 V) and Fe^{2+} (E_0=-0.44 V) were used for the reduction. According to our observations, spontaneous formation of metal oxide NPs occurs on the surface of GO without damaging the integrity of its framework. NMR integration showed that GO is only slightly reduced during the transformation and the method allowed GO-MO composites preparation with a tunable fraction of reduced carbon ranging from 26-75%.

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Ricky Tjandra
Gregory Lui, Alicia Veilleux, Josh Broughton, Gordon Chui, Aiping Yu

Department of Chemical Engineering, Waterloo Institute for Nanotechnology, Waterloo Institute for Sustainable Energy, University of Waterloo, 200 University Avenue West, Waterloo, Canada
Aipingyu@uwaterloo.ca

Enhanced Binding of Reduced Graphene Oxide to Polyurethane Sponge for Oil Absorption

In this work [1], we have shown increased binding of reduced graphene oxide onto the surface of polyurethane (PU) sponge using two different coupling agents: (3-aminopropyl)triethoxysilane (APTES) and titanium(IV)(triethanolaminato)isopropoxide (TTEAI), resulting in a composite material that can be utilized as a re-usable and efficient oil-water separation tool. The coupling agents were introduced before and after reduction of graphene oxide to compare the impact of the sequence on the absorbance capacities. The sponges were evaluated based on hydrophobicity and pump oil absorbance capacities (Figure 1). The structure and properties of the sponges were evaluated using scanning electron microscopy and Fourier transform infrared spectroscopy. Results revealed that reacting the graphene oxide PU sponge with the coupling agent titanium(IV)(triethanolaminato)iso-propoxide prior to reduction significantly improved oil absorbance. This was due to the improved binding of reduced graphene oxide to the polyurethane framework. The resulting improved rGO-coated PU sponge is a highly efficient and reusable sorbent material and is a promising alternative to current absorbent materials for oil−water separation applications (Fig 2).

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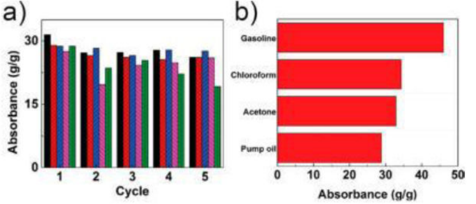

Figure 1: (a) Normalized absorbance capacities for pump oil over five cycles for (left to right) rGO, a-APTES-rGO, a-TTEAI-rGO, b-APTES-rGO, and b-TTEAI-rGO. (b) Absorbance of a-TTEAI-rGO in various organic media.

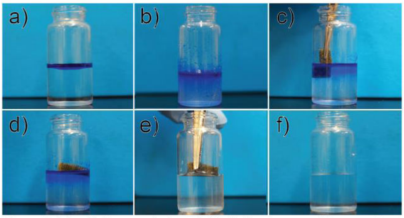

Figure 2: (a) – (f) Removal of emulsified, dyed pump oil from water using the as-prepared modified rGO/PU composite sponge

A Varonides

University of Scranton, USA

Landauer formalism modeling for total thermionic current in reverse-biased Ideal Graphene/n-Si Schottky junctions

The typical current-voltage relationship predicted by basic thermionic emission modeling, namely, $j = A^*T^2 \exp(-q\boldsymbol{\Phi_b}/kT)[\exp(qV/kT)-1]$, ($q\boldsymbol{\Phi_b}$ the junction barrier, V the applied voltage, and A* Richardson's constant) does not include specific carrier transport mechanisms that occur across the graphene/semiconductor Schottky junction (G/n-Si junction). In a reverse biased G/n-Si junction, graphene's fermi level shifts upwards relative to the semiconductor's fermi level, through the reversely applied voltage though the junction. Reverse current (G to Si side) is probable via thermionic escape. In this communication we propose a way of re-writing, from first principles, the formula for thermionic carrier transport across an ideal Schottky G/n-Si junction under reverse bias, by considering two groups of migrating carriers: those carriers that surmount over and tunnel through the junction barrier spike $q\boldsymbol{\Phi_b}$. We model carrier escape in a Landauer scheme, by expressing the current over or through the Schottky junction spike, namely, $J=(q/t_g)\int dE \times D(E) \times (f_g-f_s) \times f(E) \times v(E) \times t(E)$, where q is the electronic charge, t_g is the thickness of the graphene layer D(E) is graphene's linear DOS (density of states), $f_{g,S}$ are the Fermi-Dirac carrier probability functions, v(E) is the thermal electron velocity, and t(E) is the transmission probability over (=1) and through (less than 1) the barrier. Starting from the last integral, we model migrating carriers through two transport mechanisms (a) thermionic escape (TE) over the junction barrier and (b) thermionic field emission (TFE) through the barrier. In the first case, electrons are modeled as carriers surmounting the junction barrier with maximum probability (|t| = 1), and in the latter, carriers are modeled as tunneling through the junction barrier with a non-zero probability $|t| = \exp(-q\boldsymbol{\Phi_b}/E_{oo})$. The denominator in the probability exponential depends on semiconductor donor doping N_d according to $E_{00} = \left(\frac{q\hbar}{2}\right)\sqrt{N_d}/\sqrt{m*\varepsilon_s}$, where m* is the semiconductor's effective mass and ε_s is its dielectric constant. We calculate both TE and TFE currents explicitly: (1) TE current is explicitly derived as

$J_{TE} = A_1^* T^{1.5} exp(-q\Phi_b/kT)[1 - \exp(-qV/kT)]$
and (2) TFE current:
$J_{TFE} = A_2^* T^{2.5} exp\left(-\frac{q\Phi_b}{nkT}\right)[1 - exp(-qV/kT)]$,
where n is the junction *quality factor* directly dependent on the tunneling probability |t| through the potential spike at the junction at energy values under $q\Phi_b$.; the parameter n can be found explicitly as $n = \gamma N_d^{½}$, with γ an appropriate constant $\gamma \sim 10^{-12} m^{3/2}$ related to carrier effective mass, semiconductor dielectric constant and the kT parameter. For total current, we the sum of TE and TFE components will suffice:

$$J = [A_1^* T^{3/2} exp(-q\Phi_b/kT)+$$
$$+A_2^* T^{5/2} exp(-q\Phi_b/nkT)](1 - \exp(-qV/kT)].$$

We see from this last result, that temperature dependence is smeared between concurrent $T^{1.5}$ and $T^{2.5}$ dependences respectively, and two new Richardson-like constants A_1^*, and A_2^*, containing the thickness of the 2-D graphene layer; different Richardson's constants appear because of the graphene layer replacing the metal of a traditional Schottky junction. The quality factor n in the second exponential is related to the semiconductor's donor doping level as $N_d^{½}$; with values expected to be between 0.10 and 0.316 for semiconductor doping levels from $N_d = 10^{16}$ to 10^{17} cm^{-3} respectively. Alternatively, the total current can also be written in a more general form:

$$J = [A_1^* T^{3/2} exp(-q\Phi_b/kT)$$
$$[1 + (3/2)(kT/q\Phi_b) exp(-q\Phi_b/mkT)]$$
$$(1 - \exp(-qV/kT)]$$

kT<< qΦ_b, and m a new version of the original factor m = n/ (n-1).

P. Venezuela[1]
M. Calandra[2] and F. Mauri[2]

[1] Instituto de Física, Universidade Federal Fluminense, Niterói, RJ, Brazil
[2] Institut de Minéralogie, de Physique des Matériaux, et de Cosmochimie, Sorbonne Universités – Paris, France

vene@if.uff.br

Second-Order Overtone and Combination Raman Modes in Bilayer Graphene: First-Principles Calculations

Double-resonant Raman (DRR) spectroscopy is a very important characterization tool for graphitic materials. In general, the 2D band is the most intense DRR peak in this kind of material. The 2D band involves the creation of two optical phonons with wave-vector close to the K-point of the Brillouin Zone. Nevertheless, there are several other DRR peaks in graphitic materials that may be useful in these materials characterization.

Here, we present *ab-initio* calculations for intensities, positions and line shapes of small intensity DRR peaks in bilayer graphene. We use the methodology presented in detail in [1], where density functional theory calculations were used to investigate the 2D band in bilayer graphene.

Recently, several experimental papers reported results for small intensities DRR peaks in bilayer graphene. The fisrt measurements appeared in Refs. [2] and [3]. In these works the Raman spectra for bilayer graphene between 1650 and 2300 cm^{-1} were reported. In this region of the spectra it is possible to identify several DRR bands. More recently, these DRR bands were measured with several different laser energies [4.5].

In bilayer graphene there are also DRR bands in the region of the spectra between 160 and 200 cm^{-1}[6]. These bands are due to layer breathing phonon modes. Additionally, Stokes-anti-Stokes bands measurements have been also reported recently [7].

In our calculations we are able to reproduce the experimental measurements reported in Refs. [2] to [7] with excellent agreement. In FIG. 1, for instance, our results are shown, in the region of the spectra between 1650 and 2400 cm^{-1}, for three different laser energies. More importantly our calculations allow a full interpretation of the experimental results. We are able to unambiguously determine the origin of all DRR processes concerning the phonons branches, wave-vectors and relevant electronic processes.

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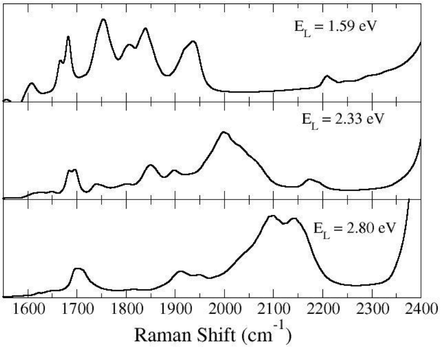

Figure 1: Calculated DRR bands for bilayer graphene for three different laser energies.

Zhongyang Wang[1]
Guqiao Ding[2], Xingxia Deng[1], Siwei Yang[2], Linfan Tian[1]

[1] Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai,201210, China
[2] State Key Laboratory of Functional Materials for Informatics, Shanghai Institute of Microsystem and Information Technology, CAS, Shanghai, 200050, China
wangzy@sari.ac.cn

Luminescence Mechanism of the doped Graphene Quantum dots

Due to the enormous merits, such as stable photoluminescence(PL), high fluorescent activity, low toxicity, excellent solubility and biocompatibility, Graphene quantum dots (GQD) have potential application in the fields of light-emitting diodes, bioimaging and electrochemical biosensing . The luminescence origin has been found to be attributed with quantum confinement effect, surface passivation, and doping. Recently our researches find that the nitrogen-doping is the effective way to improve the PL quantum efficiency of GQDs, however the electronic structure and luminescence mechanism still need to explore. In this talk, we will present some new results of the doped GQDs. Using the spectroscopic and time-resolved measurements, we try to understand the luminescence mechanism with different doping state such as pyridinic-N, pyrrolic-N and graphitic-N. We also studied the influence of oxygen-doping on PL of GQDs. The origin of the PL emission at 430nm with the PLE peak at 320nm previously explained as the carbine-like state from the free zigzag edge, is now recognized as from oxygen-doping. We also discussed the effects of the quantum confinement effect and the edge functional groups on the PL emission.

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Yu-Shu Wu
Mu-Kuen Lee, Maio-Ling Lin, Ning-Yuan Lue, and Yen-Chun Chen
National Tsing-Hua University, Hsin-Chu, Taiwan
yswu@ee.nthu.edu.tw

Valley FETs in graphene

Electrons in graphene exhibit the binary degree of freedom called valley pseudospin, due to the two-fold valley degeneracy at the Dirac points (K and K') in the graphene band structure. Protected by the large wave vector difference between K and K', the valley pseudospin has a sizable decoherence time allowing for the realization of the pseudospin-based electronics known as valleytronics.[1]. Previous theoretical works have developed a unified VOI-based methodology for the design of valleytronic devices. Such a methodology utilizes the VOI (valley-orbit interaction) that occurs between the pseudospin and an in-plane electric field and electrically manipulates the valley pseudospin to achieve device functions. Employing the methodology, a family of valleytronic devices including valley qubits [2], valley filters [2] and valley FETs [3] have been proposed. Here, for demonstration of the methodology, we consider the valley FET. The presentation will discuss its underlying principle and report our recent numerical simulation of electron transport through the structure based on the method of recursive Green's function.

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Figures

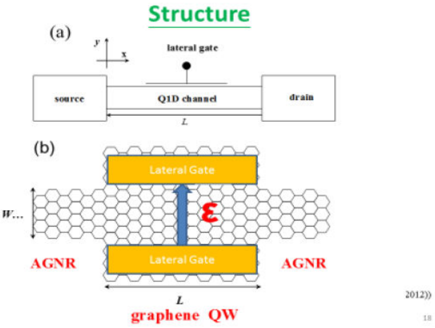

Figure 1: The proposed valley FET - a Q1D channel in gapped graphene, with the source and drain being armchair nanoribbons and the channel defined and controlled by electrical gates.

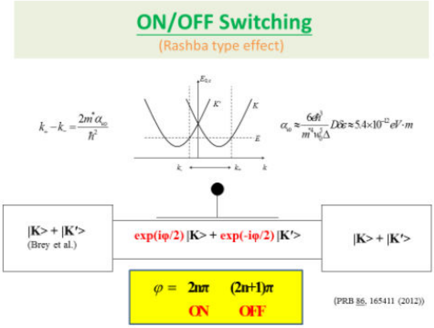

Figure 2: Source / drain inject / detect electrons in the state |K> + |K'>. The channel pseudospin is rotated due to the VOI. Depending on the angle of rotation, the channel is on (for angle = (2n+1)π) or off (for angle = 2nπ)

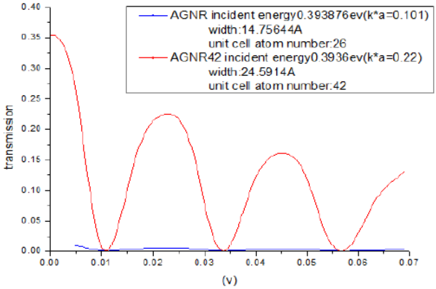

Figure 3: Electron transmission vs. gate bias, for two channel widths.

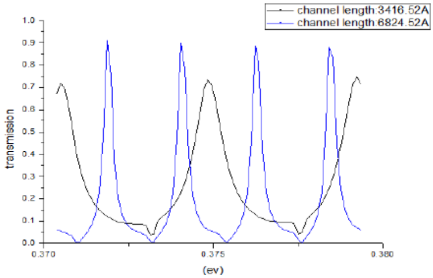

Figure 4: Electron transmission vs. gate bias, for two channel lengths.

Saifeng Zhang[1]
Yuanxin Li[1], Niall McEvoy[2], Damien Hanlon[2,3], Jonathan N. Coleman[2,3], Jun Wang[1]

[1] Key Laboratory of Materials for High-Power Laser, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China
[2] Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN) and Advanced Materials and BioEngineering Research (AMBER) Centre, Trinity College Dublin, Dublin 2, Ireland
[3] School of Physics, Trinity College Dublin, Dublin 2, Ireland

sfzhang@siom.ac.cn

Nonlinear absorption in two dimensional materials

Thin film materials with large optical nonlinearity and ultrafast response are required as the promising candidates for future integrated photonics applications. Recently, the two dimensional layered transition metal dichalcogenides (TMDs) and black phosphorus (BP) have attracted much attention due to their unique electronic, optical, mechanical, chemical and thermal properties. WS_2 and MoS_2 mono- and few-layer films were fabricated by vapor phase sulfurization and chemical vapor deposition method. We investigated the optical nonlinearity of mono- and few-layer WS_2 and MoS_2 using the Z-scan technique at a wavelength of 1030 nm, 800 nm and 515 nm with femtosecond pulses. The monolayer WS_2 exhibited giant optical nonlinearities having the two-photon absorption (TPA) coefficient of ~1.0×10^4 cm/GW. The layer number and excitation wavelength dependence of the optical nonlinearity of WS_2 and MoS_2 were investigated. The mechanism of the optical nonlinearity was discussed in detail. The damage thresholds of TMDs were also given to support potential device application in the future. We have obtained the ground-state absorption (GSA) and excited-state absorption (ESA) cross section σ_{gs} andσ_{es} of BP nanosheets, 1.25×10^{-16} cm^2 and 2.85×10^{-17} cm^2 at 515 nm, 1.23×10^{-17} cm^2 and 3.02×10^{-18} cm^2 at 1030 nm with the slow saturable absorber (SA) model. The ratio of σ_{es}/σ_{gs} is about 0.228 at 515 nm and 0.247 at 1030 nm. It is even lower when the fast SA model is applied. The σ_{gs} and σ_{es} of MoS_2 and graphene were also achieved using the slow SA model. It is clear that the ratio σ_{es}/σ_{gs} of BP is the smallest among three two-dimensional layered materials. The superior saturable absorption performance of BP over MoS_2 and graphene can be well explained from this perspective. The results fundamentally support BP to be used as a saturable absorber in nanophotonic devices, such as fiber laser mode-locking, Q-switching, optical switches, etc.

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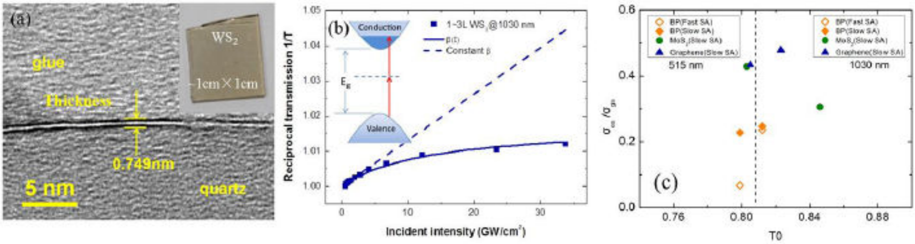

Figure 1: (a) Cross-sectional TEM of the 1~3L WS_2 film, (b) Reciprocal transmission versus irradiance of 1~3L WS2 film. Solid squares are the measured data. Solid line: theoretical variation for a hyperbolic irradiance dependence of the TPA coefficient. Dashed line: theoretical variation for a constant TPA coefficient. (c) The ratio of ground-state absorption and excited-state absorption cross section of BP, MoS_2 and graphene at 515 nm and 1030 nm.

Navjot Kaur
Kaushik Pal

Indian Institute of Technology Roorkee,
Roorkee, Uttarakhand, India – 247667

pl_kshk@yahoo.co.in

Effect of cobalt doping on the magnetic properties of graphene nanoribbons

Carbon based materials such as fullerene, carbon nanotubes, graphene and its derivatives have good candidates for various applications due to unique properties. In this work, we have focused on structural and magnetic properties of cobalt doped graphene nanoribbons (GNRs). The effect of Co on tuning of the electronic band structure which play crucial role in contribution of calculated magnetic moment of as synthesized GNRs through chemical reduction of graphene oxide nanoribbons using hydrazine and first principle calculations based on density-functional theory (DFT). Field emission scanning electron microscope with EDX analysis and X-ray diffraction pattern were confirmed the doping of Co into the GNRs matrix. Moreover, the results obtained from VSM analysis are clearly revealed that improved saturation magnetization (Ms) from $7.95\text{x}10^{-3}$ emug^{-1} to 12.68 $\text{x}10^{-3}$ emug^{-1}, due to the presence of Co content in pristine GNRs. The achieved magnetizations curve show ferromagnetic behaviour at room temperature. The Co doped GNRs exhibited interesting magnetic properties which create much attention towards its applications such as spintronics devices and some related fields.

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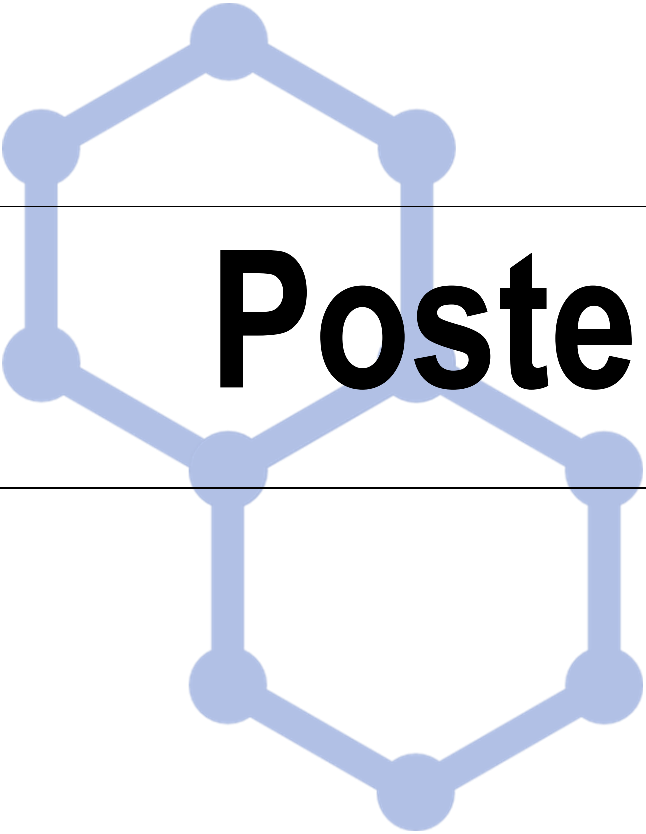

Sana Ahmed[1]
Huma Shaikh[1], Amber Solangi[1], Sirajuddin[1] and Adil Denizli[2]

[1] National Centre of Excellence in Analytical Chemistry, University of Sindh, Jamshoro, Pakistan
[2] Department of Chemistry, Biochemistry Division, Hacettepe University, Ankara, Turkey

sanaahmed127@gmail.com

Development of atrazine imprinted graphene composites based electrochemical sensor for trace level determination of atrazine in real samples

Graphene, a 2D allotrope of carbon with honeycomb structure is an exciting material for many device applications. Since its production in 2004, it has been used significantly to improve the sensitivity in various applications due to rapid electron transfer and high surface-to-volume ratio. This one atom layer thick carbon sheet exhibits many interesting electronic, optical and mechanical properties due to its two-dimensional (2D) crystal structure and possesses high conductivity despite it being an organic compound.

Therefore, an attempt was made to prepare a graphene based electrochemical sensor for trace level analysis of atrazine in real samples. In this regard; graphene oxide (GO) was prepared from graphite by modified Hummers method, later on it was further reduced using polypyrrole to prepare polymer composites. Finally, these graphene composites were made selective for atrazine by molecular imprinting using ionic liquid (1-butyl-3-methyl imidazolium) tetra fluoro borate.

After synthesis, this work includes usage of imprinted graphene composites (IGCs) for the selective and sensitive detection of atrazine in different standards and real samples. Screen Printed Electrodes were modified using the IGC. The voltammetric measurements were performed in NaH_2PO_4 electrolyte. Parameters such as type of electrolyte, its concentration, pH, SPE coating, equilibrium time, frequency, stirring, scan rate, etc were studied thoroughly to detect atrazine using the prepared sensor.

The surface morphology and composition of IGC was studied using a number of suitable techniques such as SEM, FTIR, XRD and Raman spectroscopy.

Key Words: Atrazine, imprinting, screen printed electrode, graphene.

Randy Belanger[1]
Jonghyurk Park[2], Jian Wang[3], Young-June Kim[1]

[1]Department of Physics, University of Toronto, Toronto, ON, Canada M5S1A7
[2]ICT Devices and Materials Laboratory, Electronics and Telecommunications Research Institute (ETRI), Daejeon, Korea 34129
[3]Canadian Light Source Inc., University of Saskatchewan, Saskatoon, SK, Canada S7N 2V3
belanger@physics.utoronto.ca

Thickness mapping of exfoliated graphene samples using Scanning Transmission X-ray Microscopy (STXM)

Ever since single graphene layers were isolated using the "scotch tape" exfoliation method [1], graphene has been the topic of intense research. Much of this research has been centered on trying to commercialize products which make use of the unique physical properties of graphene. However, since these properties are very sensitive to the number of graphene layers that are present in a sample it is very important to know the thickness and/or the number of graphene layers that are present. The majority of work that has been conducted on thickness measurements of graphene has been done with the use of atomic force microscopy (AFM). Unfortunately, AFM has been notoriously unreliable in determining the thickness of graphene layers. In this talk, I will discuss the use of another technique, Scanning Transmission X-ray Microscopy (STXM), to determine the number of graphene layers in a region of an exfoliated sample prepared on SiN windows. This technique consists of collecting an image by raster scanning a highly focused (40 nm) soft x-ray synchrotron beam [2,3] across a sample at defined incident photon energies. A near edge X-ray absorption fine structure (NEXAFS) spectrum is collected at each pixel by adjusting the energy through the carbon K edge, producing an image "stack". The optical density obtained in this manner can be used to quantitatively determine the number of graphene layers. In addition, the acquired spectroscopic information is useful for characterizing surface cleanliness of the graphene sample.

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Dominic Boisvert
Guido Soliveri, Eduardo Di Mauro, Sébastien Francoeur, Clara Santato
Polytechnique Montréal, 2900 boul. Édouard-Montpetit, Montreal, Canada

dominic-2.boisvert@polymtl.ca

Deposition and structural characterization of atomically-thin melanin biopigments

Mainly known for the tan it produces in humans, melanin is also responsible for a variety of functions, such as photo-protection of skin and eye, free radical scavenging and metal ion binding. It results from the combination of two monomers, DHI and DHICA, and their redox forms that polymerize in a disordered manner to form 2D oligomers that then form more complex layered structures. Recent reports in the literature point to the mixed ionic-electronic conduction properties of these structures and such properties renders melanin an interesting candidate for organic nanoscale devices featuring biocompatible and easily available “active” materials. A critical point concerns the metal ion binding properties that is not well understood despite the strategic importance it has to explain the biological role of melanin in our body as well as possible development of green solar conversion technologies. On one hand, metal ion binding can be advantageously used to tailor electrical conduction but on the other hand, it can increase the structural disorder in the pigment, thus limiting both the reproducibility of the experiments and that same conduction. Using spin coating deposition, we exploit a new synthetic approach based on the solid state polymerization of the melanin monomers [1] to produce controlled samples of 2D melanin that will have different chain lengths and nominal thickness, ranging from the sub monolayer regime to a few monolayers. Using atomic force microscopy (AFM, Figure 1) we observed that thicknesses lower than five monolayers can be obtained and we correlated deposition method and surface coverage. Capitalizing on these findings, we explore the use of other characterization methods such as Raman spectroscopy and UV-Visible absorption to gain additional insight on the structural and optical properties of the obtained oligomers.

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Figures

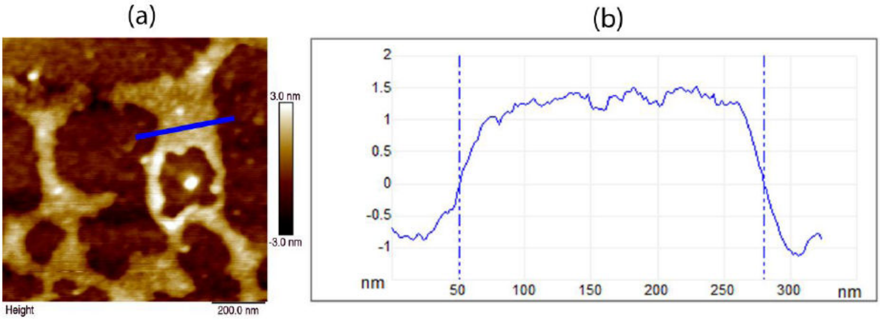

Figure 1 : 1x1 μm AFM scan of a DHI on SiO_2 sample. (a) shows the height diagram of the measured sample and (b) is the cross section shown by the line on (a) .

Carvalho, R.D.E.
Carvalho, R.H., Ferreira, M.P., Santos, A.P., Furtado, C.A.

Centro de Desenvolvimento da Tecnologia Nuclear - CDTN, Av. Presidente Antônio Carlos, 6.627
Campus da UFMG – Pampulha Caixa Postal 941 Belo Horizonte, Minas Gerais, Brazil
duque_strada@yahoo.com.br

Graphene based coatings for UV radiation shielding: an improved way of protection!!

UV radiation has short wavelength and high energy exposure. Even though UV radiation is widely used in industrial processes and applications like detectors, sensors, medical diagnosis and therapy, prolonged exposure to it may result in degradation and harmful health effects on the skin, eyes, and immune system of humans. Protection is therefore needed and alternative UV shielding materials with enhanced performance and versatility are still required [1].

In this paper, graphene oxide and few layer graphene based nanocomposite and coatings of poly (vinyl alcohol) and textiles of polyester (PY) were tested for shielding of UVC (100–280 nm), UVB (280–315 nm) and UVA (315–400 nm) radiation. The nanocomposites were characterized by scanning electron microscopy, thermogravimetric analysis, Raman spectroscopy and infrared spectroscopy. Results show no significant structural changes after radiation, indicating a good stability of the studied systems. The incorporation of graphenic nanostructures resulted in transparent films and textiles (Fig.1) with enhanced UV-shielding properties. The results are promising and the absorbance of ultraviolet light at 300 nm can be up to 80% for films with around 70% of transparency in visible spectral range and concentration as low as 0.5 wt% of nanocarbon.

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Figures

Figure 1: Photographs of graphene containing coating and textile for UV radiation shielding.

Carl-Bernard Charpin[1]
Dhan Cardinal[2], Saman Choubak[2], Pierre L. Levesque[1], Patrick Desjardins[2], Richard Martel[1]

[1] Regroupement Québécois sur les Matériaux de Pointe (RQMP) and Département de Chimie, Université de Montréal, C.P. 6128, Succursale Centre-Ville, Montréal, Québec H3C 3J7, Canada
[2] Regroupement Québécois sur les Matériaux de Pointe (RQMP) and Département de Génie Physique, Polytechnique Montréal, Montréal, C.P. 6079, Succursale Centre-ville, Montréal, Québec H3C 3A7, Canada
carl-bernard.charpin@umontreal.ca

Graphene Growth Kinetics Under Purified Conditions

Chemical vapor deposition (CVD) is one of the most promising route towards the mass production of graphene sheets for industrial applications requiring large surfaces and pristine quality. To reach the necessary standardization of the resulting material and to further model and engineer the growth, a fundamental understanding of the growth kinetics, including a reliable database of the associated parameters, is still lacking. Our group has recently shown the important role of oxidizing impurities [1, 2], even at trace level, under graphene CVD growth conditions. Using a severe control over these impurities from both the furnace and gas feedstock, we were able to identify two growth regimes: Oxidizing impurity limited growth and precursor dissociation limited growth. According to our findings, most kinetic studies in the literature using standard ultra-high purity (UHP) gases are in the oxidizing impurity limited growth regime. In order to better discretize growth parameters effects and extract fundamental kinetics values, we devised a set of experiments where purity conditions are minimized in order to set the growth conditions in the precursor dissociation limited regime. We conducted LP-CVD growth experiments with varying growth conditions: pressure, temperature and time. The samples were characterized using microscopic methods (scanning electron microscopy (SEM) and Low energy electron microscopy (LEEM)) to study the surface morphology, crystallinity, nucleation density and coverage. RAMAN spectroscopy was used to assess the graphene quality and thickness. In route toward the full model, we found out that in purified condition the growth is greatly accelerated by a factor of x50 compared to UHP condition without affecting the graphene quality. For instance, we reached >85% coverage in 10min @ 800 °C and full coverage in 20 s @ 1000 °C. We also determined that the total dose of methane is the main parameter controlling the graphene coverage (see figure below) and that the H_2 has only minor effect when O_2 is minimized. Effects of growth parameters on nucleation density and crystallite size will be further commented within the presentation. This study will allow retrieving the fundamental kinetic constants and activation energies that are essential to later model and engineer graphene for future applications and also give new fundamental insight on the catalytic activity of copper for dissociating methane.

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Figures

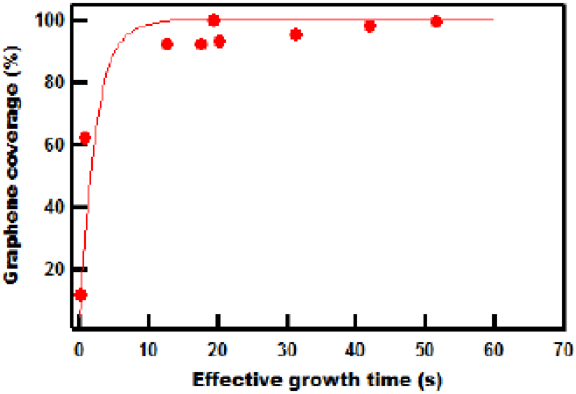

Figure 1: Effective growth time influences graphene coverage. These growths were obtained with varying total methane doses.

Maxime Daigle
Audrey Picard-Lafond, Eliane Soligo, Jean-François Morin
Université Laval, 1045 Avenue de la Médecine, Québec, Canada
maxime.daigle.3@ulaval.ca

A Photochemical Approach Towards Graphene Nanoribbons

As graphene is known to be a highly conductive zero band gap material, graphene nanoribbons (GNRs) are of interest as semiconducting materials for electronic applications owing to its high charge carrier mobilities [1]. Although challenging, a bottom-up approach towards their preparation is the only way to access well-defined structures.

Scholl cyclodehydrogenation is still the most exploited reaction for the cyclization of polyphenylene precursors, yielding to a variety of polycyclic aromatic hydrocarbons (PAHs) as well as GNRs [2]. Unfortunately, the Scholl cyclodehydrogenation reaction lack of selectivity, therefore limiting the variety of graphenic molecules that can be made [3]. Thus, new synthetic cyclization strategies need to be investigated in order to build new libraries of molecules, thus extending the chemical and physical properties of the obtained materials.

Here we propose a newly designed photochemical approach that consists in the irradiation of photosensitive halogenated polyphenylenic precursors yielding to PAHs and GNRs. The cyclodehydrochlorination reaction benefits of a precise regioselectivity as well as low byproduct release, resulting in a greener alternative to produce PAHs and GNRs [4].

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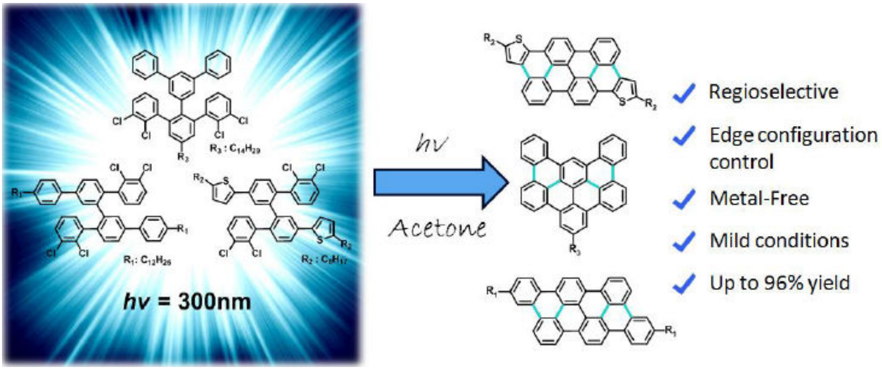

Figure 1: Multiple cyclodehydrochlorination reaction for nanographenes synthesis.

Ganesh Gollavelli
Chain Shu-Hsu
National Chiao Tung University, Dept. of Applied Chemistry, Hsinchu, 30010 Taiwan
ganesgollavelli@gmail.com

Exploring photothermal hot spots of graphene in first and second biological window to inactivate cancer cells and pathogens

Abstract: We have developed a new composite material composed of reduced graphene oxide (RGO) and poly acrylic acid (PAA) to achieve reduced graphene oxide functionalized poly acrylic acid (RGOPAA). The photothermal capability of RGOPAA was demonstrated at 808 nm and 1064 nm near infra-red (NIR) wavelengths to kill HeLa cancer cells and S. aureus. The key feature of RGOPAA is that it exhibits wide range of NIR adsorption both in first (650~950 nm) and second biological windows (1000~1300 nm). The photothermal heating curves of RGOPAA show similar temperature profiles at 808 nm and 1064 nm wavelengths. The confocal laser scanning microscopy (CLSM) images reveals the successful internalization of RGOPAA when into HeLa cancer cells. The MTT assay and heat shock protein expression studies reveal that the cytotoxicity of HeLa cancer cells was mainly attributed from the heat generation of graphene upon photoirradiation. In addition, the photothermal studies were also demonstrated in the pathogenic bacteria, S. aureus which reveals excellent killing efficiencies. Overall, our results clearly shows that graphene exhibits excitation wavelength independent photothermal therapy in the NIR region covering both the biological windows and can be a robust and economic photothermal reagent to combat against cancer and pathogens.

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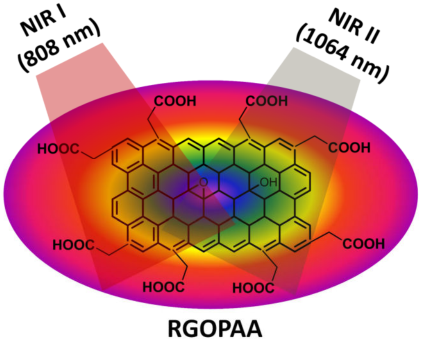

Jean-Francis Germain[1]

Alexandre Favron[2], Richard Leonelli[2], Richard Martel[2], Sébastien Francoeur[1]

[1]École Polytechnique de Montréal, 2900 boul. Édouard-Montpetit, Montréal, Canada
[2]Université de Montréal, 2900 boul. Édouard-Montpetit, Montréal, Canada
jean-francis.germain@polymtl.ca

The semiconductor-semimetal transition induced by potassium doping in atomically-thin black phosphorus studied through Raman spectroscopy

wo-dimensional materials based on Black phosphorus is a 2D semiconductor material exhibiting a thickness-tunable direct band-gap and pronounced electrical, mechanical, and optical in-plane anisotropies resulting from its puckered structure. Exposed to the high electric field induced by adsorbed potassium atoms, the band gap of few-layer black-phosphorus shrinks and electronic band dispersions change significantly: from a quadratic to linear dispersion along the armchair direction [2]. At high doping levels, a semiconductor to semimetal transition has been observed [1]. To shed light on these mechanisms, we analyse the evolution of the Raman-allowed Ag1 and Ag2 modes as a function of potassium doping, thickness, excitation wavelength, and polarization. Raman spectra reveal a clear signature of the semiconductor to semimetal transition.

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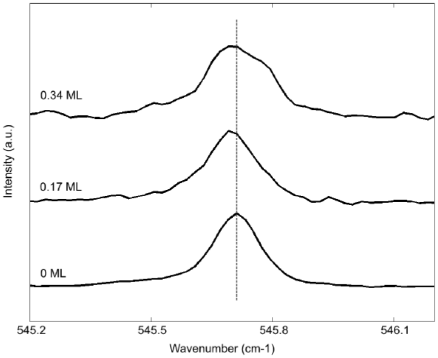

Figure 1: The Ag2 mode shifts and increases in width as a function of doping where a monolayer of potassium (ML) is 3.46 x 1014 electrons. Dash line is a guide to the eye.

Haider Haseeb
Affan Waheed, Saima Shabbir
Institute of Space Technology and University of Augsburg, Augsburg, Germany
haiderhaseeb@live.com

Polymer Based Clay Quantum Dot Multifunctional Nano-materials: Synthesis and Characterization

Nanotechnology can provide a multitude of improvements and new solutions to the polymeric material. Polymer based clay–quantum dot multifunctional nano-hybrids were synthesized using the solution intercalation method. The inclusion of inorganic fillers in a polymer improves its mechanical strength, anti-flammability, dimensional stability and other properties. With the selection of appropriate fillers, existing properties of the polymer can be enhanced or new properties can be acquired that is not usually associated with the polymer. Fundamental limits in clay composition prevent them from being used easily in applications requiring electrical/thermal conductivity or optical applications. Therefore, Multi nano-fillers, organoclay and quantum dots, were used yielding nanocomposite with enhanced mechanical, flammability, thermal, electrical and optical properties – allowing it to be a drop-in replacement for many different materials while keeping other properties intact. The dispersion of nanosized fillers (nanofillers) in the polymer is often the most challenging and crucial problem. Nanofillers tend to aggregate due to strong van der Waals attraction, which makes it necessary to develop strategies to overcome this attraction. End functionalization strategy was adopted to develop various interactions among the phases. Hydrophillic nature of montmorillonite clay was changed into organophillic by ion exchange method using various intercalating agents with different functionality and chain lengths. These modifications were carried out in order to have good compatibility, uniform and homogenous dispersion of clay and quantum dots in the polymer matrix. These intriguing new polymer composites based on clay-quantum dots will combine the favorable emission characteristics with excellent mechanical properties. The polymer based clay-quantum dot materials with high photoluminescence (PL) efficiency will be promising for use in future optoelectronic devices such aslight emitting devices, aerospace applications and solar cells.

Figures

Figure 1: Yellow luminescence under UV excitation for the solution, quantum dots and fabricated films with quantum dots and organoclay as nanofillers.

M. Hulman
M. Sojková, Š. Chromik, E. Dobročka, A. Rosová, J. Brndiarová. P. Hutár
Institute of Electrical Engineering, SAS, Dúbravská cesta 9, 841 04 Bratislava, Slovakia
martin.hulman@savba.sk

Growth of extremely thin MoS_2 films by pulsed laser deposition

Two-dimensional (2D) materials have been one of the most extensively studied classes of materials due to their unusual physical properties. The best example is graphene – a single layer of carbon atoms arranged in a two dimensional (2D) honeycomb lattice. Many graphene´s extraordinary properties have been reported including excellent electronic and thermal conductivities and mechanical properties [1]. However, pristine graphene itself is unlikely to be used for the fabrication of logical circuits because of lacking band gap.

The discovery of graphene has stimulated an extensive research on other 2D materials. It has been shown that it is not only possible to exfoliate stable, single-atom or single-polyhedral-thick 2D materials, but that these materials can exhibit unique and fascinating physical properties. The 2D structure determines the electronic properties that may exhibit correlated electronic phenomena such as charge density waves and superconductivity [2].

Some remarkable changes in the electronic properties of layered materials as their thickness is reduced down to single or only a few layers are common among these 2D crystals. In contrast to the zero band gap of pristine graphene itself, a single-layer MoS_2 sheet is a direct band gap semiconductor. The result is that MoS_2 is a promising material that has the potential to be incorporated into digital circuits and light-emitting diodes. For instance, the current on/off ratio of single-layer MoS_2 transistors exceeds 10^8 at room temperature. This is much higher than that (approximately 100) of graphene transistors [3].

Recognizing the uniqueness of the 2D structure, one may expect that such materials will reveal new and unexpected properties providing a number of innovative opportunities. Because of their distinct properties and high specific surface areas, these 2D materials are important in various applications such as optoelectronics, spintronics, catalysts, chemical and biological sensors, supercapacitors, solar cells, and lithium ion batteries [4].

Here, we present preparation of very thin MoS_2 films on various substrates (Si, sapphire, GaN) from a stoichiometric target by a pulsed laser deposition. Combined results from Raman spectroscopy and X-ray reflectivity have shown that the thinnest samples contain two MoS_2 layers. The thickness is controlled by the number of laser pulses applied albeit no simple direct proportion between the two quantities has been observed. Selected area electron diffraction studies have confirmed that the films deposited on Si (100) are nanocrystalline and oriented perpendicularly to the substrate surface while an epitaxial growth of MoS_2 films was observed on GaN substrates [5].

We also report on preliminary results on fabrication of ultrathin MoS_2 layers on graphene and graphene oxide. Hybrid systems are promising for some applications including photodetectors and field effect transistors where graphene may act as a material for electrodes with ultimate thickness.

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Acknowledgement

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Frédéric Joucken[1]
Nicolas Reckinger[1], Stéphane Lorcy[2], José Avila[2], Chaoyu Chen[2], Jérôme Lagoute[3], Jean-François Colomer[1], Jacques Ghijsen[1], Maria Carmen Asensio[2] & Robert Sporken[1]

[1]University of Namur, Namur, Belgium
[2]Soleil synchrotron, Gif-sur-Yvette, France
[3]MPQ lab, Université Paris Diderot – Paris 7, France

frederic.joucken@gmail.com

NanoARPES investigation of pristine graphene: evidencing the need for high spatial resolution in ARPES experiments

Angle-resolved photoemission spectroscopy (ARPES) is the most direct tool to measure the electronic structure of materials. In particular, fine features of the spectra can be analyzed for evaluating the electron self-energy. Owing to a setup allowing ARPES investigation with submicron resolution and state-of-the-art energy and momentum resolution operated at the Antares beamline of the Soleil synchrotron [1], we show here first that ARPES spectra of pristine and virtually undoped monolayer graphene acquired on a small spot do not display manifestations of self-energy. We next demonstrate that, although the region of the sample investigated is a unique graphene domain, it displays faint spatial inhomogeneity, both in its crystallographic orientation and its thickness, which is undetectable with conventional ARPES but renders the spectra improper for self-energy extraction. These results indicate that care should be taken when analyzing ARPES spectra obtained with poor spatial resolution.

A nanoARPES image of the graphene domain investigated is shown on Fig. 1, together with spectra acquired at different spots along the x-axis on the image. Differences between spectra are evident.

The results presented here are detailed in ref. [2].

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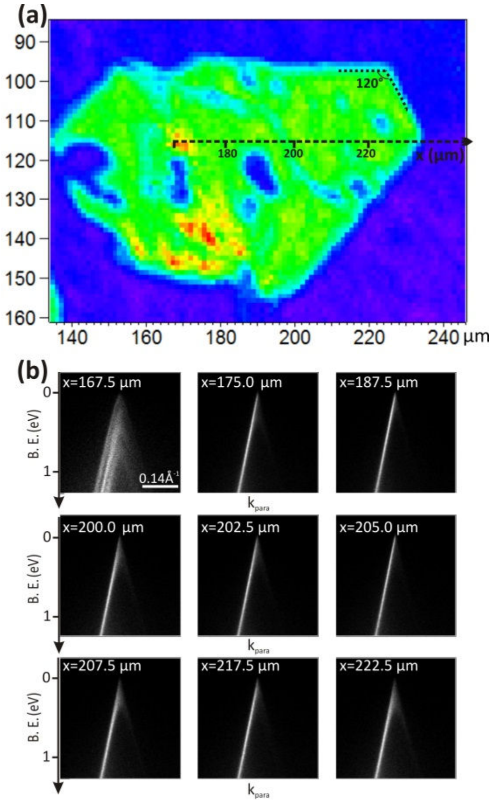

Figure 1: A nanoARPES image of the graphene domain investigated (a), together with spectra acquired at different spots along the x-axis on the image (b).

Tatsuhiro Komatsu[1]
Taiichi Otsuji[1], Victor Ryzhii[1], Dmitry Svintsov[2], Vladimir Vyurkov[2], Akira Satou[1]

[1]Research Institute of Electrical Communication, Tohoku University, Sendai, Japan
[2]Institute of Physics and Technology, Russian Academy of Sciences, Moscow, Russia
komatsut@riec.tohoku.ac.jp

A Numerical Model for Finite-Temperature Electron Self-Energy in Doped Graphene with Electron-Electron Interaction

We develop a numerical model for the electron self-energy in doped graphene due to the electron-electron interaction at finite temperatures. It numerically calculates the real and imaginary parts of the self-energy within the Hartree-Fock [1] and random phase approximations [2] taking into account the finite-temperature Fermi-Dirac distribution, so that effects of both the band renormalization and the band broadening on real graphene devices can be investigated. We showed that the calculated RPA dielectric function exhibits the peak broadening and blue shift in the plasmon dispersion as the temperature increases (Fig. 1). As a result, the imaginary part of the self-energy exhibits the broadening of peaks associated with plasmon excitation (Fig. 2). In addition, splitting of the peaks (leading to the formation of the so-called plasmaron band [2] aside from the normal band for electrons) at high temperature is found. These results indicate that the temperature plays a crucial role in the energy dispersion through the self-energy and, in turn, the carrier transport in real devices.

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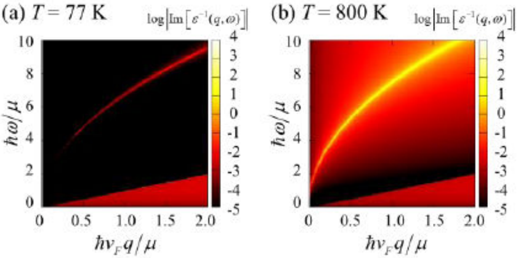

Figure 1: Logarithmic expression of imaginary part of the RPA dielectric function ε in graphene as a function of the wavevector q and the frequency ω calculated at (a) T = 77 K and (b) 800 K. Axes are normalized by the Fermi level μ.

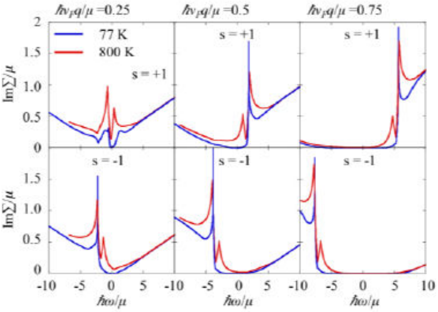

Figure 2: Imaginary part of the self-energy Σ in graphene calculated at T = 77 and 800 K (red and blue lines, respectively). Columns from left to right correspond to $\hbar vFq/\mu$ = 0.25, 0.5, and 0.75, respectively, while top and bottom rows correspond to s = 1 (the conduction band) and s = -1 (the valence band), respectively.

Alexander Krawciw

Mount Douglas Secondary, Victoria BC Canada, Grade 12

freakyfactoid@gmail.com

Graphene: The Next Generation of Printed Circuits

Reduced graphene oxide (rGO) was synthesized using various techniques in order to characterize the substance's conductive properties and suitability for printed circuit boards. It is hypothesized that reduced graphene oxide (rGO) would make a suitable material for printed circuit boards (PCBs). The resistance of an rGO film is expected to be a linear function of the ratio of length/width. Typically rGO has a sheet resistance of about 1000 Ω/□ [1].

Initially graphene oxide synthesis was attempted using liquid phase sonication [2], but this was not successful. Reduced graphene oxide was made using the Lightscribe method [3] but this procedure is fairly slow and has too great a resistance for PCB traces. Using an industrial CO_2 laser cutting [4] was more reliable and yielded a sheet resistance on the order of 3 kOhm/square. The product was analyzed using a Raman spectrometer. This material shows potential for making discrete resistors and especially capacitors.

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Tian Lan[1]

Jonathan Sayago[1], Fabio Cicoira[2], and Clara Santato[1]*

[1]École Polytechnique de Montréal, Département de Génie Physique, C.P. 6079, Succ. Centre Ville, Montréal, Québec H3C-3A7, Canada
[2]École Polytechnique de Montréal, Département de Génie Chimique, C.P. 6079, Succ. Centre Ville, Montréal, Québec H3C-3A7, Canada

*clara.santato@polymtl.ca

Electrolyte-Gated Transistors using n-type organic molecular semiconductors: the case of PCBM as Channel Material

Electrolyte-gated transistors (EGTs) using electrolytes as the gating medium are of interest since the exceptionally high capacitance of the electrical double layer forming at the electrolyte/transistor channel interface permits current modulations of several orders of magnitude, at relatively low gate voltages, sub 2V [1]. Electrolyte gating is a powerful approach to produce large charge carrier densities (ca 1014 cm^{-2}) in semiconductors. Room temperature ionic liquids (RTILs) show great potential as gating media for their physico-chemical properties, such as negligible volatility, electrochemical stability windows up to ca 5 V, ionic conductivity up to 10 mS·cm^{-1} [2]. N-type organic transistors are of relevance for organic p-n junctions, bipolar transistors and complementary integrated circuits for flexible, large-area, and low-cost electronic applications [3]. In this work, we will report on EGTs with RTILs based on bis(trifluoromethylsulfonyl)imide ([TFSI]) anions and 1-ethyl-3-methylimidazolium ([EMIM]) cations as the gating medium and phenyl-C61-butyric acid methyl ester (PCBM) as the n-type organic molecular semiconducting channel material. Cyclic voltammetries in transistor configuration (where the working electrode is the channel material included between the source and drain electrodes and the reference is a high surface area gate electrode) were recorded to bridge the gap between the redox properties of PCBM films, characterized by a certain supramolecular organization, and the modulation of the film electrical conductance. Drain-source voltages, V_{ds}, as low as 100mV and gate-source voltages, V_{gs}, within the range 0~1.25 V, 0~1.7 V, 0~1.9 V were explored. We observed typical transistor behavior for our PCBM EGTs with I_{ds}/I_{gs} and ON/OFF ratios of around 10, ON/OFF ratios increasing with the increase of the gate voltage range. A correlation has been established between the redox peaks observed in the cyclic voltammetry with the morphology/structure of the films as deduced by Atomic Force Microscopy and XRD obtained at glancing angles and the corresponding transistor behavior.

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T.L.M. Lane[1,2]
J.R. Wallbank[1,2], V.I. Fal'ko[1,2]

[1] National Graphene Institute, The University of Manchester, Manchester, M139PL, UK
[2] School of Physics and Astronomy, The University of Manchester, Manchester, M139PL, UK

thomas.lane-3@postgrad.manchester.ac.uk

Twist-controlled resonant tunnelling between monolayer and bilayer graphene

We investigate the current-voltage characteristics of a field-effect tunnelling transistor comprised of both monolayer graphene (MLG) and bilayer graphene (BLG) with well-aligned crystallographic axes, separated by three layers of hexagonal boron nitride. Using a self-consistent description of the device's electrostatic configuration we relate the current to three distinct tunable voltages across the system and hence produce a two-dimensional map of the I-V characteristics in the low energy regime. We show that this theoretical model agrees with recent experimental results and can be used to demonstrate the exact band alignment from which each prominent feature arises. The characteristics include regions of negative differential conductance which present an opportunity for the production of highly tunable devices operable in the terahertz regime [1]. Further, the use of gates either side of the heterostructure as well as varying the twist between the graphene electrodes has been shown to offer a fine degree of control over the device's rich array of characteristics [2].

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Figures

(1)

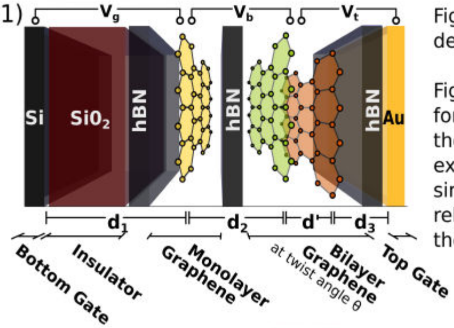
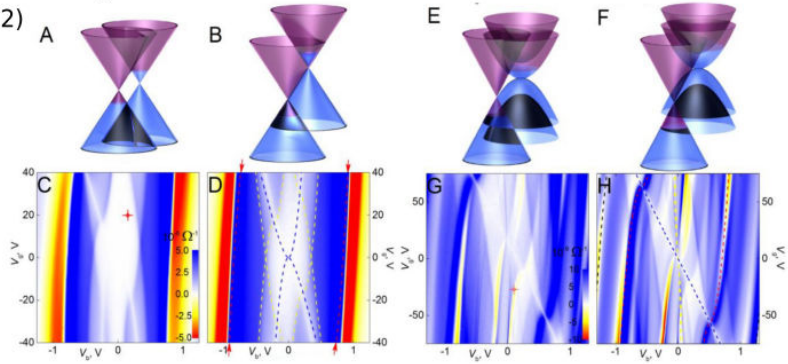


Figure 1. structure of an MLG to BLG tunnelling device with both top and bottom gates.

Figure 2. C and D show current characteristics for an MLG-MLG device and G and H depict those of an MLG-BLG structure. C and G are experimental results, whilst D and H illustrate simulated data. A, B, E and F show the relative band alignment for specific features in the characteristics.

(2)



Alejandro León[1]
Mónica Pacheco[2]*

[1]Facultad de Ingeniería, Universidad Diego Portales, Ejército 441, Santiago, Chile
[2]Universidad Técnica Federico Santa María, Av. España 1680, Valparaíso, Chile

alejandro.leon@udp.cl
*monica.pacheco@usm.cl

Electrical and optical properties of beta-graphyne nanoribbons.

The □beta-graphyne is a class of graphyne that presents an interesting difference from other graphynes shapes. The Dirac cone is not located in a point of high-symmetry in reciprocal space [1-2]. In this work we perform a theoretical study of the electronic and optic properties of bi-nanoribbons of this material, for different stacking configurations. The electronic properties of the systems are obtained using the density functional theory (DFT) code OpenMX [3], which is based on localized pseudoatomic orbitals (PAOs) [4] and norm-conserving pseudopotentials [5]. The PAO of carbon atoms consists of two sorbitals and two p-orbitals. The Perdew–Burke–Ernzerhof (PBE) exchange-correlation functional [6], which is derived within the generalized gradient approximation (GGA) is used, taking into account an energy cutoff equal to 150 Ry and a convergence criteria of 10^{-8} Hartree. All structures are relaxed using the Direct Inversion Iterative Subspace method [7] with a residual force criteria less than 10^{-4} Hartree/Bohr. Our results show that these systems have interesting optical and electrical conduction properties, depending on the stack and edges of nanoribbons. As an example, shown in Figure 1, the results of electrical conduction, of nanoribbon formed with a single layer.

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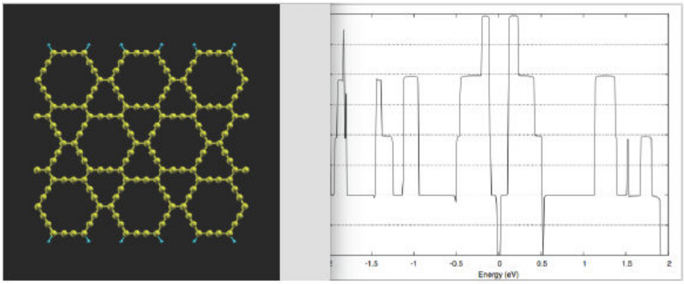

Figure 1: Schematic of beta-graphyne nanoribbon and electrical transmission.

A.B. López-Oyama[1,2]
M.A. Domínguez-Crespo[1], A.M. Torres-Huerta[1], E. Onofre-Bustamante[1], R. Gamez-Corrales[3]

[1] Instituto Politécnico Nacional. CICATA-Altamira. Grupo CIAMS, 14.5, Carretera Tampico-Puerto Industrial Altamira. C. P. 89600. Altamira, Tamps. México
[2]CONACyT- CICATA-Altamira. Grupo CIAMS, 14.5, Carretera Tampico-Puerto Industrial Altamira. C. P. 89600. Altamira, Tamps. México
[3]Universidad de Sonora Blvd. Luis Encinas y Rosales S/N, Col. Centro, C.P. 83250 Hermosillo, Sonora, México

ablopezoy@conacyt.mx

Electrochemical reduction of graphene oxide: synthesis and applications

The past decades have seen a great interest in the carbon nanostructures by its inherent features, e.g. thermal and electronic conductivities, as well as their relevance in the development of photovoltaics and fuel cells, sensors, opto-electronics, and thermoelectric devices [1,2]. Our research group has a great interest in graphene production methodologies and enhances the performance in different applications. In this way, the present work explores two ways to obtain ERGO using a combination of Hummers[3] and potentiostatic methods. In the first one, GO was obtained starting from an aqueous solution containing graphite in H_2SO_4 and thereafter reduced using different potentials from 1-4V and AISI 304L as electrodes. The second formulation, was starting from GO produced by the traditional modified Hummers method, followed by a pulse potential method at 1.6 and 2.0 V in H_2SO_4. Samples characterization were realized using Raman spectroscopy, Ultraviolet-Visible (UV-Vis), Fourier transform infrared (FTIR), X-ray diffraction patterns (XRD) and X-ray photoelectron spectroscopy (XPS). The results shown that depending of the synthesized method the ERGO can be obtained in solution or as a film on the metallic substrate, with potential applications.

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Figures

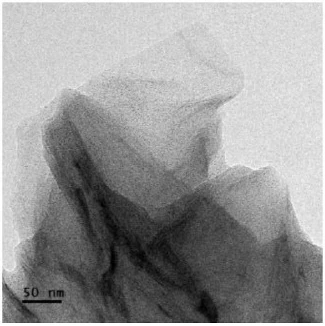

Figure 1: TEM image of ERGO at 1.6V

Olivier Malenfant-Thuot
Germain Robert-Bigras, Luc Stafford, Michel Côté
Département de physique, Université de Montréal, 2900 boul. Édouard-Montpetit, Montréal, Canada

olivier.malenfant-thuot@umontreal.ca

Ab Initio Calculations of Nitrogen Functionalization of Graphene

A new way of incorporating single nitrogen atoms in a graphene sheet, which involves a treatment in the late afterglow part of a plasma, has been developed by an experimental team [1,2]. It aims to maximize the substitutional, or graphitic, doping while minimizing the damage done to the graphene. With the software package ABINIT [3,4], we have carried out some first-principles calculations of different configurations of doping nitrogen atoms in a graphene sheet. We obtained the formation energies of the structures to study whether they are realistic configurations. We found that vacancies in the graphene sheet facilitate the incorporation of nitrogen atoms and, in particular, that the substitutional doping has the lowest formation energy. We also studied the energetic of nitrogen atoms adsorbed above the carbon plane because it plays a role in the process of functionalization. Using the Nudged Elastic Band method (NEB), we were able to calculate energy barriers for the diffusion and the in-plane absorption of these atoms.

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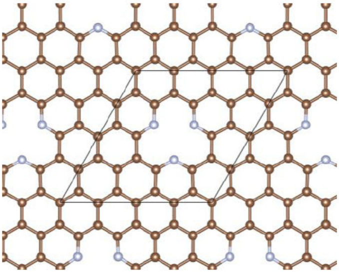

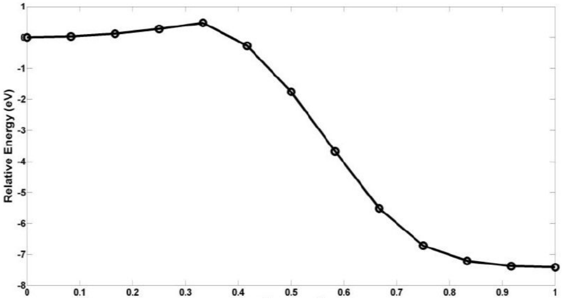

Juyeong Oh
Seong Chan Jun*
Department of Mechanical Engineering, School of Engineering, Yonsei University, 262 Seongsanno, Seodaemoon-gu, Seoul, Korea
*scj@yonsei.ac.kr

Graphene oxide based high-frequency modulation in electronics via optical illumination

Here, we report variation of scattering parametric properties in graphene oxide via illumination control. Graphene oxide was synthesized by chemically exfoliating graphite so that it has band properties while graphene has zero bandgap at K point. Since optoelectronic properties are highly affected by band gap properties, graphene oxide has been attracted tremendous attractions due to its controllable property [1-3]. Also, graphene oxide has potential for electronic devices since it partially has sp^2 orbital which is basic building block for graphene [4]. However, observation in high frequency transmission modulated via optical signal has not been explored sufficiently, particularly, for demonstration of controlling wave intensity and wavelength for high frequency optoelectronics. In this demonstration, we prepared two types of graphene oxide device; first one is thick enough for low intensity to investigate intensity relation while the other is thin to verify photon energy relation.We hope that this investigation will be a foothold for high frequency transmission experiment for graphene oxide and other materials under illumination.

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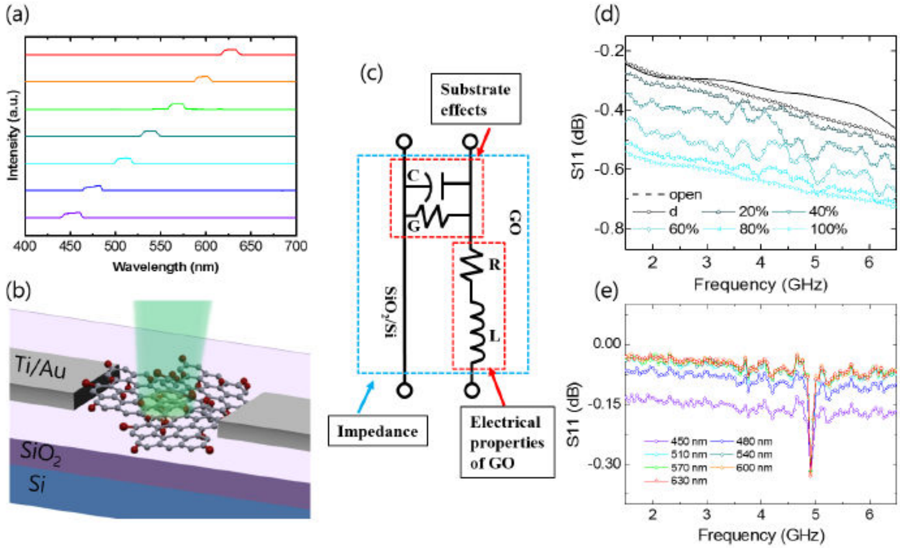
(a)
Intensity (a.u.)
400
450
500
550
600
650
700
Wavelength (nm)
(b)
Ti/Au
SiO_2
Si
(c)
Substrate effects
C
G
GO
R
L
SiO_2/Si
Impedance
Electrical properties of GO
(d)
S11 (dB)
-0.2
-0.4
-0.6
-0.8
open
d
20%
40%
60%
80%
100%
2
3
4
5
6
Frequency (GHz)
(e)
S11 (dB)
0.00
-0.15
-0.30
450 nm
480 nm
510 nm
540 nm
570 nm
600 nm
630 nm
2
3
4
5
6
Frequency (GHz)

Diby Benjamin Ossonon
Daniel Bélanger
Département de chimie, Université du Québec à Montréal, CP8888, Succursale Centre-Ville,
Montréal, QC, H3C 3P8, Canada
belanger.daniel@uqam.ca

Functionalization of Graphene Sheets During the Electrochemical Exfoliation of Graphite in Aqueous Media

Graphene [1,2] is undoubtedly one of the most attractive material for researchers, consequently the most intensively studied material due to its potential applications in several different areas. Various approaches are being developed for its large-scale preparation by green and cost-effective method [3,4]. One of the best methods is the electrochemical exfoliation of graphite in an aqueous solution [5]. However, for several applications graphene must be functionalized either non-covalently or covalently. In this work, graphene sheets produced electrochemically were functionalized during the exfoliation process. The resulting materials were characterized by several techniques such as Raman spectroscopy, thermogravimetric analysis, TEM, SEM, electronic conductivity measurements and electrochemical techniques.

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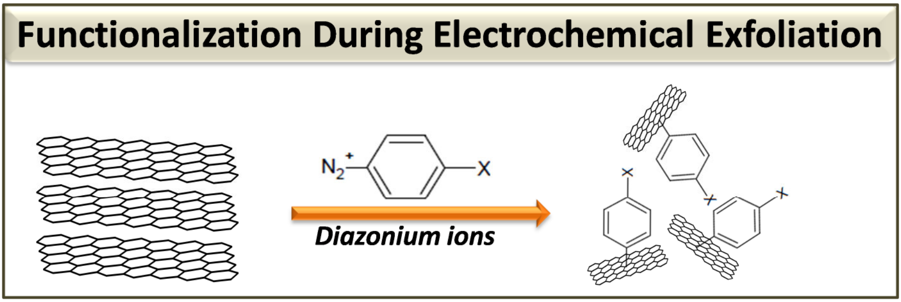

Jaewoo Park

Paul Bazylewski, Arash Akbari*, Sabastine Ezugwu, Reg Bauld, and Giovanni Fanchini

[1]Physics & Astronomy, University of Western Ontario, 1151 Richmond St, London ON N6A 3K7, Canada
*Present address: Université de Sherbrooke, 2500 boul. de l'Université, Sherbrooke, QC J1K 2R1, Canada

jpark627@uwo.ca

Copper nanoparticle-assisted etching of graphene layers

It has been demonstrated that metal etching by strong acids in the presence of graphene oxide and graphene layers promotes a certain degree of additional oxidation of the carbon backbone, breaking some C=C bonds, with the formation of C=O and C-OH groups.[1,2] If metallic nanoparticles on graphene are used for this oxidation process,[3] pores may locally form in the correspondence of locations at which C=C bonds are broken. At higher concentration of metallic nanoparticles, or when nanoparticles adhere more strongly to graphene, etching involves the complete removal of the graphene layer.[4] In this presentation we will review two distinct case studies of copper nanoparticle (Cu-NP) assisted etching of graphene, which offer a complementary vision of this startling material.

In the first study (Figure 1, [4]) Cu-NPs grown on graphene films by radio-frequency sputtering adhere well on flat graphene surfaces, but not on graphene wrinkles and ridges. By etching these Cu-NP's in nitric acid, we can remove the largest portions of flat graphene flakes, leaving behind only ridges and wrinkles as individual graphene nanoribbons (GNRs). Two classes of GNRs are formed in this way: wide GNRs from planar ridges and narrow and straight GNRs from vertical wrinkles.

In the second study (Figure 2, [3]) thermally evaporated Cu-NPs weakly adhere on graphene thin films and their etching in nitric acid leads to porous graphene-based membranes offering energy-efficient water filtration at relatively low differential pressures (down to 30 kPa) while still exhibiting high performance in terms of adsorption of metal ions at the part-per-million level.

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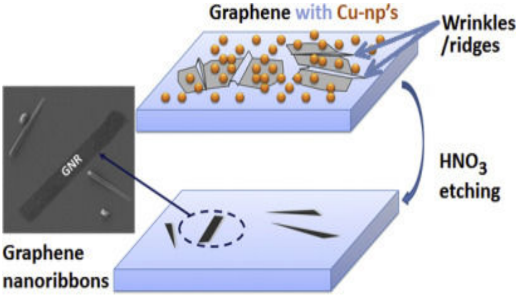

Figure 1: Etching of Cu-NP's grown by radio-frequency sputtering on graphene , leaving behind wrinkles and ridges as graphene nanoribbons.[4]

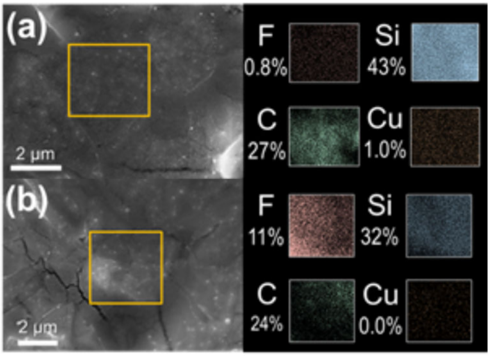

Figure 2: Etching of Cu-NP's grown by thermal evaporation on a graphene thin film on Teflon™ substrate: (a) SEM prior to and (b) after etching, which leaves behind a nano-porous graphene-based material suitable for water purification at low differential pressures. The higher detection rate of fluorine from Teflon™ substrate is a consequence of higher sub-nanoporosity[3]

Felix Rösicke[1,2]
Marc A. Gluba[1], Guoguang Sun[3], Timur Shaykhutdinov[3], Karsten Hinrichs[3], Jörg Rappich[1] and Norbert Nickel[1]

[1]Helmholtz- Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany
[2]SALSA - School of Analytical Sciences Adlershof, Berlin, Germany
[3]Leibniz - Institut für Analytische Wissenschaften - ISAS - e.V., Department Berlin, Berlin, Germany

felix.roesicke@helmholtz-berlin.de

Characterization of pre-transfer functionalized graphene

Large area graphene was successfully covalently functionalized with p-aminophenyl (AP) and p (N maleimido)phenyl residues (MP) from the respective diazonium salts, prior to transfer. The success of the functionalization was confirmed by Raman backscattering spectroscopy and infrared spectroscopic ellipsometry (IRSE). Furthermore, we have investigated the thickness and spatial distribution of the MP residues, using infrared spectroscopic ellipsometry and infrared atomic force microscopy, prior and after the transfer process. The data show consistently a MP layer thickness of 4.5-4.8 nm. Both functionalizations were used for subsequent modification with either carboxylic acids (AP-functionalized) or thiols (MP-functionalized). Furthermore, functionalized graphene sheets were transferred to various substrates, including glass, SiO_2, PTFE tape and aluminium foil. Considering the variety of available diazonium cations and possible further modifications, this opens the possibility to the use of modified graphene as tailored modular building block for specific surface functionalization.

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A. Sarkissian[1]

C. Côté[1], R. Nouar[1], R. Porter[1], S. Wolfe[1], M. Ionescu[2], K. Voon[3], K.D. Chowdhury[3], A.Nojeh[3], G.R. Bigras[4], L. Stafford[4]

[1] Plasmionique Inc, Varennes, QC, Canada
[2] NRC, London, ON, Canada
[3] UBC, BC, Canada
[4] University of Montreal, QC Canada

Various Synthesis Technologies and Applications of Nanostructured Allotropes of Carbon

Potential applications of nanostructured carbon allotropes have drawn considerable attention from various industrial fields, due to their potential applications stemmed from their outstanding electrical, mechanical, and optical properties. Example of applications in nanoelectronics, nano-electromechanical system (NEMS), flexible electronics, advanced energy production and storage devices, like batteries and fuel cells and super capacitor, as well as gas and chemical sensors, to name a few, are driving the global interest. Their unique characteristics placed them among the most promising molecular building blocks in nanotechnology. However, various applications require structures with different building blocs; therefore, it is necessary to develop specific strategies for selective growth, with excellent control over the size, electrical, mechanical and optical characteristics, in order to fully exploit their potentials. Moreover, some applications, particularly those related to nanoelectronics and NEMS, will require doping of these molecular structures with various dopants in order to modify their electrical properties.

There are variety of techniques used for fabrication of various allotropes of carbon nanostructures. Plasma- and vacuum assisted synthesis techniques have been used as a strategy to assist the synthesis at lower temperatures, since plasma-assisted dissociation of precursors used for synthesis, facilitate the nanomaterial growth at low temperatures [1]. Plasma techniques have also been used to dope graphene and CNT with nitrogen [1,2], an important step for nanoelectronic applications. In this presentation we will discuss various synthesis strategies and their specific advantages. We will also present characteristics of some samples produced from different techniques, and finally will present example of applications related to energy production using thermoelectric convertors [4] and development of gas sensors.

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Faranak Sharifi
Giovanni Fanchini
The University of Western Ontario, Physics and Astronomy Department, 1151 Richmond St. N6A 3K7, London, Canada
fsharif6@uwo.ca

Photo-Induced Open Circuit Voltage in Graphene-Based Organic Photovoltaics and Its Origin

We investigated the origin of open circuit voltage in graphene-based organic photovoltaics (OPVs) from I-V measurements at 1 Sun irradiation and using Kelvin Probe Force Microscopy (KPFM) under laser illumination. In this work we utilized poly(3-hexyl-thiophene):phenyl-C_{61}-butyric acid methyl ester (P3HT:PCBM) blend as a photoactive layer to fabricate bulk hetherojunction OPVs. Our experiments demonstrated that thin P3HT:PCBM OPVs assembled on transparent graphene electrodes exhibit higher open circuit voltages ($V_{oc} \approx 0.79 \pm 0.01$ eV) than identical ITO-based OPVs ($V_{oc} \approx 0.60 \pm 0.01$ eV). Both previously reported models, the band energy offset [1] and the metal-insulator-metal models [2], failed to explain the high open circuit voltage in graphene-based OPVs. Using KPFM under illumination, we found that the work function of graphene thin film electrodes strongly increase with increasing intensity of illumination and the amounts of photoexcited holes injected into graphene, as shown schematically in Figure 1. This result led us to propose a dynamic graphene-insulator-metal model (d-GIM) based on the specific zero-band gap semiconducting properties of graphene. Our d-GIM model predicts an open circuit voltage of $V_{oc} = 0.74$ V for thin graphene-based OPVs that compares well with the value of $V_{oc} = 0.79 \pm 0.01$ V directly measured from I-V curves. Our d-GIM model sheds light on the factors that influence controlling the open circuit voltage in virtually any ultrathin OPVs. Since the efficiency of OPVs is proportional to the open circuit voltage, our findings can be seen as a critical step towards improving the performance of organic photovoltaics.

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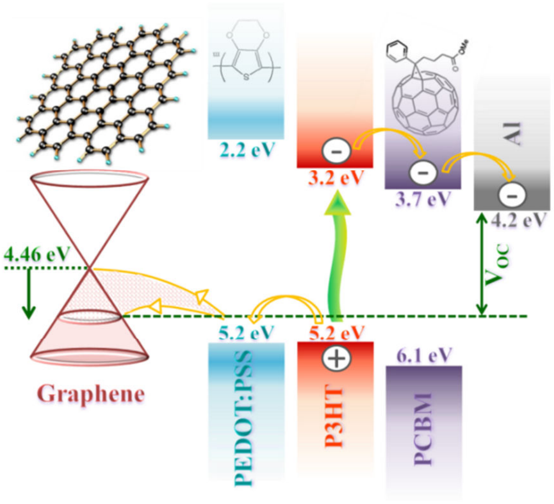

Figure 1: Schematic of hole generation and Fermi level shift in graphene-based solar cells under illumination. The shift in the Fermin level and the subsequent increase in the work function of graphene lead to larger difference between the work function of Al top contact and graphene electrode. This difference corresponds to the open circuit voltage.

Filipa Simoes[1]
Nitin Batra[1]; Bashir H. Warsama[2]; Christian G. Canlas[3]; Tahir F. Yapici[2]; Pedro M. F. J. Costa[1]

[1] King Abdullah University of Science and Technology (KAUST), Physical Science and Engineering Division, Thuwal 23955-6900, Saudi Arabia
[2] King Abdullah University of Science and Technology (KAUST), Analytical Core Laboratory, Thuwal 23955-6900, Saudi Arabia
[3] King Abdullah University of Science and Technology (KAUST), Advanced Imaging and Characterization Laboratory, Thuwal 23955-6900, Saudi Arabia
*filipa.fernandessimoes@kaust.edu.sa

Elemental quantification of wet digested Nanocarbons and characterization of their residues

It is estimated that Nanocarbons will be incorporated in our daily life in about three decades. Despite the accumulated knowledge on these materials, a pressing need to develop viable strategies for industrial-scale standardization persists. In addition, it is recognised that even minute amounts of other elements beyond carbon (e.g. transition metals used as growth catalysts) could have a deleterious effect not just on the aggregated physicochemical response of these materials but also in the environment and human health.[1]

Currently, the "gold standard" technique to quantify impurities in Nanocarbons is the neutron activation analysis.[2] However, as it relies on a neutron source, a nuclear reactor is needed, and so it is not suitable for routine analysis. On the other hand, inductively coupled plasma (ICP) techniques are widely available in chemistry laboratories and commonly applied to the study of vestigial quantities of metals in solid materials. Unfortunately, its applicability to Nanocarbons (e.g. graphene and nanotubes) has suffered from the lack of efficient digestion steps and certified reference materials (CRM).[3]

In this study, various commercial and certified graphitic carbon materials were subjected to a "two-steps" microwave-assisted acid digestion procedure. Following this, the concentration of up to 18 elements was analysed by ICP coupled to optical emission spectrometry (OES). Successful quantification of the certified elements was achieved, hence validating our wet digestion approach. Its applicability was further confirmed for a commercial single-walled carbon nanotube (SWCNT) sample. Where possible, the digestion residues were characterised by Raman spectroscopy, transmission electron microscopy and nuclear magnetic resonance (NMR) as we strived to understand the structural disintegration mechanism for graphitic carbons. It was seen that NMR, despite being less commonly used in the study of Nanocarbons, may effectively be the most valuable tool to access the digestion degree of these materials.

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M. Sojková[1]
M. Hulman[1], Š. Chromik[1], E. Dobročka[1], D. Machajdík[1,2], A. P. Kobzev[2]

[1]Institute of Electrical Engineering, SAS, Dúbravská cesta 9, 841 04 Bratislava, Slovakia
[2]Joint Institute for Nuclear Research, Dubna, Moscow Region, Russia
michaela.sojkova@savba.sk

MoS_2 and $MoSe_2$ thin films fabrication

Two dimensional (2D) materials have gained great interest in recent years due to their unique electronic, mechanical, and optical properties. Especially transition metal dichalcogenides (TMDs) with the formula MX_2, where M is a transition metal (Mo, W, and so on) and X is a chalcogen (S, Se, or Te), have attracted much attention due to their layer structure and semiconducting properties [1]. Their structure consists of a plane of metal atoms sandwiched between two planes of chalcogen atoms by covalent interaction and different layers are held together by van der Waals interactions [2]. These layered materials exhibit many distinctive characteristics such as outstanding flexibility, moderate carrier mobility and layer dependent electronic and optical properties. This makes TMD materials suitable for various applications such as transparent and flexible field effect transistors (FETs), photodetectors, photovoltaic cells, light-emitting diodes and catalysts [1].

Among metal dichalcogenides, molybdenum sulfide (MoS_2) materials have been extensively investigated for its interesting properties including enhanced optical absorption, thermoresponsive photogeneration, efficient hydrogen evolution reaction capability, valley polarization and high on/off ratio with low subthreshold swing, which can lead to ultrathin and highly efficient photovoltaics, photo-thermoelectrics, catalysis for sustainability [3]. Moreover, by quasi-continuous electrostatic carrier doping achieved by the combination of HfO_2 and ionic liquid, a superconducting state was achieved in MoS_2 [4]. Recently, monolayer $MoSe_2$ has started to gain attention because it has many interesting electronic and optical properties similar to those of monolayer MoS_2, such as a direct band gap, strong photoluminescence (PL), and a large exciton binding energy. It is known that sulfur defects in the MoS_2 monolayer greatly affect the electronic transport and optical properties [1]. Therefore, it would be meaningful to carefully compare the properties of monolayer MoS_2 and $MoSe_2$.

A large effort has been made to develop methods for growing high quality ultrathin films. Still, mechanical exfoliation is the most widely used way to prepare MoS_2 and $MoSe_2$ monolayers. On the other hand, the method is inherently not suitable for up-scaling. As an alternative, a chemical vapor deposition (CVD) is capable of producing large area MoS_2 and $MoSe_2$ films. Recently, physical vapor deposition, especially pulsed laser deposition (PLD) has successfully been used for the fabrication of MoS_2 thin films [5].

Molybdenum disulfide and molybdenum diselenide thin films were prepared by a two-step method on different substrates. First, molybdenum thin films were magnetron sputtered on different substrates (Si, sapphire). Second, the thin films were sulfurized / selenized in vapors of sulfur or selenium at high temperatures in a furnace. As-prepared films were characterized by XRD analysis, Raman spectroscopy, Rutherford backscattering and SEM techniques. Afterwards, the films were patterned to different types of structures using photolithography and ion beam etching. Electrical conductivity of the patterned structures were investigated. Finally, we discuss the structural and electrical properties of MoS_2 and $MoSe_2$ thin films and structures made of them.

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J.L. Valverde[1]
A. Romero[1], L. Sanchez-Silva[1], M.P. Lavin-Lopez[2]

[1]University of Castilla-La Mancha, Avenida Camilo Jose Cela S/N, 13071, Ciudad Real (Spain)
[2] Graphenano S.L., Calle Pablo Casals 13, 30510, Yecla, Murcia (Spain)
JoseLuis.Valverde@uclm.es

Improving the growth of monolayer CVD-graphene over polycrystalline iron sheets

Graphene is a two-dimensional carbon nanomaterial with hexagonal structure. The extraordinary properties of graphene make it an ideal material for a wide variety of applications [1]. Two different strategies can be distinguished for graphene synthesis, such as *Bottom-Up* and *Top Down*. The first one is based on the synthesis of graphene from a carbonaceous gas source. The second one is based on the synthesis of graphene using graphite as the raw material [2]. Within the *Bottom-Up* strategy is included Chemical Vapor Deposition (CVD) method. This method highlights because it is simpler and easier to scale than the other ones. In addition, it is possible to obtain large areas of graphene. Raman spectroscopy is the most widely used technique to perform quickly structural and electronic characterization of graphitic materials, besides being a non-destructive characterization method [3].

The principal aim of this work was the synthesis and optimization of CVD-graphene synthesis using polycrystalline iron as catalyst with the purpose of increase the percentage of monolayer graphene grown over the metal substrate. Although graphene is only one atom thick, it is visible at the optical microscope. In this investigation, four different colors could be distinguished in optical microscope images of the graphene samples, corresponding each one with one type of graphene. To study the quality of the samples, an Excel-VBA application was designed. This application analyzed the optical microscope images obtained and allows to know the percentage of each type of graphene deposited over the iron. Based on those percentages, the Excel-VBA application assigns values between 0 and 1000 to quantify a quality value of the sample. If 100% of the sample is coated with multilayer, few-layer, bilayer or monolayer graphene, the application assigns a value of 1, 10, 100 or 1000, respectively. Therefore, the closer to 1000 is the quality value; the higher the percentage of monolayer graphene coating the metal foil [3].

To achieve the aim of this investigation, different synthesis variables which affect the graphene quality were optimized (reaction time, CH_4/H_2 flow rate ratio and total flow (CH_4+H_2) during the reaction step at different reaction times). It was observed that the percentage of monolayer graphene and thus, the quality of graphene sample increased with the optimization of the synthesis variables, reaching the 62.4% of the sample covered with monolayer graphene and a value of 642 of graphene quality.

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Figures

% MONOLAYER GRAPHENE	62.4
% BILAYER GRAPHENE	24.7
% FEW-LAYER GRAPHENE	8.8
% MULTILAYER GRAPHENE	4.3
QUALITY VALUE	642.5

Table 1: Percentage of each type of graphene and quality value of the optimum sample. *(Synthesis conditions: 1025 ºC, CH_4/H_2= 0.25 v/v, 80 Nml/min at 7 min)*

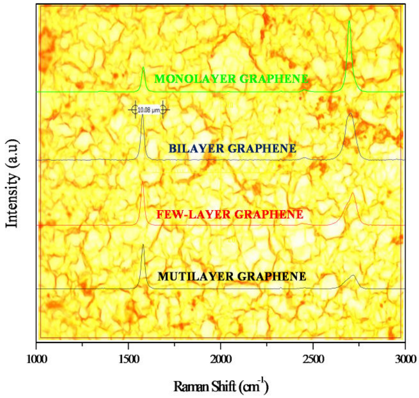

Figure 1: Optical microscopy and Raman spectroscopy of the optimum sample. *(Synthesis conditions: 1025 ºC, CH_4/H_2= 0.25 v/v, 80 Nml/min at 7 min)*

Zhongyang Wang[1]
Linfan Tian [1, 2], Siwei Yang [3]

[1.] Shanghai Advanced Research Institute, Chinese Academy of Science, Shanghai, China
[2.] School of Physical Science and Technology, ShanghaiTech University, Shanghai, China
[3.] Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Science, Shanghai, China

wangzy@sari.ac.cn

Green, simple and large scale synthesis of N-doped graphene quantum dots with uniform edge groups by electrochemical bottom-up synthesis

As a new kind of photoluminescence (PL) material, graphene quantum dots (GQDs) have drawn extensive attention of researchers because of their remarkable properties. It deserves to note that the specific PL mechanism of GQDs is the fundamental issue for its development. It has been demonstrated that the size, heteroatoms, edge carbon atoms, defects and edge groups all obviously affect the PL progress of GQDs [1, 2]. As the inevitable constituent part of GQDs, edge groups have a significant influence on the PL spectra. However, by now, the real role of edge groups in PL process is unclear. This can be due to the unavoidable different edge groups in most reported GQDs [1]. Thus, it is very necessary for the development of new preparation approach for GQDs with uniform edge groups.

We developed an electrochemical bottom-up synthesis of N-doped GQDs (N-GQDs) with large amount of well-defined edge groups for the first time [5]. This progress is green (both atom utilization and yield are higher than 95 %, without by-products), simple and suitable for large scale synthesis. The exclusive edge group of the obtained N-GQDs is -NH_2 because of the amino oxidative coupling of o-phenylenediamine, which results in unique excitation wavelength independence behavior. The high quantum yield is believed to originate from the localized π state due to the graphitic doping of nitrogen [3, 4]. The edge groups have little influence on the spectral resolved PL lifetime as well as the size distribution [5].

References

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Figures

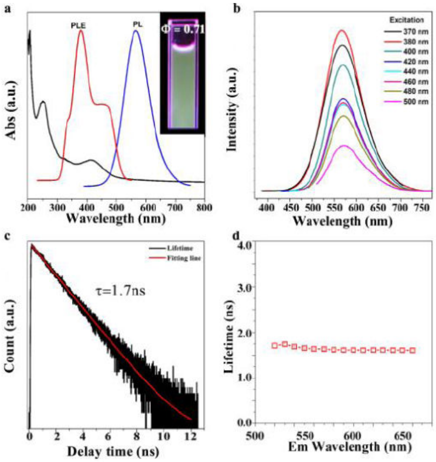

Figure 1: (a) UV-vis absorption, PL (λ_{ex}=420 nm) and PLE (λ_{em}=569 nm) spectra of N-GQDs aqueous solution, inset: the photograph of N-GQDs aqueous solution under 365 nm UV light. (b) PL spectra of N-GQDs aqueous solution at different excitation wavelengths (370-500 nm). (c) The lifetime of N-GQDs measured at 25 °C (λ_{ex} =420 nm and λ_{em}=570 nm). (d) PL decay curves of N-GQDs tested at 25 °C with 10 nm increments of progressive emission wavelength (λ_{ex}=420 nm).

Hyong Seo Yoon
Seong Chan Jun
School of Mechanical Engineering, Yonsei University, Seoul, Korea
scj@yonsei.ac.kr

Contact performance enhancement based on isotype junction effects of thickness dependent MoS_2 layers

In early 2000s, graphene was shown to have excellent carrier mobility as a zero-gap semiconductor. [1] Since its discovery, various materials in 2D crystalline structures have garnered significant attention because of their unique properties, which can be used in various fields of academics and industry. [2-4] Recently, transition metal dichalcogenides (TMDs) have become a hot topic of interest due to their remarkable electronic performances and their potential to replace silicon-based systems. Single-layer MoS_2 may be the most popular material among TMDs showing clear n-type semiconductor characteristics with a direct-energy band gap that is similar in size to silicon and can be used as an active layer in various electronic devices.

In this work, the difference in the work functions of MoS_2 nanosheets with different thickness has been observed based on the relative surface potential values measured by a scanning Kelvin probe microscopy technique. From the energy structure change, a layer-dependent MoS_2 isotype heterojunction shows small potential barriers and weak current-rectifying effects. However, the small potential barrier at the junction easily becomes negligible with a 2D contact area that is obtained by the polymer-assisted dry transfer technique showing only linear and symmetric current features. Therefore, we can obtain significant enhancement of contact performance by applying a multilayer MoS_2 structure between the MoS_2 channel and the metal electrode because two small potential barriers are more advantageous than one large Schottky barrier and they allow increment of the effective contact area.

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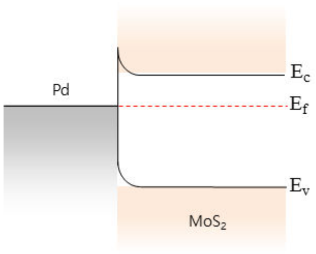
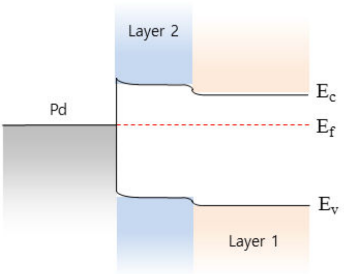
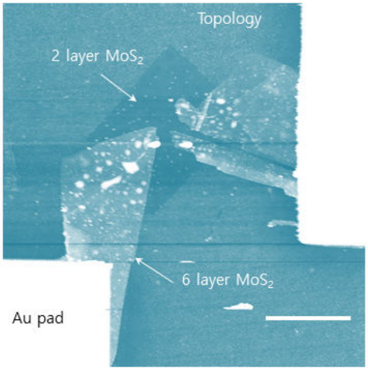
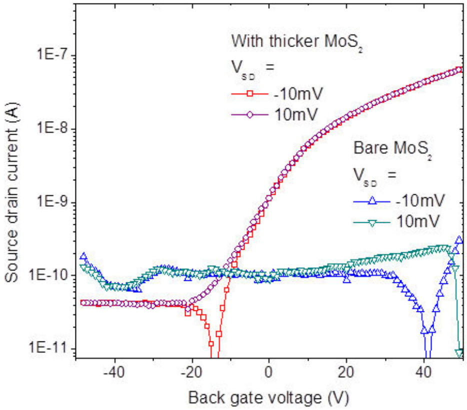
Figures


Pd
E_c
E_f
E_v
MoS_2


Layer 2
Pd
E_c
E_f
E_v
Layer 1


Topology
2 layer MoS_2
6 layer MoS_2
Au pad


With thicker MoS_2
V_{SD} =
-10mV
10mV
Bare MoS_2
V_{SD} =
-10mV
10mV
Source drain current (A)
1E-7
1E-8
1E-9
1E-10
1E-11
-40
-20
0
20
40
Back gate voltage (V)

Jin Zhang[1]
Songlin Yang[1Δ], Longyi Chen[1Δ], Paul Ogilvie[2]

[1]Department of Chemical and Biochemical Engineering, University of Western Ontario, London, Ontario, Canada, N6A 5B9
[2]Saint Jean Carbon Inc. 86 Wilson Street, Oakville, ON L6K 3G5

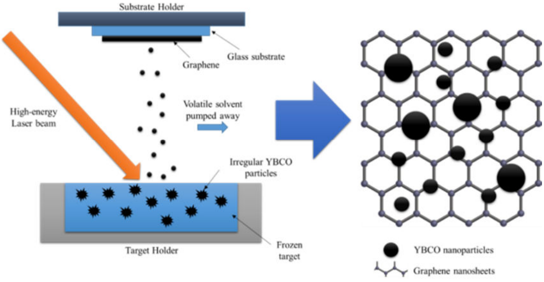
[Δ]S.L. Yang and L. Y. Chen contributed equally

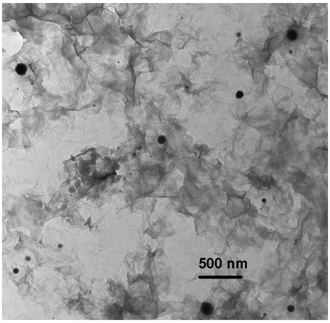
jzhang@eng.uwo.ca

Graphene/YBCO Hybrid Nanosheets Prepared by Matrix Assisted Pulsed Laser Evaporation

Hybrid graphene nanosheets by depositing yttrium barium copper oxide (YBCO) superconductor particles were developed by using the matrix-assisted pulsed laser evaporation (MAPLE), a free-contamination method. With increasing irradiation time, the amount of YBCO nanoparticles deposited on graphene is increased. In addition, the microstructures and elemental composition of YBCO nanoparticle deposited on graphene sheet by the MAPLE process were studied in terms of particle size and shape as a function of the deposition time/irradiation time (*t*). It is noted that the shape and size of the YBCO nanoparticles are more uniform with increasing t. When t increases to 2 hours, the average diameter of the spherical YBCO nanoparticles deposited on graphene sheets is around 50 ± 10 nm. This study demonstrates that MAPLE is a suitable process for depositing inorganic superconductor nanoparticles on graphene sheets without additional chemical agents.

Figures





Edited by:

PHANTOMS foundation

Alfonso Gómez 17
28037 Madrid – Spain

info@graphenecanadaconf.com
www.graphenecanadaconf.com