DS 17: Multiferroics III: Strain / New routes towards multiferroicity (jointly with MA, DF, KR, TT)

Time: Tuesday 9:30–12:45 Location: EB 301

DS 17.1 Tue 9:30 EB 301

Substrate influence on the strain in epitaxially grown $BiCrO_3$ thin films investigated using Raman spectroscopy and X-ray diffraction — •Andreas Talkenberger[1], Cameliu Himcinschi[1], Kannan Vijayanandhini[2], David Rafaja[3], Ionela Vrejoiu[2], Torsten Weissbach[1], Christian Röder[1], and Jens Kortus[1] — [1]TU Bergakademie Freiberg, Institute of Theoretical Physics, D-09596 Freiberg — [2]Max Planck Institute of Microstructure Physics, Weinberg 2, D-06120 Halle — [3]TU Bergakademie Freiberg, Institute of Materials Science, D-09596 Freiberg

$BiCrO_3$ (BCO) is an interesting candidate for multiferroic applications. Therefore a deep understanding of the material properties and the fabrication of high quality epitaxial thin films is necessary. In this work we investigated epitaxially grown BCO thin films fabricated by pulsed laser deposition on $SrTiO_3$, LSAT, $NdGaO_3$ and $DyScO_3$ by means of Raman spectroscopy and X-ray diffraction (XRD). The shift of phonon modes at room temperature indicates different strains in the BCO films grown on the different substrates. Primarily, the XRD experiments helped to quantify the elastic lattice strains caused by the lattice misfit between the substrate and the thin films. The reciprocal space mapping was employed to follow the relaxation of the lattice strain through the formation of microstructure defects. This data was correlated to the observed Raman shifts. Using density functional theory the shifts of the Raman peaks were calculated for different strain states, and compared to the experimentally observed ones. This work is supported by the German Research Foundation DFG HI 1534/1-1.

DS 17.2 Tue 9:45 EB 301

Directly probing the effect of strain on magnetic exchange interactions — •Kathrin Dörr[1,2], Andreas Herklotz[2], Hans-Martin Christen[3], and Michael Biegalski[3] — [1]MLU Halle Wittenberg, Von-Danckelmann-Platz 3, 06120 Halle — [2]IFW Dresden, Postfach 270116, 01171 Dresden — [3]CNMS, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA

Thin films of transition metal oxides of the perovskite type ABO_3 (B = 3d or 4d metal) have revealed abundant examples for strain-driven changes of magnetic ordering. In spite of strong efforts, the theoretical treatment of magnetic exchange in complex oxides has remained a challenge, and experiments continue to show unpredicted large effects of the epitaxial strains in films. In order to provide meaningful experimental data on strain dependences, epitaxial thin films should be grown in various coherent strain states on different substrates without changing anything but the strain. This is inherently difficult: possible problems arise from a strain-dependent oxidation level or microstructure. As a complementary approach, the in-plane strain of epitaxial oxide films can be controlled reversibly by 0.1-0.2 percent using a piezoelectric substrate. I will address reversible-strain studies on $La_{0.7}Sr_{0.3}MnO_3$, $La_{1-x}Sr_xCoO_3$ (x = 0, 0.2, 0.3) und $SrRuO_3$ films, showing the strain response of the magnetic Curie temperature and the magnetization and discussing the current understanding of the strain effects on magnetic ordering.

DS 17.3 Tue 10:00 EB 301

Induced magnetoelectric response in Pnma perovskites — •Eric Bousquet and Nicola Spaldin — Materials Department, ETH Zurich, Switzerland

We use symmetry analysis to show that the G, C and A-type antiferromagnetic *Pnma* perovskites can exhibit magnetoelectric (ME) responses when a ferroelectric instability is induced with epitaxial strain. Using first-principles calculations we compute the values of the allowed ME response in strained $CaMnO_3$ as a model system. Our results show that large linear and non-linear ME responses are present and can diverge when close to the ferroelectric phase transition. By decomposing the electronic and ionic contributions, we explore the detailed mechanism of the ME response.

DS 17.4 Tue 10:15 EB 301

Search for strain-induced ferroelectricity in EuO films — •Carsten Becher[1], Masakazu Matsubara[1], Andreas Schmehl[2], Jochen Mannhart[3], Darrell G. Schlom[4], and Manfred Fiebig[1] — [1]Department of Materials, ETH Zürich, Switzerland — [2]Institut für Physik, Universität Augsburg, Germany — [3]Max Planck Institute for Solid State Research, Germany — [4]Department of Materials Science and Engineering, Cornell University, USA

Ferromagnetic EuO arouses a lot of interest due to a multitude of extreme properties, such as an insulator-metal transition, a colossal-exceptional magnetoresistance effect, and nearly 100 % spin polarization of the conduction electrons in the ferromagnetic state. In addition, recent theories predict that EuO becomes ferroelectric under epitaxial strains $\geq$ 4.2 %, suggesting a route to novel multiferroics combining ferromagnetic and ferroelectric order. Here, we use optical second harmonic generation (SHG) to detect changes of the electric as well as magnetic order of EuO thin films. In search of a strain-induced spontaneous polarization, we vary the sample temperature, apply electric and magnetic fields in various configurations, and use different photon energies of the incident laser pulses. So far, we verified that samples strained below 4 % do not display ferroelectricity. However, a new sample batch allows us to present results from EuO films with tensile strains up to 7 %.

DS 17.5 Tue 10:30 EB 301

On the lattice engineering of magnetoelectric couplin — •Michael Fechner and Nicola Spaldin — ETH Zurich, Department for Material Theory,CH-8093 Zurich, Switzerland

We present results of first-principles calculations of the microscopic origin of the linear magnetoelectric (ME) effect in Cr_2O_3. In general such magnetoelectric responses – that is the electric polarization created by an applied magnetic field – are small. Since they are composed of both electronic- and lattice-mediated contributions, however, an increase in the response can in principle be achieved by phonon engineering. Here we investigate this possibility by first calculating how the magnetic interaction parameters are affected by phonon modes of different symmetry, focussing particularly on those that are active in the ME coupling. We find that the exchange interactions are most strongly modified in the non-IR active rotational phonon modes which do not contribute to the ME response. We then calculate the effect on the ME response if these phonons are disabled. Based on our results we suggest new routes for engineering materials with enhanced ME couplings.

DS 17.6 Tue 10:45 EB 301

First principles study of Mn_2O_3 under pressure: Competition between Jahn-Teller distortion and charge disproportionation — •Carmen Quiroga and Rossitza Pentcheva — Dept. of Earth and Environmental Sciences, University of Munich

Contrary to most sesquioxides, which naturally occur in the corundum structure, Mn_2O_3 has a complex crystal structure corresponding to an orthorhombically distorted bixbyite [1], associated with the presence of the Jahn-Teller active Mn^{3+} cation. It has been suggested that the Jahn-Teller effect is inhibited under pressure, which could induce a phase transition to the corundum structure [2], from where the general transformation sequence of sesquioxides to perovskite and post-perovskite should follow. So far, however, only the post-perovskite has been reported experimentally above 27 GPa [3].

Using density functional theory calculations including an on-site Coulomb repulsion term, we explore the stability of the ambient phase α-Mn_2O_3 (*Pbca*) and candidate high-pressure polymorphs: corundum ($R\bar{3}c$), perovskite phases of Rh_2O_3 II (*Pbcn*) and $GdFeO_3$ (*Pbnm*) type and post-perovskite (*Cmcm*). In particular we focus on the effect of pressure on the charge, spin and structural degrees of freedom. Parallels to the pressure induced phase transitions in $MnTiO_3$ are discussed.

Funding by DFG SPP1236 (PE883/8-1) is acknowledged.

[1] S. Geller. Acta Crystallogr. **B27**, 821 (1971).
[2] C.T. Prewitt et al. Inorg. Chem. **8**, 1985 (1969).
[3] J. Santillán et al. Geophys. Res. Lett. **33**, L15307 (2006).

DS 17.7 Tue 11:00 EB 301

Investigation of Magnetoelectric Coupling in Self Assembled Ferromagnetic/Ferroelectric Heterostructures — •Fikret Yildiz[1], Chan-Ho Yang[2], Sinan Kazan[1], Yoon-He Jeong[2], and Bekir Aktas[1] — [1]Gebze Institute of Technology, De-

partment of Physics, 41400 Gebze-Kocaeli, Turkey — [2]Pohang University of Science and Technology, Department of Physics, Pohang, 790-784 S. Korea

Creating ferromagnetic/ferroelectric heterostructures is a way for developing multifunctional materials which is called multiferroics. Exchange bias may be used to couple a normal ferromagnet to a ferroelectric antiferromagnet and thus create a multiferroic system with nonzero magnetization. In implementing this idea we developed a synthesis method for composite films of MnFe2O4 embedded in BiFeO3 [1]. The method utilizes the Bi volatility to obtain the composite films via thermal annealing of multilayer composed of BiFeO3 and BiMnO3. SEM measurements showed that the cluster size varies depending on the film thickness. The composite films possess both ferroelectric and ferromagnetic properties [1]. Magnetoelectric coupling (MEC) was investigated by Ferromagnetic resonance (FMR) technique. Analysis of FMR data showed that resonance field can be controlled by GHz range electric fields.

[1] C.H. Yang, F. Yildiz, S.H. Lee, Y.H. Jeong, U. Chon, T.Y. Koo, Apply Phys. Lett. 90, 163116 (2007).

DS 17.8 Tue 11:15 EB 301

Strain Determination in Magnetoelectric Composite Systems by X-ray Diffraction Methods — •Christian Koops[1], Madjid Abes[1], Stjepan Hrkac[1], Bridget Murphy[1], Olaf Magnussen[1], Eric Woltermann[2], Henry Greve[2], and Eckhard Quandt[2] — [1]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany — [2]Institut für Materialwissenschaft, Christian-Albrechts-Universität zu Kiel, Germany

Understanding the coupling at the interface between magnetostrictive and piezoelectric components in magnetoelectric composites (ME) is essential for the optimization of these composites for sensor applications. A large ME response is only possible if the lattice deformation induced by an external magnetic field in the magnetostrictive material can be transferred efficiently to the piezoelectric material. To study this coupling at the burried interface of ME composites we measured the lattice deformation in ZnO as the piezoelectric component by grazing incidence X-ray diffraction in an external magnetic field, using the high-resolution and high intensity X-ray beam provided by the Diamond Light Source (I16) and PETRA III (P08). We employ samples with thin layers of different magnetostrictive materials, $(Fe_{90}Co_{10})_{78}Si_{12}B_{10}$ and Terfenol-D, on the (001) surface of high quality, single crystalline ZnO substrates. From the Bragg peak positions we determined the interplanar spacings in the ZnO substrates close to the interface and the corresponding strain as a function of the applied magnetic field.

15 min. break

DS 17.9 Tue 11:45 EB 301

Relaxor ferroelectricity in pure and doped magnetite — •Eugen Ruff[1], Florian Schrettle[1], Stephan Krohns[1], Peter Lunkenheimer[1], Victor A. M. Brabers[2], and Alois Loidl[1] — [1]Experimental Physics V, University of Augsburg, 86135 Augsburg, Germany — [2]Department of Physics, Eindhoven University of Technology, 5600 MB Eindhoven, Netherlands

A possible example for a multiferroic material is the extensively studied magnetite Fe_3O_4, which shows charge-order (CO) below the Verwey transition at $T_V \approx 120\,K$ and is ferrimagnetically ordered below $500\,K$[1]. As shown in the present contribution, dielectric spectroscopy reveals a relaxation below T_V, indicating relaxorlike polar order in Fe_3O_4[2]. We find long-range ferroelectric order to be impeded by the continuous freezing of polar degrees of freedom and the formation of a tunneling-dominated glasslike state of electrons at low temperatures. To reveal the origin of the ferroelectric state, whose dielectric signature is partly superimposed by a so called Maxwell-Wagner (MW) relaxation, we have investigated doped samples with Al, Ga, and Mg. The dielectric spectra of these doped samples ($< 2\%$) provide further evidence for the relaxor ferroelectric state of Fe_3O_4 and demonstrate the influence of the MW relaxation.

[1]D. I. Khomskii, J. Magn. Magn. Mater. **306**, 1 (2006).
[2]F. Schrettle *et al.*, Phys. Rev. B **83**, 195109 (2011).

DS 17.10 Tue 12:00 EB 301

Full-potential DFT+U study of orbitally ordered systems: the importance of non-spherical contributions and double counting — •Adam Jakobsson[1,2], Biplab Sanyal[1], Ivetta Slipukhina[2], Marjana Ležaic[2], Ersoy Sasioglu[2], Gustav Bihlmayer[2], and Stefan Blügel[2] — [1]Department of Physics and Astronomy, Uppsala University, 75120 Uppsala, Sweden — [2]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

DFT+U has for many years been a standard method to calculate properties of strongly correlated systems. Initially the method [1] was implemented in DFT-codes using the atomic sphere approximation (ASA) but was later also implemented into full potential DFT-codes. Various flavours of the double counting corrections used in the DFT+U method further add to the variety of different DFT+U functionals. The double counting corrections originally derived in the context of ASA are now routinely applied in full potential codes. Using the FLEUR code [2], we have investigated the importance of the non-spherical potential and the issue of double counting for orbital ordering and magnetism, i.e. properties that play a crucial role in many multiferroic materials. A recent implementation [3] of the constrained-RPA method was used to obtain parameters for the DFT+U calculations. This work was supported by the Young Investigators Group Program of the Helmholtz Association, Germany, contract VH-NG-409.
[1] V. I. Anisimov et al. PRB **44**, 943-954 (1991) [2] www.flapw.de [3] E. Şaşıoğlu et al. PRB **83**, 121101(R) (2011).

DS 17.11 Tue 12:15 EB 301

Ab initio **calculations of the magnetic properties of ordered perovskites** — •Igor Maznichenko[1], Alberto Marmodoro[2], Martin Lüders[3], Zdzislawa Szotek[3], Walter Temmerman[3], Ingrid Mertig[1,2], and Arthur Ernst[2] — [1]Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, D-06099 Halle (Saale), Germany — [2]Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle (Saale), Germany — [3]Daresbury Laboratory, Daresbury, Warrington WA4 4AD, Cheshire, United Kingdom

Perovskites are of particular interest in condensed matter physics due to their remarkable electronic and magnetic properties. Colossal magnetoresistance, ferroelectricity, multiferroicity, superconductivity, charge ordering, orbital ordering, metal-insulator transition, Jahn-Teller, and other effects are observed in perovskites. All these properties are strongly depending on the type of cations. Here we present a first-principles study of electronic and magnetic properties of $La_{2/3}Sr_{1/3}MnO_3$ (LSMO), which is a strongly correlated $3d$ transition metal oxide with a Curie temperature of 370 K. For varying La/Sr ratios different types of antiferromagnetism are observed. Using a self-consistent KKR Green function method, we show how the electronic and magnetic properties of LSMO depend on the valency of Mn, ordering of different cations in the lattice, and their relative orientation to each other. The influence of the Mn–O–Mn angle on the double-exchange coupling was examined.

DS 17.12 Tue 12:30 EB 301

Crystal growth and scattering investigations of $YFe_2O_{4-\delta}$ — •Thomas Müller[1], Joost de Groot[1], Jörg Strempfer[2], and Manuel Angst[1] — [1]Peter Grünberg Institut PGI and Jülich Centre for Neutron Science JCNS, JARA-FIT, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany — [2]Deutsches Elektronen-Synchrotron DESY, D-22607 Hamburg, Germany

$LuFe_2O_4$ is attracting attention as proposed multiferroic compound, but there is much less known about other isostructural rare-earth ferrites. We have grown single-crystals of $YFe_2O_{4-\delta}$ in a CO/CO_2-atmosphere to tune δ. Optimized crystals exhibit a magnetic behaviour identical to highly stoichiometric powder samples, i.e. two hysteretic phase transitions at 228 K and 180 K upon cooling.

Corresponding to these phases single-crystal x-ray-diffraction shows 3D-charge-ordered states, partially not compatible with the phases observed by electron diffraction [1]. At least one additional transition below 160 K, not present in magnetisation, is found in x-ray-diffraction. On one sample we found reflections at $(\frac{1}{3}, \frac{1}{3}$,half-integer) at 10 K , identical to the superstructure reflections of $LuFe_2O_4$. Nevertheless sample differences, due to different δ, have to be reviewed.

We further searched for anisotropy by resonant x-ray diffraction and full polarization analysis on superstructure reflections at PETRA III-P09. As for $LuFe_2O_4$ no anisotropy is observed. Although YFe_2O_4 has the same structure as $LuFe_2O_4$, the phases between 100 K and 230 K are totally different, showing much more complex incommensurate ordering. [1] N. Ikeda et al. Ferroelectrics **272**, 309 (2002)