KFM 4: Focus: Domains and Domainwalls in (Multi)Ferroic II

Chair: Prof. Dr. Lukas Eng (TU Dresden)

Time: Tuesday 9:00–13:10 Location: POT 51

Invited Talk KFM 4.1 Tue 9:00 POT 51

Towards spatially resolved measurements of thermal transport and electrocaloric effects at the nanoscale in ferroelectric materials — Rebecca Kelly, Olivia Baxter, Fran Kurnia, Amit Kumar, Marty Gregg, and •Raymond McQuaid — School of Mathematics and Physics, Queen's University Belfast, Belfast, U.K.

Scanning Thermal Microscopy (SThM) is a promising Atomic Force Microscopy technique for mapping the thermal properties of materials at the nanoscale. In this talk, I will discuss how SThM could be a powerful tool for studying the role of microstructure on heat flow in ferroelectric materials.

Interest in using ferroic domain boundaries to enable active control of heat flow has been steadily growing over the last decade. However, direct, spatially resolved measurements of domain wall thermal transport have yet to be reported. In the first part of the talk, I will describe our SThM based approach to map spatial variations in thermal conductivity associated with microstructural inhomogeneity. We map the contrast in thermal response of the electrode/dielectric layers of a multilayer ceramic capacitor (MLCC) to demonstrate proof of principle thermal imaging and then use this approach to investigate the thermal transport properties of conducting domain walls in LiNbO3.

In the second part of the talk, I will discuss how SThM can be adapted to measure electric field induced temperature changes in electrocaloric materials with sub-micron spatial resolution. Using our approach, 2D spatially resolved maps of electrocaloric heating and cooling can be generated, here demonstrated in a BaTiO3 based MLCC.

KFM 4.2 Tue 9:30 POT 51

Real time polarization monitoring during growth for the design of artificial layered ferroelectrics — •Ipek Efe[1], Elzbieta Gradauskaite[1], Alexander Vogel[2], Marta D. Rossell[2], Manfred Fiebig[1], and Morgan Trassin[1] — [1]Department of Materials, ETH Zurich, Switzerland — [2]Electron Microscopy Center, Empa, Switzerland

Increasing the complexity of unit cells of ferroelectric oxides beyond the standard perovskite building block supports exotic functionalities such as superconductivity, magnetoresistance, and ferroelectricity. However, integrating complex crystal structures into epitaxial design is challenging, and routes to precisely monitor the non-perovskite systems have yet to be established. Here, we directly access the polarization dynamics of the layered ferroelectric model system Aurivillius $Bi_5FeTi_3O_{15}$ films during growth using in-situ optical second harmonic generation (ISHG). We identify the characteristic Aurivillius antipolar ordering of the dipoles along the growth direction, which leads to an oscillating intensity of the ISHG signal during the layer-by-layer deposition. In combination with reflection high-energy electron diffraction monitoring, we show how the polarization orientation of the films consistently changes from out-of-plane during the growth of perovskite blocks, to fully in-plane upon the completion of the unit-cell with the fluorite-like $(Bi_2O_2)^{2+}$ planes. We demonstrate how direct access to structure-dependent polarization dynamics during growth enables the development of novel layered systems incorporating various functional perovskite blocks into the Aurivillius structure.

KFM 4.3 Tue 9:50 POT 51

3D mapping of grain boundary chemistry in ferroelectrics — •Kasper Hunnestad, Jan Schultheiss, Anders Mathisen, Constantinos Hatzoglou, Antonius Helvoort, and Dennis Meier — Norwegian University of Science and Technology (NTNU), 7491 Trondheim, Norway

In this work, we study the impact of polar order on the local chemistry of charged grain boundaries, which naturally form in polycrystalline pyro- and ferroelectric materials during processing. Analogous to ferroelectric domain walls, these interfaces can develop new physical properties, representing intriguing functional 2D systems. Understanding the origin of the emergent interfacial phenomena, however, is a challenging task, requiring high-resolution imaging of the atomic-scale structure.

Here, we apply atom probe tomography (APT) to study the local chemistry at grain boundaries in ferroelectrics. APT combines high chemical sensitivity and accuracy with three-dimensional spatial resolution, allowing to map and quantify otherwise inaccessible changes in chemical composition. In our polycrystalline model system, $ErMnO_3$, we consistently find an enrichment of erbium and a depletion of oxygen at the grain boundaries. This trend occurs independently of the local charge state of each grain boundary and is unexpected as it implies a local violation of charge neutrality. Our results provide new insight into the defect chemistry at the grain boundaries in polar materials, suggesting pathways for local property engineering in ferroelectric oxides via grain-boundary-selective doping.

15 min. break

KFM 4.4 Tue 10:25 POT 51

Impact of defect dipoles on ferroelectric domain-walls motion — •Sheng-Han Teng and Anna Grünebohm — Interdisciplinary Centre for Advanced Materials Simulation (ICAMS) and Center for Interface-Dominated High Performance Materials (ZGH), Ruhr-University Bochum, Germany

Defect dipoles can alter the polarization switching and the dynamics of domain walls in ferroelectrics [1, 2], which may result in internal bias fields, domain-wall pinning [3] and bowing. In this work, we use *ab initio* based molecular dynamics simulations based on the effective Hamiltonian method [4, 5] to investigate the impact of defect dipoles on the domain-walls motion of tetragonal and orthorhombic phases in $BaTiO_3$. We find that the internal bias induced by defect dipoles may modify the potential energy landscape and leads to a completely different migration path for domain switching, which can pin and possibly even accelerate domain-wall motion.

[1] X. Ren, *Nat. Mater.* **3**, 91-94 (2004)
[2] A. Grünebohm, M. Marathe, R. Khachaturyan, R. Schiedung, D. C. Lupascu, and V. V. Shvartsman, *J. Phys.: Condens. Matter* **34**, 073002 (2021)
[3] A. Dimou, P. Hirel, and A. Grünebohm, *Phys. Rev. B* **106**, 094104 (2022)
[4] W. Zhong, D. Vanderbilt, and K. M. Rabe, *Phys. Rev. B* **52**, 6301-6312 (1995)
[5] T. Nishimatsu, M. Iwamoto, Y. Kawazoe, and U. V. Waghmare, *Phys. Rev. B* **82**, 134106 (2010)

KFM 4.5 Tue 10:45 POT 51

Revealing hidden ferroelectric domain walls in sub-surface regions and their electronic properties via non-destructive conductance mapping — •Jiali He[1], Manuel Zahn[1,2], Leonie Richarz[1], Ursula Ludacka[1], Erik D. Roede[1], Zewu Yan[3,4], Edith Bourret[4], István Kézsmárki[2], and Dennis Meier[1] — [1]NTNU Norwegian University of Science and Technology, Norway — [2]Universität Augsburg, Germany — [3]ETH Zurich, Switzerland — [4]Lawrence Berkeley National Laboratory, USA

Ferroelectric domain walls hold great promise for next-generation nanoelectronics. In particular, charged domain walls in improper ferroelectrics have triggered conceptually new application strategies. Although it is known that the electronic properties of domain walls are determined by their charge state, orientation, and curvature, non-destructive measurements of these parameters remain a major challenge. We investigate the correlation between electronic surface properties and hidden ferroelectric domain walls in $ErMnO_3$. By recording conductance maps using scanning electron microscopy (SEM) and conductive atomic force microscopy (cAFM) in combination with FIB-nanostructuring, we reveal that domain walls in surface-near regions give rise to distinct variations in surface contrast. The findings are rationalized in a simplified model, linking the contrast variations to the local charged state of the hidden domain walls, their orientation and distance from the surface. Our work introduces novel strategies to analyze the physical properties of ferroelectric domain walls in surface-near regions with nanoscale resolution in a non-destructive way.

KFM 4.6 Tue 11:05 POT 51

Electronic transport at pristine neutral ferroelectric domain walls in lead titanate — •Sabine Körbel[1] and Christophe Adessi[2] — [1]Institute of Condensed Matter Theory and Optics, Friedrich Schiller University Jena, Germany — [2]Institut Lumière Matière, Université Claude Bernard Lyon I, France

Ferroelectric domain walls are intrinsic interfaces that form spontaneously in ferroelectric materials, such as, for example, perovskite oxides. Whereas the ferroelectric perovskite oxide itself is an insulator, ferroelectric domain walls can be electrically conductive, as numerous experiments on different perovskite oxides have shown. This domain-wall conduction could serve, e.g., for charge-carrier transport in future photovoltaic absorbers. We investigated, using ab initio calculations based on Green's functions, the electronic transport along and through neutral ferroelectric domain walls in the prototype ferroelectric perovskite oxide $PbTiO_3$, and determined how the domain walls change the electronic transmission of bulk $PbTiO_3$ and the I-V curves of an ultrathin metal/perovskite/metal sandwich structure. We find that pristine neutral domain walls have a moderate effect on electronic transmission and I-V curve (within about one order of magnitude), much smaller than the experimentally measured conductivity increase at the walls by several orders of magnitude. We suggest that the measured conductivity increase does not directly originate in the electronic structure of the pristine neutral domain walls, but is caused by secondary effects, such as the accumulation of free charge carriers and/or the segregation of charged defects at the walls.

15 min. break

KFM 4.7 Tue 11:40 POT 51

Ferrielectric phase boundaries in antiferroelectric lead zirconate — •Gustau Catalan[1], Krystian Roleder[2], and Ying Liu[3] — [1]ICREA and ICN2, Barcelona, Catalonia — [2]Institute of Physics, University of Silesia in Katowice, Katowice, Poland — [3]ICN2-Institut Catala de Nanociencia i Nanotecnologia, Barcelona, Catalonia

When antiferroelectric PbZrO3 is cooled down from the paraelectric phase, antiferroelectric domains nucleate and grow until they coalesce. Adjacent antiferroelectric domains can differ in the phase sequence of the antipolar arrangement, meaning that there is a polar discontinuity at the domain wall, also known as translational boundary or antiphase boundary. Using high resolution transmission electron microscopy, we have characterized such phase boundaries. We find that their internal structure is ferrielectric, with a basic unit cell that can be described as having two dipoles ion one direction and one in the antiparallel direction. We also find that such translational boundaries can cluster together, forming stripe domains of an incipient ferrielectric phase that can moreover be internally switched. We therefore propose that antiphase boundaries can act as the seed for a low-temperature ferrielectric phase that has been theoretically predicted and which might explain the remnant polarization sometimes observed as triple hysteresis loops in nominally antiferroelectric lead zirconate.

KFM 4.8 Tue 12:10 POT 51

Field Effect Transistor employing the static negative capacitance of a ferroelectric nano-domain nucleus — •Pavel Mokry[1], Vit Kosina[1], and Tomas Sluka[2] — [1]Faculty of Mechatronics, Informatics and Interdisciplinary Studies, Technical University of Liberec, Liberec, Czech Republic — [2]CREAL SA, Ecublens, Switzerland

Miniaturization of conventional field effect transistors (FETs) approaches the fundamental limits beyond which opening and closing the transistor channel require such a gate voltage swing, which causes an unacceptable increase in heat generation. This problem could be reduced by placing a ferroelectric layer between the FET gate electrode and the channel. In this ferroelectric-semiconductor sandwich structure, the gate voltage can be amplified due to the negative capacitance regime of ferroelectrics. However, the original idea of using a bulk ferroelectric for voltage amplification suffers several difficulties. In this work, we provide phase-field simulations of a system that provides static negative capacitance from a nano-domain nucleus. We model the nucleus of a ferroelectric domain with reversed polarization produced by applying the voltage on a small gate electrode. We show that such a nano-domain nucleus represents a reversible system, which follows a unique path during electrical cycling and inevitably crosses a higher energy state characterized by negative static differential capacitance. Phase-field simulations confirm the robustness of this concept offering conveniently small effective negative capacitance and its compatibility with FET technology.

KFM 4.9 Tue 12:30 POT 51

Novel functionalities at twin domain crossings — •Kumara Cordero-Edwards[1,2], Philippe Tückmantel[2], Iaroslav Gaponenko[2], Sahar Saremi[3], Lane Martin[3,4], and Patrycja Paruch[2] — [1]Institut Català de Nanociencia i Nanotecnología, Barcelona, Catalonia. — [2]DQMP, University of Geneva, Geneva, Switzerland — [3]Department of Materials Science and Engineering, University of California, Berkeley, USA. — [4]Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, USA.

In ferroelectrics, domain walls (DWs) are thin interfaces separating regions with different orientations of electric polarization. These interfaces can present physical properties quite different from the surrounding domains, allowing them to be used as active components in future device applications. Recent studies of DWs using scanning probe microscopy (SPM) have focused on mapping their response to different parameters in order to understand their structure-property relationships. In particular, the role of high strain gradients present at ferroelectric twins has been shown to enhance their electrical conduction and can lead to complex rotational polarization textures.

Here, I will present our investigation of ferroelastic twin domains in epitaxial PbTiO3 thin films grown on SrTiO3, explored with SPM. Our results suggest a complex polarization structure around the twin domains, which present an unusual and distinct lateral PFM signal, associated with a distinct current signature. Moreover, twin domain crossings show a unique mechanical response distinct from the surrounding ferroelectric phase, and enhanced electrical conduction.

KFM 4.10 Tue 12:50 POT 51

Three-dimensional imaging of ferroelectric domains using digital holographic tomography — •Pavel Mokry[1,2], Marek Mach[1,2], Pavel Psota[1], and Karel Zidek[1] — [1]Faculty of Mechatronics, Informatics and Interdisciplinary Studies, Technical University of Liberec, Liberec, Czech Republic — [2]Institute of Plasma Physics of the Czech Academy of Sciences, Prague, Czech Republic

The formation and evolution of domain patterns in ferroelectrics are fascinating physical phenomena, which determine to a large extent, the macroscopic properties of ferroelectric samples. Therefore, imaging of ferroelectric domains belongs to the essential characterization techniques of ferroelectric materials. This work demonstrates the three-dimensional (3D) imaging of ferroelectric domains using Digital Holographic Tomography (DHT). Our method is based on the construction of the Digital Holographic Microscope, which allows taking several images of the domain pattern projections from different directions. The 3D image of the ferroelectric domain pattern is then numerically reconstructed using our original method called Curvilinear Filtered Back-projection. Our experimental method has been demonstrated by imagining the domain structure in periodically poled lithium niobate single crystals. The developed method allows fast and accurate 3D observations of ferroelectric domain structures in the whole volume of the ferroelectric single crystals on the centimeter scale. The recent DHM and DHT systems, which allow high-resolution optical imaging and on-chip optical imaging of ferroelectric domain patterns, are demonstrated and discussed.