

## KFM 1: Focus Session I: Ferroics - Domains and Domain Walls

The Focus Sessions: Ferroics - Domains and Domain Walls is dedicated to the detection of multiferroic and ferroelectric domain pattern, their manipulation as well as the modeling of domains. These domains and domain walls are fascinating building blocks for novel (nanoscale) electronics ranging from switches, memristive elements towards diodes and reconfigurable wires.

Chairman: Stephan Krohns (University of Augsburg)

Time: Tuesday 10:00–12:45

Location: H2

### Invited Talk

**KFM 1.1 Tue 10:00 H2**  
**Effect of humidity on the ferroelectric domain wall dynamics in BaTiO<sub>3</sub> thin films** — IRENA SPASOJEVIC<sup>1</sup>, ALBERT VERDAGUER<sup>2</sup>, GUSTAU CATALAN<sup>1</sup>, and NEUS DOMINGO<sup>1</sup> — <sup>1</sup>Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193 Barcelona, Spain — <sup>2</sup>Institut de Ciència de Materials de Barcelona (ICMAB), CSIC, Campus UAB, Bellaterra, 08193 Barcelona, Spain

The switching dynamics of ferroelectric polarization under electric fields depends on the availability of screening charges in order to stabilize the switched polarization. In ferroelectrics thin films with exposed surfaces investigated by piezoresponse force microscopy (PFM), the main source of external screening charges is the atmosphere and the water neck, and therefore relative humidity (RH) plays a major role. In this context dynamic writing of linear domains in BaTiO<sub>3</sub> thin films changes by varying scanning speeds in the range of RH between 2.5% and 60% reveal that the critical speed for domain writing increases non-monotonically with RH. Additionally, the width of line domains shows a power law dependence on the writing speed, with a growth rate coefficient decreasing with RH. The size of the written domains at a constant speed as well as the creep-factor describing the domain wall kinetics follow the behavior of water adsorption represented by the adsorption isotherm, indicating that the screening mechanism dominating the switching dynamics is the thickness and the structure of adsorbed water structure and its associated dielectric constant and ionic mobility.

**KFM 1.2 Tue 10:30 H2**

**Charged ferroelectric domain walls for a.c. signal control** — JAN SCHULTHEISS<sup>1</sup>, ERIK LYSNE<sup>1</sup>, LUKAS PUNTINGAM<sup>2</sup>, ZEWU YAN<sup>3,4</sup>, EDITH BOURRET<sup>4</sup>, STEPHAN KROHNS<sup>2</sup>, and DENNIS MEIER<sup>1</sup> — <sup>1</sup>NTNU Norwegian University of Science and Technology, 7034, Trondheim, Norway — <sup>2</sup>University of Augsburg, 86159, Augsburg, Germany — <sup>3</sup>ETH Zurich, 8093, Zurich, Switzerland — <sup>4</sup>Lawrence Berkeley National Laboratory, 94720, Berkeley, CA, USA

Ferroelectric domain walls are natural interfaces separating volumes with different orientation of the spontaneous polarization. Depending on the local charge state, the domain walls exhibit unusual direct current (d.c.) conduction ranging from insulating to metallic-like behavior. Because of their unique electronic properties, they bear great potential as nanoelectronic components, serving, e.g., as switches and synaptic devices. In contrast to the functional d.c. behavior at charged walls, their response to alternating currents (a.c.) falls into an uncharted territory. Here, we explore the a.c. characteristics of charged ferroelectric walls in ErMnO<sub>3</sub> in the adiabatic regime (kHz-MHz), using a combination of atomic force microscopy and macroscopic dielectric spectroscopy. We demonstrate a pronounced non-linear response at the electrode-domain wall junction, which correlates with the charge state of the wall. The dependence on the a.c. drive voltage and frequency enables us to reversibly switch between uni- and bipolar output signal, providing conceptually new opportunities for the application of charged domain walls as functional nano-elements in a.c. circuitry.

**KFM 1.3 Tue 10:45 H2**

**Conductivity control via minimally invasive anti-Frenkel defects in a functional oxide** — D. M. EVANS<sup>1,2</sup>, T. S. HOLSTAD<sup>2</sup>, A. B. MOSBERG<sup>2</sup>, D. R. SMÅBRÅTEN<sup>2</sup>, P. E. VULLUM<sup>3</sup>, A. L. DADLANI<sup>1</sup>, K. SHAPOVALOV<sup>4</sup>, Z. YAN<sup>5,6</sup>, E. BOURRET<sup>6</sup>, D. GAO<sup>2,7</sup>, J. AKOLA<sup>2,8</sup>, J. TORGENSEN<sup>2</sup>, A. T. J. VAN HELVOORT<sup>2</sup>, S. M. SELBACH<sup>2</sup>, and D. MEIER<sup>2</sup> — <sup>1</sup>University of Augsburg, Germany — <sup>2</sup>NTNU, Norway — <sup>3</sup>SINTEF, Norway — <sup>4</sup>ICMAB-CSIC, Spain — <sup>5</sup>ETH Zurich, Switzerland — <sup>6</sup>LBNL, USA — <sup>7</sup>Nanolayers Research Computing LTD, London, — <sup>8</sup>Tampere University, Finland

The control of conductivity is critical to any electronic device. In this context, oxide materials are particularly interesting as their conduc-

tivity can be continuously tuned via an electric field. In addition, they have a plethora of inherent functionalities arising from the electronic degrees of freedom, such as, superconductivity, magnetism, and ferroelectricity. However, utilizing both these changes in conductivity and electronic degrees of freedom simultaneously requires the ability to change one without affecting the other. Usually this is a problem, as the net redox reaction that gives the change in conductivity also affects the electronic degrees of freedom. In this talk, I demonstrate how stable, nanoscale, enhancement of conductivity can be achieved in ferroelectrics without net mass transfer, net change in stoichiometry, or the build-up of spurious electric and chemical gradients. This approach permits both the multiple orders of magnitude change in conductivity and the inherent functionality of oxides to be utilized independently and in parallel to each other.

**KFM 1.4 Tue 11:00 H2**

**Atomic-scale analysis of individual dopants in a functional oxide** — KASPER HUNNESTAD<sup>1</sup>, CONSTANTINOS HATZOGLOU<sup>1</sup>, ANTONIUS VAN HELVOORT<sup>2</sup>, and DENNIS MEIER<sup>1</sup> — <sup>1</sup>Department of Materials Science and Engineering, Norwegian University of Science and Technology (NTNU), 7491 Trondheim, Norway — <sup>2</sup>Department of Physics, Norwegian University of Science and Technology (NTNU), 7491 Trondheim, Norway

Oxide materials exhibit unique electronic and ionic properties that can readily be tuned via compositional variations and local defect chemistry. Intriguing examples are point-defect driven insulator-metal transitions, interfacial magnetism and superconductivity. However, 3D imaging of the individual point defects that are responsible for the emergent phenomena remains a challenge.

Here, we apply atom probe tomography (APT) to overcome this challenge, gaining first experimental insight into the 3D distribution of dopants in the multiferroic oxide ErMn<sub>0.998</sub>Ti<sub>0.002</sub>O<sub>3</sub>. We resolve the position of individual Ti atoms within the crystal lattice, and study local characteristics such as density fluctuations, gradient effects and clustering.

Our results establish a pathway for resolving individual dopants in functional oxides, bringing us an important step closer to understanding the complex atomic-scale physics and ultimately control lattice, charge and spin degrees of freedom at the local scale.

**15min. break.**

### Invited Talk

**KFM 1.5 Tue 11:30 H2**  
**Magnetic avalanche of non-oxide conductive domain walls** — SOMNATH GHARA<sup>1</sup>, KORBINIAN GEIRHOS<sup>1</sup>, LUKAS KUERTEN<sup>2</sup>, PETER LUNKENHEIMER<sup>1</sup>, VLADIMIR TSURKAN<sup>1</sup>, MANFRED FIEBIG<sup>2</sup>, and ISTVÁN KÉZSMÁRKI<sup>1</sup> — <sup>1</sup>Experimental Physics V, Center for Electronic Correlations and Magnetism, University of Augsburg, Augsburg, Germany — <sup>2</sup>Department of Materials, ETH Zurich, 8093 Zurich, Switzerland

Conductive domain walls have been exclusively observed in oxides, where off-stoichiometry and defects often hamper the domain wall conductivity and render the walls immobile and thus curtail their usefulness and flexibility. In this talk, we will show the giant conductivity of domain walls in the non-oxide multiferroic GaV<sub>4</sub>S<sub>8</sub>, investigated by macroscopic transport as well as microscopic PFM and c-AFM measurements. We observe a fascinating architectures of ribbon- and folded sheet-like conductive domain walls emerging in the polar rhombohedral state below its Jahn-Teller transition  $T_{JT} = 45$  K. Besides the giant negative magnetoresistance (~80%) inherent to these conductive domain walls, their high conductivity is exploited to trigger unprecedentedly large changes of the bulk resistance via on-demand magnetic or electric conversions between multi- and mono-domain states. Such a transformation to the insulating mono-domain state through an avalanche-like domain-wall expulsion process leads to an abrupt

conductance changes as large as eight orders of magnitude.

Reference: S. Ghara, K. Geirhos et al., Nature Communications 12, 3975 (2021).

KFM 1.6 Tue 12:00 H2

**Conductivity of ferroelectric domain walls in the lacunar spinel  $\text{GaV}_4\text{S}_8$**  — •LUKAS PUNTIGAM, DONALD EVANS, MARKUS ALTTHALER, SOMNATH GHARA, LILIAN PRODAN, VLADIMIR TSURKAN, STEPHAN KROHNS, and ISTVAN KÉZSMÁRKI — University of Augsburg, 86159, Augsburg, Germany

Ferroelectric domain walls, which can be written, tuned or erased at will, are being considered as functional building blocks for nano devices. Especially, the case of conductive domain walls, where their spatially confined electronic responses, that differ from the bulk of the host material, caused the idea of domain wall for nanoengineering.

To date, the origin for such increased conductivity in ferroelectrics domain walls has been attributed to the formation of screening charges driven by polar discontinuities.

Here, we discuss how other phenomena, such as strain, could induce enhanced conductivity of domain walls, using the lacunar spinel  $\text{GaV}_4\text{S}_8$  as a template system. This system exhibits ferroelectric domain pattern below the Jahn-Teller transition at 42 K. Temperature dependent conductive and piezoresponse force microscopy, as well as local  $I(V)$ -spectroscopy, are employed to understand the local conductivity. We reveal at low temperatures that the conductivity is non-trivial consistent with the conventional polar-discontinuity approach, but rather - at secondary domain walls - is consistent with a strain-based mechanism. This provides a new mechanism on generating conductivity at domain walls, which is not limited by polar discontinuity requirements.

KFM 1.7 Tue 12:15 H2

**Understanding the Electronic Structure of Lacunar Spinel  $\text{GaM}_4\text{X}_8$  by *ab initio* Multi-Configurational Calculations** — •THORBEN PETERSEN, LIVIU HOZOI, and ULRICH RÖSSLER — Institute for Theoretical Solid State Physics, Leibniz IFW Dresden, Germany

Generally, transition-metal based compounds show a large manifold of structural motifs that are based on molecular-like lattices which often lead to strong correlations due to the sharing of the valence electrons

between multiple transition metal centers [1]. In particular, lacunar spinels of the formula  $\text{GaM}_4\text{X}_8$  ( $A = \text{Ga}$ ;  $M = \text{V}, \text{Nb}, \text{Ta}$ ;  $X = \text{S}, \text{Se}$ ) are a representative class of such materials and have shown to exhibit various electronic and magnetic properties [2]. In this study, we apply *ab initio* multi-reference methods in the framework of CASSCF to understand the underlying electronic configuration of these materials. This will allow for the calculation of excited states that can be compared to experimentally available RIXS data of  $\text{GaTa}_4\text{Se}_8$  [3]. In addition, a thorough analysis of the electronic structure will path the way towards modeling inter-site couplings and associated magnetic properties of this material class.

[1] R. L. Dally *et al.*, Phys. Rev. B **102**, 014410 (2020). [2] I. Kézsmárki *et al.*, Nat. Mater. **14**, 11, 1116 (2015). [3] M. Y. Jeong *et al.*, Nat. Commun. **8**, 782 (2017).

KFM 1.8 Tue 12:30 H2

**Defeating depolarizing fields with artificial flux closure inultrathin ferroelectrics** — •ELZBIETA GRADAUSKAITE<sup>1</sup>, NATASCHA GRAY<sup>1</sup>, QUINTIN N. MEIER<sup>2</sup>, MARCO CAMPANINI<sup>3</sup>, THOMAS MORAN<sup>4</sup>, BRYAN D. HUEY<sup>4</sup>, MARTA D. ROSSELL<sup>3</sup>, MANFRED FIEBIG<sup>1</sup>, and MORGAN TRASSIN<sup>1</sup> — <sup>1</sup>Department of Materials, ETH Zurich, Switzerland — <sup>2</sup>CEA Grenoble, LITEN, Grenoble, France — <sup>3</sup>Electron Microscopy Center, Empa, Switzerland — <sup>4</sup>Department of Materials Science and Engineering, University of Connecticut, Storrs, USA

Material surfaces encompass structural and chemical discontinuities that often lead to the loss of the property of interest in the so-called “dead layers”. It is notably problematic in nanoscale oxide electronics, where the integration of ferroic materials into devices is obstructed by the thickness threshold required for the emergence of their functionality. Here, we report the stabilization of ultrathin out-of-plane ferroelectricity in oxide heterostructures through the design of an artificial flux-closure-like architecture. Inserting an in-plane polarized Aurivillius epitaxial buffer provides continuity of polarization at the interface, and despite its insulating nature we observe the emergence of polarization in our out-of-plane-polarized model ferroelectric  $\text{BaTiO}_3$  from the very first unit cell. Our model heterostructure further enables the stabilization of charged domain walls with pronounced chiral textures in multiferroic  $\text{BiFeO}_3$  films. Thus, we show that the smart integration of insulating materials can surpass standard metals in the design of the next generation ferroelectric-based oxide electronics.