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TOKYO (JAPAN) - JANUARY 29-30, 2019

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FOREWORD

On behalf of the Organising Committee we take great pleasure in welcoming you to Tokyo for the first edition of the **1&2DM 2019** International Conference.

A plenary session with internationally renowned speakers, an industrial forum with extensive thematic workshops in parallel and a significant exhibition featuring current and future Graphene, Nanotubes and other 1&2DM developments will be highlighted at the event.

1&2DM 2019 will bring together, from a global perspective, scientists, researchers, end-users, industry and policy makers in an environment of cooperation and sharing towards the challenges of 1&2DM commercialization.

We truly hope that **1&2DM 2019** serves as an international platform for communication between science and business.

We are also indebted to the following Scientific Institutions and Companies for their help and/or financial support: Grafoid Inc. and Elsevier (FlatChem).

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

One thing we have for granted: very few industries, one way or another, will escape from the influence of 1&2D Materials and the impact on businesses is here to stay.

Hope to see you again in the next edition of **1&2DM** to be held in 2020.

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In the National Innovation Council meeting on the 29th of October 2009 chaired by the Right Honorable Prime Minister, Nanotechnology was identified as one of the new growth engines for the New Economic Model (NEM).
On the 14th of February 2011, the National Innovation Council convened and agreed that a nanotechnology commercialisation agency was needed and corresponding activities must be aligned with Agensi Inovasi Malaysia's (AIM) initiatives.
NanoMalaysia Berhad was incorporated in 2011 as a company limited by guarantee (CLG) under the Ministry of Science, Technology and Innovation (MOSTI) to act as a business entity entrusted with nanotechnology commercialisation activities. Some of its roles include:

- Commercialisation of Nanotechnology Research and Development
- Industrialisation of Nanotechnology
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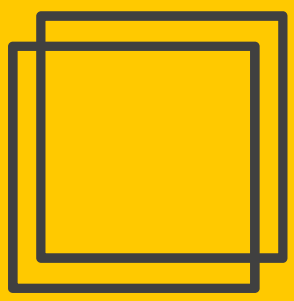
SPEAKERS



PLENARY SPEAKERS

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Materials in the Flatland

One of the most important “property” of graphene is that it has opened a floodgate of experiments on many other 2D atomic crystals: BN, $NbSe_2$, TaS_2, MoS_2, etc. The resulting pool of 2D crystals is huge, and they cover a massive range of properties: from the most insulating to the most conductive, from the strongest to the softest. If 2D materials provide a large range of different properties, sandwich structures made up of 2, 3, 4 … different layers of such materials can offer even greater scope. Since these 2D-based heterostructures can be tailored with atomic precision and individual layers of very different character can be combined together, - the properties of these structures can be tuned to study novel physical phenomena or to fit an enormous range of possible applications, with the functionality of heterostructure stacks is “embedded” in their design.
Already now such materials bring to life a number of exciting applications. In my lecture I will review some of them.

KEYNOTE SPEAKERS

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A step forward towards the large scale production and industrialization of graphene

In this talk I will illustrate the development of industrial-scale, reliable, inexpensive production processes[2] for the implementation of 2D materials in flexible (opto)electronics and energy applications.
I will show how the production of 2D materials by solution processing[2,6] represents a simple and cost-effective pathway towards the development of 2D materials-based (opto)electronic and energy devices, presenting huge integration flexibility compared to other production methods. I will first present our strategy to produce 2D materials on large scale by wet-jet milling[7] of their bulk counterpart and then an overview of their applications for flexible and printed (opto)electronic and energy devices. [3,8,9,10,11,12,13,14]

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Berry Phase Controlled Optical Responses in 1D & 2D Transition Metal Dichalcogenides

MoS_2 and related semiconducting transition metal dichalcogenide (TMD) family is regarded as a gapped graphene systems, the properties of which are controlled by the structural symmetry. The direct band gap at K and –K points that appears in monolayer MoS_2 is regarded as a mass gap opened by the in-plane broken inversion symmetry as compared with the Dirac cone in monolayer graphene. Effects of structural symmetry on TMDs can be investigated not only by thinning TMDs from bilayer to monolayer, but also by wrapping up the 2D sheets to tubular structures. In this presentation, we would like to report a couple of subjects related to the Berry phase in 2D and 1D TMD nanostructures.

As a first topic, we report the exciton Hall effect [1]. Hall effect is a well known phenomena to determine the carrier density in semiconductors, in which carriers are subjected to the transverse motion due to the Lorentz force by the externally applied magnetic field. We found that exciton in monolayer MoS_2 shows a similar Hall effect without external magnetic fields. This transverse motion of excitons are driven by the Berry curvature of the K and K' bands, which works as an internal magnetic field in the momentum space. From the real space imaging of the exciton diffusion trajectories, we found that the valley diffusion length reached 2 □m at 50 K.

The second subject is related to TMD nanotubes. TMD nanotube was first synthesized a quarter century ago [2], however, the investigation of electronic properties have been started only several years ago [3]. Very recently, our group have discovered superconductivity in individual WS_2 multiwalled nanotubes through ionic gating [4], and photovoltaic properties of p-n junctions formed on the WS_2 nanotubes [5]. In particular we focus on the unique photovoltaic properties of WS_2 multiwalled nanotubes, which clearly reflects its structural symmetry.

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Production and Functionalization of Carbon Nanotubes for Energy Devices

(Sub)millimeter-long carbon nanotubes (CNTs) are attractive for use as current collectors in electrochemical energy storage devices due to their ability forming light-weight, self-supporting, electrical conductive, sponge-like films which can capture any capacitive materials. Such long CNTs have been synthesized on 2D substrates [1,2] but their efficient synthesis in 3D reaction space is important to meet the cost and scale requirements for such devices. We have realized batch and semi-continuous production of submillimeter-long single-wall [3] and few-wall [4,5] CNTs by fluidized bed chemical vapor deposition (FBCVD), in which 70% of C_2H_2 is converted to CNTs within 0.3 s (Figure 1) [4]. We use spherical ceramic beads instead of porous powder as catalyst support to retain CNT structure similar to on-substrate CVD and to enable easy separation of CNTs from the beads. The as-synthesized CNTs have a carbon purity >99 wt% with metal impurity <0.1 wt% [5] and ready for use without purification for many purposes. A tiny amount of CNTs as small as 1 wt% can capture capacitive particles of 99 wt% without any polymeric binder nor metal foil and yield self-supporting film electrodes, and a lithium ion full cell with $LiCoO_2$-CNT cathode and graphite-CNT anode is demonstrated (Figure 2) [6]. This CNT-based electrode architecture is effective to create electrodes of the emerging active materials such as Si anode [7] and S cathode with practically high gravimetric, areal, and volumetric loadings.

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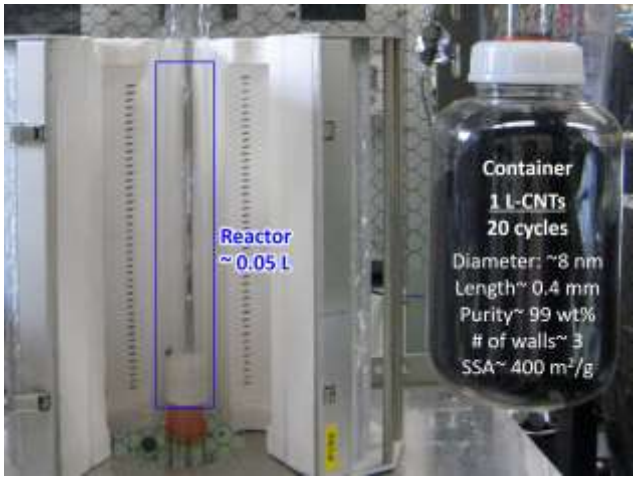

Figure 1: Semi-continuous, high-yield production of 500 μm-long, 99 wt%-pure few-wall CNTs by FBCVD [4].

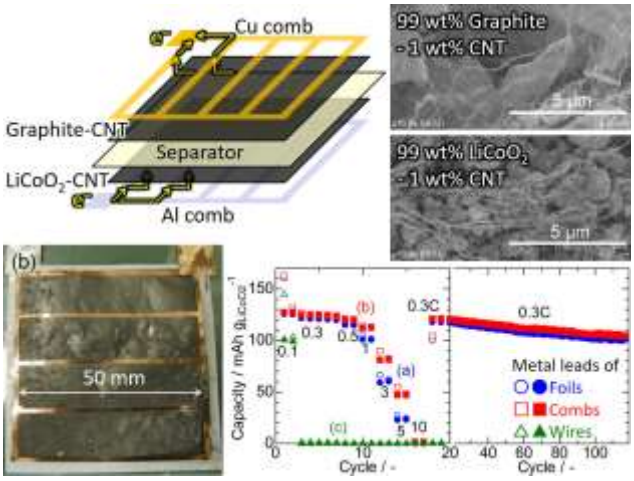

Figure 2: Lithium ion full cell with $LiCoO_2$ cathode and graphite anode based on CNT 3D current collectors [6].

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Graphene-based van der Waals Heterostructures towards a New Type of Quantum-Cascade Terahertz Lasers

Abstract
Current-injection or optical pumping makes population inversion of graphene carriers enabling lasing and/or amplification of terahertz (THz) radiation [1-3]. We've recently demonstrated 1-8-THz broadband amplified spontaneous THz emission as well as single-mode THz lasing at 5.2 THz both at 100K [3]. Introduction of a gated double-graphene-layered (G-DGL) van der Waals heterostructure in which gate-bias tuned THz radiation emission is obtained via plasmon- and/or photon-assisted quantum-mechanical resonant tunneling is a promising rout to further increase operation temperature as well as output intensity (Fig. 1) [4-5]. We experimentally demonstrated the proof of concept of such an operation mechanism [6]. The important physics behind is the acoustic plasmon modes in the DGL that can enormously enhance the quantum efficiency by orders for dc electric power to THz photo radiation power conversion in comparison with that for a simple graphene-channel transistor laser structure (Fig. 1) [5]. We have proposed a cascading of the G-DGL unit element working as a new type of THz quantum-cascade lasers (Fig. 1) [7]. The laser cavity can be structured along with the in-plane direction of the G-DGL mesa structure. The vertical G-DGL cascade structure can enlarge the mode field of the THz photon radiation to match the free-space impedance. Numerical analyses demonstrate further increase of the quantum efficiency of THz lasing by order of magnitude in comparison with a single G-DGL structure. Experimental verification is now under going. This work is financially supported by JSPS KAKENHI #16H06361, Japan.

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Figures

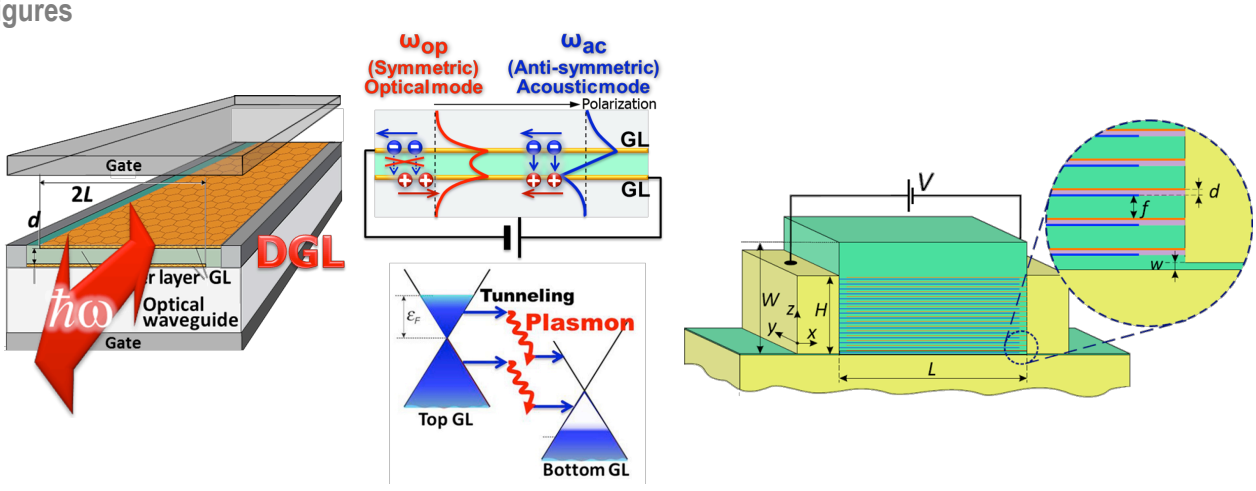

Figure 1: G-DGL structure (left), its plasmon modes and plasmon-assisted tunneling (center), and G-DGL cascade (right).

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Charge, Thermal and Spin Transport in Graphene Composites & Polycrystalline Heterostructures

Abstract

I will discuss charge, thermal and spin transport in chemically and structurally complex forms of graphene accounting from substrate effects, polycrystalline morphology of CVD graphene (and hBN), and chemically functionalization; all aspects being of crucial relevance for the development of applications in flexible and transparent electronics, energy harvesting and spintronics. Multiscale simulation and predictive modelling will be shown to enable simulations of physical properties in realistic models of very large system sizes (with up to 1 billion atoms), reaching the experimental and technology scales.

After introducing some challenges about the modelling of graphene composites I will present quantitative analysis of charge and thermal transport properties in graphene materials in presence of structural imperfections as produced during the wafer-scale production of graphene through chemical growth (CVD), the chemical transfer to versatile substrates, and the device fabrication. Fundamental properties of charge mobilities in polycrystalline graphene, accounting the variability in average grain sizes and chemical reactivity of grain boundaries as observed in real samples grown by CVD will be presented, together with their relevance for device optimisation and diversification of applied functionalities such as chemical sensing [1].

In a second part, I will also briefly explain the current state-of-the-art in understanding spin transport in graphene and how spin manipulation can be engineered through the fabrication of van der Waals heterostructures, fostering progress towards the design of non-charge based revolutionary information processing and computing

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Doped Graphene for Enhanced Raman Spectroscopy

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Doped Graphene for Enhanced Raman Spectroscopy

This talk will discuss the synthesis of large-area, high-quality monolayers of nitrogen-, silicon- and boron-doped graphene graphene sheets on Cu foils using ambient-pressure chemical vapor deposition (AP-CVD) [1-3]. Scanning tunneling microscopy (STM) and spectroscopy (STS) reveal that the defects in the doped graphene samples arrange in different geometrical configurations exhibiting different electronic and magnetic properties. Different substitutional nitrogen sites could be controlled by controlling the CVD temperature, flowrate and the duration of the doping precursor insider the reactor. Interestingly, these doped layers could be used as efficient molecular sensors in conjunction with Raman spectroscopy.

We will also show that B-doped graphene (BG) exhibits unique sensing when detecting toxic gases such as NO_2 and NH_3 [3]. The detection limit for BG can reach as low as 95 ppt for NO_2 gas and 60 ppb for NH_3 gas. BG has enhanced sensitivity values of 27 and 105 times better than graphene for NO_2 and NH_3 detection [3]. This is attributed to the presence of B atoms within the graphene lattice that results in a high affinity to both donor and acceptor molecules, leading to stronger interactions between the molecules and graphene.

Finally we will discuss the use of graphene for enhancing Raman signals for specific molecules, also known as graphene enhanced Raman scattering (GERS). NG and Si-doped graphene (SiG) could significantly enhance the Raman signal of fluorescent molecules: Rhodamine B (RhB), crystal violet (CRV) and methylamine blue (MB) [1]. SiG exhibits an enhancement factor 10-40 times higher than that of PG (see Fig. 1) [1]. We will also show that by using NG as a substrate, Raman signals of dye molecules can be detected at a record low concentration of 10^{-11} M [4]. This is very close to single molecular detection. However, this enhanced Raman sensing requires the Fermi energy of the substrate to match the LUMO level of the probe molecule, which allows an effective charge-transfer excitation [4].

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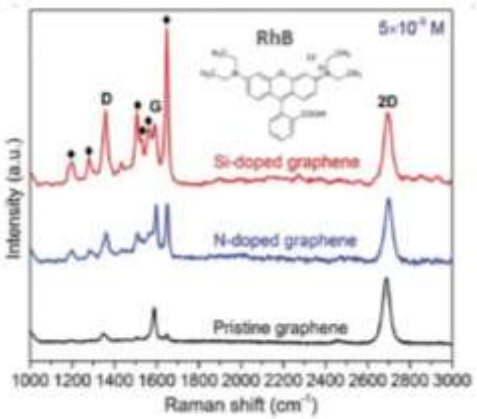

Figure 1: Comparison of Raman enhancement effect when RhB molecules are applied on top of pristine, N-doped and Si-doped graphene respectively.

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Topological Properties of Graphene and Related 2D Materials

In atomically-thin materials such as graphene and transition metal dichalcogenide nanosheet, the electronic properties crucially depend on the size, edge structures and topological properties of the system. It is well-known that graphene zigzag edges possess edge states at Fermi energy. The presence of edge states in graphene provides peculiar magnetic properties and perfectly conducting channel for electron conduction [1].

The origin of edge states can be understood from the topological properties of bulk wavefunctions. There are two fundamental topological quantities i.e. Berry curvature and Berry connection, which are understood as magnetic field and vector potential in momentum space, respectively. The origin of graphene edge states is attributed to the existence of nonzero Zak's phase (integration of Berry connection) of bulk wavefunction. This is distinct difference from conventional topological insulators where the existence of topological edge states is guaranteed by the nonzero Berry curvature owing to the presence of spin-orbit interactions.

In my talk, I will briefly give overview of edge and nanoscale effects on electronic transport properties of graphene nanostructures [1]. After that, we shall discuss a two-dimensional lattice model which exhibits a nontrivial topological phase in the absence of the Berry curvature on the basis of two-dimensional Su-Schrieffer–Heeger (SSH) model [2]. Since this system possesses both time-reversal and inversion symmetries, Berry curvature is zero but finite Berry connection. In spite of the absence of Berry curvature, the system leads to the robust edge states. Also, we discuss possible candidates of topological 2D materials, e.g. A3B biatomic sheet on the basis of first-principles calculations [3], and 2D photonic crystals [2]. Our approach will serve to design the topological 1D and 2D materials in absence of spin-orbit interactions.

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Figures

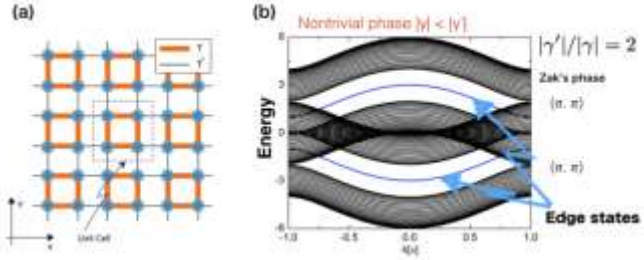
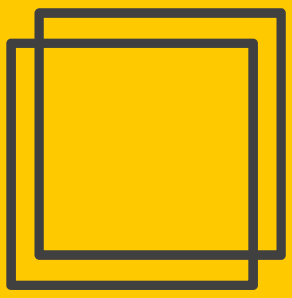


Figure 1: (a) 2D SSH model and (b) corresponding energy dispersion for ribbon structure in nontrivial phase.



INVITED SPEAKERS

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Engineering Graphene Electrodes for Flexible Display Applications

The recent advance in flexible, stretchable or wearable electronics derives the development of novel materials, which have high electrical or optical properties in addition to mechanical flexibility. In this perspective, two-dimensional (2D) materials, represented by graphene and related materials, have enormous potential to be exploited for the next-generation human-friendly electronic and optoelectronic systems due to their unique electrical, optical, and mechanical properties. During the last decade, we have demonstrated that versatile properties of graphene have been incorporated into the present-day electronic and optoelectronic technology, i.e., field-effect transistors, nonvolatile memory devices, chemical and biological sensors, plasmonic devices, and flexible display devices. However, there are still fundamental or technological issues to be addressed for the real applications of graphene to commercial electronics systems. In this talk, the recent advances in the display application of graphene from my research group will be presented. Specifically, I will discuss several engineering approaches, including reliable transfer methods for large-area graphene [1,2], surface or optical engineering for high-performance optical applications [3,4], and selective defect-healing techniques for improving the sheet resistance and mechanical property of graphene as an electrode material for flexible OLED devices [5].

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Bottom-up Chemical Synthesis of Atomically Precise Graphene Nanoribbons and Their Potentials

In contrast to zero-bandgap graphene, structurally confined graphene nanoribbons (GNRs) have open bandgaps due to the quantum confinement effect, making them interesting as next-generation carbon-based semiconductor materials.[1] The electronic and optical properties of GNRs critically depend on their chemical structures, in particular the width and edge structure, but conventional top-down methods like "cutting" of graphene sheets and "unzipping" of carbon nanotubes have failed to achieve the required precision. To this end, bottom-up chemical synthesis can provide narrow (~1–2 nm) GNRs with atomically precise structures.[1,2] In this talk, I will introduce our recent progresses in the synthesis of GNRs in solution and on surface and discuss their potentials. By the solution synthesis we can obtain long (>600 nm) GNRs that are dispersible in organic solvents and can be processed from the liquid phase.[3] The GNR edges can be functionalized with different substituents. For example, GNRs functionalized with extended aromatic units demonstrated unique rectangular self-assembly behavior[4] and introduction of organic radicals induced magnetic edge state through spin injection, interesting for spintronic and quantum computing applications.[5] On-surface synthesis of GNRs is typically performed under ultrahigh vacuum (UHV), but also possible under low vacuum to ambient pressure with an industry viable setup of chemical vapor deposition (CVD), producing high-quality, large-area GNR films.[6] We have for example achieved fabrication of low-bandgap *N* = 5 armchair GNRs (5-AGNRs) and their integration into field-effect transistor devices. Lateral fusion of 5-AGNRs into wider *N* = 10 and 15 armchair GNRs could be demonstrated upon annealing at higher temperatures, showing optical absorption over 2000 nm, which could be interesting for infrared sending applications.

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Figures

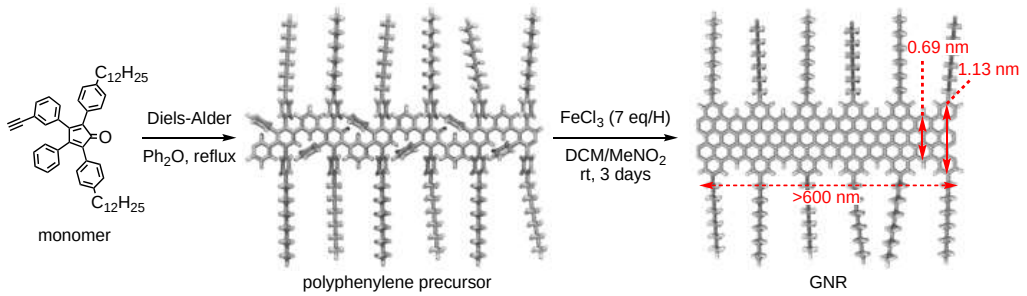

Figure 1: An example for bottom-up solution synthesis of graphene nanoribbons.[3]

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Carbon nanotube thin films for wearable electronics application

Wearable healthcare devices have the potential to revolutionize preventive medical care and health promotion technologies. Carbon nanotube thin films are promising electronic materials for transistors and integrated circuits [1,2], biosensors [3], and other passive components to build flexible and stretchable devices with excellent wearability and performance because of the high-carrier mobility, mechanical flexibility, and biocompatibility. In the presentation, after reviewing recent progresses of carbon nanotube-based electron devices, our recent works on flexible integrated circuits and biosensors for wearable devices are introduced. A concept to design carbon nanotube-based analog integrated circuits, which are indispensable for sensor devices, is presented, with a demonstration of the first carbon nanotube differential amplifiers on a flexible plastic film. Energy harvesting technologies, which harvest electricity from small energy sources existing in environment, may be useful for the power source of wearable devices. The CNT-based transparent and stretchable triboelectric generators, utilizing the contact electrification and electrostatic induction, will be introduced. Some demonstrations such as driving 100 blue LEDs with a 5x5 cm^2 triboelectric generator, a generator-equipped gloves lighting with hand claps, and so on will be shown in the presentation.

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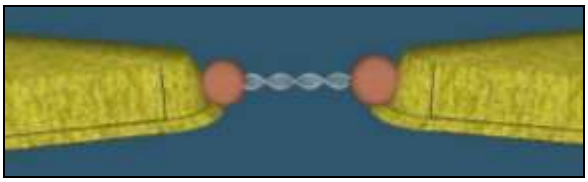
From 1D DNA nanowires to 2D nanotube networks



Charge transport through molecular structures is interesting both scientifically and technologically. To date, DNA is the only type of polymer that transports significant currents over distances of more than a few nanometers in individual molecules. Nevertheless and in spite of large efforts to elucidate the charge transport mechanism through DNA a satisfying characterization and mechanistic description has not been provided yet. For molecular electronics, DNA derivatives are by far more promising than native DNA due to their improved charge-transport properties.
In recent years we have invested great efforts to address the above issues. Measuring the charge transport in DNA was elusive due to great technical difficulties leading to various results. We recently devised an experiment in which double-stranded DNA is well positioned between metal electrodes. Electrical measurements give surprisingly high currents over 100 base-pairs (~30 nm) elevated from the surface. The temperature dependence indicates backbone-related band-like transport.
In collaboration with the Kotlyar group, We were also able to synthesize and measure long (hundreds of nanometers) DNA–based derivatives that transport significant currents when deposited on hard substrates. Among the molecules, metal containing DNA, which is true metal-organic hybrid, a smooth and thin metal coated DNA and G-quadruplex DNA.
Step by step we improve the synthesized constructs and the measurement methods of single DNA-based molecules.
Using the same measurement methods we characterize a 2D carbon nanotube network to understand the role of doping at the junctions as a basis for transparent electronics.

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Structures and properties of one-dimensional materials

One-dimensional materials can be as small as a single atom in width and are the simplest possible orderable structures, which makes them ideal platforms for the study of fundamental properties of matter [1]. Analytical technique at single atom level is crucial to diagnose the atomic structures of one-dimensional materials and then to predict their physical/chemical performance. In my presentation, single atom imaging and spectroscopy by means of electron energy-loss spectroscopy (EELS) inside TEM/STEM will be shown to discriminate individual atoms in one-dimensional materials. Examples for intriguing physical phenomena from this emerging class of materials, such as Peierls distortions [2, 3], phase changes [4, 5], structural confinements [6, 7], will be presented. Also their transport or optical properties are attempted to correlate with their atomic structures [4, 8, 9].

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Figures

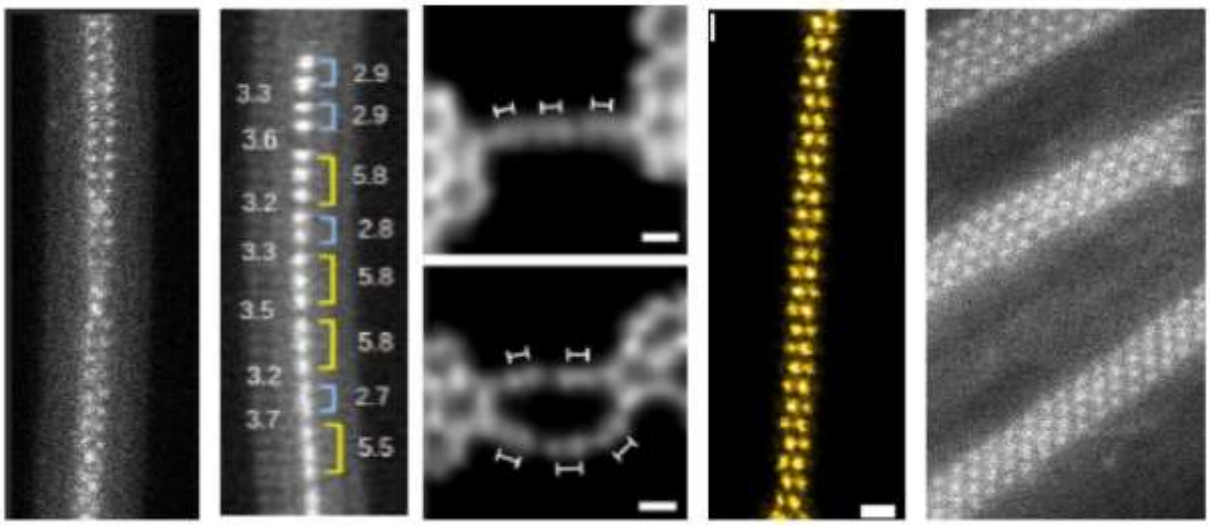
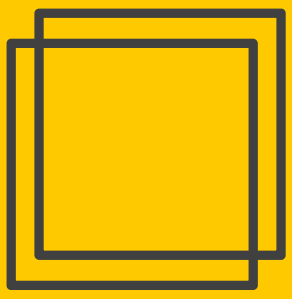


Figure 1: Examples for one-dimensional structures: (Left to right) Eu double atomic chains, iodine mono-atomic chains, carbon chains, MoSe nanowire, WS_2 nanoribbons.



INVITED SPEAKERS

PARALLEL SESSION

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Correlated Tip-Enhanced Optical Spectroscopy and SPM on 2D materials

Raman spectroscopy and confocal Raman microscopy have already proved to be essential characterization tools in many areas of advanced research, with a number of these applications extending into industry. As time moves on, new applications that are not addressed by existing technologies arise. Indeed, at the nanoscale, materials exhibit different properties than at the macro level, often quite dramatically different.

The characterization of nanomaterials naturally requires imaging techniques with resolution at the same scale or better, so that local property variations can be discerned and defects properly detected; only with this understanding can the material properties be engineered to meet the performance requirements of next-generation devices. In this talk, we will present new nano-imaging capabilities to perform such measurements. Tip-enhanced optical spectroscopies (TEOS) such as TERS (tip-enhanced Raman spectroscopy) and TEPL (tip-enhanced photoluminescence) provide a unique capability for the characterization of 2D materials [1, 2]. We will demonstrate the power and importance of the cross-correlation of nanoscale hyperspectal imaging with data from other scanning-probe techniques such as topography, surface potential, conductivity and photocurrent [3, 4].

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Figure

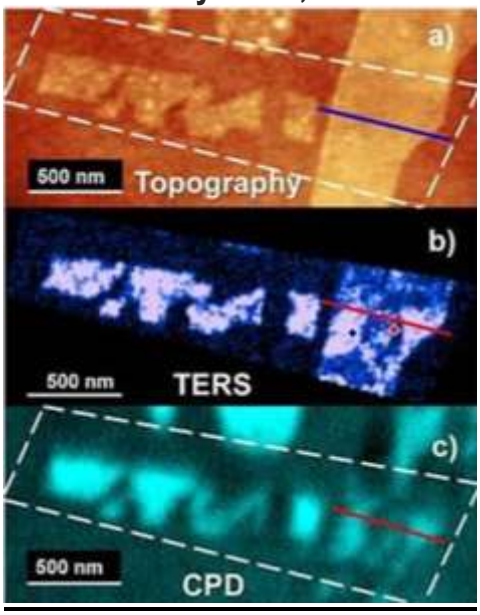

Figure: AFM topographic, TERS and surface potential images of WSe2 flakes

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Introducing 2D Materials for Magnetic Tunnel Junctions

The recent discovery of graphene, and other 2D materials, has opened novel exciting opportunities in terms of functionalities and performances for spintronics devices. While to date, it is mainly graphene properties for efficient spin transport which have been put forward, we will present here experimental results on another avenue for 2D materials in spintronics. We will show that a thin graphene passivation layer, directly integrated by low temperature catalyzed chemical vapor deposition (CVD), can prevent the oxidation of a ferromagnet [1]. This in turn enables the use of novel humid/ambient low-cost processes for spintronics devices, which would usually lead to oxidation during the fabrication and thus a quenching of the spintronic performances. We will illustrate this property by demonstrating the use of ozone based ALD processes to fabricate efficient spin valves protected with graphene [2]. Importantly, the use of graphene on ferromagnets allows to preserve a highly surface sensitive spin current polarizer/analyzer behavior and adds new enhanced spin filtering property [1][2]. Furthermore, we will present results concerning another 2D material isomorph to graphene: the atomically thin insulator hexagonal boron nitride (h-BN). Characterizations of complete spin valves making use of monolayer h-BN tunnel barriers grown by CVD will be presented [3]. These different experiments unveil promising uses of 2D materials for spintronics [4].

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Quantum-hybridization negative differential resistance from halide perovskite nanowires and vertical graphene junctions

In the effort to develop advanced electronic, optoelectronic, energy, and bio devices based on emerging low-dimensional materials, first-principles or *ab initio* simulations are playing an increasingly important role by providing atomistic information that are not easily accessible in experiments. For this purpose, a key ingredient that is still relatively immature and should be further developed is the capability to treat non-equilibrium open junction systems under finite bias in a first-principles manner. In this talk, I will first apply the existing machinery that combines density functional theory (DFT) and nonequilibrium Green's function (NEGF) formalisms and predict that ultrahigh negative differential resistance (NDR) can be obtained from the nanowires derived from recently synthesized one-dimensional (1D) halide perovskites [1]. Next, I will discuss the limitations of DFT-NEGF in simulating the finite-bias nonequilibrium electronic structure in nanoscale junctions and introduce the novel multi-space constrained search DFT (MS-DFT) formalism that we have recently developed. The MS-DFT formulation goes beyond the standard DFT-NEGF in several aspects, and as an application example I will consider graphene-based two-dimensional (2D) vertical heterosturcture tunneling transistors, which show NDR and are a promising platform to realize next-generation "More Moore" and "More than Moore" devices [2]. It will be emphasized that in both cases NDR arises from the novel "quantum-hybridization" NDR mechanism (Fig. 1).

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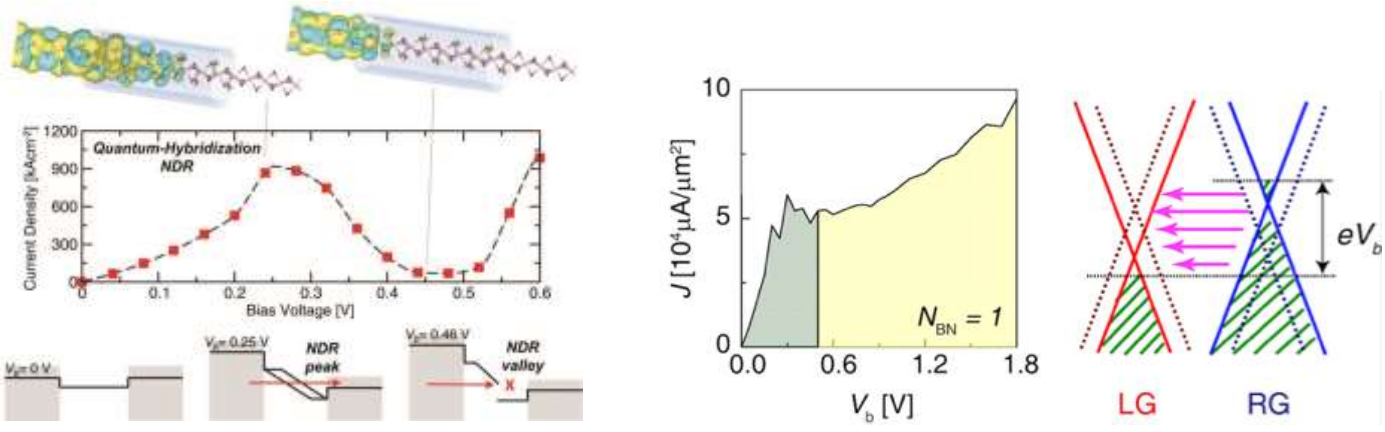

Figure 1: Quantum-hybridization NDR from 1D halide perovskite nanowires [1] (left) and graphene electrode-based 2D vertical tunneling transistors [2] (right).

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Graphene synthesis on 200 mm wafers

Graphene and graphene-based devices have demonstrated great potential to extend the functionality of a large variety of microelectronic applications [1, 2]. However, wafer-scale high quality graphene on CMOS compatible materials (dielectrics or semiconductors) is not yet solved. In this paper, we examine the potential of 200 mm, in-house grown epitaxial Ge(100)/Si(100) and Ge(110/Si(110) substrates for the development of Si-CMOS compatible graphene synthesis methods. CVD experiments have been performed at a deposition temperature of 885 °C using CH_4 as carbon source. The Raman results are shown in Fig. 1, where good quality graphene was detected over the entire Ge(100)/Si(100) substrate. The full development details and complementary electrical characterizations of both substrates will be presented in this work. Finally, it should be also stressed that the experiments in this work were carried out in a standard BiCMOS pilot-line, making this study unique, as its results might directly pave the way to further graphene integration and graphene-based device prototyping in mainstream Si technologies.

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Figures

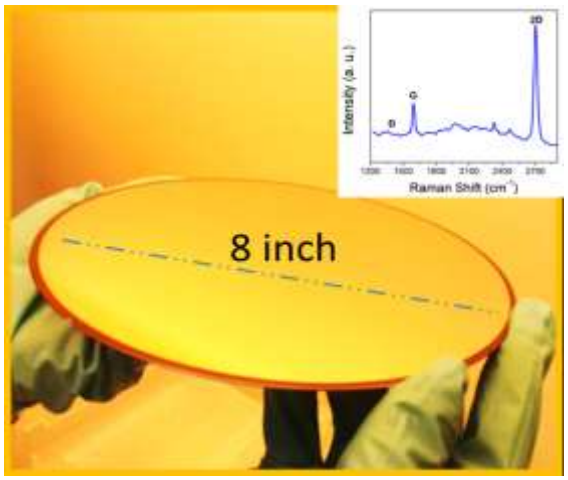

Figure 1: Graphene, grown on 200 mm Ge(100)/Si(100) wafer. A typical Raman spectrum of the Graphene is presented in the inset of Fig.1

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Topological Transport in Graphene/hBN Superlattices

Abstract

Heterostructures of atomic two-dimensional materials have attracted great interests because they form superlattice structures which allow band engineering as well as use of multiple degenerate degrees of freedom. In graphene with hexagonal boron nitride (hBN), energy gaps are induced when the crystal orientation is aligned with an angle of almost zero degree. The broken inversion symmetry induces Berry curvature in graphene, and the Berry curvature also induces topological current (valley Hall effect), which has been recently observed via non-local resistance measurement [1]. Here, we have fabricated hBN/single- or bi-layer graphene/hBN heterostructures with one-dimensional edge contacts as well as a Hall-bar geometry [2, 3]. Longitudinal- and Hall-conductance oscillations in magnetic fields have been observed, originating from the Hofstadter's butterfly which indicates the good alignment of graphene and hBN crystal orientation (Fig.1). We observed the non-local resistance at both a Dirac point and a secondary Dirac point (SDP) that is generated by band modulation due to the moiré superlattice [2, 3]. In single-layer graphene/hBN superlattices, giant non-local resistance at the SDP with the order of quantum resistance was observed even at zero magnetic field, indicating the occurrence of the quantum valley Hall state [2]. As a result of all transport measurements, we conclude that the mechanism driven by the edge states is a more likely scenario for the giant nonlocal resistance in the quantum limit than a bulk-related interpretation.

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Figures

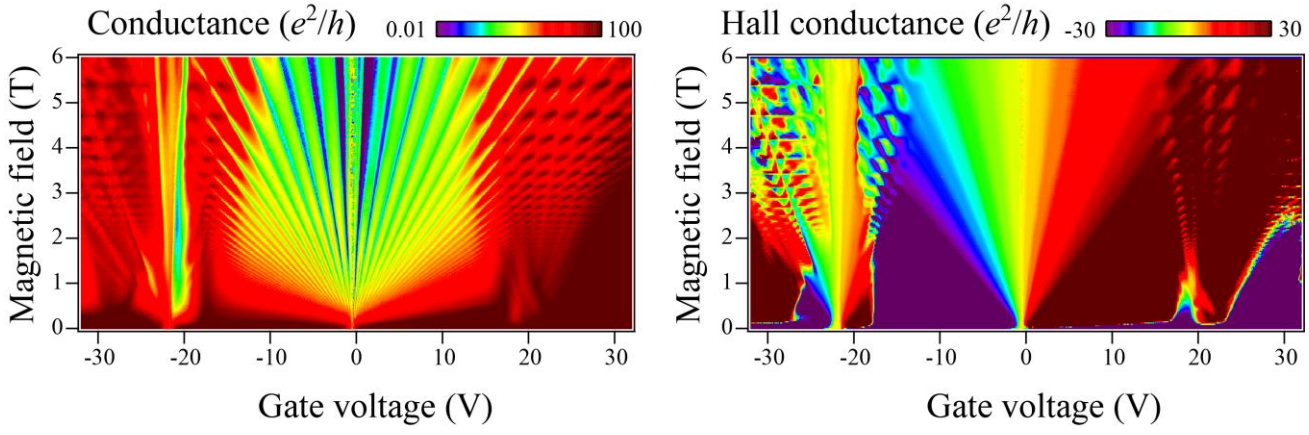
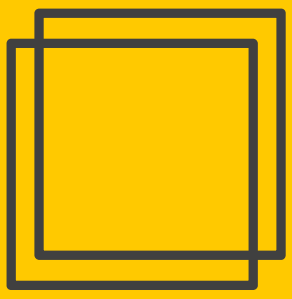


Figure 1: Longitudinal conductance and Hall conductance as a function of a gate voltage and a magnetic field applied perpendicular to the substrate at 6 K in single-layer graphene/hBN superlattice devices.



INVITED SPEAKERS

INDUSTRIAL FORUM

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Exploration of Single-layer Nanosheets and Their Assemblies with Emergent Properties

I will introduce our recent progress on the preparation, characterization and diverse application prospects of single-layer nanoshees derived from layered oxides and hydroxides. In particular, we succeed in the controllable synthesis and chemical exfoliation of highly crystallized layered hydroxides with various metallic compositions (Mg, Fe, Co, Ni, Zn, Al, etc.) by rationally designing the oxidation state as well as by modifying the coordination geometry of metal cations. To harvest a synergistic effect, a superlattice-like nanocomposite through molecular-scale hetero-assembly of single-layer nanosheets and graphene derivatives was fabricated. The combination of conductive graphene directly adjacent to redoxable nanosheets can greatly improve the overall charge transfer efficiency of the composites. Electrochemical characterizations identified the nanocomposites as ideal electrode materials for the development of high-performance energy storage devices, as well as efficient electrocatalysts. We also show that, upon exfoliation, single-layer hydroxide nanosheets exhibited exceptionally high in-plane ion conductivities approaching 10^{-1} S cm^{-1}, which were the highest among anion conductors and comparable to proton conductivities in commercial proton exchange membranes (*e.g.*, Nafion). In contrast, cross-plane conductivities of restacked nanosheets were much poorer, which were 4-5 orders of magnitude lower than the in-plane values. These results clearly revealed an exceptionally high and anisotropic hydroxyl ion conduction in single-layer nanosheets as a benefit of exposing the whole 2D surfaces. The exotic 2D conducting properties might promise a great potential as inorganic solid ionic conductors in a large array of energy- or environment-related applications.

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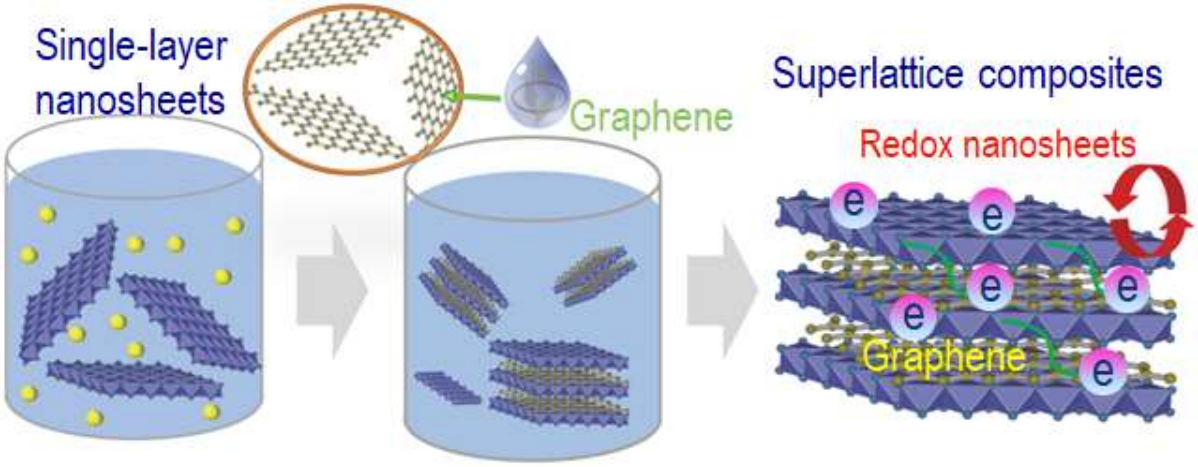

Figure 1: Molecular-scale hetero-assembly of single-layer nanosheets and graphene for superlattice-like composites.

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High-throughput synthesis of graphene by plasma CVD and its commercialization

It is necessary to establish high-quality and high-throughput graphene synthesis technique for the practical application of graphene transparent films. In this talk development of high-throughput plasma-enhanced CVD for high quality graphene and its commercialization will be discussed.
The plasma CVD is characterized by high-growth rate graphene atomic membrane compared with conventional thermal CVD (fig.1), which is suitable for the high-throughput production for the industrial use [1,2,3]. We have achieved a graphene membrane with a transmittance of 95% (two-layer) for visible light and sheet resistance of 130Ω (gold chloride doped) in A4 size by developing an original plasma CVD method. The grain boundary and residual strain in graphene synthesized by plasma CVD with high-growth rate is analyzed by scanning transmission electron microscopy (STEM) and Raman spectroscopy [4]. The connection between the grains of graphene by high-throughput synthesis was confirmed. The compressive strain remained in graphene, which affects electrical conductivity, was observed. (fig.2). We have established a start-up company for the commercialization of high-throughput synthesized graphene.

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Figures

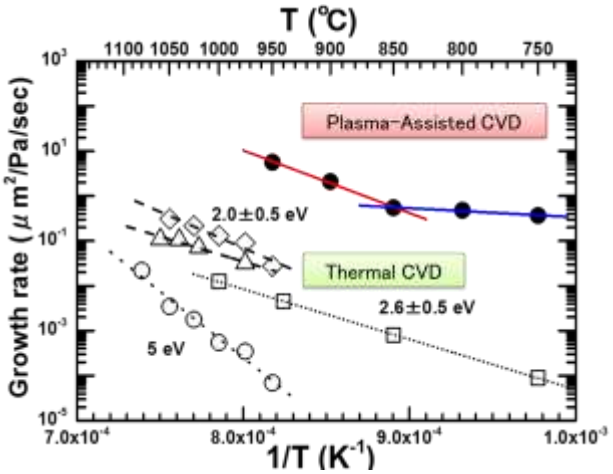

Figure 1: Temperature dependence of graphene growth rate for thermal CVD and plasma–assisted CVD which are normalized by CH4 partial pressure.

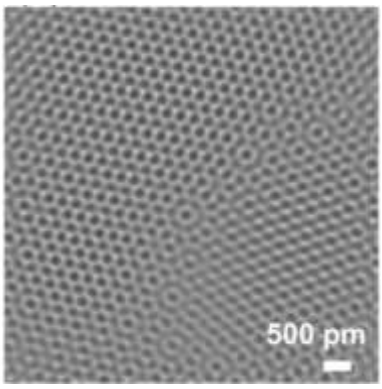

Figure 2: Grain boundary of graphene synthesized by high-throughput plasma CVD

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Synthesis and application of carbon nanotubes, graphene, and graphene nanoribbons

We work on growth of carbon nanotubes (CNTs), graphene, and graphene nanoribbons (GNRs), and their application to electronic devices, such as transistors, interconnects, and sensors. We explain some of our recent results. As for CNTs, we have been working on application of CNTs to interconnects [1, 2], and thermal interface materials (TIMs) [3]. In particular, we fabricated TIMs consisting of bundles of vertically aligned CNTs, where the density of CNTs was increased by a newly-developed compressing method [3]. The thermal resistance of the CNT-TIM was found to be as low as that of indium film [3]. We also work on applications using graphene and graphene nanoribbons (GNRs). In fact, graphene can be used for high-frequency wave detection [4]. We actually proposed a diode consisting of a GNR heterojunction (Fig. 1) for such a purpose [4] The heterojunction consists of a hydrogen-terminated armchair-edge GNR (H-AGNR) and fluorine-terminated armchair-edge GNR (F-AGNR). Since there is a difference in electron affinity between them, we can construct a staggered-type lateral-heterojunction p-n diode. Simulations show that, due to band-to-band tunneling, the diode has a nonlinear reverse current of the order of kA/m. The junction capacitance is extremely small due to the small junction area. It has been found that the diode can have a much better sensitivity for terahertz wave than a GaAsSb/InAlAs/InGaAs heterojunction diode [5]. Furthermore, we recently developed a graphene-gate transistor, where the gate of a Si transistor was replaced with single-layer graphene (Fig.2) [6]. This graphene-gate transistor can be used as a gas sensor. In fact, when gas molecules adsorb on the graphene-gate surface, the Fermi level or work function of graphene can change, thus shifting the threshold of the Si transistor. This causes changes in the drain current if the gate voltage is kept constant. This graphene-gate sensor is very sensitive to NO_2 and NH_3. In fact, we found that the sensor can detect NO_2 with concentrations less than 1 ppb.
This research was partly supported by JST CREST Grant Number JPMJCR15F1, Japan.

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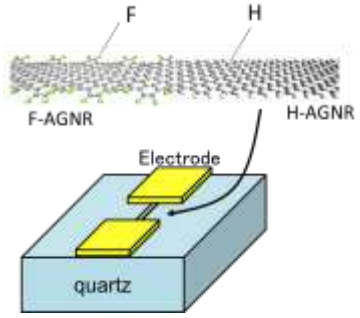

Figure 1: Illustration of a diode using a heterojunction of F-AGNR and H-AGNR.

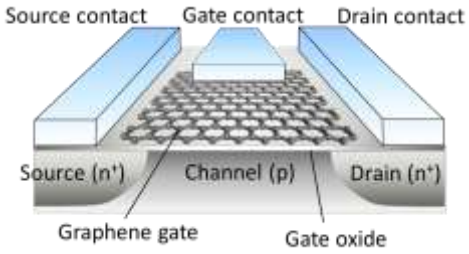

Figure 2: Schematic illustration of a graphene-gate transistor (sensor)

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Graphene Oxide and Graphene Oxide Derivatives, Properties and Applications

Abstract
Graphene oxide (GO), as prepared by the so called Hummers method[1] is a solid acidic compound that can be modified in a number of ways. It can be reduced, thermally, chemically or by light to become graphene-like rGO, partly reduced or fully reduced, it can be functionalized to become for example organophilic and it can be doped with N and B. The acidity of both GO and rGO can be reduced by washing with water or even neutralized by treatment with ammonium hydroxide or other bases. The sheets can be small or large, all these variations giving rise to a large family of related compounds with different properties, suitable for different application. Our company aims at offering all these varieties of GO to end-users, in Kg-quantities. Keeping in mind that GO is a metastable material, it is essential to understand the stability or shelf-life of all these different forms, as well as the most suitable storage conditions. In Figure 1 and 2 below we compare dispersibility, color and X-ray diffractograms of standard graphene oxide stored for 3 months, 3 years and 6 years respectively, observing that standard GO is fairly stable for years, still being fully dispersible after 6 years, although its composition has slightly changed as manifested by the color becoming darker. Now, we have undertaken a much wider study where we store a range of derivatives under different conditions (frozen, cool and ambient). These samples will be analyzed regularly over the coming years in order to exactly define the changes that occur and how fast. Our aim is to establish well defined standardized products spanning the entire parameter space of graphene oxide derived materials.
Potential applications of graphene oxide and graphene oxide derivatives include such diverse technologies as load-speaker membranes, water treatment, polymer composites, protective coatings, sports equipment, energy storage and medical applications. We see several applications now being piloted around the globe.

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Figures

Figure 1: Suspensions of GO aged 6, 3 and 0.3 years.

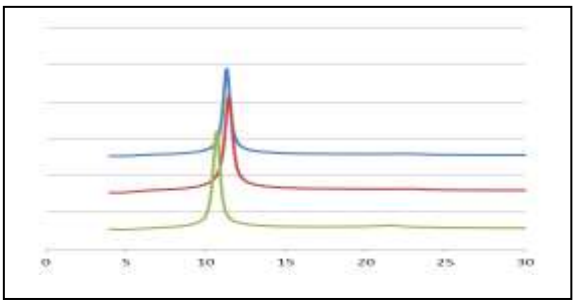

Figure 2: XRD patterns of GO aged 6, 3 and 0.3 years.

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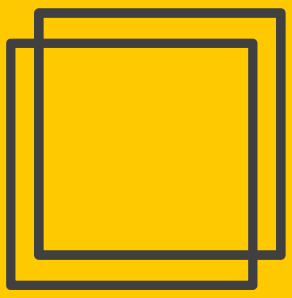
Grafoid and Focus Graphite: A Global Mine-to-Market Platform for Green Technology

Quality, price and security of supply will be the key drivers of the critical materials market for new energy applications. Those companies that can build a green technology supply platform to serve the global market will be the winners. Focus Graphite Inc. and Grafoid Inc. are doing just that: delivering value-added graphite products and graphene applications to meet the needs of a growing market - with unsurpassed quality, low prices and guaranteed supply for their customers.

Focus Graphite's Lac Knife natural graphite resource, located in northern Quebec, is formally recognized by the province as a key economic development project under Plan Nord's revitalization of the mining sector. Ranked by an independent study as the #1 natural graphite resource in the world, Lac Knife's high-grade (15%), high-purity (98.3%) output will offer one of the lowest costs of production in the world, at $335/ton. When in operation in 2020, annual production will be approximately 44,300/tpy. Lac Knife's spherical graphite products are unique when compared with other commercial grades of natural flake and synthetic graphite. With the successful purification of its fine flake graphite from to 99.99%, through a proprietary in-house process, all grades are battery quality. Focus is in battery testing programs with a number of potential customers. Over the past two years, an robust battery material testing program has been undertaken which demonstrates the superior quality of Focus's products. It shows extremely low irreversible capacity losses, nearly theoretical reversible capacities, and essentially zero capacity loss in long-term cycling tests. These improved capacities when compared with using synthetic graphite in the anode offer customers competitive advantages.

Focus Graphite owns 18% of Grafoid Inc., a world-leading graphene R&D, applications development and technology licensing company. It produces graphene and processes for transformative, industrial-scale graphene applications in partnership with leading corporations and institutions around the world. Grafoid targets four emerging graphene commercialization sectors: energy creation and storage, composites, coatings and membrane applications and technology. Founded in 2011, and headquartered in Kingston Ontario, Grafoid's Global Technology Centre (GGTC) houses the company's production facilities for Grafoid's proprietary suite of Mesograf™ graphene products, its research facilities in material science applications, and a center for JV application development. Grafoid has two security of supply graphite off-takes in place with Focus Graphite, one known as the "energy" off-take and the other known as the "polymer" off-take.

Together, both companies are posied to supply customers with valued-added graphite and graphene products to serve the Lithium-ion battery needs of EV and energy storage customers.



ORAL CONTRIBUTIONS

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Acoustic Implosions for Synthesis of Carbon Materials and Oscillations between Diamond and Graphite phases

Nonlinear modulation of microstructures concerns questions also relevant for understanding of the origin of life, material science, geo- and bio- science. Recent examples are the formation of chiral and hierarchically structured porous metal composites, epitaxial strain induced transitions in layered oxides, switchable infrared nanophotonic elements based on phase change materials, design of autonomous motors, etc.
However, the main question is, how to establish a dynamic control of useful characteristics, for example dynamic control of crystal / grain size and composition modulation in solids. A possible answer is to develop a new generation of dynamic impactors that can trigger oscillations of structures and functions.
In my talk I focus on ultrasonically triggered cavitation, that can be defined as growth and collapse of microbubbles, as a unique approach to generating a strong shock impact and, thus, a rapid increase of temperature and pressure at a localized area (<0.02 μm). At 20 kHz bubbles oscillate with a period of 50 μs. Adiabatic collapse of a bubble leads to electron temperature up to tens of eV. Thus, shock impact of oscillating bubbles creates highly non-equilibrium conditions for a dynamic modification of liquids and solids at microseconds time scale.
I will talk about the linearity of cavitation driven microstructural changes in metals, namely changes in Ni grain sizes and transformations of Ni phases in Ni based alloys vs. time of ultrasonic treatment.
Our work shows that the Interaction of microbubbles with surfaces drive several forces that lead to both grain growth and grain size reduction. The main questions are: Which forces drive grain growth and which forces trigger grain size reduction? What is the coupling mechanism that allows periodic switching between forces in the cavitating medium and leads to nonlinear effects in solids?
Furthermore, the efficiency of the ultrasonically modified Ni compounds in synthesis of carbon phases can be established as a tool for monitoring of the effects of cavitation on solids. I will demonstrate cavitation driven sp2 and sp3 carbon transformations on Ni surface.

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Laser-irradiated 2D materials: a new path towards straintronic devices

Graphene and transition metal dichalcogenides (TMDs) offer a unique platform to enable novel opto-electronic devices. Thanks to their peculiar physical and electronic properties such as bandgap tuneability, chemical functionalisation and the ability to sustain high levels of strain, such materials can enable flexible photodetectors, highly-efficient solar cells and unforeseen "straintronic" devices, i.e. devices which electronic properties are tuned by strain. Laser-irradiation can be used to modify the properties of 2D materials to enable new devices. Using this technique, we are able to define photoactive junctions in functionalised graphene which display an unprecedented linear dynamic range of 44 dB. This is achieved through the efficient quenching of hot-carrier effects in graphene and demonstrates the first purely photovoltaic graphene-based photodetector [1,2]. Laser-irradiation is then used to induce photo-oxidation in ultra-thin HfS2. In this way, a spatially varying bandgap can be engineered using a local strain field to enable the first observation of the so-called "inverse charge-funnelling" [3], as illustrated in Figure 1. This effect allows photo-excited charges to be driven away from the excitation area, towards regions of smaller gap, where they can be efficiently separated and collected. We observe an enhanced signal, with a 350 % improvement in the responsivity with respect to the pristine device, indicating efficient extraction of photogenerated carriers. The bias dependence of the photocurrent demonstrates that the measured signal is due to the inverse charge-funnelling enabled by the strain-engineered gradient of energy gap in the planar HfS2/HfO2 interface. Strain-engineering in 2D materials represents a new field with promising applications for a new generation of electronic devices, in particular when applied to superlattice heterostructures [4].

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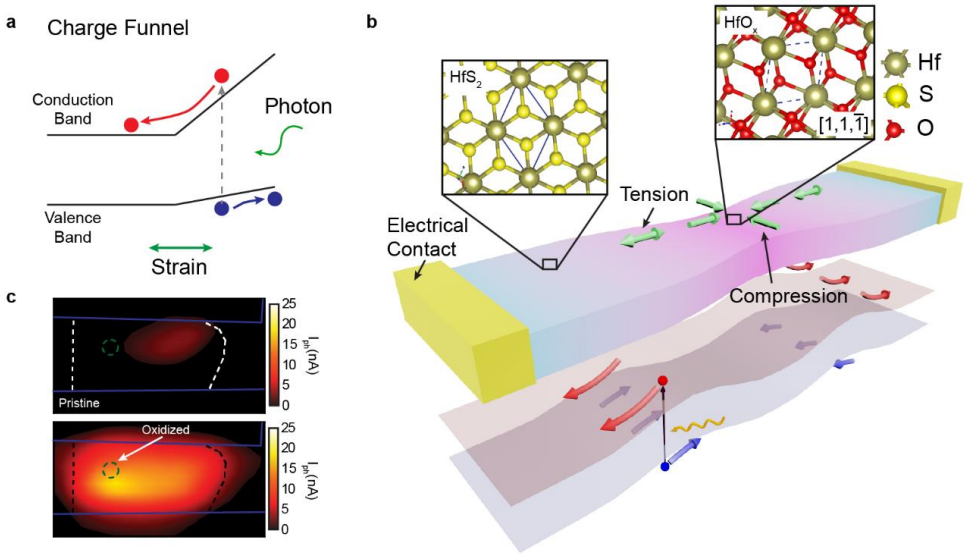

Fgure 1: a) Charge-funnel concept. b) Selective oxidation lead to lattice mismatch at the interface between HfS2 and HfOx creating a strain gradient which modulates the bandgap. c) Enhanced photoresponse is observed in proximity of the oxidised area as a result of charge-funnelling.

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Functionalization of the CVD-grown graphene: towards the on-surface chemistry reactions and motion

Controlled tuning of the CVD-grown graphene properties may open the gate for its potential application in optoelectronics, spintronics or sensors. Functionalization [1] or heterostructure formation [2] of the carbon nanostructures is currently considered as the most convenient method for efficient manipulation with their unique properties. It has been shown that the hydrogenation of graphene leads to the opening of the bandgap, enhancement of the spin-orbit coupling and appearance of the magnetic moment. Moreover, the prior hydrogenation of graphene enables its further functionalization with compounds that do not react with pristine graphene [3]. Namely, we have shown using Raman spectroscopy reactivity of the partially hydrogenated graphene with $KMnO_4$, KIO_4, as well as attachment of the benzyl groups due to alkylation with BnBr what was not achieved without the hydrogenation process. Moreover, functionalized graphene can be used as a substrate for the process of the dynamic covalent motion of the fluorescent nanodiamonds (fNDs) using fluorescence microscopy [4]. Multivalent imine linkages formed between aldehyde-decorated nanoparticles and monolayer graphene grafted with amino groups allowed reversible binding–unbinding upon periodic pH changes and oriented motion of the fND in pH gradient.

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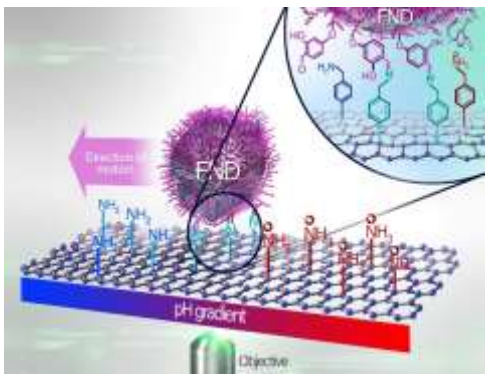

Figure 1: Directional motion of the aldehyde-decorated fND at amino-functionalized graphene as a result of the applying an external pH gradient in a microfluidic channel

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Magneto-photoconductivity in 2D strong spin-orbit coupling systems

Optoelectronic properties of 2D semiconductors and 3D topological insulators are of great interest. Among 2D semiconductors, transition metal dichalcogenides (TMDs) have band gap corresponding to the visible range of the electromagnetic spectrum. For instance, in molybdenum disulphide (MoS2) – a 2D semiconductor TMD the band gap energy is about 2 eV in monolayers. Monolayer MoS2 has a non-centrosymmertic crystal, inherent broken inversion symmetry which leads to a large spin-orbit coupling. At low temperatures, we observed a crossover from positive magneto-photoconductivity (MPC) to negative MPC of monolayer MoS2 backgated FET devices. This crossover was not observed for bilayer MoS2 devices. We explain this observation in terms of illumination and localization effects based on a recent theory. On the other hand, magnetic field dependence of the photocurrent in a 3D topological insulator was also studied. Among the 3D topological insulators bismuth telluride (Bi2Te3) has unique hexagonal warping and spin texture which has been studied by photoemission, scanning tunnelling microscopy and transport. We measured magneto-photoconductivity of 3D Bi2Te3 excited by 532 nm laser along the magnetic field perpendicular to the sample plane, at temperatures between 4 K and 30 K. As the magnetic field increases the measured photocurrent at zero degree angle of photon polarization exhibits a negative MPC which turns to be a constant above 4 T. When the thickness of the sample is decreased, the negative MPC turns to a positive magneto-photoconductivity above 4 T. This behavior of MPC indicates there are two kinds of conduction; one due to the bulk states and the other due to the surface states in Bi2Te3 and they can be distinguished by MPC measurements.

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Structure analysis of Ca-intercalated bilayer graphene by total-reflection high-energy positron diffraction

Recently, superconductivity (SC) induced in graphene has been attracting attention [1-3]. Many of the Graphene intercalation compounds show superconductivity. An example is Ca-intercalated bilayer graphene; the resistance steeply drops at 4 K and reaches zero at 2 K under zero magnetic field [1]. The SC mechanism has been understood in analogy with graphite intercalation compounds (GICs). However, the structure of Ca-intercalated bilayer graphene is not just a 2D limit of GICs because of the existence of buffer layer between graphene and SiC substrate. It is unclear whether or not the structure of Ca-intercalated bilayer graphene on SiC substrate is the same as that of GIC. Here we have analyzed the structure of Ca-intercalated bilayer graphene on SiC(0001) by using total-reflection high-energy positron diffraction (TRHEPD) at KEK. The positive charge of the positron allows TRHEPD to be a remarkably surface-sensitive tool [4], as the penetration depth of the positron under the total reflection condition is less than approximately 2 Å. Figure 1(a) shows a TRHEPD pattern from Ca-intercalated bilayer graphene. The $\sqrt{3}\times\sqrt{3}$-*R*30° streaks and spots originate from the intercalated Ca atoms. Figure 1(b) shows a TRHEPD rocking curve which is glancing angle dependence of a specular (00) spot intensity. The theoretical curve based on the structural model of Fig. 1(c) is in good agreement with the experimental result. This model where the Ca atoms are intercalated between graphene and the buffer layer is different from the previous expectation [1]. In the presentation, we show the details of the experiment and discuss the structure compared with GIC.

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Figures

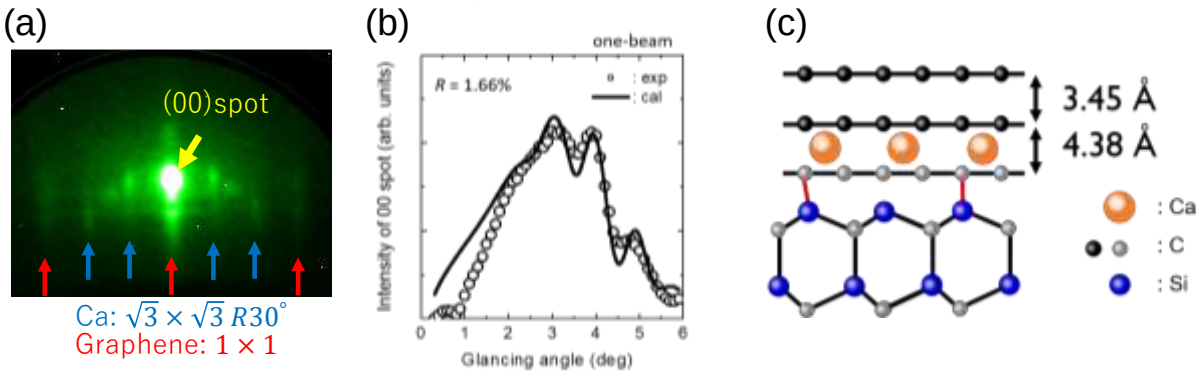

Figure 1: Structure analysis of Ca-intercalated bilayer graphene on SiC(0001). (a) TRHEPD pattern, (b) experimental rocking curve (circles) and theoretical curve (line), (c) structural model determined by TRHEPD.

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Thermally reduced graphene oxide for CO_2 capture technology

CO_2 capture process represents typically about 70% of the total cost of the carbon capture and storage (CCS) chain. One of the most promising technologies for CO2 capture is based on the adsorption process using solid sorbents. As a member of this family, the metal–organic frameworks (MOFs) are well recognized for their extraordinary surface area, ultrahigh porosity, and most importantly the flexibility to tune the porous structure as well as the surface functionality. For more efficient utilization of MOFs sorbents, several hybrid systems based on MOFs with other solid sorbents have been investigated in order to utilize the synergism between the two sorbents and therefore ultimately improve the overall performance in CO_2 separation. Moreover, sorbents such as activated carbons, reduced graphene oxide (rGO) and Carbon nanotubes (CNTs) provide the added feature of high surface area and easily functionalized sites, which contribute to the tuning of the final properties of the composite material. The rGO is an important carbonaceous functional materials that has attracted considerable attention owing to its high aspect ratio, high mechanical strength, unique electrical properties, and chemical stability.
Abalonyx is a partner of a Horizon 2020 project entitled “CARMOF” in which aims to combine the advantage of physical and chemical adsorbents in a hybrid structures composed by functionalized MOFs, and rGO for CO2 capture[1].
In this work, we present some parts of the results obtained for synthesis of rGO through thermal heat treatment. The thermal heat treatment in this work has been performed with two different strategies: in the first one, the GO samples were heated at different temperature for different soaking times. In the second approach, as reduced GO (rGO) was annealed at 800 °C and 1000 °C for 1hr under the vacuum. Indeed, the second approach combines an initial flash pyrolysis of GO at lower temperatures and a subsequent ramp-heating treatment (annealing) up to the selected final temperature (800 °C and 1000 °C). The results of characterizations obtained for annealed rGO and heat treated GO, have been compared and discussed.

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Figures

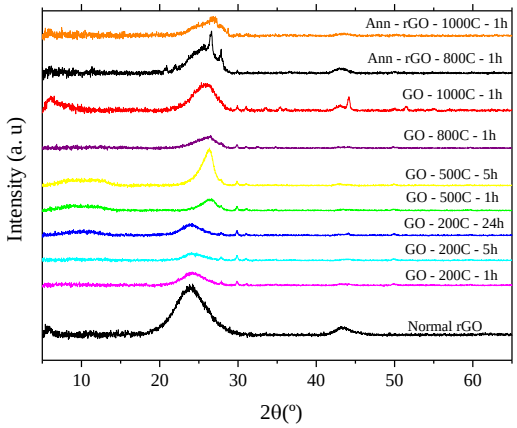

Figure 1: XRD patterns for GO and rGO powder treated at different temperatures for different soaking times

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Functionalization of Graphene Oxide for Industrial Applications

Graphene and its functional derivatives (GO and rGO) have attracted attention in the scientific community owing to their remarkable chemical and physical properties. [1] Importantly, the excellent electrical and thermal conductivity of these compounds has opened new possibilities in scientific fields such as electrochemistry, optics, capacitors and biological sensing. Particularly, graphene oxide, an oxidized graphite sheet containing oxygen-type functionalities, both on the basal plane and on the edge, offers a unique opportunity to modulate the chemical and physical properties by modification and engineering of its surface. [2],[3]. In this work graphene oxide was prepared using Hummer's methodology. [4] Subsequently, it was chemically modified using appropriate organic or inorganic molecules. In one example; GO is functionalized using an amino based polymer such as polyethyleneimine (PEI), covalently forming an amide with the carboxylic acid residues on the GO (Figure 1). [5] The reduction of GO was also explored using an economical and environmentally friendly electron source: ascorbic acid. These approaches (reduction and surface modification) represent excellent opportunities for the fine tuning of the chemical and physical characteristics of GO and rGO to confer the desired properties into the material. Finally, different methodologies are employed to characterize and define these modified materials (GO/rGO) in order to allow a systematic study of the morphological, structural and functional state of the new materials. Various techniques were employed for this purpose, such as FT-IR, SEM, XPS, XRD; NMR solid state, FT-Raman and TGA.

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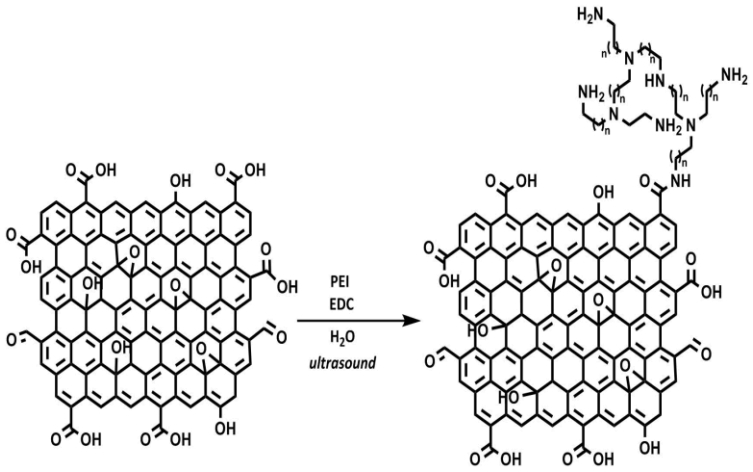

Figure 1: General scheme for GO functionalization with polyethyleneimine (PEI).

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Momentum Conservation Driven Ultrafast Charge Transfer Dynamics of Interlayer Excitons in vdW Heterostructure

Heterostructures comprising vdW stacked transition metal dichalcogenide (TMDC) monolayers are a fascinating class of two dimensional (2D) materials with a host of unique properties [1-3]. Presence of interlayer exciton is an intriguing feature of these heterostructures. In an interlayer exciton, the electron and the hole remain spatially separated in the two layers due to the ultrafast charge transfer, which compete with other relaxation channels [4]. Inevitably, the effciency of devices with 2D heterostructures is critically dependent on the charge transfer dynamics. Investigating MoS_2/WSe_2 heterostructures with monochromated low-loss electron energy loss spectroscopy (EELS) combined with aberration-corrected scanning transmission electron microscopy (STEM), we report for the first time that momentum conservation is a critical factor in the charge transfer dynamics of TMDC heterostructures. From the low-loss electron energy loss (EEL) spectra of the heterostructures with various rotation angles, we demonstrate that- in the aligned case, the charge transfer rate can be about three times faster than the anti-aligned case. Our results provide a deeper insight into the role of the fundamental principle of momentum conservation in the 2D TMDC heterostructure charge transfer dynamics, which could have substantial implications in the rational design of effcient devices based on interlayer excitons. This work also demonstrates the strength of combined STEM-EELS with aberration-correctors and monochromators for investigating optical properties at high spatial resolution discriminating local nanoscale inhomogeneities far beyond the resolution limits of conventional optical techniques.

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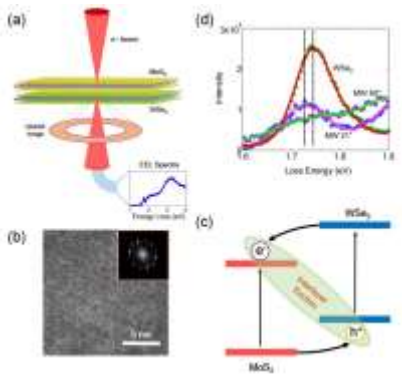

Figure 1: (a) Schematic of the STEM-EELS experimental set-up. (b) HAADF STEM image of the Moiré superlattice due to 29˚ relative rotation angle between the MoS_2 and WSe_2 monolayers (inset show the FFT pattern). (c) Schematic representation of interlayer exciton formation after charge transfer to the other layer. (d) Comparison of representative low-loss EEL spectra for monolayer WSe_2, aligned MoS_2/WSe_2 heterostructure (60˚), and mis-aligned MoS_2/WSe_2 heterostructure (21˚).

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Direct Electron Microscopic Analysis of Individual Reaction Events in Single-Walled Carbon Nanotubes

Observation of the motions and reactions of molecules, being quantum mechanical entities, has long been an impossible dream. Through advances in transmission electron microscopy (TEM) and methods to anchor molecules on a single-walled carbon nanotube (CNT), we can visually study the structural changes of molecules in situ using single-molecule atomic-resolution real-time TEM (SMART-EM) imaging [1,2]. To go one step further to study reaction kinetics, we need statistical information over time and temperature—information that is so far inaccessible by many of the single-molecule analytical methods. Here we chose to study [2 + 2] electrocyclic conversion of a van der Waals complex of C_{60} in a CNT to an [2+2] dimer (step 1, Fig 1) [3]—a well-known reaction without mechanistic information [4]. Through counting reaction events one by one, we provided the direct proof that the ensemble behavior of the random events conforms to the Rice-Ramsperger-Kassel-Marcus theory. One observed at 393–493 K (step 1_H) involves a singlet excited state [2 + 2] cycloaddition reaction with an activation energy of 33.5 $\pm$ 6.8 kJ/mol. Here the CNT acts as a sensitizer of the cycloaddition. Another pathway found at 103–203 K (step 1_L) to occur after CNT was heavily damaged by the electron beam. This reaction takes place with an activation energy of only 1.9 $\pm$ 0.7 kJ/mol. Here an ionized form of the damaged CNT is considered to act as an oxidant to generate a reactive radical cation of C_{60}.

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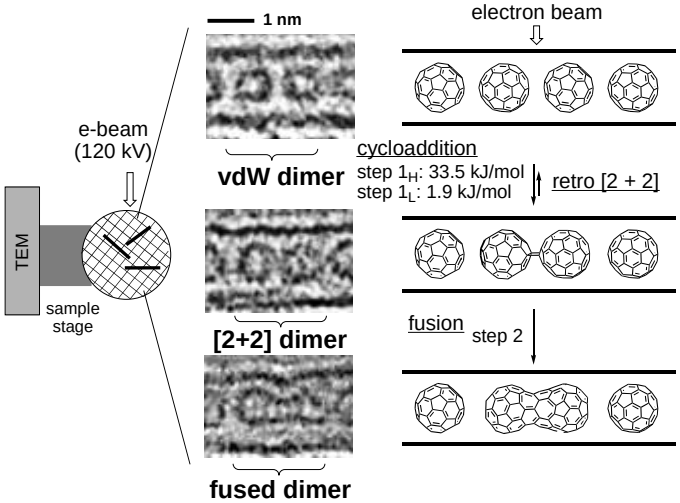

Figure 1: A schematic illustration of the SMART-EM imaging of the dimerization of C_{60}@CNT.. (b) TEM images of intermediates in a 1.4-nm CNT. Four types of reactions observed using TEM.

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MOF derived interconnected structured porous carbon for high-performance supercapacitor

The accessible surface area, conductivity, and pore size of carbon materials determine the capacitive performance. Although metal-organic frameworks (MOF) derived porous carbon has been widely used as electrode materials for supercapacitor, the limited ion diffusion/transportation as well as relatively low conductivity have hampered its high capacitance achieved.[1] To address this issue, an interconnected hierarchical nanoporous carbon structure (HNPC) derived from nanosized MOF crystals is proposed, which presents high ion-accessible surface area and ion diffusion/transportation rate as well as enhanced electric conductivity. Benefiting from the interconnected structure, the resulting HNPC exhibits an ultrahigh capacitance in both aqueous electrolyte and organic electrolyte. More importantly, the high energy density, excellent long-term cycle stability are achieved. This simple and cost-effective process can be readily scaled up to produce HNPC materials commercially.

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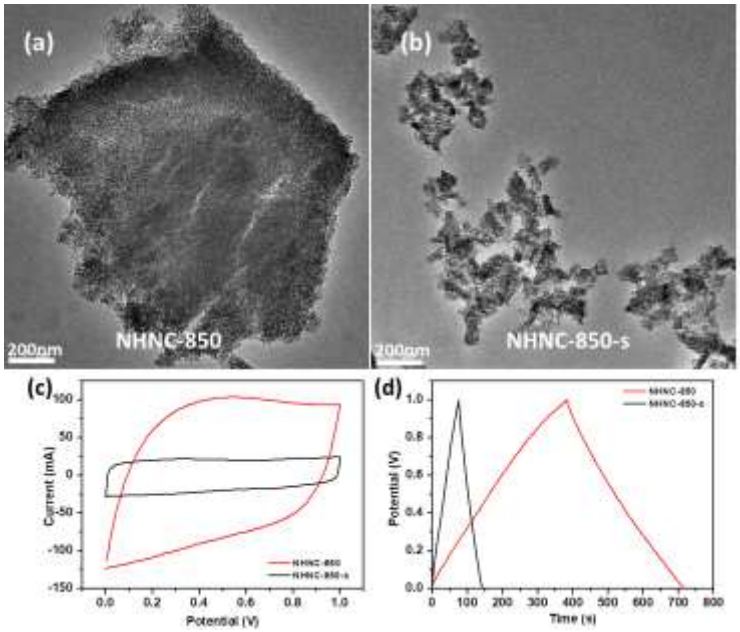

Figure 1: TEM images of (a) HNPC with interconnected structure (b) HNPC with separated structure. Electrochemical performance of HNPC with interconnected and separated structure of (c) CV cures (d) Charge/discharge profiles.

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Transient SHG microscopy on atomically thin 2D materials

Abstract

A pump-probe-based time-resolved spectroscopy has been extensively adopted to study ultrafast carrier dynamics of nanosized materials requiring sub 10 ps level time resolution. More recent research in this area has focused on two-dimensional (2D) materials. Potential applications include energy storage and the enhancement of optical susceptibility and carrier transfer efficiency. Recently, the usage of time-dependent transient second-harmonic generation (TSHG) spectroscopy was suggested for the observation of carrier dynamics and acoustic phonons with diffraction-limited spatial resolution in MoS_2 crystalline structures [1]. Compared with this work, our technique involves a simple measurement system that can deliver high pulse power density at low beam energies, whereas the use of a raster scanning system enabled the pulse exposure time to be reduced to approximately 10 μs per pixel, leading to reduced sample damage [2,3].

Specifically, we report the ultrafast carrier dynamics in atomically thin molybdenum sulphide (MoS_2) crystals, triggered by creating an A-excitonic resonance condition with pump beam. The power dependence suggests the highly dominant nature of χ_3 term in TSHG signals while the polarization dependence resolves a crossed contribution of χ_2. The thickness dependent monitoring on the target sample critically allowed us to claim the early evolution of acoustic phonons in the thicker layers. Also, interestingly enough, the dynamics of different chiral edges were resolved through spectral analysis and were supported by first principal calculation and TEM.

Theory predicts there is an additional gap in the density of states near the edge of the work function of the atomically thin ReS_2 system, which has not yet been fully identified in an experimental fashion. In this paper, we suggest one way to probe the existence of it by employing the time-resolved SHG microspectroscopy where separately-controllable high energy pump and probe pulses were accessible [4]. We also interpret the origin the ultrafast time dependent carrier dynamic behaviors of ground-state-depletion and excited-state-absorption from a viewpoint of the allowance of each electronic transition by adopting the angular momentum selection rule.

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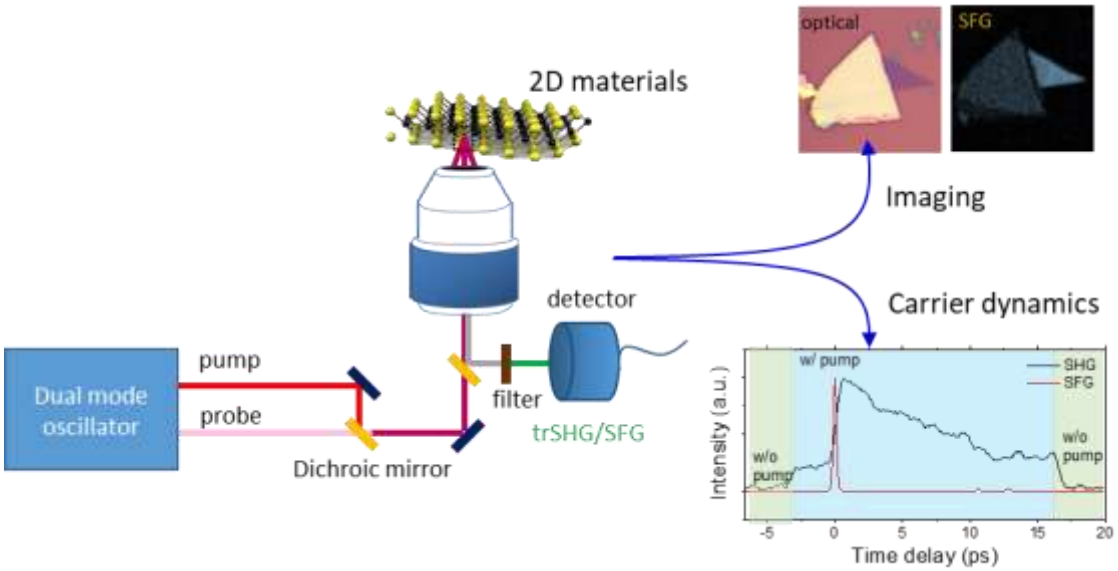

Figure 1: Scheme of time-resolved SHG microscopy on ultrafast carrier dynamics of 2D materials

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Thermal interface materials made of vertically-aligned multi-walled carbon nanotubes

Carbon nanotubes (CNTs) are promising materials for next generation of electronics by virtue of their supreme physical properties such as tolerance for high current density and thermal conductivity on the same level with diamond [1, 2]. Actually, applications of CNTs to interconnects and thermal interface materials have been studied [3-4]. Unfortunately, there are many issues caused by difficulties in connecting CNTs to other materials like metal electrodes, where small electrical/thermal contact resistance is necessary. Here, we focus on the application of vertically-aligned CNTs to thermal interface materials (TIMs).
Vertically-aligned multi-walled CNTs were grown on a silicon substrate with a thermal silicon dioxide layer by chemical vapor deposition (CVD), as shown in figure 1(a). After peeling CNTs off the substrate, the CNTs were annealed at temperatures above 2000°C and pressed to form a CNT sheet, as shown in figure1b). The thermal property of CNT sheets was measured by a temperature gradient method. Figure 1(c) shows the dependence of the thermal resistance of CNT sheets (annealing temperature: 2400°C) including the interface resistance on the pressure applied at the time of the measurement. It was found that the thermal conductivity of CNT sheets was estimated to be approximately 80 W/mK. The results of Raman spectroscopy showed that the annealed CNTs are of high quality, supporting this high thermal conductivity of CNT-TIMs. Our results indicate that CNT sheets annealed above 2000 °C are really promising for future TIM applications.

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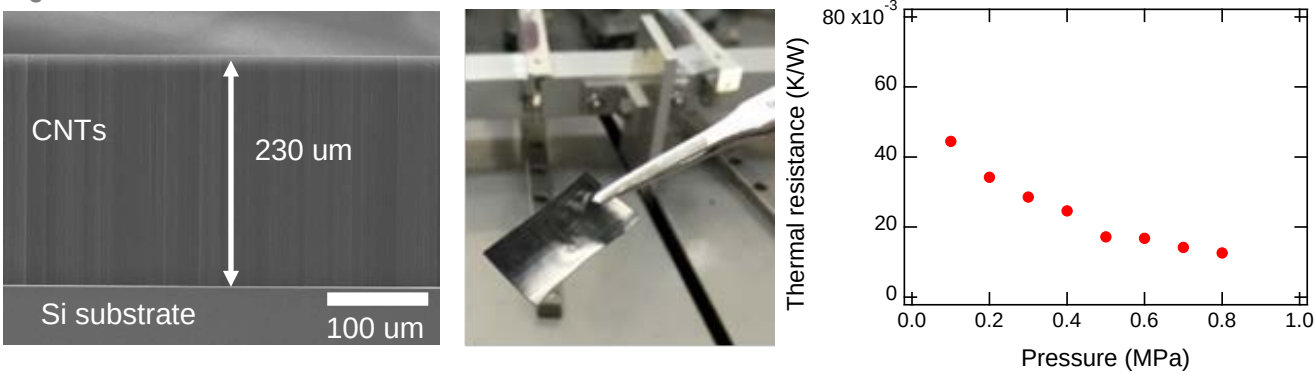

Figure 1(a): SEM image of CNT sheets after annealing at 2400 °C. (b): Photo image of CNT sheets. (c): Thermal conductivity of CNT sheets dependent on pressure.

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Creation and manipulation of valley magnetic domain toward valleytronic current processing

First-principles density functional theory provides significant insights into an interplay between an applied strain and the Berry curvature reconstruction in the uniaxially strained monolayer MoS_2, which leads to the unbalanced Berry curvatures centered at **K** and **–K** points and eventually the homogeneous valley ferromagnetism, i.e., valley magnetic domain (VMD) under an external static electric field. This is the valley Edelstein effect (VEE) to explain a recent experimental observation of the strain-induced valley magnetoelectricity [1]. Here we demonstrate (i) the migration of VMD (i.e., domain wall moving) and (ii) the inversion of VMD (i.e., domain switching) in terms of controlling a strain streangth and an external electric field direction, respectively. We further extensively investigate the electric current responses depending on those various manipulations of VMD and suggest relevant potential applications. It is proposed that the VMD manipulation should be a key ingredient of valleytronic realization and application.

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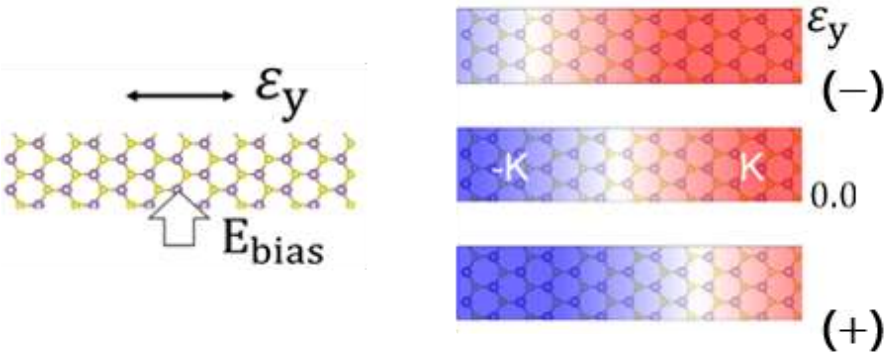

Figure 1: Strain-induced valley magnetic domain in monolayer MoS_2 under an external electric field (E_{bias})

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Facile Catalyst Deposition Using Mist for Fluidized-Bed Production of Sub-Millimeter-Long Carbon Nanotubes

Abstract
Fluidized-bed chemical vapor deposition (FBCVD) has enabled mass-production of carbon nanotubes (CNTs) [1]. By using spherical ceramic beads as catalyst support and depositing Fe/Al_2O_3 catalyst on them, we have realized semi-continuous production of sub-millimeter-long CNTs [2,3]. To improve the controllability over catalysts, a new method of catalyst deposition on ZrO_2 beads in fluidized bed by feeding water or organic catalyst mist is reported. In the water system, low-cost $Fe(NO_3)_3$ and $Al(NO_3)_3$ aqueous solutions mist is supplied and deposited on the beads. CNTs of 5-10 nm in diameter, triple-wall on average, 0.6 mm in length are synthesized with a yield of 16.6 mg_{CNTs}/g_{Beads} (Figures. 1 and 2). These results benefit from the uniform catalysts realized by mist-deposition and proper amount of Al, which is considered to play an important role in controlling the diffusion and aggregation of Fe atoms (Figure. 2). Besides, quick deposition of catalysts are realized by feeding aluminium isopropoxide and ferrocene ethanol solution mist. CNTs with 0.5 mm in length are synthesized with a yield of 17.0 mg_{CNTs}/g_{Beads}. These results indicate that mist deposition to be a facile and effective method toward high-yield produciton of CNTs.

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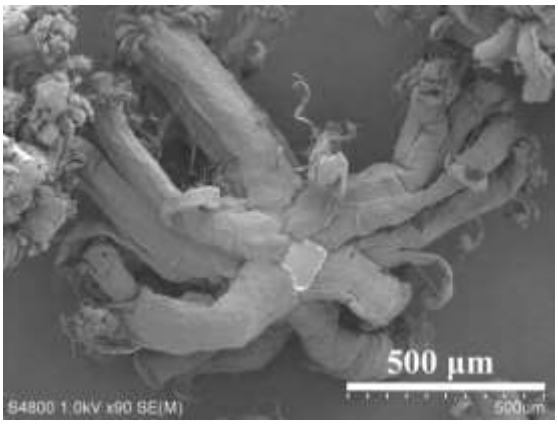

Figure 1: SEM image of CNTs on the beads.

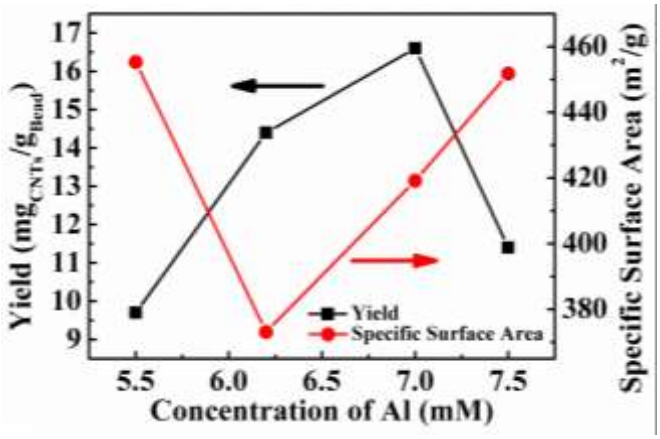

Figure 2: Effect of Al source concentration on the yield and specific surface area of CNTs.

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Multiscale modeling on the mechanical properties of graphene based two dimensional nanostructures

Abstract: With the rise of nanotechnology and the advances in interdisciplinary research, two dimensional (2D) nanomaterials, such as graphene, have received intense global interest due to their unique physical and chemical properties over traditional materials. The mechanical properties of graphene based nanomaterials are characterized using density functional theory, molecular dynamic simulation as well as theoretical predictions. Atomistic molecular dynamics (MD) models will be constructed for the in-plane mechanical properties of surface functionalized and planar hybridized graphene. The effect of grain boundary on mechanical properties of polycrystalline 2D materials will also be addressed in this project by the stress contour analysis using MD simulations and modified dislocation theory. Surface functionalization and in-plane hybridization are investigated for the manipulation of mechanical as well as thermal properties of 2D graphene structures. Investigations about the principle and mechanism for the mechanical properties and behavior of graphene based nanomaterials are critical to its functional design and application in flexible devices and next generation electronics.

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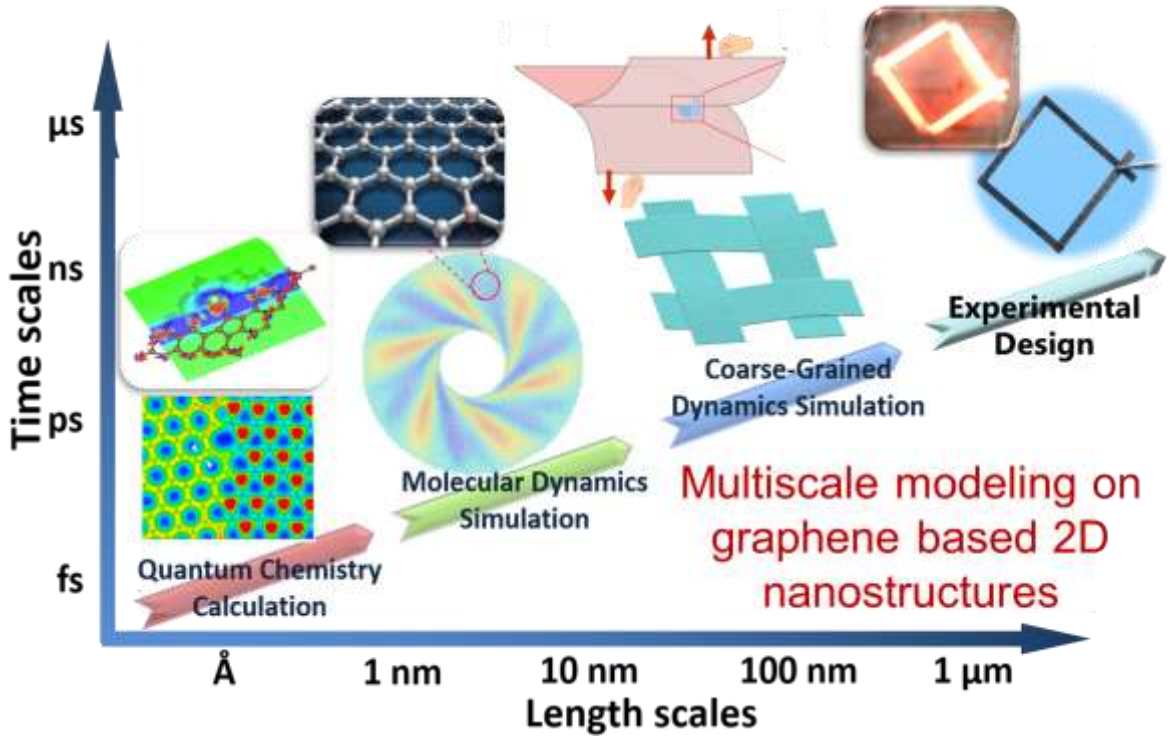

Figure 1: Multiscale modeling on the mechanical properties (tensile strength and torque capacity) of graphene based two dimensional nanostructures (graphene, planar CBN heterostructure, woven nanostructures and graphene welded structures)

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Topological dipoles and quadrupoles

Topology offers us a unique dimension of designing solid-state materials. One famous example is the Quantum spin Hall effect (QSHE) where electrons of opposite spins propagate oppositely. The origin of QSHE comes from a geometric field strength in momentum space that is the so-called Berry curvature. Besides QSHE, the geometric vector potential whose curl yields the Berry curvature – the Berry connection, can induce an electric dipole even under zero Berry curvature. The topological electric dipoles result fractional surface charges that manifest as topological edge states, which are robust to defects and edge roughness. Furthermore, a pair of such the topological dipoles can form a high order multipole – quadrupole, which corresponds to the topological corner states.

In this talk we will discuss a simple tight-binding model that possesses topological dipoles and quadrupoles in zero Berry curvature [1]. Experimental realizations based on solid-state material and dielectric photonic crystal are proposed [2]. Furthermore, we show that in a pure quadrupole phase, topological edge state is pseudo-spin polarized in general [3].

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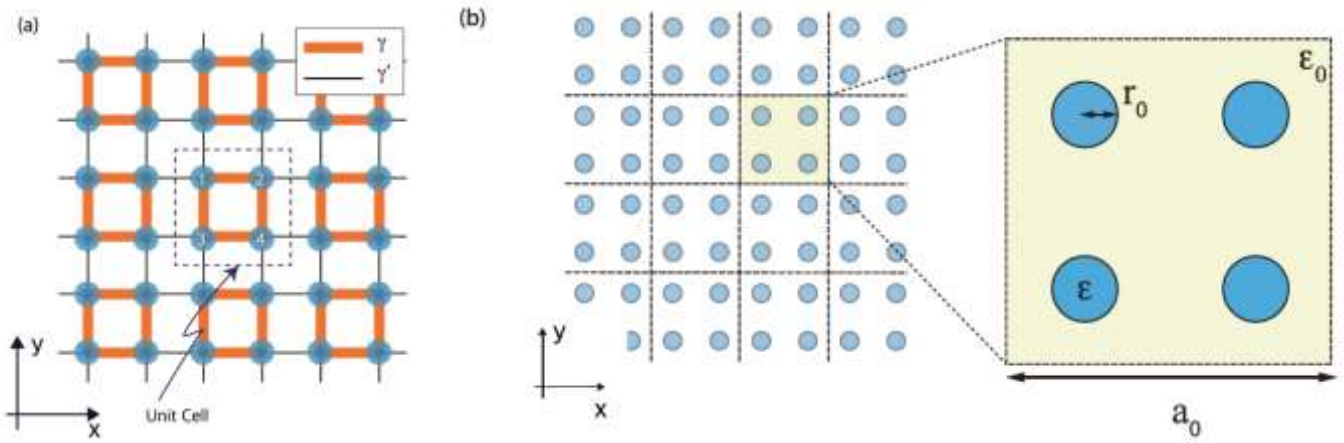

Figure 1: (a) Schematic lattice structure of the 2D SSH model. The model is specified by the intracellular hopping γ and the intercellular hopping γ'. (b) Schematic of the dielectric crystal mimicking the 2D SSH model. The unit cell consists of four identical dielectric cylinders of radius r_0 placed at (±0.25, ± 0.25)r_0. The lattice constant is a_0.

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Carbon based nanostructures - thermal , electrochemical, adsorption, photocatalytical response.

In our research we aim at designing, synthesis and physicochemical properties of innovative carbon based nanostructures. The Department focus on the studies of challenging nanocomposites flame retardant and designing electrodes and adsorbents with emphasis on correlation between nanomaterials structure with highest capacity and efficient gas adsorption. Conducted research engage all efforts to broaden understanding of nanomaterials synthesis and properties along with the improved nanomaterials development process. The work is mainly focused on:
- improving the development of different polymer nanocomposites with high flame retardant, thermal conductivity and thermal stability by innovative admixtures like 2D molybdenum disulphide nanostructures functionalized with carbon nanotubes and metal oxides (e.g. Ni_2O_3 and Fe_2O_3) [1,],
- synthesis novel highly porous carbon nanostructures by carbonization of metal organic frameworks and their functionalization for applications such as gases and waste adsorbents, and for supercapacitors and Li-ion battery electrodes [2,3],
- design of different novel catalysts for photocatalysis in UV-Vis region.enhancing the highly porous carbon nanosphere electorchemial and gases adsorption properties by functionalization them with different inorganic nanomaterials e.g. metal oxides (iron oxide), metal (platinum/palladium) nanoparticles and graphene structures [4],
- nanomaterials waste-free synthesis for environmental friendly manufacturing [5].

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Developement of nitrogen doped graphene derivatives for renewable nano-structured membranes for direct alkaline ethanol fuel cell

Fuel cells are electrochemical devices that convert a fuel's chemical energy directly to electrical energy, and have been identified as one of the most promising technologies for the clean energy industry. Abalonyx is a partner of an ERA-NET project entitled “NanoElMem” [1]. The overall concept of the NanoElMem project relates to developing novel stable and highly effective materials for the direct alkaline ethanol fuel cell (DAEFC), which directly converts ethanol to electric power. The enhancement of the performance of DAEFCs is based on the development of platinum (Pt)-free electrode catalysts and nano-composite membranes by using environmental-friendly inorganic and polysaccharide-based materials and technologies. The enormous technical and scientific potential of graphene was explored by producing novel graphene-polysaccharide based membranes. The emphasize is on Abalonyx activities in producing nitrogen doped- graphene oxide (NGO) and reduced graphene oxide (NrGO) (Figure 1). To make the membaranes, the GOs and NGO/NrGOs were dispersed in 1 wt% chitosan solution (Figure 2). Eventually, the fuel cells equipped with these membranes were assembled and the cell performance was evaluated.

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Figures

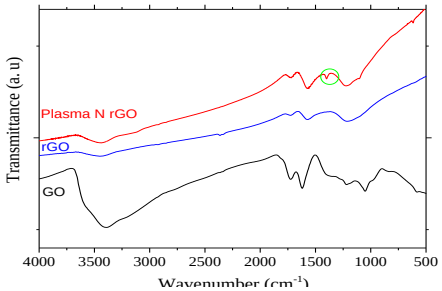

Figure 1: FTIR spectra of GO, rGO and plasma NrGO.

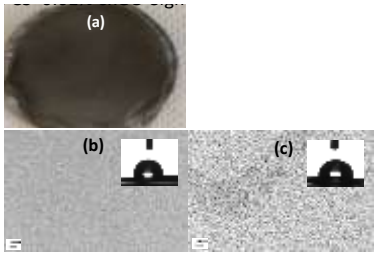

Figure 2: Optical and SEM images of pristine 1 wt% of chitosan membrane (b) and chitosan membrane in the presence of GO (a, c).

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Valley-Mixing-Induced Splitting in Conductance Resonances through a Single Magnetic Quantum Dot in Graphene

Charge and spin transport through quantum dots is one of major research interests in mesoscopic and condensed matter physics. In particular, graphene has received the attention as a promising playground for both experimental and theoretical investigations due to its extraordinary transport properties and stability[1,2]. Also, the presence of an additional degree of freedom in graphene, so-called `valley, leads to another aspect of graphene in terms of research: valley transport and its device applications. In this study, we have shown that Dirac fermions are strongly localized in a magnetic quantum dot (MQD), which is created by screening out an external magnetic field with the circular geometry, and conductance through the MQD exhibits resonances when Dirac fermion energies meets eigenenergies of the MQD. Interestingly, the conductance resonances are revealed to be split into two distinct resonant peaks: symmetric Breit-Wigner and asymmetric Fano resonances. We have demonstrated that such a dual resonant feature is because of the two-level splitting as a consequence of valley mixing in the MQD, in a finite-size system.

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Figures

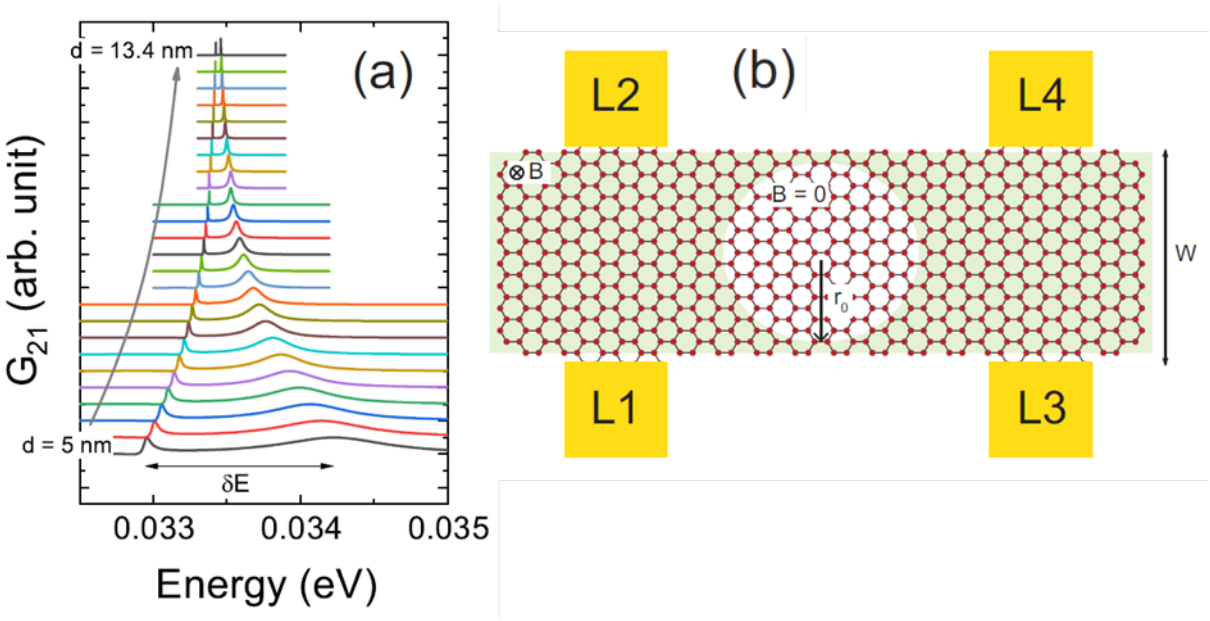

Figure 1: (a) Conductance spectra through the MQD for various MQD-edge distances. Each curves are calculated from the S-matrix approach between L1 and L2 leads. (b) Schematic model of the system. Uniform magnetic fields are applied to the graphene Hall bar with four leads, and screened out in the circular region, resulting in the MQD.

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Calculation of the Electromechanical Properties of CNTs Under Deformation by Means of a Novel Numerical Model

The effects of mechanical deformation on the electron transport behavior of carbon nanotubes (CNTs) are of primary interest due to the enormous potential of nanotubes in making electronic devices and nanoelectromechanical systems (NEMS). Moreover it could help to evaluate the presence of defects or to assess the type of CNTs that were produced. Conventional atomistic simulations have a high computational expense that limits the size of the CNTs that can be studied with this technique. Here we present a novel numerical approach able to simulate the electromechanical behavior of SWNTs and MWNTs of the dimensions used in nano-electronic devices. The numerical model was designed to realize orders-of-magnitude savings in terms of computational time in the calculation of the mechanical behavior and of the changes induced by the deformation in the electrical transport properties of the nanotubes. The computational method has been validated with respect to fully atomistic simulations and to a simulation of laboratory experiments. In this paper, simulation results are presented for different types of SWCNT and MWCT in a variety of boundary conditions.

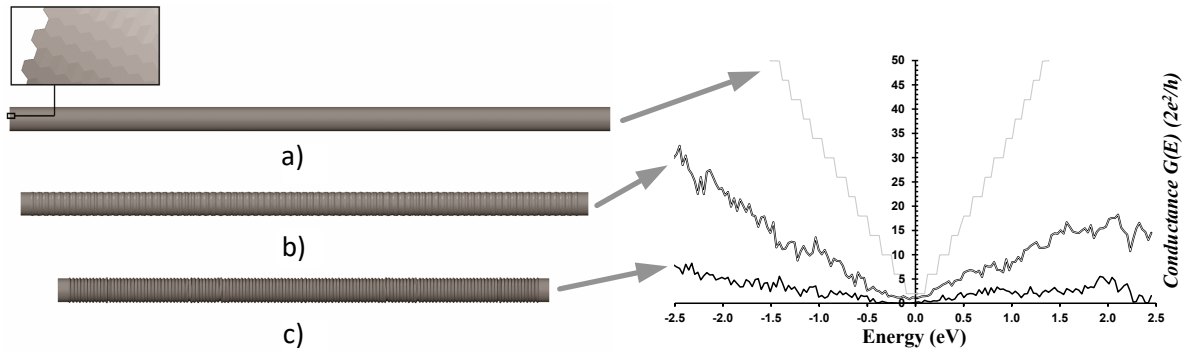

Figure 1: Model reproducing a MWCNT: a) in the undeformed configuration, b) compressed axially by 20 nm, c) compressed axially by 50 nm. The inset shows a magnification of the outermost wall revealing the hexagonal cells.

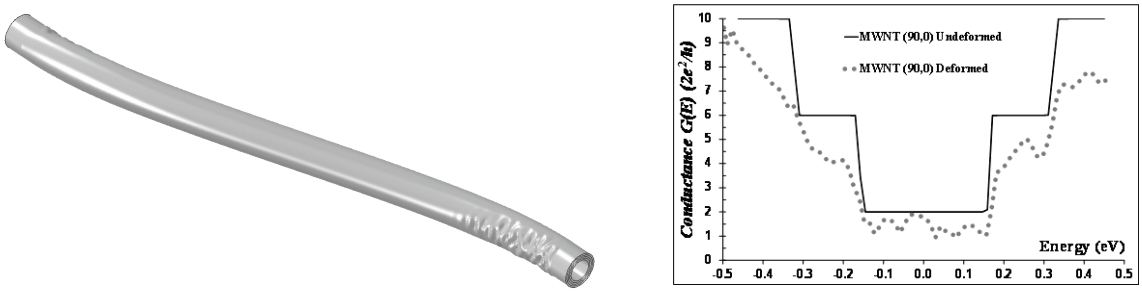

Figure 2: Simulations of a MWCNT where the two ends of the tube have been misaligned by 30 nm.

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Scattering-type scanning near-field optical microscopy as a versatile tool for the characterization of 1D and 2D materials.

Materials with strong confinement in one or two dimensions have shown large potential for new application due to their exotic properties, which are directly related to their nanoscale dimensions. It is often desirable to correlate properties, such as electronic structure and vibrational modes, to the material or nanostructure thickness and the nanoscale morphology.
One method that yields both local topography and can probe different optically excited transitions is scattering-type scanning near-field optical microscopy (s-SNOM). A metal-coated standard atomic force microscopy (AFM) probe is illuminated with light, which creates a nanofocus at the tip apex in which the electrical field of the incident light is amplified. The back-scattered light contains the information of the interaction of the sample with the nanofocus, with a spatial resolution dependent only on the tip apex radius[1]
With s-SNOM direct imaging of 2D material surface polaritons became possible for the first time. The graphene surface plasmon polaritons can be launched in the tip near-field and the interference of the waves directly imaged[1,2]. The discovery of the visualization of the polariton waves opened the field to the examination of all kinds of light–matter–interaction in 2D materials, including phonon polaritons and exciton polaritons[4] and also of phonon plaritons in 1D materials, such as InAs nanowires[5].
While the imaging of polariton waves is the most prominent application example, s-SNOM has recently also been used to characterize the conductivity of both carbon nanotubes[6] and single-layer graphene[7].

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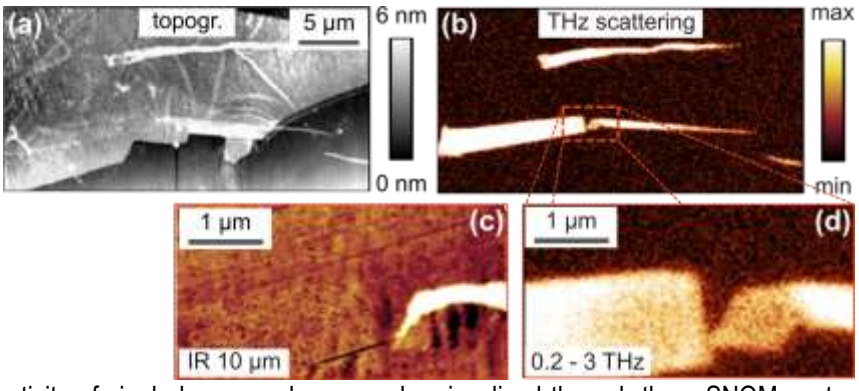

Figure 1: The conductivity of single layer graphene can be visualized through the s-SNOM contrast in the IR and THz spectral range. High reflectivity in the IR shows highly conductive areas, while those areas that are reflective in the THz spectral range are expected to have higher conductivity than the non-reflecting surroundings but lower conductivity than the areas that reflect IR light.

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Vertical Injection in Black Phophorus-Graphene Heterostructures for Terahertz Lasing

The heterostructures based on graphene-layers (GL) with optical and injection pumping can be used as active elements in the sources of terahertz (THz) radiation, including THz lasers [1-3]. One of the most important advantages of such lasers with lateral lateral carrier injection from the side p- and n-contacts in comparison with the lasers using the optical pumping is associated with relatively low energies of the injected carriers.
In this communication, we propose and evaluate the GL-based THz lasers with the lateral injection from the side n-contacts and the vertical hole injection. To prevent a marked heating of the electron-hole system in the GL due to the injection relatively hot holes, we propose to use a p^+-p- injector with the black phosphorus layers (PLs) [4]. The energy spacing between the GL Dirac point and the top of the PL valence band is about 0.1 eV. The latter value is smaller than the optical phonon energy in the GLs (about 0.2 eV). This can result not to the heating of the electron-hole system in the GL, but to a substantial cooling of the latter, promoting a strong interband population inversion and, hence, forming the conditions for the effective THz lasing.
Using the developed device model, we calculate the spectral dependences of the dynamic conductivity as functions of the structural parameters and the injection current and the gain of the plasmon-polariton modes propagating along the GL. We demonstrate that the heterostructures in question can surpass the GL-based heterostructures with the lateral double injection.

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Universal Characters of Superconductivity in the 2-D (Quasi-2-D) Systems of Strongly Correlated Electrons based on the SU(2) Gauge Theory of Slave-boson Approach to t-J Hamiltonian

The two-dimensional systems of strongly correlated electrons have been of great interest as various physical problems are still unresolved. As an example, one of the major theoretical challenges in 2-D (or quasi 2-D) high-Tc superconductivity is to reproduce the observed phase diagrams which display the monotonously decreasing pseudogap temperature and the dome shaped superconducting phase transition temperature. in the plane of temperature vs. hole concentration and, in addition, to consistently fit other observed physical properties of the cuprate superconductors. Earlier Lee and Salk [1,2] reported a successful reproduction of the phase diagrams by proposing a realistic gauge theoretic slave-boson approach to the Heisenberg term in the t-J Hamiltonian. Based on the same theory we have been able to consistently reproduce physical properties associated with both spin dynamics and charge dynamics, namely the spin susceptibility [3] and the optical conductivity [4] respectively. Most recently, Salk [5] presented comprehensive descriptions on the well predicted universal scaling behaviours of high temperature superconductivity and suggested a possibility of room temperature superconductivity. We first make a brief review of these works. Admitting the fact that electron has two apparent "faces", the spin and the charge degrees of freedom we pay attention to interplay between charge and spin dynamic to explain physics involved with the observed phased diagrams, optical conductivity and spin susceptibility among others. In connection, derived universal characteristics [5] of physical properties are further appraised in connection with the interplay of the two degrees of freedom involving the spin susceptibility and the optical conductivity. In extension, such universalities in association with the physically well predicted propensity of the larger the antiferromagnetic coupling J, the higher the superconducting transition temperature equally lead to the anticipation of feasible room temperature superconductivity.

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Lightweight high-performance Cu/Carbon Nanotube composites

We present high-performance copper-matrix carbon nanotube composites (Cu/CNT) lighter than copper [1-3] as promising copper-substitutes. Compared to copper, our composites are at least 2/3rd as light and show competitive room-temperature electrical resistivities, higher temperature- and current-stabilities, and at par mechanical strengths. Our data suggest that composite performances may depend on microstructure (Cu spatial distribution) and CNT attributes. For instance, our Cu/SWCNT composites show room-temperature electrical resistivities (ρ_{RT}) as low as 3.3 × 10^{-6} Ohm cm (~ × $2\rho_{RT}$Cu) and temperature coefficient of resistivity (TCR) as low as 4.4 x 10^{-4} /K (~10% Cu_{TCR}). In contrast, our Cu/MWCNT show higher ρ_{RT} (× 10 ρ_{RT}Cu) and TCR (50% Cu_{TCR}). Besides ρ_{RT} and TCR, the current carrying capacity of our composites is higher than Cu. In addition to electrical performances, the composite mechanical and thermal expansion properties are also favorable. Our composites show mechanical strengths (~ 300 MPa) similar to annealed and drawn commercial Cu wires. The composite's coefficient of thermal expansion (CTE) is ~ 4-7 ppm/K, which is more similar to Si (~3 ppm/K) and lower than Cu (~17 ppm/K).

We have fabricated these Cu/CNT composites as microscale pillars and macroscopic wires, which are additions to planar Cu/CNT microlines and sheets reported by our group previously [4-6]. We believe our array of Cu/CNT composites show immense potential to fulfill a growing demand for lightweight electrically conducting Cu-substitutes. The macroscopic Cu/CNT wires could replace heavy copper electrical wiring in aircrafts and automobiles to achieve better fuel efficiencies and reduced CO_2 emissions. Our composite's temperature-stable resistivity (TCR < Cu) specifically makes it a reliable conductor for high-temperature operation e.g., in motor windings. Meanwhile, our microscale Cu/CNT structures with CTE ~ Si could serve as better interconnects than Cu in high-power electronics, facilitating the development of smaller and more powerful next-generation devices.

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Nanoscale optical and vibrational spectroscopy of low-dimensional materials

Physical properties of low-dimensional materials are strongly influenced by the quasiparticle excitations at non-perfect structures like boundaries, edges, defects or irregular stacking sequence. Unfortunately, such local information has been usually averaged in conventional inelastic scattering techniques using x-ray, neutron and light sources because of their inferior spatial resolution. Here we demonstrate the nanoscale optical and vibrational spectroscopy of 1D/2D materials by using a monochromatic electron source mounted in a scanning transmission electron microscope. Its energy resolution, better than 30 meV, allows to access the quasiparticle excitations (i.e. phonon, exciton and plasmon) of low-dimensional materials by electron energy-loss spectroscopy (EELS). The spatial and momentum resolutions are dependent on each other and can be tuned freely. For instance, by integrating a wide momentum space, an atomic sized probe can be formed and the local spectroscopy on a single defect is possible. Indeed, we have successfully measured the optical gap transitions from a defect of an individual semiconducting carbon nanotube [1,2]. The optical conductivity extracted from an EEL spectrum via Kramers-Kronig relation for a certain type of defect presents a characteristic modification near the first exciton peak (Fig. 1). The line-width of exciton peak shows a variety of broadening at different defect sites and suggests different degrees of shortening of its lifetime. In contrast, an electron probe with a higher momentum resolution can provide a full phonon dispersion of 2D materials such as hexagonal boron nitride or graphene at a few tens nanometre scale. This local spectroscopy with a large flexibility will open up a wide possibility to unravel the defect physics of quantum matters.

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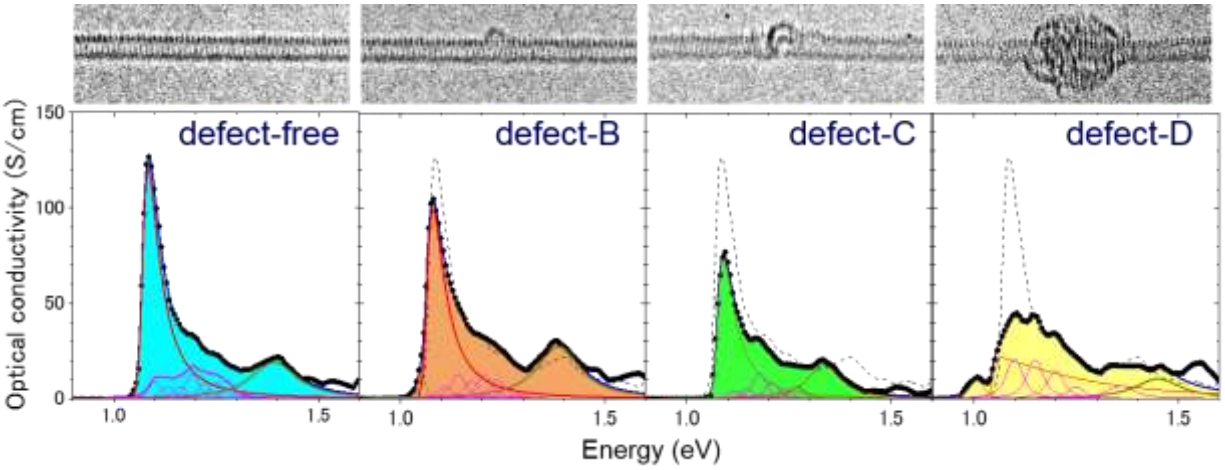

Figure 1: Absolute optical conductivity of a semiconducting (9,2) at three different defects as well as a defect-free region. This shows how the gap transition and exciton lifetime depend on the type of local defect [2].

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Ab-initio modeling of metal interaction with 2D materials

Employing density functional theory we studied microscopic mechanisms governing binding of metal atoms and their nanostructures at selected 2D materials. First, we considered Li interaction with monolayers of several transition metal oxides and dichalcogenides, carbides of group XIV elements, functionalized graphene, silicene and germanene, as well as black phosphorus and Ti_2C MXene. We found that the general trend in Li binding can be estimated from positions of conduction band minima of 2D materials, since the energy of the lowest empty electronic states shows a nice correlation with the strength of Li adsorption [1]. At variance to the majority of studied surfaces where occurs a simple electron transfer from Li to the substrate, in monolayers of carbides of group XIV elements Li adsorbates profoundly modify substrates, creating well-localized mid-gap states. The second part of presented work describes growth mode of Li, Ti and Ca on graphene [2] and structural and electronic properties of graphene/MoS_2 heterostructure intercalated with Au [3]. The tendency towards the planar or three-dimensional growth of metals on graphene is rationalized based on the atomic-scale description of the interaction between metal adatoms, as well as adsorption geometries of their smallest nanostructures on graphene [2]. Finally, we show that graphene/MoS_2 represents a suitable template to produce a quasi-free-standing 2D Au monolayer with the electrons in the vicinity of Fermi level closely resembling features of 2D free electron gas [3].

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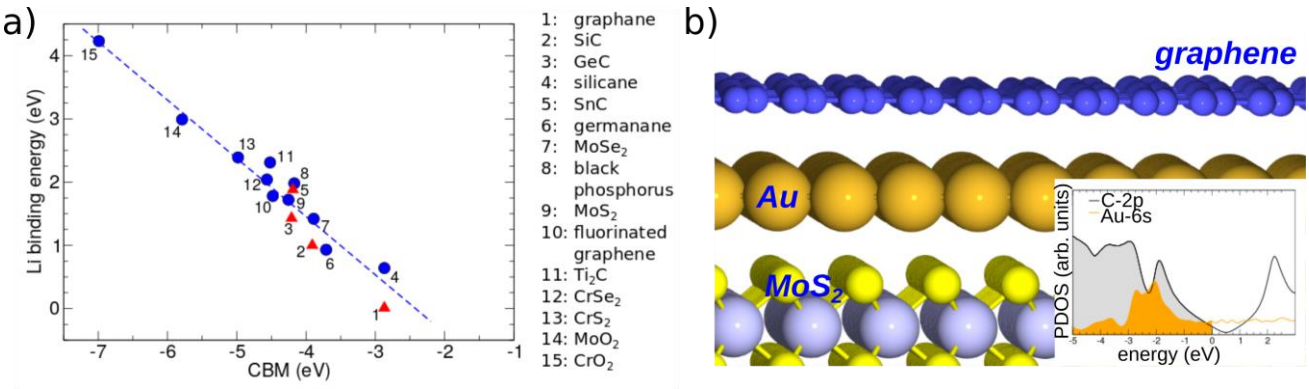

Figure 1: a) Correlation between Li binding energy and conduction band minimum (CBM) of studied 2D materials; b) atomic structure and (inset) density of electronic states of graphene/MoS_2 intercalated with Au monolayer.

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An interdigitated electrode with dense carbon nanotube forests on conductive supports for electrochemical biosensors

An interdigitated electrode (IDE) has an advantage for high sensitivity due to the amplified current by the redox cycling of analytes. As a material for the electrodes, carbon nanotubes (CNTs) have several advantages such as high electrical conductivity and fast electron transfer kinetics. Direct growth of CNTs on substrates by chemical vapor deposition (CVD) is especially a suitable way to integrate the CNTs into the IDE. We have developed low temperature growth technique (450 °C) of ultra-high mass density CNT forests (1.6 g cm^{-3}) on conductive supports.[1,2] They are attractive for the electrode material in IDE as the CNTs and supports have an ohmic contact which is different from the conventional CNT forests on insulators (e.g. SiO_2 or Al_2O_3).

In this report, we applied the dense CNT forests to the IDE by combining the UV lithography and the CVD process (Fig. 1a and 1b). By optimizing the geometry of the electrodes (width and gap), the performance of the IDE was significantly improved. The cyclic voltammetry (CV) measurements of $K_4[Fe(CN)_6]$ showed that the current of IDE with CNTs (CNTF-IDE) reached to the steady-state current much more rapidly compared to that of conventional gold IDE (Au-IDE) (Fig. 1c and 1d). As a model case of the biomolecules, dopamine (DA) was measured under coexistence of ascorbic acid (100 μM). The selective detection of DA was achieved with the linear range of 100 nM – 100 μM, the sensitivity of 14.3 mA mol^{-1} L, and the limit of detection (LOD, S/N=3) of 42 nM. In addition, the CNTF-IDE showed superior anti-fouling property with a negligible shift of half-wave potential ($\Delta E_{1/2}$< 1.4 mV) for 30-times repeated CV measurement of DA at high concentration (100 μM).[3]

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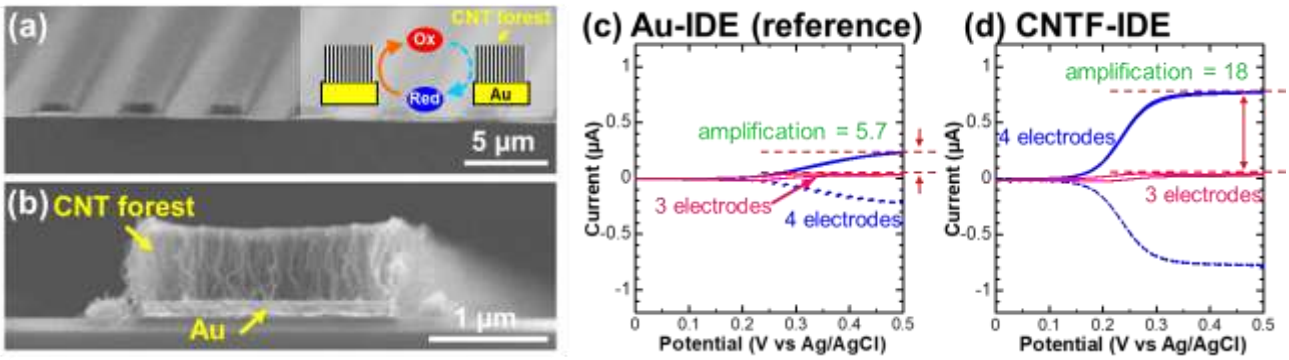

Figure 1: (a, b) Side-view SEM images of the CNT forests on Au electrodes. CV results of (c) Au-IDE (reference) and (d) CNTF-IDE with the $K_4[Fe(CN)_6]$ (100 μM) in KCl (100 mM) at the scan rate of 10 mV s^{-1}.

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Graphene Composite Fibers for Flexible Supercapacitors

Abstract
Flexible energy storage devices are indispensable for flexible and wearable electronics. As the prevailing type, flexible supercapacitors have attracted much attention over the last decade.[1] Among them, fiber-shaped supercapacitors may possess an inherent advantage in flexibility, which can be facilely woven into clothing.[2] To this end, various fibers have been utilized, e.g., metal fibers, chemical fibers, carbon fibers, carbon nanotube fibers and graphene fibers.[2] Here, we would like to introduce some progress on graphene composite fibers for fiber-shaped supercapacitors in our lab. Graphene, a superior material for electrochemical energy storage, however suffers some problems that prevent graphene fibers from high electrochemical properties. The first one is the re-stacking of graphene sheets in the process of fiber formation, leading to a largely deteriorated performance. Therefore, some nanofillers are introduced between graphene sheets to make full use of graphene and/or in the meantime the nanofibers make a contribution to the whole energy output.[3, 4] The second problem is the hydrophobic character of graphene sheets that hinders the wetting of graphene sheets by electrolytes. This can be addressed by adding hydrophilic components.[5] In addition, several facile methods of fabricating graphene composite fibers are illustrated herein.

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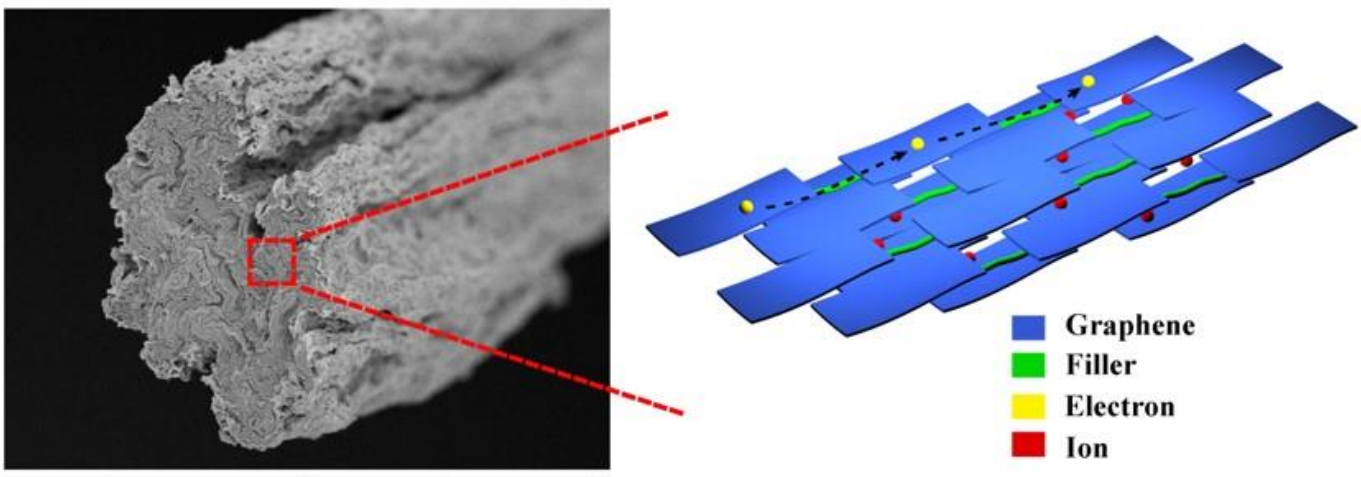

Figure 1: SEM image and scheme of graphene composite fiber.

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Multilayer-Graphene Fillers for Improved Gas Barrier Properties in Polymer Composites

Graphene as a 2D carbon structure has emerged as the strongest lightweight material with excellent charge mobility for many electronic, optoelectronic and energy storage applications [1]. Unlike its other carbon family members such as carbon nanotubes and fullerenes, impermeable platelet structure of graphene can effectively separate gas molecules or completely halt its transportation through polymer matrix when mixed with it. Therefore, an increasing trend of use of layered-graphene structures into polymer composites has been noticed for several applications [2]. In this abstract, a comparison of gas permeability of thermoplastic polyurethane filled with few-layer and multi-layer graphene nanofillers has been presented. Graphene with the different number of stacked layers were mixed with TPU solutions with the help of ultrasonication. Chloroform was used as a common solvent during all preparations. The as-prepared TPU/graphene composites demonstrated a trend of enhanced O_2 gas barrier properties with the increasing number of graphene layers. As such, graphene with 1.3 nm thickness corresponding to ≈ 3 or few-layer graphene displayed 42% reduction in O_2 gas permeability at 5 wt.% concentration. On the other hand, graphene or graphene nanoplatelets with 5.3 nm thickness (≈ 16 layers) produced 54% reduction in O_2 gas permeability at a similar concentration, given that lateral sizes were close to 450 nm in both cases. As shown in Figure 1, graphene with the least number of layers is rolled-up into tube-like structures when mixed with TPU solution. In contrast, graphene with ≥ 10 layers remained equally dispersed throughout the TPU matrix and preserved its platelet geometry as shown in Figure 1. Another multilayered graphene with 13 nm thickness and 740 nm lateral size was also incorporated into TPU/graphene composites for comparison and could not produce any substantial increase in barrier properties. As a result, one can conclude that graphene with more number of layers is highly favorable for gas barrier properties in polymer composites, provided that lateral size is kept constant [3]. In addition to gas barrier properties, the as-prepared TPU composites with 5.3 nm thick graphene fillers demonstrated improvement in mechanical properties. For instance, Young's modulus of TPU film was increased from 6 MPa (for neat) to 15 MPa after 5wt.% graphene loading. However, at more than 5 wt.% graphene concentration the elongation at break was reduced from 600% to 300% [4]. The prepared TPU/graphene composites with enhanced gas barrier and mechanical properties can be used in packaging, pneumatic tubes in industrial process or vehicles and gas container applications.

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Figure

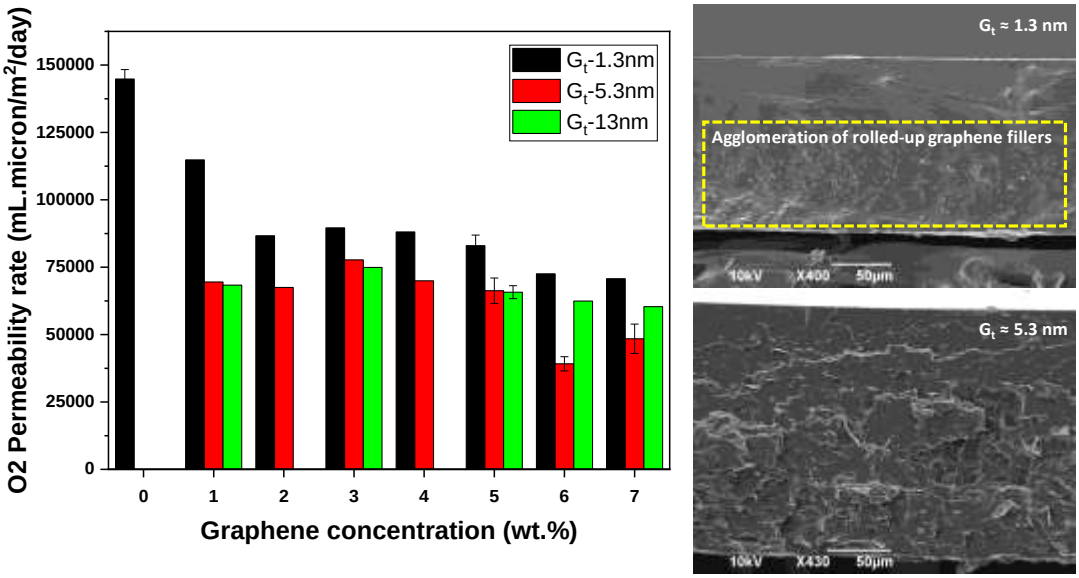
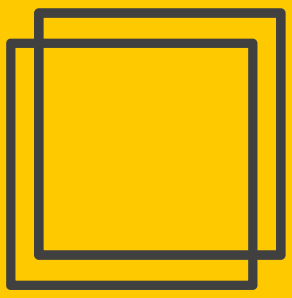


Figure 1: O_2 Gas barrier properties of the TPU/graphene composites loaded with graphene of different thickness or number of layers and their corresponding cross-sectional SEM images. (G_t indicate graphene thickness in nm).

"This project has received funding from the European Union's Horizon 2020 research and innovation program under grant agreement No. 785219 – GrapheneCore2".



POSTER CONTRIBUTIONS

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Neutron Scattering Instruments for Nanostructure Characterization at HANARO, Korea

Small angle neutron scattering(SANS) technique non-destructively probes structures in materials on nano-meter length scales(1nm ~ 1000 nm) and has been a very powerful tool in various scientific and engineering research areas of polymers including biopolymers, complex fluids, colloidal systems, protein folding and protein complexes, nano-magnetic materials including magnetic recording media, metals and alloys, ceramics and the flux-line lattice in superconductors. Neutron reflectometery (NR) is a technique for probing the surface and buried interface of thin films with a 10–5000 Å thickness by measuring the reflected neutrons at glancing angles from the surface. The technique can be used to examine characteristics such as a thickness, surface roughness, interfacial roughness, composition and any defects with a resolution down to 10 Å along the depth direction. The 30MW HANARO research reactor at the Korea Atomic Energy Research Institute (KAERI) is a world class research reactor which has a peak thermal neutron flux of 5.4 x 10^{14} n/cm^2/sec and has cold neutron source. There are three small angle neutron scattering instruments and two neutron reflectometers which are operational at HANARO. In this study, the 40m SANS instrument and the vertical reflectometer are introduced [1,2] and the research results related to 2-dimensional nanostructure analysis using two low-q neutron scattering instruments are presented.

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Figures

Figure 1: Cold neutron guide hall at HANARO

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First-Principle Studies of Ni/hBN/Ni nano-spin-valve-like Structure for Spin Electronics

Hexagonal boron nitride (hBN) sandwiched between magnetic nickel layers has been theoretically studied within the framework of generalized gradient approximations of density functional theory. Possible thirty-six stacking arrangements of nickel layers with respect to hBN were considered to determine the most stable stacking structure. Our results show that strong hybridization between hBN and nickel slabs occurred when nickel atoms of the first nickel layer placed on the top or below nitrogen atoms. The strong hybridization leads the N-top stacking arrangement as the most favored energy state. This result is an analog to previous theoretical studies of Ni/hBN interface which found that nitrogen atom on-top sites of nickel layer with boron atom on the hcp- or fcc-sites (second or third layer respectively) are the most energetically favored stacking arrangement[1]. Interestingly, hBN-nickel sandwich has an asymmetric stacking arrangement on the second and third nickel layer between upper and lower nickel slab as the most favored energy as shown in figure 1. This asymmetric stacking arrangement is in constrast with graphene-nickel sandwich that has a symmetric structural arrangement[2]. Furthermore, within the most stable structure, the anti-parallel spin configuration system has an energy lower than parallel spin configuration which is in agreement with previous experimental study[3]. Starting from this point, further calculation to explain the current path of Ni/2D/Ni nano-spin-valve system can be derived.

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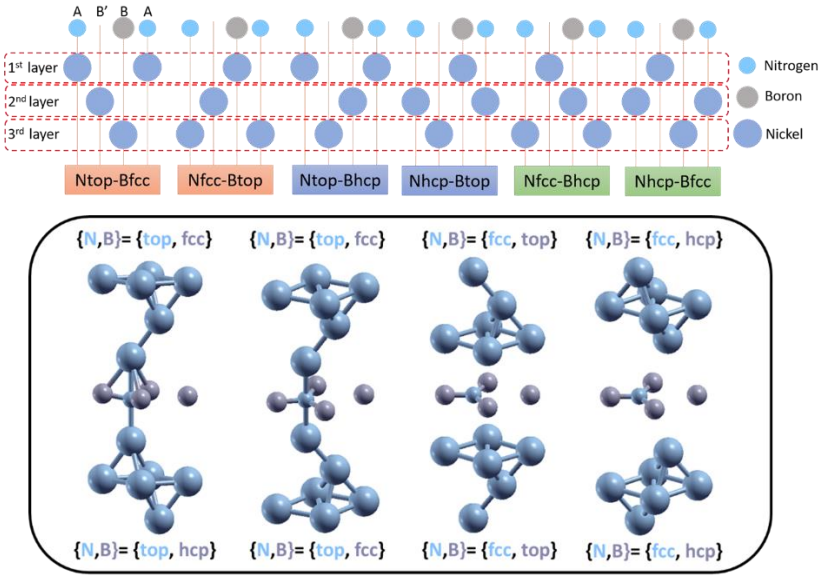

Figure 1: Various stacking arrangement on Ni/hBN interface (upper). Some representative figures as the results from thirty-six configurations. From left to right, the total energy increasing from the most stable to the unstable structure (lower). The combination of top-fcc on the top side and top-hcp on the bottom side are the most stable structure.

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Preparation of Stable and Impurity-Free Graphene Oxide Dispersion in Various Organic Solvents

In this study, we demonstrate a simple two-step method for producing stable graphene oxide (GO) dispersions in various organic solvents without stabilizers or chemical modifications. In this method, the exfoliation and dispersion processes, which are commonly performed simultaneously, are separated. Perfectly exfoliated single-layer GO is firstly prepared in water and then water is carefully replaced with each solvent. Although this new method is simple and requires no stabilizers or chemical modifications, it achieved GO dispersions with much improved long-term stability in organic solvents compared to those prepared by the conventional method (Figure 1). Additionally, impurity contents such as S and N were minimal. A simple GO/PVC composite is also presented in this study. The GO/PVC composite prepared by the new method showed better mechanical and electrical performances compared to that prepared by the conventional method. This facile process for forming stable GO dispersions will fulfill the needs of scientists and engineers in developing applications using GO.

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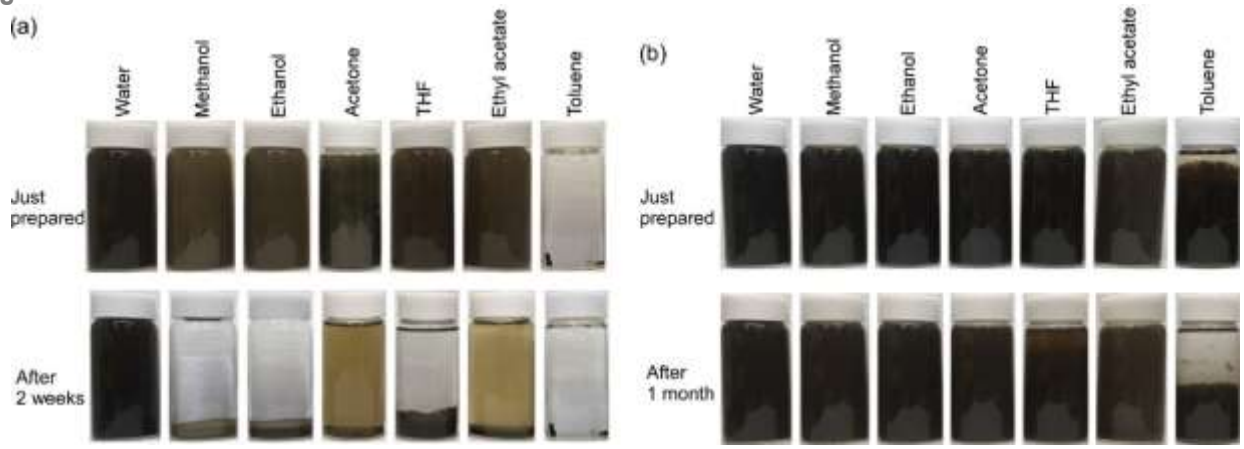

Figure 1: Photographs of (a) GO dispersions prepared by the conventional method, just prepared and after two weeks, and (b) GO dispersions prepared by the new method, just prepared and after one month.

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Graphene Synthesis on Single Crystal Copper Foil

Generally, graphene is synthesized by CVD method using a polycrystalline copper thin film as a catalytic metal. Copper films are preferred because of their low cost, low carbon content and relatively high melting temperatures.
However, it is known that graphene synthesized from polycrystalline copper thin films has many wrinkles and defects. This is because the surface of the copper film is composed of crystal grains of various planes. Recently, graphene synthesis has been attempted on the surface of monocrystalline copper since there is no grain boundary. However, monocrystalline copper is very expensive in cost, and hard to obtain. In this study, we investigated the effect of oxygen on monocrystallization of polycrystal copper film using SEM, Raman, XPD, XRD and SIMS. Single crystal graphene was synthesized and the I-V curve of FET was measured to characterize the graphene.

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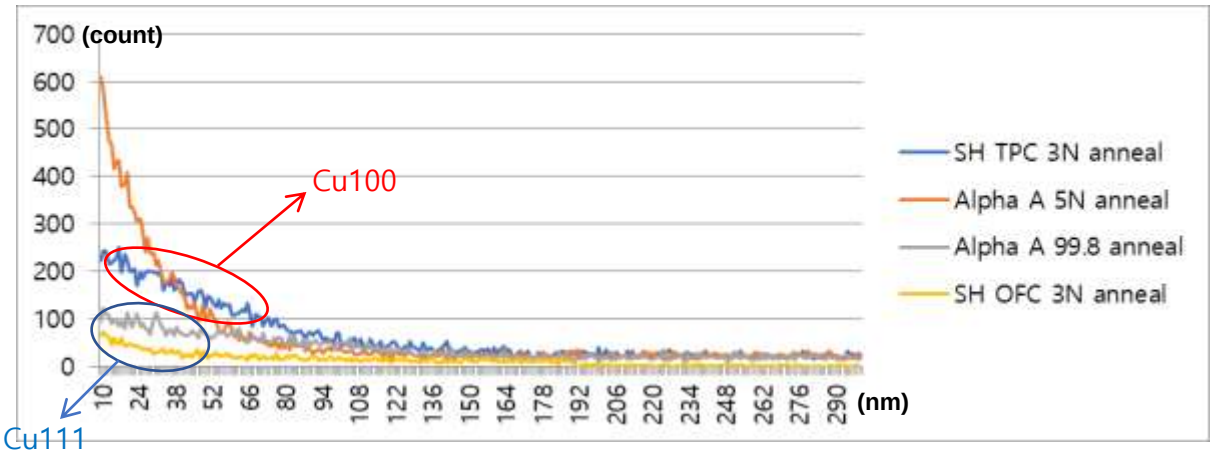

Figure 1: Oxygen content in annealing Cu foil (TOF-SIMS)

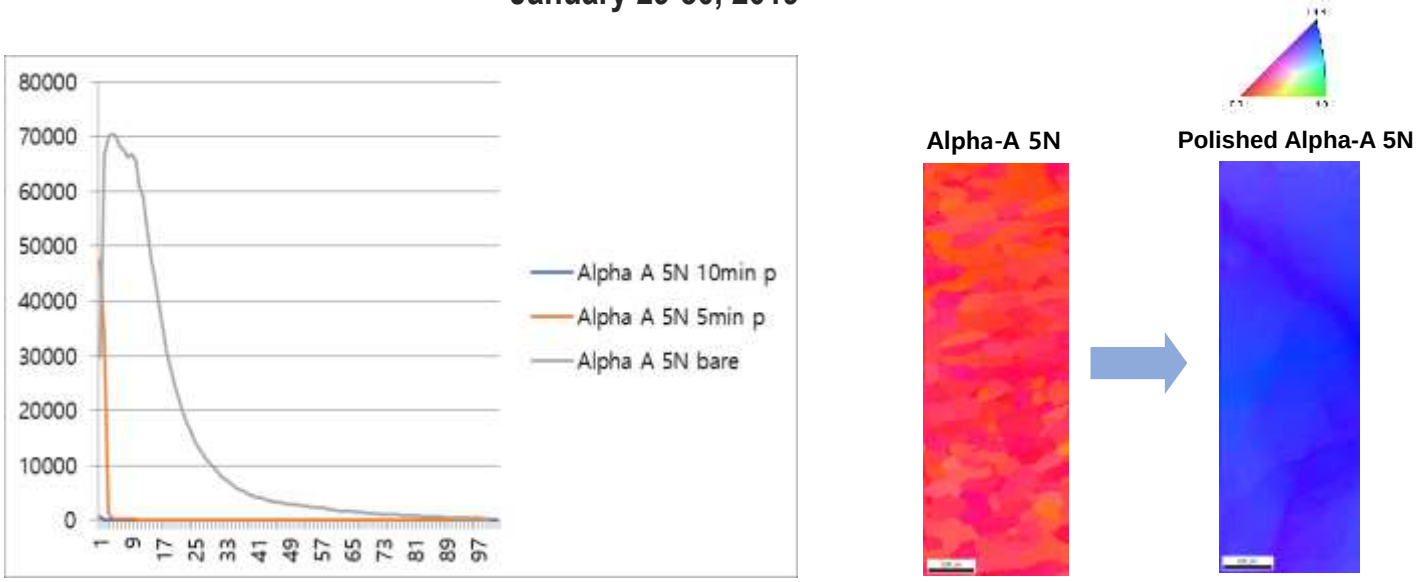

Figure 2: (a) Oxygen content in polished Cu foil (TOF-SIMS) (b) phase orientation of annealing Cu foils (EBSD)

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Soft Aerogels Supported by ~1 mass% Carbon Nanotubes for Thermal Interface Materials

Thermal interface materials (TIMs) are used for enhancing heat transfer between solid surfaces by creating thermal paths. Their important characteristics are high thermal conductivity and softness for filling the air gaps. However, many conventional TIMs consist of the thermal conductive fillers dispersed in polymer matrix, which has disadvantages of low thermal conductivity and poor thermal stability. Here we propose aerogel TIM replacing polymer matrix with air matrix. Air matrix is released upon pressing and conductive fillers can directly contact each other. A soft sponge like self-supporting film can be fabricated using carbon nanotubes (CNTs), and this structure is able to support fillers 100 times larger in mass compared with their own mass [1]. Moreover, the CNTs do not disturb heat conduction between fillers because CNTs have high thermal conductivity, and the CNTs also have high thermal stability (500 °C in air). We selected silver having the highest thermal conductivity among the metals as conductive filler. The aerogel TIM was fabricated by hybridizing Ag particles with CNTs (Fig. 1a, b). The TIM was self-supporting even when the amount of fillers was 200 times as large as that of CNTs. From the result of steady-state thermal resistance measurement, the thermal resistance of 99.5 mass% Ag-0.5 mass% CNT-TIM between two Cu rods was 40 mm^2 K/W under 0.8 MPa (Fig. 1c). TIM showed lower thermal resistance with lower CNT content. In addition, selecting the surfactant for keeping Ag particle clean and dispersing CNTs well is one of the key factor to obtain lower thermal resistance. Optimization of the structure is now underway, and the latest results will be reported.

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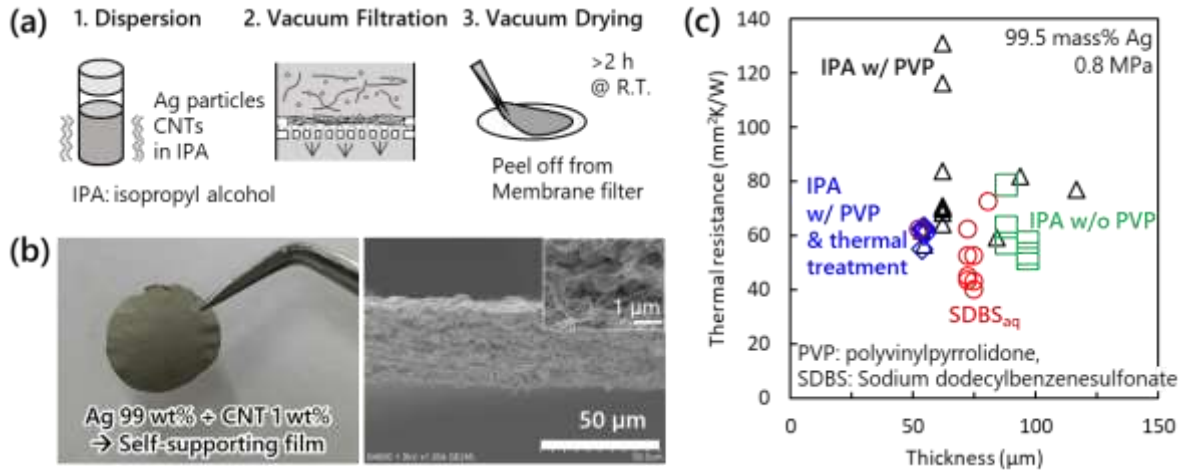

Figure 1: Fabrication process and thermal resistance of Ag-CNT aerogel TIMs. (a) Fabrication process. (b) Digital and SEM images. (c) Thermal resistance of the TIMs with different surfactants.

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CNT and graphene-based oxide composites for energy storage applications

Abstract
Numerous efforts have been reported regarding improving the rate capabilities of oxide-based intercalation materials by decreasing Li+ or Na+ diffusion length and increasing electronic conductivity. For example, nano-structured oxides were coated with conductive carbon layer to improve the electronic conductivity of oxides, thus providing a continuous electronically conductive network in the electrode. Various organic carbon sources such as glucose, sucrose, and citric acid were used to coat carbon layer on $NaTi_2(PO_4)_3$ NTP particles.[1] However, there is a growing concern that a carbon layer may hinder ion transport to oxide, thus limiting the rate capability, since thicker coating layer could act as a physical barrier to the transport of ions to the oxides.[2] There is an interplay of carbon layer between electron conduction pathways and physical barrier to the transport of Na+ ions.
As an alternative to conductive carbon coatings, nanocarbons such as 0 D carbon black, 1 D CNT and 2 D graphene have been employed to prepare metal-oxide/nanocarbon composites as an approach to provide electron conduction pathways to oxides, in which nanosized oxides were deposited on nanocarbons using solution-based synthesis.[3, 4] Since the morphology, size of metal oxides, and their contact nature with nanocarbons can be tuned by the surface properties of the underlying nanocarbon substrates, electrochemical properties of metal-oxide/nanocarbon composites should be compared and analyzed on the basis of not only different nanocarbon dimensions, but also surface properties of the underlying nanocarbon substratesIn this study, we report on the influence and mechanism of different dimensions and surface properties of the underlying nanocarbon substrates on the active NTP nanoparticles are investigated and rational design of the composites using nanocarbons are proposed.

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Multi-channel (NIR, DUV) Photodetectors Based on Quasi-Two-Dimensional Graphene/β-Ga_2O_3 Schottky Junction

Photodetectors are considered one of the most widely applied optoelectronic devices. As the demand for solar-blind ultraviolet (UV) photodetectors in various military and civilian applications increases, wide bandgap semiconductor materials have gained significant attention. Beta-gallium oxide (β-Ga_2O_3) is one of the promising materials for such applications. It has an ultrawide bandgap of 4.8-4.9 eV and therefore intrinsic solar-blindness. Its high chemical and thermal stabilities and superior breakdown field are some of other advantages in fabricating optoelectronic devices [1]. Integrating with other semiconductor materials can help β-Ga_2O_3-based devices overcome limitations such as low responsivity and thermal conductivity. Interestingly, bulk β-Ga_2O_3 can be mechanically exfoliated into microflakes although it is not a van der Waals material. Its unique lattice structure allows mechanical cleavage along (100) direction, and this provides quasi-two-dimensional (quasi-2D) property [2]. β-Ga_2O_3 can now be easily stacked with other 2D materials using dry-transfer method. In this experiment, 2D graphene was applied to form heterojunction with quasi-2D β-Ga_2O_3 flakes. Based on graphene/β-Ga_2O_3 Schottky junction, metal-semiconductor field-effect phototransistors were fabricated. The phototransistors showed excellent performance and high response to UV light (254 nm). The highly transparent graphene gate was efficient in controlling the photodetector parameters and achieved very low dark current while the rejection ratio and detectivity were superior among other solar-blind UV photodetectors. In addition to deep-UV photodetection by β-Ga_2O_3, near-IR detection from graphene was enabled through graphene/β-Ga_2O_3 Schottky diode. Further results and discussion will be presented in detail.

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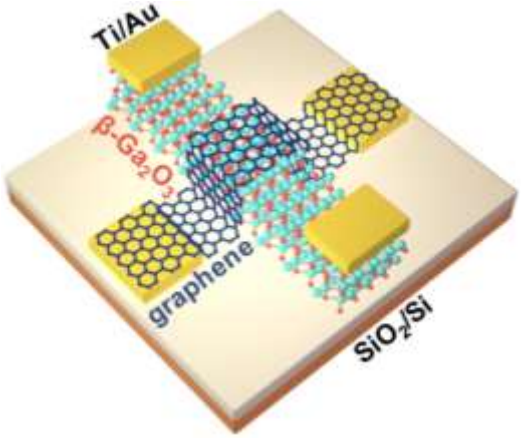

Figure 1: Schematic of metal-semiconductor phototransistor based on graphene/β-Ga_2O_3 heterojunction

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Two-dimensional materials-based double heterojunction bipolar transistors with high current amplification

Two-dimensional material (2DM) based heterojunctions have received great attention because of their fundamental physical and electrical properties. Especially, the properties of 2DMs with weak van der Waals interaction, atomically sharp interface and no dangling bond make them a suitable material for the heterojunction devices. These characteristics also have advantages to fabricate high-performance devices such as heterojunction bipolar transistors (HBTs), which have been difficult to implement in conventional epitaxy due to the problems of lattice constant mismatch.

In our study, we successfully fabricated npn double HBTs by vertically stacking MoS_2 (n-type) and WSe_2 (p-type) flakes. At first, bottom n-type MoS_2 flake was dry-transferred onto the SiO_2/Si substrate. Then p-type (WSe_2) and n-type (MoS_2) flakes were vertically stacked in order on to the previously transferred MoS_2 flake using micro-manipulator. After that, electrodes (Ti/Au for MoS_2 and Pt/Au for WSe_2) for the emitter, base, and collector were fabricated using standard e-beam lithography. The formation of the two p-n junctions in base-emitter and base-collector was experimentally observed. Our fabricated npn double HBT showed excellent electrical characteristics with highly amplified current modulation (β = ~100, α = ~1), which is comparable to that of conventional semiconductor devices. The detail of our experiment and results will be presented at the conference.

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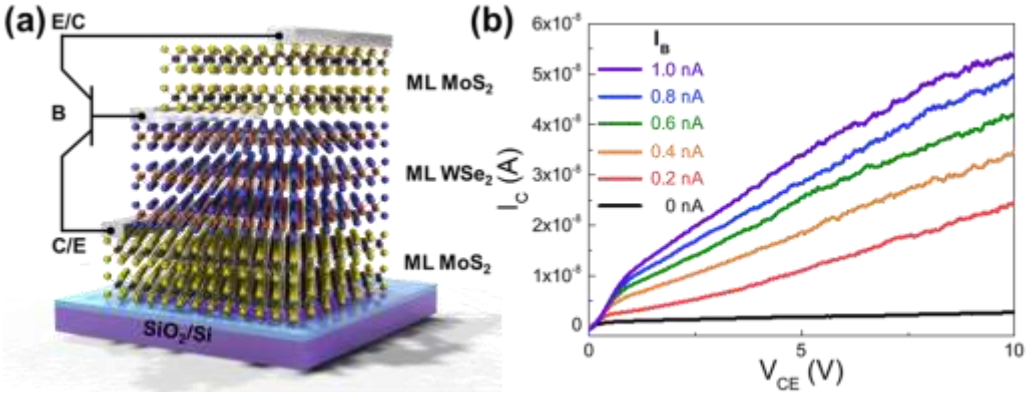

Figure 1: (a) schematic of an npn double HBT (b) output characteristics of a fabricated npn double HBT

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Controllable and reproducible Major Carriers of few-layered MoTe2 Transistors by Self-Heating treatment

Through electro-thermal doping (E-doping) processes, accurately controllable and reversible p/n-type electronic doping of 2H-molybdenum ditelluride (2H-$MoTe_2$) transistors is realized without any chemical dopant at room temperature.[1] E-doping processes include electron (n-type) doping and hole (p-type) doping, achieved by an electric field in a vacuum chamber and exposure to air. It is exactly beneficial to modern manufacture by using this simplicity doping processes. Predictably, complementary metal oxide semiconductor-like (CMOS-like) logic circuits were successfully achieved, such as an inverter, a NOR gate, and a NAND gate, through controllable and reversible p/n-type doping of $MoTe_2$ transistors.[2] Based on the method of p/n-type doping presented in this study, a technical feasibility was provided to develop novel 2D-based optoelectronic devices. E-doping is therefore potentially useful for optoelectronic nanodevice applications.

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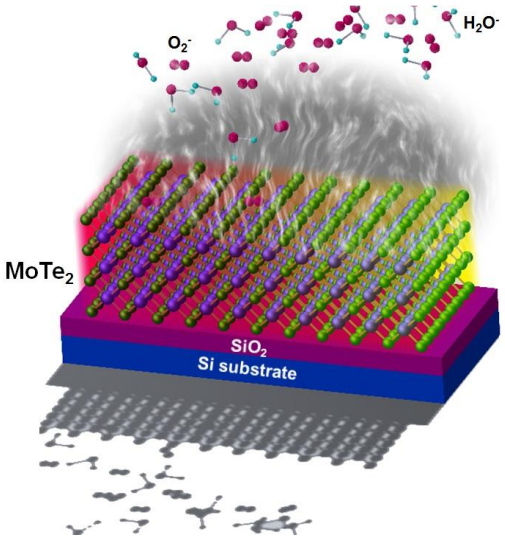

Figure 1: E-doping processes of MoTe2 conduc-ting channels. Schematic of a MoTe2 channel processed by E-annealing under an electric field in vacuum.

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High Efficiency Single Wall Carbon Nanotube Synthesis Using Single- and Multi-layered Porous Substrates

Abstract
Over the past 25 years since the discovery of the single-wall carbon nanotube (SWCNT), the field has taken great strides in the development of SWCNT-based applications. However, the SWCNT cost is one of the primary bottlenecks for establishing a SWCNT-based industry. Therefore, the research and development of SWCNT mass production technologies (high growth efficiency synthetic technology) to achieve low production cost, are essential for realizing the practical use of "SWCNT products". Super-growth (Water-assisted) CVD affords a method of high growth rate, high efficiency synthesis, and high purity of millimeter-tall SWCNTs [1]. For further improvements of the SWCNT growth efficiency have been reported by a number of researchers using various approaches. In this study, we focused on the substrate designs as a new approach for high efficiency SWCNT growth. Specifically, we investigated the use of using porous substrates composed of wire meshes on increasing SWCNT growth rates as well as real growth yield.
Here we show an exceptionally high-efficiency synthesis of long SWCNT forests using porous substrates (metal meshes) in place of nonporous flat substrates. This study examined the dependence of the growth efficiency on various mesh structures, including wire diameter, aperture size, and total surface area. We demonstrated that the synthesis of SWCNT forests is highly dependent on the initial porosity as well as maintaining the open pores throughout the duration of the growth; we achieved the high efficiency synthesis of SWCNT forests (height: >3.47 mm, average growth rate: 301 mm min^{-1}, and yield: 12.7 mg cm^2 in 10 min growth time). Furthermore, we showed that the initial growth rates exceeded 1 millimeter per minute (1,000 mm/min) [2]. Based on these results, we also implemented the high yield SWCNTs using a multi-layer, three-dimensional (3-D) metal mesh porous substrate consisting of eight parallel and evenly spaced mesh substrates. SWCNT yield in this synthesis improved 78-times compared to that of a single flat nonporous substrate. In addition, the total SWCNT yield was greater than eight-fold (mesh layered number). This yield improvement caused from the two reasons. First, mesh substrate provided a higher surface area substrates than that of a flat nonporous substrate. Second, the individual substrate layers acts as heating elements for subsequent catalyst layers proving the uniform delivery of the reacted source gases. CNTs grown possessed identical single-walled structure from the first layer to the final (8th) layer [3]. This architecture led to the achievement of the total carbon conversion efficiency of above 80% with single-walled structure (average: 1.2 number of walls). We believe this represents an interesting alternative approach for highly efficient synthesis and the cost reduction of SWCNTs, resulting in the acceleration of SWCNT application developments in our future life.

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Transfer-free fabrication of nanocrystalline graphene field-effect transistor gas sensor arrays

Gas sensor arrays for highly sensitive and selective detection of toxic gases have become a major topic in the recent years. Graphene, a two-dimensional material with high surface-to-volume ratio has shown to be a good candidate for that purpose [1]. Yet the successful fabrication of gas sensor arrays requires a scalable approach other than mechanical exfoliation.
In this contribution we demonstrate the transfer-free mass fabrication of hundreds of nanocrystalline graphene field-effect transistors (ncGFETs). Figure 1a shows the respective measured backgate input characteristics of 524 ncGFETs in ambient environment. Materials characterization using Raman spectroscopy, near-edge x-ray absorption fine structure and atomic force microscopy has confirmed the presence of a bi- to trilayer nanocrystalline graphene [2]. Characterization of the intrinsic gas sensing properties of the ncGFETs using a vacuum probing station have shown detection capabilities for toxic species including ammonia (NH_3), nitrogen dioxide (NO_2) and carbon monoxide (CO). The respective sensitivities under gaseous influence can be seen from backgate input characteristics of single ncGFETs as shown in figures 1b-1d. By the use of gas sensor arrays selectivity can be achieved by tuning the sensitivities of different ncGFETs by controlling the backgate bias. Moreover, from the hysteresis loop of the devices one can conclude on the gaseous species.

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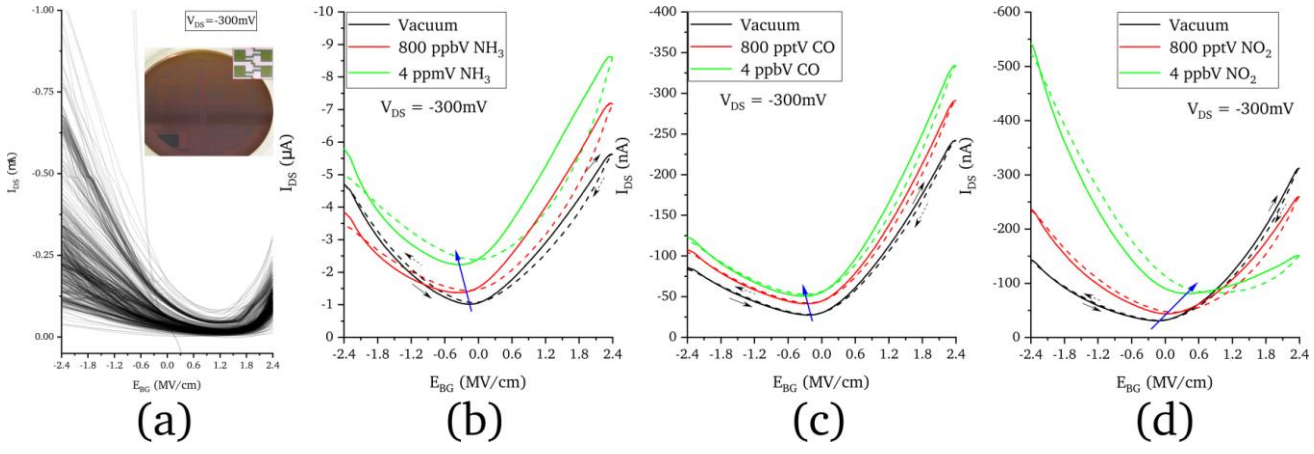

Figure 1: (a) Input characteristics of 524 ncGFETs measured in ambient air, inset shows 2" wafer with the respective devices. Input characteristics of different devices under influence of (b) NH_3 (5.0), (c) 1,000 ppm CO in dry technical air, (d) 1,000 ppm NO_2 in dry technical air. Concentrations have been calculated from the vacuum pressure in the chamber

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Cytotoxicity of different nanocarbon materials tested on HeLa cells

Cytotoxicity for HeLa cells [1] was investigated in the case of different nanocarbon materials, i.e. graphite(S1), expanded graphite (S2), graphene oxide prepared using a modified Hummers method from graphite (S3) and expanded graphite (S4), graphene oxide reduced under various conditions with sodium borohydride (S5), hydrazine (S6), hydrazine under microwave pressure conditions (S7) and vitamin C (S8) using MTT assay (Fig. 1).

The surface of the investigated specimens studied using X-ray photoelectron spectroscopy (XPS) and reflection electron energy loss spectroscopy (REELS) revealed chemical and structural differences, i.e. in the number of carbon sp^2/sp^3 hybridizations, content of chemical groups like hydroxyl (C-OH), epoxy (C-O), carbonyl (C=O), carboxyl (C-OOH) and average number of graphene layers in stacking nanostructures [2]. The average cytotoxicity for HeLa (cell death percent) is directly proportional to the number of C sp^2 bonds and inversely to C sp^3 hybridizations, epoxy and hydroxyl groups (Fig. 2). No dependence on the average number of graphene layers in graphene stacking nanostructures was observed.

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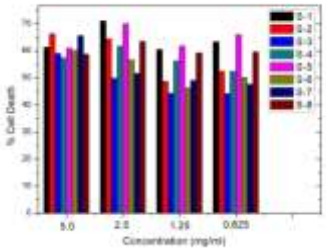

Figure 1: Cytotoxicity for HeLa cells of different nanocarbon materials.

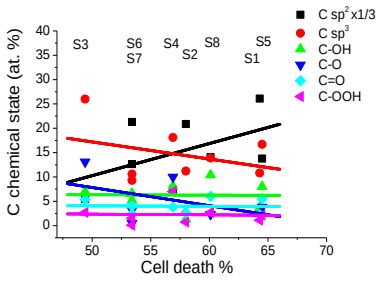

Figure 2: Dependence of average cytotoxicity for HeLa cells on carbon chemical state content at the surface of nanocarbon materials.

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A Minute Amount of Noble Metals Added into Iron Catalyst Enabling the Highly Efficient Synthesis of Single-walled Carbon Nanotube Forest without Reducing Gases

Abstract
Pre-annealing iron thin film in reducing gases such as hydrogen (H_2) are one of the common factors in most of the previous reports preparing the catalyst suitable for the synthesis of tall single-walled carbon nanotube (SWNT) forest [1,2]. To study the fundamental requirement for the preparation of active catalyst, we survey the effect of metal composition of catalyst materials. Here, we report a minute amount (< 0.5 at%) of noble metals (NM = iridium, rhodium, and platinum) added into iron catalyst unexpectedly enabled a highly-efficient synthesis of tall (>600 μm) CNT forests without any reducing gases. Raman and TEM analyses suggested that those CNT forests mainly composed of SWNT. X-ray photoelectron spectroscopy (XPS) analyses on the catalyst films suggested that NM addition invoked the reduced oxidation states of iron atoms in prior to SWNT synthesis. This work was supported by JSPS KAKENHI Grant Number JP17K14090.

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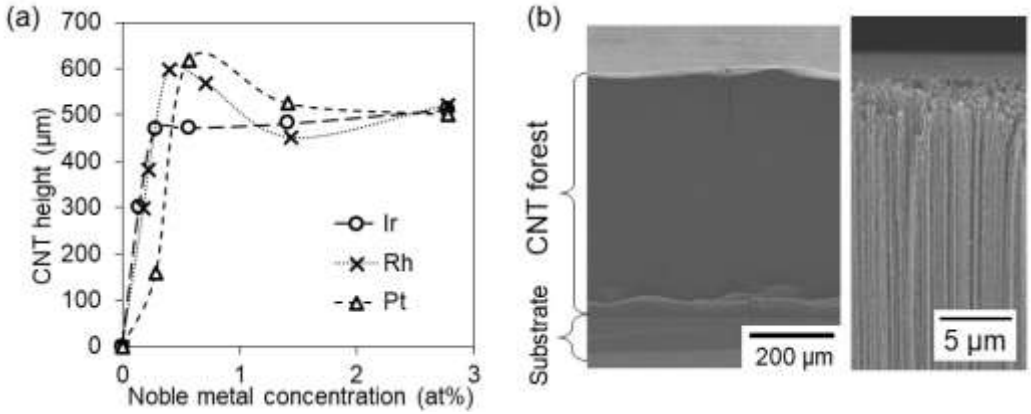

Figure 1: (a) Height of CNT forest (Grown within 10 minutes) vs Atomic concentrations of noble metal (Circle: Iridium, Cross: Rhodium, Triangle: Platinum) added into iron catalyst. (b) SEM images of CNT forest from Rh 0.4 at% catalyst (inset: Photograph from top of the forest).

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Magnetic Field Dependence of Opening and Closing Dirac Cone in Ni/Graphene/Ni nano-spin-valve-like structure

We present magnetic properties and electronic structure studies of a graphene-based nano-spin-valve-like structure theoretically. Magnetic nickel layers on both sides of graphene are considered. A spin-polarized generalized-gradient-approximation determines electronic states. In the most energetically stable stacking arrangement of graphene and two nickel layers, the anti-parallel spin configuration of the underlayer and overlayer magnetic moments has the lowest energy[1], which is in agreement with previous experimental studies.[2] The spin density mapping and obtained band-structure results show that when upper and lower Ni(111) slabs have anti-parallel (parallel) magnetic-moment configuration, the carbon atoms of sublattice A and B will have antiferromagnetic (ferromagnetic) spin configuration. A band gap at the Dirac cone is open when the alignment is anti-parallel configuration, and it is closed when the alignment is parallel configuration[1]. Therefore, the in-plane conductance of the graphene layer depends on the magnetic alignment of two nickel slabs when the Fermi level is adjusted at the Dirac point. This result will give arise the reconsideration of the Klein paradox on graphene[3] as we consider the conductance of graphene nano-ribbon with the partially sandwiched by magnetic nickel layers from upper and lower side as shown in figure 1.

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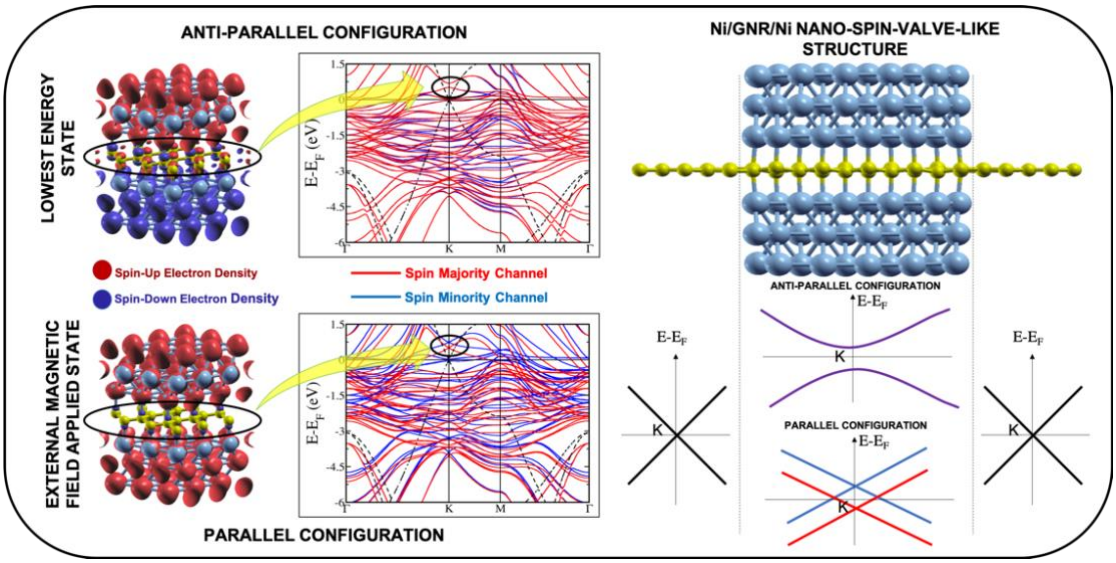

Figure 1: Spin-dependent charge density mapping (left) and bandstructure of Ni/Graphene/Ni nanostructure (middle). When the magnetic moment between upper and lower nickel slabs is antiparallel (parallel), the Dirac cone of graphene is open (close). The Klein paradox on graphene need to be reconsidered by proposed partially sandwiched structure of graphene nano-ribbon with nickel layers from upper and lower side (right).

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Separation of Graphite and Graphene Particles By Hydrocyclone

Hydrocyclones have been used for mineral processing for more than 100 years. Nowadays hydrocyclones are extensively used in the industry to remove or classify particles, and to separate particles by density or size [1-3]. Hydraulic residence times for hydrocyclones are about 1-2 seconds, compared to several minutes for traditional gravity separator. Therefore, it is benefit to use hydrocyclone as a classifier in industrial processing.

In this study, hydrocyclone (Figure 1) is used to separate graphite and graphene particles. The underflow diameters for Type A and Type B hydrocyclones are 10 and 5 mm, respectively. The comparison of the two separators in the same operation conditions is discussed. For 2.6% total solid weight concentration of mixed graphite and graphene particles, the experimental results show that Type B hydrocyclone could separate raw materials to 1.9% overflow and 4.6% underflow, which meet the process demand of Nitronix Nanotechnology Corp. in Taiwan.

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Figures

Figure 1: Hydrocyclone used as a classifier to separate graphite and graphene.

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Enhancing Photovoltaic Performance of Solution-Based Si Heterojunction Solar Cells by 1-min Surface Passivation

Solution-based heterojunction is emerging for facile fabrication of Si-based solar cells due to the room temperature manufacturing and solution processing capability [1]. Surface passivation of Si substrate is established well for the photovoltaic (PV) performance enhancement of the conventional bulk Si cells but not of the heterojunction cells. Pristine Si usually has high-concentration, non-saturated dangling bonds at the surface that causes high local carrier recombination rates. Inserting a thin oxide layer between Si and contact materials could provide an intermediate "i" region for the p-i-n device, which greatly suppresses carrier recombination and build an internal electrical field. For the emerging solution-based Si heterojunction solar cells, the surface oxide layer has been introduced by exposure to air or chemical oxidation, which resulted in passivation layer of insufficient quality [2]. Therefore, it is essential to realize a high-quality passivation layer via a simple process.

Here, we report a facile and repeatable method to passivate the Si surface by a simple 1-min annealing process in vacuum, and integrated it into the heterojunction cell with poly(3,4-etyhlenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) and that with carbon nanotube (CNT). A 1-nm-thin and dense oxide layer was introduced on the Si surface to provide high-quality hole transport and passivation, which enhanced the power conversion efficiency (PCE) from 9.34% to 12.87% (1.38-times enhancement) for the PEDOT:PSS/n-Si cells (Figure 1) and 6.61% to 8.33% (1.26-times enhancement) for the CNT/n-Si cells. The simple surface passivation will enhance the PV performance of the Si-based heterojunction cells with various materials without losing the easiness of the cell fabrication.

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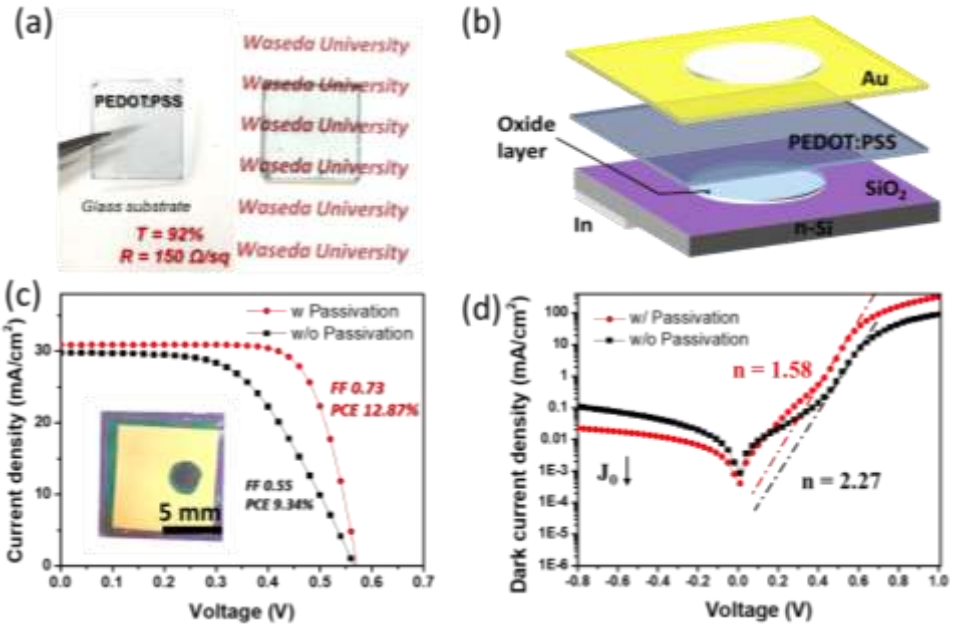

Figure 1: Solution-based PEDOT:PSS-/n-Si heterojunction solar cell with 1-min surface passivation.

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Carrier Transport Mechanisms in Atomically Thin van der Waals Tunnel Field-Effect Transistors and Its Potential for Applications

Van der Waals (vdW) heterostructures present a promising application for tunneling devices.[1,2] Here, we demonstrate vertical broken-gap (type-III) BP/MoS_2 vdW heterostructure, as shown in Figure 1, for the investigation of its transport mechanism. Through temperature dependent electrical measurements, the band-to-band tunneling (Figure 2A) as well as negative differential resistance (Figure 2B) can be observed in our vdW transistors. Besides, a low subthreshold swing can be achieved using such the type-III transistor (Figure 2C). We further interpret the carrier transport mechanism in detail using the variation of energy band diagrams (Figure 2D). This work undoubtedly gives insight into the fundamental understanding of carrier transport in the type III heterostructure transistors and also pave an avenue for developing power-saving electronics.

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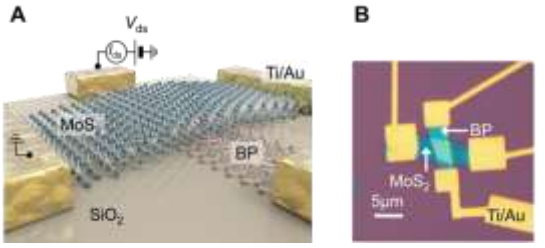

Figure 1A: Schematic of a BP/MoS_2 vdW transistor with a circuit load.
Figure 1B: An optical image of the as-fabricated vdW transistor.

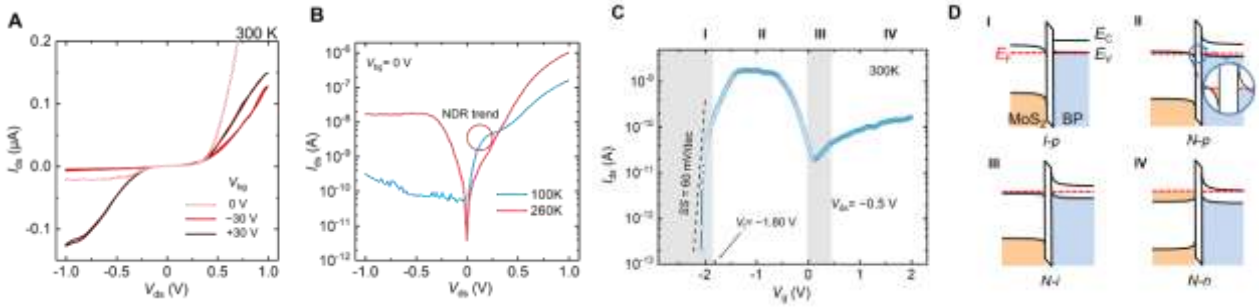

Figure 2A: Output characteristics of the vdW transistor in linear scale at different V_{bg} at 300 K.
Figure 2B: Output characteristics in log scale at various temperatures under V_{bg}= 0 V.
Figure 2C: Transfer characteristic of the vdW transistor with a top gate configuration at V_{ds}= –0.4 V at room temperature.
Figure 2D: Energy band diagram variations of the BP/MoS_2 vdW transistor at various V_g conditions.

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