Wednesday Morning, October 23, 2019

Nanometer-scale Science and Technology Division
Room A222 - Session NS-WeM

Optics and Scattering on the Nanoscale

Moderators: Alex Belianinov, Oak Ridge National Laboratory, Nancy Burnham, Worcester Polytechnic Institute

8:00am **NS-WeM-1 Semiconductor Nanowires for Optoelectronics Applications***, **Chennupati Jagadish**[1] ,* Australian National University, Australia **INVITED**

Semiconductors have played an important role in the development of information and communications technology, solar cells, solid state lighting. Nanowires are considered as building blocks for the next generation electronics and optoelectronics. In this talk, I will introduce the importance of nanowires and their potential applications and discuss about how these nanowires can be synthesized and how the shape, size and composition of the nanowires influence their structural and optical properties. I will present results on axial and radial heterostructures and how one can engineer the optical properties to obtain high performance lasers, THz detectors and solar cells. Future prospects of the semiconductor nanowires will be discussed.

8:40am **NS-WeM-3 Photonic-Plasmonic Fiber Probe for Nanoscale Chemical Imaging***, B Birmingham, K Minn, B Ko, H Lee, **Zhenrong Zhang**,* Baylor University

Probing light-matter interaction in nanoscale regime requires the efficient delivery and collection of electromagnetic energy to and from the nanoscale region of interest. Metallic plasmonic nano-probes can efficiently excite and detect the near-field at nanoscale for near-field imaging and sensing applications such as tip-enhanced Raman spectroscopy (TERS). We have studied the interaction of molecules with bulk MoS_2, a semiconductor, using TERS. MoS_2, has attracted tremendous attention due to promising applications in electronics, photonics, and catalysis. Here we have compared the difference in the interaction of sub-monolayer copper phythalocyanine (CuPc) molecules with MoS_2 and Au. The relative Raman peak ratio and Raman peak position shift from spatial TERS mapping show the difference in the adsorbates-adsorbates interaction and the adsorbates-substrates interaction on Au and MoS_2 substrates.

We also propose a photonic-plasmonic probe for nanoscale confinement of light. In our device, light in a fiber couples with the surface plasmons of a nano-antenna. The coupled plasmonic mode then propagates down the conical waveguide to the narrow apex where it gets localized and strongly focused, exhibiting immense field enhancement. By changing the structures at the fiber-antenna interface, the linearly polarized fiber mode is converted to radial surface plasmon polaritons (SPP's) through asymmetric coupling. The probe can be implemented into TERS setup to obtain spectroscopic information at the nanoscale.

9:00am **NS-WeM-4 Nanoscale Infrared Confinement Using Surface Phonon Polaritons***, **Vanessa Breslin**, A Grafton,* National Research Council Postdoctoral Fellow*; D Ratchford, A Giles, K Fears, C So, S Katzer, C Ellis, J Tischler,* U.S. Naval Research Laboratory*; J Caldwell,* Vanderbilt University*; A Dunkelberger, J Owrutsky,* U.S. Naval Research Laboratory

Plasmonic materials can be used for surface enhanced infrared absorption, a particularly useful technique for chemical sensing applications, but these materials typically suffer from high optical loss due to the fast scattering of electrons, which results in broad optical resonances. In contrast, surface phonon polaritons (SPhPs) have much lower losses because of the slower scattering rates of phonons, resulting in narrower resonance bands. In particular, our group is investigating polar dielectric inorganic crystals that have mid-IR Reststrahlen bands, frequency ranges where the crystals' optical constants resemble metals and can support SPhP resonances. Currently, our efforts are focused on studying the optical properties of $W(CO)_6$ and calcite ($CaCO_3$) crystals in the mid-IR. We are also experimenting with using a helium ion microscope, a gallium focused ion beam, and other lithographic techniques to nanostructure the surface of these polar dielectric materials in order to generate SPhPs for sub-diffraction optical confinement of mid-IR incident light. The results of these studies will allow us to better understand how to tune SPhPs in a broader spectral range with different inorganic materials and provide a basis for exploring how these resonances interact with other chemical systems through enhanced spectroscopies and energy transfer.

9:20am **NS-WeM-5 Actuating and Probing a Single-molecule Switch at Femtosecond Timescales***, D Peller, L Kastner, T Buchner, C Roelcke, F Albrecht, R Huber, **Jascha Repp**,* University of Regensburg, Germany **INVITED**

Accessing ultra-fast non-equilibrium phenomena is enabled by terahertz (THz) scanning tunneling microscopy [1] (THz-STM) through combining STM with lightwave electronics. In THz-STM, the electric field of a phase-stable single-cycle THz waveform acts as a transient bias voltage across an STM junction. These voltage transients may result in a net current that can be detected by time-integrating electronics. The recent development of this lightwave STM has enabled the combined femtosecond and sub-angstrom resolution in observing matter [2].

We now demonstrate the first combined femtosecond and sub-angstrom access in the control of matter. Ultrafast localized electric fields in lightwave STM enable exerting atom-scale femtosecond forces to selected atoms. By shaping atomic forces on the intrinsic timescale of molecules, coherent atomic motion can now be excited. Utilizing this coherent structural dynamics, we can modulate the quantum transitions of a single-molecule switch by up to 39%. We directly visualize the coherent excitation of the switch in the first femtosecond single-molecule movie [3].

To resolve the impact of coherent control of the single-molecule switch, alongside, we introduce single-shot action spectroscopy in lightwave STM as the first concept resolving individual path-selective reaction events of a single molecule in space and time. With this novel concept, we detect the outcome of every single laser shot and further separate the statistics of the two inverse reaction paths.

Our results open a new chapter in the control and observation of reactions of individual molecules directly on the relevant ultrafast and ultrasmall scales.

References:

[1] T. L. Cocker et al., Nature Photon. 7, 620 (2013).

[2] T. L. Cocker et al., Nature 539, 263 (2016).

[3] D. Peller et al., in preparation.

11:00am **NS-WeM-10 Nanoscale Structural Imaging through Bragg Diffraction Microscopy***, **Martin Holt**,* Argonne National Laboratory **INVITED**

The development of x-ray nanobeam instrumentation at synchrotron x-ray light sources has created a wide range of opportunities in understanding nanoscale phenomena in materials science, chemistry, and condensed matter physics. Such instruments, including the Center for Nanoscale Materials/Advanced Photon Source Hard X-ray Nanoprobe (HXN) employ highly brilliant x-ray beams with focal spot sizes on the order of tens of nanometers and sufficient phase-coherent intensity to produce high-dynamic range scattering patterns from individual nanoscale objects. The far higher brilliance resulting from the APS Upgrade project promises to enable new classes of nanodiffraction experiment and to bring new challenges in the management and predictive analytic interpretation of large coherent scattering datasets. The scientific use of these instruments has required the creation of advanced x-ray analysis techniques based on combinations of coherent diffraction and ptychography with the unique optical conditions of tightly focused x-ray beams enabling correlation of structural and chemical mapping. The classes of scientific questions that are addressable by these techniques and the potential impact of diffraction limited storage rings such as the APS-Upgrade project will be explored within the context of recent results.

11:40am **NS-WeM-12 First Launch of XTIP - The World's First User Program for the Combination of Scanning Tunneling Microscopy with Synchrotron Radiation***, **Volker Rose**, N Shirato, D Rosenmann, M Fisher, S Hla,* Argonne National Laboratory

The combination of the ultimate spatial resolution of scanning probe microscopy with the chemical and magnetic sensitivity of synchrotron x-rays has opened the prospect for an entirely new way of nanoscale materials' characterization. Over the last couple of years, Argonne National Laboratory has pioneered the development of synchrotron x-ray scanning tunneling microscopy (SX-STM). The technique has demonstrated imaging with direct elemental contrast down to the level of single atom height as well as imaging of nanoscale magnetic domains of thin films.

In order to open up this new capability to the entire science community, and to fully exploit the special capabilities of the technique, XTIP, a dedicated beamline for SX-STM has been construction at Argonne's

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[1] **NSTD Recognition Award**

Advanced Photon Source. To meet the scientific objective of the nanoscience and nanomagnetism communities most effectively, XTIP offers full polarization control over the 500-1600 eV photon energy range. The dedicated XTIP beamline, inaugurated in the summer of 2019, provides researchers access to a one-of-a-kind instrument. Among the potential breakthroughs are “designer” materials created from controlled assemblies of atoms and molecules, and the emergence of entirely new phenomena in chemistry and physics. The easy process for obtaining access to the XTIP beamline will also be covered.

This work was performed at the Advanced Photon Source and the Center for Nanoscale Materials, a U.S. Department of Energy Office of Science User Facility under Contract No. DE-AC02-06CH11357.

12:00pm **NS-WeM-13 Application of Scanning Tunneling Microscopy and Tip-Enhanced Raman Spectroscopy to the Study of Intermolecular and Molecule-Substrate Interactions***, **Jeremy Schultz**[1], N Jiang,* University of Illinois at Chicago

Molecular self-assembly on surfaces is defined by the unique set of circumstances that arise from the complicated interplay of molecule-molecule and molecule-substrate interactions. These interactions are defined by their highly localized chemical environments. As a result, it becomes necessary to apply spatially resolved techniques. In this work we have applied two primary techniques to the study of intermolecular and molecule-substrate interactions. Scanning tunneling microscopy (STM) reveals local electronic effects and structure, while tip-enhanced Raman spectroscopy (TERS) defines the vibrational fingerprint of a molecule which is highly sensitive to localized chemical effects. In combination with gas phase Density Function Theory (DFT) calculations it is possible to define the effects of molecule-substrate interactions on the molecules’ vibrations. Three different systems involving organic molecules on single crystals have been examined: boron subpthalocyanine, 3,6-dibromo-9,10-phenanthrenequinone, and rubrene. Through the tandem technique of STM-TERS, intermolecular interactions that result in self-assembly, specifically hydrogen bonds, halogen bonds, and van der Waals interactions have been characterized. Similarly, molecule-substrate effects on molecular configuration and binding strength have been considered through comparison with DFT simulated Raman spectra to obtain a detailed description. Ultimately, the application of complementary techniques results in highly descriptive vibrational fingerprints with spatial resolution.

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[1] **NSTD Graduate Student Award Finalist**

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