2D Materials Technical Group
Room 303 - Session 2D+EM+MI-WeA

2D Materials: Charge Density Waves, Magnetism, and Superconductivity

Moderators: An-Ping Li, Oak Ridge National Laboratory, **Xiaomeng Liu**, Princeton University

2:20pm **2D+EM+MI-WeA-1 Tunable Electronic Structure and Correlations in Quasi-Freestanding Monolayer Transition Metal Disulfides*, Thomas Michely,*** *Universität zu Köln, Germany* **INVITED**

In situ reactive molecular beam epitaxy using single crystal graphene as a substrate enables to grow transition metal disulfides as quasi-free standing monolayers under well controlled conditions. Thereby access to their undisturbed electronic properties of as well as to those of their intrinsic defects is provided.

A non-invasive technique to shift the chemical potential in semiconducting transition metal disulfide layers like MoS_2 or WS_2 through p- and n-type doping of graphene is presented, while graphene remains a well-decoupled 2D substrate. These shifts induce giant band gap renormalizations, insulator-to-metal to insulator transitions and affects the metallic states in mirror twin boundaries.

Electronic correlations are known to be strong for dimensional reasons in transition metal dichalcogenide monolayers and often give rise to charge density waves and other competing electronic phases. The dependence of charge density waves on the environment and its layer dependence are investigated for several transition metal disulfides. We show that monolayers of VS_2 realize a CDW which stands out of our expectations. It displays a full CDW gap residing in the unoccupied states of monolayer VS_2 and the CDW induces a topological metal-metal (Lifshitz) transition. Non-linear coupling of transverse and longitudinal phonons is essential for the formation of the CDW and the full gap above the Fermi level.

Lastly we will focus to mirror twin boundaries in MoS_2 and investigate how to determine quantized polarization charges on these domain boundaries, whether they are subject to Fermi level pinning and how shifts in the Fermi level change screening of their line charge.

Contributions to this work by Clifford Murray, Camiel van Efferen, Wouter Jolie, Jeison Fischer, Timo Knispel, Joshua Hall, Stefan Kraus, Felix Huttmann, Carsten Busse, Niels Ehlen, Boris Senkovskiy, Martin Hell, Alexander Grüneis, Hannu-Pekka Komsa, Arkady Krasheninnikov, Jan Berges, Erik van Loon, Arne Schobert, Malte Rösner, Tim Wehling, Nico Rothenbach, Katharina Ollefs, Lucas Machado Arruda, Nick Brookes, Gunnar Schönhoff, Kurt Kummer, Heiko Wende, Philipp Weiß, Fabian Portner, and Achim Rosch are gratefully acknowledged.

3:00pm **2D+EM+MI-WeA-3 Dopants Modulated Interplay of Charge Density Wave and Superconductivity in 2D VdW Layered ZrTe3*, Xiao Tong,*** . Center of Functional Nanomaterials, Brookhaven National Laboratory*; Y. Liu, Z. Hu,* Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory*; D. Leshchev,* National Synchrotron Light Source II, Brookhaven National Laboratory*; X. Zhu, H. Lei,* Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory*; E. Stavitski, K. Attenkofer,* National Synchrotron Light Source II, Brookhaven National Laboratory*; C. Petrovic,* Condensed Matter Physics and Materials Science, Department, Brookhaven National Laboratory

Two-dimensional transition metal trichalcogenides ZrTe3 holds atomic chains in the crystal structure give rise to quasi one-dimensional (quasi 1D) conduction, and features the charge density wav (CDW) below TCDW ¼ 63 K and filamentary superconductivity below 2 K, Here, we report that superconductivity(SC) is enhanced as the consequence of suppressed CDW for Hf dopped ZrTe3, in contrast, SC is suppressed as the consequence of enhanced CDW for the Se dopped ZrTe. Our XPS and Raman studies suggested the suppressed CDW is due to Hf caused disorder in Te2-Te3 atomic chains, while the enhanced CDW is due to Se induced enhanced electron-phonon coupling in unperturbed periodicity of the Te2-Te3 chains, respectively.

3:20pm **2D+EM+MI-WeA-4 Magnetic Order in a Coherent Kondo Lattice in 1T/1H $TaSe_2$ Heterostructures***, W. Wan,* ***Rishav Harsh****, P. Dreher, S. Sajan,* Donostia International Physics Center , Spain*; A. Menino, I. Errea,* Centro de Física de Materiales (CSIC-UPV-EHU), Spain*; F. de Juan, M. Ugeda,* Donostia International Physics Center , Spain

Kondo lattice systems are of fundamental importance for our understanding of quantum criticality and unconventional superconductivity. At the heart of their complexity lies the competition between the opposing forces of Kondo screening and magnetic interactions, which is revealed at very low temperatures as the moments start behaving coherently and eventually determines the fate of the ground state. While our understanding of Kondo lattices has traditionally relied on technically challenging strongly correlated bulk f-electron systems, new light is being shed on the problem thanks to heterostructures of 2D transition metal dichalcogenides, which realize a tunable Kondo lattice platform in a simple material. Here, we study the 1T/1H-$TaSe_2$ heterostructure by high-resolution Scanning Tunneling Spectroscopy at 300 mK, and show a well resolved splitting of the Kondo peak, which increases monotonically in a nonlinear fashion in the presence of an out-of-plane magnetic field. This behavior is unexpected for a fully screened Kondo lattice, and it originates instead from a ground state with residual magnetic order, consistent with a Kondo coupling much below the critical point in the Doniach phase diagram.

4:20pm **2D+EM+MI-WeA-7 Structural and Magnetic Properties of Ultrathin $Cr_{(1+\delta)}Te_2$ Films Grown by Van Der Waals Epitaxy***, Kinga Lasek*, P. Coelho,* University of South Florida*; P. Gargiani , M. Valvidares,* ALBA Synchrotron Light Source, Spain*; K. Mohseni , H. Meyerheim , I. Kostanovskiy,* Max Planck Institute of Microstructure Physics, Germany*; K. Zberecki,* Warsaw University of Technology, Poland*; M. Batzill,* University of South Florida

Over the past years, researchers have proved that the layered structure of transition metal dichalcogenides (TMDs) enables the synthesis of novel materials. Specifically, introducing extra transition metal atoms into the vdW gap of the TMDs host lattice leads to various structural, electrical, and magnetic properties modulations. Exploring the latter, in particular, aligns with a recent search for ferromagnetic 2D materials.

In this talk, we will explore the epitaxial growth, structural, surface, and magnetic properties of a bi- to few-layer thick $Cr_{(1+\delta)}Te_2$ ($0 < \delta < 1$)films that represent a group of self-intercalated TMDs materials. These materials demonstratewell-known ferromagnetic ordering and exist in different compositional phases that vary by the amount of Cr intercalated (δ) between $CrTe_2$ layers.By detailed compositional and structural characterization, using scanning tunneling microscopy (STM), and high-resolution Rutherford backscattering (HR-RBS) we will show that the amount of self-intercalated Cr atoms can be controlled by post-growth annealing. Such modified films are characterized by an increased T_c up to 190K, a coercive field being reduced from 0.5 T to 0.3 T, and an isotropization of the magnetic anisotropy confirmed by XMCD measurements.

Finally, we will demonstrate that ultrathin vdW films can be prepared with the ultimate limit of a single self-intercalated layer by vdW epitaxy. These vdW materials maintain their ferromagnetic properties with desirable out-of-plane anisotropy and thus are potential ferromagnetic 2D materials that can be combined in vdW heterostructures by a bottom-up growth process.

4:40pm **2D+EM+MI-WeA-8 Transition Metal Silicates as a Platform for Robust Single Layer, Two-Dimensional Ferromagnetism*, Nassar Doudin,*** *K. Saritas,* Yale University*; P. Shafer, A. T. N'Diaye,* Lawrence Berkeley National Laboratory (LBNL)*; S. Ismail-Beigi, E. Altman,* Yale University

Two-dimensional (2D) materials have received extensive attention and rapid development since the discovery of graphene in 2004.[1] Magnetism in two dimensions has long been at the heart of theoretical, experimental, and technological advances, where great efforts have been made to realize magnetism in 2D materials.[2] To date, 2D magnetic materials have been obtained via exfoliation from bulk samples; however, this makes it difficult to precisely control the thickness and domain size of the samples. Moreover, the most intensely studied 2D magnetic materials (e.g. CrI_3, $Cr_2Ge_2Te_6$) are unstable in air and are only ferromagnetic below ~50 K. Hence, exploring new 2D magnetic materials is of grand significance. Recently, vdW epitaxy techniques have fulfilled the growth of 2D vdW materials on metal substrates, such as 2D silica bilayers and related transition metal silicates which break inversion symmetry guaranteeing at least a piezoelectric response. The arrival of these 2D vdW materials opens up exciting opportunities for preparing 2D multiferroic materials. In this talk, we demonstrate air stable, single layer 2D $M_xSi_2O_5(OH)_y$ (M= Fe, Cr) structures consisting of a silica-capped metal oxide grown on Pd and Au substrates that exhibit ferromagnetic order at room temperature as measured by x-ray magnetic circular dichroism (XMCD) spectroscopy. Application of small magnetic fields proves that the observed magnetic structures follow a hysteretic behavior. SQUID magnetometry confirms these results and reveals high spin-polarization at room temperature with in-plane magnetic anisotropy. The measurements are further supported by first-principles theoretical calculations which highlight approaches to stabilize the magnetic order. Thus $M_xSi_2O_5(OH)_y$ based 2D materials represent a new platform for single layer 2D ferromagnetism with potential multiferroic behavior.

1. Meyer, J. C. *et al.* The structure of suspended graphene sheets. *Nature* **446**, 60–63 (2007).
2. K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. G. and A. A. F. Electric Field Effect in Atomically Thin Carbon Films. *Science.* **306**, 666–669 (2004).

5:00pm **2D+EM+MI-WeA-9 Novel Materials for Quantum Computing Devices: Monolayer Topological Superconductors***, **Yi-Ting Hsu,*** University of Notre Dame **INVITED**

Atomically thin superconductors that possess 'topological properties' have attracted extensive attention since many of them are predicted to host exotic zero-energy quasi-particles called Majorana zero modes (MZMs). MZMs are proposed to be the building blocks in a type of fault-tolerant quantum computation scheme known as topological quantum computation (TQC). One of the pressing challenges in the field of TQC is to experimentally realize such a superconducting film and detect the existence of MZMs. In this talk, I will first discuss what a topological superconductor is and why it is a promising platform for a TQC device. I will then talk about candidate materials in the family of transition metal dichalcogenides. Finally, I will discuss our theory-experiment combined effort to systematically identify more candidates in two-dimensional material databases.

5:40pm **2D+EM+MI-WeA-11 Tuning Magnetism and Superconductivity in Single Layer FeSeTe by Chemical Pressure***, **Basu Oli**, Q. Zou, H. Zhang,* West Virginia University*; T. Shishidou, M. Weinert,* University of Wisconsin, Millwaukie*; L. Li,* West Virginia University

The interplay of topology, magnetism, and superconductivity in a single-layer FeX (X = S, Se, Te) epitaxially grown on $SrTiO_3$ (STO) substrate provides a model system for investigating a wide range of quantum phenomena. This work explores the impact of chemical pressure on magnetism and superconductivity in single-layer $FeSe_{1-x}Te_x$/STO grown by molecular beam epitaxy using *in-situ* angle-resolved photoemission spectroscopy and scanning tunneling microscopy/spectroscopy. We find that the Fermi surface consists of only an electron pocket at the M point, which decreases in size with increasing Te concentration and disappears completely for $x > 0.75$. At the Γ point, a hole pocket appears with $x > 0.65$, while the bands changes from parabolic to linear up to $x = 0.9$, where it reverts back to parabolic. Accompanying the changes in the band structure, the top of bands at the Γ also shift towards and then away from the Fermi level, indicative of a topological phase transition in $FeSe_{1-x}Te_x$/STO. At 4.3 K, while the FeTe films are non-superconducting, dI/dV tunneling spectra indicate the emergence of superconductivity when Se concentration is greater than 25%. Our spin spiral calculations indicate that the FeTe system exhibits long-range bi-collinear antiferromagnetic (AFM) order, which is tuned toward the checkerboard (CB) AFM fluctuations with the incorporation of Se. Our findings indicate that CB AFM fluctuations are critical for superconductivity in epitaxial single layer iron chalcogenide superconductors on STO.

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6:00pm **2D+EM+MI-WeA-12 Peculiar Near-Contact Regime of Andreev Reflection at the Breakdown of a Tunnel Junction***, **Petro Maksymovych**, S. Song,* Oak Ridge National Laboratory*; J. Lado,* Aalto University, Finland*; W. Ko,* Oak Ridge National Laboratory

Recently we introduced non-contact Andreev reflection (NCAR) - a new experimental approach to quantify Andreev reflection in a tunable tunnel junction [1]. The technique utilizes the fundamental connection between the physics of the scattering process and the strength of exponential non-linearity of the tunneling current, and therefore adopts the tunneling current decay rate as the observable. NCAR simultaneously achieves spectroscopy of quasiparticle density of states, atomic-scale resolution and quantitative Andreev reflection, while avoiding the necessity to form invasive and mesoscale mechanical contacts.

One surprising observation of NCAR is that Andreev reflection does not have a monotonous dependence on tip-sample separation. In particular, the decay rate signature of Andreev reflection proceeds through a maximum just before the mechanical contact. In this talk, we will explore in detail the origin of this near-contact regime and its significance for the characterization of superconductivity. In particular, we will rationalize the observations within the accepted BKT model as well as accurate tight-binding simulations, revealing the fundamental connection between the tunneling barrier dependence of Andreev reflection and the order in perturbation theory responsible for the tunneling current. Furthermore, we will highlight the importance of higher order Andreev reflection for the measurement of unconventional superconductors. In general, tunneling and near-contact regimes will qualitatively differ from each other in the case of unconventional superconducting order parameters, enabling direct inference of their symmetry.

At the same time, we will demonstrate how quantitative comparison of statistical distributions of decay rate enables a complementary, probabilistic analysis of the Andreev reflection [2]. This purely informational approach is particularly important given the narrow parameter space that separates the now rich variety of techniques to directly probe superconductivity, enabling robust control over crossovers between non-contact, near-contact, multiple Andreev reflection as well as quasiparticle tunneling in atomic-scale junctions.

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1. W. Ko, J. Lado, P. Maksymovych, Nano Lett. 22, 4042 (2022).

2. W. Ko, E. Dumitrescu, P. Maksymovych, Phys. Rev. Res. 3, 033248 (2021).

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