

Thursday Afternoon, October 25, 2018

2D Materials Focus Topic

Room 201B - Session 2D+EM+MN+NS-ThA

Novel Quantum Phenomena in 2D Materials

Moderator: Hsin Lin, Institute of Physics, Academia Sinica

2:20pm **2D+EM+MN+NS-ThA-1 Double Indirect Interlayer Exciton in a MoSe₂/WSe₂ van der Waals Heterostructure, Aubrey Hanbicki, H Chuang, M Rosenberger, C Hellberg, S Sivaram, K McCreary, I Mazin, B Jonker, Naval Research Laboratory**

Tailoring semiconductor heterostructures for specific functionalities has led to varied opto-electronic devices including solar cells, photodetectors, light-emitting diodes and lasers. An emerging class of heterostructures involves monolayer semiconductors such as many of the transition metal dichalcogenides (TMDs) which can be combined to form van der Waals heterostructures (vdWHs). vdWHs offer novel functionalities making them promising hosts for future devices. One unique new heterostructure property is an interlayer exciton (ILE), a spatially indirect, bound electron-hole pair with the electron in one TMD layer and the hole in the other. Here, using state-of-the-art preparation techniques, we are able to resolve emission from the ILE in a MoSe₂/WSe₂ heterostructure into two distinct peaks separated by 24 meV at zero field. These peaks have nearly equal intensity, indicating they are of common character, and have *opposite* circular polarizations when excited with circularly polarized light. *Ab initio* calculations successfully account for these observations – they show that both emission features originate from excitonic transitions that are indirect in momentum space and are split by spin-orbit coupling. Also, the electron is strongly hybridized between both the MoSe₂ and WSe₂ layers, with significant weight in both layers, contrary to the commonly assumed model. Thus, the transitions are not purely interlayer in character. This work represents a significant advance in our understanding of the static and dynamic properties of TMD heterostructures.

This research was performed while H.-J.C. held an American Society for Engineering Education fellowship and M.R.R and S.V.S held a National Research Council fellowship at NRL. This work was supported by core programs at NRL and the NRL Nanoscience Institute. This work was also supported in part by a grant of computer time from the DoD High Performance Computing Modernization Program at the U.S. Army Research Laboratory Supercomputing Resource Center.

2:40pm **2D+EM+MN+NS-ThA-2 Comparison of A- and B-exciton Intensity and Polarization in Transition Metal Dichalcogenide Monolayers and Heterostructures, Kathleen McCreary, A Hanbicki, S Sivaram, B Jonker, U.S. Naval Research Laboratory**

We survey a large number of monolayer TMDs to better understand the conditions responsible for various emission characteristics that have been reported in literature. We find that the intensities for both A- and B- peak emission vary widely as a result of sample-to-sample variations. However, a measurable B-peak intensity is evident in all samples. There is a clear linear relationship between the two peak intensities. The emission from the dominant A-peak is commonly several orders of magnitude higher than B-peak emission, resulting in B/A-intensity ratios well below 1%. Yet, as the A-peak intensity decreases, the ratio of B/A monotonically increases, and we observe a B/A ratio up to 30% in monolayer MoS₂. The A-excitonic emission is further quenched when MoS₂ is incorporated into an MoS₂/MoSe₂ heterostructure, where we observe comparable A- and B-peak intensities. We attribute these variations to differences in exciton recombination times, clarifying contradictory reports regarding the accessibility and significance of B-peak emission. Furthermore, we observe a high degree of valley polarization in both B-exciton emission in isolated monolayers and A-exciton emission in heterostructures, consistent with our model detailing the rapid exciton lifetimes in B-emission and van der Waals heterostructures.

Supported by core programs at NRL and the NRL Nanoscience Institute

3:00pm **2D+EM+MN+NS-ThA-3 Optospintronics and Magnetism with 2D Materials and Heterostructures, Roland Kawakami, The Ohio State University**

INVITED

I will review our latest developments in spintronics, optospintronics and magnetism in two-dimensional (2D) materials and heterostructures. Graphene continues to exhibit improved properties for spin transport and demonstrates additional functionality through the use of vertically stacked heterostructures. One of the interesting new directions is optospintronics enabled by heterostructures of graphene and transition metal dichalcogenides (TMD) [1]. Due to the valley optical selection rules of TMDs

and the large spin-orbit coupling, the helicity of the photon is coupled to the valley spin polarization of electrons. Thus, circularly polarized optical excitation into a TMD/graphene heterostructure generates spin polarization in the TMD that subsequently transfers to the graphene. This optical spin injection into graphene is an example of new functionality for the expanding field of 2D spintronics. In the area of 2D magnets, we have used molecular beam epitaxy (MBE) to deposit monolayer MnSe₂, which exhibits ferromagnetism at room temperature [2]. These results open the door for new possibilities for magnetoelectronic applications with low dimensional materials.

[1] Yunqiu Kelly Luo, Jinsong Xu, Tiancong Zhu, Guanzhong Wu, Elizabeth J. McCormick, Wenbo Zhan, Mahesh R. Neupane, and Roland K. Kawakami, *Nano Lett.* 17, 3877 (2017).

[2] Dante J. O'Hara, Tiancong Zhu, Amanda H. Trout, Adam S. Ahmed, Yunqiu Kelly Luo, Choong Hee Lee, Mark R. Brenner, Siddharth Rajan, Jay A. Gupta, David W. McComb, and Roland K. Kawakami, *Nano Lett.* doi: 10.1021/acs.nanolett.8b00683 (2018).

4:00pm **2D+EM+MN+NS-ThA-6 Giant Electromechanical Response in Van-der-Waals Layered Crystals, Sabine Neumayer, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory; E Eliseev, National Academy of Sciences of Ukraine; A Tselev, CICECO and Department of Physics, University of Aveiro, Portugal; A Morozovska, National Academy of Sciences of Ukraine; M Susner, M McGuire, Oak Ridge National Laboratory; J Brehm, S Pantelides, Vanderbilt University; N Balke, P Maksymovych, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory** Obtaining ultrathin electromechanically active materials for memory and energy applications encounters numerous challenges as significant downscaling of classical ferroelectrics such as perovskite oxides is severely constrained by size and screening effects. Moreover, interfacing pseudocubic ferroelectrics with 2D electronic materials faces challenges related to defect and impurities, which limit performance. Van der Waals ferroelectrics, especially transition metal thiophosphates such as copper indium thiophosphate (CIPS) yield promising prospects for applications as ultrathin piezoelectric structures and interface materials due to their stable surfaces, layered structure and transition temperatures near room temperature. Here, we use scanning probe microscopy to explore the remarkable functional properties of CIPS across the transition temperature. At low temperatures, strong electromechanical response is measured despite the small polarization values and the material contracts in electric fields rather than expanding. These findings point to giant negative electrostrictive coefficients, which were quantified using Landau-Ginzburg-Devonshire analysis. Above the transition temperature, CIPS shows dielectric tunability comparable to BST at low frequencies. In addition, electromechanical strain exceeding 10 nm displacement was measured upon fully reversible field induced formation of particles on the surface. Complimentary DFT calculations provide further insight into the role of ionic displacement in electromechanical behavior.

Research conducted at the Center for Nanophase Materials Sciences, which is a DOE Office of Science User Facility. Research was sponsored by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U. S. Department of Energy.

4:20pm **2D+EM+MN+NS-ThA-7 A Universal Method for Measuring Valleytronic Quality of 2D Materials using Conventional Raman Spectroscopy, Steven Vitale, J Varghese, D Nezich, M Rothschild, MIT Lincoln Laboratory**

Valleytronics offers a new information processing paradigm based on the momentum index of real or quasi-particles in 2D materials as the fundamental unit of information storage instead of charge. A major challenge to realize valleytronic computing is the development of deterministic material growth processes which yield valleytronic-quality material with the requisite valley relaxation lifetime (T₁) and valley dephasing time (T₂). Unfortunately direct measurement of T₁ and T₂ requires complex instrumentation to perform ultrafast spectroscopic measurements and thus is not practical for routine material analysis. In this paper, we demonstrate that an accurate and reproducible measurement of T₁/T_{exc} (where T_{exc} is the exciton recombination lifetime) can be performed a simple Raman microscope. By simultaneously measuring the photoluminescence of the 2D material and the Raman transition of the underlying silicon substrate as a function of the incident laser polarization angle, one can remove sources of error and equipment-to-equipment variability. This technique is completely general and can be applied to any valleytronic material which can be grown-on or transferred-to a Raman-

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active crystalline substrate, such as silicon. Using this technique we show that valley relaxation in a sample of CVD-grown MoS₂ is an order of magnitude slower at 4 K than at 100 K. Oxidation of MoS₂ left exposed to the ambient environment severely decreases the valleytronic quality of the material. Two-dimensional mapping of the valley relaxation time of CVD MoS₂ domains at 4 K shows a three-fold spatial symmetry which is suggestive of new valley physics phenomena which arise in 2D crystals of finite size. MoS₂ domain size also affects the valley relaxation time, which has significant material-growth implications for real valleytronic applications. Finally we compare these measurements to our calculated requirements for valley relaxation time in a practical information processing device and quantify the challenges for future valleytronic material growth.

4:40pm **2D+EM+MN+NS-ThA-8 Discovery of Intrinsic Ferromagnetism in 2D van der Waals Crystals**, *Xiang Zhang, C Gong*, University of California, Berkeley **INVITED**

In this talk, I will present our discovery of the intrinsic ferromagnetism in 2D van der Waals (vdW) crystals, including the prominent dimensionality effect and unprecedented magnetic field control of the Curie temperature in the nearly-ideal 2D Heisenberg ferromagnet. Significant fundamental physics in 2D magnetism and the corresponding exotic phenomena we observed will be expounded. Updated research on the complex magnon scatterings, material level engineering of 2D magnetism, and the development of novel concept of spintronic devices will be further discussed. Finally, I will envision the possible directions towards advancing 2D magnets for practical spintronic applications.

5:20pm **2D+EM+MN+NS-ThA-10 Spectroscopic Evidence of Pair-mediated Bosonic Modes in Superconductor FeSe/SrTiO₃(100) Film**, *Minjun Lee*, Seoul National University, Republic of Korea; *M Oh, H Jeon, S Yi, I Zoh*, Seoul National University, Republic of Korea; *C Zhang*, Seoul National University, Republic of Korea; *J Chae, Y Kuk*, Center for Quantum Nanoscience, Institute for Basic Science, Republic of Korea

Single layer FeSe on SrTiO₃(100) is atypical but noticed system in superconductivity. This has unique properties due to the substrate phonon. Unlike other bulk systems, the presence of the interface allows the substrate phonons to affect the superconducting layer. We have investigated substrate phonon effects on superconducting FeSe layer by using scanning tunneling spectroscopy and Eliashberg theory. We were able to measure acoustic, optical and substrate phonons in d^2I/dV^2 spectroscopy. We found these phonon modes attribute to the paring of electrons in this superconducting layer. These results are analyzed by Eliashberg model and we will discuss the coupling strength of these bosonic features. We have found that the substrate phonon has major contribution to increase the transition temperature of this system.

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