

# Tuesday Evening, December 10, 2024

## Nano and 2D Materials

### Room Naupaka Salon 4 - Session NM1-TuE

#### Synthesis and Manipulation

Moderator: Sarah Burke, University of British Columbia

#### 6:20pm NM1-TuE-3 On-Surface Formation of One-Atom Thick Carbon-Based Low-Dimensional (1D/2D) Nanomaterials: A Surface and Nanoscale Science Approach, *Maryam Ebrahimi*, Lakehead University, Canada

The second quantum revolution encompasses novel approaches to manipulate matter at the quantum level which is determined by their design, chemical building units, and lattice symmetry. Low-dimensional nanomaterials including zero-, one-, and two-dimensional (0D/1D/2D) materials have revolutionized the world of materials over the past two decades. On-surface synthesis approach offers a bottom-up platform for the discovery, creating, and the manipulation of carbon-based 1D/2D quantum materials with stable quantum states. Our group works on the rational design and formation of ordered 1D/2D carbon-based nanomaterials with one-atom thickness on single crystalline surfaces. These materials include 2D covalent organic frameworks (2D-COFs), 2D metal-organic frameworks (2D-MOFs), 1D/2D polymers, and 2D self-assembled molecular networks, with different lattice symmetry. We employ the state-of-the-art tools of surface and nanoscale science and technology including scanning tunneling microscopy and spectroscopy, atomic force microscopy, and surface characterization spectroscopy techniques to unveil the structural properties of these nanomaterials. The lattice symmetry of 1D/2D networks together with the chemical nature of their building units determine their electronic band structure and properties, promising for quantum technology and organic electronic applications. In this talk, I will review our recent 1-7 and current research on the formation of several 1D/2D molecular-based nanomaterials with different lattice symmetry. I will present the role of molecular symmetry, functional groups, and surfaces, in the formation of molecular self-assembly, metal-organic frameworks, and covalent networks. These ordered nanomaterials are formed and characterized in-situ, on Au(111), Ag(111), Cu(111) under ultra-high vacuum, or at solid-liquid interface on HOPG and Au(111)/mica. In addition to the nanoscale science experimental techniques, we use ab-initio theoretical calculations to provide simulated models for the chemical phenomena, reaction pathways, molecular structure, and electronic properties of these 1D/2D low-dimensional materials. 1 N. Cao et al., *Nanoscale*, 2021, 13, 19884-19889. 2 G. Galeotti et al., *Nature Materials*, 2020, 19, 874-880. 3 G. Galeotti et al., *Chemical Science*, 2019, 10, 5167-5175. 4 F. De Marchi et al., *Nanoscale*, 2018, 10, 16721-16729. 5 D. Cui et al., *Journal of the American Chemical Society*, 2017, 139, 16732-16740. 6 M. Ebrahimi, *Nature Chemistry*, 2022, 14, 3-47. 7 C. Jing et al., *Angewandte Chemie International Edition*, 2019, 58, 18948-18956

#### 6:40pm NM1-TuE-4 Synthesis of Uniform Borophene: In Situ Spectroscopic Analysis and Ex Situ Macroscopic Transfer, *Marko Kralj, S. Kamal, B. Radatovic, V. Jadrisko, D. Novko, N. Vujicic, M. Petrovic*, Center for Advanced Laser Techniques, Institute of Physics, Croatia

Borophene (Bo), a recently realized polymorphic mono-elemental two-dimensional (2D) material, holds promise for diverse applications, including metal-ion batteries, supercapacitors, hydrogen storage, gas sensors, and freshwater production. We present a study of Bo fabricated via segregation-assisted chemical vapor deposition (CVD) epitaxy, where an Ir(111) substrate was exposed to borazine vapors in ultra-high vacuum (UHV) at elevated temperatures. This process yielded structurally uniform and high-coverage  $\chi_6$  polymorph of Bo, as evidenced by electron diffraction and scanning probe microscopies. Such Bo samples were further subjected to extensive spectroscopic analysis and post-synthesis manipulation, both of which are scarce due to experimental challenges posed by sample inhomogeneity and significant chemical reactivity of epitaxial Bo systems.

In the first part of this talk, we characterize Bo on Ir(111) (Bo/Ir), focusing on electronic properties influenced by the nanoscopic modulation of the Bo sheet. X-ray photoelectron spectroscopy (XPS) and scanning tunneling spectroscopy (STS) data reveal inhomogeneous binding of Bo to Ir, creating a stripe-like structure. This structure acts as a one-dimensional (1D) grating, causing Umklapp scattering of photoelectrons detected in ARPES experiments [1]. Density functional theory (DFT) calculations support our findings, providing insights into the pristine electronic structure of Bo, unaffected by the Ir substrate.

In the second part, the challenges of developing large-area Bo applications are addressed. We demonstrate large-area growth followed by

electrochemical transfer of macroscopic single-layer Bo sheets from the growth substrate to a target Si wafer [2]. Our results show that deterministic manipulation of Bo layers is feasible despite their inherent chemical and mechanical instability, advancing Bo research and utilization. Post-transfer Bo displayed minimal mechanical defects, such as cracks and holes, mostly inherited from the synthesis substrate. Successful Bo transfer was further confirmed by Raman spectroscopy, which showed very good overlap of Raman peaks before and after the transfer, indicating preservation of Bo's original crystal structure.

References:

[1] ACS Appl. Mater. Interfaces 14 (2022) 21727-21737

[2] ACS Appl. Mater. Interfaces 15 (2023) 57890-57900

#### 7:00pm NM1-TuE-5 Fabrication of Advanced Nano and 2D Material Devices - Utilizing the Next Generation NanoFrazor Capabilities, *Nicholas Hendricks, E. Çağın*, Heidelberg Instruments Nano AG, Switzerland

Nano and 2D materials are of intense research interest by both academic and industrial communities as these materials provide great promise for next generation electronic devices and various other applications. When fabricating such devices, patterning the electrical contacts with conventional fabrication techniques (electron beam lithography (EBL), photolithography, focused-ion beam (FIB) lithography) becomes challenging and time consuming due to overlay requirements. The use of energetic particles, such as electrons, photons, or ions, can also lead to less than desired device performance due to damage from the charged particles or ultraviolet irradiation as well as contamination from residual resist. The time intensive processing often comes from the random positioning of nanomaterials or 2D flakes on substrates which makes locating such materials and overlaying the intended patterns challenging.

To confront these challenges, thermal scanning probe lithography (t-SPL), enabled by the NanoFrazor, is offering an alternative direct-write nanolithography method that utilizes thermal energy to perform the patterning process [1-4]. t-SPL generates patterns by scanning an ultrasharp tip over a sample surface to induce local changes with a thermal stimulus. By using thermal energy as the stimulus, it is possible to perform various modifications to the sample via removal, conversion, or addition of/to the sample surface. Along with an ultrasharp tip, the t-SPL cantilever contains several other important functions such as an integrated thermal height sensor and an integrated heating element. To complement the nanopatterning capabilities of t-SPL, the NanoFrazor has a direct laser sublimation (DLS) module to pattern larger (>500nm) features such as electrical traces and contact pads. With nano and microlithography capabilities in a single tool, the NanoFrazor provides the resolutions needed for complete device fabrication, which is interesting for nanoelectronics, photonics, molecular sensing, and quantum computing applications.

In this presentation, the background and workings of t-SPL will be introduced along with the fabrication and electrical performance of nanowire and 2D transition metal dichalcogenide (TMD) devices. To complete the presentation, the next generation NanoFrazor capabilities in parallelized large area patterning and automation of user operations will be shown.

1. S. Howell et al., *Microsystems & Nanoengineering*, 6, 21 (2020)
2. Talha-Dean et al., *ACS Applied Materials & Interfaces*, 16, 31738 (2024)
3. B. Erbas et al., *Microsystems & Nanoengineering*, 10, 28 (2024)
4. L. Shani et al., *Nanotechnology*, 35, 255302 (2024)

## Nano and 2D Materials

### Room Naupaka Salon 4 - Session NM2-TuE

#### 2D Materials Based on Carbon and Boron

Moderator: Akitoshi Shiotari, Fritz-Haber Institute

#### 7:40pm NM2-TuE-7 First-Principles Study of Adsorption and Reaction on the Hydrogen Boride Sheet, *Ikutaro Hamada*, Osaka University, Japan

INVITED

The hydrogen boride (HB) sheet [1] is a newly synthesized two-dimensional material composed of hydrogen and boron atoms with their ratio of 1:1. The HB sheet has been attracted increasing attention, not only because of its high gravimetric hydrogen content as a hydrogen storage material and its ability to release hydrogen through light [2] and electrochemical [3] means, but also due to its catalytic activities [4]. Furthermore, the HB sheet has

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been shown to be chemically stable against water [5], unlike many boron hydrides that undergo hydrolysis reactions. The stability is a fascinating considering the practical application of the HB sheet. However, the molecular and mechanistic details of the chemical processes occurring on the HB sheet have yet to be fully addressed. In this talk, I will discuss the electronic and structural properties of the HB sheet [5], as well as the adsorption and reaction on the HB sheet based on our density functional theory calculations. Special emphasis is placed on the interaction with water molecules, and the mechanism of the stability of the HB sheet against water will be discussed [6,7].

References:

- [1] H. Nishio, *et al.*, J. Am. Chem. Soc. **139**, 13761 (2017).
- [2] R. Kawamura, *et al.*, Nat. Commun. **10**, 4880 (2019).
- [3] S. Kawamura, *et al.*, Small **20**, 2310239 (2024).
- [4] T. Goto, *et al.*, Commun. Chem. **5**, 118 (2022).
- [5] L. T. Ta, Y. Morikawa, and I. Hamada, J. Phys. Condens. Matter **35**, 435002 (2023).
- [6] K. I. M. Rojas, *et al.*, Commun. Mater. **2**, 81 (2021).
- [7] K. I. M. Rojas, Y. Morikawa, and I. Hamada (submitted).

8:20pm **NM2-TuE-9 N-doped Graphene Synthesis through Nitrogen Ion Irradiation**, *Zbynek Novotny*, Pacific Northwest National Laboratory; *B. Alupothe Gedara, P. Evans, Z. Dohnalek*, PNNL

Hydrogen (H<sub>2</sub>) is one of the most promising clean and renewable energy sources. Nevertheless, the storage of hydrogen shows poor performance due to the low gravimetric and volumetric densities. Nitrogen-doped graphene (Gr) has been identified as a potential material for H<sub>2</sub> storage. We study the growth of Gr on a Ru(0001) surface by chemical vapor deposition (CVD) of pyridine (H<sub>5</sub>C<sub>5</sub>N) and N-doping through N<sub>2</sub><sup>+</sup> ion irradiation using scanning tunneling microscopy (STM) and x-ray photoelectron spectroscopy (XPS). A high-quality Gr film with low N densities was obtained by pyridine CVD on Ru(0001) at 1100 K. Higher concentrations of N-dopants were introduced on the Gr/Ru(0001) through low-energy N<sub>2</sub><sup>+</sup> irradiation at 100 eV. Nitrogen can be embedded in the Gr lattice preferentially in two configurations, namely graphitic N (N substituted in the C lattice) and pyridinic N (substitutional N next to a C vacancy). Atomically-resolved STM images of graphitic and pyridinic-N defects demonstrate their preferential locations within the Gr Moiré. XPS shows that coverage of up to 3.9% of pyridinic-N and 2.3% of graphitic N can be embedded into the high-quality Gr film using N<sub>2</sub><sup>+</sup> irradiation at room temperature, indicating a preferential formation of pyridinic N over graphitic N. Only graphitic N was observed upon annealing the ion-irradiated Gr/Ru(0001) to 1063 K, revealing higher thermal stability of graphitic N over pyridinic N. Our current efforts center on the adsorption studies of atomic hydrogen, its interactions with N dopants, and thermally induced diffusion.

8:40pm **NM2-TuE-10 Nanoscale Investigation of N-Heterocyclic Carbene Monolayers on Metal Surfaces**, *Francesco Tumino*, Queen's University, Canada, Italy; *E. DesRoche, M. Aloisio, D. Nanan, A. McLean, C. Crudden*, Queen's University, Canada

Molecular monolayers on metal surfaces are two-dimensional (2D) systems of great interest for advanced applications in several fields, such as molecular electronics, photovoltaics and catalysis[1]. The physical and chemical properties of these systems critically depend on the choice of the molecular unit that constitutes the basic building block of the resulting 2D assembly[2].

Recently, N-heterocyclic carbenes (NHCs) have been proposed for the fabrication of robust molecular monolayers, based on their ability to form strong covalent bonds with transition metals[3]. Studies conducted on gold surfaces have shown that NHC-based monolayers can have greater thermal and chemical stability than alkanethiolate 2D layers[4], making them promising for applications in biosensing, surface patterning and corrosion protection[3].

The fabrication of NHC-based monolayers, however, requires a detailed understanding of how the NHC structure and the surface properties affect the molecular adsorption and self-assembly behavior on metals.

Therefore, we have conducted a nanoscale investigation of NHCs on single-crystal metal surfaces under ultra-high vacuum conditions (UHV). Our study includes NHCs with different backbones and N-substituents, deposited onto atomically clean surfaces using a vapor phase method (supplemental document, Fig.1). Low-temperature scanning tunneling microscopy (STM)

was used to image in situ the resulting monolayers and obtain molecular-scale information on binding modes, adsorption geometries, and self-assembly patterns.

We observed different supramolecular assemblies (Fig. 2), in which the binding mode and lattice geometry vary depending on the N-substituents, the backbone structure, and the molecular density on the surface. Moreover, the metal surface properties play an important role, affecting the ability of NHCs to extract metal adatoms from the surface and form mobile NHC-metal complexes.

By discussing different factors which influence the NHC behavior on metals, our work contributes to a wider understanding of the coordination of these ligands to metal surfaces and the formation of 2D NHC-based layers.

- [1] Goronzy D.P., *et al.* "Supramolecular assemblies on surfaces: nanopatterning, functionality, and reactivity." ACS Nano 12.8 (2018): 7445-7481.
- [2] Barth J.V., *et al.* "Engineering atomic and molecular nanostructures at surfaces." Nature 437 (2005).
- [3] Smith C.A., *et al.* "N-heterocyclic carbenes in materials chemistry." Chem Rev 119 (2019).
- [4] Crudden C.M., *et al.* "Ultra stable self-assembled monolayers of N-heterocyclic carbenes on gold." Nat Chem 6 (2014).

## Nano and 2D Materials

### Room Naupaka Salon 4 - Session NM1-WeM

#### Surface Engineering and Characterization

Moderator: Maryam Ebrahimi, Lakehead University

8:20am **NM1-WeM-2 Synthesis, Doping, and Encapsulation of 2D Transition Metal Dichalcogenides**, *Yu-Chuan Lin*, National Yang Ming Chiao Tung University (NYCU), Taiwan

**INVITED**

Two-dimensional (2D) transition metal dichalcogenides (TMDC), such as  $\text{MoS}_2$  and  $\text{WSe}_2$ , exhibit useful material properties and versatile material chemistry for optoelectronic devices, quantum information, and energy missions. To realize these applications, we need to make them in large areas and be able to control their impurity concentrations. First, I will introduce our metalorganic chemical vapor deposition (MOCVD) process for deposition of TMDC epitaxial monolayers and the approach for growth of epitaxial TMDC multilayers on sapphire. Next, I will discuss how we introduced Re or V dopants into the cation sites of TMDC by MOCVD and their impact on the quality and properties of TMDC films.[3] In addition to cation substitutional doping, we can create Janus TMDC with an intrinsic dipole moment by replacing the elements at the anion sites (i.e., S and Se). I will present our result [4,5] of the conversion of 2D  $\text{WS}_2$  and  $\text{MoS}_2$  into 2D Janus  $\text{WSSe}$  and  $\text{MoSSe}$  by pulsed laser deposition and explain how we confirmed the presence of a dipole moment in 2D Janus TMDC optically. To improve the interface quality between TMDC and oxide dielectric materials for electronic applications, we developed thermal atomic layer deposition (ALD) of amorphous boron nitride (aBN) on both traditional and van der Waals surfaces. In the end, I will talk about nucleation and growth of aBN on  $\text{MoS}_2$  surface in our thermal ALD and aBN/ $\text{MoS}_2$  integration for improve field-effect transistor performance and quantum well fabrication.[6]

#### References

(1) Lin *et al.*, *ACS Nano* **2018**, *12*, 965–975. (2) Eichfeld *et al.*, *ACS Nano* **2015**, *9*, 2080–2087. (3) Torsi *et al.*, *ACS Nano* **2023**, *17*, 15629–15640. (4) Lin *et al.*, *ACS Nano* **2020**, *14*, 3896–3906. (5) Zheng *et al.*, *ACS Nano* **2022**, *16*, 4197–4205. (6) Chen *et al.*, *Nat. Comm.* **2024** *15*, 4016.

9:00am **NM1-WeM-4 Small Clusters of Molecular Anions: Locally Probing a Model Hubbard System**, *Sarah Burke*, University of British Columbia, Canada

**INVITED**

Electronic correlations drive many important phenomena in materials that cannot be described by a single-electron picture. The Hubbard model captures a remarkable range of this behaviour by introducing an on-site interaction – the “Hubbard U” – that competes with the kinetic energy of the electrons, or delocalization through wavefunction overlap – described by the hopping parameter  $t$ . While Hubbard models are not generically analytically or even exactly solvable, model systems such as clusters often are and can lend insights into more complex extended structures. The success of such models in describing correlated electron phenomena like Mott-insulator transitions, charge density waves, magnetically ordered states, and superconductivity<sup>1</sup>, is typically assessed by comparing to macroscopic quantities. However, as a model that captures the local character of electron-electron interactions, local tools provide a unique perspective. Noncontact atomic force microscopy (ncAFM) offers a particularly intriguing view through the ability to probe charge states and electrostatic interactions locally.

We used clusters of PTCDA molecules adsorbed on NaCl bilayer films on silver as an experimental prototype of a 4-site Hubbard model in a regime where  $U \gg t$ . PTCDA clusters have weak in-plane hybridization ( $t$  small), and isolated molecules have a Hubbard U in gas phase of  $\sim 3\text{eV}$ , reduced to  $\sim 1.4\text{eV}$  by screening of the nearby silver<sup>2</sup>. The large electron affinity of PTCDA leads to electron transfer from the silver substrate, so that isolated molecules carry a charge of  $-1$ . Thus, these clusters represent a half-filled Hubbard model where localization is expected, connected to a reservoir of charge. We used STM, STS, ncAFM and electrostatic force spectroscopy to probe the structures, charge states, and charging energies of two different geometries of 4-molecule clusters as well as isolated PTCDA molecules. Equilibrium charge distributions of asymmetric clusters show charge segregation, while symmetric clusters show uniform charge. A 4-site extended Hubbard model was mapped onto the experimental results to identify the necessary interactions required to describe the ground state and charge excitations of the system. Once above a threshold where  $U/t$  drives localization, intersite energy differences and repulsion compete with hopping, driving the observed charge segregation and unexpected correlated behaviour.

1. M. Qin, *et al.*, *Annu. Rev. Condens. Matter Phys.* **13**, 1–28 (2021).

2. K. A. Cochrane, *et al.*, *Nat. Commun.* **6**, 8312 (2015)

9:40am **NM1-WeM-6 Tunable Areal Density and Defined Morphology Regimes of Langmuir Monolayers of PEGylated Gold Nanoparticles**, *H. Cameron*, *I. Curtis*, *R. Takai*, Mount Allison University, Canada; *M. Radford*, Simon Fraser University, Canada; *A. Williams*, Mount Allison University, Canada; *B. Gates*, Simon Fraser University, Canada; *M.-Vicki Meli*, Mount Allison University, Canada

The interfacial behaviour and control of nanoparticles has important consequences on their assembly into thin films, superstructures and subsequent materials suitable as metamaterials and nanophotonics applications. Our investigations into the interfacial self-assembly of nanoparticles coated with polyethylene glycol (PEG) and tetradecane thiols as a function of ligand shell and subphase composition will be presented. Methyl- and carboxylic acid-terminated PEG thiols are compared as a function of subphase pH. Film morphology is demonstrated as a function of ligand composition, transitioning from island-forming to isolated individual nanoparticles with controllable areal density. Transmission electron microscopy of the self-assembled films will be presented with corresponding surface pressure vs. area compression isotherms. Insights into NP film purity and (in)stability as it relates to the mentioned parameters will also be discussed. Compression-expansion isotherms with exceptionally large onset areas were measured. The effect of added tetradecanethiol into the ligand shell is shown to improve the stability and maintain the separation of the nanoparticles in the films.

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### Room Naupaka Salon 4 - Session NM2-WeM

#### Properties of 2D Materials

Moderator: Zbynek Novotny, Pacific Northwest National Laboratory

10:20am **NM2-WeM-8 Atomic-Scale Control of Plasmon-Driven Single-Molecular Switch**, *Akitoshi Shiotari*, Fritz-Haber Institute, Germany

**INVITED**  
Localized surface plasmons provide effective molecule–light interactions that are highly motivating for the realization of advanced molecule-based optoelectronic devices [1]; however, innovative precision in plasmon control is still required to induce local selective reactions at the single- or sub-molecule level. Scanning tunneling microscopy (STM) using a plasmonic metal tip not only creates a local photochemical reaction field, but also enhances Raman signals from molecules within the STM junction, namely tip-enhanced Raman spectroscopy (TERS). Low-temperature STM-TERS is a powerful tool to evaluate the structure and photoreactivity of single molecules, even on a semiconductor silicon surface [2].

In this talk I will mainly report on a plasmon-induced single-molecule switch in a Ag-tip–anhydride-molecule–Si-surface junction characterized and controlled by low-temperature STM-TERS [3]. The localized surface plasmon at the tip can dissociate the O–Si bonds between the molecule and the Si(111)-7x7 surface, resulting in the conductance switching between OFF (tunnelling) and ON (point-contact) states of the tip–adsorbate junction. Sub-angstrom scale positioning of the plasmonic tip over the target molecular can tune the switching rate. We also demonstrated that the switching ability can be changed by the single-atom-level substitution of the molecular structure; imide/Si(111) is inert against the tip plasmon. Our demonstrations of the tip-plasmon control and the chemical tailoring for the single-molecule switch pave the way to the fusion of single-molecule electronics and plasmonics, towards ultimate miniaturization of optoelectronics, namely “pico-optoelectronics.”

#### Reference:

1. Xiaona Xu, Chunyan Gao, Ramya Emusani, Chuancheng Jia, and Dong Xiang, *Adv. Sci.* **2400877** (2024) DOI: 10.1002/adv.202400877
2. Borja Cirera, Shuyi Liu, Youngwook Park, Ikutaro Hamada, Martin Wolf, Akitoshi Shiotari, and Takashi Kumagai, *Phys. Chem. Chem. Phys.* (2024) DOI: 10.1039/d4cp01803f.
3. Youngwook Park, Ikutaro Hamada, Adnan Hammud, Takashi Kumagai, Martin Wolf, and Akitoshi Shiotari, *Nat. Commun.* (2024) DOI: 10.1038/s41467-024-51000-w.

# Wednesday Morning, December 11, 2024

11:00am **NM2-WeM-10 Tunable Metasurface with Gap and Collective Surface Plasmon Modes**, *Anatoliy Pinchuk*, University of Colorado at Colorado Springs

We investigate a tunable metasurface composed of a monolayer of gold nanoparticles on a glass substrate in close proximity to a thin aluminum film, both numerically and experimentally. The extinction spectra of the metasurface exhibit three angle- and polarization-dependent peaks. Using the finite-difference time-domain (FDTD) method, we confirm the positions of both the collective surface plasmon and gap modes. Altering the polarization of the incident light causes a shift in the wavelength of these peaks.

A recent study reported strong coupling between the gap and collective surface plasmon polariton modes in a system consisting of a monolayer of gold nanoparticles supported on a glass substrate, separated from a thin aluminum film by a shellac polymer spacer film of varying thickness [1,2]. Two optical extinction bands were identified: a long-wavelength gap mode and a short-wavelength collective surface plasmon mode. The position and amplitude of both modes were found to depend strongly on the thickness of the shellac dielectric spacer. Varying the spacer thickness causes a shift in the two modes and their degeneration into a single mode as the thickness increases. These findings reveal a complex interplay between the gap and collective surface plasmon polariton modes.

Despite detailed numerical and experimental studies on various geometries of plasmonic metasurfaces, all such studies have been conducted with the probing light beam incident normally. There have been no reports on the polarization dependence of the extinction spectra for monolayers of gold nanoparticles in close proximity to a thin metal film. Our study addresses this gap by exploring the effects of polarization on the optical properties of these metasurfaces, providing new insights into their tunable characteristics and potential applications in nanophotonic devices.

## References

1. Yeshchenko, O.A., Kozachenko, V.V., Tomchuk, A.V., Haftel, M., Knize, R.J., and Pinchuk, A.O. "Plasmonic metasurfaces with tunable gap and collective surface plasmon resonance modes," *The Journal of Phys. Chem. C*, Vol. 123, No. 20, 13057-13062, 2019.
2. Yeshchenko, O. A., Kozachenko, V. V., Naumenko, A. P., Berezovska, N.I., Kutsevol, N.V., Chumachenko, V.A., Haftel, M., Pinchuk, A.O., "Gold nanoparticle plasmon resonance in near-field coupled Au NPs layer/Al film nanostructure: Dependence on metal film thickness," *Photonics and Nanostructures- Fundamentals and Applications*, Vol.29, 1-7, 2018.

11:20am **NM2-WeM-11 Probing Inherent Optical Anisotropy in Transition Metal Dichalcogenide Substrates via Mie Scattering-Induced Surface Analysis (MISA)**, *H. Woo*, Korea Research Institute of Standards and Science, Republic of Korea; *J. Han, S. Ji, B. Shin*, Sungkyunkwan University (SKKU), Republic of Korea; *S. Lee*, Nanyang Technological University, Singapore; *Young Jae Song*, Sungkyunkwan University (SKKU), Republic of Korea

We present a novel approach to investigate the optical anisotropy of transition metal dichalcogenide (TMD) substrates using Mie scattering-induced surface analysis (MISA). By employing scattering-type scanning near-field optical microscopy (s-SNOM) and finite-difference time-domain (FDTD) simulations, we systematically studied and directly visualized the Mie scattering patterns of superspherical gold nanoparticles (s-AuNPs) at the nanoscale.

Our research revealed distinct differences in the optical properties of molybdenum disulfide (MoS<sub>2</sub>) and rhenium disulfide (ReS<sub>2</sub>) substrates. While MoS<sub>2</sub> exhibited optical isotropy, ReS<sub>2</sub> demonstrated significant anisotropic behavior, particularly in the near-infrared energy range. This anisotropy was manifested through more pronounced spectral and angular responses in the satellite peaks of Mie scattering patterns.

We observed that the distances between satellite peaks in the Mie scattering patterns varied with the incident angle of light for ReS<sub>2</sub>, but remained constant for MoS<sub>2</sub>. This angular dependence in ReS<sub>2</sub> was consistently observed across different excitation energies, providing strong evidence of its inherent optical anisotropy.

Our findings highlight the potential of MISA as a powerful, non-destructive tool for probing the intrinsic dielectric properties of substrates at the nanoscale. This technique offers superior visualization of nanoscale anisotropies and enables a more nuanced approach to substrate characterization compared to traditional spectroscopic methods.

The insights gained from this study not only advance our understanding of light-matter interactions at the nanoscale but also have significant implications for the development of advanced nanophotonic devices and the field of optical metrology.

Keywords: superspherical-AuNP, scattering-type scanning near-field optical microscope, Mie scattering, anisotropy, substrate

11:40am **NM2-WeM-12 Enhancement of Photocatalytic Water Splitting Upon Induced Structural Evolution and Increase of Phase Polarity of Two-Dimensional Covalent Organic Frameworks**, *Jrjeng Ruan*, National Cheng Kung University (NCKU), Taiwan

The evolution of smectic liquid crystals of newly designed two-dimensional covalent organic frameworks (COFs) has been disclosed for the first time. With obtained changes of phase morphology, selected-area electron diffraction patterns, and Young's modulus via compressive testing, this smectic phase is realized to transfer to nematic phases upon the ph. changes of surrounding solutions. Furthermore, the coordination of selected metal ions for the preparation of M-COFs is also stimulated by conducted ph. changes, which is surprisingly found able to greatly adjust the out-of-plan phase polarity and thus the piezoelectric responses of evolved liquid crystals of synthesized M-COFs. The controlled adjustment of phase polarity has been clarified able to modify energy bandgap, enhance absorption of visible lights, and increase the efficiency of stimulated electron transition to metal active centers upon the absorption of visible lights. As a result, the coordination of metal ions and accompanied elevation of phase polarity significantly promote the photocatalytic water splitting via synthesized M-COF as photocatalysts. Hence, with the coordination of metal ions on evolved COF liquid crystals, this research has investigated the availability of various levels of phase polarity of M-COF liquid crystals, and comprehensively studies achievable impacts and contributions of phase polarity on photocatalytic water splitting.

## Nano and 2D Materials

### Room Naupaka Salon 1-3 - Session NM-WeP

#### Nano and 2D Materials Poster Session

**NM-WeP-1 Introduction to Measurement Uncertainty Evaluation Method and Results of Silicon Nitride Thin Film Layer Thickness and Complex Dielectric Constant, *Yong Jai Cho, W. Chegal*, Korea Research Institute of Standards and Science, Republic of Korea**

SE (Spectroscopic Ellipsometer) has excellent measurement resolution, but measurement quality control of SE requires careful attention because measurement uncertainty can be easily affected by small changes in the device. For SE measurement quality control, thin film layer thickness certified reference materials are mainly used. SE has the advantages of being non-destructive, ultra-fast, and non-contact, but also has excellent high-precision characteristics that can detect a single atomic layer even before it is completely grown, so demand is increasing as a core measurement equipment for the semiconductor device manufacturing process. As semiconductor device manufacturing technology has recently developed due to the complexity of semiconductor device structures, the introduction of atomic layer-level processes, and the introduction of new materials, there is a need to improve quality control of TTTM (Tool-To-Tool Matching) related to measurement uncertainty. Therefore, in order to improve the measurement quality control level of SE equipment for semiconductor processing, it is necessary to develop higher quality thin film layer thickness certified reference materials. As a first research step, our research team developed a unified uncertainty evaluation method for the measurands of various types of rotating-element spectroscopic ellipsometers with excellent real-time measurement performance[1]. This method allows the measurement uncertainty of ellipsometric transfer quantities (e.g. Psi and Delta) to be calculated directly from observed ellipsometric transfer quantities, so there is no need for the analysis results of unknown optical properties of the sample required to calculate the measurement uncertainty. As a second research step, we developed a method for assessing uncertainty in the values of unknown optical properties of the sample, obtained using a nonlinear least-squares fit from the spectra of the observed ellipsometric transfer quantities[2]. The implicit function theorem was adopted to describe the propagation of uncertainty for nonlinear least-squares calculations[3]. We would like to introduce the method and results of applying the developed uncertainty evaluation method to the uncertainty evaluation of the thin film layer thickness and complex dielectric constant of silicon nitride thin film sample obtained through SE.

#### References

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- [2] Y. J. Cho and W. Chegal, *Opt. Express* **29** 394428 (2021).
- [3] J. A. Fessler, *IEEE Trans. Image Processing* **5**, 493 (1996).

**NM-WeP-2 A New Tool for Single Ion Implantation and Nanoscale Materials Engineering: System Design and Source Development, *Paul Blenkinsopp*, Ionoptika Ltd., UK; *K. McHardy*, Ionoptika, Ltd., UK; *G. Aresta*, Ionoptika Ltd., UK**

Quantum computing is the next great frontier of science. It has the potential to revolutionize many aspects of modern technology, including digital communications, "quantum-safe" cryptography, and incredibly accurate time measurements.

Single impurity atoms in semiconductors are receiving attention as potential quantum technologies, and proof-of-concept devices have shown promise. However, such devices are incredibly challenging to manufacture, as single atoms must be placed within  $\sim 20$  nm of each other within a pure  $^{28}\text{Si}$  matrix.

All working devices thus far have been fabricated using hydrogen lithography with an STM followed by atomic layer deposition. This is labour-intensive and requires several days of meticulous preparation to create just a single quantum bit (qubit). Real-world devices will require arrays of hundreds or thousands of impurity atoms, highlighting the requirement for a scalable method of positioning single atoms with nanometer precision.

We report on a new commercial instrument for the fabrication of quantum materials and devices via single ion implantation.

The instrument features a high-resolution mass-filtered focused ion beam (FIB), a high-sensitivity deterministic implantation system, 6-inch wafer

handling, and a high-precision stage. The deterministic implantation system allows single ion implantation with confidence levels as high as 98%.

The ion dose delivered to the sample can be adjusted across a wide range, providing many nanoscale materials engineering capabilities in a single tool, from single ion implantation to direct-write capabilities such as isotopic enrichment and targeted ion-implantation of nanomaterials.

The liquid metal alloy ion sources, coupled with and mass filtered column will enable for the implantation of many different elements with isotopic resolution. Available sources include silicon, erbium, gold, and bismuth, while many others of technological interest are in development. We will report on the LMIG source development carried out at Ionoptika in collaboration with our partners.

**NM-WeP-3 Graphene-Incorporated Dielectric Composites by Varying the Mixing Method and Degree of Oxidation of Graphene, *S. Jun, Kwangsin John Ahn, S. Yu*, Hankuk University of Foreign Studies, Republic of Korea**  
Graphene-incorporated dielectric composites by varying the mixing method and degree of oxidation of graphene

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To obtain high-performance dielectric properties of dielectric composites, graphene and barium titanate ( $\text{BaTiO}_3$ , BTO) fillers were mixed in the polymer matrix, by varying the degree of reduction of graphene and the mixing method. Two kinds of graphene materials, i.e., graphene oxide (GO) and reduced graphene oxide (rGO); and two kinds of mixing methods for graphene and BTO, i.e., encapsulation of BTO in graphene and subsequent mixing, and simple mixing of BTO and graphene in the polymer matrix. This yielded to four kinds of graphene-incorporated samples, which was compared with the reference sample, i.e., a sample with only BTO fillers in the polymer matrix without graphene. Encapsulation process was performed by the self-assembly of positively functionalized BTO and negatively functionalized GO. After the encapsulation, GO-encapsulated BTO fillers were chemically reduced to become rGO-encapsulated BTO fillers [1,2]. All the four graphene-incorporated samples exhibited better dielectric constant values, i.e., 1.5 to 2.2 times higher than the reference sample. It was thought to be caused by the presence of graphene via the interfacial polarization or micro-capacitor effects [1]. However, the increase in dielectric loss, which had frequently occurred for the simple mixing cases of conducting nano-fillers, was successfully suppressed for the encapsulated samples. Here, the dielectric loss was controlled under lower 20%, which was caused by removal of free graphene platelets during the chemical process of the encapsulation [2]. The best performance one is the encapsulation method for rGO material. The next one is GO encapsulation case. The highest enhancement of 120% in the dielectric constant and moderate dielectric loss was obtained for rGO-encapsulation case. Therefore, we can say that encapsulation of BTO in graphene is very promising mixing method for high-performance polymer-based dielectric composite materials.

#### References:

- [1]S.-Y. Jun et al., *Carbon* **199**, 23 (2022).
- [2]S.-Y. Jun et al., *Mater. Chem. Phys.* **255**, 123533 (2020).

**NM-WeP-4 Deep Learning-Based Prediction of Adsorption Energies for  $\text{MoO}_2\text{Cl}_2$  Precursor on  $\text{SiO}_2$  Surface Using Density Functional Theory, *Do-Hyun Kwon, J. Lee*, Korea University of Technology and Education, Republic of Korea; *J. Kim*, Pohang University of Science and Technology (POSTECH), Republic of Korea; *Y. Kim*, Korea University of Technology and Education, Republic of Korea**

A search for appropriate adsorption geometries of a  $\text{MoO}_2\text{Cl}_2$  precursor on an -OH terminated  $\beta\text{-SiO}_2$  surface was conducted using a deep-learning approach. The adsorption geometries were configured by translating and rotating the precursor located near the surface. A deep-learning model was developed to learn important properties of the adsorption geometries; the model consists of three main units to learn positional features of the precursor atoms, to learn distance features among the precursor and surface atoms, and to predict the corresponding adsorption energy. A dataset of adsorption geometry and the adsorption energy pairs was collected by grid search. Using this dataset, the model was trained to predict the adsorption energies with minimal error and optimized to achieve a root mean square error (RMSE) of less than 0.011%. Energetically more favorable adsorption geometries were found using the trained model. By inputting arbitrary adsorption structures into the trained model and

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obtaining the predicted adsorption energies, recommendations for energetically stable adsorption structures were received. Through actual calculations, it was confirmed that the model could identify energetically stable adsorption structures that were not included in the dataset. The proposed method is applicable to finding appropriate adsorption geometries of other types of precursors and surfaces.

**NM-WeP-5 Induced Self-Assembly of Small (3 - 5 nm) Nanoparticles Into Flexible Nanofilms at Air- and Oil-Water Interfaces**, *H. Cameron, Y. Zhang, K. Leslie, B. Scott, I. Curtis, L. Gamble, M.-Vicki Meli*, Mount Allison University, Canada

Thin-film assemblies of gold nanoparticles (AuNPs) at the liquid interfaces have been an exciting research area over the past decade due to their promising applications in optics, sensors, shape-shifting and metamaterials, and catalysis. Autonomous self-assembly at such interfaces is an attractive method of synthesizing these superstructures, however this is a complex process that has proven to be difficult to control. Small AuNPs with core sizes <10 nm pose greater difficulties than larger AuNPs because they have a small free energy of adsorption that is comparable to their thermal energy. Therefore, there are few published examples of the self-assembly of such AuNPs at the liquid-liquid interface. The present research analyzes the effects of various experimental parameters on the adsorption of ~3 nm AuNPs to the oil-water interface, including ligand shell composition, alcohol additive structure and amount, oil phase composition and the phase of NP introduction. Film quality is qualitatively assessed and compared with a recent thermodynamic model to elucidate the primary driving forces of adsorption. Films with tunable optical density ranging from lightly coloured to gold and lustrous, are obtained with variation over a single parameter. Determining approaches that promote the adsorption of 3 nm AuNPs to an aqueous interface and furthering the understanding of its thermodynamic mechanism is necessary to gain precise control and to work towards the industrial use of such thin films.

**NM-WeP-6 Molecular Structure and Vapor Pressure of Molybdenum Pentachloride Using Ab-Initio Thermodynamics**, *N. Lee*, Korea University of Technology and Education, Republic of Korea; *S. Kim, J. Kim, Yeong-Cheol Kim*, Korea University of Technology and Education, Republic of Korea

This study aims to elucidate the molecular structure and vapor pressure of molybdenum pentachloride in the gas phase using ab-initio thermodynamics. Molybdenum pentachloride can exist as  $\text{MoCl}_5$  (monomer) and/or  $\text{Mo}_2\text{Cl}_{10}$  (dimer). The monomer is thermodynamically favored above 215 K, but Cl-Cl bond breakage is required to dissociate a dimer into two monomers. This suggests that the dimer is kinetically favorable even above 215 K, and the calculated dimer vapor pressure agrees well with the experimental data.

## Nano and 2D Materials

### Room Naupaka Salon 5 - Session NM1-ThM

#### Nanomaterials - Properties and Applications I

Moderator: Santosh KC, San Diego State University

8:40am **NM1-ThM-3 Different Directions In Layered Materials, Joshua Goldberger**, The Ohio State University **INVITED**

Layered and 2D materials are currently one of the most well-studied classes of solid-state compounds, due to the plethora of unique physical phenomena found in these anisotropic materials. Here we will describe our recent work in the synthesis, properties, and applications of layered materials that exhibit either n-type or p-type conduction behavior depending on the crystallographic direction, a phenomenon we refer to as "goniopolarity". We will establish the origin of this exotic behavior and the band structure design principles for identifying new goniopolar materials.<sup>[1]</sup> This has led to a significant expansion in the number of compounds that we have experimentally demonstrated to exhibit this effect, such as NaSn<sub>2</sub>As<sub>2</sub>, NaSnAs, WSi<sub>2</sub> and PdSe<sub>2</sub>.<sup>[2-5]</sup> Finally, considering that most modern electronic devices require the integration of p-type and n-type regions for functionality, we will show how the unique charge separation in goniopolar materials have the potential to create new efficient energy-harvesting and electronics technologies.

[1] Y. Wang; K. G. Koster; A. M. Ochs; M. R. Scudder; J. P. Heremans; W. Windl; J. E. Goldberger, *J. Am. Chem. Soc.* **142**, 2812-2822 (2020).

[2] B. He; Y. Wang; M. Q. Arguilla; N. D. Cultrara; M. R. Scudder; J. E. Goldberger; W. Windl; J. P. Heremans, *Nat. Mater.* **18**, 568-572 (2019).

[3] K. G. Koster; Z. Deng; C. E. Moore; J. P. Heremans; W. Windl; J. E. Goldberger, *Chemistry of Materials* **35**, 4228-4234 (2023).

[4] R. A. Nelson; Z. Deng; A. M. Ochs; K. G. Koster; C. T. Irvine; J. P. Heremans; W. Windl; J. E. Goldberger, *Mater. Horiz.* **10**, 3740-3748 (2023).

[5] A. M. Ochs; P. Gorai; Y. Wang; M. R. Scudder; K. Koster; C. E. Moore; V. Stevanovic; J. P. Heremans; W. Windl; E. S. Toberer; J. E. Goldberger, *Chem. Mater.* **33**, 946-951(2021).

9:20am **NM1-ThM-5 Evaluation of Vapor Pressure of MoO<sub>2</sub>Cl<sub>2</sub> and Its Initial Chemical Reaction on a SiO<sub>2</sub> Surface by Ab Initio Thermodynamics, H. Kim, N. Lee, Yeong-Cheol Kim**, Korea University of Technology and Education, Republic of Korea

The vapor pressure of MoO<sub>2</sub>Cl<sub>2</sub> and its initial chemical reaction on a SiO<sub>2</sub> surface were evaluated and analyzed using *ab initio* thermodynamics. The vapor pressure of MoO<sub>2</sub>Cl<sub>2</sub> was calculated using the Gibbs free energy, while considering the zero-point energy, temperature-dependent enthalpy change, and entropy. The initial surface reaction was also studied as a function of temperature and partial pressure. The calculated sublimation temperature of MoO<sub>2</sub>Cl<sub>2</sub> was 410 K, and its vapor pressure at 350 K was 8.2 torr. The partial pressure calculated for MoO<sub>2</sub>Cl<sub>2</sub> agreed reasonably well with the experimentally measured value. The surface reaction energy barrier between MoO<sub>2</sub>Cl<sub>2</sub> and SiO<sub>2</sub> was 0.8 eV.

9:40am **NM1-ThM-6 Development of TiAl Alloys: A Future Light-Weight Material for Extreme Condition, Seong-Woong Kim**, Korea Institute of Materials Science, Republic of Korea

Research on developing new TiAl alloys for high temperature (> 900°C) applications is introduced. TiAl alloys have been gained interest for automobile and aerospace applications due to their low density, good oxidation resistance and high temperature strength. However, lack of room temperature ductility and the limitation of operating temperature hindered the practical applications of TiAl alloys. At KIMS, we have developed new TiAl alloys which have excellent room temperature and high temperature properties. Especially, the alloy #16 showed excellent oxidation resistance in the temperature range from 900 to 1000°C by forming stable Al<sub>2</sub>O<sub>3</sub> oxidation layer. Moreover, alloy #16 exhibit room temperature ductility up to 0.78% solely by casting.

In addition, we introduced some in-situ transmission electron microscopy experiments in order to understand an underlying mechanism on room temperature ductility of TiAl alloys. Also, molecular dynamics simulation was conducted to calculate the stacking fault energy of TiAl alloys and to show which deformation mode is dominant. The difference in deformation mode was explained by stacking fault energy of the TiAl alloys which was calculated by molecular dynamics. Furthermore, the role of lamellar orientation of tensile direction on deformation behavior was examined using Schmid factor of each orientation. Finally, we proposed the important microstructural factors to have room temperature ductility of TiAl alloys.

## Nano and 2D Materials

### Room Naupaka Salon 5 - Session NM2-ThM

#### Nanomaterials - Properties and Applications II

Moderator: Yu-Chuan Lin, National Yang Ming Chiao Tung University (NYCU)

10:20am **NM2-ThM-8 2D Metal Carbides (MXenes) for Catalysis, Yue Wu**, Iowa State University

MXenes, a new group of 2D transition metal carbides, are of considerable interest in catalysis due to their rich surface chemistry, tunable electronic structures, and thermal stability. In this presentation, we will discuss our latest discovery on utilization of metal-support interactions between noble metal and 2D metal carbides to enhance the catalytic reactions, such as alkane dehydrogenation, methane coupling, and electrochemical catalytic reactions. In addition, the nature of active sites in the MXene-based catalysts will be discussed.

10:40am **NM2-ThM-9 Investigating 2D-Materials Using Correlative Spectroscopy & Microscopy, James Lallo, L. Ping, T. Nunney, P. Mack, R. Simpson, H. Tseng**, Thermo Fisher Scientific

Across a wide range of application areas, understanding the chemistry and structure of surfaces and interfaces is crucial. In the last fifty years, X-ray photoelectron spectroscopy (XPS) has become established as one of the key techniques for measuring surface and interface chemistry, and advances in instrumentation have enabled it to keep pace with the requirements for both academia and industry. XPS can deliver quantified surface chemistry measurements, and by using depth profiling, an understanding of layer and interfacial chemistry, but the limit on spatial resolution for XPS can prevent it from determining how the surface structure is related to the measured chemical properties. For example, how the changing morphology of the surface during a depth profile could influence the measured composition would be challenging to determine using just XPS.

Other experimental techniques which are unable to match the surface selectivity of XPS are able to provide complementary information to extend the data from XPS. Electron microscopy can provide high resolution imaging, with elemental composition provided by energy dispersive X-ray microanalysis, but without the same surface selectivity seen with XPS or Auger electron spectroscopy (AES). This can be a perfect complement to XPS analysis, so long as the same points of interest can be identified. Molecular spectroscopy, such as FTIR or Raman, can also provide complementary information to XPS, albeit with different sampling depths, which can be extremely useful to validate measurements or confirm particular molecular structures using the wide range of spectral libraries available for those techniques.

In this presentation, we will describe how a correlative approach using both surface analysis instrumentation and scanning electron microscopy can be used to characterize 2D nanomaterials. Samples of MoS<sub>2</sub> grown on Si substrates have been investigated using XPS, Raman and SEM to determine their composition and structure. To facilitate co-alignment of the analysis positions when moving between the instruments, special sample carriers and software alignment routines have been developed.

11:00am **NM2-ThM-10 Electronic, and Optical Properties of 2D Metal Chalcogenophosphates, H. Chiu, Santosh KC**, San Diego State University

Recently, there have been significant research activities on two-dimensional (2D) materials for their potential use in electronic and optical devices. The structure, electronic, and optical properties of 2D metal chalcogenophosphates are investigated based on the Density Functional Theory. The lattice dynamics, surface, and interface properties with metal and oxides are investigated. Moreover, the electronic properties are tuned with substitutional impurities for metals and chalcogens. Our results indicate that the electronic and optical properties of metal chalcogenophosphates exhibit them as an emerging candidate material for devices.

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