TT 4: Focus Session: Correlated van-der-Waals Magnets

Reducing the dimensionality of electronic materials often yields novel phenomena and exceptional physical properties. In layered van-der-Waals (vdW) materials which are formed by structurally stable but out-of-plane only weakly coupled crystalline layers this is, e.g., demonstrated by the presence of long-range magnetic order down to the bilayer in $Cr_2Ge_2Te_6$ and down to the monolayer in CrI_3. For VSe_2, the presence of ferromagnetism even at room temperature has been reported for monolayers while the bulk material is paramagnetic. Due to their quasi-2D, layered vdW-magnets do not only allow to investigate fundamental aspects of electronic correlation in structurally reduced dimensionality, but also hold a promise for technological applications, as demonstrated, e.g., by $Cr_2Ge_2O_6$/NiO heterostructures or $NiPS_3$-based field-effect transistors. Furthermore, the recent discovery of magnetic topological insulators (MTIs) in the $(MnBi_2Te_4)(Bi_2Te_3)$n (n = 0, 1, 2) family of vdW compounds has provided a rich experimental basis for the realization of new emerging physical phenomena such as the quantum anomalous Hall effect, the topological magnetoelectric effect, and majorana fermions emerging in MTIs due to a coexistence of the long-range magnetic order and the topologically nontrivial electronic band structure.

Organizers: Bernd Büchner (IFW Dresden), Rüdiger Klingeler (Heidelberg University)

Time: Monday 13:30–16:15 Location: H7

TT 4.1 Mon 13:30 H7

Topological states in $MnBi_2Te_4$-based magnetic van der Waals materials — •Hendrik Bentmann[1], Raphael. C. Vidal[1], Philipp Kagerer[1], Sebastian Buchberger[1], Celso Fornari[1], Anna Isaeva[2], and Friedrich Reinert[1] — [1]Experimentelle Physik VII and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg — [2]Van der Waals – Zeeman Institute, IoP, University of Amsterdam, 1098 XH Amsterdam, The Netherlands

Magnetic van der Waals materials down to a single monolayer have attracted considerable attention in recent years. In this talk, we will discuss electronic and magnetic properties of $MnBi_2Te_4$-based systems, in which Mn local moments and strong spin-orbit interaction of Bi and Te yield an interplay of magnetism and non-trivial band topology. Using angle-resolved photoemission and X-ray magnetic dichroism, we provide evidence that 3D $MnBi_2Te_4$ realizes an antiferromagnetic topological insulator [1]. Incorporation of non-magnetic Bi_2Te_3 spacer layers in $MnBi_4Te_7$ and $MnBi_6Te_{10}$ yields modified magnetic properties and surface-termination-dependent topological surface states [2]. In the 2D regime, $MnBi_2Te_4$ is a candidate for realizing the quantum anomalous Hall state. We will present ongoing efforts to grow $MnBi_2Te_4$ thin films using molecular beam epitaxy (MBE) [3].

[1] Nature 576, 416 (2019)
[2] Phys. Rev. X 9, 041065 (2019), Phys. Rev. Lett. 126, 176403 (2021)
[3] J. Appl. Phys. 128, 135303 (2020)

TT 4.2 Mon 14:00 H7

Static and dynamic magnetic properties of $(MnBi_2Te_4)(Bi_2Te_3)_n$ ($n = 0, 1$) probed by electron spin resonance technique. — •Alexey Alfonsov[1], Kavita Mehlawat[1,2], Jorge I. Facio[1], Ali G. Moghaddam[1,3], Rajyavardhan Ray[1], Alexander Zeugner[4,5], Manuel Richter[1,5], Anna Isaeva[1,6], Jeroen van den Brink[1,2,5], Bernd Büchner[1,2,5], and Vladislav Kataev[1] — [1]Leibniz IFW Dresden, 01069 Dresden, Germany — [2]Würzburg-Dresden Cluster of Excellence ct.qmat — [3]IASBS, Zanjan 45137-66731, Iran — [4]H.C. Starck Tungsten GmbH, 38642 Goslar, Germany — [5]TU Dresden, 01062 Dresden, Germany — [6]University of Amsterdam, 1098 XH Amsterdam, The Netherlands

$(MnBi_2Te_4)(Bi_2Te_3)_n$ ($n = 0, 1$) represent a family of van der Waals materials which exhibit a coexistence of topologically nontrivial surface states with intrinsic magnetism. Such unusual combination of properties renders this natural heterostructures very attractive for investigations since it enables a number of exotic phenomena. In this work we address static and dynamic magnetic properties of the title materials in the ordered and disordered states using multifrequency and high field electron spin resonance technique. We show that the spin dynamics of the magnetic building blocks of these compounds, the Mn-based septuple layers (SLs), is inherently ferromagnetic (FM) featuring persisting short-range FM correlations far above the magnetic ordering temperature as soon as the SLs get decoupled either by introducing a nonmagnetic quintuple interlayer, as in $MnBi_4Te_7$, or by applying a moderate magnetic field, as in $MnBi_2Te_4$.

TT 4.3 Mon 14:15 H7

Tuning Magnetic and Transport Properties in Quasi-2D $(Mn_{1-x}Ni_x)_2P_2S_6$ Single Crystals — •S. Aswartham[1], Y. Shemerliuk[1], Y. H. Zhou[2], Z. R. Yang[2], G. Cao[3], A.U.B. Wolter[1], and B. Büchner[1,4] — [1]Institut für Festkörperforschung, Leibniz IFW Dresden, Helmholtzstraße 20, 01069 Dresden, Germany — [2]Anhui Province Key Laboratory of Condensed Matter Physics at Extreme Conditions, High Magnetic Field Laboratory, Chinese Academy of Sciences, Hefei 230031, China — [3]Department of Physics, University of Colorado at Boulder, Boulder, CO 80309, USA — [4]Institut für Festkörper-und Materialphysik and Würzburg-Dresden Cluster of Excellence ct.qmat, Technische Universität Dresden, 01062 Dresden, Germany

We report an optimized chemical vapor transport method to grow single crystals of $(Mn_{1-x}Ni_x)_2P_2S_6$ where x = 0, 0.3, 0.5, 0.7, and 1. The structural characterization shows that all crystals crystallize in monoclinic symmetry with the space group $C2/m$ (No. 12). The magnetic measurements of the all as-grown single crystals show long range antiferromagnetic order along all principal crystallographic axes. Overall, the Néel temperature T_N is non-monotonous; with increasing Ni^{2+} doping, the temperature of the antiferromagnetic phase transition first decreases from 80 K for pristine ($Mn_2P_2S_6$ (x = 0) up to x= 0.5 and then increases again to 155 K for pure $Ni_2P_2S_6$ (x = 1). We show that, the magnetic anisotropy switches from out-of-plane to in-plane as a function of composition.

15. min. break

Invited Talk TT 4.4 Mon 14:45 H7

2D Magnetism and Its Efficient Control — •Cheng Gong — University of Maryland, College Park, USA

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 provide ideal platforms to enable the atomic-thin, flexible, lightweight magneto-optical and magnetoelectric devices. Though many have hoped that the ultra-thinness of 2D magnets should allow an efficient control of magnetism, the state-of-the-art has not achieved notable breakthroughs to this end. In this talk, I will speak on our experimental discovery of the first 2D ferromagnet, and discuss the strategies of the efficient control of 2D magnetism.

TT 4.5 Mon 15:15 H7

Coulomb-Engineered Magnetism in CrI_3 Monolayer — David Soriano, Alexander Rudenko, Mikhail Katsnelson, and •Malte Rösner — Radboud University, Nijmegen, Netherlands

We present a detailed study on the microscopic origin of magnetism in suspended and dielectrically embedded CrI_3 monolayer. To this end, we down-fold minimal generalized Hubbard models from *ab initio* calculations using the constrained random phase approximation. Within mean-field approximation, we show that these models are ca-

pable of describing the formation of localized magnetic moments in CrI_3 and of reproducing electronic properties of full *ab initio* calculations. We find a multi-orbital super-exchange mechanism as the origin of magnetism in CrI_3 resulting from a detailed interplay between effective ferro- and anti-ferromagnetic Cr-Cr d coupling channels, which is decisively affected by the ligand (I) p orbitals. We show how environmental screening such as resulting from encapsulation with hexagonal boron nitride (hBN) of the CrI_3 monolayer affects the Coulomb interaction in the film and how this successively controls its magnetic properties. Driven by a non-monotonic interplay between nearest and next-nearest neighbour exchange interactions we find the magnon dispersion and the Curie temperature to be non-trivially affected by the environmental dielectric screening.

TT 4.6 Mon 15:30 H7

Magnetoelastic coupling in the ferromagnetic van-der-Waals material CrI_3 — •Jan Arneth[1], Martin Jonak[1], Sven Spachmann[1], Mahmoud Abdel-Hafiez[2], Yaroslav Kvashnin[3], and Rüdiger Klingeler[1] — [1]Kirchhoff Institute of Physics, Heidelberg University, Germany — [2]Department of Physics and Astronomy, X-ray Photon Science, Uppsala University, Sweden — [3]Department of Physics and Astronomy, Materials Theory, Uppsala University, Sweden

We present high-resolution thermal expansion and magnetostriction studies on the layered van-der-Waals (vdW) ferromagnet CrI_3 in magnetic fields up to 15 T. Distinct anomalies in the thermal expansion coefficient at the ferromagnetic ordering temperature signal magnetoelastic coupling and allow us to quantify the uniaxial pressure dependencies $\partial T_C/\partial p_i$. While T_C reduces at a rate of -0.4 K/GPa upon the application of in-plane pressure, ferromagnetism is stabilized and the effect is about 4 times larger for out-of-plane pressure. The results are compared with numerical studies. We also observe macroscopic length changes associated with field-induced flipping of antiferromagnetically coupled surface layers in the magnetostriction data. We construct the magnetic phase diagram of bulk CrI_3 and show that magnetostriction is also sensitive to the saturation fields of the FM bulk and AFM surface phases.

TT 4.7 Mon 15:45 H7

Probing magnetic states in 2D layered van-der- Waals materials under pressure — Anirudha Gosh[1], Deobrat Singh[1], Qingge Mu[2], Sergey Medvedev[2], Rajeev Ahuja[1], Olle Eriksson[1], and •Mahmoud Abdel-Hafiez[1] — [1]Uppsala University, Department of Physics and Astronomy, Box 516, SE-751 20 Uppsala, Sweden — [2]Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany

Two-dimensional van der Waals materials offer a plethora of functional properties that are not only of fundamental interest but are essential for the development of new technological applications. Through combined complementary experimental techniques supplemented with theoretical calculations on high quality CrI3 single crystals, we derive a previously not discussed pressure-temperature phase diagram. TC increases to ~ 66 K with pressure up to ~ 3 GPa and then decreases to ~ 10 K at 21.2 GPa. The origin of this behavior is associated with a decrease in the calculated bond angle from 95^o at ambient pressure to ~ 85^o at 25 GPa. At a pressure above ~ 22 GPa, the magnetically ordered state is highly quenched, possibly driving the system to a Kitaev spin-liquid state at low temperature. Pressure-dependent Raman and resistivity measurements also reveal suppression of the phonon modes and semiconductor to metal transition, respectively above ~ 22 GPa. Furthermore, we will describe our recent experiments on CrCl3 single crystals.

TT 4.8 Mon 16:00 H7

Magnetic- and structural properties of α-$RuCl_3$ under hydrostatic He-gas pressure — •Bernd Wolf[1], Anja Wolter-Giraud[3], Gael Bastien[3], Anna Isaeva[4], David Kaib[2], Aleksandar Razpop[2], Kira Riedl[2], Sananda Biswas[2], Roser Valentí[2], Bernd Büchner[3], and Michael Lang[1] — [1]Physikalisches Institut, GU, SFB/TR 288, D-60438 Frankfurt (M) — [2]Institut für theoretische Physik, GU, SFB/TR 288, D-60438 Frankfurt (M) — [3]Leibniz-Institut für Festkörper- und Werkstoffforschung (IFW) Dresden, 01171 Dresden, Germany — [4]Fakultät für Chemie und Lebensmittelchemie, TUD, 01062 Dresden, Germany

α-$RuCl_3$ is a material to probe fundamental aspects of Kitaev physics despite the occurrence of magnetic order at low temperatures. We followed the idea that the suppression of magnetic order in α-$RuCl_3$ by using external parameters like magnetic field or pressure gives rise to a range where Kitaev physics prevails. We present magnetic susceptibility measurements on α-$RuCl_3$ single crystals under almost ideal hydrostatic pressure conditions. We find that the susceptibility strongly increases with increasing pressure. Furthermore, the magnetic ordering temperature T_N becomes rapidly reduced with pressure but cannot be fully suppressed to $T_N = 0$ due to the occurrence of a pressure-induced dimerization transition. We explain both results microscopically by employing a combination of first principles and finite-temperature Lanczos methods. Importantly, thorough investigations of the experimentally observed magnetic transition at varying pressure and magnetic fields reveal clear indications for a first order transition.