MA 37: Multiferroic Oxide Thin Films and Heterostructures II (joint session KFM/TT/MA)

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

Time: Wednesday 15:00–18:15 Location: EMH 225

Invited Talk MA 37.1 Wed 15:00 EMH 225

Merging Nonlinear Optics and Multiferroic Heterostructure Design — •Manfred Fiebig — Department of Materials, ETH Zurich, Vladimir-Prelog-Weg 4, 8093 Zurich, Switzerland

Despite the large variety of valuable tools that are at our disposals for characterizing oxide thin films, some of their functionalities remain invisible. Buried layers and their ordering and interactions are difficult to access. We show how optical second harmonic generation (SHG) allows us to detect such hidden properties. We show how the real-time dynamics of a domain patterns in multiferroic $BiFeO_3$ is tracked by SHG through a ferromagnetic metallic cover layer, thus identify the magnetoelectric domain coupling nondestructively during the poling process [1]. SHG furthermore resolves the domain-wall architecture in tetragonal ferroelectric thin films. In $PbZr_{0.2}Ti_{0.8}O_3$ films it quantifies the buried distribution of a- and c-domains and reveals that c-domain walls exhibit a mixed Ising-Néel-type transverse rotation of polarization across the wall [2]. Finally, by coupling a laser beam into the deposition chamber, SHG follows the evolution of the spontaneous polarization of complex multiferroic heterostructure in real time and with monolayer sensitivity throughout the entire deposition process. Such in-situ SHG allows us tailor heterostructures with an arbitrary sequence of ordered states which could become the key to whole new class of functional ferroelectric materials [3].

[1] M. Trassin *et al.*, Adv. Mater. **27**, 4871 (2015). **[2]** G. De Luca*et al.*, M. Trassin, Adv. Mater. **29**, 1605145 (2017). **[3]** G. De Luca*et al.*, Nature Comm. **8**, 1419 (2017)

MA 37.2 Wed 15:30 EMH 225

Real-time observation of polarization emergence in ultrathin ferroelectric heterostructures — •Gabriele De Luca[1], Nives Bonacic[1], Johanna Nordlander[1], Corinne Bouillet[2], Manfred Fiebig[1], and Morgan Trassin[1] — [1]Department of Materials, ETH Zurich, Vladimir-Prelog-Weg 4, 8093, Zurich, Switzerland — [2]Institut de Physique et Chimie des Matériaux de Strasbourg CNRS UMR 7504, 67034, Strasbourg, France

The integration of functional properties into oxide multilayer architectures demands for atomic precision. In-situ diagnostic tools guarantee high structural quality but are usually insensitive to the functionality targeted with the actual deposition. The conventional optimization process requires multiple samples and ex-situ analysis. Here, we take advantage of the non-invasive nature of optical probes and monitor the functionality during growth. Taking ferroelectricity as a representative case, we show that optical in-situ second harmonic generation (ISHG) analysis can be performed simultaneous to the pulsed-laser-deposition growth operation. We follow the evolution of the spontaneous polarization in real time and with monolayer resolution throughout the deposition process [1]. Such direct access allows validating the growth of oxide heterostructures with an arbitrary sequence of up- and down-polarized ferroelectric layers. This is only the first step in the implementation of ISHG as a growth diagnostic tool. The in-situ access to emerging properties enables an unprecedented degree of control that can promote the engineering of oxides functionalities to a completely new level.

[1] G. De Luca et al., Nat. Commun. 8, 1419 (2017)

MA 37.3 Wed 15:45 EMH 225

Controlling the effect of the depolarizing field in $BaTiO_3$-$SrTiO_3$ multilayers — •Nives Bonacic[1], Gabriele De Luca[1], Shovon Pal[1], Marco Campanini[2], Marta D. Rossell[2], Morgan Trassin[1], and Manfred Fiebig[1] — [1]ETH Zurich, Department of Materials — [2]EMPA, Switzerland

The demand for ever-smaller devices has been approaching the fundamental limits of ultrathin ferroelectric films. In the low-thickness regime, maintaining a large, stable and switchable ferroelectric polarization relies on the control of the strain state, thickness, interface termination and electrostatic conditions. Achieving a robust polarization or a controlled domain state remains, however, challenging. Imperfect charge screening at interfaces results in non-cancellation of internal fields that can in extreme case annihilate ferroelectricity. Taking ($BaTiO_3$-$SrRuO_3$) capacitor-like heterostructures as a model system, we directly access the polarization and the domain state during the film deposition using optical second harmonic generation [1]. We observe a previously elusive impact of the evolving electrostatic environment on the $BaTiO_3$ domain state simultaneously with the growth. The initial phase of the top-electrode deposition is accompanied by temporary enhancement of built-in fields in the ferroelectric layer resulting in 180° domain formation. We discuss ways to manipulate the depolarizing field and control the polarization during the growth as it presents a possible route towards a novel class of oxide-electronic devices. [1] G. De Luca et al., Nat. Commun. 1419 (2017).

MA 37.4 Wed 16:00 EMH 225

In-situ characterization of improper ferroelectricity in ultrathin multiferroic h-$YMnO_3$ films — •Johanna Nordlander[1], Marta D. Rossell[2], Rolf Erni[2], Manfred Fiebig[1], and Morgan Trassin[1] — [1]ETH, Zürich, Switzerland — [2]EMPA, Dübendorf, Switzerland

Improper ferroelectrics are materials whose ferroelectricity is driven by another, primary, order parameter. This type of ferroelectricity can lead to exotic properties that do not exist in standard ferroelectrics. In the case of bulk hexagonal manganites, the structural trimerization results in a topologically protected vortex domain structure. Due to their potential for extending existing technological applications with complex functional properties, there has been a revival of interest in hexagonal manganite thin films. Here we demonstrate the growth of highly oriented, epitaxial hexagonal $YMnO_3$ thin films using pulsed laser deposition. We use in-situ optical second harmonic generation (SHG) to non-invasively probe, in real time during and after the deposition process, the ferroic state of the films in the ultrathin regime. With the complementary use of reflection high-energy electron diffraction (RHEED), the emerging polarization of $YMnO_3$ is resolved with monolayer precision. The characteristic improper ferroelectric domain pattern in the ultrathin $YMnO_3$ films is investigated using scanning transmission electron microscopy. This work provides new insights in the early stage of improper ferroelectricity and domain state in hexagonal $YMnO_3$ thin films - especially the drastic influence of epitaxial strain and reduced dimensions on the ferroelectric Curie temperature.

MA 37.5 Wed 16:15 EMH 225

Multiferroic and magnetoelectric nanocomposites for data processing — •Wolfgang Kleemann — Physics Department, University Duisburg-Esssen, 47048 Duisburg, Germany

Switching of magnetism with electric fields and magnetic control of electric polarization are challenging tasks for multiferroic and magnetoelectric materials. Various composite realizations appear most promising for data processing applications: (1) We propose 2-2 nanocomposites based on magnetoelectric (ME) chromia (111) films (Cr2O3), which allow electric switching of the magnetization of epitaxially grown ultrathin ferromagnetic Co/Pt/Co trilayers via interfacial exchange bias. Random access memory (MERAM) and logic cell MEXOR have been approved [1]. (2) Regular 2-1 composites of magnetostrictive cobalt ferrite (CoFe2O4) nanopillars are PLD-grown in a piezoelectric film of barium titanate (BaTiO3). In a transverse magnetic field they exert a staggered shear stress-induced surface polarization pattern in the BaTiO3 environment [2]. Possible data storage applications will be discussed. (3) Ceramic 0-3 composites of antiferromagnetic-ferroelectric Bi(Fe,Co)O3 nanoclusters embedded in K0.5Bi0.5TiO3 reveal giant linear magneto-electric response via bilinear piezo-magneto-electric coupling, $M = \alpha E$ with α = 10-5 s/m [3]. They are candidates for future electrically addressable nanodot mass memory devices. [1] US Pat. 7,719,883 B2 (2010). [2] Nature Comm. 4, 2051 (2013). [3] Adv. Funct. Mater. 26, 2111 (2016).

15 min. break

MA 37.6 Wed 17:00 EMH 225

Local observables in inhomogenous systems — •Raffaele Resta[1] and Antimo Marrazzo[2] — [1]IOM-CNR, Trieste, Italy —

[2]THEOS, EPF Lausanne, Switzerland

When addressing inhomogenous systems (e.g. heterostructures) a key issue is which physical properties do (or do not) admit a local expression. It is known since long time that spin magnetization is indeed local, and that the density of spin magnetization is well defined quantity. It is also known (since the early 1990s) that instead polarization density is an ill defined concept. Bulk electric polarization P is a Berry phase of the electronic wavefunction: as such it does not admit any local representation. In analogy with P, orbital magnetization is a geometrical property of the electronic ground state; but at variance with P, it also admits a local representation with a well defined density in coordinate space [1,2]. Here we address one further property: the insulating/metallic character of a region in an inhomogeneous system. A well known tool to investigate this property is the local density of states, but it is not a ground-state property. According to Kohn (1964) the insulating/metallic character of a material stems from a different organization of the electrons in their ground state. We define a local "marker" which probes such organization, and we validate it by means of computer simulations.

[1] R. Bianco and R. Resta, Phys. Rev. Lett. 110, 087202 (2013)
[2] A. Marrazzo and R. Resta, Phys. Rev. Lett. 116, 137201 (2016)

MA 37.7 Wed 17:15 EMH 225

Magnetoelectric coupling and multicaloric effects in $SrMnO_3$ — Alexander Edström and •Claude Ederer — Materials Theory, ETH Zürich, Switzerland

$SrMnO_3$ is a G-type antiferromagnet where ferroelectricity can be induced by epitaxial strain or Ba-substitution. Furthermore, a transition to ferromagnetic order has been predicted under large tensile strain [1], and the two ordering temperatures can in principle be tuned to coincide by varying both strain and composition. $SrMnO_3$ is thus a very rare example of a multiferroic with proper ferroelectric and magnetic order and similar ordering temperatures.

We use first principles electronic structure calculations in combination with first-principles-derived effective model Hamiltonians to obtain the temperature and strain-dependent ferroelectric/magnetic phase diagram of $SrMnO_3$. We then explore coupling effects between the polar and magnetic order. A particular focus thereby are possible multi-caloric effects, i.e., adiabatic temperature changes induced by applied electric and/or magnetic fields, that are very promising for future solid state cooling devices [2].

[1] J. H. Lee and K. M. Rabe, Phys. Rev. Lett. 104, 207204 (2010).
[2] X. Moya, S. Kar-Narayan, and N. D. Mathur, Nature Mater. 13, 439 (2014).

MA 37.8 Wed 17:30 EMH 225

Octahedral tilting, phonons and Goldstone modes in 111-strained perovskites — Magnus Moreau[1], Astrid Marthinsen[1], Sinead Majella Griffin[2], Tor Grande[1], Thomas Tybell[1], and •Sverre Magnus Selbach[1] — [1]NTNU Norwegian University of Science and Technology, Trondheim, Norway — [2]Lawrence Berkeley National Laboratory, Berkeley, California, USA

Epitaxial strain has been extensively explored to enhance existing and enable new functional properties in perovskites oxide thin films, with the majority of the work done on 001-oriented films. Recent advances in film growth has made other epitaxial orientations possible, and particularly 111-oriented films show interesting properties because of the different symmetry and chemical bonding at the terminating (111) facet. We use density functional theory (DFT) calculations to study the different response to 111- and 001-strain of the octahedral tilt system and the crystal field splitting of perovskite oxides. Unlike 001-strain, 111-strain is parallel to the edges of the oxygen octahedra, and tensile 111-strain can emulate negative hydrostatic pressure, which is not easily realised experimentally for bulk materials. General trends for how 111-strain affects polar and rotational modes are outlined based on calculations of twenty common perovskites. Furthermore, we show that in SrMnO3 compressive 111-strain give rise to Goldstone-like phonon modes with a Mexican hat-shaped energy surface, while large tensile strain can induce polar Goldstone modes. The chemical and structural requirements for engineering structural Goldstone modes in 111-strained perovskites are finally discussed.

MA 37.9 Wed 17:45 EMH 225

Voltage controlled magnetization dynamics in nanostructured multiferroic multilayer systems — •Alexander F. Schäffer and Jamal Berakdar — Institute of Physics, Martin-Luther-Universität Halle-Wittenberg, 06099 Halle (Saale), Germany

We investigate the control of magnetization dynamics by applying localized voltages in the framework of nanostructured multiferroic heterostructures. Manipulations of the magnetic anisotropy are spatially controlled by nanostructuring. This combination is utilized as a toolbox to excite or manipulate magnetic systems in order to manipulate multiple magnetic phenomena, such as the magnon dispersion, local spin current sources, or local non-collinear magnetic textures. Full-fledged numerical calculations for realistic systems along with basic analytical models will be shown in order to point towards opportunities for further experimental investigations and applications.

MA 37.10 Wed 18:00 EMH 225

Electronic and magnetic properties of $BaFeO_3$-Pt(111) in a quasicrystalline approximant structure — •Waheed A. Adeagbo, Igor V. Maznichenko, Hichem Ben-Hamed, Ingrid Mertig, and Wolfram Hergert — Institute of Physics, Martin Luther University Halle-Wittenberg, Germany

The first reported formation of an oxidic quasicrystal (OQC) $BaTiO_3$ (BTO) on Pt(111) has led to finding of other quasicrystalline (QC) perovskites like $SrTiO_3$ on the same substrate. Since these are non-magnetic, it is interesting to investigate the properties of magnetic perovskites in corresponding approximant structures. $BaFeO_3$ (BFO) has a very good lattice match with Pt(111) and also a robust magnetic properties which could add new interesting features to the QC systems. In this work we have carried out first principles study on the periodic BFO bulk crystals properties in the cubic (c-BFO) and the hexagonal ($6h$-BFO) phases. The derived OQC thin film which exhibits strong similarities to the BTO-derived OQC with respect to the local tiling geometry of Kepler's approximant was also studied both in the free standing and in the supported phase on Pt(111) surface. Our results shows that the anti-ferromagnetic $6h$-BFO bulk phase is preferable ground state to c-BFO phase. Like in BTO-OQC approximant, the BFO also shows all four Fe atoms surrounded by three O atoms with the FeO_3 units separated by Barium atoms with the total stoichiometry $Ba_5Fe_4O_{10}$. Since the exact oxidation states of the Fe and the role of O vacancy in the stabilization is unknown for these systems, the results of these will be presented together with the magnetic contribution.