HL 60: Focus Session: Physics of the van der Waals Magnetic Semiconductor CrSBr II (joint session HL/MA)

The joint focus session of the divisions HL and MA presents the latest developments of the rapidly growing community working with the van der Waals magnetic semiconductor CrSBr with distinct excitonic and magnetic properties, and it is organized by Shengqiang Zhou (HZ Dresden-Rossendorf), Farsane Tabata-Vakili (TU Braunschweig), and Florian Dirnberger (TU Munich).

Time: Friday 9:30–13:00 Location: H17

Invited Talk HL 60.1 Fri 9:30 H17

Constructing Artificial Matter in the Electron Microscope - Atomic Fabrication at Scale in CrSBr — •Julian Klein — Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, 02319 MA, USA

The ability to control the arrangement of individual atoms has transcended naturally occurring configurations of matter, enabling experimental breakthroughs in quantum physics. I will show how we can now use scanning transmission electron microscopy to construct artificial atomic arrangements at scale and demonstrate it with the layered magnetic quasi-1D semiconductor CrSBr. By developing strategies to position the electron beam with picometer precision and perform rapid, targeted beam actions, we achieve deterministic control over the movement of Cr atoms in space and time. With this capability, we selectively steer Cr atoms into interstitial positions, forming localized quantum states while simultaneously monitoring atomic movements in real time with microsecond resolution. Fully automating the electron microscope enables us to construct ordered arrays of Cr interstitial superlattices atom by atom as well as nonperiodic structures, spanning hundreds of locations over tens of nanometers, all within minutes. Our results show that atomic fabrication at scale in the electron microscope is now a reality, unlocking unprecedented opportunities to construct quantum defects and phases, atom by atom, in the solid state, that extend over macroscopic length scales.

Invited Talk HL 60.2 Fri 10:00 H17

Tuning the structure and magnetism in CrSBr via external pressure — •Ece Uykur — Helmholtz-Zentrum Dresden-Rossendorf, Institute of Ion Beam Physics and Materials Research, 01328 Dresden, Germany

As one of the two-dimensional (2D) van der Waals (vdW) magnets, CrSBr regained significant attention recently because it is air-stable even in the monolayer form making this compound very attractive. It shows strong coupling between its magnetic, electronic, structural, and optical properties [1]. Several ab initio calculations put forward the importance of the balance between Cr-Cr direct exchange and Cr-anion-Cr superexchange interactions and showed that the A-type antiferromagnetism in this compound is delicately balanced with these short- and long-range magnetic interactions [2]. Therefore, studies exploring the tunability of the inter- and inralayer coupling are important and one plausible experimental strategy is the external pressure.

In this talk, I will summarize our recent efforts on high-pressure single crystal XRD and magnetization studies on CrSBr. We performed single crystal XRD studies up to ∼20 GPa, which reveals a non-monotonous behavior of Cr-ion in the structure along with a structural phase transition above 17 GPa. The movement of this Cr-ion has also direct link with the magnetization of the compound that is studied with the high-pressure magnetic susceptibility measurements up to ∼8 GPa.

[1] K. Lin et al., ACS Nano 18, 2898 (2024) [2] J. Cenker et al., Nat. Nanotechnol. 17, 256 (2022).

Invited Talk HL 60.3 Fri 10:30 H17

A theoretical perspective on exciton-magnon coupling and its implications — •Akashdeep Kamra — Department of Physics, Rheinland-Pfälzische Technische Universität (RPTU) Kaiserslautern-Landau, Kaiserslautern, Germany

The dependence of exciton energies on the magnetic order in CrSBr has opened avenues for controlling optical properties using magnetic fields. Conversely, it has enabled an optical time-resolved sensing of the magnetic degrees of freedom. This has further been exploited to investigate the interplay between excitons and magnons, the excitations of the magnetic order. We will discuss how excitonic energies offer a convenient access to coherent as well as thermal magnon dynamics. Focusing on transport, we will discuss the recent observation of the magnon-exciton drag effect that makes it feasible to leverage thermal magnon currents for transporting excitons at unexpectedly fast velocities. Finally, we will conclude with a brief discussion of emergent non-linearities in exciton energies mediated by the magnonic modes in the canted magnetic state of CrSBr.

References:

1. F. Dirnberger et al., Magneto-optics in a van der Waals magnet tuned by self-hybridized polaritons, Nature 620, 533 (2023).

2. F. Dirnberger, S. Terres, Z. A. Iakovlev, K. Mosina, Z. Sofer, A. Kamra, M. M. Glazov, and A. Chernikov, Exciton transport driven by spin excitations in an antiferromagnet (unpublished).

3. B. Datta et al., Magnon-mediated exciton-exciton interaction in a van der Waals antiferromagnet, arXiv:2409.18501.

15 min. break

Invited Talk HL 60.4 Fri 11:15 H17

Exciton and valley properties of monolayer transition metal dichalcogenides on the van der Waals magnetic semiconductor CrSBr — •Yara Galvao Gobato — Universidade Federal de Sao Carlos, Sao Carlos, Brazil

Chromium sulfide bromide is a promising van der Waals (vdW) magnetic material, undergoing a magnetic phase transition to an A type antiferromagnetic state below the Neél temperature of about 132K in its bulk form. VdW heterostructures composed of monolayer transition metal dichalcogenides (TMDs) and vdW magnetic materials such as CrSBr are an interesting platform to modify valley and excitonic properties of non-magnetic TMDs. In this talk, we will present our recent results on optical and magneto-optical properties of monolayer TMDs on CrSBr under different magnetic field orientations. Remarkably, we have observed a clear influence of the CrSBr magnetic order on the exciton and valley properties of monolayer TMDs, such as an anomalous linear polarization dependence, unsual temperature dependence of emission energies, magnetic field dependence of the emission intensity, and valley g-factor values with clear signatures of an asymmetric magnetic proximity exchange interaction. Our results are explained by asymmetric magnetic proximity effects, charge transfer and a possible contribution of exciton/trion magnon coupling. Our studies suggest that vdW heterostructures with antiferromagnetic nonmagnetic interfaces are interesting platforms to modify the valley and excitonic properties of TMDs for possible applications in opto-spintronics and quantum technology.

HL 60.5 Fri 11:45 H17

Ab initio studies on the electronic and optical properties of magnetic CrSBr — •Marie-Christin Heissenbüttel[1], Pierre-Maurice Piel[2], Julian Klein[3], Thorsten Deilmann[1], Ursula Wurstbauer[2], and Michael Rohlfing[1] — [1]Institute of Solid State Theory, University of Münster, Germany — [2]Physical Institute, University of Münster, Germany — [3]Department of Materials Science and Engineering, MIT, Massachusetts, USA

CrSBr recently emerged as a van der Waals layered material exhibiting intriguing electronic and optical properties arising from the intricate interplay between crystal structure and layered magnetic order. A thorough understanding of these effects is essential to assess its potential for applications in spintronic and quantum devices. Due to the large crystal anisotropy, the monolayer, multilayer, and bulk crystal CrSBr show a quasi-one-dimensional behaviour of effective masses and exciton wavefunctions [1]. The interlayer antiferromagnetic (AFM) coupling suppresses layer to layer interactions in the magnetic ordered low temperature phase, resulting in strong quantum confinement of electrons and excitons within the individual layers [2]. Using ab-initio GW/Bethe-Salpeter equation calculations, we analyze electronic and excitonic properties on the same footing and elucidate how the AFM van der Waals stacking, symmetry properties and the large crystal

anisotropy govern the electronic and optical properties of this material.

[1] https://doi.org/10.1021/acsnano.2c07316
[2] https://doi.org/10.48550/arXiv.2403.20174

HL 60.6 Fri 12:00 H17

Internal structure and ultrafast dynamics of quasi- 1D excitons controlled by magnetic order — •N. Nilforoushan[1], M. Liebich[1], M. Florian[2], F. Mooshammer[1,3], A. D. Koulouklidis[1,3], L. Wittmann[1], K. Mosina[4], Z. Sofer[4], F. Dirnberger[5], M. Kira[2], and R. Huber[1,3] — [1]Dept. of Physics, University of Regensburg, Germany — [2]Dept. of Electrical Engineering and Computer Science, University of Michigan, USA — [3]RUN, University of Regensburg, Germany — [4]Dept. of Inorganic Chemistry, University of Chemistry and Technology Prague, Czech Republic — [5]Dept. of Physics, Technical University of Munich, Germany

In van der Waals (vdW) layered crystals, Coulomb correlations are often tuned by structural engineering, giving rise to emergent phenomena such as tightly bound excitons and exotic electronic and magnetic phases. Magnetic vdW materials offer a unique platform for in situ control of Coulomb correlations enabled by their intrinsic magnetic order. Here, we present quantitative experiment-theory proof that excitonic correlations can be tailored through spin order in the vdW magnet CrSBr. By probing internal transitions of excitons with phase-locked mid-infrared pulses, we reveal their binding energy and strong anisotropy of their quasi-1D orbitals resulting in significant fine-structure splitting. We switch excitons from monolayer-localized to interlayer-delocalized species by pushing the system from the antiferromagnetic to the paramagnetic phase. The exciton's ultrafast dynamics further support this scenario. In future applications, excitons may be interfaced with spintronics enabling on-demand phase transitions.

HL 60.7 Fri 12:15 H17

Raman controlled lithium intercalation into CrSBr van der Waals structure — •Ksenia Mosina, Aljoscha Söll, Martin Veselý, Jiří Šturala, and Zdenek Sofer — Department of Inorganic Chemistry, University of Chemistry and Technology Prague, 166 28 Prague 6, Czech Republic.

Lithium intercalation into the van der Waals crystalline structure of layered transition metal dichalcogenides by means of chemical and electrochemical intercalation is a well-known method for studying semiconductor-metallic phase transitions. The layered semiconductor chromium sulphur bromine (CrSBr) in recent years becomes an ultimate playground for the studies of low-dimensional magneto-optical properties. The interlayer distance of CrSBr allows the easy cleavage and intercalation of the guest molecules within the crystalline structure. Conveniently air-stable, this material exhibits a direct band gap of 1.5 eV, an antiferromagnetic state in bulk and ferromagnetism in the monolayer. Here, we present the lithium intercalation method into the CrSBr structure by lithium-solvated electron solution. To monitor the lithiation process in real-time, we investigated the Raman spectra evolution upon lithium ion intercalation into a few-layered CrSBr flake. Our findings suggest that the quasi-one dimensional nature of CrSBr leads to weak interlayer hybridization along the b-direction, which facilitates the diffusion of guest ions by lowering the migration energy barrier and enables anisotropic Li+ diffusion. The reliable intercalation methodology allows tracking the intercalation process directly in the desired area favorable for device fabrication.

Invited Talk HL 60.8 Fri 12:30 H17

Electric field control of intra- and interlayer excitons in CrSBr — •Nathan Wilson[1], Amine Ben Mhenni[1], Ferdinand Menzel[1], Alain Dijkstra[1], Zdenek Sofer[2], and Jonathan Finley[1] — [1]Walter Schottky Institute, TU Munich, Garching, Germany — [2]Institute of Chemistry and Technology, Prague, Czech Republic

In the 2D magnetic semiconductor CrSBr, the interplay between a direct bandgap for all layer thicknesses and layered antiferromagnetism with strong magneto-electronic coupling give rise to rich but poorly understood excitonic physics. So far, the presence of two closely spaced conduction bands and existence of both intra and interlayer excitons in multilayers has complicated interpretation of the optical spectrum of its excitons. Here, we study monolayers and bilayers of CrSBr in dual-gated structures, allowing for independent tuning of electric field and charge doping. Our study reveals the existence of the previously unobserved ground state exciton in monolayers, which is darkened both by charge doping and electric field. We find that both intralayer and hybrid intra/interlayer excitons are highly sensitive to the vertical electric field, implying a reasonably large exciton polarizability and control over wavefunction symmetry. With this information, we are able to form a more complete picture of the real space and band character of the excitons in CrSBr.