DS 19: Multiferroics II (jointly with MA, DF,KR, TT)

Time: Wednesday 9:30–13:00 Location: HSZ 04

DS 19.1 Wed 9:30 HSZ 04

An Engineered Polar Oxide Heterostructure Built from Isosymmetric Magnetically Ordered Components — •Matthew S Dyer[1], Jonathan Alaria[1], Pavel Borisov[1,5], Troy D Manning[1], Serban Lepadatu[2], Markys G Cain[2], Elena D Mishina[3], Natalia E Sherstyuk[3], N A Ilyin[3], Joke Hadermann[4], David Lederman[5], John B Claridge[1], and Matthew J Rosseinsky[1] — [1]University of Liverpool, Liverpool, UK — [2]National Physical Laboratory, Teddington, UK — [3]Moscow State Technical University, Moscow, Russia — [4]University of Antwerp, Antwerp, Belgium — [5]West Virginia University, Morgantown, USA

Theory predicts that certain layered heterostructures consisting of perovskite blocks have non-centrosymmetric structures. The breaking of spatial inversion symmetry arises through a combination of octahedral tilting and A site ordering. Following this prediction, we grow a thin-film of the $[(YFeO_3)_5(LaFeO_3)_5]_{40}$ heterostructure using RHEED monitored pulsed layer deposition. Polar domains are present in the thin-film, as demonstrated by second harmonic generation and piezoelectric force microscopy measurements. We experimentally confirm that the heterostructure is also magnetically ordered at room temperature with a finite magnetization, retaining the magnetic structure of the individual $YFeO_3$ and $LaFeO_3$ components.

DS 19.2 Wed 9:45 HSZ 04

First-principles study of the $BaTiO_3/BaFeO_3$ perovskite interface — •Igor Maznichenko[1], Sergey Ostanin[2], Arthur Ernst[2], and Ingrid Mertig[1,2] — [1]Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, D-06099 Halle, Germany — [2]Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

Epitaxial growth can combine a robust ferroelectric, such as $BaTiO_3$, and strong ferromagnets into the so called composite multiferroic films. The switching properties of artificial multiferroics sandwiched between metallic contacts make them excellent candidates for the room temperature four-state memories.

Regarding the ferromagnetic side of composite multiferroics, we suggest to use the cubic perovskite $BaFeO_3$ whose epitaxial growth has been recently reported. Here, from the basis of *ab-initio* electronic structure calculations, within the Korringa-Kohn-Rostoker method, we study the magnetic properties of bulk $BaFeO_3$. The approach allows us to accurately monitor the evolution of the Curie temperature upon both the tetragonal distortions and presence of oxygen vacancies. Finally, we examine magnetoelectricity at the $BaTiO_3/BaFeO_3$ interface.

DS 19.3 Wed 10:00 HSZ 04

Behaviour of Raman modes in $BiFeO_3$ epitaxial thin films with respect to azimuthal orientation — •Andreas Talkenberger[1], Cameliu Himcinschi[1], Ionela Vrejoiu[2,3], Florian Johann[2], and Jens Kortus[1] — [1]TU Bergakademie Freiberg, Institute of Theoretical Physics, Leipziger Str. 23, D-09596 Freiberg, Germany — [2]Max Planck Institute of Microstructure Physics, Weinberg 2, D-06120 Halle, Germany — [3]Max Planck Institute for Solide State Research, Heisenbergstr. 1, D-70569 Stuttgart, Germany

$BiFeO_3$ (BFO) is an interesting candidate for multiferroic applications. In this work we focus on the Raman spectroscopic investigation of epitaxially grown thin films of BFO by pulsed laser deposition on different substrates, belonging to the group of scandates ($DyScO_3$, $SmScO_3$, $GdScO_3$). The Raman spectra were recorded using the 442 nm emission line of a He-Cd laser. Some phonon modes show changes in the position, full width at half maximum (FWHM) and intensity depending on the azimuthal angle. We found a 90 degree periodicity of the peak position and of the FWHM for particular modes. For both parallel and crossed polarisation the four maxima in positions correspond to the minima in FWHM. Such a behaviour can be explained considering a twin family of domains with a very well defined orientation to each other. Our results are supported by piezoresponse-force microscopy and X-ray diffraction measurements as well.

DS 19.4 Wed 10:15 HSZ 04

$BiFeO_3/LaFeO_3$: a magnetoelectric multiferroic — •Zeila Zanolli[1,3], Jacek Wojdel[2], Jorge Iniguez[2], and Philippe Ghosez[3] — [1]Forschungszentrum Jülich, PGI and IAS, Jülich, Germany — [2]ICMAB-CSIC, Bellaterra, Spain — [3]Université de Liège, Physics Department, Liegé, Belgium

Transition-metal oxides of perovskite structure present a wide variety of physical properties. In particular, there is a strong interest in multiferroic materials that are simultaneously ferroelectric and magnetic (*magnetoelectrics*). Due to the scarcity of natural magnetoelectric multiferroics and thanks to recent advances in epitaxial growth techniques, designing new magnetoelectric multiferroic heterostructures is a promising way to succeed in this quest.

First-principles techniques are used to investigate electric control of the magnetization in the $BiFeO_3/LaFeO_3$ perovskite oxide superlattice (SL) on a (001)-$SrTiO_3$ substrate. Our results [1] show that the $BiFeO_3/LaFeO_3$ SL exhibits a trilinear coupling of a polar mode with two different rotations of the oxygen cages (*hybrid improper ferroelectricity*). Non-collinear spin calculations reveal that the ferroelectric ground state also presents weak ferromagnetism with easy axis along the [1 -1 0] direction. The microscopic mechanism allowing one to manipulate the magnetization with an electric field in such systems is presented and its dependence on strain and chemical substitution is discussed. The $BiFeO_3/LaFeO_3$ SL is found to be a good candidate to attain electric switching of magnetization at room temperature.

[1] Phys. Rev. B **88**, 060102(R) (2013)

DS 19.5 Wed 10:30 HSZ 04

The influence of strain on the optical properties of pseudo-tetragonal $BiFeO_3$ thin films — •Cameliu Himcinschi[1], Akash Bhatnagar[2], Andreas Talkenberger[1], Dietrich R.T. Zahn[3], Jens Kortus[1], and Marin Alexe[2,4] — [1]TU Bergakademie Freiberg, Institute of Theoretical Physics, D-09596 Freiberg, Germany — [2]Max Planck Institute of Microstructure Physics, Weinberg 2, D-06120 Halle, Germany — [3]Semiconductor Physics, Technische Universität Chemnitz, D-09107 Chemnitz, Germany — [4]Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom

Tetragonally distorted $BiFeO_3$ recently attracted a lot of attention because of its interesting multiferroic properties and the larger spontaneous polarization as compared to its rhombohedral counterpart. Highly strained (when grown on $LaAlO_3$ substrates) and nearly pseudomorphic (when grown on $TbScO_3$ substrates) $BiFeO_3$ films were deposited by pulsed laser deposition. The symmetry of the tetragonally distorted $BiFeO_3$ films is discussed based on polarisation dependent Raman measurements and the comparison with Raman spectra measured for films deposited on $TbScO_3$. The evaluation of ellipsometric spectra reveals that the films deposited on $LaAlO_3$ are optically less dense and the dielectric function is blue-shifted by more than 0.3 eV as compared to the films deposited on $TbScO_3$. By analyzing the absorption edge using a bandgap model, bandgaps of 3.10 eV and 2.75 eV were determined for the films deposited on $LaAlO_3$ and $TbScO_3$, respectively. This work is supported by the German Research Foundation DFG HI 1534/1-1.

DS 19.6 Wed 10:45 HSZ 04

Electrically induced magnetic transiton at the LSMO/BTO interface — •Markus Schmitz, Alexander Weber, Paul Zakalek, Markus Waschk, and Thomas Brückel — Jülich Centre for Neutron Science JCNS and Peter Grünberg Institut PGI, JARA-FIT, Forschungszentrum Jülich GmbH, 52425 Jülich Germany

The magnetoelectric coupling is one of the most fascinating and active research areas today. The control of the magnetism due to an applied electric field may lead to new device concepts. First principles calculations of $La_{(1-x)}Sr_xMnO_3/BaTiO_3(001)$ interfaces show magnetic reconstructions due to the change of the polarization of BTO by applying an external electric field. The different electron densities influence the equilibrium between super- and double-exchange favoring a ferromagnetic or an antiferromagnetic order at the interface for the two different orientations of the polarization. Here we report on LSMO/BTO, grown with an Oxide Molecular Beam Epitaxy system. The epitaxial layer-by-layer growth was confirmed by in-situ RHEED analysis and the crystalline quality of the surface was investigated by LEED and Atomic Force Microscopy. The structural characterization was carried out by X-ray reflectometry and diffraction. We could prove the possibility to electrically polarize $BaTiO_3$ substrates due to an ap-

plied voltage of 400V by optical methods. The macroscopic magnetic properties were determined by MOKE and SQUID magnetometry. The magnetic formation at the interface with respect to the polarization of the $BaTiO_3$ was investigated by Polarized Neutron reflectometry measurements performed at MARIA (FRM II).

15 min. break

DS 19.7 Wed 11:15 HSZ 04

Growth and structure characterization of double perovskite Sr2FeMoO6 thin films — •Hakan Deniz[1], Dietrich Hesse[1], Marin Alexe[1], Robert Lowndes[2], and Lucian Pintilie[2] — [1]Max-Planck Institute of Microstructure Physics, Weinberg 2, D-06120, Halle (Saale), Germany — [2]National Institute of Materials Physics, Atomistilor 105bis, Magurele 077125, Romania

The double perovskite Sr2FeMoO6 (SFMO) has drawn considerable attention recently owing to some of its unique features such as high Curie temperature (˜410K) and half-metallic ferrimagnetic nature with a high saturation moment of 4μB. The low-field room temperature magnetoresistance observed in SFMO makes it an attractive candidate for oxide spintronics applications. However, the broad distribution of results reported so far on SFMO films suggests that an optimal structure is attainable only within a narrow window of growth conditions; and magnetic/transport properties are highly akin to Fe and Mo atomic site disorder. Pulsed laser deposition was employed to grow SFMO thin films on vicinal SrTiO3 substrates from a custom-made stoichiometric target using argon as ambient gas. X-ray diffraction data revealed that the SFMO films were grown epitaxially with respect to the substrate, including, however, a small percentage of secondary phases. The morphology of the films shows flat plains with embedded grain- or needle-like structures, which are most likely the result of spurious phases. The nature of these defects and their interfaces with the SFMO matrix are under investigation by transmission electron microscopy. This work is supported by the EU-FP7 project IFOX.

DS 19.8 Wed 11:30 HSZ 04

Magnetic Anisotropy in Multiferroic Lu_2MnCoO_6 — •Martin Lonsky[1], Merlin Pohlit[1], María Antonia Señarís Rodríguez[2], and Jens Müller[1] — [1]Physikalisches Institut, Goethe-Universität, Frankfurt (M), Germany — [2]Dpto. Química Fundamental U. Coruña, Coruña, Spain

Lu_2MnCoO_6 recently has been introduced as a new type-II multiferroic with ferroelectricity due to charge ordering and magnetostriction related to magnetic Mn^{4+} and Co^{2+} ions which are arranged alternately in the form of Ising chains along the c-axis of the crystal [1]. The magnetic properties, however, remain puzzling, which in particular is due to the lack of measurements on single crystals, that have not yet successfully been synthesized. Here, we present for the first time measurements of the magnetic anisotropy by employing micro-Hall magnetometry on a few micrograins of dimensions $\sim 1\,\mu$m only. Our results reveal a strong dependence of magnetic hysteresis on temperature and the applied field direction. This anisotropy is also reflected in the observation of a variety of unusual effects as for instance wasp-waisted hysteresis loops, sharp jumps in magnetization at about $T = 300$ mK and an exchange bias, ocurring in each case in only one field direction. Additionally, the observation of a pronounced maximum in the coercive field at $T_{SF} \sim 12$ K indicates a significant change in the spin dynamics of the system below T_{SF}, similar to the behavior of the related compound $Ca_3Co_{2-x}Mn_xO_6$ ($x \approx 0.95$) [2].

[1] S. Yáñez-Vilar et al., Phys. Rev. B. 84, 134427 (2011).
[2] T. Lancaster et al., Phys. Rev. B 80, 020409 (2009).

DS 19.9 Wed 11:45 HSZ 04

The multiferroic $CuCrO_2$ compound: interlayer exchange and domain population — •Matthias Frontzek — Laboratory for Neutron Scattering, Paul Scherrer Institut, 5232 Villigen-PSI, Switzerland

Multiferroic materials have become of interest for their unusual low-temperature properties in general, and the tunability of the magnetic structure through an electric field and the electric polarization through a magnetic field in particular. The most promising candidates for such controllable multiferroics have been found among the materials with inherent geometric magnetic frustration.

Among these, the delafossite $CuCrO_2$, which crystallizes in the rhombohedral $R\bar{3}m$ space group, is a multiferroic compound with an apparent strong coupling of spin and charge. In contrast to other multiferroic compounds $CuCrO_2$ shows a spontaneous electric polarization upon antiferromagnetic ordering without an accompanying structural phase transition, thus the magnetic ordering alone breaks the inversion symmetry. The peculiar magnetic structure of $CuCrO_2$ allows the direct quantitative analysis of the domain population.

In our contribution, we present a detailed study on $CuCrO_2$ single crystals using neutron diffraction in applied electric and magnetic fields. With the fields we were able to tune the multiferroic states in $CuCrO_2$ and could directly relate them to the underlying domain physics. Our results allow the re-interpretation of macroscopic measurements and show that the $p-d$ hybridization is the dominant spin-charge coupling mechanism.

DS 19.10 Wed 12:00 HSZ 04

Structure and Magnetic Coupling in $YBaFeCuO_5$ — •Andrea Scaramucci[1], Mickael Morin[2], Ekaterina Pomjakushina[2], Marek Bartkowiak[2], Denis Sheptyakov[2], Lukas Keller[2], Juan Rodriguez-Carvajal[3], Michel Kenzelmann[2], Kazimierz Conder[2], Marisa Medarde[2], and Nicola A. Spaldin[1] — [1]Materials Theory, ETH-Zurich, Zurich, Switzerland — [2]Paul Scherrer Institute, Villigen, Switzerland — [3]Institute Laue Langevin, Grenoble, France

We theoretically study the structure and exchange couplings in multiferroic $YBaFeCuO_5$ (YBFCO). Using density functional theory we calculate energies of configurations with various Fe^{3+}/Cu^{2+} orderings in the bilayered perovskite structure of YBFCO. We find that configurations with different distribution of Fe^{3+} and Cu^{2+} ions fall into two groups with distinctly different energies. The energies of those in the lowest energy group are close to that of the ground state (relative to the growth temperature) suggesting Fe^{3+} and Cu^{2+} to be disordered in YBFCO. Finally, we calculate exchange coupling constants for all the low energy configurations and show that the magnetic ordering resulting from these couplings is compatible with the experimentally-observed high-temperature magnetic ordering. However, they do not explain the existence of the experimentally observed low-temperature incommensurate magnetic structure.

DS 19.11 Wed 12:15 HSZ 04

Hybrid-functional study of the structural, magnetic and electronic properties of rare-earth nickelates — •Konstantin Z. Rushchanskii, Stefan Blügel, and Marjana Ležaić — Peter Grünberg Institut, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

Rare-earth nickelates ($ReNiO_3$) are very promising functional perovskite crystalline materials, exhibiting metal-insulator (MI) transition, which can be continuously controlled by composition, bi-axial strain and(or) electric field. Unfortunately, conventional *ab initio* DFT+U results fail to reproduce their magnetic ground state as well as the effect of epitaxial strain on MI transition temperature. We present results of our comprehensive study of structural, magnetic and electronic properties of bulk $ReNiO_3$ (Re=Y, Gd, Eu, Sm, Nd and Pr) and strained $SmNiO_3$ films [1], performed with HSE06 functional. We show correlation between MI transition temperature and structural parameters of bulk and films, which nicely fits known experimental data. We also analyze the difference in the electronic structure obtained in DFT+U and with the hybrid functional and their influence on the resulting magnetic ordering in the ground state.

We acknowledge the support by Helmholtz Young Investigators Group Programme VH-NG-409, JSC and JARA-HPC.

[1] F.Y. Bruno, K.Z. Rushchanskii, S. Valencia, Y. Dumont, C. Carrétéro, E. Jacquet, R. Abrudan, S. Blügel, M. Ležaić, M. Bibes, and A. Barthélémy, Phys. Rev. B 88, 195108 (2013).

DS 19.12 Wed 12:30 HSZ 04

Magnetic properties of multiferroic $TbMnO_3$ — •Natalya Fedorova, Andrea Scaramucci, Claude Ederer, and Nicola Spaldin — ETH Zurich, Materials Theory, Wolfgang-Pauli-Strasse 27, CH-8093 Zurich, Switzerland

We use ab-initio calculations to investigate the magnetic properties of multiferroic $TbMnO_3$.

At low temperatures $TbMnO_3$ demonstrates an incommensurate spiral ordering of Mn spins which is accompanied by appearance of spontaneous electric polarization driven by applied magnetic field [1]. The establishment of such spin ordering is usually described within the framework of a Heisenberg model with competing nearest-neighbor and next-nearest-neighbor exchange interactions. However, our theoretical estimations of these interactions by ab-initio calculations demonstrate a clear deviation from Heisenberg model.

We consider first the coupling between magnetic and orbital orderings as a main source of non-Heisenberg behavior in $TbMnO_3$, but conclude that it does not explain the observed deviation. We find that higher order interactions (biquadratic and four-body spin couplings) should be taken into account for proper treatment of the magnetism in $TbMnO_3$.

[1] T. Kimura et al., Nature 426, 55-58 (2003)

DS 19.13 Wed 12:45 HSZ 04

Coupling of epitaxial strain and point-defect formation in perovskites — •Ulrich Aschauer, Philipp Baumli, and Nicola A. Spaldin — Materials Theory, ETH Zurich, Zürich, Switzerland

Using density functional theory calculations we recently established the existence of a strong coupling between epitaxial strain and the formation energy of oxygen vacancies in the model perovskite $CaMnO_3$ (Phys. Rev. B. 88, 054111, 2013). Here we investigate the generality of this concept for other oxides including metallic perovskites and also investigate the effect of strain on the formation of cation vacancies. We find that in general the response of the defect profile follows the behavior expected from chemical-expansion arguments, with tensile strain favoring oxygen vacancies and compressive strain favoring cation vacancies. We show, however, that material-specific details of the electronic structure can cause deviations from this trend under both tensile and compressive strain.