MA 20: Multiferroics

Time: Wednesday 14:00–18:15 Location: EB 202

MA 20.1 Wed 14:00 EB 202

Observation of ferrotoroidic order in $LiCoPO_4$ — •Manfred Fiebig[1,2], Bas B. Van Aken[1,2], Jean-Pierre Rivera[3], and Hans Schmid[3] — [1]HISKP, Universität Bonn, Nussallee 14–16, 53115 Bonn, Germany — [2]Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin, Germany — [3]Department of Chemistry, University of Geneva, 30 quai Ernest-Ansermet, 1211 Geneva 4, Switzerland

Domains are an essential property of any ferroic material. Three forms of ferroic order (ferromagnetism, ferroelectricity, ferroelasticity) are widely known. It is currently debated whether to include an ordered arrangement of magnetic vortices as fourth form of ferroic order termed ferrotoroidicity [1]. Although there are reasons to do this from the point of view of thermodynamics a crucial hallmark of the ferroic state, *i.e.*, ferrotoroidic domains, has never been observed. Here ferrotoroidic domains are spatially resolved by optical second harmonic generation in $LiCoPO_4$ where they coexist with independent antiferromagnetic domains [2]. The origin of ferrotoroidicity in $LiCoPO_4$ is discussed. Their space- and time asymmetric nature relates ferrotoroidics to multiferroics with magnetoelectric phase control and other systems in which space and time asymmetry leads to exciting possibilities for future application. — Work supported by the SFB 608 of the DFG.

[1] C. Ederer, N.A. Spaldin, arXiv:0706.1974v1 [cond-mat.str-el], Phys. Rev. B, in press (2007)

[2] B.B. Van Aken, J.P. Rivera, H. Schmid, M. Fiebig, Nature **449**, 702 (2007)

MA 20.2 Wed 14:15 EB 202

Towards a microscopic theory of toroidal moments in periodic crystals — •Claude Ederer[1] and Nicola A. Spaldin[2] — [1]School of Physics, Trinity College Dublin, Dublin 2, Ireland — [2]Materials Department, University of California, Santa Barbara, CA 93106, USA

A toroidal moment breaks both time- and space-inversion symmetries simultaneously, and thus facilitates coupling between magnetization and electric polarization in magnetoelectric multiferroics. Furthermore, the recent observation of toroidic domains suggests that "ferrotoroidicity"represents a fundamental form of ferroic order, in addition to ferromagnetism, ferroelectricity, and ferroelasticity [1].

Here we review the basic definitions of toroidal moments and illustrate the difficulties in evaluating the toroidal moment of an infinite periodic system. We show that periodic boundary conditions give rise to a multivaluedness of the toroidal moment per unit cell, in close analogy to the case of the electric polarization in bulk periodic crystals. We then evaluate the toroidal moments of several multiferroic and magnetoelectric materials ($BaNiF_4$, $LiCoPO_4$, $GaFeO_3$, and $BiFeO_3$) in the "localized dipole limit", where the toroidal moment is caused by a time and space reversal symmetry-breaking arrangement of localized magnetic moments [2].

[1] B. B. Van Aken, J. P. Rivera, H. Schmid, and M. Fiebig, Nature 449, 702 (2007).

[2] C. Ederer and N. A. Spaldin, arXiv:0706.1974v1 [cond-mat.str-el], Phys. Rev. B, in press (2007).

MA 20.3 Wed 14:30 EB 202

Pyroxenes: a new class of multiferroics — •Sven Jodlauk[1], Petra Becker[1], John Mydosh[2], Daniel Khomskii[2], Thomas Lorenz[2], Sergej Streltsov[2], and Ladislav Bohatý[1] — [1]Inst. f. Kristallographie, Universität Köln — [2]II. Physikalisches Institut, Universität Köln

We present the results of our dielectric and magnetic investigations on pyroxenes with the general formula $AMSi_2O_6$ (A = mono- or divalent metal, M = di- or trivalent metal). They are shown to be a new class of multiferroic materials. In particular, we have found so far that $NaFeSi_2O_6$ becomes ferroelectric in a magnetically ordered state below ≈ 6 K. Similarly, magnetically driven ferroelectricity is also detected in the Li homologues, $LiFeSi_2O_6$ ($T_C \approx 18$ K) and $LiCrSi_2O_6$ ($T_C \approx 11$ K). In all these monoclinic systems the electric polarization can be strongly modified by magnetic fields. We present measurements of magnetic susceptibility, pyroelectric current and dielectric constants performed in various magnetic fields using a natural crystal of aegirine ($NaFeSi_2O_6$) and synthetic crystals of $LiFeSi_2O_6$ and $LiCrSi_2O_6$ grown from melt solution. For $NaFeSi_2O_6$ we propose a temperature versus magnetic field phase diagram. The possibility of a spiral magnetic structure caused by frustration to be the origin of the multiferroic behaviour taking into account computed exchange constants is discussed. We propose that other pyroxenes may also be multiferroic, and that the versatility of this family offers an exceptional opportunity to study general conditions for and mechanisms of magnetically driven ferroelectricity. This work was supported by the DFG via SFB 608.

Reference: J. Phys.: Condens. Matter **19** (2007) 432201

MA 20.4 Wed 14:45 EB 202

(Sr,Mn)TiO_3 - a magnetoelectric multiglass — •Subhankar Bedanta[1], Vladimir Shvartsman[1], Pavel Borisov[1], Wolfgang Kleemann[1], Alexander Tkach[2], and Paula Vilarinho[2] — [1]Angewandte Physik, Universität Duisburg-Essen, D-47048 Duisburg, Germany — [2]Department of Ceramics and Glass Engineering, CICECO, University of Aveiro, 3810-193 Aveiro, Portugal

In recent years there has been a growing interest in studies of multiferroic materials, in which two or more ferroically ordered states - usually long-range polar and magnetic order - exist simultaneously. In this work we extend this frame onto the simultaneous occurrence of two different glassy states, hence, looking for a "multiglass"scenario rather than for a multiferroic one. This is realized in $Sr_{0.98}Mn_{0.02}TiO_3$ ceramics [1] where the Mn^{2+} dopand ions are at the origin of both a polar cluster and a magnetic spin glass. Both the temperature dependencies of the magnetization, $M(T)$, and of the magnetic susceptibility, $\chi(T)$, show an anomaly around 38 K [2]. On the other hand, this temperature corresponds to the polar glass transition, indicating a correlation between magnetic and polar order. Both glassy states are unambiguously and independently evidenced by their specific aging and memory effects. In addition, higher order magnetoelectric coupling between the polar and magnetic degrees of freedom is observed [2].

[1] A. Tkach et al., Appl. Phys. Lett. **86**, 172902 (2005).

[2] S. Bedanta et al., in press

MA 20.5 Wed 15:00 EB 202

Relaxation dynamics of multiferroic rare-earth manganites — •Florian Schrettle[1], Peter Lunkenheimer[1], Joachim Hemberger[2], and Alois Loidl[1] — [1]Experimental Physics V, Center for Electronic Correlations and Magnetism, University of Augsburg, 86135 Augsburg, Germany — [2]Institute of Physics II, University of Cologne, 50937 Cologne, Germany

In recent years, multiferroic compounds attracted an enormous scientific and technological interest [1]. This rare class of materials combines ferroelectricity with (anti-)ferromagnetism in a single phase. In rare cases both order parameters are strongly coupled, leading to pronounced magnetocapacitive effects. Prominent examples for such systems are rare-earth manganites like, e.g., $TbMnO_3$. In these compounds partial frustration of the Mn-spins leads to spiral magnetic structures, which allow for a finite ferroelectric polarization. Besides the transition to the multiferroic phases, dielectric spectroscopy reveals a relaxation along the c-direction in these materials. We provide the results of a detailed investigation of the relaxation dynamics in several rare-earth manganites ($GdMnO_3$, Eu:$YMnO_3$, $TbMnO_3$, $DyMnO_3$). This dynamics seems to play an important role for the magnetocapacitive properties of these compounds.

[1] T. Kimura *et al.*, Nature **426**, 55 (2003); T. Lottermoser *et al.*, Nature **430**, 541 (2004); T. Goto *et al.*, Phys. Rev. Lett. **92**, 257201 (2004).

MA 20.6 Wed 15:15 EB 202

Magnetoelectric coupling in $La_{2/3}Ca_{1/3}MnO_3$-$BaTiO_3$ superlattices — •Kai Gehrke[1], Vasily Moshnyaga[1], Konrad Samwer[1], and Alexandr Belenchuk[2] — [1]I. Physikalisches Institut, Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany — [2]Institute of Applied Physics, Academieis 5, MD-2028 Chisinau, Moldova

Artificial composites of ferromagnetic and ferroelectric phases are promising for the study of magnetoelectric phenomena. We have prepared LCMO-BTO superlattices on MgO substrates, using the metalorganic aerosol deposition technique. Very high values of magnetocapacitance, $MC = [C(7T) - C(0)]/C(0) = 800\%$, were observed. The treatment in terms of an equivalent circuit shows that the majority

of this effect originates from the colossal magneto resistance (CMR), present in the LCMO-layers. Considering the resistance of LCMO, one can separate the "CMR-part" from the "coupling-part" of the MC. The analysis shows that the coupling is present in the ferromagnetic state only and results in a linear dependence of the BTO-layer capacitance on magnetic field ($C_{BTO} = C_0 + \alpha|H|$). This MC is as large as MC=16% at T=TMI, and MC=5% at low temperature.

The work was supported by the Deutsche Forschungsgemeinschaft via SFB 602, project A2.

MA 20.7 Wed 15:30 EB 202

Characterization of multiferroic $HoMnO_3$ films by second harmonic generation — •Tobias Kordel[1], Christian Wehrenfennig[1], Dennis Meier[1], Thomas Lottermoser[1], Catherine Dubourdieu[2], Isabelle Gelard[2], Kathrin Dörr[3], Jong-Woo Kim[3], and Manfred Fiebig[1] — [1]HISKP, Universität Bonn, Germany — [2]LMPG, Grenoble, France — [3]IFW, Dresden, Germany

The magnetic and electric properties of hexagonal $HoMnO_3$ films were investigated by optical second harmonic generation (SHG). Symmetry arguments relate different polarized SH contributions uniquely to either the electric or magnetic properties of $HoMnO_3$.

Epitaxial hexagonal $HoMnO_3$ and $YMnO_3$ films with 50-1000nm thickness were grown by PLD and MOCVD on YSZ(111) substrates. The "nonmagnetic" SHG signal in the films is similar to that of bulk $HoMnO_3$ pointing to the presence of ferroelectric ordering. However, a "magnetic" SHG signal has not been observed. At present, this can be explained by two models: (i) The film has anitferromagnetic domains with lateral dimensions much smaller than the wavelength of the light, so that the magnetic signal cancels out by interference; (ii) the film is in a ferroelectric single domain state. This induces ferromagnetic ordering of the Ho^{3+} spins and a Mn^{3+} spin configuration for which no SHG signal is observable. The latter model is supported by the observation of misaligned crystallites in the film with a preferred direction of polarisation.

MA 20.8 Wed 15:45 EB 202

Spindynamics of multiferroic $HoMnO_3$ — •Tim Hoffmann[1], Tim Günter[1], Takuya Satoh[2], Thomas Lottermoser[1], and Manfred Fiebig[1] — [1]HISKP, Universität Bonn, Nussallee 14-16, 53115 Bonn, Germany — [2]Department of Basic Science, The University of Tokyo, Komaba, Meguro-ku, Tokyo 153-8902, Japan

$HoMnO_3$ is a promising multiferroic because of the unusually pronounced coupling of electric and magnetic order manifesting as giant magnetoelectric effect. Static measurements have shown that the mangetic state of the Ho spins can be controlled by an external electric field [1]. We have investigated the dynamical properties of the magnetoelectric coupling of hexagonal $HoMnO_3$ using optical pump-probe experiments with 130 fs time resolution. Around 37 K a reorientation of the Mn spins is induced by competition of Ho-order, Mn-order and ferroelectric distortion, thus reflecting the multiferroic coupling. The reorientation is triggered by a pump pulse at 1.55 eV and probed time-resolved by optical second harmonic generation. Results point to an optically induced reorientation of Mn spins within a few hundreds of picoseconds. Is is superimposed by an oscillation with a 10 ns period around the new orientation. Possible explanations of these effects will be discussed.

[1] T. Lottermoser et al., Nature 430, 541 (2004)

15 Min Session Break

MA 20.9 Wed 16:15 EB 202

Electromagnons in rare earth multiferroics $DyMnO_3$ and $TbMnO_3$ — •Alexey Shuvaev[1], Alexander Mukhin[2], Anatoli Balbashov[3], Alois Loidl[4], and Andrei Pimenov[1] — [1]Experimentelle Physik IV, Universität Würzburg, 94074 Würzburg, Germany — [2]General Physics Institute of the Russian Academy of Sciences, 119991 Moscow, Russia — [3]Moscow Power Engineering Institute, 111250 Moscow, Russia — [4]Experimentalphysik V, EKM, Universität Augsburg, 86135 Augsburg, Germany

Dielectric permittivity spectra of rare earth multiferroic manganites $DyMnO_3$ and $TbMnO_3$ have been investigated in the terahertz frequency range. Similar to other multiferroics well-defined magnetoelectric excitations (electromagnons) are observed in the incommensurate magnetic phases below T = 40 K. Electromagnons are excitations of the incommensurate magnetic structure and, contrary to magnons, can be excited solely by electric component of the electromagnetic wave. Application of external magnetic fields strongly modifies the permittivity spectra in both compositions leading to magnetoelectric effects down to zero frequencies. In $TbMnO_3$ and in moderate fields additional structure appears close to $\nu \sim 10$ cm^{-1} and is suppressed again for magnetic fields above 5 T. Manifestations of different magnetic phase boundaries in the spectra of the dielectric permittivity are discussed.

MA 20.10 Wed 16:30 EB 202

Nonlinear optics on multiferroic tricolor superlattices — •Thomas Lottermoser[1,2], Hiroyuki Yamada[3], Jobu Matsuno[2], Takahisa Arima[2,4], Masashi Kawasaki[3,5], Manfred Fiebig[1], and Yoshinori Tokura[2,3,6] — [1]HISKP, Universität Bonn, 53115 Bonn, Germany — [2]ERATO, Multiferroics Project, JST, Tokyo 113-8656, Japan — [3]CERC, AIST, Tsukuba 305-8562, Japan — [4]IMRAM, Tohoku University, Sendai 980-8577, Japan — [5]Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan — [6]Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan

While multiferroics propose new physical phenomena and technical applications, materials exhibiting pronounced magnetoelectric coupling are still rare. Here, a new concept based on tricolor superlattices composed of repeated sequences of the transition-metal oxides $SrMnO_3$, $LaMnO_3$ and $LaAlO_3$ is used. Electron transfer at the interfaces of the antiferromagetic insulators $SrMnO_3$ and $LaMnO_3$ leads to a modification of their magnetic and electronic behavior. The electronic state near the interface is modified to be a ferromagnetic metal like (La, Sr)MnO_3. In addition, the charge transfer produces electric dipole moments along the stacking direction that do not cancel out in the ABCABC stack. Therefor the tricolor superlattices are polar ferromagnets. The artificial symmetry breaking allows (non-)magnetic contributions to optical second harmonic generation (SHG) which we used to probe directly the magnetic properties of the superlattice interfaces. The dependence on layer thickness and stress induced by different kinds of substrates is reported.

MA 20.11 Wed 16:45 EB 202

Magnetic field dependent structural modulations in $RFe_3(BO_3)_4$ — •Martin Philipp[1], Olga Kataeva[1,2], Christian Hess[1], Jorge E. Hamann-Borrero[1], Rüdiger Klingeler[1], Natalia Tristan[1], Bernd Büchner[1], Martin von Zimmermann[3], Alexander Vasiliev[4], Elena Popova[4], and Leonard N. Bezmaternykh[5] — [1]Leibniz-Institute for Solid State and Materials Research, IFW-Dresden, 01171 Dresden, Germany — [2]A.E.Arbuzov Institute, Russian Academy of Sciences, Arbuzov Str. 8, 420088 Kazan, Russia — [3]HASYLAB@DESY, Notkestr. 85, 22603 Hamburg, Germany — [4]Physics Faculty, Moscow State University, 119992 Moscow, Russia — [5]L.V.Kirensky Institute of Physics, Siberian Branch of RAS, 660036 Krasnoyarsk, Russia

Recently, rare earth iron borates $RFe_3(BO_3)_4$ (R: Rare Earth) attracted attention as candidates for possible multiferroic behavior. Most of the $RFe_3(BO_3)_4$ crystals exhibit antiferromagnetic order below $\sim$ 40 K. In this ordered phase several magnetic transitions are taking place, in dependence of temperature and magnetic field.

We have studied these magnetic transition by means of hard x-ray diffraction ($h\nu$ = 100 keV) for the compounds $TbFe_3(BO_3)_4$, $GdFe_3(BO_3)_4$ and $YFe_3(BO_3)_4$. Several superlattice reflections could be observed, which indicate commensurate stuctural modulations along a- and c-axis. We compare the field and temperature dependence of these superlattice reflections with results from thermodynamics and transport measurements.

MA 20.12 Wed 17:00 EB 202

Magnetization density in $TbMnO_3$ single crystals — •de Groot Joost[1], Kentzinger Emmanuel[1], Goukassov Arsène[2], Gillon Beatrice[2], and Möchel Anne[1] — [1]Forschungszentrum Jülich Gmbh D52425 Jülich IFF-4: Streumethoden — [2]Laboratoire Léon Brillouin (LLB), CEA-CNRS, CE Saclay, F-91191 Gif-sur-Yvette Cedex, France

$TbMnO_3$ belongs to a class of rare earth manganites showing multiferroic behaviour. While neutron diffraction [1] gives evidence for 4f Tb magnetization below the ferroelectric phase transition temperature 26K, hard x-ray resonance exchange scattering [2] clearly shows a magnetic polarization in the Tb 5d states below T_N=41K. In [2] it is argued that a strong coupling exists between the Mn magnetic order and the Tb magnetization. In order to evidence this coupling and to search for induced magnetization on the Oxygen atoms, we performed polarized neutron diffraction using the spin flip method in an external

field on a $TbMnO_3$ single crystal. These experiments were performed on the diffractometer 5C1 at the LLB. We detected a clear antiferromagnetic coupling between the field induced magnetization at the Tb and Mn sites in the paramagnetic phase. We will discuss these results compared to resonant x-ray scattering.

[1] M. Kenzelmann et al.,PRL 95, 087206 (2005)
[2] J. Voigt, J. Persson, J. W. Kim, G. Bihlmayer, Th. Brückel; Phys. Rev. B, 76 (2007), 104431-1

MA 20.13 Wed 17:15 EB 202

Coexistence of ferroelectric and long-wavelength magnetic ordering in $MnWO_4$ — •Dennis Meier[1], Michael Maringer[1], Thomas Lottermoser[1], Gouliang Yuan[1], Petra Becker[2], Ladislav Bohatý[2], and Manfred Fiebig[1] — [1]HISKP, Universität Bonn — [2]Institut für Kristallographie, Universität zu Köln

The strong interest in magnetoelectric multiferroics is due to their potential concerning the design of novel multifunctional devices, as well as to their unusual physical properties. Among these, $TbMnO_3$, $Ni_3V_2O_8$, and $MnWO_4$ form a particularly challenging group: The key factor for ferroelectricity lies in the long-wavelength magnetic order. However, the nature of the ferroelectric (FE) state in such a *spiral magnet* and its relation to the magnetic ordering is largely unclear. Here we report about the spatial distribution of FE domains in $MnWO_4$, revealed by optical second harmonic generation (SHG). Although the spontaneous polarization in this ferroelectric is magnetically induced, 180° domains as in a conventional ferroelectric are observed. This work was supported by the DFG through SFB 608

MA 20.14 Wed 17:30 EB 202

Polarization-dependent X-Ray Absorption Spectroscopy of $MnWO_4$ — •Nils Hollmann[1], Zhiwei Hu[1], Ladislav Bohaty[2], Petra Becker-Bohaty[2], Arata Tanaka[3], and Liu Hao Tjeng[1] — [1]II. Physikalisches Institut, Universität zu Köln — [2]Kristallographisches Institut, Universität zu Köln — [3]Department of Quantum Matter, Hiroshima University

Multiferroic materials which combine magnetism and ferroelectricity currently attract considerable attention. One of the recently discovered multiferroic materials is $MnWO_4$ (Hübnerite). It belongs to the group of multiferroics where a spontaneous electric polarization is caused by a spiral magnetic structure with a spin rotation axis not coinciding with the propagation vector.
To investigate the details of this astonishing combination of electronic and magnetic properties, we look at the electronic structure with the use of polarization-dependent X-ray absorption spectroscopy on single crystals of $MnWO_4$. The analysis of the experimental data on the L-edge of Mn is done by a configuration interaction calculation and will be discussed during the talk.

MA 20.15 Wed 17:45 EB 202

Growth and Characterization of $La_{0.67}Ca_{0.33}MnO_3$ and $BaTiO_3$ multilayers using PLD with in-situ RHEED — •Alexander Hirsch, Heiko Fasold, Ralf Koppert, Christian Wille, Frank Ludwig, and Meinhard Schilling — TU Braunschweig, Institut für Elektrische Messtechnik und Grundlagen der Elektrotechnik,

One way to design multiferroics, i.e., materials with ferroelectric and ferromagnetic properties, for new sensor applications is to grow superlattices with alternating ferromagnetic and ferroelectric layers.

Using pulsed laser deposition (PLD) $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) and $BaTiO_3$ (BTO) multilayers are grown on $SrTiO_3$ (100) substrates. To obtain atomically flat and single terminated substrate surfaces a chemical and subsequent annealing treatment is applied. All targets were prepared by standard ceramic synthesis. The growth of the films is monitored by in-situ reflection high energy electron diffraction (RHEED), which is a powerful tool for optimizing thin films with the objective of superlattice growth. In addition, the thin films are characterized by X-ray diffraction and atomic force microscopy.

Superlattices with alternating LCMO and BTO layers are grown, with BTO layer thicknesses ranging from 4 u.c. to 16 u.c. and LCMO layer thicknesses of 5 u.c.. RHEED intensity oscillations are used to control the thickness of the multilayers. Superlattices up to thicknesses of approx. 825 nm were grown. The influence of the BTO layer and the superlattice thickness, respectively, on the properties is analyzed.

MA 20.16 Wed 18:00 EB 202

Superconducting quantum interference device (SQUID) setup for magnetoelectric measurements — •Pavel Borisov, Andreas Hochstrat, Vladimir V. Shvartsman, and Wolfgang Kleemann — Angewandte Physik, Universität Duisburg-Essen, 47048 Duisburg, Germany

The magnetoelectric (ME) effect in its original sense refers to the intrinsic property of some material to react on an electric field by establishing a magnetic moment, and vice versa, to generate an electric polarization by applying a magnetic field. The best investigated ME material is Cr_2O_3, where Astrov measured the linear ME effect for the first time [1]. In Astrov's setup, the magnetic *ac* response signal, which was induced by an external electric field, was detected. Our method of the ME measurements is mainly based on Astrov's approach, but at the same time takes advantage of modern SQUID magnetometry [2]. We modified a commercial SQUID setup (MPMS 5S from Quantum Design), equipped with a magnetic *ac* susceptibility option, for measurements of the linear ME effect and tested it on a Cr_2O_3(111) single crystal. The results are in excellent agreement with previously reported data. The main advantage is the high sensitivity of the SQUID magnetometer combined with lock-in measuring technique. In addition, investigation of field-induced and higher order ME effects is possible.

[1] D. N. Astrov, Sov. Phys. JETP **11**, 780 (1960); **13**, 729 (1961)
[2] P. Borisov *et al.*, Rev. Sci. Instr. **78**, 106105 (2007)