Nanoscale Science and Technology
Room 320 - Session NS-TuA

Multimodal Techniques in Surface and Interface Engineering at the Nanoscale II

Moderators: Dr. Son Le, University of Maryland, **Dr. Taisuke Ohta**, Sandia National Laboratories

2:15pm **NS-TuA-1 Multimodal Investigation and Interface Engineering of Near-Surface Diamond Quantum Defects***, Nazar Delegan,* Argonne National Laboratory **INVITED**

Solid-state spin defects, such as the nitrogen-vacancy (NV) center in diamond, are premier candidates for quantum sensing and information processing due to their long spin coherence times and robust room-temperature operation. However, when brought within nanometers of the surface to maximize coupling to external targets, their quantum properties degrade due to interface defects, uncontrolled surface terminations, and parasitic "dark" electron spins. Overcoming these challenges requires a multimodal approach that pairs sub-nanometer characterization with precise chemical control of the interface.

In this talk, we address this problem by leveraging atomic layer etching (ALE) and atomic layer deposition (ALD) as a dual framework for highly sensitive surface diagnostics and precise interface engineering. We show that sub-angstrom ALD nucleation profiles serve as an analytical probe capable of distinguishing between common diamond (001) surface terminations. Expanding on this capability, we deploy ALD-grown ultrathin titanium oxide (TiO_2) films to passivate the diamond interface, drastically reducing the background density of deleterious dark surface spins.

This controlled processing allows for the realization of pristine diamond surfaces. Enabled by this approach, we present direct atomic-scale imaging, spectroscopic characterization, and charge-state manipulation of individual near-surface NV centers using scanning tunneling microscopy (STM). By utilizing a conductive graphene capping layer to circumvent diamond's bulk insulating nature, we identify clear spectroscopic signatures of NV- defects. Critically, we demonstrate local control over the defect's electronic environment, dynamically tuning the charge state from NV^- to NV^0 via tip-induced gating. Together, these complementary nanoscale imaging and conformal engineering paradigms offer a robust toolkit for mitigating surface noise and optimizing diamond-based quantum architectures.

2:45pm **NS-TuA-3 Multi-Technique Nanoscale Probe Studies of Intercalated Phases of Pb, Gd and Dy Under Graphene on SiC***, Shen Chen, Salma Khatun, Umamahesh Thupakula,* Ames National Laboratory*; Zhe Fei,* Iowa State University*; Marek Kolmer,* Ames National Laboratory*; Michael C. Tringides,* Iowa State University

Metal intercalation beneath epitaxial graphene on SiC (Gr/SiC) can host different subsurface phases, which is a way to control the band structure, subsurface superstructure, metal nucleation and charge transfer. Realizing and controlling these subsurface phases requires identifying the galleries occupied by the intercalants and the types of intercalated phases formed. Here, we use complementary techniques, including STM/STS, SPA-LEED, and ARPES to address the previous questions. Among Pb-intercalated phases, $(10\times10)_{gr}$ is of particular interest because the large supercell generates replica Dirac cones at the corners of mini-Brillouin zones. However, establishing reproducible growth conditions for the $(10\times10)_{gr}$ phase remains challenging. By monitoring deposition and annealing cycles, we quantitatively show that during growth the $(10\times10)_{gr}$ and Pb(10) intensities are correlated, suggesting that Pb(111) islands preferentially nucleate on $(10\times10)_{gr}$ domains[1]. For Gd intercalation, multiple phases with different gallery occupancies form, depending on growth conditions of coverage and annealing temperature, all of which have been comprehensively studied with the previously listed techniques. Multiprobe STM transport confirm the decoupling of the top graphene by the large displacement field generated by intercalated Gd[2]. Recent experiments show the formation of pre-designed Gd intercalated phases by kinetically controlling the Gd subsurface occupation[3]. Dy [4] intercalation shares similar intercalated phases and growth as Gd and both metals are ideal metals to control graphene magnetism. These experiments show that metal intercalation is a very promising approach for further engineering graphene's exceptional electronic properties. They establish a framework to identify metal intercalated phases in Gr/SiC heterostructures and to generate 2-d quantum materials with tunable electronic properties.

[1] S. Chen, M. Kolmer, L. Wang, Y. Han, M. Blank, J.W. Evans, M.C. Tringides, *Phys. Rev. B*. **112**, 045418 (2025).

[2] M. Kolmer, W. Ko, J. Hall, S. Chen, J. Zhang, H. Zhao, L. Ke, C.-Z. Wang, A.-P. Li, M.C. Tringides, *J. Phys. Chem. Lett.* **13**, 11571 (2022).

[3] S. Khatun, S. Chen, J. Hall, M. Fralaide, U. Thupakula, M. Hupalo, Y. Han, Z. Fei, M.C. Tringides, M. Kolmer, *Carbon*. 121670 (2026).

[4] S. Chen, Y. Han, M. Kolmer, J. Hall, M. Hupalo, J.W. Evans, M.C. Tringides, *Phys. Rev. B*. **107**, 045408 (2023).

3:00pm **NS-TuA-4 Multidimensional Characterizations of Pyrrolidine Dissociations on Cu(100) Surface with the Single-Bond Resolution***, Liya Bi, Hao Zhou, Zihao Wang, Shaowei Li,* University of California San Diego

Here, we report the development of a multidimensional characterization platform for identifying surface adsorbents with the single-bond resolution. This platform is based on low-temperature scanning tunneling microscopy, encompassing the bond-resolved STM (BR-STM), STM-inelastic electron tunneling spectroscopy (STM-IETS) and infrared-integrated STM (IRiSTM). We demonstrated both its chemical sensitivity and spatial resolution through fully characterizing the dissociation products from a pyrrolidine (C_4H_9N) to a dehydrogenated pyrrole (C_4H_4N) on Cu(100) surfaces. We unambiguously assigned a series of products after step-by-step dehydrogenation to dehydrogenated pyrrolidine (C_4H_8N), 1-pyrroline (C_4H_7N), dehydrogenated 2-pyrroline (C_4H_6N), 3H-pyrrole (C_4H_5N) and dehydrogenated 1H-pyrrole (C_4H_4N), which was supported by bond-resolved topographic imaging and/or IETS mapping and/or single-molecule IR spectroscopy. Our experimental scheme is expected to aid in chemical identification in on-surface synthesis or catalysis with both structurally and vibrationally sensitive probes.

3:15pm **NS-TuA-5 Linking Sers Spectral Variability to Reliable Detection of Low-Affinity Analytes via Plasmonic Substrate Physics and Machine Learning***, Amit Kumar, Fengbo MA,* University of Georgia*; Susu M. Zughaier,* Qatar University, Qatar*; Xianyan Chen, Yiping Zhao,* University of Georgia

Surface-enhanced Raman spectroscopy (SERS) provides ultrasensitive molecular detection through localized plasmonic field enhancement; however, reliable sensing of low-affinity analytes remains limited by spectral variability arising from heterogeneous electromagnetic enhancement and adsorption-orientation-dependent Raman tensor projections. In this work, we establish a physics-informed and machine-learning-assisted framework linking SERS spectral variability with sensing reliability on oblique-angle-deposited silver nanorod (AgNR) substrates. A comprehensive SERS dataset was constructed using 1,2-bis(4-pyridyl) ethylene (BPE) under six controlled experimental conditions, including defect mapping, batch-to-batch variation, nanorod-length variation (200-1000 nm), concentration-dependent drop-casting (10^{-3}-10^{-12} M), static immersion (10^{-3}-10^{-10} M), and real-time adsorption (10^{-5} M). Hierarchical cluster analysis (HCA) partitioned the spectra into seven reproducible spectral clusters spanning high- and low-SNR regimes, while principal component analysis (PCA) revealed that cluster separation originates from systematic redistribution of Raman modes rather than stochastic noise. The results show that geometry-dependent plasmonic enhancement redistributes Raman peak intensities through electromagnetic weighting, while tensor-based orientation analysis demonstrates orientation-selective enhancement governed by Raman polarizability anisotropy. Building on this spectral variability framework, HCA and PCA were further applied to concentration-dependent bacterial biomarker datasets to identify distinct spectral evolution regimes associated with adsorption affinity and concentration variation. Strongly adsorbing biomarkers, including 2,3-DHBA, 2,5-DHBA, and pyocyanin, produced stable high-intensity spectra, whereas weakly adsorbing biomarkers, including lipoteichoic acid (LTA), enterobactin, and β-carotene, exhibited weak enhancement and non-monotonic spectral evolution that limited conventional calibration-based quantification. Using HCA/PCA-derived spectral patterns, convolutional neural network (CNN) classification and regression models were implemented directly on the SERS spectra. The CNN classification model achieved 99.99% classification accuracy, while regression models yielded concentration prediction performance exceeding R^2 >0.97 with MAE <0.27 despite substantial spectral variability. These results establish SERS spectral variability as a physically interpretable phenomenon governed by coupled plasmonic and molecular-orientation effects and demonstrate a scalable framework for robust nanoscale sensing in complex chemical and biological environments.

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