

# Sunday Afternoon, January 13, 2019

PCSI

Room Ballroom South - Session PCSI-SuA

## The Future of Computing

**Moderator:** Christopher Palmstrøm, University of California, Santa Barbara

3:00pm **PCSI-SuA-1 Chiraltronics: Chiral Domain Wall and Anti-skyrmion Data Storage Elements for High Performance Racetrack Memories, Stuart Parkin**, Max Planck Institute, Germany **INVITED**

Over the past few years there have been remarkable discoveries in spin-based phenomena that rely on spin-orbit coupling that could spur the development of advanced magnetic memory devices<sup>1-3</sup>. One of the most exciting of these is Racetrack Memory<sup>1,4</sup> that stores digital data in the form of the presence or absence of chiral domain walls. Recent discoveries in the emerging field of chiraltronics makes possible the current induced motion of series of chiral domain walls at high speeds that exceed 1 km/s in atomically engineered racetracks<sup>2</sup>. The same type of Dzyaloshinskii-Moriya exchange interactions that stabilize chiral Néel domain walls in such racetracks also enable the formation of topological spin textures such as spin helices and skyrmions. Recently we have discovered magnetic antiskyrmions in a tetragonal Heusler compound, using Lorentz transmission electron microscopy<sup>5</sup>. Such antiskyrmions are potential storage elements in racetrack memories. These recent discoveries in chiraltronics makes Racetrack Memory extremely attractive as a dense, high performance, low energy consuming, non-volatile memory technology<sup>1</sup> that could make computing systems more compact and yet more powerful.

1. Parkin, S. S. P. & Yang, S.-H. Memory on the Racetrack. *Nat. Nano.* **10**, 195-198, (2015).
2. Yang, S.-H., Ryu, K.-S. & Parkin, S. S. P. Domain-wall velocities of up to 750 ms<sup>-1</sup> driven by exchange-coupling torque in synthetic antiferromagnets. *Nat. Nano.* **10**, 221-226, (2015).
3. Ryu, K.-S., Thomas, L., Yang, S.-H. & Parkin, S. S. P. Chiral spin torque at magnetic domain walls. *Nat. Nano.* **8**, 527–533, (2013).
4. Parkin, S. S. P., Hayashi, M. & Thomas, L. Magnetic Domain-Wall Racetrack Memory. *Science* **320**, 190-194, (2008).
5. Nayak, A. K. et al. Magnetic antiskyrmions above room temperature in tetragonal Heusler materials. *Nature* **548**, 561-566, (2017).

3:40pm **PCSI-SuA-9 Neuromorphic Computing, Angel Yanguas-Gil**, Argonne National Laboratory **INVITED**

4:20pm **PCSI-SuA-17 Panel Discussion I,**

## PCSI

### Room Ballroom South - Session PCSI-SuE

#### III-V Growth

Moderator: Lincoln J. Lauhon, Northwestern University

7:30pm **PCSI-SuE-1 Understanding the Kinetics of III-V Semiconductor Nanowire Growth using *in-situ* TEM**, *C Maliakkal, D Jacobsson, M Tornberg, A Persson, J Johansson, R Wallenberg, Kimberly Thelander*, Lund University, Sweden

INVITED

Semiconductor nanowires have emerged as a highly promising technology for next-generation electronics and photonics, in particular due to their potential for forming novel metastable crystal phases, complex alloys and heterostructure combinations not achievable in bulk semiconductors. The most common method of fabricating these structures is the vapor-liquid-solid mechanism, which makes use of a catalytic liquid metal droplet. The development of these materials requires a fundamental understanding of how they form. Since nanowire growth is performed in a vapor phase atmosphere at high temperature, the dynamic processes controlling their formation cannot be directly deduced by analyzing only the final grown nanostructure. As such, the current mechanistic understanding of the synthesis process is insufficient for achieving the promised level of control.

In order to address this challenge, we use *in-situ* TEM imaging combined with *in-situ* compositional analysis to study the processes occurring at the interface between the metal droplet and nanowire, and how these control the final structure. Nanowires are grown in a Hitachi HF3300S aberration-corrected environmental TEM connected to a chemical vapor deposition system designed for III-V semiconductor growth. Growth is performed on a SiNx-based MEMS heating chip mounted on a holder with two separate microtubes for supplying the precursor gases. High resolution, high-frame-rate videos enable us to determine the rate at which individual semiconductor bilayers form, along with the interface morphology, catalyst geometry and nanowire crystal structure. The elemental composition of the catalyst is measured by energy dispersive X-ray spectroscopy as a function of the growth parameters. We identify different 'regimes' in which growth occurs that can be identified by the composition and structure of the catalyst, and in which different steps in the growth process control the overall formation rate as well as the structure of the material.

8:00pm **PCSI-SuE-7 *In situ* Studies of Surface Morphological Evolution During Indium Nitride Growth by Atomic Layer Epitaxy**, *Charles R. Eddy, Jr., N Nepal*, U.S. Naval Research Laboratory; *S Rosenberg*, American Association for Engineering Education; *V Anderson*, Sotera Defense Solutions; *J Woodward*, American Society for Engineering Education; *C Wagenbach*, Boston University; *A Kozen*, American Society for Engineering Education; *Z Robinson*, College at Brockport SUNY; *L Nyakiti*, Texas A&M University; *S Qadri*, U.S. Naval Research Laboratory; *M Mehl*, U.S. Naval Academy; *K Ludwig*, Boston University; *J Hite*, U.S. Naval Research Laboratory

Nitride semiconductors have had significant commercial success, but full bandgap engineering of these materials is prohibited by the high temperatures used in conventional growth methods. Recently, we have developed a growth method – low temperature atomic layer epitaxy (ALEP) – that has empirically produced crystalline semiconductor films with properties comparable to those from conventional growth processes, but at roughly half the growth temperature [1,2]. This has eliminated miscibility gaps in ternary III-N semiconductor films and enabled the realization of full bandgap engineering from 0.7 eV to 6.1 eV.

Despite these empirical successes, the fundamental mechanisms involved in ALEP are unknown and the full promise of the method unrealized. To obtain such enabling knowledge we have employed synchrotron-based grazing incidence small angle x-ray scattering (GISAXS) to study the low temperature atomic level processing (ALP) of GaN substrate surfaces for epitaxy and ALEP nucleation and growth of InN on said surfaces. GISAXS allows real-time, *in situ* monitoring of the surface morphology during these processes.

In this presentation, we will introduce the GISAXS method and the apparatus we have developed to conduct *in situ* GISAXS measurements of the aforementioned ALP and ALEP processes. We monitor the evolution of GaN substrate surface morphology during a series of low-temperature ALPs including: gallium flash off (GFO), hydrogen plasma clean, and nitrogen plasma nitridation. We learn that the optimum surface results from a GFO conducted at 500° C for only 10 cycles followed by a hydrogen plasma clean. Further, we learn that conventional plasma nitridations are detrimental to smooth surface evolution. When employed to study ALEP

InN nucleation and growth, GISAXS data, coupled with Porod[3] and 2D Fourier Transform analysis, affords a clear picture that the growth proceeds by island nucleation and growth and not by the conventionally accepted layer-by-layer growth associated with atomic layer deposition. We have monitored the evolution of island nucleation density, island spacing, island shape and island size as a function of key ALEP growth parameters. We observe that the islands are generally tens of nm or less in size and evolve from a spherical mound shape to a cylindrical shape. Finally, we will present the variations between 2D and 3D growth modes with growth parameter variations that provide insights on process modifications to promote higher quality electronic materials growth.

1. Nepal et al., Cryst. Growth and Des., 13, 1485 (2013)
2. Nepal et al., Thin Solid Films 589, 47 (2015).
3. G. Porod, Kolloid Z., 124, 83 (1951).

8:05pm **PCSI-SuE-8 Growth Strategies for Modifying Heterovalent Interfaces**, *Kirstin Alberi, K Park*, National Renewable Energy Laboratory  
Integration of dissimilar semiconductor materials is becoming an increasingly important pathway for pushing the boundaries of device performance by expanding the options for material selection. The major challenge that must be overcome is the formation of interfaces between two materials with different valences. Charge imbalances arising at interfaces between conventional semiconductors are often alleviated through re-distribution of atoms, which can lead to intermixing-induced degradation of the adjoining layers.

Here, we show that a combination of elemental treatments combined with UV photon exposure can be used to tailor the properties of model ZnSe/GaAs interfaces formed by molecular beam epitaxy. X-ray diffraction and photoluminescence were used to assess the interface roughness and degree of intermixing. Treatment of an As-terminated GaAs surface with UV light and a Se flux results in both an abrupt interface and passivation of the underlying GaAs epilayer. We propose that this improvement over interfaces grown under dark conditions and treatment with a Zn flux is triggered by light-induced desorption of As atoms followed by enhanced Ga-Se bond formation [1]. Thus, the combination of light and elemental treatment during interface initiation offers a highly tunable approach to significantly alter interface properties.

8:10pm **PCSI-SuE-9 InAs QD Formation on GaAs(110) by Bi-surfactants**, *W Martyanov*, Technische Universität Berlin, Germany; *R Lewis*, Paul-Drude-Institut für Festkörperelektronik, Germany; *H Janssen, P Farin, R Zielinski, C Schulze, A Lenz*, Technische Universität Berlin, Germany; *L Geelhaar*, Paul-Drude-Institut für Festkörperelektronik, Germany; *Holger Eisele*, Technische Universität Berlin, Germany

8:15pm **PCSI-SuE-10 Total Tomography of Nonplanar III-As Heterostructures**, *L Lähnemann*, 1Paul-Drude-Institut für Festkörperelektronik, Leibniz-Institut im Forschungsverbund Berlin e.V., Germany; *M Hill*, Northwestern University; *J Herranz*, 1Paul-Drude-Institut für Festkörperelektronik, Leibniz-Institut im Forschungsverbund Berlin e.V., Germany; *O Marquardt*, Weierstraß-Institut für Angewandte Analysis und Stochastik, Germany; *A Al Hassan, A Davtyan*, Naturwissenschaftlich-Technische Fakultät der Universität Siegen, Germany; *O Hruszkewycz, M Holt*, Argonne National Laboratory; *C Huang*, Northwestern University; *U Jahn*, Paul-Drude-Institut für Festkörperelektronik, Leibniz-Institut im Forschungsverbund Berlin e.V., Germany; *U Pietsch*, Naturwissenschaftlich-Technische Fakultät der Universität Siegen, Germany; **Lincoln J. Lauhon**, Northwestern University; *L Geelhaar*, Paul-Drude-Institut für Festkörperelektronik, Germany

Nanowire core-shell heterostructures are promising candidates for developing on-chip photonic light sources. Here we investigate the emission characteristics of InGaAs quantum wells grown on GaAs nanowires and the dependence on nanowire polytype, indium mole fraction, quantum well morphology (width), and strain. We have measured each of these structural characteristics through correlated atom probe tomography and x-ray nanodiffraction. Correlated cathodoluminescence was used to map local optical properties, providing a comparison with a model of emission built from the atom probe and x-ray analyses. We find that both composition and polytype play a role in explaining differences in emission between zinc blende and wurtzite regions.

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# Sunday Evening, January 13, 2019

8:20pm PCSI-SuE-11 Superconducting Proximity Effect in Two-Dimensional Semiconductor-Superconductor Structures, *Javad Shabani*, New York University **INVITED**

Progress in the emergent field of topological superconductivity relies on synthesis of new material combining superconductivity, low density, and spin-orbit coupling. Theory indicates that the interface between a one-dimensional semiconductor with strong SOC and a superconductor hosts Majorana-modes with nontrivial topological properties. We present our recent developments in materials synthesis and growth of density-controlled surface 2D InAs quantum wells with epitaxial superconducting Aluminum. These developments have led to unprecedented control over proximity effect in semiconductors where electron densities can be tuned using a gate voltage. We demonstrate Josephson junctions with IV characteristic indicating highly transparent contacts. We focus on multiterminal junctions and superconducting quantum interference device (SQUID) geometry to investigate the subtle interplay between supercurrents and spin orbit interaction in these materials. The amplitude and shape of current phase plot is varied as critical currents in each arm are independently controlled using gate-tunable junctions. The data can be well described using solution of Bogoliubov–de Gennes equation for our SQUID junctions. We discuss potential applications in gate-based qubits as well as exploring topological superconductivity for computation.

## PCSI

### Room Ballroom South - Session PCSI-1MoM

#### 2D Plasmonics

**Moderator:** Anders Mikkelsen, Lund University, Sweden

8:30am **PCSI-1MoM-1 Controlling Light at the Atomic Scale with 2D Polaritons**, *Javier García de Abajo*, ICFO-Institut de Ciències Fotoniques, Av. Carl F. Gauss 3, 08860 Castelldefels (Barcelona), Spain **INVITED**

Two-dimensional materials have been recently shown to host robust polaritonic modes, ranging from plasmons in highly doped graphene to excitons in transition metal dichalcogenides. The electromagnetic behavior of these modes can be well understood in terms of an effective surface conductivity, in which we can capture their strong dependence on temperature and external static electric and magnetic fields. In this talk, I will overview the general characteristics of the optical response of these materials, which we can understand in terms of simple theoretical descriptions. We will also cover more sophisticated descriptions, aiming at exploring genuinely quantum-mechanical effects. We will further overview recent advances in ultrafast optical response and nonlinear optics, as well as the potential application of these materials for quantum-optics and optical sensing.

9:05am **PCSI-1MoM-8 An Optical transformer-based Campanile Near-field Probe on an AFM Cantilever**, *K Le*, aBeam Technologies; *S Bilent*, Lawrence Berkeley National Lab; *C Pina-Hernandez*, aBeam Technologies; *S Cabrini*, Lawrence Berkeley National Lab; *Keiko Munechika*, aBeam Technologies

Near-field scanning optical microscopy (NSOM) is a powerful and unique approach to characterize the chemical, physical and biochemical properties of materials with the nanometer scale resolution in real-time. NSOM has so far played only a niche role as characterization technique due to one main bottleneck: The need of reliable, efficient, and broadband near-field probes. An optical transformer-based Campanile near-field probes are a novel class of nano-optical tips, which proved to operate extremely reliably, can be employed by non-experts, are completely independent of the substrate type, and combine superior performance in terms of spatial resolution, near-field enhancement, bandwidth, and signal to noise ratio. Since the invention of the probes, the tips have been successfully used for multidimensional spectroscopic imaging of nanostructures with nanoscale resolution, providing so far in-accessible insights into optoelectronic process. The original probes are fabricated on top of an optical fiber. Our aim is to develop the fabrication technology to incorporate the Campanile optical transformer into a standard atomic force microscopy (AFM) platform as a novel class of NSOM probes.

We report a realization of Campanile near-field scanning optical probe on an AFM platform. The fabrication process was based on standard micro-lithography techniques allowing a straightforward production of multiple cantilevers on a wafer scale. Figure 1 shows a scanning electron micrograph (SEM) image of an optical transformer on a cantilever. The measured resonance frequencies of the AFM cantilevers are in excellent agreement with theoretical calculations over different cantilevers as shown in Figure 2, which justifies our characterization. This work paves the way for low cost and reproducible manufacturing of near-field probes suitable for high-resolution hyperspectral imaging.

9:10am **PCSI-1MoM-9 Localized Surface Curvature Artifacts in Gap-mode Tip-enhanced Nanospectroscopy**, *Darya Stepanichsheva*, Tomsk Polytechnic University, Russia

Tip-enhanced Raman spectroscopy (TERS) allows for chemical analysis to exceed the limit of light diffraction and to reach nanoscale spatial resolution. The high-spatial sensitivity is provided by the metal substrate in the so-called gap-mode TERS. However, in this case, the connection between the tip and the sample could lead to distortions in the image of the nanostructure during visualization. The purpose of this work is to provide a generalized view of such image artifacts in TERS imaging and to find out whether these effects occur and to what extent. We used ultrathin molecular layers and self-assembly monolayers as Raman-active probes deposited on Au and Si films and Au nanostructures. In addition to the 6-fold amplification of the Raman signal, we found that the sample curvature in gap-mode induces imaging artifacts visible as distortions in the electromagnetic field distribution.

Our results show that the use of gap-mode significantly increases the signal strength, but that at the same time, the sample curvature makes an impact to the TERS image contrast which was not considered until now. Beyond metal nanoparticles functionalized with organic molecules, our conclusions

impact the nanoscale chemical visualization of molecular and inorganic systems using vibrational spectroscopy.

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## PCSI

### Room Ballroom South - Session PCSI-2MoM

#### Atomic Layer Deposition & Etching I

**Moderator:** Anders Mikkelsen, Lund University, Sweden

9:15am **PCSI-2MoM-10 How Chemistry Drives Microstructure: Probing the Structure of sub-nm ALD Materials using *in-situ* FTIR and Synchrotron Techniques**, *Angel Yanguas-Gil*, Argonne National Laboratory **INVITED**

Atomic layer deposition is a thin film technique that is well known for its ability to coat high surface area materials.[1] However, it is also an ideal model system to understand how surface interactions drive the growth and evolution of microstructure of materials at the nanoscale: processes based on self-limited precursor-surface interactions are ideally suited for multimodal characterization, allowing the design of experiments and reactors that are optimized for specific characterization techniques.

To complement *in-situ* techniques such as quartz crystal microbalance, FTIR, or XPS, which focus primarily on the surface chemistry and electronic properties of materials, we have developed a modular *in-situ* reactor at the Advanced Photon Source that gives us access to multiple techniques that can provide detailed microstructural information, including X-ray scattering, XAFS, and PDF. [2]

In this talk I will show how we can apply these techniques to understand the interplay between surface chemistry and microstructure. I will focus on two examples: the synthesis of materials with a controlled distribution of dopants, and the evolution of microstructure from isolated cations to a bulk-like structure. The primary target of our research has been oxide-based materials such as ZnO, In<sub>2</sub>O<sub>3</sub>, MgO, HfO<sub>2</sub>, and nanolaminates.[2-4] However, recently we have expanded to other materials systems, including hybrid organic/inorganic systems and 2D materials such as MoS<sub>2</sub>. [5]

Overall, the development of new *in-situ* characterization tools at synchrotron radiation facilities worldwide is going to be an enabling capability that will help us understand the driving forces behind the emergence of crystallinity and the control of microstructure at low temperatures.

[1] A. Yanguas-Gil, *Reactive transport in nanostructured materials*, (Springer, New York, 2017).

[2] J. Klug et al, *Rev. Sci. Instr.* **86**, 113901 (2015)

[3] A. Yanguas-Gil et al, *Chem. Mater.* **23**, 4295 (2011)

[4] A. Yanguas-Gil et al, *Chem. Mater.* **25**, 4849 (2013)

[5] S. Letourneau et al, *ACS Appl. Nano Mater.* **1**, 4028 (2018)

9:45am **PCSI-2MoM-16 Mechanism of Hydrogen Plasma Modified ALD Growth of Metal-enriched Oxides Studied by *In-Situ* Mass Spectrometry**, *Thomas Larrabee*, S Prokes, Naval Research Laboratory

Modifying atomic layer deposition (ALD) growth by inserting argon/hydrogen plasma steps has been shown to alter the stoichiometry of oxides, driving metal-enriched films and lower oxidation states. Altering oxide stoichiometry is useful for changing the electrical conductivity, optical properties, bandgaps, and other device characteristics; often inaccessible from typical ALD growth. It has been unclear what the mechanism of the plasma-surface reactions and modifications to standard chemical (or "thermal") ALD growth has been in these processes, however. To elucidate this mechanism, a quadrupole mass spectrometer has been used to examine both reactant and product gas species during the ALD and plasma-modified ALD reactions of ZnO growth from diethyl zinc (DEZ), water, and H<sub>2</sub>/Ar plasmas.

Comparison was made between thermal reactions, inserting H<sub>2</sub>/Ar plasma after the metal precursor step, after the oxidant step, between sequential metal precursor steps before the oxidant step, and of argon-only plasma between sequential metal steps. Evidence for a mechanism involving metal-organic ligand desorption and further reaction with the plasma upon H<sub>2</sub>/Ar plasma exposure is to be presented. No evidence demonstrating removal of oxygen atoms from the growing film, either by O<sub>2</sub> or H<sub>2</sub>O gas evolution was found. Zinc metal enriching in ZnO growth occurs when the H<sub>2</sub>/Ar plasma is inserted between sequential DEZ steps before the water steps. When the ethyl-terminated surface (after DEZ reaction) is exposed to plasma, primarily a small amount of methane (M/Z = 16) is produced, with

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a trace of ethane ( $M/Z = 28$ ). This is due to ethane desorption and fast reaction with the large excess of hydrogen radicals from the plasma, cracking ethane to methane. After desorption of ethyl ligands, sites open up for additional adsorption of Zn in the second DEZ step. Plasmas containing hydrogen are required, as no ethane nor methane was observed with argon-only plasma.

To verify the generality of this mechanism for plasma-modified ALD using hydrogen plasmas to metal enrich ALD-grown oxides, additional modified oxide materials will also be presented (*i.e.*  $Nb_2O_5$  to  $NbO_2$ , etc.).

9:50am **PCSI-2MoM-17 *In Situ* Investigation of Doping of 2D Semiconductors During Atomic Layer Deposition of Dielectrics**, Michael Moody, J Shang, J Chen, A Henning, T Lohr, T Marks, L Lauhon, Northwestern University

The sensitivity of ultrathin and 2-dimensional (2D) semiconductors to the surrounding environment provides a key opportunity for control of material and device behavior. Especially as substitutional dopants may be difficult to control and lead to increased scattering, adlayers are a promising approach to tuning the Fermi level in 2D semiconductors [1]. Despite the growing body of results using oxide dielectrics to this end, there is a lack of mechanistic investigation and understanding of scope and limitations.

In this talk, we build on results using atomic layer deposition of a tunable oxide to dope  $MoS_2$  [2], and investigate mechanisms of growth and doping via *in situ* electrical measurements. Using a modified atomic layer deposition (ALD) reactor, we can measure field-effect transistors during dielectric growth at temperatures up to 300°C. As well as being more efficient than *ex situ* measurements for some studies (e.g. carrier concentration vs. thickness [3]), it enables otherwise-impossible observation of dynamics and changes with each half-cycle of deposition. We are therefore also positioned to learn about growth and reactivity. The first and to date only other such *in situ* electrical measurements identified that physisorption of ozone promotes dielectric growth on graphene [4]. We further observe reversible adsorption of metal-organics for nucleation of dielectrics on  $MoS_2$ . Still, while physisorptive nucleation of ALD is not unique to graphene, neither is it universal to van der Waals materials. Even moderately air-stable transition metal dichalcogenides such as  $MoTe_2$  can differ notably in reactivity, growth mechanism, and thus semiconductor-dielectric interface. As such, *in situ* measurements are a powerful tool to understand growth on 2D materials.

[1] X. Zhang, Z. Shao, X. Zhang, Y. He, J. Jie, *Adv. Mater.* **28**, 10409 (2016).

[2] M. J. Moody, A. Henning, T. Jurca, J. Y. Shang, H. Bergeron, I. Balla, J. N. Olding, E. A. Weiss, M. C. Hersam, T. L. Lohr, T. J. Marks, L. J. Lauhon, *Chem. Mater.* **30**, 3628 (2018).

[3] W. Luo, M. Zhu, G. Peng, X. Zheng, F. Miao, S. Bai, X.-A. Zhang, S. Qin, *Adv. Funct. Mater.* **28**, 1704539 (2018).

[4] S. Jandhyala, G. Mordji, B. Lee, G. Lee, C. Floresca, P.-R. Cha, J. Ahn, R. M. Wallace, Y. J. Chabal, M. J. Kim, L. Colombo, K. Cho, J. Kim, *ACS Nano*. **6**, 2722 (2012)

9:55am **PCSI-2MoM-18 The Impact of the Annealing Temperature of the Seed Layer on the Growth and the Electrical Properties of the Main Layer in Atomic Layer Deposition of  $SrTiO_3$  Films**, Sang Hyeon Kim, Seoul National University, Republic of Korea; W Lee, Northwestern University; C An, D Kwon, D Kim, S Cha, S Cho, C Hwang, Seoul National University, Republic of Korea

The atomic layer deposition of  $SrTiO_3$  (STO) films was studied with  $Sr(Pr_3Cp)_2$  and  $Ti(CpMe_3)(OMe)_3$  (Pr, Cp, and Me are propyl, cyclopentadienyl, and methyl groups, respectively) on Ru and Si substrates at 370 °C. The second STO layer (main layer) was grown on the annealed first 3-5 nm-thick STO layer (seed layer) to induce the in-situ crystallization. The electrical properties and the growth behavior of the main layer were studied with the variations in the seed layer condition, which was varied by varying the annealing temperature of the seed layer in the range of 450-650 °C. The STO films were remained in amorphous by annealing below 500 °C. They were started to be crystallized at 550-575 °C, and well crystallized at 600 °C or above. The growth rate of the main layer on the crystallized seed layer (0.15 nm/cycle) was 50 % higher than that on the amorphous seed layer (0.10 nm/cycle). As a result, the root-mean-square roughness of the main layer increased from 0.5 nm on the amorphous seed layer to 2.1 nm on the mixed phase (amorphous and crystalline) seed layer, where the main layer growth on the crystallized portion was higher than that on remaining amorphous portion. When the seed layer was well crystallized, the main layer roughness decreased again to 1.0 nm by the uniformly high

growth rate across the entire surface. The dielectric constants of the main layers increased from 20 to 100 when the annealing temperature of the seed layer increased from 450 to 650 °C due to the improvement of the film crystallinity.

## PCSI

### Room Ballroom South - Session PCSI-3MoM

#### Topological Materials

Moderator: Javad Shabani, New York University

11:00am **PCSI-3MoM-31 Topological Heterostructures by Molecular Beam Epitaxy**, Susanne Stemmer, University of California, Santa Barbara **INVITED** Topology, both in real space and in reciprocal space, has emerged as a new design principle for materials that can host a wealth of novel properties. Interfaces and heterostructures with topological materials offer opportunities to control and manipulate their electronic states and associated phenomena, for example, via electric field effect, strain, or symmetry breaking. In this presentation, we will discuss recent progress in the growth of thin films of the three-dimensional Dirac semimetal  $Cd_3As_2$  by molecular beam epitaxy. We show that high-mobility, epitaxial  $Cd_3As_2$  films can be grown and discuss some of the phenomena that can be observed, such as an unusually large negative longitudinal magnetoresistance under parallel electric and magnetic fields. The heterostructures allow for experimental tests of theoretically predicted transitions between topological states by manipulating parameters, such as confinement and film strain. For example, as the film thickness is reduced, a band gap opens in the bulk Dirac electronic states and we observe a quantum Hall effect. Using electric field gating and Landau level spectroscopy, we demonstrate the Dirac dispersion of these two-dimensional states.

11:30am **PCSI-3MoM-37 Structural Distortions and Surface/Bulk Competition in Quasi-2D  $SnSe-TiSe_2$  Nanolayered Heterostructures**, Sage Bauers, National Renewable Energy Laboratory; D Hamann, D Merrill, J Ditto, M Esters, University of Oregon; D Roberts, University of Colorado at Boulder; A Zakutayev, National Renewable Energy Laboratory; D Johnson, University of Oregon

Low dimensional materials and their heterostructures are becoming increasingly important due to their emergent properties not observed in bulk analogues. As the fraction of surface or interface atoms become comparable to the bulk, so too do their contributions to the free energy. Since the most stable interface structure does not necessarily agree with the most stable bulk structure, competition between structures can cause size-dependent distortions or phase transitions. These structural behaviors can fundamentally alter the electronic environment, significantly affecting the material properties.

I will present on multilayer heterostructures formed by self-assembly of designed precursors. This will focus on a system containing quasi-2D layers of  $SnSe$  interleaved with  $TiSe_2$ , which exhibits size-dependent structures and transport properties. When a single bilayer of  $SnSe$  is present all atoms reside at a  $TiSe$  interface, templating is observed and long-range 3-dimensional order is established in the heterostructure.[1] However, as the number of  $SnSe$  layers increases and “bulk” atoms are introduced, distortions, additional structures, and new stacking sequences are observed.[2] These findings are corroborated by DFT calculations. The changing  $SnSe$  structure has profound effects on the composite transport properties with a shift in carrier type, as shown by Seebeck coefficient and Hall measurements.

This designed precursor approach has also been used to study thermoelectric enhancement[3] and quantum phase transitions[4] as a function of layer thickness in other selenides. Work extending this approach to sulfides is currently underway at NREL, and I will briefly present our initial results synthesizing similar heterostructures via combinatorial sputter deposition.

[1] D. M. Hamann, *et al*, *Inorg. Chem.* 2017, *56*, DOI 10.1021/acs.inorgchem.6b03063.

[2] D. M. Hamann, *et al*, *ACS Nano* 2018, 12, DOI 10.1021/acsnano.7b07506.

[3] S. R. Bauers, *et al*, *J. Mater. Chem. C* 2015, 3, 10451.

[4] M. Falmbigl, *et al*, *Nano Lett.* 2015, 15, 943.

11:35am **PCSI-3MoM-38 Gold-gold Dimer Buckling and Electronic Structure of Epitaxial LaAuSb Films.**, *Patrick Strohbeen, D Du, C Zhang, E Shourov*, University of Wisconsin-Madison; *F Rodolakis, J McChesney*, Argonne National Laboratory; *P Voyles, J Kawasaki*, University of Wisconsin-Madison

Recently, a new subset of hexagonal Heuslers with composition  $L_n\text{AuSb}$  ( $L_n = \text{La-Nd, Sm}$ ) was predicted to exist and shown to be stable in bulk polycrystalline samples<sup>1</sup>. These compounds contain 19 electrons per formula unit, typically thought to be unstable through traditional electron counting rules, however these compounds have been predicted to be stabilized through by highly localizing the 19<sup>th</sup> electron in Au-Au dimer bonds. Of these new phases, LaAuSb was also predicted through bandstructure calculations to be a Topological Dirac Semimetal (TDS). Here, we will (1) investigate this unique crystal structure and (2) study the electronic properties and structure of single phase epitaxial LaAuSb thin films.

In the original study, LaAuSb was grown in a bulk crystal, however it was both polycrystalline and full of impurity phases and thus it did not lend itself well to study. Here we use molecular beam epitaxy (MBE) to grow single crystalline thin films on c-plane sapphire to investigate the proposed “dimerized” structure. Growth was monitored in-situ by reflection high energy electron diffraction (RHEED) in which the resultant film patterns were streaky and well-resolved Kikuchi lines were observed (Fig. 1a,b), indicating the well-ordered and relatively flat surface of the film. Strong RHEED oscillations were observed throughout growth as well as Kiessig fringes (Fig. 1d,e) which is also consistent with a layer-by-layer growth mode resulting in a smooth surface and substrate/film interface. Magnetotransport measurements were conducted and show the expected metallic behavior with a residual resistivity ratio (RRR) of 2.25 (Fig. 1f). Angle-resolved photoemission spectroscopy (ARPES) measurements were conducted at BL-29ID at the Advanced Photon Source, using a vacuum suitcase to protect the film surface. The valence band density of states is compared to DFT calculations assuming both the dimerized and non-dimerized versions of the LaAuSb crystal. The crystal structure, magnetotransport, and electronic structure will be discussed.

Acknowledgement: this work was supported by a grant from the Army Research Office, W911NF-17-1-0254

11:40am **PCSI-3MoM-39 MBE Growth of  $\text{Cd}_3\text{As}_2$  on GaAs(001) Substrates**, *Anthony Rice, K Alberi*, National Renewable Energy Laboratory

The Dirac semimetal  $\text{Cd}_3\text{As}_2$  has become a useful material in the scientific community, as it provides access to a variety of interesting phenomena ranging from topological superconductivity to massless Dirac fermions. It is also potentially useful for energy-related applications due to its high electron mobility and large phonon-phonon scattering. Both single crystal and thin film synthesis has been achieved, however thin film research to date has been confined to non-traditional substrates, such as mica [1], or zinc blende (111) orientations [2,3] due to the preferred (112) growth plane of  $\text{Cd}_3\text{As}_2$ . To aid integration of  $\text{Cd}_3\text{As}_2$  into current technological applications, growth on a (001) substrate must be pursued.

Here, we explore the use of a  $\text{InTe}/\text{GaAs}(001)$  platform to support  $\text{Cd}_3\text{As}_2$ .  $\text{CdTe}$  grown directly onto  $\text{GaAs}(001)$  results in a (111) orientation due to the large lattice mismatch (14.6%), as shown by X-ray diffraction (XRD). A high temperature anneal prior to  $\text{CdTe}$  growth, resulting in a Ga-rich surface reconstruction, further improves the interface and resulting film morphology. Atomic force microscopy reveals film roughness comparable to  $\text{CdTe}$  structures on  $\text{GaAs}(111)$ . Growth of  $\text{Cd}_3\text{As}_2$  on this platform is explored and compared to similar films on  $\text{GaAs}(111)$  substrates.

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## PCSI

### Room Ballroom South - Session PCSI-4MoM

#### Surface Characterization

Moderator: Javad Shabani, New York University

11:45am **PCSI-4MoM-40 The Direct Band Gap of  $\alpha$ -Sn Investigated by Infrared Ellipsometry**, *Rigo Carrasco, C Zamarripa, S Zollner*, New Mexico State University; *J Menendez*, Arizona State University

Interest in gray tin has been revived because it is the endpoint constituent of  $\text{Ge}_{1-x}\text{Sn}_x$  alloys, which have potential applications as mid-infrared detectors and lasers,[1][2] and becomes a topological insulator or Dirac semimetal due to stress[3][4]. The band structure for gray tin is similar to Ge, but the  $\Gamma_7$  s-antibonding band, which is the conduction band in Ge, moves downward in gray tin and appears in between  $\Gamma_8^*$  and  $\Gamma_7^*$  with a negative energy  $E_0$  if we conventionally choose  $\Gamma_8^*$  as the zero energy level.

Using Fourier-transform infrared ellipsometry, we provide spectroscopic evidence about the valence band structure of  $\alpha$ -tin. The mid-infrared dielectric function of  $\alpha$ -tin grown pseudomorphically on InSb or CdTe by molecular beam epitaxy shows a strong  $\tilde{\epsilon}_0$  peak near 0.41 eV (Fig. 2). The peak can be attributed to the allowed intravalence band transitions from the  $\Gamma_7$  (electron-like) to the  $\Gamma_8^*$  heavy hole valence band and/or interband transitions from the  $\Gamma_7$  band to the  $\Gamma_8^*$  light “hole” conduction band. Possible sources for the strength of the peak, and its temperature dependence will be discussed. We would like to thank Dr. Arnold M. Kiefer and Stephanie Chastang for providing us the strained  $\alpha$ -tin samples.

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11:50am **PCSI-4MoM-41 Advanced ARPES Analyzer and Momentum Microscope KREIOS 150 – Concepts and First Results on Layered Materials and Topological Insulators**, *Thomas Schulmeyer*, SPECS-TII, Inc.

Modern ARPES analyzers provide a high degree of parallelization in data acquisition, recording hundreds of energy and angle channels simultaneously. Additionally, integrated deflectors enable users to perform angle scanning perpendicular to the analyzer’s entrance slit to record (kx, ky, E) data sets without sample rotation. However, the design of conventional analyzers implies a limited acceptance angle and corresponding accessible momentum space volume. Due to the trade-off between acceptance angle and angle resolution multiple changes in sample position and lens modes are necessary during a typical high resolution ARPES experiment. The new KREIOS 150 Energy Analyzer uses an extractor zoom lens design to overcome these limitations.

This new lens provides a full solid acceptance angle with highest angular resolution. In contrast to standard ARPES measurements with conventional hemispherical analyzers, electronic structure data from and beyond the 1st Brillouin zone is recorded without any sample movement. In addition, the lens of such an instrument can work in a lateral imaging mode for microscopy as well. This enables navigation on the sample and reduces the size of the area under investigation in ARPES down to a few micrometers in diameter. This combination of large acceptance angle, high angular resolution, and small acceptance area makes this instrument the ideal tool for electronic structure studies on small samples or sample areas. The design is compact with a straight optical axis.

The capabilities of this instrument were tested at the UE 56/2 at the Bessy II synchrotron in Berlin. Specification tests show excellent angle and lateral resolution as well as small spot capability down to  $2\mu\text{m}$  FOV. Subsequently real live samples like Graphene on Germanium were measured. Even on macroscopically rough surfaces like Graphene on NbSe2 excellent ARPES

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and X-PEEM results could be obtained. By taking advantage of the small spot capability of the KREIOS 150 meaningful band structure data has been recorded on such patchy samples.

Acknowledgements: We thank Yu. Dedkov (University of Shanghai, China) and M. Fonin (University Konstanz) for providing beamtime and samples for the measurements with KREIOS 150 at BESSY II.

11:55am **PCSI-4MoM-42 Investigating Relative Binding Strengths of Various Dye Attachment Chemistries at the Titania-Dye Interface in Dye-Sensitized Solar Cells**, *Gregory Smith, B Harvey, J Placzek*, Angelo State University

Dye-sensitized solar cells (DSSC) continue to be a promising area of research in our society's quest for clean and renewable energy. Typically, in these solar cells light is harvested by a dye molecule which is covalently bound to a mesoporous semiconducting material, most commonly titania. The excited electron is injected into the semiconductor and moves down to the electrode. One of the most important parts of these solar cells is the interface between the dye and the semiconductor. Optimally, the bond between dye and semiconductor should be strong, yet allow for rapid electron transport from excited dye to semiconductor. The most common chemistry for this attachment is with one or more carboxylate groups. There is a surprising lack of information about the performance of other chemistries in the literature, particularly experimental evidence. Some theoretical calculations have been performed<sup>1</sup>, but it would be very useful to have a library of the experimental performance of various attachment chemistries that could be used when considering specific dyes or in the synthesis of new dye molecules.

Here, we discuss a systematic method to compare the strength of the attachment of different chemistries to titania surfaces. To be able to directly compare chemistries, a model set of molecules were used, each 18 carbons in length capped with the group being studied. These molecules were used to form a self-assembled monolayer on a flat titania surface, then exposed to a typical solvent used in DSSCs at a slightly elevated temperature for 24 hours. The quality of the monolayer was measured before and after exposure using goniometry and ellipsometry. Table 1 shows the amount of degradation seen in each group studied. A description of the method, a comparison of the results, and how the results compare to previous theoretical studies will be included in discussion.

Terminal group:	Carboxyl	Thiol	Silane	Phosphonic acid
Degradation:	15.7 %	53.9 %	33.2 %	38.1 %

Table 1. Degradation of each monolayer after exposure to ethanol at 30°C for 24 hours.

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## PCSI

### Room Ballroom South - Session PCSI-1MoA

#### Device Interface Characterization

Moderator: Kimberly Thelander, Lund University

#### 2:00pm PCSI-1MoA-1 Sequential and In-Situ Atom Probe Tomography and Optical Spectroscopy on Single Luminescent Nanoscale Objects, *Lorenzo Rigutti*, University of Rouen Normandie, France **INVITED**

Correlating two or more microscopy and spectroscopy techniques on the same nanoscale object may yield an amount of information difficult to achieve by other means. In this contribution, we present selected studies of micro-photoluminescence ( $\mu$ -PL), high-resolution scanning transmission electron microscopy (HR-STEM) and laser-assisted atom probe tomography (APT) performed *sequentially* on single nano-objects containing quantum confined systems based on III-V and II-IV materials. This approach can be applied to the study of heterostructure interface definition, presence of extended defects such as stacking faults or dislocations, carrier localization and optical emission in quantum confined systems [1-4]. Furthermore, the use of complementary techniques may be extremely helpful for a correct interpretation of atom probe results and for understanding its limitations [3,5]. Finally, we will show that the study of PL *in situ* in an atom probe opens up novel possibilities, such as the discrimination - with a spatial resolution beyond the diffraction limit - of the optical signal of two quantum wells (QWs) and the manipulation of the optical signal of color centers by the application of an electrostatically-induced stress [6].

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#### 2:30pm PCSI-1MoA-7 The Three-dimensional Shape of Antiphase Domains in GaP on Si(001), *Pascal Farin*, Technische Universität Berlin, Germany

The integration of III-V semiconductors on Si(001) has been a long standing research aim to lower the cost of optoelectronic devices and simultaneously improve their performance. As the lattice mismatch between Si and GaP is smaller than 4%, this particular III-V semiconductor is used preferentially. However, due to charged, three-dimensional defects called antiphase domains (APDs) in GaP arising at the interface, its integration has proven to be quite challenging. While the search for growth conditions to avoid pronounced formation of these defects has been successful, the exact shape of the remaining ones is not yet fully understood.

In this work, APDs in GaP on Si(001) are investigated by means of transmission electron microscopy (TEM) and cross-sectional scanning tunneling microscopy (XSTM), two methods that offer unique insight into the appearance of APDs' cross sections due to their high surface sensitivity and resolution. The progression of the cross sections of the antiphase boundaries (short: antiphase boundaries) that separate the crystal's mainphase from the antiphase could be analyzed all the way down to the atomic level, allowing for an identification of the individual crystal planes along which the antiphase boundaries form. The accurate analysis of their progression by means of XSTM is illustrated by Fig. 1. After a thorough investigation of the antiphase boundaries' appearances on plane-view TEM and on the (110) and the (1-10) cleavage planes by means of XSTM, it has been possible to develop a true-to-scale, three-dimensional model of antiphase domains.

This project was supported by BMBF "MehrSi 03SF0525C", and by DFG through SFB 1083 and LE 3317/1-2.

#### 2:35pm PCSI-1MoA-8 Atom Probe Tomography of GaN Vertical Power Diodes: Impurity Distribution near Regrowth Interfaces, *Alexander Chang*, Northwestern University; *M Nami, B Li, J Han*, Yale University; *L Lauhon*, Northwestern University

GaN has demonstrated its utility in lighting and RF application and are being considered as the next generations of vertical power devices. However, high leakage currents, current crowding followed by premature breakdown, and degradation of the device[1] have hindered the GaN technology from entering the market. For nonplanar vertical power diodes, active regions of the device are formed through ex-situ processes, e.g. regrowth and selective area growth. Therefore, precise control of intentional and unintentional impurity incorporation near these interfaces is essential for achieving high performance devices[2]. Analysis of the 3D

spatial distribution and concentration of solute impurity atoms can be difficult, especially for nonplanar interfaces; thus, tomographic analysis becomes necessary. We describe atom probe tomography (APT) characterization of planar n/n and p/n GaN homojunctions with emphasis on detection sensitivity of dopants.

GaN homojunctions were grown by metal-organic chemical vapor deposition at Yale University, and analyzed using APT at Northwestern University. n-GaN templates were exposed to air prior to the regrowth of n-GaN or p-GaN to test subsequent cleaning procedures, and APT detection limits for impurities in GaN substrates and planar regrowth were analyzed by comparison with secondary ion mass spectroscopy (SIMS). Si impurities were observed with a surface density of  $10^{12}\text{cm}^{-2}$  for both n/n and p/n homojunctions and attributed to contamination from reactor parts and air. However, detection of Si in GaN using APT was limited due to the overlapping  $^{28}\text{Si}^+$  and  $^{14}\text{N}_2^+$  mass to charge ratios. The detection limits of other intentional and unintentional impurities in APT varied from  $10^{17}\text{cm}^{-3}$  to  $10^{19}\text{cm}^{-3}$ , depending on the peak positions relative to the Ga and N ion thermal tails. Analysis on spatial distribution of p-type dopant atoms did not reveal any evidence of clustering. Within the reconstructed APT data, the location of the regrowth interface of n/n homojunction was identified by spatial variations in charge states of evaporated Ga ions (Ga CSR). Although noticeable fluctuations in Ga CSR also coincided with the p/n homojunction, no clear correlation between Ga CSR and Mg concentration was observed. We will discuss efforts to extract the built-in field near p/n junctions and correlate with dopant concentrations.

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#### 2:40pm PCSI-1MoA-9 Surface/Subsurface Identification and Control of Ga<sub>2</sub>O<sub>3</sub> Native Point Defects, *Hantian Gao*, *S Muralidharan*, *N Pronin*, *M Karim*, *S White*, *T Asel*, *G Foster*, *S Krishnamoorthy*, *S Rajan*, *L Cao*, The Ohio State University; *M Higashiwaki*, National Institute of Information Communications Technology, Japan; *H Von Wenckstern*, *M Grundmann*, Universität Leipzig, Germany; *H Zhao*, The Ohio State University; *D Look*, Wright State University; *L Brillson*, The Ohio State University

Ga<sub>2</sub>O<sub>3</sub> has been considered a promising material for next generation high power devices due to its wide band gap enabling very high breakdown fields and n-type doping ranging from intrinsic to degenerate, both features of which lead to numerous solid-state electronic applications. However, surface/subsurface plays an important role in electronic applications of such material. Particularly, native point defects at surface/subsurface remain relatively unexplored. We used two surface sensitive techniques, depth-resolved cathodoluminescence spectroscopy and surface photovoltage spectroscopy to measure the optical signatures related to native point defects at surface/subsurface of Ga<sub>2</sub>O<sub>3</sub>. With near-surface treatments including plasma processing, annealing, and neutron irradiation on  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> grown by different methods, the near-surface sensitivity and depth resolution of these optical techniques enabled us to identify spectral changes associated with removing or creating these defects up to 180 nm below the material surface. This leads to measurement, identification, and control of one oxygen vacancy-related and two gallium vacancy – related energy levels in the  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> band gap. The combined near-surface detection and processing of Ga<sub>2</sub>O<sub>3</sub> suggests an avenue for identifying the physical nature and controlling the density of native point defects in this and other semiconductors at surface and subsurface.

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#### 2:45pm PCSI-1MoA-10 Electrically Detected Magnetic Resonance Study of Leakage Currents in a-SiN:H, *Ryan Waskiewicz*, *P Lenahan*, Pennsylvania State University; *S King*, Intel Corp.

Electronic transport in dielectric thin films are important concerns in semiconductor device technology. We have initiated a study of the defects involved in electron transport through a-SiN:H thin films of various stoichiometries utilizing electrically detected magnetic resonance (EDMR) measurements at multiple frequencies. The primary defect responsible for



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electronic transport (the K center) through SiN films has been identified using conventional electron paramagnetic resonance (EPR), but a detailed understanding of the effects of varying nitrogen incorporation in very thin films is not complete on the atomic scale. The EDMR measurement involves spin dependent trap assisted tunneling (SDTAT) on a sample structure of p-Si/a-SiN:H/Ti where the N/Si ratio in the a-SiN:H film was varied from 0.432 (N poor) up to 1.268 (nearly stoichiometric). In EDMR, we detect spin-dependent changes in device current under the influence of a slowly varying magnetic field. When the resonance condition is met, defect spins “flip” and a forbidden tunneling event becomes allowed, resulting in a change in device current that allows us to identify the chemistry and local structure of the defect responsible for leakage current. These measurements were made at low frequency ( $\nu=350\text{MHz}$ ) and high frequency ( $\nu=9.5\text{GHz}$ ) over the entire range of film stoichiometries. Fig. 1 shows the EDMR results at low frequency (dashed line, filled squares) and high frequency (solid line, open diamonds). The trend of increasing linewidth with increasing N/Si ratio is observed in both sets of measurements. A comparison between the high and low frequency EDMR measurements allows us to determine the contribution to linewidth broadening from spin orbit coupling and electron-nuclear hyperfine interactions [1]. The linewidth broadening is nearly identical for both sets of measurements, so we can conclude that a majority of the linewidth comes from hyperfine interactions and not spin orbit coupling (we would expect a significant increase in broadening at high field compared to low field). This result is consistent with the prediction of increasing hyperfine interactions with increasing average number of nitrogen atoms in K center defects.

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2:50pm **PCSI-1MoA-11 Internal Mechanical Stresses Relaxation in the Si-SiO<sub>2</sub> System and its Influence on the Interface Properties**, *Daniel Kropman, V Seeman*, Tartu University, Estonia; *A Medvids, P Onufrievs*, Riga Technical University, Latvia

It is known that internal mechanical stresses (IMS) due to the differences in the thermal expansion coefficients between films and substrates and lattice mismatch appear in the Si-SiO<sub>2</sub> system during the process of its formation and that point defects (PD) generation and redistribution could be used to reduce partially the surface stress. However, this process on the atomic scale is still not studied. The goal of the present report is to investigate the stress relaxation mechanism in the Si-SiO<sub>2</sub> system using EPR, IR absorption spectroscopy, scanning electron microscopy (SEM) and samples deflection measurements. PD density and stresses in the Si-SiO<sub>2</sub> system were varied by oxidation condition (temperature, time, cooling rate, ambient) and by Si<sub>3</sub>N<sub>4</sub> deposition on SiO<sub>2</sub>. Different sign of the thermal expansion coefficient of the SiO<sub>2</sub> and Si<sub>3</sub>N<sub>4</sub> on Si allow to modify the IMS at the interface. It has been found that samples deflection decreases or increases simultaneously with EPR signal intensity depending on the oxidation condition (temperature).

At oxidation temperature 1100°C the deflection of the samples(h) decreases with the increase of EPR signal intensity (vacancies), while at a oxidation temperature 1200°C EPR signal (I) and deflection increase simultaneously. Those allows to suggest that at lower oxidation temperature PD (vacancies) reduce the tensile IMS in Si, while at higher oxidation temperature compressive IMS created PD in SiO<sub>2</sub> (E' centers). At an intermediate oxidation temperature tensile stresses in Si and compressive stresses in SiO<sub>2</sub> may be equal and compensate each others (Fig.1).

It has been found that at oxidation temperature 1130°C IMS at the Si-SiO<sub>2</sub> interface are lower than at 1100°C and 1200°C. Lower defect density on samples cross-section microphotos obtained by SEM and PD density diminishing in samples oxidized at 1130°C confirmed this suggestion. In Fig.2 the EPR signal and IR absorption line-width dependence on the oxidation time is shown. It can be seen that EPR signal and IR absorption line-width at 1100 cm<sup>-1</sup> dependence on the oxidation time (oxide thickness) is nonmonotonous and depended on the cooling rate. In slowly cooled samples the increase of the EPR signal is accompanied by the decrease of  $\Delta\nu$  but, in fast cooled samples EPR signal and  $\Delta\nu$  increase simultaneously with increase oxidation time. Absence of the cooling rate influence on the PD density and  $\Delta\nu$  dependence on the oxidation time at I(t) and  $\Delta\nu(t)$  dependence intersection points show, that IMS by an appropriate choice of the SiO<sub>2</sub> film thickness disappear.

## PCSI

### Room Ballroom South - Session PCSI-2MoA

#### Spectroscopy of 2D Materials

Moderator: Kimberly Thelander, Lund University

2:55pm **PCSI-2MoA-12 Light Matter Interaction in Tunable 2D Materials and Artificial van der Waals Solids**, *Ursula Wurstbauer*, University of Münster, Germany **INVITED**

Atomically thin two-dimensional layered materials receive great interest because of their unique properties. Particularly, monolayers of semiconducting transition metal dichalcogenides (SC-TMDs), such as MoS<sub>2</sub>, excel due to their strong light-matter interaction that is dominated by exciton phenomena [1-3]. Key to the integration of SC-TMD and related artificial van der Waals solids into circuitries is the possibility to tune and engineer their properties on demand and on-chip e.g. by defects, dielectric environment or doping [4-7].

We apply inelastic light scattering together with emission, absorption and transport measurements to study the manifold coupling mechanism in van der Waal hetero- and hybrid structures. We introduce the influence of the dielectric environment, the charge carrier density as well as defects on the optical properties of these atomically thin materials and discuss consequences for their integration into optoelectronic circuits [8]. Moreover, optical properties of direct and indirect interlayer excitons in van der Waals heterostructures are addressed.

We acknowledge the financial support by the Deutsche Forschungsgemeinschaft (DFG) via excellence cluster Nanosystems Initiative Munich (NIM) as well as DFG project WU 637/4-1.

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3:25pm **PCSI-2MoA-18 Ultrafast Enhancement of Interfacial Exchange Coupling in Ferromagnetic Co<sub>2</sub>FeAl/(Ga,Mn)As Bilayer**, *Gunter Luepke*, College of William & Mary

Fast spin manipulation in low-dimensional magnetic heterostructures, where magnetic interactions between different materials often define the functionality of devices, is a key issue in the development of ultrafast spintronics. Although recently developed optical approaches such as ultrafast spin-transfer and spin-orbit torques open new pathways to fast spin manipulation, these processes do not utilize the unique possibilities offered by interfacial magnetic coupling effects in ferromagnetic multilayer systems. Here, we experimentally demonstrate ultrafast photoenhanced interfacial exchange interactions in the ferromagnetic Co<sub>2</sub>FeAl/(Ga,Mn)As system at low laser fluence levels [1]. The excitation efficiency is 30-40 times higher than without the (Ga,Mn)As layer due to *p-d* exchange interaction between photoexcited holes and Mn spins (Fig. 1). The coherent spin precessions persist to room temperature, indicating that proximity-induced ferromagnetism plays a key role in the optical excitation mechanism. The results highlight the importance of considering the range of interfacial exchange interactions in low-dimensional heterostructures and how these magnetic coupling effects can be utilized for ultrafast, low-power spin manipulation.

## PCSI

### Room Ballroom South - Session PCSI-3MoA

#### Magnetism in 2D Materials and Interfaces

Moderator: Anders Mikkelsen, Lund University, Sweden

4:30pm **PCSI-3MoA-31 Magnetism in Monolayer Transition Metal Dichalcogenides**, *Matthias Batzill*, University of South Florida **INVITED**

Van der Waals heterostructures hold the promise of combining materials with only weak interlayer interaction. This could allow integration of materials with diverse properties and minimize the influences of chemical interface interactions on materials and device properties. Of particular interest is the integration of magnetic materials in such van der Waals

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heterostructures. Although it has been recently demonstrated that ferromagnetism may be preserved down to the monolayer limit, there exists no known layered van der Waals material with Curie temperatures close to room temperature-- which is a pre-requisite for most applications. Thus there has been excitement by theoretical (DFT) predictions of high Curie temperature ferromagnetism in monolayer materials such as  $VSe_2$ , a material that is known to be paramagnetic in bulk-form. In addition, there exist increasing evidence of dilute magnetic semiconductors (DMS) in van der Waals materials. In this presentation, we illustrate direct growth of  $VSe_2$  monolayers on van der Waals substrates (Fig. 1) and demonstrate that these (sub)monolayers indeed exhibit ferromagnetic properties to above room temperature [1], while multilayers exhibit strongly suppressed magnetization (Fig. 2). Recent angle resolved photoemission spectroscopy (ARPES) characterization, however, indicate that  $VSe_2$  monolayers do not exhibit spin-split bands as predicted by DFT. In this talk we discuss possible alternative explanations for the observed magnetism. Moreover, we show evidence for magnetic coupling between monolayer  $VSe_2$  and a van der Waals substrate, which gives rise to an exchange bias that can be measured by magnetometry. Finally, we discuss an alternative approach to induce magnetism in 2D materials. Specifically, we show the formation of DMS by incorporation of magnetic impurities (Fig. 3) by a novel doping mechanism [2] in  $MoTe_2$ .

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5:00pm **PCSI-3MoA-37 Epitaxial Growth and STM Characterization of 2D Magnet  $MnSe_2$  and  $VSe_2$** , *Tiancong Zhu*, The Ohio State University; *D O'Hara*, University of California, Riverside; *J Repicky, J Cobbert, J Gupta*, The Ohio State University; *R Kawakami*, Ohio State University-Columbus  
Magnetism in 2D materials is a fascinating topic. Although extensively studied theoretically, the experimental realization of 2D magnets was not achieved until 2017 [1, 2]. Being only a few atoms thick, the high surface to volume ratio makes 2D magnets extremely sensitive to the surface environment, thus their magnetic properties can be strongly manipulated through proximity with other materials. Recently, the demonstration of epitaxial growth of 2D magnets [3,4] further brings more potential to the material. By directly synthesizing 2D magnet on different substrates, one could manipulate the electrical and magnetic properties of 2D magnet, explore new phenomena at their interface, as well as novel device structures for spintronic applications.

In this talk, we will present the epitaxial growth of 2D magnet  $MnSe_2$  and  $Vse_2$  on various substrate materials and characterize some of them with scanning tunneling microscopy (STM). First, we will present our recent discovery of  $MnSe_2$  [3], one of the first 2D magnets with intrinsic ferromagnetic ordering at room temperature (Fig. 1). The magnetic properties of  $MnSe_2$  grown on GaSe and SnSe<sub>2</sub> is compared. Furthermore, we will also show our exploration of the epitaxial growth of 2D magnet  $Vse_2$  on different substrates, including HOPG, GaAs(111) and Bi<sub>2</sub>Se<sub>3</sub>. Despite of the large lattice mismatch between  $Vse_2$  and the substrates, all the growths show epitaxial registry, as demonstrated by low energy electron diffraction (LEED) and STM (Fig. 2). The epitaxial alignment and clean interface between  $Vse_2$  and the substrate material make it possible to host proximity interaction between them. Finally, some preliminary result of characterizing the 2D magnet materials with spin-polarized STM will be discussed.

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5:05pm **PCSI-3MoA-38 Investigation of Low-Energy Ion-Implanted Multilayer Epitaxial Graphene**, *P Miceli, Alessandro Mazza*, University of Missouri

There is considerable interest in integrating magnetism with graphene in the hope of creating a new class of spintronic materials. With the recent discovery of a large band-gap semiconducting form of graphene on SiC substrates [1], there are new possibilities for epitaxial graphene-based electronics and integrating magnetism could introduce a new technological dimension. One potential avenue is to make graphene magnetic using defects to generate p-orbital magnetism. Indeed, atomic hydrogen [2]

attached to the graphene surface as well as vacancies [3] in graphene have been shown to induce magnetism, while high energy (MeV) proton irradiation can produce ferromagnetism at room-temperature in graphite [4]. However, detailed investigations of these systems are absent so that little is known about the density and configuration of defects, the role of interfaces, or how these relate to magnetism. Moreover, the p-orbital magnetism in these materials is unusual and of scientific interest.

In this talk, we will discuss our investigation [5] of atomic hydrogen implanted at low energies into multilayer epitaxial graphene grown on C-face SiC. The flat interface of this epitaxial system is conducive to x-ray and neutron reflectivity studies where the latter is sensitive to both the H density and magnetism. X-ray diffraction, measured *in situ* during 500 eV implantation, shows that the spacing between the graphene layers increases significantly with the ion dose. Most of the H ions remain within the sample after dosing as revealed by unpolarized neutron reflectivity. It is found from SQUID magnetometry that low-energy H implantation induces a ferromagnetic moment at room temperature. The implications of these results and others, such as ARPES and electron microscopy, will be discussed.

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5:10pm **PCSI-3MoA-39 Large Positive Linear Magnetoresistance in the Two-dimensional  $t_{2g}$  Electron Gas at the  $EuO/SrTiO_3$  Interface**, *Alexander Demkov*, The University of Texas

The high mobility two-dimensional  $t_{2g}$  electron gas (2DEG) present at oxide/oxide interfaces is currently under intense investigation [1-2]. In this talk, we will discuss the integration of highly spin-split ferromagnetic semiconductor  $EuO$  onto perovskite  $SrTiO_3$  (001). A careful deposition of  $Eu$  metal by molecular beam epitaxy results in crystalline  $EuO$  growth via oxygen out-diffusion from  $SrTiO_3$  [3]. This in turn leaves behind a highly conductive interfacial layer through generation of oxygen vacancies. Below the Curie temperature of 70 K of  $EuO$ , this spin-polarized two-dimensional  $t_{2g}$  electron gas at the  $EuO/SrTiO_3$  interface displays very large positive linear magnetoresistance (MR). Soft x-ray angle-resolved photoemission spectroscopy (SX-ARPES) reveals the  $t_{2g}$  nature of the carriers. First principles calculations strongly suggest that Zeeman splitting, caused by proximity magnetism and oxygen vacancies in  $SrTiO_3$ , is responsible for the MR [4]. This system offers an as-yet-unexplored route to pursue proximity-induced effects in the oxide two-dimensional  $t_{2g}$  electron gas [5].

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## PCSI

### Room Ballroom South - Session PCSI-4MoA

#### Oxide Growth and Properties

Moderator: Anders Mikkelsen, Lund University, Sweden

5:15pm **PCSI-4MoA-40 Invited Speaker, Bharat Jalan**, University of Minnesota **INVITED**

5:45pm **PCSI-4MoA-46 Strain Enhancement of the Electro-optical Response in Semiconductor-integrated Perovskites, Alexander Demkov**, The University of Texas

Interest in perovskite ferroelectrics such as BaTiO<sub>3</sub> (BTO) for use in nonlinear optic devices lies in its extremely large electro-optic (Pockels) coefficients >100 pm/V [1]. Even more importantly, the monolithic integration of BTO on semiconductors has paved the way to several types of entirely different devices ranging from ferroelectric memory to electro optical modulators [2-4]. Together, these developments have raised a possibility for applications of BTO in silicon nanophotonics, a hybrid technology combining semiconductor logic with fast broadband optical communications and optical information technologies .

I will discuss the possibility of significantly enhancing the nonlinear electro-optical response in strained perovskite BaTiO<sub>3</sub> and SrTiO<sub>3</sub> [5]. For BaTiO<sub>3</sub>, first principles calculations predict the enhancement for both compressive and tensile strain. The physical origin can be traced to strain-induced phonon softening that results in diverging first order susceptibility. Within the Landau-Ginzburg-Devonshire formalism we demonstrate how, in turn, this divergence results in a diverging second order susceptibility and Pockels coefficient. In epitaxially strained SrTiO<sub>3</sub> the electro-optical response is calculated for biaxial strain values ranging from -2.0% to 2.0% relative to the theoretically-optimized lattice constant. Under 1.0% tensile strain, the Pockels tensor components that are zero without strain due to the centrosymmetric structure of SrTiO<sub>3</sub>, increase dramatically. Experimentally, we study the nonlinear optical response in a strained thin film ferroelectric oxide BaTiO<sub>3</sub> using piezoelectric PMN-PT as a variable strain substrate and La-doped SrTiO<sub>3</sub> as a conductive buffer layer [6]. The rotation-anisotropic second harmonic intensity profile shows hysteretic modulation corresponding to strain variation from the inverse piezoelectric response of the substrate. Our results suggest a promising route to enhance the performance of nonlinear optical oxides for the development of future nano-opto-mechanical devices.

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5:50pm **PCSI-4MoA-47 Synthesis of Large Area Single-crystalline Freestanding Oxide Membranes, Prastuti Singh, A Swartz, D Lu, S Hong**, Stanford University; *K Nishio*, Geballe Laboratory for Advanced Materials; *Y Hikita*, Stanford Institute for Materials and Energy Sciences; *H Hwang*, Stanford University

Recently, a number of 2D materials have been the focus of intense study for the development of nanoelectronics [1]. One potential platform for study is the complex oxide perovskites. Complex oxides are known to exhibit a rich variety of properties such as superconductivity and ferroelectricity due to the local entanglement of the charge, spin and orbital degrees of freedom [2]. In addition, these properties have been shown to be highly tunable by strain, which is an advantage of the 2D geometry compared to bulk samples [3]. The synthesis of complex oxide nanomembranes, while highly challenging due to the 3D nature of the oxygen octahedral bonding, may offer pathways towards highly controllable flexible optical and electronic devices.

In order to take advantage of the unique properties of complex oxides, the final membrane or device must consist of high quality single-crystalline oxide materials. Recently, we have developed a new synthesis technique for crystalline complex oxide membranes and heterostructures compatible with state-of-the-art atomic-scale thin-film growth. The technique makes

use of pH-neutral water to selectively etch the sacrificial layer (Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>) to release the epitaxially grown thin-film from the substrate [4].

In this process, the lattice mismatch between the sacrificial layer and the thin-film plays a critical role in the growth process, membrane release, resulting nanocrystallite size, and material quality. A key question for further developing this highly promising route is to understand the scope of materials and lattice constants which can be accessed via the Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> family of materials. Here we take a representative transparent conducting oxide, La-doped BaSnO<sub>3</sub>, as a model system. The large lattice mismatch of 3.9% with Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> leads to high crack density when released due to the large epitaxial strain at the sacrificial layer and thin-film interface. Employing a lattice matched sacrificial layer (Sr,Ba)<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> (mismatch 0.4%) leads to successful fabrication of millimeter-scale large-area membranes (Fig. 1a) with crystallinity and electronic properties comparable to the epitaxial thin films (Fig. 1b), paving the way for the development of nanomembrane devices.

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## PCSI

### Room Ballroom South - Session PCSI-MoE

#### 2D Materials Characterization and Devices

Moderator: Ursula Wurstbauer, University of Münster

7:45pm **PCSI-MoE-1 Determining Chirality of Non-Centrosymmetric FeGe and MnGe with Spin-polarized Imaging of MnGe Spin Spirals via STM**, *J. P. Corbett*, *J Repicky*, *T Zhu*, *A Ahmed*, *R Bennet*, The Ohio State University; *J Guerrero-Sanchez*, Universidad Nacional Autonoma de Mexico, Mexico; *R Kawakami*, Ohio State University-Columbus; *J Gupta*, The Ohio State University

Recent interest in the 'B20' phase of FeGe and MnGe stems from noncollinear magnetic states, such as magnetic Skyrmions[1]. Here we present a joint STM and DFT study of FeGe and MnGe films grown by molecular beam epitaxy. An average surface lattice constant of  $\sim 6.8$  Å (FeGe) and of  $\sim 6.9$  Å (MnGe), in agreement with the bulk values, was observed via and LEED, as well as in situ RHEED during the MBE growth. Atomic resolution images of each of the four possible chemical terminations in the FeGe films were characterized by distinct image contrast and corrugation, as well as local density of states in tunneling spectra [2]. Likewise, three of the four possible chemical terminations of MnGe were identified in atomic resolution STM images. These assignments were confirmed by the good agreement between the STM images and DFT-simulated images using the Tersoff-Haman approximation. Having determined the surface terminations, STM images of the atomic layering order and registry across step edges allows us to uniquely determine the stacking order, and hence chirality of these films, which is difficult with conventional crystallography techniques.

The noncollinear spin state of MnGe stems from 3 interacting spin spirals, the triple- $\mathbf{q}$  state[1]. We have imaged spin-spirals of MnGe by SP-STM whose structure persists across quad-layer atomic steps which necessarily have the same surface termination. In contrast, the observed spiral contrast changes direction across non-quad-layer steps between different surface terminations. Spin-polarized tunneling spectroscopy reveals a pronounced field-dependence on one of these terminations, which is in good agreement with the calculated spin-resolved partial densities of states from DFT. These studies lay the groundwork for tuning Skyrmion size and thermal stability in B20-based interfacial heterostructures [1]

[1] Kanazawa, N., et al. Phys. Rev. B **96**, (2017): 220414

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8:00pm **PCSI-MoE-4 Ultrafast Spin and Charge Transfer in Monolayer WSe<sub>2</sub>-Graphene Heterostructure Devices**, *Michael Newburger*, *K Luo*, Ohio State University-Columbus; *K McCreary*, Naval Research Laboratory; *I Martin*, *E McCormick*, Ohio State University-Columbus; *B Jonker*, Naval Research Laboratory; *R Kawakami*, Ohio State University-Columbus

Monolayer transition metal dichalcogenides (TMDs) such as WSe<sub>2</sub> have garnered much attention due to their long spin/valley lifetimes and ability to optically excite spin/valley polarization. Additionally, one of the great strengths of TMDs is their ability to compliment other materials, such as graphene, by acting as a means of optical spin injection or proximity coupling. Recently multiple groups have observed efficient, proximity mediated charge transfer in TMD/graphene heterostructures [1]. Moreover, magnetoresistance measurements have demonstrated the ability to optically inject spins from TMD to graphene [2]. However, little work has been done to quantify the speed of spin transfer across a TMD/graphene interface or its effect on spin/valley lifetime.

Here we use time-resolved Kerr rotation (TRKR) microscopy to image the spatial dependence of spin/valley dynamics in monolayer WSe<sub>2</sub>/graphene heterostructures. Under p-type gating, Kerr rotation spatial maps of bare CVD grown WSe<sub>2</sub> demonstrate a characteristically spatial dependent, long-lived spin/valley signal [3]. Surprisingly though, these Kerr maps show strong quenching of spin/valley density at the WSe<sub>2</sub>/graphene interfaces. Time delay scans of the interfaces reveal lifetimes as low as several picoseconds, up to 3 orders of magnitude lower than typical lifetimes in bare WSe<sub>2</sub>. In addition, photoluminescence maps show quenched emission at the interfaces, whereas photoconductivity is enhanced at the same locations, demonstrating efficient charge transfer from WSe<sub>2</sub> to graphene. From these results, we conclude that the ultrafast quenching of spin/valley lifetime is due to transfer of spin information by the efficiently conducted charge carriers.

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8:05pm **PCSI-MoE-5 Probing Quantum Hall and Quantum Valley Hall Effect in Bilayer Graphene Nanostructures**, *Jing Li*, Los Alamos National Laboratory; *J Zhu*, The Pennsylvania State University

The lowest  $N = 0$  and  $1$  Landau levels (LLs) in bilayer graphene carry particularly rich physics due to the eight-fold degeneracy resulting from the spin, orbital isospin and valley degree of freedom. External knobs such as perpendicular electric field and tilted magnetic field put the ordering of electronic degree of freedom in bilayer graphene into a competitive fashion, which leads to a rich variety of many-body ground states such as postulated canted anti-ferromagnet, and a quantum spin Hall liquid have been observed. However, a general LL diagram in this system is still missing. We parameterize all the interaction effects and demonstrate an effective single-particle LL diagram for the quantum Hall (QH) octet in bilayer graphene in the presence of an electric field [1], which provides a framework to interpret a diverse group of experimental findings in the literature. This could serve as a starting point to explore more sophisticated effects of electron-electron interactions. Moreover, we demonstrate gate-controlled tunneling of QH edge states between two lateral QH systems in bilayer graphene by using a pair of dual split gates [2], which allow us to independently control the filling factors of the QH systems and also the potential profile in the tunnel junction. We observe sequential pinch-off of individual edge states, and the potential profile in the junction can be well understood using finite element simulations. The gate-controlled transmission of edge states is the foundation towards realizing more sophisticated nanostructures which enable further exploration of the intriguing fractional QH states in bilayer graphene.

In a separate project, we have demonstrated the existence of theory predicted valley-momentum locked edge states, i.e. quantum valley Hall (QVH) kink states, in a line junction of two oppositely gated bilayer graphene [3]. More recently, we have obtained ballistic QVH kink states with quantized conductance of  $4e^2/h$ , and furthermore valleytronic operations such as valley valve and electron beam splitter have been realized using these high quality kink states [4]. The versatile controls and potential scalability of this new helical 1D system open a door to many exciting possibilities in low dimensional topological applications.

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8:10pm **PCSI-MoE-6 Optoelectronic Modulation in 2D Mo<sub>1-x</sub>W<sub>x</sub>Te<sub>2</sub> Monolayers**, *Zakaria Al Balushi*, UC Berkeley

Alloys of transition metal dichalcogenides (TMDs), such as Mo<sub>1-x</sub>W<sub>x</sub>Te<sub>2</sub>, exhibit a wide range of electronic properties (semiconducting, semimetal and metallic phases) with unique polymorphs that depend on atomic stacking and coordination. However, reversible tunability between such phases is challenging. This is largely due to a combination of differences in the free energy between TMD polymorphs and phase transition kinetic barriers. Phase modulation in MoTe<sub>2</sub> has, however, gained recent interest due to the low barrier for transition (40 meV) between its semiconducting and semimetal phases when compared to other TMDs. Evidently, dynamic control between the thermodynamically favorable semiconducting 2H phase and the metastable semimetal 1T' phase in MoTe<sub>2</sub> is achieved with a variety of external stimuli, including strain, temperature and electrostatic doping. In the latter case, theory predicts that this phase transition occurs when the charge density exceeds  $\sim 10^{14}$  cm<sup>-2</sup>, which has been experimentally verified in monolayer MoTe<sub>2</sub> using ionic gating. However, reversible switching between these phases with a solid-state electrostatic gate still remains elusive. To circumvent the charge density requirements, theory predicts that the barrier for phase transition can be reduced in MoTe<sub>2</sub> when alloyed with tungsten. Evidence of this was demonstrated by Zhang et al. (arXiv:1709.03835), where an all solid-state resistive random-access memory device fabricated with Mo<sub>1-x</sub>W<sub>x</sub>Te<sub>2</sub> showed reversible switching between high and low resistance states. In addition to changes in conductivity, another feature of the phase transition in MoTe<sub>2</sub> includes in-

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plane structural changes that should give rise to district optical anisotropic responses in monolayers which remains highly unexplored.

We investigate and compare changes in the optical response of 2H-MoTe<sub>2</sub> and 2H-Mo<sub>1-x</sub>W<sub>x</sub>Te<sub>2</sub> alloys as they undergo phase transition between semiconductor-to-semimetal phases in an all vdW device structure using an hBN gate dielectric and graphene contacts. The optical response of the vdW stack are investigated using angle- and polarization-dependent reflection measurements as a function of gate voltage. Initial results showed changes in the optical response of the vdW stack between (1.1–1.4 μm) with applied gate voltage in alloys with x=0.09. Furthermore, polarization and temperature-dependent Raman measurements are also performed to map out structural changes in Mo<sub>1-x</sub>W<sub>x</sub>Te<sub>2</sub> as a function of applied gate voltage. These results will provide new insight into the optical response of these materials to enable new avenues for application in low-voltage and ultrafast modulators and other nanophotonic devices.

**8:15pm PCSI-MoE-7 Pressure-controlled Photoluminescence and Identification of an Electronic State in Hydrated Methyl-Terminated Germanane, B.A. Noesges, T Asel, W Huey, S Jiang, K Krymowski, Y Wang, W Windl, J Goldberger, L Brillson, The Ohio State University**

Two-dimensional (2D) materials continue to be an exciting area of research due to their interesting properties including large surface-to-volume ratio, high free carrier mobility and ease of stacking with other 2D materials to modular device design. Germanane, a germanium analogue to graphene, has a direct band gap around 1.6 eV that can be tuned by modifying the ligand termination on the Ge scaffold creating opportunity for optoelectronic applications [1]. Germanane, synthesized by deintercalating CaGe<sub>2</sub> with an acid chosen for a desired ligand termination, is sensitive to many factors which impact the behavior of this material. Defects in the Ge scaffold and incomplete reactions can create electronically active states within the band gap [2]. External factors such as ambient pressure and contaminants sitting between 2D layers can also play a large role in the observed electronic properties. We used an atmosphere-controlled photoluminescence spectroscopy (PLS) setup combined with depth-resolved cathodoluminescence spectroscopy (DRCLS) and density functional theory (DFT) calculations to investigate how atmospheric conditions affect germanane. Both pressure and atmospheric composition effects were seen. First, we observed the intensity of the photoluminescence measurement of germane vanish above a few hundred millitorr. Our original PLS measurements taken in low vacuum agree well with DRCLS collected in ultra-high vacuum (UHV) conditions [2]. If the germanane is in a water-free gas such as N<sub>2</sub>, O<sub>2</sub>, or Ar atmosphere, the luminescence stays quenched and no further states develop. However, once H<sub>2</sub>O is introduced either by simply using ambient air or bubbling a gas through water, a new hydrated state emerges with higher than band gap energy photons emitted at 1.8-1.9 eV which can be removed upon returning the germanane to vacuum. This demonstrates pressure-reversible control of the germanane electronic structure with atmospheric conditions. This work supported by NSF MRSEC under award number DMR-1420451.

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**8:20pm PCSI-MoE-8 Ultrafast Hot Electron Dynamics in InAs Nanowires with Variable Crystal Phases Investigated by Time-resolved Photoelectron Emission Microscopy, L Wittenbecher, J Vogelsang, S Lehmann, K Thelander, D Zigmantas, Anders Mikkelsen, Lund University, Sweden**

The III-V nanowire (NW) technology platform has reached a level of advancement that allows atomic scale tailoring as well as flexible device integration. In particular, controlled axial stacking of Wurtzite (Wz) and Zincblende (Zb) crystal phases is uniquely possible in the NWs[1]. We have previously found that multiphoton electron excitations can be controllably varied across the NW crystal segments [2] and that segments retain their unique electronic properties to the smallest possible scales[3]. In the present study we employ ultrafast time resolved PEEM utilizing femtosecond laser sources to explore the initial stages of the photo-carrier relaxation dynamics in InAs nanowires consisting of segments with different crystal structure. To this end, we combine PEEM with a tunable femtosecond laser source delivering sub 20fs pulses in the visible spectral range and perform spatially resolved one-color pump-probe measurements on individual nanowires. The photoelectron yield is found to increase

within approximately 100fs after excitation, followed by an exponential decay with a dominant time constant of about 400fs. We tentatively attribute these signals to the thermalization and the cooling of the photo-excited electrons. Furthermore, we demonstrate local variations of the relaxation times within individual nanowires as a function of excitation light polarization and crystal structure.

[1] D. Jacobsson et al, Nature, 531 (2016) 317

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**8:25pm PCSI-MoE-9 Van der Waals Integration beyond 2D Materials, Xiangfeng Duan, UCLA**

**INVITED**

The heterogeneous integration of dissimilar materials is a long pursuit of material science community and has defined the material foundation for modern electronics and optoelectronics. The current material integration strategy such as chemical epitaxial growth usually involves strong chemical bonds and is typically limited to materials with strict structure match and processing compatibility. Materials with substantially different lattice structures cannot be epitaxially grown together without generating too much interfacial defects that seriously alter/degrade their intrinsic properties. Alternatively, *van der Waals* integration, in which pre-formed building blocks are physically assembled together through weak *van der Waals* interactions, offers a bond-free material integration approach. The flexible “physical assembly” process used in *van der Waals* integration is not limited to materials that have similar lattice structures or require similar synthetic conditions. It can thus open up vast possibilities for damage-free integration of highly distinct materials beyond the traditional limits posed by lattice matching or process compatibility requirements, as exemplified by the recent blossom in the *van der Waals* integration of a broad range of 2D heterostructures. Here I will discuss *van der Waals* integration as a general material integration approach for creating diverse heterostructures with minimum integration-induced damage and interface states, enabling high-performing devices (including high speed transistors, diodes, flexible electronics) difficult to achieve with conventional “chemical integration” approach [1-10]. Recent highlights include the formation of *van der Waals* metal/semiconductor contacts free of Fermi level pinning to enable the first experimental validation of the Schottky-Mott rule since the initial prediction in 1930s [9]; and the development of *van der Waals* thin films for high performance large area electronics [10]; and the creation of a new class of *van der Waals* 2D-molecular superlattices with radically different layers yet atomic precision in each layer [7].

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## PCSI

### Room Ballroom South - Session PCSI-1TuM

#### Quantum Emitters and Excitations

**Moderator:** Javier García de Abajo, ICFO-Institut de Ciències Fotoniques

8:30am **PCSI-1TuM-1 The NV Center in Diamond: A Versatile Quantum Technology**, *Ania Bleszynski Jayich*, University of California, Santa Barbara; *A Jenkins, D Bluvstein, S Meynell, S Baumann, Z Zhang*, University of California, Santa Barbara

INVITED

The nitrogen vacancy (NV) center in diamond is an atomic-scale defect that exhibits remarkably coherent quantum properties in a uniquely accessible way: at room temperature, in ambient conditions, and even immersed in biological environments. NV centers are being explored for a variety of quantum technologies, including quantum sensing and quantum information processing. In this talk, I introduce the physics and materials science behind the success of the NV center and I highlight some of the major achievements of NV-based quantum sensors, the most advanced of NV-based technologies. I present a versatile NV-based imaging platform where we have incorporated an NV center into a scanning probe microscope and used it to image vortices in superconductors [1], skyrmions in thin film magnetic multilayers, and conductivity on the nanoscale [2].

I also outline the challenges facing the widespread use of NV centers in quantum applications, including spin decoherence [3] and charge state instabilities near interfaces. Using the NV center as a quantum probe of its local environment, we have identified several of the microscopic mechanisms responsible for reduced quantum functionality of near-surface NV centers, thus guiding the ongoing development of quantum control techniques and materials design, pushing towards the ultimate goal of NV-based single nuclear spin imaging.

9:05am **PCSI-1TuM-8 Stark Tuning of Single Photon Emitters in Hexagonal Boron Nitride**, *G Noh, D Choi*, Ajou University, Korea; *J Kim, D Im, Y Kim*, POSTECH, Korea; *H Seo, Jeun Lee*, Ajou University, Korea

Single photon emitters are fundamental resources of quantum optics and quantum information technologies. Recently, the emergence of single photon emission in atomic defects in hexagonal Boron Nitride (*h*-BN) at room temperature has evoked great interests in 2D-material-based single photon sources. For full exploitation of 2D single photon emitters for quantum technologies, however, the ability to control each atomic defect individually is critical. In this work, we show the electrical control of single photon emission in *h*-BN induced by an out-of-plane electric field [1]. This has been possible by fabricating a vertical heterostructure of *h*-BN containing atomic defects with graphene gates. A diverse spectral trail of Stark shifts is measured, providing information on defects' dipole transitions. The effect also persists at room temperature. We will also show possible ground states of defect structures that can induce the observed Stark shifts.

9:20am **PCSI-1TuM-11 Quantum Magnonics in V[TCNE]<sub>2</sub>**, *Ezekiel Johnston-Halperin*, The Ohio State University

The study of quantum coherent magnonic interactions relies implicitly on the ability to excite and exploit long lived spin wave excitations in a magnetic material. That requirement has led to the nearly universal reliance on yttrium iron garnet (YIG), which for half a century has reigned as the unchallenged leader in high-Q, low loss magnetic resonance, and more recently in the exploration of coherent quantum coupling between magnonic and spin [1] or superconducting [2] degrees of freedom. Surprisingly, the organic-based ferrimagnet vanadium tetracyanoethylene (V[TCNE]<sub>2</sub>) has recently emerged as a compelling alternative to YIG. In contrast to other organic-based materials V[TCNE]<sub>2</sub> exhibits a Curie temperature of over 600 K with robust room temperature hysteresis with sharp switching to full saturation. Further, since V[TCNE]<sub>2</sub> is grown via chemical vapor deposition (CVD) at 50 C it can be conformally deposited as a thin film on a wide variety of substrates with Q rivaling the very best thin-film YIG devices [3], which must be grown epitaxially on GGG substrates at temperatures over 800 C. Work in preparation shows that this Q can be as high as 8,000 (linewidth of 0.50 Oe at 9.86 GHz). Here, we will present evidence of coherent magnonic excitations in V[TCNE]<sub>2</sub> thin films and nanostructures, pointing to magnon-magnon coupling that can be tuned into the strong coupling regime and spin-magnon coupling that allows for the transduction of quantum information from 0D to extended quantum states. These results demonstrate the remarkable potential for these structures to play a major role in the emerging field of quantum magnonics, with applications ranging from the creation of highly coherent magnon crystals to quantum sensing and information. This work is

supported by DARPA/MTO MESO program and NSF Grant No. DMR-1507775 and EFRI NewLAW EFMA-1741666.

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## PCSI

### Room Ballroom South - Session PCSI-2TuM

#### Atomic Scale Characterization

**Moderator:** Javier García de Abajo, ICFO-Institut de Ciències Fotoniques

9:25am **PCSI-2TuM-12 Surface Potential and Hydrophilicity Measurements on Titanium Dioxide before and after Ultraviolet Irradiation**, *Takuya Furukawa, K Noda*, Keio University, Japan

Surface properties of titanium dioxide (TiO<sub>2</sub>) have been studied extensively due to their attractive phenomena such as photocatalysis and photo-induced superhydrophilicity. However, relations between these phenomena and photo-induced carriers in TiO<sub>2</sub> have not been fully understood yet. In this work, we discuss the relationship between surface potential (SP) and hydrophilicity on various TiO<sub>2</sub> surfaces before and after ultraviolet (UV) irradiation, by investigating the relaxation process of SP with Kelvin probe force microscopy (KFM) and water contact angle measured with an inkjet deposition system.

9:30am **PCSI-2TuM-13 Atomic-scale Observations of Reduced Graphene Oxide Nanosheets Dispersed on HOPG Substrates**, *Shaoxian Li, T Hirano, K Kawai, K Yamamura, K Arima*, Osaka University, Japan

Reduced graphene oxide (rGO) is a promising catalyst in both an electrode for fuel cells and a chemical tool for a semiconductor surface. In order to obtain higher catalytic performances, we need to control the structure of an rGO sheet on the atomic-scale. In this study, we aimed at obtaining highly resolved images of rGO sheets on HOPG by scanning tunneling microscopy (STM). First, we found four kinds of nanostructures in rGO. Secondly, both edges and defect sites of the rGO sheet implied to have high local density of states around the Fermi level.

9:35am **PCSI-2TuM-14 Diamond Coated Tips for Scanning Tunneling Microscopy**, *J Owen*, Zyvex Labs; *Ben Stein, O Auciello*, University of Texas at Dallas

Scanning Tunneling Microscopy (STM) has shown promise as an emerging tool for nanotechnologists to enable atomically-precise control over surface modification in the nanometer regime via surface lithography for device fabrication, and atomic-scale surface imaging and microanalysis. However, researchers have reported tip performance issues such as wear, oxidation and damage from electrostatic discharge, among other effects. While polycrystalline tungsten (W) wires have been typically used for tip fabrication via KOH-etching, diamond has long been considered an ideal potential candidate for numerous applications in scanning probe microscopy due to its well-documented hardness, chemical inertness and corrosion resistance, high Young's modulus, low coefficient of friction, and potential for controlled conductivity through selective doping via either Boron or Nitrogen atom insertion in grain boundaries. Numerous methods have been developed for fabricating diamond-based STM probes. However, no standardized practices have been established due to lack of repeatability, scalability and doping requirements. Clearly, there is room for improvement in the design and fabrication of diamond-based STM tips.

In this presentation, we report the development, application, and characterization of diamond coated STM tips and demonstrate their superior functionality in scanning imaging and lithography modes. Polycrystalline tungsten probe tips are prepared using the established method of electrochemical etching terminated at drop-off automated by the Zyvex ZEtcher system, followed by a secondary self-limiting Field-Directed Sputter Sharpening (FDSS) step, which utilizes an unfocused Ar<sup>+</sup> ion beam directed at a positively biased tip. Ultra-nanocrystalline diamond (UNCD) is then directly grown onto the tips without prior seeding in a microwave plasma chemical vapor deposition (MPCVD) process utilizing Bias Enhanced Nucleation (BEN) and Bias Enhanced Growth (BEG). We have found that by lowering the process temperature and plasma pressure, the extreme point of the tip can be quickly coated with a UNCD film tapering to a point with a radius of curvature <10 nm. While further sharpening of the diamond tip is possible with a lower energy FDSS step, it has been found that with ideal growth conditions no further sharpening is needed, nor is any extra doping step required to achieve tip conductivity. Furthermore,

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the tip can be used immediately for surface scanning and hydrogen depassivation lithography. UNCD film-based tips exhibit excellent durability, maintaining consistent scanning performance over very large scan areas. Tip morphology is characterized via STEM and ED.

9:40am **PCSI-2TuM-15 Surface Physical and Chemical Processes with an Optical Scanning Tunneling Microscope**, *Shaowei Li*, University of California, Irvine; *W Ho*, Northwestern University

The desire for observing finer details using optical microscopy particularly in bio-science has pushed technology developments toward the joint spatial-temporal resolution. The combination of optical excitation with scanning probe techniques provides a new window for viewing the unique properties of individual nano-scale objects. The optically excited atoms or molecules can be locally probed by a scanning probe microscope (SPM). The combination of a femtosecond (fs) laser with the scanning tunneling microscope (STM) would enable the study of laser photochemistry to attain simultaneous spatial and temporal resolutions.

Here, we demonstrate the laser photochemistry at single molecule level with a femtosecond laser STM, and ultimately probe the coherence molecular dynamics with joint fs-Å sensitivity. Irradiation of the STM junction with femtosecond laser pulses can generate energetic photo-assisted tunneling electrons. The coupling of laser to the electron tunneling process can shatter the diffraction limited resolution in light induced phenomena and reach the atomic scale resolution of the STM. We show that photo-assisted tunneling electrons can selectively activate individual C-H bonds in an azulene molecule adsorbed on a Ag(110) surface at 8.6 K. Electron energy required to break the bond decreased by 1.5 eV under 820 nm laser illumination, indicating that an electron can be coupled with one photon to induce the reaction. The C-H bond to be activated can be chosen by positioning the tip over the molecule with sub-Å resolution. The inelastic tunneling probe (itProbe) images taken before and after the reaction provide unambiguous structural identifications of the reaction products.

The coupling of femtosecond laser pulses to the tunneling process enables the investigation of coherence chemistry with joint fs-Å resolution. We observed the coherent vibration driven structural transition of single pyrrolidine molecules adsorbed on a Cu(001) surface with joint fsÅ resolution. The molecular dynamics, including vibrational frequency and lifetime, are determined to drive the coherent structural transition in the molecule. When two molecules are spatially close to each other, the intermolecular interactions lower the vibrational lifetime and downshift the vibration energy. We expect our work with joint fs-Å resolution to open a new avenue into probing single molecule coherent chemistry.

## PCSI

### Room Ballroom South - Session PCSI-3TuM

#### 2D Materials and Heterostructure Growth

**Moderator:** Susanne Stemmer, University of California, Santa Barbara

11:00am **PCSI-3TuM-31 Chemically and Atomically Ordered States in 2D Crystal Alloys**, *Nasim Alem*, Penn State University **INVITED**

Alloying and doping are considered versatile strategies for tuning charge and heat transport in nanostructures. Whether the resulting alloy structure is random or ordered can have a profound impact on the macroscale electronic, optoelectronic, vibrational, and transport properties of the material. In this talk, we present structural and chemical ordering<sup>1,2</sup> as a mechanism to design anisotropy in the family of 2D transition metal dichalcogenides (TMDs) alloys (Fig. 1). Leveraging recent advancements in atomic resolution scanning/transmission electron microscopy (S/TEM) imaging and spectroscopy, we show the formation of chemically ordered states and vacancy/dopant coupling that leads to unusual relaxation effects around dopant-vacancy complexes leading to local strain and symmetry breaking around individual dopant sites. In addition, we will further uncover the defect structure, i.e. grain boundaries and anti-phase boundaries, and their stability and dynamics in monolayer TMD crystals and their alloys. This understanding can have a strong impact on the synthesis and functionality of novel nanostructure alloys and provides the key to properly design devices for heat dissipation applications, energy storage, electronics, optoelectronics, and thermoelectrics.

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11:30am **PCSI-3TuM-37 Defect-Assisted Heteroepitaxial Growth of Monolayer Tungsten Diselenide Films with Preferential Orientation on Hexagonal Boron Nitride**, *Xiaotian Zhang, F Zhang, Y Wang, D Schulman, T Zhang, A Bansal, N Alem, S Das, V Crespi, M Terrones, J Redwing*, The Pennsylvania State University

The rapid development of device technologies based on 2D transition metal dichalcogenides (TMDs) causes increasing demand for synthesis of high quality large area monolayer and few layer films. Our previous work demonstrated epitaxial growth of large area monolayer WSe<sub>2</sub> films on c-plane sapphire using gas source chemical vapor deposition (CVD). However, the optical and electrical properties of coalesced monolayer films grown on sapphire are negatively impacted by the existence of anti-phase boundaries (APBs) as well as non-uniformities arising from steps and charge-induced doping associated with the sapphire surface. Prior studies demonstrated a preferred domain orientation for TMDs grown on hBN and first-principle calculations suggest this phenomenon originates from single atom vacancies on the hBN surface that act as nucleation sites. In this study, we further investigate the mechanism of defect-assisted domain alignment of 2D TMDs on hBN and demonstrate the growth of fully-coalesced WSe<sub>2</sub> films on hBN with a reduced density of APBs and improved optical and electrical properties compared to films grown on sapphire.

WSe<sub>2</sub> monolayer films were grown by gas source CVD at 800°C using W(CO)<sub>6</sub> and H<sub>2</sub>Se in a H<sub>2</sub> carrier gas employing a multi-step process to separately control nucleation density and lateral growth and coalescence of domains. Single crystal hBN flakes exfoliated from bulk crystals and transferred onto c-plane sapphire were used as substrates. He plasma treatment and NH<sub>3</sub> annealing were used to modify the surface defect density of hBN. Detailed studies of WSe<sub>2</sub> deposition on hBN as a function of growth conditions and substrate pre-treatment confirm that domain nucleation is controlled by the surface defect density rather than the precursor concentration. Over 90% of WSe<sub>2</sub> domains have consistent orientation via the defect-assisted growth. Through careful control of nucleation and extended lateral growth time, fully coalesced WSe<sub>2</sub> monolayer films on hBN were produced for subsequent characterization. High resolution scanning transmission electron microscopy (S/TEM) analysis demonstrates the absence of APBs in coalesced regions formed by the merging of 0° oriented domains. Temperature-dependent photoluminescence measurements show sharp and enhanced exciton and trion emission peaks, with no defect-related bound exciton emission from monolayer WSe<sub>2</sub>/hBN down to 80K. Backgated FET devices fabricated on WSe<sub>2</sub>/hBN films transferred to SiO<sub>2</sub>/Si substrates show an order of magnitude increase in room temperature carrier mobility (~5 cm<sup>2</sup>/V-s) compared to similar devices fabricated using monolayer WSe<sub>2</sub> films transferred from sapphire.

11:35am **PCSI-3TuM-38 Novel Sulfide Heterostructures from Designed Precursors**, *D Roberts*, University of Colorado at Boulder; *Sage Bauers, J Perkins*, National Renewable Energy Laboratory; *C Stoldt*, University of Colorado at Boulder; *A Zakutayev*, National Renewable Energy Laboratory  
Composite 2D material systems present not only unique structural and electrical phenomenon but can be intentionally designed to elicit properties desirable for a specific application. Existing reports of chalcogenide heterostructures have shown the ways in which varying thicknesses of constituent layers from monolayer to bulk-like affects the resistivity, [1] transport properties, [2] and overarching band structure [3] of such systems. To date, systems in this vein are limited to selenides and tellurides. Extension to sulfide systems is important to harness properties of emerging 2D materials such as MoS<sub>2</sub> and WS<sub>2</sub>

In this work we demonstrate the synthesis of multilayer sulfide heterostructures crystallized from amorphous precursors via sputter deposition. We utilize a combinatorial approach to depositions in order to reduce time and specificity needed in calibrating the time needed to deposit constituent components of such structures. A schematic of this setup can be seen in Fig. 1. Here we will focus on spatially resolved structural characterization of these combinatorial samples with respect to changes in long range ordering indicative of improved crystallinity. Additionally, we investigate the role of off-stoichiometry in as-deposited precursors on the structure and quality of our crystalline end product. Challenges inherent in sulfur-based depositions are also addressed.

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11:40am PCSI-3TuM-39 Rotational Alignment of Epitaxially-grown hBN on Macrostepped Graphene/SiC(0001) Single-Crystal Substrates, *Daniel Pennachio*, University of California, Santa Barbara; *C Ornelas-Skarin*, University of California, Irvine; *N Wilson*, *E Young*, *A McFadden*, *T Brown-Heft*, University of California, Santa Barbara; *K Daniels*, *R Myers-Ward*, *K Gaskill*, *C Eddy, Jr.*, U.S. Naval Research Laboratory; *C Palmstrom*, University of California, Santa Barbara

Many of the intriguing properties of 2D devices rely on the relative rotational alignment between layers. For instance in the graphene/hBN system, band structure modulation can occur at specific alignments [1], but a misalignment may be beneficial if innate graphene properties are to be examined. To allow for scalable graphene/hBN heterostructure formation, this work investigates hBN growth on single-crystal epitaxial graphene (EG) on macrostepped SiC(0001) substrates. The presented results suggest these macrosteps may influence the hBN epitaxial relation such that a metastable, 30° in-plane hBN/EG alignment is more favorable with certain growth conditions than the direct 0° alignment between hBN/graphene, despite their similar crystal structures.

Plasma-enhanced chemical beam epitaxy (PE-CBE), an ultra-high vacuum (UHV) compatible process, was utilized to provide a clean environment for examination of the hBN structural, electrical, and chemical properties via *in-situ* and *in-vacuo* characterization methods. To determine the effect of substrate macrostep morphology, EG on SiC (0001) substrates with no offcut and with a 4° offcut toward  $\langle 11\bar{2}0 \rangle_{\text{SiC}}$  were tested. The alignment of the hBN/EG/SiC(0001) heterostructure was studied by relating *in situ* electron diffraction to nuclei edge directions. In addition, cross-sectional transmission electron microscopy (TEM) confirmed registry of the hBN to the EG/SiC substrate, while plan-view TEM showed in-plane alignment and uniformity. The macrostep-directed epitaxy of hBN on EG highlighted in this work highlights the possibility of various rotational alignment during van der Waals epitaxy, a promising feature for direct growth of 2D heterostructures.

[1] M. Yankowitz, *et al.*, Nat. Phys. **8**, 382 (2012).



# Tuesday Evening, January 15, 2019

## PCSI

### Room Ballroom South - Session PCSI-TuE

#### Quantum Materials?

**Moderator:** Debdeep Jena, Cornell University

7:00pm **PCSI-TuE-1 Magnetic Weyl Semimetals!**, *Claudia Felser, J Gooth, K Manna, E Lui, Y Sun*, Max Planck Institute, Germany **INVITED**

Topology a mathematical concept became recently a hot topic in condensed matter physics and materials science. One important criteria for the identification of the topological material is in the language of chemistry the inert pair effect of the s-electrons in heavy elements and the symmetry of the crystal structure [1]. Beside of Weyl and Dirac new fermions can be identified compounds via linear and quadratic 3-, 6- and 8- band crossings stabilized by space group symmetries [2]. Binary phosphides are the ideal material class for a systematic study of Dirac and Weyl physics. Weyl points, a new class of topological phases was also predicted in NbP, NbAs, TaP, MoP and WP2. [3-7]. In magnetic materials the Berry curvature and the classical AHE helps to identify interesting candidates. Magnetic Heusler compounds were already identified as Weyl semimetals such as Co<sub>2</sub>YZ [8-10], in Mn<sub>3</sub>Sn [11,12] and Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> [13].

The Anomalous Hall angle helps to identify even materials in which a QAHE should be possible in thin films. Besides this k-space Berry curvature, Heusler compounds with non-collinear magnetic structures also possess real-space topological states in the form of magnetic antiskyrmions, which have not yet been observed in other materials [14].

[1] Bradlyn et al., Nature 547 298, (2017) arXiv:1703.02050

[2] Bradlyn, et al., Science 353, aaf5037A (2016).

[3] Shekhar, et al., Nature Physics 11, 645 (2015)

[4] Liu, et al., Nature Materials 15, 27 (2016)

[5] Yang, et al., Nature Physics 11, 728 (2015)

[6] Shekhar, et al. preprint arXiv:1703.03736

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[10] Kübler and Felser, EPL 108 (2014) 67001 (2014)

[11] Nayak, et al., Science Advances 2 e1501870 (2016)

[12] Nakatsuji, Kiyohara and Higo, Nature 527 212 (2015)

[13] Liu, et al. preprint arXiv:1712.06722

[14] Nayak, et al., Nature 548, 561 (2017)

7:30pm **PCSI-TuE-7 Status of Purity for Bulk Samples and Implications for Quantum States**, *Arthur Ramirez*, University of California Santa Cruz **INVITED**

Present interest in topological materials, some with quantum entangled ground states, is driven in part by a proliferation of new bulk compounds. These materials include structures that lack inversion or time-reversal symmetry, leading to Weyl semimetal (WSM) behavior, and triangular or honeycomb systems capable of supporting a quantum spin liquid (QSL) states. The realization of either undoped WSM or fully entangled QSL phases will likely require levels of crystallinity far beyond that realized in present samples. I will sketch out a few examples in both classes of systems and discuss approaches to achieve high purity bulk material.

8:00pm **PCSI-TuE-13 Rump Session**,

## PCSI

### Room Ballroom South - Session PCSI-1WeM

#### Hybrid and Mixed-Dimensional Interfaces I

Moderator: A. Alec Talin, Sandia National Laboratories

##### 8:30am PCSI-1WeM-1 Nanoimaging and Spectroscopy of Emerging Photovoltaic, Marina Leite, University of Maryland INVITED

Solar energy can deliver the >184 000 TWh per annum required to meet the current worldwide energy needs. Yet, High-performance and low-cost photovoltaics (PV) are still required to successfully substitute fossil fuel-based technologies. Hybrid perovskites and thin-film polycrystalline materials are promising options for high-performance and low-cost PV. These materials present mesoscale constructs, with nano- and microscale grains and boundaries that individually contribute to the devices' optoelectronic behavior [1,2]. For hybrid perovskites, their stability is the primary limiting factor towards commercialization. Thus, we resolve the dynamic optical [3] and electrical [4] responses from the macro- to the nanoscale, as these materials are exposed to: water, oxygen, temperature, bias, and illumination. We further propose a machine learning (ML) tactic to deconvolute the individual and combined effects of each parameter on device stability. We elucidate ion motion within the perovskite grains by a fast scanning probe microscopic new method that enables us to spatially (<20 nm) and temporally (msec) resolve the devices' open-circuit voltage (Voc) (Fig 1) [4,5]. In the realm of polycrystalline materials, we demonstrate a novel AFM-based approach to image local Voc variations, based on illuminated Kelvin-probe force microscopy [6]. Our functional imaging paradigm provides a new platform to map device performance with nanoscale spatial resolution.

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S. Leite. Advanced Energy Materials 5, 1501142(2015).

##### 9:05am PCSI-1WeM-8 Electronic Charge Transport in Solution-processed Vertically Stacked 2D Perovskite Quantum Wells, H Tsai, Rice University; R Asadpour, Purdue University; M Kanatzidis, Northwestern University; M Alam, Purdue University; A Mohite, Rice University; Wanyi Nie, Los Alamos National Laboratory

State-of-the-art quantum well based devices such as photovoltaics, photodetectors and light emission devices were enabled by understanding the nature and the exact mechanism of electronic charge transport. Ruddlesden-Popper halide perovskites are two-dimensional solution-processed quantum wells and have recently emerged as highly efficient semiconductor for solar cell approaching 13% in power conversion efficiency. However, further improvements will require an understanding of charge transport mechanisms, which are currently unknown and further complicated by the presence of strongly bound excitons. In our study, we discovered that the carrier transport is closely related to the structure and stacking of those quantum wells. Combining systematic device characterization and simulation, we conclude that the photo generated electron-hole pairs need to overcome the multiple internal potential barriers for collection to occur. The potential barriers that block the efficient separation of electron-hole pairs are attributed to the misalignment between crystalline slabs. Both of those properties are unfavorable for photovoltaic cell operation. Here we elucidate the critical role of field-assisted charge carrier separation that overcomes these bottlenecks leading to the efficient photocurrent collection. On the other

hand, the structure of the 2D perovskites could promote the radiative recombination by spatially localizing the electrical injected carriers, which makes them an excellent material for light emitting diodes.

##### 9:10am PCSI-1WeM-9 N-type Doping in Organic Semiconductor Thin Films by using a Dendritic Oligoarylamine-substituted Benzimidazole Dopant, Yuji Yoshihashi, Keio University, Japan

Organic n-type doping with donor dopants has not been established well because of air instability of conventional donor dopants. With this background, we have focused on imidazole-based compounds as solution-processable and pure organic n-type dopants. These compounds are known as strong single-electron reductants. However, these dopants are quickly oxidized in solution phase in ambient condition. In this work, we report an approach for increasing the stability of benzimidazole dopant molecule in solution phase by attachment dendritic oligoarylamine (N,N-bis[4-(di-4-anisyl-amino) phenyl]amine) at the 2-position of the imidazole ring (N3-DMBI-H shown in Fig. 1)[1].

N3-DMBI-H was newly synthesized by ourselves. N3-DMBI-H solution in chlorobenzene (CB) revealed much better stability in ambient air than solutions of other benzimidazole dopants. [6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM) solution (in CB) was dropcast onto Si substrates covered with 200 nm-thick SiO<sub>2</sub> layer. Doping concentrations of N3-DMBI-H were 0, 0.5, 2 and 5 wt%. After that, the dropcast films were dried and heated at 100°C overnight to activate the dopants. The above processes were performed in a N<sub>2</sub>-filled glovebox. After Au electrodes (thickness: 25 nm) were deposited onto the PCBM film surfaces by vacuum evaporation, we measured two-terminal current-voltage (*I-V*) characteristics of PCBM films under a vacuum condition (10<sup>-2</sup> Pa).

Figure 2 shows two-terminal *I-V* characteristics of undoped and N3-DMBI-H doped PCBM films. The gap distance between the two contact electrodes was 50 μm. As shown in Fig. 2, doping N3-DMBI-H into PCBM increased the current value by more than five orders of magnitude. The electrical conductivity of the N3-DMBI-H-doped PCBM thin film was reported to be 6.39 × 10<sup>-4</sup> cm<sup>-1</sup>(undoped PCBM: 6.62 × 10<sup>-9</sup> S cm<sup>-1</sup>). This result suggests that the electric resistance of PCBM thin films considerably decreased by N3-DMBI-H doping, due to carrier (electron) doping effects.

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##### 9:15am PCSI-1WeM-10 Multi-scale Modeling of Molecule-Surface Interactions for Improved Charge Transfer across Photoelectrochemical Interfaces, A Iyer, K Kearney, University of Illinois at Urbana-Champaign; A Rockett, Colorado School of Mines; Elif Ertekin, University of Illinois at Urbana-Champaign INVITED

The design of efficient, stable photoelectrochemical devices requires the use of complex heterojunctions composed of semiconducting, protective, and catalytic layers. Understanding the nature of charge transport across these interfaces is challenging, due to the complexity of the interfaces and possibility of competing charge transfer mechanisms. We present our recently developed multiscale approach that combines first-principles density functional theory (DFT) and solid-state drift/diffusion device scale modeling to give insights into the nature of charge transport across photoelectrode interfaces [1-3], Fig. 1. DFT is used to estimate the surface dipole induced by the functionalization, and the device software wxAMPS is used to predict experimentally measurable features such as the JV curve and the open-circuit voltage. Using this approach, we are able to identify the mechanism of hole transfer across *n*-Si(111)-R|TiO<sub>2</sub> photoanodes where -R is a series of mixed aryl/methyl monolayers containing an increasing number of methoxy units (mono, di, and tri). In collaboration with experiment, we find that hole transport is limited at the *n*-Si(111)-R|TiO<sub>2</sub> interface and occurs by two processes—*thermionic emission* and/or *intraband tunneling*—where the interplay between them is regulated by the interfacial molecular dipole. Finally we will highlight how molecule/surface interactions can be used to model doped silicon slabs at experimental doping densities in a computationally tractable manner.

##### 9:45am PCSI-1WeM-16 Scanning Electrochemical Microscopy of Graphene-based Hybrids: Insights into Physicochemical Interfacial Processes and Electroactive Site Density Distribution, Sanju Gupta, Western Kentucky University

Surface (and interfacial) science is found in various environments of scientific significance including biomembranes, ocean and atmospheric chemistry, applied electrochemistry and at the grand challenges of *energy-water-nexus* [1]. Molecular redox behavior on the surface and at the interface is drastically different than bulk counterparts.

# Wednesday Morning, January 16, 2019

In this talk, we demonstrate scanning electrochemical microscopy (SECM) is a powerful tool to investigate dynamic physical-chemical processes at surfaces and buried interfaces. This technique helps to determine ion (or electron) transfer kinetic rate, diffusion coefficient, imaging electrochemical redox reactions and activity. The significant advantage offered by SECM is its capability of probing chemical information at the solid/liquid interfaces. A constant potential is applied to the tip and electrochemical working electrode (*i.e.* the substrate in electrolyte) to drive reaction of redox species (or mediator) in bulk electrolyte solution to probe the surfaces of graphene-based hybrids, relevant for electrochemical energy systems. The micro-configured cyclic voltammograms, probe approach (current versus tip-substrate distance) curves in feedback and imaging modes, for graphene/CNT, graphene/transition metal oxide and aerogels are chosen as case examples to probe ion adsorption, charge transfer dynamics and to map highly electroactive ('hot spots') edge sites. The SECM setup has a resolution of ~40 nm and can locate and relocate areas of interest precisely after a coarse image. The heterogeneous electrode surfaces comprised of graphene nanosheets (conducting)/other nanomaterials (semiconducting) exhibit peak and valley tip current behavior and site distribution and the data is analyzed in terms of reactive basal plane and edge plane sites distribution, and help to determine heterogeneous rate constant using modeling and fitting experimental data. The findings reinforce the available electron density of states in the vicinity of the Fermi level contributing to higher electroactivity, faster interfacial diffusion, and shorter distances for electron transfer, facilitated through molecular and chemical bridges [1].

[1] S. Gupta and S. B. Carrizosa, *Appl. Phys. Lett.* **109**, 243903 (2016).

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[mailto:sanju.gupta@wku.edu]

## PCSI

### Room Ballroom South - Session PCSI-2WeM

#### Topological Materials II

**Moderator:** Lincoln J. Lauhon, Northwestern University

11:00am **PCSI-2WeM-31 Epitaxial Nitride Semiconductor/Superconductor Heterostructures**, *Debdeep Jena*, Cornell University **INVITED**

A new development in MBE growth of lattice-matched NbN/AlN/GaN heterostructures has allowed the integration of GaN based high-electron mobility transistors (HEMTs) with MBE grown NbN superconductors. Structural and electronic properties of these heterostructures and the effect of interfaces will be discussed. Attempts to obtain the quantum Hall effect in the 2DEGs epitaxially grown on the superconductor will be discussed. Finally, progress towards the MBE growth of NbN/AlN/NbN Josephson junctions will be reported.

11:30am **PCSI-2WeM-37 Wafer Bonding Approach for Epitaxial Al/GaAs(001)/Al Tri-layers**, *Anthony McFadden*, *M Seas*, University of California, Santa Barbara; *C McRae*, *R Lake*, National Institute of Standards and Technology; *J Wen*, *J Wang*, *I Arslan*, Argonne National Laboratory; *D Pappas*, National Institute of Standards and Technology; *C Palmstrøm*, University of California, Santa Barbara

Superconductor-insulator-superconductor (Josephson) junctions utilizing amorphous oxide barriers have been studied extensively, however relatively little work has been done using single crystal semiconductors in place of amorphous oxide barriers. This is likely due to difficulty in fabrication of such structures including symmetry mismatch of the semiconductor to the superconductor and the reactions and roughening that may occur at the temperatures needed for semiconductor growth. This work focuses on a wafer bonding approach, subsequent substrate removal, and superconductor regrowth for fabrication of Al/GaAs(001)/Al Josephson junctions. AlGaAs/GaAs/Al structures are grown by molecular beam epitaxy on GaAs(001) substrates and wafer-bonded to Si. The substrate and sacrificial AlGaAs layers were removed by selective wet etching followed by surface cleaning in ultrahigh vacuum and aluminum regrowth. The wafer bond and Al/GaAs interfaces are studied by transmission electron microscopy (TEM). X-ray photoelectron microscopy (XPS) is used to determine GaAs surface cleaning conditions compatible with the wafer bonding process following substrate removal. X-ray diffraction (XRD) and reflection high energy electron diffraction (RHEED) is used to assess crystalline quality and orientation of the epitaxial aluminum.

11:35am **PCSI-2WeM-38 Growth and Nucleation of Low-Loss Titanium Nitride Superconductors on Silicon (111) using Plasma Assisted MBE**, *Chris Richardson*, *A Alexander*, *C Weddle*, University of Maryland; *B Arey*, *M Olszta*, Pacific Northwest National Laboratory

Titanium nitride (TiN) is a known superconducting material that has demonstrated low-microwave-loss and used as passive components in superconducting quantum circuits for quantum information devices [1]. In contrast to conventional synthesis techniques, plasma assisted molecular beam epitaxy is used to produce low-loss TiN on bare silicon wafers. Using a rf-plasma source to crack the nitrogen molecules, and a conventional high temperature effusion cell for titanium, TiN growth is completed under nitrogen rich conditions to produce polycrystalline thin films that sit on an amorphous nitride layer. The motivation and activities pursued to synthesize epitaxial TiN on silicon that is compatible with the requirements of superconducting quantum circuits will be discussed.

A number of techniques are used to characterize the structure of the material, while cryogenic tests down to 80 mK characterize the superconducting properties. Coplanar waveguide resonators operating at 5 GHz demonstrate single photon quality factors above 1M, and high-power quality factors that approach 7M without observing saturation.

#### REFERENCE:

[1] J. B. Chang, M. R. Vissers, A. D. Córcoles, M. Sandberg, J. Gao, D. W. Abraham, J. M. Chow, J. M. Gambetta, M. B. Rothwell, G. A. Keefe, M. Steffen, and D. P. Pappas, *Appl. Phys. Lett.* **103**, 012602 (2013).

## PCSI

### Room Ballroom South - Session PCSI-3WeM

#### Exploiting Ions in Devices

**Moderator:** Lincoln J. Lauhon, Northwestern University

11:40am **PCSI-3WeM-39 Non-volatile Redox Memory for Brain Inspired Computing**, *Elliot Fuller*, Sandia National Laboratories; *S Keene*, *A Melianas*, Stanford University; *Z Wang*, University of Massachusetts Amherst; *S Agarwal*, *Y Li*, Sandia National Laboratories; *Y Tuchman*, Stanford University; *C James*, *M Marinella*, Sandia National Laboratories; *J Yang*, University of Massachusetts Amherst; *A Salleo*, Stanford University; *A Talin*, Sandia National Laboratories

The end to Moore's law is driving interest in developing novel memory and logic devices with dramatically improved energy efficiency. Electrochemical systems can improve efficiency and imbue novel functionality through ion-induced transformations that are inaccessible to traditional semiconductor devices. Here, I will discuss two electrochemical systems that are combined to execute neuromorphic computing: one based upon proton insertion/extraction and redox reaction in a polymer matrix for synaptic weight storage[1, 2], and a second based upon ion migration and diffusion for synapse addressability and retention[3]. The first device is a synaptic transistor based upon PEDOT:PSS (Fig. 1 green, blue, orange) that is programmed via protonation/de-protonation of the channel. Addressable programming of the polymer synapse is mediated by current injection through a Ag-based diffusive electrochemical-metallization cell (d-EMC) (Fig. 1, purple). Combined these systems comprise a non-volatile redox memory (NVRM) with unique advantages over other memory proposed for neuromorphic computing (*i.e.* phase change cells and filament forming metal oxides). For example, NVRM is capable of operating at voltages <500mV with currents <10nA. Due to low circuit parasitics and linear and symmetric programmability (Fig. 2), NVRM is capable of efficient online learning with fully-parallel inference and weight update operations. A path towards realizing a fully flexible and biocompatible neuromorphic array will be discussed.

[1] E. J. Fuller, *et al.*, *Advanced Materials*, 2017

[2] Y. van de Burgt, *et al.*, *Nature Materials*, 2017

[3] Midya, *et al.*, *Advanced Materials*, 2017

11:45am **PCSI-3WeM-40 Non-volatile Electrochemical Memory Operating near the Thermal Voltage Limit**, *Yiyang Li*, *E Fuller*, *S Agarwal*, *A Talin*, Sandia National Laboratories

Non-volatile memories like flash, phase-change, and filament forming metal oxides are desirable as synapses for neuromorphic analog computation, which can consume much less power than a digital processor. One criterion for non-volatile memory is to minimize the switching voltage, both to reduce energy consumption and to prevent dielectric breakdowns. Due to the Boltzmann distribution of electrons, it is unclear if any memory

can switch significantly below 1V; developing non-volatile memory operating near the thermal voltage limit is a grand challenge.

In this work, we use two silicon-free electrochemical devices to build non-volatile memory that operates below 300 mV (Fig. 1a). The first device is an ion insertion transistor which electrochemically shuttles lithium ions and electrons between the gate and the channel, analogous to a battery. Because the ions are mobile and move with the electrons, the process is charge neutral, and up to  $10^{21}\text{cm}^{-3}$  of electrons and ions can be reversibly shuttled between the gate and channel without electrostatic charging. The second device is a diffusive memristor operating based on  $\text{Ag}^+$  migration and filament formation in the ON state, and diffusive filament dissolution to the OFF state. The low activation energy for  $\text{Ag}^+$  motion enables the device to switch with low applied voltages. The combination of high charge density of the ion insertion transistor and low leakage current of the diffusive memristor enables these two electrochemical devices to retain memory. Because both devices operate at low voltage, we can linearly tune the electronic conductance using sub-300-mV voltage pulses (Fig. 1b). This device is also compatible with a V/2 crossbar select scheme without a select transistor, and demonstrates that non-volatile synaptic memory near the thermal voltage limit is attainable for low-power electronics.

**11:50am PCSI-3WeM-41 Simultaneous Topographical and Electrochemical Mapping using Scanning Ion Conductance Microscopy – Scanning Electrochemical Microscopy (SICM-SECM),** W Shi, Byong Kim, K Lee, Park Systems; G Mendoza, Park Systems, Mexico

Lately, scanning ion conductance microscopy (SICM), has emerged as a versatile non-contact imaging tool. To obtain spatially-resolved electrochemical information, scanning electrochemical microscopy (SECM), also known as the chemical microscope, has been developed [1]. Hybrid SICM-SECM techniques have been developed, in which the SICM compartment provides the accurate probe-sample distance control, while the SECM compartment measures the faradaic current for electrochemical information collection [2].

In this work, we demonstrate the use of an Atomic Force Microscopy (Park NX10) in combination with an ammeter for concurrent topography imaging and electrochemical mapping. The SICM-SECM probe consisted of a Au crescent electrode (AuE) on the peripheral of a nanopipette. High resolution probe-substrate distance control was obtained by the ion current feedback from SICM, while simultaneous electrochemical signal collection was achieved via the AuE from SECM. As a proof-of-concept experiment, a Au/Pyrex pattern standard sample was imaged with the SICM-SECM technique. The Au bar and the Pyrex substrate were clearly resolved from the SICM topography image, with the bar height and pitch width closely matching the actual values. In terms of the electrochemical property mapping, higher Faradaic current was seen when the probe was scanned over Au bar as a result of redox cycling, while lower Faradaic current was observed when the probe was over Pyrex substrate due to hindered diffusion. The capability of the SICM-SECM technique described here holds promise of many exciting applications in the field of electrochemistry, material science and battery research.

[1] P. K. Hansma, et al., *Science*, 1989, **243**, 641.

[2] Shi, W. et al., *Faraday discussions*, 2016, **193**, 81-97.

PCSI

Room Ballroom South - Session PCSI-1WeA

## Atomic Layer Deposition & Etching II

Moderator: Angel Yanguas-Gil, Argonne National Lab

2:00pm **PCSI-1WeA-1 Thermal Atomic Layer Etching of Silicon Using an Oxidation and "Conversion-Etch" Mechanism**, *Steven M. George, A Abdulagatov*, University of Colorado at Boulder **INVITED**

Thermal atomic layer etching (ALE) is based on sequential, self-limiting surface reactions. Thermal ALE is the reverse of atomic layer deposition (ALD). Thermal ALE has been demonstrated for many materials including  $\text{Al}_2\text{O}_3$ ,  $\text{HfO}_2$ ,  $\text{ZrO}_2$ , TiN and W. This talk will focus on thermal Si ALE using oxidation and "conversion-etch". During this process, the Si surface is oxidized to a silicon oxide layer using  $\text{O}_2$  or ozone. The silicon oxide layer is then converted to an  $\text{Al}_2\text{O}_3$  layer using trimethylaluminum (TMA) [1]. Subsequently, the  $\text{Al}_2\text{O}_3$  layer is fluorinated by HF to an  $\text{AlF}_3$  layer prior to the removal of the  $\text{AlF}_3$  layer by a ligand-exchange reaction using TMA [1]. This reaction sequence is shown in Figure 1.

Si ALE was studied using silicon-on-insulator (SOI) wafers in a warm wall reactor with a hot sample stage. *In situ* spectroscopic ellipsometry (SE) was employed to monitor the thickness of both the silicon and the silicon oxide layer during Si ALE. These studies observed that the silicon film thickness decreased linearly with number of reaction cycles while the silicon oxide thickness remained constant. Using an  $\text{O}_2$ -HF-TMA reaction sequence, the Si ALE etch rate was  $0.4 \text{ \AA}/\text{cycle}$  at  $290^\circ\text{C}$  as shown in Figure 2. Comparable etching rates were observed using ozone instead of  $\text{O}_2$  as the oxidant.

Thermal Si ALE should be useful in advanced semiconductor fabrication. Thermal Si ALE could also be utilized for atomic-scale polishing and cleaning of silicon surfaces. In addition, there may be applications in other areas such as silicon-based optoelectronics, photonics and MEMS fabrication

2:30pm **PCSI-1WeA-7 Fundamental Properties for Enhanced Etching of Ge Surfaces in Water Assisted by Single Sheets of Reduced Graphene Oxide**, *Tomoki Hirano, Y Nakata, H Yamashita, S Li, K Kawai, K Yamamura, K Arima*, Osaka University, Japan

Metal-assisted chemical etching is an emerging technology for fabricating various three-dimensional nanostructures on a semiconductor surface for future electronic and optical devices. Thus far, our group has reported that a Ge surface in contact with noble metals, including Pt and Ag, is selectively etched in  $\text{O}_2$ -containing water. However, it is extremely difficult to remove residual metallic particles on the processed Ge surfaces. To resolve this issue, graphene is used, which supposedly functions as a catalyst free from metals.

In this study, fundamental properties of graphene-assisted chemical etching, which is the preferential etching method for a Ge surface in contact with single sheets of reduced graphene oxide (rGO) in  $\text{O}_2$ -containing water were described. An rGO solution was prepared by chemical reduction of a GO solution and spin-coated on a Ge(100) surface. The sample was immersed in  $\text{O}_2$ -containing water for 24 h. We show the change in surface morphology of rGO sheet-loaded Ge upon immersion in  $\text{O}_2$ -containing water. The sheet thickness ranged from 0.8 to 1.2 nm and single rGO sheets were well dispersed after our sample preparation. Then the Ge surface was preferentially etched under the rGO sheets in  $\text{O}_2$ -containing water. This enhanced etching did not occur using water without dissolved  $\text{O}_2$  molecules, indicating that rGO-assisted chemical etching is mediated by an oxidant ( $\text{O}_2$  molecules) in a solution (water). Next, the dependence of etching rate on water temperatures was investigated. The etching rate considerably depended on water temperatures (e.g.,  $1.9 \text{ nm}/\text{h}$  at  $22^\circ\text{C}$  and  $15.1 \text{ nm}/\text{h}$  at  $48^\circ\text{C}$ ). The Arrhenius plot from this data permitted the estimation of activation energy for this etching mode.

2:35pm **PCSI-1WeA-8 Laser-patterning of Graphene Oxide Beyond the Diffraction Limit**, *Maksim Fatkullin*, Tomsk Polytechnic University, Russia

Graphene oxide (GO), same as graphene itself, attracted a tremendous attention since its re-discovery in 2004. [1]. Main advantages of GO are: water dispersible, flexible and the greatest one its tunable conductivity. Conductivity changing due to reduction process, during which dielectric GO becomes to conductive reduced graphene oxide (rGO). Reduction could be done in different ways, but all of them are either chemical or thermal [2].

The most part of reduction methods are expensive and not scalable, for example reduction using atomic force microscopy (AFM) tip or electron beam [3][4]. The other limitation of these methods is that they attracted all volume of material.

We used laser irradiation to provide external energy for the thermal annealing at the local area of GO thin film (selective reduction). From other side this method is cheap, eco-friendly and easy to perform.

Using this method, anyone can make electrical circuits of different shapes at the flexible transparent substrates with carbon nanowires, made beyond the diffraction limit without special equipment and conditions. To pattern rGO conductive lines we used different substrates and different lasers:  $\text{CO}_2$  laser with  $\lambda=1064 \text{ nm}$  and portable laser engraver with  $\lambda=405 \text{ nm}$ . Figure 1 demonstrated rGO patterning by the laser irradiation.

The key result of this contribution came out after applying the highest laser power. Even though the patterned material was completely ablated in some regions, we still observed a significant electrical conductivity along the pattern (Figure 2). This observation implies that by optimizing the laser parameters, and under higher power than has been reported previously, it is possible to achieve confined rGO regions: the edges of the patterns. Conductive rGO patterns can thus be achieved with spatial dimensions much smaller than the laser spot size.

[1] A.K. Geim, Graphene: status and prospects, *Science*. 324 (2009) 1530–1534.

[2] Songfeng Pei, Hui-Ming Cheng, *Carbon*, 50 (2012) 3210 – 3228

[3] Jeffrey M. Mativetsky et al., *Am. Chem. Soc.*, 132 (2010) 14130 –14136

[4] Gil Gonçalves et al., *Carbon*, 129 (2018) 63 - 75

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2:40pm **PCSI-1WeA-9 Maskless Si Nano-wall Formation by Wet-etching Process using a Femtosecond Laser Irradiation**, *S Lee*, Pusan National University, South Korea; *Hyun Hwi Lee*, Pohang Accelerator Laboratory, South Korea; *H Kim*, Pusan National University, South Korea

Recently, ultrashort pulsed laser direct writing technique has been intensively investigated to develop a nanoscale fabrication enabling high aspect ratio. In the referred applications, laser induced nanostructures are formed with well manipulated laser beams in small area. On the other hand, Laser Induced Periodic Surface Structure (LIPSS) has attracted increasing attention for the possibility to generate periodic nanoscale structures in large area with relatively simple optical configurations. LIPSS is a self-organized one-dimensional periodic structure which is formed when multiple laser pulses with the laser fluence near ablation threshold are exposed. These self-induced nanostructures are often categorized into Long Spatial Frequency LIPSS (LSFL) and High Spatial Frequency LIPSS (HSFL) by their periodicity. The periodicity of LIPSS is varied by the wavelength of the laser and the material properties of the substrates. The orientation of the LIPSS is affected by the laser polarization. In case of the LSFL, the orientation is observed almost perpendicular with respect to the incident laser polarization. Therefore, LIPSS with direct-write scheme has been applied in rapid production of functional surfaces on large area such as superhydrophobic surfaces, enhanced absorption, cell adhesion, and tribological applications.

However, these demonstrations utilized the LIPSS property to increase the surface area by desirable surface roughening or enhancing surface properties. Relatively less attention was paid to the morphological changes of LIPSS followed by a chemical etching.

This presentation will introduce a new mask-free patterning technique of Si, which is combined with a conventional wet-etching process and direct-write LIPSS patterns of Si. The technique is a two-step process. First, a femtosecond laser irradiates a Si surface to generate one-dimensional LIPSS pattern on the surface. Conventional wet-etching solvents, such as KOH and TMAH (tetramethylammonium hydroxide), etch the LIPSS to form a periodic micro-cell surrounded by nanowalls with the height of a few hundred nanometers. The distance between the nanowalls was approximately one micrometer and the bottom surface of the cell was atomically flat which is sufficient to grow organic semiconducting thin films for organic devices. The bottoms of micro-cells surrounded by the nanowalls were considerably flat with a 3.10 nm surface roughness. A pentacene layer was deposited on the micro-cells of a Si surface to evaluate the film properties by grazing incidence wide angle x-ray scattering measurements.

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2:45pm **PCSI-1WeA-10 Epitaxial (Bi,Sb)<sub>2</sub>Te<sub>3</sub>/Graphene/2D-Ga Heterostructures Towards Topological Superconductivity, Brian Bersch, N Briggs, J Jiang, Y Zhao, Y Chuang, C Li, Y Wang, The Pennsylvania State University; M Fu, Q Zou, Z Gai, A Li, Oak Ridge National Laboratory; M Chan, C Chang, V Crespi, J Zhu, J Robinson, The Pennsylvania State University**

The interface of a conventional BCS superconductor (SC) and a topological insulator (TI) is predicted to host an exotic state of matter known as a topological superconductor, the elemental excitations of which could potentially enable quantum computing schemes that are robust against error produced by decoherence. The direct synthesis of a SC-TI heterostructure, however, is challenging due to structural dissimilarities and high interface reactivities between common superconductors and topological insulators. Here, we report on the synthesis and properties of wafer-scale (Bi,Sb)<sub>2</sub>Te<sub>3</sub>/graphene/2D-Ga heterostructures grown in-part via a new method, i.e. confinement heteroepitaxy (Chet), pioneered by the Robinson group [1]. Chet enables us to intercalate and form atomically thin gallium (Ga) layers at the interface of 2L epitaxial graphene (Gr) and its native SiC (0001) substrate. The graphene-encapsulated 2D-Ga films are predominantly 2-3L thick, crystalline, and epitaxially registered to the SiC, as confirmed by high-resolution scanning transmission electron microscopy (HR-STEM) (Fig. 1a). The Gr/Ga films exhibit a superconducting state with zero resistance at a transition temperature of  $T_c \sim 4$  K (Fig. 1c), which is higher than that of bulk  $\alpha$ -Ga ( $T_c = 1.08$  K). In addition to serving as a capping layer for the 2D-Ga film, the Gr layer is both a reaction barrier and an ideal substrate for the subsequent molecular beam epitaxial (MBE) growth of a TI material (Bi,Sb)<sub>2</sub>Te<sub>3</sub>. HR-STEM and energy-dispersive X-ray spectroscopy (EDS) mapping shown in Fig. 1b demonstrates the atomically sharp interfaces and high-quality layer-by-layer growth of all constituent layers in a heterostructure of 6 quintuple-layer (Bi,Sb)<sub>2</sub>Te<sub>3</sub> grown on Gr/Ga. Reflection high energy electron diffraction pattern and angle-resolved photoelectron spectroscopy measurements verify its crystalline integrity and the Dirac surface bands of the TI film. First-principles calculations reveal the electronic band structure of the heterostructure, which is conducive to proximity-induced superconductivity in the TI film. Our approach circumvents several key challenges in making high-quality SC-TI heterostructures to offer a new route towards the realization of topological superconductivity.

[1] Nature Materials 15, 1166–1171 (2016)

3:00pm **PCSI-1WeA-13 Tracking Defects through Ultra-Thin Layered Complex Oxides, B.A. Noesges, The Ohio State University; J Lee, C Eom, University of Wisconsin-Madison; L Brillson, The Ohio State University**  
Improving ultra-thin films and devices requires characterization techniques capable of resolving defects at near-nanoscale depths. Defect formation in and nearby ultra-thin layers can produce effects that permeate the layer and alter interface properties. Resolving defects on this scale is especially important for complex oxide heterostructures like LaAlO<sub>3</sub>/SrTiO<sub>3</sub> where just nanometers thick layers of LAO and STO produce a highly conductive interface between the insulating materials. Interfacial majority carriers are controlled through choice of neighboring atomic planes at the interface. These interfaces are referred to as two-dimensional electron or hole gases (2DEG/2DHG) depending on carrier type since conduction is confined in-plane. The performance of the 2DEG is strongly influenced by nearby defects like oxygen vacancies which are donors and increase carrier concentration while reducing mobility. The 2DHG interface is even more strongly affected and can't form unless nearby oxygen vacancies are sufficiently suppressed [1]. We used depth-resolved cathodoluminescence spectroscopy (DRCLS) alongside advanced growth techniques designed to carefully control defect formation to systematically study the interplay between native point defects, defect complexes and interface carrier conductivity. DRCLS is an ideal technique to investigate defects in ultra-thin layers since probe depth is tunable on the near-nanoscale by adjusting voltage of an incident electron beam. With this control, cathodoluminescence can be obtained from nanometers below the film surface, through interfaces and hundreds of nanometers into the substrate providing information about the defects present in each layer. DRCLS guided by DFT calculations can identify oxygen vacancy (V<sub>o</sub>) defects as well as assorted metal cation related defects like Sr vacancies and Ti on Sr antisite (Ti<sub>Sr</sub>) defects [2]. Certain defects can suppress others, i.e., Ti<sub>Sr</sub> formation suppressing oxygen vacancy defects creating extremely clean, high mobility 2DEG interfaces. DRCLS is a unique tool for tracking defect concentration through ultrathin oxide layers to determine the role of defects in space charge regions. Growth control combined with near-nm DRCLS distribution measurements inside complex oxide heterostructures provides a powerful

approach to achieve advanced electronic features and improved device quality. Support by NSF DMR-18-00130.

[1] H.Lee, et al, Direct observation of a two-dimensional hole gas at oxide interfaces. Nature Materials 17, 231–236 (2018)

[2] D. Lee, et al, Identification of a functional point defect in SrTiO<sub>3</sub>. Phys. Rev. Materials 2, 060403\* (2018)

## PCSI

### Room Ballroom South - Session PCSI-2WeA

#### Applications of 2D Defects and Interfaces

**Moderator:** Ania Bleszynski Jayich, University of California, Santa Barbara

4:15pm **PCSI-2WeA-28 Hexagonal Boron Nitride for Quantum and Nonlinear Optics, Alexander Solntsev, I Aharonovich, University of Technology Sydney, Australia**

**INVITED**

Photonic integrated circuits that process information encoded in particles of light are poised to revolutionize information processing, communications and sensing. A promising, emerging class of quantum technologies is based on solid-state, on-demand, single photon emitters (SPEs) coupled to optical resonators and waveguides that serve as building blocks for high density, on-chip quantum photonic circuits [1]. Nevertheless, despite years of research, existing systems are inadequate for real-world applications, and there is a significant effort to find high-performance emitters hosted by materials that enable integration in photonic devices. Recently, the SPE family expanded upon the discovery of quantum emitters in two-dimensional (2D) materials [2]. These materials are atomically thin and hence offer new possibilities for scientific exploration and device engineering. Later, hexagonal boron nitride (h-BN) emerged as a compelling 2D host of SPEs offering bright single photon emission and robust operation [3].

Another important sphere of 2D material applications is nonlinear optics (NLO). Most widely used integrated photonic platforms do not possess quadratic optical nonlinearity, which significantly limits NLO applications such as wavelength conversion and all-optical switching. Integrating 2D materials with strong NLO response into photonic circuits resolves this problem [4]. Here, h-BN is particularly well positioned, since unlike other popular 2D materials, it offers both significant NLO susceptibility and transparency in the visible range. This presentation will focus the latest advances in h-BN nonlinear and quantum optics.

[1] P. Lodahl, S.Mahmoodian, & S. Stobbe, "Interfacing single photons and single quantum dots with photonic nanostructures," Reviews of Modern Physics 87, 347-400 (2015).

[2] "Single photons for all," Nature Nano., 10, 481-481 (2015).

[3] T. T. Tran, K. Bray, M. J. Ford, M. Toth, & I. Aharonovich, "Quantum emission from hexagonal boron nitride monolayers," Nature Nano. 11, 37-41 (2016).

[4] A. Autere, H. Jussila, Y. Dai, Y.Wang, H. Lipsanen, Z. Sun, "Nonlinear Optics with 2D Layered Materials," Adv. Mater. 30, 1705963 (2018).

4:45pm **PCSI-2WeA-34 Detection of Thermodynamic "Valley Noise" in Monolayer Semiconductors: Access to Intrinsic Valley Relaxation Timescales, Mateusz Goryca, National High Magnetic Field Laboratory; N Wilson, University of Washington; P Dey, National High Magnetic Field Laboratory; X Xu, University of Washington; S Crooker, National High Magnetic Field Laboratory**

The new class of atomically-thin transition-metal dichalcogenide (TMD) semiconductors such as monolayer MoS<sub>2</sub> and WSe<sub>2</sub> has focused broad attention on the concept of "valleytronics", founded on the idea of encoding information in an electron's valley degree of freedom. The key parameter for any valleytronic technology is the intrinsic timescale of an electron's inter-valley relaxation, and recent optical pump-probe studies have shown long (exceeding microseconds at low temperatures) valley relaxation of resident carriers in monolayer TMDs [1,2]. However, a significant drawback of all such experiments is that they are by design perturbative: they require optical pumping to intentionally drive the electrons out of equilibrium. Such pumping inevitably introduces so-called "dark" exciton states, whose presence can mask carriers' intrinsic valley relaxation.

Here we present a completely alternative approach, based on the idea of passively "listening" to the random spontaneous scattering of carriers between *K* and *K'* valleys in strict thermal equilibrium. We demonstrate

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that the stochastic *valley noise* is measurable by optical means and encodes the true intrinsic timescales of valley relaxation, free from any pumping or excitation [3].

Using this new fluctuation-based methodology we measure very long valley relaxation dynamics of both electrons and holes in a single electrostatically-gated monolayer of WSe<sub>2</sub>. Valley noise frequency spectra reveal long intrinsic valley relaxation with a single sub-microsecond time scale. Moreover, they validate both the relaxation times and the spectral dependence of conventional pump-probe measurements, thereby resolving concerns about the role of dark excitons and trions in studies of long-lived valley relaxation.

[1] J. Kim *et al.*, *Science Advances* **3**, e1700518 (2017).

[2] P. Dey *et al.*, *Phys. Rev. Lett.* **119**, 137401 (2017).

[3] M. Goryca *et al.*, *arXiv:1808.01319* (2018).

5:00pm **PCSI-2WeA-37 The Electronic Structure of 2D Materials, Justin Wells**, Norwegian University of Science and Technology, Norway **INVITED**

The electronic bandstructure contains complete information about the occupied electronic states which exist in a material. i.e. intrinsically it includes information on doping, Fermi velocities, confinement, the orbital nature of the bands, spin-coupling and all possible interactions. Measuring the electronic bandstructure is possible using techniques derived from photoelectron emission spectroscopy; and due to the exceptionally short probing length, this works particularly well for 2D materials and surface phenomena.

In this talk, I will describe some recent examples from our own research. In particular, I will describe how certain materials allow a smooth 3D to 2D transition, such that the role of dimensionality can be disentangled. Specifically, I will discuss the unusual phonon mediated scattering/coupling in graphene/graphite [1-3], the spin texture of monolayer transition metal dichalcogenides (TMDCs), and how the origins of this are derived from local symmetry breaking in the parent bulk compounds [4,5]. Finally, I will discuss the 3D to 2D transition in group-IV semiconductors (silicon and diamond) created as a result of a high-density dopant plane within the bulk material (see figure), and the implications of this confinement for the ultimate miniaturisation of classical and quantum devices [6-8].

[1] Mazzola *et al.*, *PRL* **111**:216806 (2013)

[2] Mazzola *et al.*, *Phys. Rev. B* **95**:075430 (2017)

[3] Hellsing *et al.*, (2018) [arxiv.org/abs/1808.08620](https://arxiv.org/abs/1808.08620)

[4] Riley *et al.*, *Nature Physics* **10**:835 (2014)

[5] Bahramy *et al.*, *Nat. Mater* **17**:21 (2018)

[6] Miwa *et al.* *PRL* **10**:136801 (2013)

[7] Cooil *et al.*, *ACS Nano* **11**:1683 (2017)

[8] Mazzola *et al.* *PRL* **120**:046403 (2018)

PCSI

Room Ballroom South - Session PCSI-ThM

## Hybrid and Mixed-Dimensional Interfaces II

Moderator: Marina Leite, University of Maryland

8:30am PCSI-ThM-1 Excitons and Exciton Confinement in Organic Heterojunctions, **Stephen Forrest**, University of Michigan **INVITED**

Excitons play a central role in photogeneration in organic detectors and solar cells. The efficiency of the exciton dissociation process is directly related to molecular structure of the donor and acceptor molecules and the film morphology. In our work, we have focused on understanding the microscopic nature of these interactions in both fullerene and non-fullerene based junctions. We have developed quantum mechanical models coupled with molecular dynamics simulations to understand the role that morphology plays on the energy of the charge transfer state – i.e. the intermediate between exciton and free polaron – and ultimately its ability to generate free charge at a low expense of energy. We find that quantum confinement of the exciton by crystalline domains in dilute donor-acceptor blends can result in substantial energy shifts in the state, thereby impacting its binding energy.[1, 2] Further, we have developed models that quantitatively relate the energy loss in non-fullerene acceptors to the molecular structure itself. Ultimately, this understanding of exciton and CT state energetics has led to very high efficiency (>15%) organic solar cells, with prospects of reaching 20% in the not distant future.[3] The models, experiments and implications of these findings will be discussed in my presentation. Finally, if there remains time, I will discuss confinement effects in novel quantum well structures comprising a combination of organic and inorganic semiconductors, including 2D solids.[4]

[1] X. Liu, K. Ding, A. Panda, and S. R. Forrest, "Charge transfer states in dilute donor-acceptor blend organic heterojunctions," *ACS Nano*, vol. 10, p. 7619, 2016.

[2] K. Ding, X. Liu, and S. R. Forrest, "Charge Transfer and Collection in Dilute Organic Donor-Acceptor Heterojunction Blends," *Nano letters*, 2018.

[3] X. Che, Y. Li, Y. Qu, and S. R. Forrest, "High Fabrication Yield Organic Tandem Photovoltaics Combining Vacuum- and Solution-Processed Subcells with 15% Efficiency," *Nature Energy*, vol. 3, p. 422, 2018.

[4] A. Panda, C. K. Renshaw, A. Oskooi, K. Lee, and S. R. Forrest, "Excited State and Charge Dynamics of Hybrid Organic/Inorganic Heterojunctions. II. Experiment," *Phys. Rev. B*, vol. 90, p. 045303, 2014.

9:10am PCSI-ThM-9 Panel Discussion II - Forrest, Leite, Ertekin,



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