DF 17: Ferroics - Domains, Domain Walls and Skyrmions IV

Chairs: Claude Ederer and Petro Maksymovych

Time: Thursday 9:30–13:30 Location: WIL B321

Topical Talk DF 17.1 Thu 9:30 WIL B321

Magnetic and orbital excitations in the multiferroic skyrmion host GaV_4S_8 — Dieter Ehlers[1], Zhe Wang[1], Hans-Albrecht Krug von Nidda[1], Vladimir Tsurkan[1], Peter Lunkenheimer[1], Istvan Kézsmárki[2], Ioannis Stasinopoulos[3], Dirk Grundler[4], and •Alois Loidl[1] — [1]Experimental Physics V, Center for Electronic Correlations and Magnetism, University of Augsburg, Germany — [2]Department of Physics, Budapest University of Technology, Hungary — [3]Lehrstuhl für Physik funktionaler Schichtsysteme, Physik Department, TUM, Munich, Germany — [4]EPFL, Institut des matériaux, Lausanne, Switzerland

The lacunar spinel GaV_4S_8 exhibits a complex magnetic phase diagram, including ferromagnetic, cycloidal and Néel-type skyrmion phases [1,2]. Orbital ordering induces ferroelectricity [3] and spin-driven excess polarizations emerge in all magnetic phases, including skyrmions with ferroelectric polarization [2]. We present a detailed discussion on orbital and skyrmion excitations using THz and coplanar waveguide (CPW) absorption spectroscopy. At THz frequencies dipolar relaxations are strongly coupled to the orbital dynamics, establishing an orbitally-driven ferroelectric phase [3]. Using broadband CPW absorption spectroscopy we study the temperature and frequency dependence of skyrmion excitations [4].

[1] I. Kézsmárki *et al.*, Nature Materials **14**, 1116 (2015).
[2] E. Ruff *et al.*, Science Advances **1**, E1500916 (2015).
[3] Zhe Wang *et al.*, Phys. Rev. Lett. **115**, 207601 (2015).
[4] D. Ehlers *et al.*, Phys. Rev. B **94**, 014406 (2016).

DF 17.2 Thu 10:00 WIL B321

Investigating the structural phase transition of GaV_4S_8 — •Jonathan Döring[1], Ádám Butykai[2], Istvan Kézsmárki[2], Peter Milde[1], Susanne C. Kehr[1], and Lukas M. Eng[1] — [1]Technische Universität Dresden, Institut für Angewandte Physik — [2]MTA-BME Lendület Magneto-optical Spectroscopy Research Group

GaV_4S_8 (GVS) is a lacunar spinel. As most materials of this group, it features several magnetic phases at low temperatures, most notably a skyrmion lattice (SkL) phase between 9 K and 13 K under low magnetic fields [1]. Upon cooling below $T = 42$K, GVS exhibits a Jahn-Teller phase transition, changing the crystal structure from paraelectric cubic to ferroelectric rhombohedral. The multiferroic nature of the skyrmions offers interesting application perspectives in magnetoelectronics.

We study GVS by means of low-temperature piezoresponse force microscopy (LT-PFM) [2] and low-temperature scattering scanning near-field optical microscopy (LT-s-SNOM) [3] in order to characterize the ferroelectric domain configurations on the nanometer length scale. LT-PFM measurements are performed at temperatures both below and above the Jahn-Teller phase transition on as-grown (100) and (111) surfaces of single crystalline GVS. We observe several types of domain configurations in the rhombohedral phase. Applying LT-s-SNOM shows an enhanced response at wavelengths around $\lambda = 31.5\ \mu m$ close to a GVS phonon mode, enabling IR-imaging of the crystal anisotropy.

[1] Kezsmarki, I. et al., Nature Materials 14, 1116-1122 (2015)
[2] Butykai, A. et al., submitted
[3] Döring, J. et al., Applied Physics Letters 105, 053109 (2014)

DF 17.3 Thu 10:15 WIL B321

Searching for prospective multiferroic compounds hosting skyrmion lattices — •Erik Neuber[1], Peter Milde[1], Istvan Kézsmárki[2], and Lukas Eng[1] — [1]Institut für Angewandte Physik, TU Dresden, D-01069 Dresden, Germany — [2]Dep. of Physics, Budapest Univ. of Technol. and Econ. and MTA-BME Lendület Magneto-optical Spectroscopy Research Group, 1111 Budapest, Hungary

Following earlier predictions, skyrmion lattices (SkL), i.e. non-collinear periodic arrays of (chiral) spin vortices, are now reported to exist in various magnetic crystals of mostly chiral structure [1]. Nevertheless, also non-chiral but polar materials possessing the C_{nv} symmetry have been identified as ideal SkL-supporting hosts. GaV_4S_8 (GVS), a multiferroic polar magnetic semiconductor with rhombohedral (C_{3v}) symmetry, constitutes the first such candidate that has thoroughly been investigated [2]. GVS not only shows easy axis anisotropy that firmly pins SkL vortices even under external magnetic fields, but, furthermore, surprises with an unusally-broad temperature range that exhibits SkL ordering. Motivated by this great success, more such unusual SkL-materials with multiferroic properties must exist, even in the family of lacunar spinels [3]. In this contribution, we will briefly review on the real-space exploitation of such SkLs in GVS by scanning probe techniques, and report on our fascinating search for further such materials exhibiting a similar behavior to GVS.

[1] P. Milde et al., Science **340**, 1076 (2013). [2] I. Kézsmárki et al., Nat. Mater. **14**, 1116 (2015). [3] H.-S. Kim et al., Nat. Commun. **5**, 3988 (2014).

15 min. break

DF 17.4 Thu 10:45 WIL B321

Dielectric properties of organic charge-transfer salts — •Jonas K. H. Fischer[1], Peter Lunkenheimer[1], Rudra Manna[2], Harald Schubert[3], Michael Lang[3], Jens Müller[3], Stephan Krohns[1], John A. Schlueter[4], Cecile Mézière[5], Patrick Batail[5], and Alois Loidl[1] — [1]Experimental Physics V, EKM, University of Augsburg, Augsburg, Germany — [2]Experimental Physics VI, EKM, University of Augsburg, Augsburg, Germany — [3]Phys. Inst. Univ. Frankfurt, SFB/TR 49, Frankfurt, Germany — [4]Materials Research, National Science Foundation, Arlington, Virginia, United States — [5]Laboratoire MOLTECH, UMR 6200 CNRS-Université d'Angers, Bt. K, UFR Sciences, Angers, France

The EDT-TTF-based charge-transfer salts have attracted considerable attention due to their intriguing dielectric properties [1]. An example is κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl, which exhibits multiferroicity [2]. Its polar moment was suggested to arise from molecular dimerization, combined with charge order. Other interesting recent examples include α-(BEDT-TTF)$_2$I$_3$, which shows the signature of relaxor-ferroelectric behavior [1]. Here, we will present an overview of the dielectric properties of the above systems and provide new results on κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl, which also seems to show ferroelectric behavior in its charge-ordered state. In addition, we present measurements of δ-(EDT-TTF-CONMe$_2$)$_2$Br. This compound lacks dimerization, but exhibits charge order already at room temperature.

[1] P. Lunkenheimer and A. Loidl, J. Phys.: Condens. Matter **27**, 373001 (2015). [2] P. Lunkenheimer *et al.*, Nat. Mater. **11**, 755 (2012).

DF 17.5 Thu 11:00 WIL B321

Probing ferroelectricity in multiferroic $Dy_{0.7}Tb_{0.3}FeO_3$ using second harmonic generation — •Ehsan Hassanpour Yesaghi[1], Yusuke Tokunaga[2], Thomas Lottermoser[1], Yasujiro Taguchi[3], and Yoshinori Tokura[3,4] — [1]Department of Materials, ETH Zürich, Zürich, Switzerland — [2]Department of Advanced Materials Science, University of Tokyo, Kashiwa, Chiba 277-8561, Japan — [3]RIKEN Center for Emergent Matter Science (CEMS), Wako 351-0198, Japan — [4]Department of Applied Physics and Quantum-Phase Electronics Center (QPEC), University of Tokyo, Tokyo 113-8656, Japan

Multiferroic $Dy_{0.7}Tb_{0.3}FeO_3$ is reported to host a magnetically-driven spontaneous polarization below 2.65 K. This polarization emerges as a result of exchange interaction between Fe^{3+} spins and rare-earth moments. At about 2 K the polarization vanishes due to the change in the magnetic phase of iron (ferromagnetic to antiferromagnetic). Some models have been proposed on the magnetoelectric coupling in this material but a profound experimental study is yet to be done. Here we investigate the magnetic and electric order at the level of domains using spatially resolved techniques. We employ optical Faraday effect and second harmonic generation (SHG) to investigate the magnetic and electric order respectively. Our results reveal that the transition at 2 K is not sharp and the two types of ordering for the Fe^{3+} spins coexist simultaneously from which only one (ferromagnetic phase) contributes to the spontaneous polarization. The volume ratio of these two phases, and hence the amplitude of the ferroelectric polarization, can be controlled via an external magnetic field.

DF 17.6 Thu 11:15 WIL B321

Strong magnetoelectric coupling within ceramic core-shell

structures — •Leonard Henrichs[1], Torsten Scherer[1], James Benett[2], Andrew Bell[2], Oscar Cespedes[2], and Christian Kübel[1] — [1]Karlsruhe Insitute of Technology, Karlsruhe, Germany — [2]University of Leeds, Leeds, United Kingdom

In perovskite ceramics of the composition $BiFe_{0.9}Co_{0.1}O_3)_{0.4}$ – $Bi_{1/2}K_{1/2}TiO_3)_{0.6}$, novel nano-sized regions called multiferroic clusters (MFC) were recently discovered. These MFC belong to so-called core-shell structures as known from other relaxor ferroelectrics, where $BiFe_{1-x}Co_xO_3$-rich cores are surrounded by a $Bi_{1/2}K_{1/2}TiO_3$-rich shell within one grain. The MFC exhibit exceptionally large direct and converse local ME coupling. The observed electric-field induced switching of magnetization is especially interesting in terms of applications, since it enables in principle electrically driven magnetic memory, one of the 'holy grails' in information technology research. It is assumed that the strong magnetism stems from ferrimagnetic order of Fe and Co in MFC, which requires a superstructure of Fe and Co on the B lattice site. The main unsolved question in this system is, why the exceptional multiferroic properties occur in the $BiFe_{1-x}Co_xO_3$-rich Cores, but have never been observed in pure $BiFe_{1-x}Co_xO_3$ compounds. An explanation might be epitaxial strain originating from the core-shell structure. It is anticipated, that deeper understanding of the MFC might give valuable insights for the design e.g. of a thin-film material with similar multiferroic properties like the MFC.

DF 17.7 Thu 11:30 WIL B321

Nuclear magnetic and electric interactions in multiferroic $Ba_2CoGe_2O_7$ — •Martina Schädler[1], Titusz Fehér[2], Norbert Büttgen[1], Vilmos Kocsis[2], Yoshinori Tokura[3], Yasujiro Taguchi[3], Alois Loidl[1], and István Kézsmárki[2] — [1]Center for Electronic Correlations and Magnetism, Institute of Physics, University of Augsburg, Germany — [2]Department of Physics, Budapest University of Technology and Economics, Hungary — [3]RIKEN Center for Emergent Matter Science, Wako 351-0198, Japan

In the multiferroic compound $Ba_2CoGe_2O_7$ the electric polarization is induced magnetically by the spin-dependent hybridization mechanism. As a result the polarization arises locally inside the CoO_4 tetrahedra and is dependent on the orientation of the external magnetic field H. Nuclear Magnetic Resonance (NMR) allows access to the local electric field gradient (EFG) via the nuclear quadrupole moment. We performed ^{59}Co NMR measurements on $Ba_2CoGe_2O_7$ for various orientations of the applied field in order to determine the local microscopic properties of magnetic spin order and electric polarization at the cobalt site: (1) we were able to model the local hyperfine field at the two magnetically inequivalent cobalt sites and (2) observed the displacement of the surrounding ions by the NMR quadrupolar effect.

DF 17.8 Thu 11:45 WIL B321

The effect of strain on the magnetic and ferroelectric properties of orthorhombic $TbMnO_3$ — •Amadé Bortis[1], Natalya Fedorova[2], Alessandro Vindigni[3], Andrea Scaramucci[2], and Nicola Spaldin[2] — [1]Laboratory for Multifunctional Ferroic Materials (M. Fiebig), Departement of Materials, ETH Zurich, Switzerland — [2]Materials Theory, Departement of Materials, ETH Zurich, Switzerland — [3]Microstructure Research, Departement of Physics, ETH Zurich, Switzerland

We model the effect of strain on magnetic and ferroelectric properties of orthorhombic $TbMnO_3$ using ab initio electronic structure calculations and Monte-Carlo simulations. $TbMnO_3$ is a magnetoelectric multiferroic, where the ferroelectricity is triggered by the magnetic ordering, which allows manipulating the electric polarization by a change in the magnetic order. It was shown experimentally that bulk samples of $TbMnO_3$, at low temperature, show a spiral magnetic order, which drives a weak polarization. Recent experiments showed that by applying strain one can change the magnetic order to E-AFM, which leads to a higher polarization. In order to understand the transition from spiral to E-AFM order, we perform ab initio calculations to extract the relevant exchange couplings for a bulk and strained sample, and we use these couplings to perform Monte-Carlo simulations to find the corresponding ground state spin configuration. This allows the determination of the magnetic order and the investigation of its effect on the polarization. We found that applying strain can drive the transition from spiral to E-AFM ordering and can enhance the polarization.

15 min. break

Topical Talk DF 17.9 Thu 12:15 WIL B321

Role of charged defects on conduction and dynamics of domain walls in $BiFeO_3$ — •Tadej Rojac[1], Andreja Bencan[2], Goran Drazic[2], Naonori Sakomoto[2], Hana Ursic[2], Bostian Jancar[2], Gasper Tavcar[2], Maja Makarovic[2], Julian Walker[2], Barbara Malic[2], and Dragan Damjanovic[2] — [1]Electronic Ceramics Department, Jozef Stefan Institute, 1000 Ljubljana, Slovenia — [2]See author list of the paper in Nature Materials

Domain walls in ferroelectrics tend to interact with charged point defects, such as oxygen vacancies, resulting in pinning effects. In practice, this "hardening" mechanism represents one of the most important ways of controlling properties in ferroelectrics. For example, doping $Pb(Zr,Ti)O_3$ or $BaTiO_3$ with an acceptor will create oxygen vacancies which by forming re-orientable defect complexes act as pinning sites for domain walls, affecting profoundly the switching behavior and piezoelectric response. It is widely accepted that oxygen vacancies play the major role in pinning effects and hardening. It has been recently established that undoped polycrystalline $BiFeO_3$ behaves as a "hard" ferroelectric. Using atomic-scale structural and chemical analysis, we will show that, in contrast to the usually assumed oxygen vacancies, the dominant defects in $BiFeO_3$ mainly responsible for the pinning effect are electron holes, associated with the presence of Fe^{4+}, and bismuth vacancies. Direct identification of these charged defects using Cs-corrected microscopy also showed that they have a tendency to accumulate in the domain wall region, revealing the p-type hopping conduction at domain walls associated with Fe4+ defects. Discussion will be provided on how this local conductivity affects domain-wall dynamics and thus the piezoelectric response of $BiFeO_3$.

Rojac, Tadej, et al. "Domain-wall conduction in ferroelectric $BiFeO_3$ controlled by accumulation of charged defects." Nature Materials (2016)

DF 17.10 Thu 12:45 WIL B321

Mechanically soft domain walls in hard ferroelectrics — •Neus Domingo, Kumara Cordero-Edwards, James Zapata, and Gustau Catalan — Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and The Barcelona Instituteof Science and Technology, Campus UAB, Bellaterra, 08193 Barcelona, Spain

All ferroic materials can display regions (domains) with different polarity of the order parameter. The boundaries between domains are known as domain walls. Domain walls may possess functional properties not existent in the host material, such as conductivity in the walls of insulators, ferromagnetism in the walls of antiferromagnets, or polarization in the walls of ferroelastics. This could potentially be used to make new electronic devices at an unprecedented small scale, where the "active ingredient" are not the domains but the domain walls.

Among the many properties of domain walls, mechanical response appears to have been largely neglected, and there are very few, if any, studies specifically aimed at determining the local mechanical properties of domain walls.

In this presentation, we will show our first experimental measurements of the stiffness of domain walls in ferroelectric lithium niobate and related perovskite ferroelectrics, as measured by atomic force microscopy using a mechanical resonance spectroscopy mode. The key result is that even purely ferroelectric (non-ferroelastic) 180 degree domain walls in uniaxial ferroelectrics are considerably softer than the domains they separate.

DF 17.11 Thu 13:00 WIL B321

Reversibly tuning the domain wall conductivity in lithium niobate: from insulating to metallic-like and back again — •Christian Godau, Alexander Haussmann, and Lukas Eng — Institute of Applied Physics, Technische Universität Dresden, D-01062 Dresden, Germany

Ferroelectric domain walls (DWs) have become a central topic of research. Especially their tunable electronic properties stay in focus for the last couple of years, reporting domain wall conductivity (DWC) in both thin films [1] and single crystals [2]. Our recent research established DWC in lithium niobate (LNO) as promising building blocks for prospective electronic devices [3]; as theoretically predicted [4], DWs in fact become more conductive the larger their inclination angle is rendered. We will present here protocols how to fabricate such highly-conductive transport channels. In fact, our high-voltage treatment allows reversibly switching the conductive channels on and off, i.e. varying the inclination angle on will. This allows to fabricate a variety of novel nanoelectronic devices, for instance memory devices based on resistive switching [5] of DWC, or unidirectional / bidirectional diode/Ohmic-like 2-dimensional transport channels across wide

band-gap semiconductors.

[1] J. Seidel et al., Nat. Mater. 8, 229 (2009) [2] T. Sluka et al., Nat. Comm. 4, 1808 (2013) [3] C. Godau et al., (2016) submitted [4] E. A. Eliseev et al., Phys. Rev. B 83, 235313 (2011) [5] R. Waser et al., Adv. Mater. 21, 2632 (2009)

DF 17.12 Thu 13:15 WIL B321

Imaging of multiferroic domains at the optical resolution limit — •Stefan Günther, Martin Lilienblum, Thomas Lottermoser, and Manfred Fiebig — Department of Materials, ETH Zürich, Zürich, Switzerland

Multiferroics accommodate strongly coupled electric and magnetic ordering. In a fundamental work on the hexagonal manganites (h-$RMnO_3$) from 2002, it was shown, that this coupling can extend to the electric and magnetic domain structure [1]. However, in those measurements the coupling was not resolved down to the archetypal ferroelectric vortex domain structure that was established for this system a decade later. In this talk, both type of domains (ferroelectric and antiferromagnetic) are resolved with second harmonic generation (SHG) in multiferroic h-$ErMnO_3$. As verified by piezo-response force microscopy the compound shows ferroelectric as-grown domains in the few μm-size and can thus be imaged with SHG close to the optical resolution limit. The measurements show the ferroelectric vortex domain structure, a partial coupling of magnetic and electric domain walls and give significantly new insights in the fine structure of antiferromagnetic domain walls. In particular, they demonstrate a thermally induced instability of these walls.

[1] M. Fiebig et al., Nature **419**, 818 (2002)