WORKSHOP #4

Magneto-Optical-Electronic Properties of Nanomaterials and Their Applications

Mingxing Li (BNL), Mircea Cotlet (BNL), Deep Jariwalla (UPenn), and Don DiMarzio (NGC Next)

Low dimensional nanomaterials such as quantum dots, nanowires and monolayers of two-dimensional (2D) materials have garnered considerable attention owing to their low dimensional physics which includes unique optical, electronic, and magnetic properties not seen in their bulk state of matter. Electro-optic effects based on 2D materials are increasingly used in optoelectronic devices such as photodetectors, light-emitting diodes, solar cells, and optical modulators. Magneto-optical effects based on 2D materials are now applied in optical communications and data storage while spin-based optical changes in single photon emitters of 2D materials provide platforms for quantum information science. Much of these nanoscale applications have their origin in basic studies of the understanding of optical, electronic and magnetic properties of said nanomaterials. CFN is planning to develop a state-of-the-art magneto-optical microscopy facility for the investigation of low dimensional nanomaterials, including 2D materials, and is planning to prospect the interest of the CFN/NSLS2 science user community in the future utilization and additional development of this facility with particular focus on fundamental studies of low dimensional materials.

This workshop aims to bring together leaders in the field of magneto-optical microscopic studies of low dimensional materials to discuss the latest research results in this field and to engage the scientific user community in potential partnerships with CFN staff in the utilization of the soon to come magneto-optical facility.

Start Time (ET)	Title	Speaker (Affiliation)
10:00 – 10:05 a.m.	Introduction/opening remarks by organizers	
10:05 – 10:40 a.m.	Imaging and Characterizing Magnetic Van der Waals Semiconductors	Deep Jariwala University of Pennsylvania
10:40 – 11:15 a.m.	Magneto-spectroscopy of interlayer excitons in TMDC heterostructures	Tobias Korn University of Rostock,
11:15 – 11:30 a.m.	BREAK	
11:30 – 12:05 p.m.	2D Magnets and Heterostructures	Cheng Gong University of Maryland-College Park
12:05 – 12:40 p.m.	Quantum emitters in two-dimensional materials	Chitraleema Chakraborty University of Delaware
12:40 – 12:45 p.m.	Edwards Vacuum	
12:45 – 2:00 p.m.	LUNCH	
2:00 – 2:35 p.m.	Tunable excitons and electron correlation in rhombohedral stacked graphene	Long Ju MIT
2:35 – 3:10 p.m.	Terahertz spectroscopy evidence of an electromagnon in honeycomb Dirac magnet CoTiO3	Rolando Valdes Aguilar Ohio State University
3:10 – 3:20 p.m.	BREAK	
3:20 – 3:55 p.m.	Surprises in Van Der Waals Moires, and Approaches to More Controlled Fabrication	David Goldhaber-Gordon Stanford University
3:55 – 4:30 p.m.	Controlling excitons in 2D semiconductor heterostructures	John Schaibley University of Arizona
4:30 – 4:35 p.m.	Edwards Vacuum	

4:35 – 4:45 p.m.	Discussion	

Abstracts

Imaging and Characterizing Magnetic Van der Waals Semiconductors

Prof. Deep Jariwala,

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The isolation of a growing number of two-dimensional (2D) materials has inspired worldwide efforts to integrate distinct 2D materials into van der Waals (vdW) heterostructures. Early focus remained on graphene and semiconducting 2D layers but as the number of materials grew, so did the electronic flavours and characters. Layered 2D magnetic semiconductors and were identified and isolated in 2018.1, 2 Since then, a large number of magnetic 2D materials have been isolated and studied as well. Among them, the magnetic chalcogenides and phopho-trichalcogenides are particularly appealing due to their semiconducting nature with band gaps in the visible frequency range. Semiconducting ferromagnets and anti-ferromagnets are particularly interesting for electronic and photonic applications and can also allow field and strain tunable magneto-electric and magneto-optical properties.

In this context I will present our collaborative works on 2D ferromagnetic Cr2Ge2Te6 (CGT) and also on anti-ferromagnets (AFMs) MnPS3 and FePS3 all of which are semiconductors. Our work on CGT done in collaboration with BNL3 reports the first observation of topologically non-trivial spin-textures in CGT in a Lorentz TEM below its Curie temperature. Evolution of these skyrmion-like "bubble" spin-textures was observed as a function of magnetic field including formation of closed lattice of bubble textures whose diameter was tuned with magnetic field. In our recent work on FePS3 which has a zig-zag type AFM order, we observe presence of large and tunable linear dichroism (LD) in the visible part of the spectrum.4 The LD coincides with the magnetic transition temperature of ~118 K and approaches 98% at specific wavelengths which vary as function of thickness of the FePS3. We identify that this large LD is a result of cavity effect of the crystal and the substrate and therefore can be tuned both spectrally and in magnitude. This LD and its spatial map can be used to probe spin-order in a simple and non-destructive way in 2D zig-zag type AFMs. Conversely in MnPS3 which has Neel type AFM order there is no measurable LD but instead the broken inversion symmetry produces a large second harmonic generation signal which can be used as a probe to map the spin-order.5 I will end by giving a broad perspective on future opportunities of 2D magnetic materials.

References:

1. Gong, C.; Zhang, X. Science 2019, 363, (6428), eaav4450.
2. Gong, C.; Li, L.; Li, Z.; Ji, H.; Stern, A.; Xia, Y.; Cao, T.; Bao, W.; Wang, C.; Wang, Y.; Qiu, Z. Q.; Cava, R. J.; Louie, S. G.; Xia, J.; Zhang, X. Nature 2017, 546, (7657), 265-269.
3. Han, M.-G.; Garlow, J. A.; Liu, Y.; Zhang, H.; Li, J.; DiMarzio, D.; Knight, M. W.; Petrovic, C.; Jariwala, D.; Zhu, Y. Nano Letters 2019, 19, 7859-7865.
4. Zhang, H.; Ni, Z.; Stevens, C. E.; Bai, A.; Peiris, F.; Hendrickson, J. R.; Wu, L.; Jariwala, D. Nature Photonics 2022, 10.1038/s41566-022-00970-8.

5. Ni, Z.; Zhang, H.; Hopper, D. A.; Haglund, A. V.; Huang, N.; Jariwala, D.; Bassett, L. C.; Mandrus, D. G.; Mele, E. J.; Kane, C. L.; Wu, L. Physical Review Letters 2021, 127, (18), 187201.

Magneto-spectroscopy of interlayer excitons in TMDC heterostructures

Prof. Tobias Korn,

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Transition-metal dichalcogenide (TMDC) monolayers are direct-gap semiconductors with peculiar spin-valley coupling. They are characterized by inequivalent valleys at the corners of the Brillouin zone (K points), which can be selectively excited via circularly polarized light. Combining two different TMDCs can lead to a type-II band alignment, so that optically excited electron-hole pairs form interlayer excitons (ILE).

In heterobilayers consisting of the TMDCs MoSe2 and WSe2, optically bright ILE are only observable if the interlayer twist angle is close to 0 (also called aligned or R-type) or 60 (also called anti-aligned or H-type) degrees. Optical spectroscopy of the ILE in these systems in large magnetic fields gives us a lot of insight into their structure.

In aligned structures, interlayer transitions between the MoSe2 conduction band and the WSe2 valence band K valleys with the same valley index are direct in reciprocal space. In anti-aligned structures, interlayer transitions between valleys with different indices are k-space direct. This allows us to engineer the ILE g factor using the interlayer twist, changing its magnitude [1] and even its sign [2]. Additionally, applied magnetic fields induce a valley polarization of the ILE, and its buildup can directly be observed in helicity- and time-resolved photoluminescence (PL). Remarkably, for aligned structures, we observe a nontrivial behavior of the circular polarization degree of PL, which changes its sign as a function of time. This can be explained by taking into account the dependence of ILE optical selection rules on interlayer registry.

A peculiar feature is observed for both, R-type and H-type ILEs: at an applied field of about 24 Tesla, the valley polarization is resonantly enhanced in both structures, even though their g factors are markedly different. This observation hints at a scattering process involving single carriers within the ILE and zone-boundary acoustic phonons [3].

[1] P. Nagler et al., Nature Comm. 8, 1551 (2017).

[2] J. Holler et al., Phys. Rev. B 105, 085303 (2022).

[3] D. Smirnov et al., in preparation (2022).

2D Magnets and Heterostructures

Prof. Cheng Gong,

Department of Electrical and Computer Engineering, Quantum Technology Center, and Maryland Quantum Materials Center, University of Maryland, College Park, MD 20742

Magnetism, one of the most fundamental physical properties, has revolutionized significant technologies such as data storage and biomedical imaging, and continues to bring forth new phenomena in emerging materials of reduced dimensionalities. The recently discovered magnetic 2D van der Waals materials [1, 2] provide ideal platforms to enable the atomic-thin, flexible, lightweight magneto-optical and magnetoelectric devices [3, 4, 5]. The seamless integration of 2D magnets with dissimilar electronic and photonic materials further opens up exciting opportunities for unprecedented properties and functionalities [6, 7]. In this talk, I will speak on our experimental discovery of the first 2D ferromagnet, and discuss the new properties unraveled in 2D magnetic heterostructures.

[1] C. Gong et al. *Nature 546, 265* (2017).

[2] C. Gong et al. *Science* 363, eaav4450 (2019).

[3] S. Gong et al. *PNAS* 115, 8511(2018).

[4] C. Gong et al. *Nat. Comm. 10, 2657* (2019).

[5] E. Du et al. *Nano Lett. 20, 7230* (2020).

[6] Z. Tu et al. *npj 2D Mater. Appl.* 5, 62 (2021).

[7] Z. Tu et al. *Appl. Phys. Lett.* 120, 043102 (2022).

Quantum emitters in two-dimensional materials

Prof. Chitraleema Chakraborty

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Quantum emitters in solids are promising building blocks for quantum information processing, quantum communications, and quantum sensing. Atomically thin materials that host quantum emitters, when compared to three-dimensional materials, have the advantage of reduced total internal reflection and easy coupling with interconnects. In this talk, I will share the story of the discovery and control of quantum emitters in two-dimensional materials. The possibility of leveraging strain-, electric- and magnetic- field tunable van der Waals heterostructures for dynamic modulation of their photophysical properties will be discussed. Further, I will also discuss the progress in their ab-initio prediction, deterministic generation, and integration with photonic devices, which offers a compelling solution to scalable solid-state quantum photonics. Our work opens the frontier of quantum optics in two-dimensional materials with the potential to revolutionize solid-state quantum devices.

Tunable excitons and electron correlation in rhombohedral stacked graphene

Prof. Long Ju

Department of Physics, MIT, Cambridge, MA, 02139

Rhombohedral stacked multi-layer graphene is a versatile material that exhibits fascinating electrical and optical properties. By applying a vertical electric field, a bandgap can be opened continously--making it a rare electrically tunable semimetal/semiconductor. I will first introduce our observation of tunable excitons in bilayer graphene. These strong optical resonances feature narrow linewidth, wide tuning range, unusual optical selection rules and large magnetic moments. In the second half of my talk, I will discuss the recipe to get correlated electron states by integrating a trilayer rhombohedral stacked graphene with hexagonal boron nitride (hBN). We observed electrically tunable flat bands in such a moire superlattice, as well as spectroscopy evidence of electron correlation.

Terahertz spectroscopy evidence of an electromagnon in honeycomb Dirac magnet CoTiO3

Prof. Rolando Valdes Aguilar

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In CoTiO3, the Co2+ ions form a honeycomb lattice in the ab plane. Scattering measurements indicate in-plane ferromagnetic interactions and interplane antiferromagnetic interactions. The measured magnon dispersion relation shows nodal lines around the K points, just like the electrons in graphene. This makes this material a potential platform for the existence of topological magnons.

We report on time-domain THz spectroscopy measurements on single crystals of CoTiO3. By controlling the polarization of the THz wave with respect to the crystal axes, we can determine the nature of the magnetic excitations. At zero magnetic field, we find that the 1.3 THz (~5.4 meV) magnon is excited only by the in-plane electric field of the THz wave, making this magnon an electromagnon. In addition, we determine the zone-center gap via the measurement of the in-plane magnetic-dipole antiferromagnetic resonance at ~230 GHz (~0.9 meV).

Surprises in Van Der Waals Moires, and Approaches to More Controlled Fabrication

Prof. David Goldhaber-Gordon (accepted invitation)

Department of Physics and Stanford Institute for Materials and Energy Sciences, Stanford University, Stanford, California, 94305

Stacks of van der Waals materials can be engineered to have nearly-flat electronic minibands, yielding novel topological and correlated electron states. Some such states have proven challenging to reproduce across devices. For example, orbital ferromagnetism in twisted bilayer graphene [requires not only a narrow range of graphene-graphene twist angles, but also crystallographic alignment between graphene and a hBN cladding layer. Rotational alignment between two different materials is difficult to engineer and to verify, limiting reproduction and further exploration of ferromagnetic twisted bilayer graphene.

We are combining (i) automated stacking in vacuum with (ii) optical characterization of flake orientation prior to stacking and (iii) optical and scanned probe characterization of twist angle in heterostructures after stacking. We hope this will offer key advantages for systematically forming stacks with consistent twist angle, and specifically reproducing and explicating orbital ferromagnetism in twisted bilayer graphene. We are also beginning to explore the surprisingly dramatic effects of anisotropy introduced by uniaxial strain in moires.

This work has been supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division, under contract DE-AC02-76SF00515; by an

LDRD at SLAC; and by the Gordon and Betty Moore Family Foundation. Key collaborators include Andy Mannix, Marc Kastner, and Tony Heinz at Stanford, and Aaron Sharpe at Sandia National Lab.

Terahe Controlling excitons in 2D semiconductor heterostructures

Prof. John Schaibley

Department of Physics, University of Arizona, Tucson, AZ, 85721

Two dimensional (2D) semiconductors, such as MoSe2 and WSe2, host tightly bound excitons (electron-hole pairs) that interact strongly with light. These monolayer semiconductors can be stacked together to realize heterostructures that exhibit new excitonic effects. In this presentation, I will discuss the optical response of two different 2D semiconductor heterostructures. First, I will review the progress towards understanding interlayer excitons in MoSe2 -WSe2 heterobilayers. These interlayer excitons host a rich moiré physics associated with the spatially modulated interactions between layers. I will discuss our recent work investigating the nature of localized interlayer excitons in these heterostructures. Interlayer excitons also possess a large permanent dipole moment that allows for their energy to be tuned with an out-of-plane electric field. By nano-patterning a gate on top of the MoSe2-WSe2 heterostructure, we are able to realize a ~30 nm diameter spatial trap for IXs, which has potential applications toward realizing deterministic single photon emitters. Finally, I will discuss our recent work developing excitonic diodes and excitonic transistor operation based on interlayer excitons.