TT 86: Ferroics and Multiferroics (joint session KFM/TT/MA)

Time: Thursday 9:30–13:30 Location: EMH 225

TT 86.1 Thu 9:30 EMH 225

A piezoresponse force microscopy study of Bi(Fe,Sc)O3 multiferroic ceramics — •Vladimir Shvartsman[1], Andrei Salak[2], Dmitry Khalyavin[3], and Doru Lupascu[1] — [1]Institute for Material Science, University of Duisburg-Essen, Essen, Germany — [2]Department of Materials and Ceramic Engineering/CICECO, University of Aveiro, Aveiro, Portugal — [3]ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, UK

Bismuth ferrite (BFO) has attracted an immense attention as a rare room-temperature single-phase multiferroics. The magnetic and ferroelectric structure of BFO can be tuned by cationic substitutions. In particular, using the high-pressure synthesis method BiFe(1-x)Sc(x)O3 ceramics can be sintered. The material appears in different polymorphs. The phase obtained by quenching under pressure is antipolar, but can be irreversibly turned into a polar one by thermal cycling at normal pressure. The resulting modification is a rare example of coexistence of canted ferroelectric and ferromagnetic states. We have addressed ferroelectric properties of these materials by piezoresponse force microscopy (PFM). The post-annealed Bi(Fe0.5Sc0.5)O3 ceramics show a strong PFM signal and posses a well-developed domain pattern typical of a ferroelectric state. The quenched ceramics, however, demonstrate no piezoresponse that is in line with its antiferroelectric state. We found that this state can be transferred to a ferroelectric one by application of a strong enough electric field. The temporal and temperature stability of the induced states are studied.

TT 86.2 Thu 9:50 EMH 225

Electronic Ferroelectricity in Organic Charge-Transfer Salts — •Jonas K. H. Fischer[1], Peter Lunkenheimer[1], Rudra Manna[2,3], Harald Schubert[3], Jens Müller[3], Michael Lang[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]Department of Physics, IIT Tirupati, Tirupati 517506, India — [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 often intriguing dielectric properties of the EDT-TTF-based charge-transfer salts have attracted considerable attention in recent years [1]. Examples are κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl, which exhibits multiferroicity [2], as well as 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 shows ferroelectric behavior in its charge-ordered state. In addition, further organic candidates for ferroelectricity as well as recent results on δ-(EDT-TTF-CONMe$_2$)$_2$Br are presented. The latter compound exhibits charge order but lacks dimerization. It displays interesting glasslike relaxation dynamics.
[1] P. Lunkenheimer and A. Loidl, J. Phys.: Condens. Matter **27**, 373001 (2015). [2] P. Lunkenheimer *et al.*, Nat. Mater. **11**, 755 (2012).

TT 86.3 Thu 10:10 EMH 225

Superconductivity and ferroelectric quantum criticality in KTaO3 — •Tobias Esswein, Awadhesh Narayan, and Nicola Spaldin — Materials Theory, ETH Zurich, Wolfgang-Pauli-Strasse 27, CH-8093 Zurich, Switzerland

Electron doped cubic perovskite KTaO3 has recently been shown to become superconducting [1]. In the closely related material SrTiO3, a ferroelectric quantum critical point was proposed to be the origin of superconductivity [2]. In this work, using first-principles calculations, we show that a ferroelectric quantum critical point emerges with electron doping in KTaO3, lying at doping values close to the top of the superconducting dome. We examine the effects of larger spin-orbit coupling and absence of crystal field splitting in KTaO3, in comparison to SrTiO3, on the phonon spectrum, electron-phonon coupling and quantum oscillations. Our findings contribute to the growing understanding of superconductivity around quantum critical points and could help in designing materials with higher superconducting critical temperatures.

[1] Ueno, K. et al. Discovery of Superconductivity in KTaO3 by Electrostatic Carrier Doping. Nature Nanotechnology 6, 408 (2011). [2] Edge, J. M., Kedem, Y., Aschauer, U., Spaldin, N. A. & Balatsky, A. V. Quantum Critical Origin of the Superconducting Dome in SrTiO3. Physical Review Letters 115, 247002 (2015).

TT 86.4 Thu 10:30 EMH 225

In-operando study of polarization reversal in ferroelectric thin films — •Christelle Kwamen[1], Matthias Rössle[2], Matthias Reinhardt[1], Wolfram Leitenberger[2], Flavio Zamponi[2], Marin Alexe[3], and Matias Bargheer[1,2] — [1]Helmholtz Zentrum Berlin — [2]Institute of physics and astronomie, University of Potsdam — [3]Department of physics, University of Warwick

The mechanisms associated with polarization reversal in ferroelectric materails are still under investigations because the microscopic dynamics are not yet fully understood. The permanent quest for energy efficient technologies drives investigations on making a ferroelectric operational under lowest switching voltage. There are many studies which investigate either the electrical signature of the switching or the structural changes of the crystal lattice associated with the switching. We present here a simultaneous study of the electrical and structural responses of a lead-zirconate-titanate-based capacitor heterostructure during charging, discharging, and polarization reversal, using time-resolved X-ray diffraction. Concomitant with the ferroelectric current peak, we observe the switching is characterized by a transient disorder evidenced by a decrease of the Bragg peak intensity. A peak width increase reveals the domain dynamics during the reversal. Our investigations show how the incomplete screening of the depolarization charges affect the piezoelectric response, measured via the Bragg peak position. We examine the interplay between charge flow, atomic motion in real time during device operation. We investigate how ultrashort laser pulse excitation can increase the charge flow in a biased device.

TT 86.5 Thu 10:50 EMH 225

Domains Properties in Thin Ferroelectric Films Related to Surface Screening, Flexoelectric and Vegard Effects — •Ivan S. Vorotiahin[1,2], Anna N. Morozovska[2], Eugene A. Eliseev[3], Sergei V. Kalinin[4], Qian Li[4], Yevhen M. Fomichov[3,5], and Yuri A. Genenko[1] — [1]Institut für Materialwissenschaft, Technische Universität Darmstadt, Darmstadt, Germany — [2]Institute of Physics, National Academy of Sciences of Ukraine, Kyiv, Ukraine — [3]Institute for Problems of Materials Science, National Academy of Sciences of Ukraine, Kyiv, Ukraine — [4]The Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, USA — [5]Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic

Ferroelectric domains is a topic of undying interest in the research community, since their properties and formation conditions still remain not fully understood. Among those conditions, surface screening charges, flexoelectric effect and chemical stresses can be named. Their influence is well observable in films of several to several tens of nanometres thicknesses, i.e. in the forefront of the phenomenological theories.

A series of modelling experiments has been performed to predict the effects that those physical qualities can make on a shape of ferroelectric domains in the most well-known perovskite materials, as well as their impact on the electromechanical properties, phase diagrams, and field distributions. Their influence has been numerically and analytically estimated to provide a roadmap for future measurements and compared with each other to obtain a stronger understanding of the physical processes occurring in perovskites.

TT 86.6 Thu 11:10 EMH 225

Screening in metallized ferroelectrics — •Hongjian Zhao[1], Alessio Filippetti[2,3], Carlos Escorihuela-Sayalero[1], Pietro Delugas[4], Enric Canadell[5], Laurent Bellaiche[6], Vincenzo Fiorentini[2,3], and Jorge Íñiguez[1] — [1]Materials Research and Technology Department, Luxembourg Institute of Science and Technology, Luxembourg — [2]Dipartimento di Fisica, Università di Cagliari, Cittadella Universitaria, Italy — [3]CNR-IOM SLACS, Cittadella Universitaria, Italy — [4]Scuola Internazionale Superiore di Studi Avanzati, Italy — [5]Institut de Ciència de Materials de Barcelona, Spain — [6]Physics Department and Institute for Nanoscience and Engineering, University

of Arkansas, USA

Ferroelectric materials are characterized by spontaneous polar distortions. The behavior of such distortions in the presence of free charge is the key to the physics of metallized ferroelectrics in particular, and of structurally-polar metals more generally. Using first-principles simulations, here we show that polar distortions resist metallization and the attendant suppression of long-range dipolar interactions in the vast majority of a sample of eleven representative ferroelectrics. We identify a novel meta-screening effect, occurring in the doped compounds as a consequence of the charge rearrangements associated to electrostatic screening, as the main factor determining the survival of a non-centrosymmetric phase. Our findings advance greatly our understanding of the essentials of structurally-polar metals, and offer guidelines on the behavior of ferroelectrics upon field-effect charge injection or proximity to conductive device elements.

20 min. break

TT 86.7 Thu 11:50 EMH 225

Pressure-induced insulator-metal transition in $EuMnO_3$ — •Andres Cano — CNRS, Univ. Bordeaux, ICMCB, Bordeaux, France

We study the influence of external pressure on the electronic and magnetic structure of $EuMnO_3$ from first-principles calculations. We find a pressure-induced insulator? metal transition at which the magnetic order changes from *A*-type antiferromagnetic to ferromagnetic with a strong interplay with Jahn?Teller distortions. This unexpected pressure-induced insulator-to-metal transition, although similar to the observed in CMR $LaMnO_3$, is unprecedented within the multiferroic RMnO$_3$ series. In addition, we find that the non-centrosymmetric $E^?$-type antiferromagnetic order can become nearly degenerate with the ferromagnetic ground state in the high-pressure metallic state. These features make $EuMnO_3$ an unique compound among the manganites because it behaves differently with respect to physical and "chemical" pressure, and hosts a genuinely new type of ferroelectric-like metallic state.

[1] *Pressure-induced insulator-metal transition in EuMnO3*, R. Qiu, E. Bousquet and A. Cano, J. Phys.: Condens. Matter 29, 305801 (2017).

TT 86.8 Thu 12:10 EMH 225

Far infrared studies on a diluted rare-earth langasite — •Lorenz Bergen[1], Evan Constable[1], Lukas Weymann[1], Alexander A. Mukhin[2], Nadezhda Kostyuchenko[1], and Andrei Pimenov[1] — [1]Institute of Solid State Physics, Vienna University of Technology, 1040 Vienna, Austria — [2]Prokhorov General Physics Institute, Russian Academy of Sciences, 119991 Moscow, Russia

Rare-earth langasites demonstrate fascinating structural and magnetic effects such as geometric frustration and are possible candidates for the spin-liquid state. To better understand the interplay between the structural and magnetic properties it is important to study the phonon and crystal electric field spectra that can be observed in the far infrared (FIR) range. The langasite structure crystallizes in the P321 space group with a general formula $A_3BC_3D_2O_{14}$. Here we present spectra of the diluted rare-earth langasite $La_{2.91}Ho_{0.09}Ga_5SiO_{14}$ using polarized far infrared radiation along different crystallographic directions and under a broad temperature range. The observed phonon frequencies are compared with model calculations. We compare the results on the holmium doped crystal and on pure $La_3Ga_5SiO_{14}$ langasite.

TT 86.9 Thu 12:30 EMH 225

Magnetic Excitations and High-Order Magnetoelectric Effect in Holmium Langasite — •Lukas Weymann[1], Thomas Kain[1], Alexey Shuvaev[1], Artem Kuzmenko[2], Alexander Mukhin[2], Evan Constable[1], Lorenz Bergen[1], Nadezhda Kostyuchenko[1], Anna Pimenov[1], and Andrei Pimenov[1] — [1]Institute of Solid State Physics, Vienna University of Technology, 1040 Vienna, Austria — [2]Prokhorov General Physics Institute, Russian Academy of Sciences, 119991 Moscow, Russia

Recently, compounds of the langasite family (prototype $La_3Ga_5SiO_{14}$) have attracted considerable attention due to their intriguing magnetic and magnetoelectric properties. The geometric frustration of the magnetic ions lies in the focus of the investigation of rare-earth langasites, since this makes them promising candidates for spin liquids.

In this work we show that in diluted rare-earth langasite $La_{2.91}Ho_{0.09}Ga_5SiO_{14}$ (3%Ho-LGS), where no magnetic frustration is present, unusual properties can be observed. 3%Ho-LGS single crystals reveal a substantial magnetoelectric effect comparable with other rare-earth langasites. The symmetry and the field dependence of the effect can only be explained by taking into account the higher order expansions of the crystal field theory. Terahertz measurements with a Mach-Zehnder interferometer reveal a series of characteristic magnetic excitations and a strong zero-field mode of presently unknown origin.

TT 86.10 Thu 12:50 EMH 225

Structural phase transition and domain formation in the hybrid improper ferroelectric $Ca_3Mn_{1.9}Ti_{0.1}O_7$ — •Mads C. Weber[1], Thomas Lottermoser[1], Morgan Trassin[1], Bin Gao[2], Sang-Wook Cheong[2], and Manfred Fiebig[1] — [1]ETH Zurich, Switzerland — [2]Rutgers University, Piscataway, New Jersey, USA

One of the bottlenecks for the application of magneto-electric multiferroics is the lack of materials with both, a robust coupling between ferroelectricity and magnetism, and a sufficiently large polarization. This problem may be overcome in layered perovskite systems, where octahedral rotations can give rise to improper ferroelectricity as well as a net-magnetization. Accordingly, ferroelectricity and magnetic order are linked by non-polar structural distortions. Essential for the understanding of the potential coupling of both parameters is an in-depth comprehension of the structural distortions and the formation of domains. In this work, we present a combined Raman spectroscopy (RS) and optical second harmonic generation (SHG) study of the improper ferroelectric phase transition and the related domain formation. Using RS, we trace the evolution of the non-polar structural distortions across the phase transition by probing the lattice vibrations of the system. Furthermore, we investigate the emergence of ferroelectricity by SHG a technique highly sensitive to breaking of inversion symmetry. Hence, RS and SHG represent a unique combination to investigate improper ferroelectric phase transitions.

TT 86.11 Thu 13:10 EMH 225

Lead Palladium Titanate: A new room-temperature magnetoelectric multiferroic — •Elzbieta Gradauskaite[1,2], Jonathan Gardner[3], Rebecca M. Smith[3], Finlay D. Morrison[3], Stephen L. Lee[1], Ram S. Katiyar[4], and James F. Scott[1,3] — [1]School of Physics and Astronomy, University of St Andrews, United Kingdom — [2]Present address: Department of Materials, ETH Zürich, Zürich, Switzerland — [3]School of Chemistry, University of St Andrews, United Kingdom — [4]Department of Physics, SPECLAB, University of Puerto Rico, USA

Magnetoelectric multiferroic materials combine the advantages of FeRAMs (speed, low power) and MRAMs (non-destructive readout) due to the linear (magnetoelectric) coupling between ferroelectricity and ferromagnetism. Despite the worldwide interest and effort, very few single-phase materials have been discovered that exhibit magnetoelectric coupling at room temperature. Until very recently $BiFeO_3$ was the only one, however it is not suitable for real practical device applications due to high leakage currents and weak coupling. Here, we demonstrate that $PbTi_{1-x}Pd_xO_3$ ($0 < x < 0.3$) is multiferroic up to 400 K and possesses a strong magnetoelectric coupling. This observation is remarkable because Pd is difficult to substitute into ABO_3 perovskite oxides and it is magnetic only under unusual conditions (strain or internal electric fields). Dielectric spectroscopy and magnetization studies will be discussed in detail, while paying particular attention to secondary phases present in the bulk specimen, identified as PdO, $PbPdO_2$ and Pd_3Pb using PXRD, SEM, EDX and XPS.