O 64: Focus Session Atomic Scale Investigation of Magnetic 2D Materials

The rapid expansion of the family of magnetic two-dimensional (2D) materials led to the observation of various types of magnetic order in the 2D limit, such as (anti-)ferromagnetism, noncolinear structures, and magnetic moiré effects. On the fundamental level, there are various open questions regarding the mechanisms that underlie these ground states, as well as the understanding of the role of the interface and dimensionality.

Epitaxial growth of 2D magnetic materials on inert substrates under ultrahigh vacuum conditions and respective in situ investigation allows direct and unambiguous comparison between experimental findings and theoretical calculations. Experimentally, (spin-polarized) scanning tunneling microscopy methods are ideal to explore the electronic structure and magnetic properties of emerging 2D magnetic phases with ultimate real-space and energy resolution at low temperatures. These results are often corroborated by averaging techniques such as X-ray magnetic circular dichroism or magneto-optic Kerr effect. Theoretically, the use of model Hamiltonians requires atomically well-defined systems to precisely predict the electronic and magnetic properties. Combining these complementary approaches helps to elucidate the role of the substrate, defects, and the coupling between quasiparticles in stacked heterostructures.

This focus session aims at highlighting recent progress in the growth and characterization of magnetic states in epitaxial 2D materials and 2D heterostructures on metal substrates, bringing together experts from the fields of scanning probe techniques and first-principles calculations. The overall goal of the session is to gain fundamental insights into the driving mechanisms of 2D magnetic phases.

Organized by
Jeison Fischer, University of Cologne, Germany,
Wouter Jolie, University of Cologne, Germany.

Time: Wednesday 15:00–18:00 Location: H2

Invited Talk O 64.1 Wed 15:00 H2

Topological spin structures in two-dimensional van der Waals magnets and heterostructures — •Stefan Heinze — Institute of Astrophysics and Theoretical Physics, University of Kiel, Germany

Two-dimensional (2D) van der Waals (vdW) magnets offer exciting opportunities for topological magnetism due to high-quality interfaces, the possibility of single-atomic layer systems, and easy control of magnetism via external stimuli [1]. Here, we explore nano- and atomic-scale topological spin structures in 2D vdW magnets and heterostructures based on first-principles calculations and atomistic spin simulations. A focus is given to heterostructures based on the 2D vdW magnet Fe_3GeTe_2 which is experimentally accessible and exhibits favorable properties such as a high transition temperature. An essential prerequisite to apply topological spin states such as skyrmions in future applications is a sufficient stability which we quantify by calculating their lifetime using transition-state theory [2,3]. All-electrical skyrmion detection is proposed via the tunneling anisotropic and non-collinear magnetoresistance considering both a scanning tunneling microscopy geometry and a planar tunnel device structure [4]. Finally, the all-magnetic vdW heterostructure $Fe_3GeTe_2/Cr_2Ge_2Te_6$ is studied and the stability of bimerons in $Cr_2Ge_2Te_6$ is discussed [5].
[1] Q. H. Wang *et al.*, ACS Nano **16**, 6960 (2022).
[2] D. Li *et al.*, Nano Lett. **22**, 7706 (2022).
[3] D. Li *et al.*, Phys. Rev. B **109**, L220404 (2024).
[4] D. Li *et al.*, Nano Lett. **24**, 2496 (2024).
[5] D. Li *et al.*, arxiv: 2408.15974 (2024).

Invited Talk O 64.2 Wed 15:30 H2

Ferromagnetic Order in 2D Layers of Transition Metal Dichlorides — Andrea Aguirre[1], Andres Pinar[2], Diego Soler[2], Carmen Gonzalez-Orellana[1], Jon Ortuzar[3], Oleksandr Stesovych[2], Celia Rogero[1], Jose Ignacio Pascual[3], Pavel Jelinek[2], Maxim Ilyn[1], and •Martina Corso[1] — [1]Centro de Fisica de Materiales, San Sebastian, Spain — [2]Institute of Physics, Czech Academy of Sciences, Prague, Czech Republic — [3]CIC NanoGUNE, San Sebastian, Spain

Transition metals dihalides are an ideal class of van der Waals materials that enable the study of magnetic phases as function of the transition metal and halide composition as predicted by theory. Here, we characterize the magnetic and electronic properties of 2D magnets based on metallic dichlorides. The materials form flat epitaxial layers on Au(111) with semiconducting character. By X-Ray Magnetic Circular Dichroism (XMCD) measurements we find that single layers of FeCl2 and NiCl2 are soft ferromagnets on Au(111) and their magnetization can be switched from out-of-plane to in-plane by substituting the metal ion from Fe to Ni. Using low temperature scanning tunnelling microscopy with tips functionalized with a nickelocene molecule as magnetic sensor, we confirm the magnetic order of the materials at the atomic scale even at zero applied magnetic field. We thus established a correlation between the mesoscopic magnetic properties probed by XMCD and the atomic spins. Our results suggest that these 2D semiconducting magnets could be implemented in van der Waals heterostructures for applications in spintronics and opto-spintronics.

Invited Talk O 64.3 Wed 16:00 H2

Tailoring spin lattice in van der Waals monolayer crystals — •Ying-Shuang Fu — Huazhong University of Science and Technology, Wuhan, Chinaina

Spin lattices in monolayer van der Waals (vdW) crystals provide a paradigm for exploring both fundamental spin physics at the 2D limit and miniaturized spintronic applications. Here, we present our recent research in the construction of spin lattices with molecular beam epitaxy and addressing their spin states with both spin-averaged and spin-polarized scanning tunneling microscopy. We explored artificial Kondo lattices in monolayer vdW crystals aiming to emulate protocol heavy fermion systems. We realized quasi-1D Kondo lattice in monolayer stripe-phase 1T-NbSe2 and superconducting 2D Kondo lattice in monolayer VSe2 grown on NbSe2. We also investigated intrinsic magnetic order in monolayer vdW crystals using spin-polarized scanning tunneling microscopy. We identified an antiferromagnetic order in monolayer CrTe2, unraveling the indispensable role of interlayer coupling in determining the magnetic order. Based on that finding, we further fabricated Janus CrTeSe monolayer, and regulates the magnetic anisotropy energy via the symmetry breaking introduced from the Janus structure. Our study resolves intrinsic magnetism with atomic-scale resolution down to monolayer limit, opening up an avenue for studying the unusual spin excitations.

Invited Talk O 64.4 Wed 16:30 H2

Spin excitations in 2D heterostructures from realistic fermionic models — •António Costa — Center of Physics of the University of Minho, Braga, Portugal — International Iberian Nanotechnology Laboratory

Spin excitations dominate the magnetic response of ferromagnetic two-dimensional crystals. The interplay between low dimensionality, reduced symmetry and spin-orbit coupling endows spin excitations in those materials with intriguing properties, such as non-trivial topology and non-reciprocity. Moreover, spin-orbit coupling connects spin

and charge degrees of freedom, opening up paths to electrical control and detection of magnetic states. I will present a microscopic description of the spin response of nanostructured materials based on Hamiltonians for itinerant fermions, derived from DFT calculations. This approach incorporates spin-orbit coupling, does not rely on postulated spin models, and can be applied to insulating or conducting 2D heterostructures, with any kind of magnetic order. I will discuss the properties of magnons in insulating and metallic van der Waals ferromagnets, as well as in 2D molecular crystals. I will also discuss the proximity effect in a heterostructure formed by a 2D ferromagnet and graphene.

O 64.5 Wed 17:00 H2

Monolayer Multiferroic - Superconductor van der Waals Heterostructures — •Büşra Gamze Arslan, Mohammad Amini, Ziying Wang, Robert Drost, and Peter Liljeroth — Department of Applied Physics, Aalto University, P.O. Box 15100, 00076 Aalto, Espoo, Finland

Topological superconductivity, a quantum phase hosting robust edge modes like Majorana zero modes (MZMs), holds promise for fault-tolerant quantum computation due to its stability against disturbances. Stacked van der Waals materials offer a promising platform to engineer topological superconductors, as they arise from the interactions between superconductors and magnetic materials. With its helical magnetic order and intrinsic spin-orbit coupling, multiferroic NiI_2 is an excellent component for heterostructures realising topological superconductivity.

Here we present our study on monolayer NiI_2 grown on superconducting bulk $NbSe_2$. This system is characterized using scanning tunneling microscopy (STM) and spectroscopy (STS). The effect of doping due to the growth of NiI_2 on different substrates is investigated. Our observations revealed that NiI_2 does not show ferroelectricity, likely due to the charge transfer from the $NbSe_2$ substrate. In addition, we have investigated the edges of the NiI_2 islands, where we found clear signatures of edge modes in the $NiI_2/NbSe_2$ system. Whether these arise from topological effects remains to be seen in future studies. Our results show that combining 2D materials can create custom materials with relevant properties.

O 64.6 Wed 17:15 H2

Engineering the electronic and magnetic properties of MPS_3 (M=Fe, Ni, Co, Mn) materials through alkali metal doping — Jonah Nitschke[1], •Preeti Bhumla[2], Till Willershausen[1], Patrick Merisescu[3], Lasse Sternemann[1], Valentin Mischke[1], Michele Capra[1], Mira Arndt[1], David Janas[1], Giovanni Zamborlini[4], Silvana Botti[2], and Mirko Cinchetti[1] — [1]TU Dortmund — [2]Research Center Future Energy Materials and Systems, ICAMS, Ruhr University Bochum — [3]University of Bath — [4]Universität Graz

Transition metal phosphorus trichalcogenides, MPX_3 (where M represents a transition metal and X represents a chalcogen), have emerged as promising candidates for exploring two-dimensional (2D) magnetism. In this study, we focus particularly on MPS_3 (M = Fe, Ni, Co, Mn) materials, both above and below the Néel temperature TN. We investigate the electronic and magnetic properties of these materials using micrometer-scale angle-resolved photoelectron spectroscopy (ARPES) and density functional theory (DFT+U) calculations. We observe an increase in the band gaps and shifts in the M d and S p states below TN in the antiferromagnetic (AFM) phase. The density of states reveals the orbital character of the observed bands, and the strong hybridization between the M d and S p orbitals suggests that the superexchange mechanism, in which the S atom mediates the magnetic interaction between neighboring M ions, is relevant for these materials. Further, we examine the effect of alkali metal doping on the magnetic properties of these transition metal phosphorus trichalcogenides.

O 64.7 Wed 17:30 H2

Conflicting magnetic anisotropy in 2D metal-organic networks — •Diego Radillo[1,2], Céline Hensky[1,2], Quy Hien Le[1,2], Manfred Parschau[3], Egzona Isufi Neziri[3], Christian Wäckerlin[1,2], and Pierluigi Gargiani[4] — [1]EPFL, Switzerland — [2]PSI, Switzerland — [3]Empa, Switzerland — [4]ALBA, Spain

This talk presents insights into the properties of 2D metal-organic coordination networks embedded with atoms of conflicting magnetic anisotropies. In particular, we look into the coordination networks of tetracyanoethylene with Ni atoms (NiTCNE), Fe atoms (FeTCNE) and a mixture of both (NiFeTCNE), on a gold(111) surface. NiTCNE is known to be a ferromagnet with an out-of-plane easy axis of magnetization. Whereas, for the conflicting in-plane ferromagnet, we propose FeTCNE as a suitable candidate. In our mixed-network experiments, where the magnetic exchange energy enforces that the preferences of the two metal ions cannot be perfectly satisfied, we observe that the magnetization along the out-of-plane and in-plane axes at the Ni and Fe centers mutually influence each other in a rather rational way. This interaction suggests the potential for fine-tuning the magnetization axis and highlights the importance of investigating spin alignment at the per-atom scale. Scanning probe microscopy (SPM) and non-contact atomic force microscopy (AFM) are employed to inspect the morphology of the networks, while X-ray absorption spectroscopy (XAS) and X-ray mag- netic circular dichroism (XMCD) were utilized to analyze the magnetic properties.

O 64.8 Wed 17:45 H2

Non-van der Waals 2D Materials: Magnetic State Control — •Tom Barnowsky[1,2] and Rico Friedrich[1,2,3] — [1]TU Dresden — [2]Helmholtz-Zentrum Dresden-Rossendorf — [3]Duke University, Durham, USA

Non-van der Waals (non-vdW) 2D materials – exfoliated from non-layered bulk structures [1] – offer unique opportunities for exploring magnetic properties and their surface-assisted manipulation.

In recent data-driven studies [2,3], we predict several dozen exfoliable candidates. Many of these materials exhibit intrinsic magnetism, notably due to magnetic surface cations, which lead to strong surface spin polarization. Furthermore, the exposed "dangling" bonds at their surfaces – created by bond breaking during exfoliation from the bulk – enable passivation that can significantly modify their electronic and magnetic properties [4]. This passivation-based tuning can, for example, switch the magnetic state of these materials, *i.e.*, alter their local spin symmetry. Most notably, non-vdW 2D $CdTiO_3$ – a diamagnetic compound in its pristine form – becomes ferromagnetic upon hydrogenation. Using data mining and autonomous density functional theory, we demonstrate the potential of these materials as a powerful platform for magnetic state control, opening new possibilities for spintronics.

[1] A. Puthirath Balan *et al.*, Nat. Nanotechnol. **13**, 602 (2018).
[2] R. Friedrich *et al.*, Nano Lett. **22**, 989 (2022).
[3] T. Barnowsky *et al.*, Adv. Electron. Mater. **9**, 2201112 (2023).
[4] T. Barnowsky *et al.*, Nano Lett. **24**, 3874 (2024).