Strain and polarization coupling in ferroelectric defect structures revealed by high-resolution HAADF-STEM

A Lubk[1], M Rossell[2], G Catalan[3], B Noheda[3], J Seidel[4], Y H Chu[4], R Ramesh[4], C Magen, M Hÿtch[5] and E Snoeck[5]

1. Triebenberg Lab, Institute for Structure Physics, Technische Universität Dresden, Germany.
2. Electron Microscopy Center, Empa, Swiss Federal Laboratories for Materials Science and Technology, CH-8600 Dübendorf, Switzerland.
3. Zernike Institute for Advanced Materials, University of Groningen, 9747 AG Groningen, The Netherlands.
4. Department of Materials Science and Engineering, University of California, Berkeley, USA.
5. Laboratory of Advanced Microscopy INA - University of Saragossa C/ Mariano Esquillor, Zaragoza, Spain
6. CEMES-CNRS, Université de Toulouse, 29 rue Jeanne Marvig, 31055 Toulouse, France.

Axel.Lubk@triebenberg.de
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The performance of ferroelectric devices, e.g. the ferroelectric field effect transistor, is strongly influenced by the presence of crystal defects such as edge dislocations (EDs) and domain walls (DWs). For instance the limited mobility of DWs determines polarization switching and introduces loss; misfit EDs lead to the formation of ferroelectric dead-layers at interfaces and provide pinning centres for domain walls hampering their mobility. Hereby, the intrinsic coupling between strain and polarization plays a crucial role. It also facilitates functional optimization by an adequate choice of substrate in ferroelectric thin films (strain engineering) [1] and high piezoelectricity in compositionally engineered morphotropic phase boundary ferroelectrics. The details of the coupling are still under debate, i.e., only recently the important role of strain gradients has been highlighted (flexoelectricity) [2].

Here, we investigate strain-polarization coupling around DWs and EDs in ferroelectric $BiFeO_3$ (BFO) and $PbTiO_3$ (PTO) thin films, both archetypal ferroelectric materials with particularly large electric polarization. We use a combination of aberration-corrected high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and a dedicated model-based structural analysis algorithm to determine atomic column positions with a precision *O*(pm). Unit-cell wise resolved strain and polarization are readily obtained by analysing suitable column distances. The results are subsequently interpreted by means of density functional theory (DFT) and phase field simulations.

The investigated epitaxial PTO thin film contains a twinned a-c domain structure relaxing the mismatch to the substrate. HAADF-STEM revealed strain gradients in vertical and horizontal direction in the domains. In addition we observe a polar rotation of the polarization vector, usually characteristic for morphotropic phase boundaries, which is here attributed to flexoelectricity [3].

In BFO we provide direct experimental proof for the straight DW structure predicted by DFT as well as the recently proposed theory of diffuse DWs [4]. In that theory abrupt 90° DW steps (Miller-Weinreich model), facilitating DW motion, are replaced by diffuse slopes (Fig. 1), which are shifted much easier along the DW and hence resolve the long standing discrepancy between experimentally measured and theoretically predicted DW mobilities.

An intricate situation occurs at EDs observed in BFO, where the ED topology induces a strong anisotropic strain field. Strain and polarization maps reveal a wealth of material states. Within the highly strained region close to the ED core orthorhombic BFO nanodomains with large aspect ratios form [5]. We identify a particular piezoelectrically driven pinning mechanism of 109° (and 180°) DWs parallel (and inclined) to the Burgers vector of the ED (Fig. 2). This observation suggests a new route to DW engineering via ED arrays tuned by substrate film lattice misfits. Furthermore, unusual head-to-head and tail-to-tail DWs coupled to EDs have been observed. [6]

References

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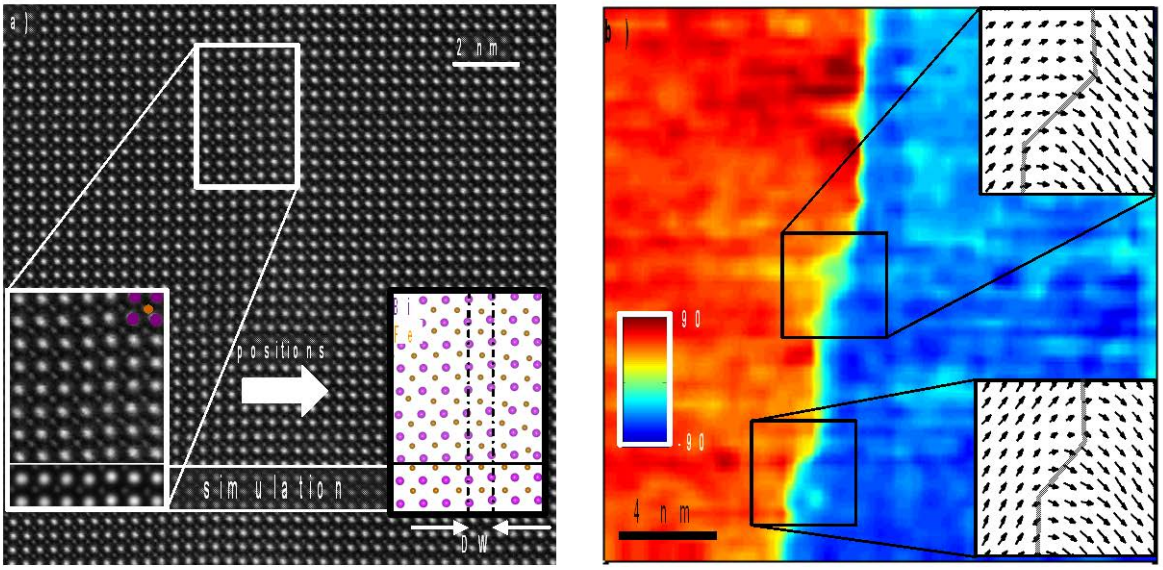

Figure 1. *a) HAADF-STEM containing a BFO 109° DW. The insets show a magnified straight part of the DW and the determined positions. Results of STEM imaging simulations based on the DFT structure show excellent agreement with the experiment. b) Polarization rotation field at 109° DW containing steps. The polarization vector field (insets) shows a diffuse rotation and attenuation across a diagonal boundary (slopes).*

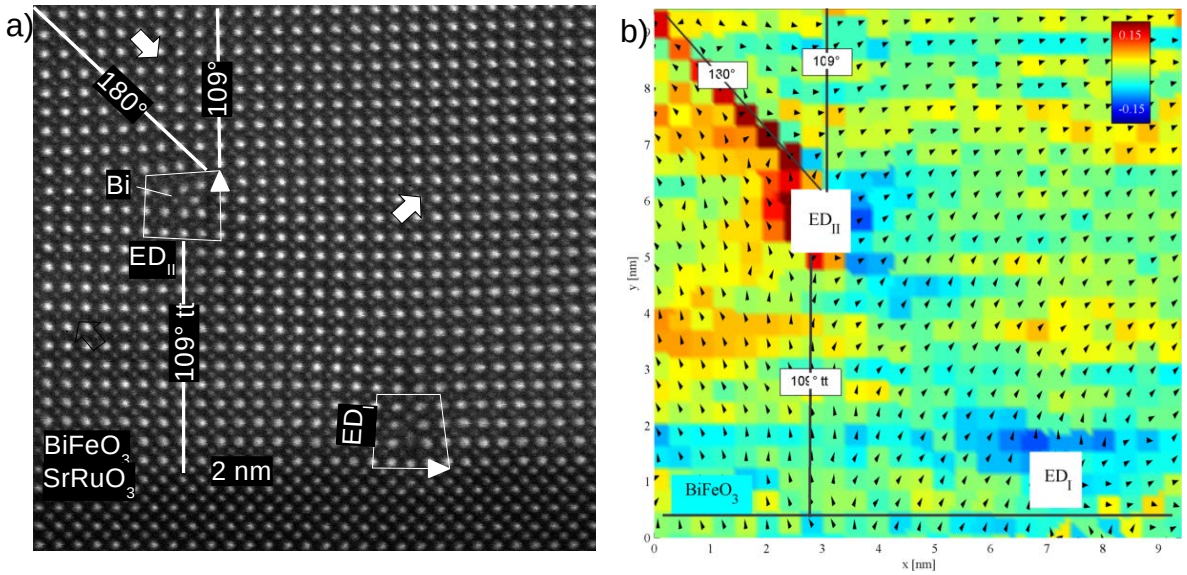

Figure 2. *a) HAADF-STEM of 2 perpendicular EDs. Burgers loops indicate the position of the EDs. b) Measured ε_{yy} strain component (color) and electric polarization (arrows). The DWs are indicated by straight lines and their nominal rotation angle. An unusual tail-to-tail DW is visible below ED_{II}.*