

## TT 55: Multiferroic Oxide Thin Films and Heterostructures I (joint session KFM/TT/MA)

Organizers: César Magén - University of Zaragoza, Aragón (Spain); Kathrin Dörr - Martin-Luther-Universität Halle-Wittenberg - Halle

Multiferroic oxide thin films and magnetoelectrically coupled oxide heterostructures are among the most attractive topics in the field of Complex Oxides. Within this extensive family of compounds, which are characterized by an unprecedented wealth of physical phenomena upon subtle variations of the structure or chemistry, multiferroics stand out due to the exciting novel physics underlying the coexistence and coupling of multiple ferroic orders. This exotic behavior bestows inherent multifunctionality upon these systems (either single-phase or heterostructure multiferroics), providing strong potential for future nanoelectronic devices.

Time: Wednesday 9:30–12:45

Location: EMH 225

**Invited Talk** TT 55.1 Wed 9:30 EMH 225

**Oxygen vacancy controlled functionalities at interfaces of multiferroic tunnel junctions.** — ●JACOBO SANTAMARIA — GFMC. Universidad Complutense 28040 Madrid

Oxygen vacancies are the most common defect in oxide perovskite oxides. Important applications are associated to their controlled generation and transport in electrochemical energy (fuel cells and batteries) and memory (memristors) devices. At interfaces oxygen vacancies can accumulate under the action of external electric fields and, especially in nanostructures be the source of novel, yet unreported, functionalities. Here we demonstrate the dynamic control of the vacancy profile in the nanometer thick barrier of a ferroelectric tunnel junction. Oxygen vacancies generated at an electrochemically active electrode accumulate towards the asymmetric interfaces of a ferroelectric tunnel barrier under the action of an external electric field and their ensuing doping effect modify the stability of ferroelectric polarization. I will further show that oxygen vacancies in a ferroelectric tunnel barrier may stabilize unexpected domain structures which control the tunneling transport providing a major step forward towards the new concept \*The Wall is the Device\* , to exploit the electronic properties of domain walls for ferroelectric tunnel barriers with new functionalities.

TT 55.2 Wed 10:00 EMH 225

**Structure and Magnetism of the Co/PZT/LSMO Interface** — ●HOLGER MEYERHEIM<sup>1</sup>, ARTHUR ERNST<sup>2</sup>, KATAYOON MOHSENI<sup>1</sup>, ANDREY POLYAKOV<sup>1</sup>, NATHALIE JEDRECZY<sup>3</sup>, ANDY QUINDEAU<sup>1</sup>, VICTOR ANTONOV<sup>1</sup>, MANUEL VALVIDARES<sup>4</sup>, HARI VASIL<sup>4</sup>, and PIERLUIGI GARGIANI<sup>4</sup> — <sup>1</sup>MPI f. Mikrostrukturphysik, D-06120 Halle — <sup>2</sup>Inst. für Th. Physik, JKU, A-4040 Linz, Austria — <sup>3</sup>INSP, UPMC-Sorbonne Univ., 75005 Paris, France — <sup>4</sup>Alba, 08290 Cerdanyola del Vallés, Spain

Using surface x-ray diffraction, x-ray absorption fine structure and x-ray circular dichroism (XMCD) experiments we have studied the geometric and magnetic properties of the Co/Pb(Ti<sub>0.8</sub>Zr<sub>0.2</sub>)O<sub>3</sub> interface. Co deposition in submonolayer amounts on the 2 unit cells thick (Ti,Zr)O<sub>2</sub> terminated Pb (Ti<sub>0.8</sub>Zr<sub>0.2</sub>)O<sub>3</sub> (PZT) layer leads to the formation of a perovskite type structure with Co-O distances of approximately 2.0 Å (octahedral) and 2.8 Å (cubic) in addition to a metallic Co-Co correlation near 2.4 Å. Co-L<sub>2,3</sub>-XMCD spectra also reveal different Co environments, especially two Co-O contributions (A) and (B) related to the octahedral coordination ( $m=2.69\mu_B$ ) and the cubic coordination ( $m=2.33\mu_B$ ). The XMCD analysis also evidences an anti-FM oriented induced moment at the PZT top layer Ti site ( $m=-0.005\mu_B$ ) related to the negative tunneling electro resistance effect. This result supports the "hybridization model" suggested by D.Pantel et al., Nat. Mat. 11, 289 (2012).

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TT 55.3 Wed 10:15 EMH 225

**Interfacial mechanisms in magneto-electric bismuth iron garnet thin films** — ●LAURA BOCHER<sup>1</sup>, ADRIEN TEURTRIE<sup>1,2</sup>, ELENA POPOVA<sup>2</sup>, ODILE STÉPHAN<sup>1</sup>, ALEXANDRE GLOTER<sup>1</sup>, and NIELS KELLER<sup>2</sup> — <sup>1</sup>Laboratoire de Physique des Solides - UMR 8502 CNRS, Université Paris-Sud, Orsay, FR — <sup>2</sup>Groupe d'Etude de la Matière Condensée - UMR8635 CNRS, UVSQ, Université Paris-Saclay, FR

Bismuth iron garnet (BIG) is ferrimagnetic with a relatively high magnetization (1600 G at 300 K), a magnetic ordering temperature from 650 K, and a giant Faraday rotation [1]. More recently, we evidenced a strong magneto-electric coupling at 300 K and above in BIG thin

films opening new perspectives for an electric control of the magnetization [2]. However BIG can solely be elaborated in thin film form using non-equilibrium growth techniques and no bulk reference exists for conventional investigations. Hence precise knowledge on the atomic and electronic structures of BIG thin films remains a key challenge to understand better their structure-property relationships.

Here we will shed light on BIG thin films using advanced electron spectro-microscopy techniques, i.e. Cs-STEM/EELS, to identify how its cubic structure can accommodate locally different lattice mismatches through a variety of relaxation mechanisms and verify down to the scale of the atomic columns any possible cation interdiffusion and/or electronic reconstruction at the film/substrate interface [3].

[1] M. Deb, et al. J. Phys. D 45 (2012) 455001. [2] E. Popova et al. APL 110 (2017) 142404. [3] E. Popova et al. JAP 121 (2017) 115304

TT 55.4 Wed 10:30 EMH 225

**Nonlinear spin-lattice coupling in EuTiO<sub>3</sub>: novel two-dimensional magneto-optical device for light modulation** —

●ANNETTE BUSSMANN-HOLDER<sup>1</sup>, KRYSYAN ROLDER<sup>2</sup>, and JÜRGEN KÖHLER<sup>1</sup> — <sup>1</sup>Max-Planck-Institute for Solid State Research, Heisenbergstr. 1, D-70569 Stuttgart, Germany — <sup>2</sup>Institute of Physics, University of Silesia, ul. Uniwersytecka 4, 40-007 Katowice, Poland

EuTiO<sub>3</sub> is antiferromagnetic at low temperature, namely below  $T_N=5.7K$ . In the high temperature paramagnetic phase the strongly nonlinear coupling between the lattice and the nominally silent Eu 4f7 spins induces magnetic correlations which become apparent in muon spin rotation experiments and more recently in birefringence measurements in an external magnetic field. It is shown here, that high quality films of insulating EuTiO<sub>3</sub> deposited on a thin SrTiO<sub>3</sub> substrate are versatile tools for light modulation. The operating temperature is close to room temperature and admits multiple device engineering. By using small magnetic fields birefringence of the samples can be switched off and on. Similarly, rotation of the sample in the field can modify its birefringence  $\Delta n$ . In addition,  $\Delta n$  can be increased by a factor of 4 in very modest fields with simultaneously enhancing the operating temperature by almost 100K. The results can be understood in terms of paramagnon phonon interaction where spin activity is achieved via the local spin-phonon double-well potential.

TT 55.5 Wed 10:45 EMH 225

**Complexity in the structural and magnetic properties of almost multiferroic EuTiO<sub>3</sub> thin films** — ZURAB GUGUCHIA<sup>1</sup>,

ZAHER SALMAN<sup>2</sup>, ●HUGO KELLER<sup>3</sup>, KRYSYAN ROLEDER<sup>4</sup>, JÜRGEN KÖHLER<sup>5</sup>, and ANNETTE BUSSMANN-HOLDER<sup>5</sup> — <sup>1</sup>Department of Physics, Columbia University, New York, New York 10027, USA — <sup>2</sup>Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland — <sup>3</sup>Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland — <sup>4</sup>Institute of Physics, University of Silesia, ul. Uniwersytecka 4, PL-40-007 Katowice, Poland — <sup>5</sup>Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

In a number of recent publications hidden magnetic properties at high temperatures have been reported for EuTiO<sub>3</sub> (ETO), which orders antiferromagnetically below  $T_N=5.7K$ . In addition, structural phase transitions have been discovered which correlate with the magnetic responses and can be tuned by a magnetic field. In order to identify the magnetic properties of ETO at temperatures well above  $T_N$ , low-energy muon-spin rotation ( $\mu$ SR) experiments have been performed on thin films of ETO which exhibit all properties observed in bulk materials and are thus well suited to conclude about the magnetic order

of the bulk. The  $\mu$ SR data reveal anomalies at 282 and 200 K related to the structural phase transitions in accordance with birefringence results. In addition, a transition to some kind of magnetic order below 100 K was observed as previously indirectly deduced from conductivity and dielectric constant measurements.

TT 55.6 Wed 11:00 EMH 225

**Surface reconstructions and related local properties of a BiFeO<sub>3</sub> thin film** — ●PENGXIANG XU<sup>1</sup> and LEI JIN<sup>2</sup> — <sup>1</sup>Institute for Theoretical Physics, ETH Zurich — <sup>2</sup>Peter Grünberg Institute (PGI-5), Forschungszentrum Juelich

Coupling between lattice and order parameters, such as polarization in ferroelectrics and/or polarity in polar structures, has a strong impact on surface relaxation and reconstruction. However, up to now, surface structures that involve the termination of both matrix polarization and polar atomic planes have received little attention, particularly on the atomic scale. Here, we study surface structures on a BiFeO<sub>3</sub> thin film using atomic-resolution scanning transmission electron microscopy and spectroscopy. Two types of surface structure are found, depending on the polarisation of the underlying ferroelectric domain. On domains that have an upward polarisation component, a layer with an Aurivillius-Bi<sub>2</sub>O<sub>2</sub>-like structural unit is observed. Dramatic changes in local properties are measured directly below the surface layer. On domains that have a downward polarisation component, no reconstructions are visible. Calculations based on ab initio density functional theory reproduce the results and are used to interpret the formation of the surface structures.

15 min. break

TT 55.7 Wed 11:30 EMH 225

**Domain engineering in BFO films** — ●YESEUL YUN<sup>1,2</sup>, NIRANJAN RAMAKRISHNEGOWDA<sup>1,2</sup>, DAVID KNOCHE<sup>1,2</sup>, DAESUNG PARK<sup>1,2</sup>, and AKASH BHATNAGAR<sup>1,2</sup> — <sup>1</sup>Zentrum für Innovationskompetenz SiLi-nano, Halle (Saale), Germany — <sup>2</sup>Martin Luther Universität Halle-Wittenberg, Halle (Saale), Germany

Multiferroic materials have attracted great attention due to their unusual physical properties and potential in device applications. The lead-free bismuth ferrite (BiFeO<sub>3</sub>) is one of the most promising candidates. The domain structure plays a crucial role in determining ferroelectric and magnetic properties. Domains and domain walls can be modulated by parameters such as epitaxial strain, film thickness, substrate termination and presence of conductive layers.

In this study, we investigate the role of plume-related characteristics in obtaining long range order of ferroelastic domains in BiFeO<sub>3</sub> films. BFO/LSMO hetero-structures were fabricated using PLD on STO (001) substrate with different O<sub>2</sub> partial pressures. Preferential nucleation and long range ordering of 71° domain walls was achieved by varying the plume density, indicating the importance of plasma plume dynamics for the evolution of domain structure in the films. The role of strain and electrostatic energies was also analyzed in conjunction. The thickness of BFO was varied to modulate the extent of strain, while the electrostatic conditions were tuned by the thickness of LSMO.

TT 55.8 Wed 11:45 EMH 225

**Domain Engineering of the Bulk Photovoltaic Effect in Bismuth Ferrite** — ●DAVID KNOCHE<sup>1,2</sup>, NIRANJAN RAMAKRISHNEGOWDA<sup>1,2</sup>, YESEUL YUN<sup>1,2</sup>, and AKASH BHATNAGAR<sup>1,2</sup> — <sup>1</sup>Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany — <sup>2</sup>Zentrum für Innovationskompetenz SiLi-nano, Halle (Saale), Germany

The photovoltaic (PV) effect in multiferroic bismuth ferrite (BFO) can be largely attributed to the bulk photovoltaic (BPV) mechanism. The mechanism is associated to the absence of inversion symmetry in these materials. The principle of the BPV effect, that results in an above-bandgap open circuit voltage ( $V_{oc}$ ), differs from the well-known photovoltaic effect observed in semiconductors like silicon, and still demands in-depth analysis. In this regard, the contribution of ferroic aspects, such as orientation of domains, is crucial and can be potentially used as a tuning parameter.

Thin films of single crystalline BiFeO<sub>3</sub> were grown epitaxially via pulsed laser deposition. Planar electrodes with different in-between distances were deposited on top of the sample. The domain orientations within the measurement gap were manipulated by applying high electric fields across the electrodes. Gradual increment in the

applied electric field was crucial in obtaining intermediate domain architectures, that were visualized with piezo force microscope (PFM). Photoelectrical response was measured in conjunction to evaluate the influence on Voc and short circuit current.

TT 55.9 Wed 12:00 EMH 225

**Investigation of a-domain formation in Pb(Zr,Ti)O<sub>3</sub> thin films** — ●NIRANJAN RAMAKRISHNEGOWDA<sup>1,2</sup>, YESEUL YUN<sup>1,2</sup>, DAESUNG PARK<sup>1,2</sup>, and AKASH BHATNAGAR<sup>1,2</sup> — <sup>1</sup>Zentrum für Innovationskompetenz SiLi-nano, Halle (Saale), Germany — <sup>2</sup>Martin Luther Universität Halle-Wittenberg, Halle (Saale), Germany

Strain engineering of ferroelectric/ferroelastic domains is an active area of research nowadays, as it provides an exotic pathway to tune the resultant properties of ferroic materials. Since the domains can be also ferroelastic, the extent of strain, applied via the substrate-film lattice parameter mismatch, can be used to define the domain width, orientation and position. However, the persistence of the strain across the thickness of the film is largely affected by growth related process parameters.

In the case of Pb(Zr,Ti)O<sub>3</sub>, one of the most widely investigated ferroelectric, recent studies involving asymmetric substrates allowed to fine tune the nucleation of a-domains, and the associated domain wall thickness. The proposed prerequisite condition of  $a_{film} < a_{substrate} < c_{film}$  was satisfied. In this work we attempt to further analyze this condition by growing PZT films on symmetric SrTiO<sub>3</sub> substrates. The role of depolarization field was evaluated by the use of conductive oxide layers sandwiched between the film and the substrate. The usually neglected contribution of target density and purity will be also elaborated.

TT 55.10 Wed 12:15 EMH 225

**Continuous control of morphotropic phases by strain doping** — ●ANDREAS HERKLOTZ<sup>1</sup>, STEFANIA FLORINA RUS<sup>2</sup>, ER-JIA GUO<sup>3</sup>, KATHRIN DÖRR<sup>1</sup>, and THOMAS ZAC WARD<sup>3</sup> — <sup>1</sup>Martin-Luther-Universität Halle-Wittenberg, Halle, Germany — <sup>2</sup>National Institute for Research and Development in Electrochemistry and Condensed Matter, Timisoara, Romania — <sup>3</sup>Oak Ridge National Laboratory, Oak Ridge, USA

The realization of a strain-driven morphotropic phase boundary in epitaxial BiFeO<sub>3</sub> (BFO) films has broadened this definition to single-phase materials and opened up great potential for advanced applications. However, a greater success of morphotropic systems in thin film technologies would require a *ex situ* control of the thin film's composition or strain state that is practically impossible with standard epitaxy approaches. Here we demonstrate that *ex situ* strain doping via low-energy helium implantation induces a complete phase transition from epitaxial rhombohedral-like to supertetragonal BFO films. This control over morphotropic phases is highly tunable and fully reversible via a high temperature anneal. We argue that strain doping of morphotropic films creates a new phase space based on internal and external lattice stress that can be seen as an analogue to temperature-composition phase diagrams of classical morphotropic ferroelectric systems.

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TT 55.11 Wed 12:30 EMH 225

**Mechanical reading of ferroelectric polarity** — ●GUSTAU CATALAN<sup>1</sup>, KUMARA CORDERO<sup>2</sup>, and NEUS DOMINGO<sup>2</sup> — <sup>1</sup>ICREA-Institutio Catalana de Recerca i Estudis Avançats, Barcelona, Catalunya — <sup>2</sup>ICN2-Institut Catala de Nanociencia i Nanotecnologia, Barcelona, Catalunya

Flexoelectricity is polarization induced by strain gradient. It is closely related to piezoelectricity (polarization induced by strain), a phenomenon for which it was originally viewed as potential substitute. More recently, however, it has become apparent that very exciting new functionalities can be achieved when we combine both flexoelectricity and piezoelectricity in ferroelectrics.

One such functionality, reported in 2012, was the seminal discovery that strain gradients induced by the tip of an atomic force microscope (AFM) could mechanically \*write\* ferroelectric domains without applying any voltage. Here, we would like to report the complementary effect: the combination of flexoelectricity and piezoelectricity allows \*reading\* the polar sign of ferroelectric domains from pure (voltage-free) mechanical response.