PCSI

Room Redondo - Session PCSI-TuM1

Electronic and Structural Properties of Films and Surfaces

Moderator: Jia Li, Brown University

8:30am **PCSI-TuM1-1 UPGRADED: Strain Soliton Formation in Epitaxial Bismuth Thin Films***, **Jason Dong**, H. Inbar, C. Dempsey, A. Engel, C. Palmstrøm,* University of California Santa Barbara

Strain solitons are topological defects which have been observed in van der Waals materials [1]. Strain solitons are generated as a result of thin film strain and form as ripples in the films, separating commensurate and incommensurate regions. The solitons are analogous to misfit dislocations and cause local relaxation of the crystal. Solitons have been of interest due to their potential for generating novel spatially confined states within two-dimensional materials [2].

In this talk, we report the formation of strain solitons in epitaxial (111) bismuth thin films grown by molecular beam epitaxy on InSb(111)B substrates. The 0.9% lattice mismatch between bulk bismuth and the InSb(111)B substrates results in strain relaxation via strain soliton formation and in-plane edge dislocation formation. Edge dislocations are observed to induce large angle bending of the solitons. The solitons appear to form at the interface and propagate throughout the film. The strain state of the bismuth thin films is characterized by *in-situ* low temperature scanning tunneling microscopy and ex-situ X-ray diffraction. The effect of the solitons on local strain and lattice rotation is mapped from the STM images using the Lawler-Fujita algorithm [3]. Partial relaxation of bismuth is observed to begin at 2 bilayers, and the thin films become fully relaxed by 30 bilayer thick. The evolution of the soliton network and their strain fields as a function of thickness is characterized. Our results indicate that bismuth primarily relaxes by strain soliton formation and enables future investigation of novel states hosted in the bismuth solitons.

References

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8:50am **PCSI-TuM1-5 Epitaxial Growth and Surface Studies of Bi/ Hexagonal ζ- Phase Mn_2N/MgO (001) using Molecular Beam Epitaxy and Scanning Tunneling Microscopy***, **Ashok Shrestha**, A. Abbas, A. Smith,* Ohio University

The growth and structure of the cubic manganese nitrides, namely θ- MnN, η- Mn_3N_2, and ε- Mn_4N, has already been investigated intensively on MgO (001) substrates [1,2]. However, the hexagonal ζ- Mn_2N has remained unexplored. Thin films of hexagonal ζ- Mn_2N were grown successfully on MgO (001) using molecular beam epitaxy (MBE) and used as a substrate to grow a very thin layer of bismuth.

In this presentation, the epitaxial growth of ζ- Mn_2N and multilayer of bismuth will be discussed. The sample growth process was monitored by *in-situ* reflection high energy electron diffraction. RHEED shows a streaky pattern indicating the smooth sample surface throughout the sample growth. During the Mn_2N growth, 1/4th order fractional streaks are observed along $[100]_{MgO}$ + R30° direction. These fractional streaks indicate the 4× reconstruction on the Mn_2N surface. The presence of a 4× reconstructed surface is also verified by room temperature scanning tunneling microscopy. Moreover, the RT-STM images show the atomically flat terraces, steps, and the atomic resolution of the hexagonal array of the Mn_2N surface. Various *in-situ* and *ex-situ* measurements are performed to calculate the *in-plane* and *out-of-plane* lattice constants. The calculated lattice parameters are in good agreement with the theoretically reported values of ζ- Mn_2N ($a=b=$ 2.83 Å, c= 4.54 Å) [3]. Furthermore, the surface chemistry of the samples was determined by *in-situ* Auger electron spectroscopy at different locations on the sample surface. The stoichiometric ratio of Mn: N on the film is nearly 2:1 which is consistent with ζ- Mn_2N. After successfully growing ζ- Mn_2N, a very thin film of Bi is deposited, and the surface is studied by RT-STM. Interestingly, the measurements show multiple steps with a height of ~1.6 Å which is not matching the step height of bulk Bi (111) (3.94 Å) [4], but it is consistent with the step height of bismuthene grown on Ag (111) as reported by Sun *et al.* (2022) [5]. Although the step heights suggest multilayer bismuthene, atomic resolution images show a rectangular lattice.

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8:55am **PCSI-TuM1-6 One-Dimensional Spin-Polarized Surface States on Bi(112) Compared to States on Other Vicinal Surfaces of Bi***, **Anna Cecilie Åsland**, J. Bakkelund,* Norwegian University of Science and Technology (NTNU), Norway*; E. Thingstad,* University of Basel, Switzerland*; H. Røst,* Norwegian University of Science and Technology (NTNU), Norway*; S. Cooil,* University of Oslo, Norway*; J. Hu,* Norwegian University of Science and Technology (NTNU), Norway*; I. Vobornik, J. Fujii,* Instituto Officina dei Materiali (IOM)-CNR, Italy*; A. Sudbø,* Norwegian University of Science and Technology (NTNU), Norway*; J. Wells,* University of Oslo, Norway*; F. Mazzola,* Instituto Officina dei Materiali (IOM)-CNR, Italy

Vicinal surfaces of bismuth have been found to be great test-beds for investigating one-dimensional (1D) spin-polarized surface states that can be used in spintronic devices [1,2]. We used angle-resolved photoemission spectroscopy (ARPES) and spin-resolved ARPES together with tight-binding (TB) and density functional theory (DFT) calculations to investigate two such surface states on Bi(112). The surface states resemble elongated Dirac cones: 1D and close to dispersionless in the k_y direction, but dispersing in energy along the k_x-direction, forming elongated, X-like features (see Figure). The spin-polarization is pointing mainly in-plane and along the 1D lines, but there are signs of an out-of-plane component for one of the two states. In this talk, we will compare the energy vs. momentum (E vs. k) dispersion and spin-polarization of the measured surface states of Bi(112) to those calculated from the tight-binding model, and the surface states on two other vicinal surfaces of bismuth [1,2]. Similar surface states were found on all three surfaces, suggesting that their existence and general properties are robust properties of bismuth vicinal surfaces. In addition, differences in the details of the states, especially regarding spin-polarization, indicate that their properties may be tuned simply by cutting and polishing the crystal precisely along different high-symmetry directions.

9:00am **PCSI-TuM1-7 Epitaxial Growth and Electronic States of Ultrathin Bi (0001) Films on InSb (111)B***, H. Inbar, J. Dong, A. Engel,* UC Santa Barbara*; M. Zubair,* University of Delaware*; C. Dempsey, Y. Chang,* UC Santa Barbara*; A. Fedorov,* ALS-LBNL*; A. Janotti,* University of Delaware*; **Chris J. Palmstrøm**,* UC Santa Barbara

Synthesizing high-quality single-domain bismuth films with controlled strain, interface chemistries, and thin thickness remains a fundamental materials science challenge with significant consequences such as obtaining high spin-polarization in spintronic devices [1] and 1D-edge transport in the quantum spin Hall phase [2]. Tensile strain in quantum-confined (0001) bismuth films is predicted to lead to a band gap opening [3], allowing surface states to contribute mainly to transport signatures. Yet, only small-area planar Bismuthene wetting layers [4] or thick (>6 bilayers, BL) compressively strained films were studied on semiconducting substrates [5].

In this work, ultrathin large-area Bi (0001) layers were grown on InSb (111)B substrates by molecular beam epitaxy (MBE) and in vacuo transferred for synchrotron-based angle-resolved photoemission spectroscopy (ARPES). In vacuo scanning tunneling microscopy (STM) and ex situ x-ray diffraction (XRD) confirm the successful synthesis of single-crystal Bi epitaxial films. Through a core-level photoemission study, we show that large-area single-domain ultrathin Bi films can be stabilized through strong film-substrate interactions. With decreasing film thickness from 200 BL to 1 BL, we quantify the confinement-induced shifts in the bulk band structure and trace the quantum well energy levels with a phase accumulation model. Significant film-substrate bonds breaking inversion symmetry affect the surface state dispersion leading to a surface state degeneracy which allows us to assign the topological order in Bi (0001) thin films. The findings of this study offer a new route for epitaxial growth and integration of band-engineered Bi films with III-V substrates.

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9:05am **PCSI-TuM1-8 Thermal Stability and Sn Segregation in GeSn Structures***, **Hryhorii Stanchu,*** Institute for Nanoscience and Engineering, Department of Electrical Engineering, University of Arkansas*; O. Olorunsola, A. Kuchuk, A. Said, F. de Oliveira, Y. Mazur, M. Benamara, S. Yu, G. Salamo,* University of Arkansas

Group IV GeSn alloys are extensively studied for efficient Si-based photonics. The interest is elevated recently due to the demonstration of direct bandgap emission in Ge [1]. Specifically, a combination of strain and *n*-type doping reduces the small separation (~140 meV) between the indirect (L) and direct (Γ) valleys in Ge [2] and raises the Fermi level for sufficient electron population in the Γ valley. Nonetheless, the high level of strain (2%) and doping (10^{19} cm^{-3}) only allows a tunable Ge bandgap between 1.3 and 1.6 µm. Alternatively, alloying Ge with the negative bandgap α-Sn results in a fundamental direct bandgap GeSn material with better tunability.The theoretical and experimental estimations indicate an indirect to direct bandgap transition for Sn compositions above 6%.

Achieving Sn-rich GeSn alloys is difficult. This relates to (i) the low equilibrium solid solubility of Sn in Ge (<1%) and (ii) the large lattice mismatch between Ge and α-Sn (15%). However, Sn-rich GeSn alloys have been demonstrated by non-equilibrium growth techniques. The thermal stability of such structures is an issue. We study the structural and optical properties of CVD grown GeSn films in detail. The strain relaxation and Sn segregation is reported for thermal treatment at 300 °C. We discuss on the mechanism of Sn segregation trough the interaction with dislocation.

9:10am **PCSI-TuM1-9 Growth of Ge$_{1-x}$Sn$_x$ Heteroepitaxial Layers with High Sn Content on InAs (001) Substrate***, **Nirosh M Eldose,*** Nanoscale Material Science Engineering, University Of Arkansas*; H. Stanchu,* Department of Electrical Engineering, University Of Arkansas*; C. Gunder, C. Li, F. Maia de Oliveira, Y. Mazur, M. Benamara,* Nanoscale Material Science Engineering, University Of Arkansas*; S. Yu,* Department of Electrical Engineering*; G. J. Salamo,* Nanoscale Material Science Engineering, University Of Arkansas

Group IV alloys of Ge and Sn (Ge$_{1-x}$Sn$_x$) are extensively studied for various electronic and optoelectronic applications on a Si platform. Alloying GeSn with α-Sn concentrations as low as 6% [1] allows for an indirect to direct optical transition compared to Ge. Higher Sn content makes possible mid and even long-range infrared optical emission and detection [2,3]. At the same time, due to the low solid solubility of Sn in Ge (~1%) as well as the large lattice mismatch of α-Sn with Ge (~14%), the realization of Sn-rich Ge$_{1-x}$Sn$_x$ structures is challenging.

We will discuss the growth and investigation of crystalline Ge$_{1-x}$Sn$_x$ heteroepitaxial layers with Sn percentage higher than 14% on InAs (001) substrates. Utilizing the small lattice mismatch between Ge$_{1-x}$Sn$_x$ layers and the InAs (001) substrate, we achieved epitaxial growth of Ge$_{1-x}$Sn$_x$ with a high Sn percentage (>30%) by applying a high Sn gradient during growth. The strain induced in the GeSn layers was investigated and the effect of gradually increasing the Sn content of the GeSn lattice significantly affected the composition and morphology of the GeSn layers. Supporting data (Figs 1-3) on the structural properties of Ge$_{1-x}$Sn$_x$alloys using X-ray diffraction, SIMS analysis, XTEM and Raman spectra will be discussed and are shown below.

9:15am **PCSI-TuM1-10 Black Phosphorus/GaAs Heterojunctions for Infrared Detection***, **Emma Renteria,*** University of New Mexico*; S. Addamane,* Center for Integrated Nanotechnologies, Sandia National Laboratories*; T. Rotter, G. Balakrishnan, F. Cavallo,* University of New Mexico

We report our recent results on the development of ultra-thin black Phosphorus (bP)/GaAs junction photodiodes for infrared (IR) detection. The devices comprise an epitaxially grown n-type GaAs nanomembrane and a p-type bP flake exfoliated from commercially available bulk crystals. bP is a promising material for the realization of visible-to-mid-IR detectors as its direct bandgap can be tuned from 0.3 eV to 2 eV by varying its thickness at the nanoscale [1]. The GaAs nanomembrane is multifunctional in that it serves as the n-side of the IR junction photodiode, and it is expected to shield the device from radio-frequency (RF) waves as it has been heavily doped. The fabricated devices have a strong potential to implement RF-waves hardened electronic sensing of IR radiation, a capability of tremendous interest for military and commercial applications. An additional value of the GaAs/bP ultra-thin photodiodes is that they can conform to non-planar surfaces such as aircraft windows and domes of IR cameras, and they can be readily integrated into soldiers' uniforms.

We have established a process to integrate large-area bP flakes onto 220 nm-thick GaAs nanomembranes and used conventional top-down photolithography to fabricate bP-GaAs p-n diodes with areas up to 1.27 x 10^{-4} cm^2. Measured dark-current-voltage characteristics showed a turn-on voltage of 1.4 V with a maximum forward current density of ~ 90 A/cm^2 at 2 V and a dark current density of ~ 2 A/cm^2 at -2 V. We correlate the materials and devices parameters extracted from the current-voltage characteristics with electronic band-structure diagrams and the structure of surfaces and interfaces probed by electron microscopy and x-ray photoelectron spectroscopy. In this talk, we will show the geometry of the bP/GaAs photodiode and the physical-chemical structure of surfaces and interfaces at the nanoscale determine the dominant transport mechanisms and the performance parameters of the device.

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9:20am **PCSI-TuM1-11 UPGRADED: Sub-Nanoscale Chemical Analysis with Nano-Confined Localized Surface Plasmons***, **Nan Jiang,*** University of Illinois - Chicago

It is of significant importance but a challenging task to probe how local chemical environments affect single-molecule behaviors with angstrom scale resolution. Tip-Enhanced Raman Spectroscopy (TERS) affords the spatial resolution of traditional Scanning Tunneling Microscopy (STM) while collecting the chemical information provided by Raman spectroscopy. By using a plasmonically-active probe tip, the Raman signal at the tip-sample junction is incredibly enhanced, allowing for single-molecule probing. This method, further aided by the benefits of ultrahigh vacuum, is uniquely capable of controlling localized plasmons via an atomistic approach. We are able to obtain (1) single-molecule chemical identification;[1] (2) adsorbate-substrate interactions of individual molecules on the surface;[2] (3) local strain effects in 2D heterostructures;[3] (4) atomic-level insights into surface oxidation reactions.[4] By investigating substrate structures, molecular superstructures, 2D material lattices, and adsorption geometries obtained from vibrational modes, we extract novel surface-chemistry information at an unprecedented spatial (< 1 nm) and energy (< 10 wavenumber) resolution. Another important application of localized surface plasmons is to achieve site-selective chemical reactions at sub-molecular scale. Recently, we selectively and precisely activated multiple chemically equivalent reactive sites one by one by scanning probe microscopy tip-controlled plasmonic resonance.[5] Our method can interrogate the mechanisms of forming and breaking chemical bonds at the angstrom scale in various chemical environments, which is critical in designing new atom- and energy-efficient materials and molecular assemblies with tailored chemical properties.

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9:40am **PCSI-TuM1-15 Light Induced Surface Tension Gradients for Hierarchical Assembly of Particles from Liquid Metals***, **Jiayun Liang**, Z. Al Balushi,* University of California at Berkeley

Achieving control over the motion of dissolved particles in liquid metals is of importance for the meticulous realization of hierarchical particle assemblies in a variety of nanofabrication processes. Brownian forces can impede the motion of such particles, impacting the degree of perfection that can be realized in assembled structures. Here we show that light induced Marangoni flow in liquid metals (i.e., liquid-gallium) with Laguerre-gaussian (LG_{pl}) lasers as heating sources, is an effective approach to overcome Brownian forces on particles, giving rise to predictable assemblies with high degree of order. We show that by carefully engineering surface tension gradients in liquid-gallium using non-gaussian LG_{pl} lasers, the Marangoni and convective flow that develops in the fluid drives the trajectory of randomly dispersed particles to assemble into 100-µm wide ring-shaped particle assemblies. Careful control over the parameters of the LG_{pl}laser (i.e., laser mode, spot size, and intensity of the electric field) can tune the temperature and fluid dynamics of the liquid-gallium as well as the balance of forces on the particle. This in turn can tune the structure of the ring-shaped particle assembly with a high degree of fidelity. The use of light to control the motion of particles in liquid metals represents a tunable and rapidly reconfigurable approach to spatially design surface tension gradients in fluids for more complex assembly of particles and small-scale solutes. This work can be extended to a variety of liquid-metals, complementary to what has been realized in particle assembly out of ferrofluids using magnetic fields.

9:45am **PCSI-TuM1-16 Charge Transport in $SrTiO_3$:Rh and $BiVO_4$ Nanoparticle Photocatalysts for Z-scheme Water Splitting***, **Brian Zutter**,* Sandia National Laboratories*; Z. Chen,* University of California Irvine*; L. Barrera,* University of Michigan, Ann Arbor*; A. Lapp, A. Bhandarkar,* Sandia National Laboratories*; K. Watanabe, A. Kudo,* Tokyo University of Science, Japan*; D. Esposito,* Columbia University*; R. Chandran,* University of Michigan, Ann Arbor*; S. Ardo,* University of California Irvine*; A. Talin,* Sandia National Laboratories

Solar-powered water splitting using nanoparticle photocatalyst suspensions is a promising route to economical, clean hydrogen production. In the Z-scheme approach, hydrogen and oxygen-evolving photocatalysts, such as $SrTiO_3$:Rh and $BiVO_4$, are coupled with a redox mediator to improve light absorption compared to single-photocatalyst systems. A key step in the water-splitting process is the separation and transport of photo-excited electrons and holes to the photocatalyst surface. Here we characterize charge transport in individual $SrTiO_3$:Rh and $BiVO_4$ nanoparticles using a nanoprobe within a scanning electron microscope, and directly map internal electric fields with electron-beam induced current. Charge transport in $SrTiO_3$:Rh particles is limited by bulk defect states within the nanoparticle, in contrast to nearly Ohmic conduction in $BiVO_4$ nanoparticles. $SrTiO_3$:Rh particles contain insignificant built-in E fields, while $BiVO_4$ nanoparticles contain built-in E field between different facets of the nanoparticle which can efficiently separate e-h pairs. Inefficient charge transport and lack of built-in electric field explain why the H_2-evolving $SrTiO_3$:Rh nanoparticles are the limiting component within this Z-scheme system.

9:50am **PCSI-TuM1-17 Improvement of Thermal Stability of Ultrathin NiAl Films***, **Kyeong Youn Song**,* Sungkyunkwan University, Republic of Korea

The current copper interconnect technology beyond the 7nm node is facing a crucial challenge of rapidly increasing metal line resistance as the downscaling of device dimensions [1]. In a narrow metal linewidth, the electrical resistivity of Cu increases rapidly due to its relatively high electron mean free path increasing the surface electron scattering. Recent studies have disclosed that binary intermetallic compounds such as NiAl have a high potential to replace Cu due to their low resistivity and high reliability at small dimensions [2]. The critical drawback of NiAl, however, is its rapid increase of resistance upon post-annealing due to the formation of surface oxide and subsequent composition shift of the compound [3].

In this talk, we propose the employment of an aluminum oxide capping layerwith thickness varied from 1-5 nm to improve the thermal stability of ultrathin NiAl films. We found that employing a capping layer effectively prevented the Al out-diffusion from NiAl to aluminum oxide thus the compositional change was no occurred during thermal annealing. Thickness-dependent resistivity of NiAl films was measured, and the NiAl film with 1-nm-thick capping layer shows remarkably low resistivity (49.7mW·cm) for 3.2 nm NiAl film after annealing at 450°C.

9:55am **PCSI-TuM1-18 Frequency-Dependent Conductivity of Granular Metals***, **Laura Biedermann**, M. McGarry, S. Gilbert, W. Bachman, J. Flicker,* Sandia National Laboratories*; P. Kotula,* Sandia*; M. Siegal,* Sandia National Laboratories

We explore the frequency-dependent conductivity, σ, of granular metals (GMs). Granular metal comprise nanoscale metal islands embedded in a dielectric matrix. We target the metal volume fraction, ϕ, so that the few-nm diameter metal islands are separated by a ~1 nm dielectric barrier. These metal/insulator composites can be modelled as a complex resistor-capacitor network with complementary tunneling and capacitive conduction paths [1]. At low frequencies, ideal GMs are highly insulating with thermally-assisted tunneling between islands occurring at high E-fields. At high frequencies, capacitive transport dominates; conductivity increases orders of magnitude.

We compare M-SiN_x and M-YSZ (M = Mo or Co; YSZ = yttria-stabilized-zirconia) GM thin films grown via rf co-sputtering on sapphire. Frequency-dependent conductivity is evaluated using impedance spectroscopy at ambient and cryogenic temperatures. Temperature and field-dependent DC conductivity provide insight into tunneling mechanisms. Granular metal structure (Fig. 1a, b) and composition are determined by scanning transmission electron microscopy (STEM) and x-ray photoemission spectroscopy (XPS).

Through careful selection of the metal-insulator system and synthesis optimization, we prepared GM films having σ_{MHz}/σ_{DC} >10^5 (Fig. 1c). This σ_{MHz}/σ_{DC} ratio was achieved for Mo-SiN_x using deposition conditions that significantly reduced the vacancies present in the sputtered insulator. Analysis of metal/insulator interfaces via XPS showed metal-oxide formation in M-YSZ and metal-silicide formation in M-SiN_x [2]. Control of deposition conditions can minimize these defects, allowing σ_{MHz}/σ_{DC} optimization for high-pass filters.

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Author Index

Bold page numbers indicate presenter