KFM 1: Focus: Domains and Domainwalls in (Multi)Ferroics I

The focus session is dedicated to advanced nanoscale characterization, property engineering, and modelling methods of (multi)ferroic materials. Typical examples may include ferroic domain walls, microstructural levers, or strain effects. Further, applications in novel nanoelectronic devices and nano-related engineering concepts for macroscopic properties of multiferroics are of interest.

Chair: Dr. Jan Schultheiß (University of Augsburg)

Time: Monday 9:00–12:15 Location: POT 51

Invited Talk KFM 1.1 Mon 9:00 POT 51

Investigations of multiferroic behavior within domains and domain walls of a multiferroic Aurivillius phase system — •Lynette Keeney — Tyndall National Institute, University College Cork, Lee Maltings Complex, Dyke Parade, Cork, Ireland, T12 R5CP

Single-phase multiferroics intertwine ferroelectric and ferromagnetic properties, providing novel ways to manipulate data and store information and provide opportunities for exploring new chemistry and physics. In recent years, my team reported the design of a room temperature multiferroic material with an Aurivillius phase structure demonstrating reversible magnetoelectric switching of ferroelectric domains under the influence of a cycled magnetic field. Our previous work used atomic structure determinations of preferred cation locations to advance comprehension of key mechanisms governing ferromagnetism within multiferroic domains. In this presentation, I will discuss fundamental electronic characteristics at differing bonding environments within this complex layered system. We reveal how crystal field splitting of the titanium cation is influenced by its position within the Aurivillius unit cell, correlating with the extent of tetragonal distortion, octahedral tilting and ferroelectric polarisation within the domains. I will discuss how electrostatic strain and elastic energy variations close to bismuth oxide interfaces and defect regions are not only influential in promoting magnetic cation partitioning and multiferroic behaviour, these also influence the formation of exotic charged domain walls and polar vortex domain walls, further initiating technology prospects for this intriguing multiferroic system.

KFM 1.2 Mon 9:30 POT 51

Understanding sensitivity of ferroelectric domain walls to atmospheric parameters — •Leonie Richarz[1], Jan Schultheiss[1,2], Edith Bourret[3], Zewu Yan[3,4], Antonius T.J. van Helvoort[1], and Dennis Meier[1] — [1]NTNU Norwegian University of Science and Technology, Trondheim, Norway — [2]University of Augsburg, Augsburg, Germany — [3]Lawrence Berkeley National Laboratory, Berkeley, CA, USA — [4]ETH Zurich, Zurich, Switzerland

Ferroelectric domain walls are natural interfaces, separating volumes with different orientation of the spontaneous polarization. The walls can develop completely different electronic properties than the surrounding domains. In ferroelectric oxides, oxygen off-stoichiometry is an additional versatile control parameter, reflected by neutral ferroelectric domain walls in hexagonal manganites: Depending on the oxygen content, their conductance varies from insulating to conducting.

In this work, we change the conductance state of neutral ferroelectric domain walls in high-quality $Er(Mn,Ti)O_3$ single crystals from insulating to conducting by annealing the samples in reducing conditions, e.g., nitrogen. The process can be reversed by annealing under oxidizing conditions, reflecting the outstanding chemical flexibility of the domain walls. The sensitivity to off-stoichiometry can be exploited to utilize the domain walls for sensing applications.

Our results provide new insight into the impact of environmental parameters on the electronic domain wall properties. This is of interest for the development of atmospheric sensors, adding a new direction to the field of domain wall nanotechnology.

KFM 1.3 Mon 9:50 POT 51

Thermal and elastic stability of acceptor dopants in BaTiO3 — •Aris Dimou[1], Aldo Raeliarijaona[2], R. E. Cohen[2], and Anna Grünebohm[1] — [1]Interdisciplinary Center for Advanced Materials Simulation (ICAMS) and Center for Interface-Dominated High-performance Materials (ZGH), Ruhr-University Bochum, Germany — [2]Extreme Materials Initiative, Earth and Planets Laboratory, Carnegie Institution for Science, Washington, DC 20015-1305, USA

The presence of defects, such as vacancies and aliovalent substitutions significantly impacts the functional properties of ferroelectric materials. Defect dipoles formed by acceptor dopants and oxygen vacancies have gained interest after the demonstration of superlative, reversible piezoelectric [1], and electrocaloric [2] responses. But so far a deep understanding of their stability is missing.

We compare the thermal stability of Fe, Cu, and Mn dopants, and explore the impact of biaxial strain on the defect stability by density functional theory and *ab-initio* molecular dynamics. We found the largest barrier for the $Cu_{Ti} - V_O$ defect complexes, and exhibit the enhanced barrier height under compressive biaxial strain.

References

[1] X. Ren, Nature Materials, 3(2):91-94, 2004
[2] A. Grünebohm et al., Phys. Rev. B, 93(13):134101, 2016

KFM 1.4 Mon 10:10 POT 51

Probing hidden order in ferroelectric oxide thin films with single crystal diffuse X-ray scattering — •Joohee Bang[1], Nives Strkalj[2], Martin Sarott[1], Morgan Trassin[1], and Thomas Weber[1] — [1]Department of Materials Science, ETH Zurich, Zurich, Switzerland — [2]Department of Materials Science and Metallurgy, Cambridge University, Cambridge, United Kingdom

Ferroelectric thin films have attracted great attention for its rich applications in energy-efficient electronic devices because of functional properties such as high dielectric constants and electrically switchable polarization. Recently, ferroelectric oxide superlattices with complex topologies such as long-range vortex-antivortex arrays of polarization have garnered much interest as they hold promise for alternative device configurations for microelectronics [1]. Here, we report on a newly discovered local order state in ferroelectric superlattices using a complete three-dimensional diffuse X-ray scattering data, which was collected by taking advantage of high-energy synchrotron X-rays in ultra-small grazing incidence geometry. The data was analyzed with the 3D-deltaPDF method [2], which not only gives a three-dimensional view of the disorder, but also the access to weak disorder that was previously not accessible. This work will contribute to understanding structure-property correlations of ferroelectric oxide superlattices and lay groundwork for developing a novel solid-state characterization technique.

[1] Yadav et al., Nature 530, 206; S. Das et al., Nature 568, 2019
[2]Weber and Simonov, Z. Kristallogr. 227, 2012

15 min. break

Invited Talk KFM 1.5 Mon 10:45 POT 51

Domains or no Domains in Wurtzite-Type Ferroelectrics — •Simon Fichtner[1,2], Niklas Wolff[1], Tom-Niklas Kreutzer[2], Georg Schönweger[1,2], Adrian Petraru[1], Hermann Kohlstedt[1], Lorenz Kienle[1], and Fabian Lofink[1,2] — [1]Faculty on Engineering, Christian Albrechts University, Kiel, Germany — [2]Fraunhofer ISIT, Itzehoe, Germany

For decades, the wurtzite structure served as a posterchild for pyroelectric, i.e. spontaneously polarized materials which are yet not ferroelectric. The resulting inability to control the dipole ordering after synthetization has largely limited research on wurtzite-type polarization domains and –walls to their suppression. Therefore, the growth of single domain materials of defined polarity has been one of the ultimate goals for the main applications of wurtzite-type semiconductors in the fields of MEMS, RF-, power- and optoelectronics.

Today though, the discovery of ferroelectricity in wurtzite-type solid solutions promises unprecedent possibilities in terms of reconfigurable polarization control in wurtzite-type materials, but also creates the necessity for renewed scientific attention to the domains and -walls of this structure. This contribution aims to provide a vantage point for this attention by giving a glimpse into the literature on this subject and by reporting on our preliminary experimental work on domains in

wurtzite-type ferroelectrics. Regarding the latter, transmission electron and piezoelectric force microscopy next to chemical anisotropy studies allow us to draw first conclusions regarding the nucleation, coalescence and distribution of domains in this particular material class.

KFM 1.6 Mon 11:15 POT 51

The role of interfacial stress on the polarization stability of lead-free relaxor ceramics — •Julia Glaum[1,2], Yooun Heo[2], Matias Acosta[3], Pankaj Sharma[2], Jan Seidel[2], and Manuel Hinterstein[2,4] — [1]NTNU, Trondheim, Norway — [2]UNSW Australia, Sydney, Australia — [3]TU Darmstadt, Darmstadt, Germany — [4]KIT, Karlsruhe, Germany

The unique structural, dielectric and electromechanical properties displayed by canonical relaxor systems, make these materials fascinating objects for fundamental studies as well as for industrial applications. Contributors to these unique properties are the multiple crystallographic phases present simultaneously even in individual grains, as well as the ferroelectric-relaxor phase transformation, which can become reversible in the vicinity of the transition temperature.

Here, we report on the thermal evolution of the crystallographic phases in a (Bi1/2Na1/2)TiO3-BaTiO3 relaxor ceramic. This system exhibits two polar phases, with the minority phase embedded into the majority phase. While the majority phase retains a stable poling state with increasing temperature up to the transition to the relaxor state, a gradual de-texturization of the poling state is observed for the minority phase over the whole temperature range. The surface domain structure decays already at significantly lower temperatures than expected from bulk observations. Development of interfacial stresses between majority and minority phases and differences in local stress state between surface and bulk are discussed as driving factors of the phase transition dynamics.

KFM 1.7 Mon 11:35 POT 51

Effect of strain on the 3D domain structure of hexagonal manganites — Nelly Natsch, •Aaron Merlin Müller, Amadé Bortis, Manfred Fiebig, and Thomas Lottermoser — Department of Materials, ETH Zurich, 8093 Zurich, Switzerland

We simulate and visualize the three-dimensional domain structure of strained hexagonal manganites. Due to the improper nature of their ferroelectric order, hexagonal manganites exhibit unconventional vortex domain patterns. In 3D, these vortex domain patterns are characterized by vortex lines, which are 1D topological defects that form arbitrarily oriented loops in bulk materials. We show that when strain is applied in the ab-plane, the domains exhibit an additional stripe-like order that is oriented perpendicular to the ab-plane. In addition, we show that the strain also acts on the vortex lines, forcing them to form closed loops aligned in the same plane as the domain walls. In our simulations, we observe two types of loops: one type that expands and another type that contracts and collapses. We relate both the formation of the stripe-like domains and the evolution of the vortex-line loops to a force acting directly on the vortex lines which is induced by the applied strain. Our numerical investigation is performed with a phase-field model to simulate the domain structure in three dimensions using a Landau-free-energy expansion.

KFM 1.8 Mon 11:55 POT 51

Tracing property-determining structural alterations via scanning electron diffraction — •Ursula Ludacka[1], Jiali He[1], Emil Frang Christiansen[1], Shuyu Qin[2], Manuel Zahn[1,6], Zewu Yan[3,4], Edith Bourret[4], Antonius van Helvoort[1], Joshua Agar[2,5], and Dennis Meier[1] — [1]NTNU, Trondheim, Norway — [2]Lehigh University, Bethlehem, USA — [3]ETH Zurich, Zurich, Switzerland — [4]Lawrence Berkeley National Laboratory, Berkeley, USA — [5]Drexel University, PA, USA — [6]Augsburg University, Germany

Ferroelectricity originates from polar displacements of lattice atoms, suggesting a one-to-one correlation between electronic and structural properties at the atomic level. Scanning electron diffraction (SED), a subcategory of 4D-STEM, in combination with direct electron detection (DED) is a powerful tool for probing functional properties on the atomic scale. We demonstrate the potential and opportunities of this innovative SED approach using improper ferroelectric ErMnO3, an ideal model system as its basic ferroelectric properties, such as local conductivity changes and atomic-scale structure are well understood. By utilizing a convolutional neural network on the SED dataset, we can deconvolve intrinsic scattering phenomena on the atomic level (ferroelectric domains and domain walls) from extrinsic scattering phenomena, such as thickness, bending, or scan distortions. Our results on this topic contain new insights into atomic scale property-structure relations for ferroelectrics. However, the gained knowledge is applicable to complex oxides in general.