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Ilka Hermes

Piezoresponse force microscopy for novel computing components

In the future, ferroelectric components, such as ferroelectric field-effect transistors or tunnel junctions, could act as building blocks in novel computing concepts including neuromorphic computing. These applications require an in-depth understanding of the used ferroelectric materials, from domain structure to polarization switching behavior. Piezoresponse force microscopy, a functional atomic force microscopy technique, can access this information with a nanoscale resolution and even manipulate or tailor customized domain patterns.

In their native form, ferroelectrics have no macroscopic polarization. Instead, the materials form many small domains of opposite polarization orientation that compensate the overall polarization to lower the energy. However, if you subject ferroelectrics to an external electric field, you can switch the polarization orientation either globally or locally, and even draw images or tailored patterns of domains with different polarization orientations (Figure 1b) [1]. This adaptability is what makes ferroelectrics so interesting for modern computing technologies: Ferroelectric random access memories (FeRAMs) store information by inscribing information into the ferroelectric storage medium via local polarization switching [2]; Ferroelectric field-effect transistors (FeFETs) use ferroelectric materials in their gate stack to adjust the threshold voltage via the local polarization direction [3]; Ferroelectric tunnel junctions (FTJs) change the tunneling resistance based on the polarization and could serve as building bock in artificial synapses for neuromorphic computing [4].

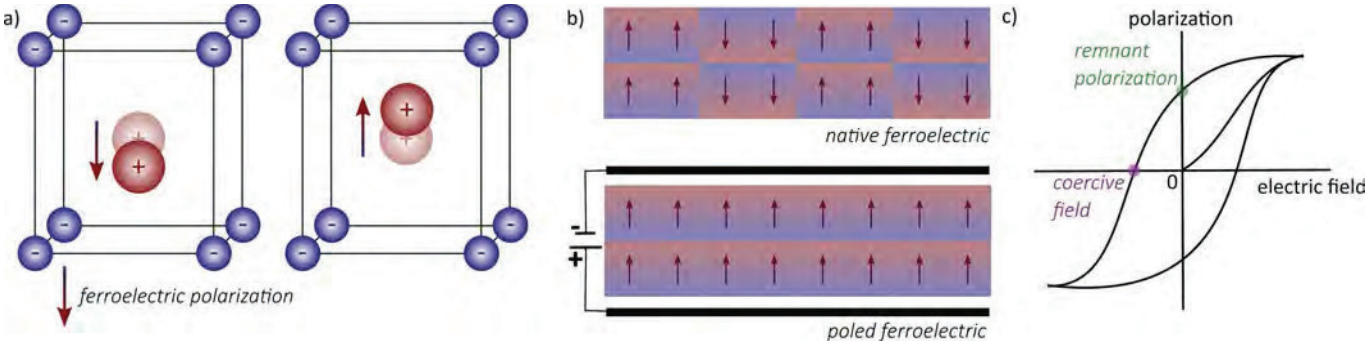

Fig. 1: a) Upwards or downward ferroelectric polarization state in a tetragonal unit cell, due to off-centering of a positive center-ion with respect to negative counter ions. b) Compensation of macroscopic polarization due to domains with opposite polarization orientation in the native state of ferroelectrics and poled state after external field application. c) Ferroelectric hysteresis.

Ferroelectric materials feature an intrinsic electrical polarization that is stable in two or more orientations: Imagine a crystalline material with a tetragonal unit cell, in which a positively charged center-ion experiences a directional electrostatic pull from some of the coordinating negative counterions or a steric hindrance due to a size mismatch between center-ion and counterions. These electrostatic interactions or steric effects can force the center-ion off its original position – for example by moving it a little bit up or down. The off-centering consequently induces a spatial misbalance between the negative and positive charges in the unit cell which leads to an electrical polarization pointing either upwards or downwards, depending on the shift of the center-ion (Figure 1a). If the center-ion has the same energy in its upward and downward shifted state, you now have two oppositely oriented, but equally stable ferroelectric polarization states [1].

To push these potential applications into commercialization, we have to understand ferroelectric domain structures and their switching behavior precisely and down to the nanoscale. Here, piezoresponse force microscopy (PFM) comes into play. PFM is a functional atomic force microscopy (AFM) method, which means simultaneous to the surface structure (topography), it can deliver functional information on local ferroelectric properties. In a PFM measurement, an electrically conductive tip attached to a cantilever scans the surface in contact mode with a constant interaction force (Figure 2). This constant force is enabled by a height feedback that keeps the cantilever deflection constant by adjusting the tip-sample distance at each pixel of the scan. At the same time, an AC voltage is applied between the tip and the back electrode *below* the ferroelectric sample.

Dr. Ilka Hermes
Institut für Physikalische Chemie & Physik der Polymere
Leibniz Institut für Polymerforschung
Hohe Straße 6, D-01069 Dresden
hermes@ipfdd.de

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Since all ferroelectrics are piezoelectric [1], this AC voltage induces a mechanical oscillation in the material at the same frequency as the applied AC voltage. The oscillating material piezoresponse can be monitored in the cantilever deflection and decoupled from the topography using a lock-in amplifier [5].

Although difficult to quantify, the amplitude of the oscillating piezoresponse – the PFM amplitude – can give us information on the piezoelectric coefficient of the ferroelectric, i.e., how much the material extends or contracts in a given direction per applied Volt. In addition, the PFM amplitude shows the positions of domain walls as local minima. The phase of the piezoresponse – the PFM phase – captures the polarization orientation of the domains. For example, domains with opposite out-of-plane polarization would result in a 180° PFM phase contrast [5].

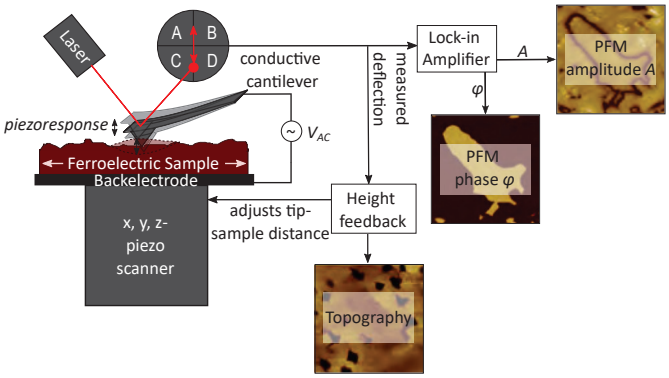

Fig. 2: Experimental setup of PFM, capturing the sample's topography and ferroelectric domains from the local piezoresponse induced by the AC voltage V_{AC}. The PFM amplitude shows the positions of domain walls as local minima and the PFM phase the polarization orientation in the domains. The exemplary images were measured on a bismuth ferrite thin film.

Since the piezoresponse per applied Volt is often times only a few tens of picometers or even below, we can enhance the signal by moving the AC excitation to the contact resonance of the cantilever and thereby use the natural amplification of oscillating systems. However, particularly for rough material surfaces, the contact resonance is not very stable. In fact, we have observed shifts in the contact resonance of up to 10 kHz on the same sample, in dependence of the local topography [6]. The resulting changes in PFM amplitude and phase can be easily mistaken for real sample responses. In order to avoid this topographic crosstalk, it is best practice to either measure far from the resonance for those sample with sufficiently large piezoresponse, or to employ dual amplitude resonance tracking (DART). DART PFM introduces an additional frequency feedback that adjusts the frequency of the AC excitation on each pixel of the scan to match the contact resonance [7]. Since its implementation requires additional lock-in amplifiers, DART is only offered on a few commercial AFMs or in combination with additional external electronics.

Besides domain imaging, PFM also allows insights on the local polarization switching behavior of ferroelectrics, which are important for their application in modern computing as FeRAM, FeFETs as well FTJs. In general, the polarization switching in ferroelectrics is described by the ferroelectric hysteresis (Figure 1c). A DC voltage is swept back and forth while the polarization is monitored. The polarization will switch, once the applied voltage exceeds the coercive field, positive and negative. Macroscopically, the polarization is measured via a reference capacitor; Microscopically, we can detect the polarization in the PFM phase in PFM switching spectroscopy. In order to do so, we stop the AFM scan at the position of interest and alternatingly apply DC voltages of increasing magnitude and AC voltages for PFM detection between tip and sample. The reason why we are alternating between excitation and detection is to minimize electrostatic crosstalk during the PFM detection. In a more recent development, researchers were able to automatize PFM switching spectroscopy using machine learning approaches for higher throughput material characterization. In the future this high throughput characterization may aid to fine-tune material composition to required ferroelectric properties for certain applications [8].

An innovative PFM approach for polarization switching and domain customization has just been presented by a group of researchers from the US (Figure 3). They combined the local writing of domain patterns via a conductive AFM tip with unusual scan trajectories: Instead of scanning back-and forth in a rectangular pattern as common in AFM, they were able to move the tip in a spiral motion or in a flower-like pattern, while applying a DC voltage to the tip. These scan trajectories together with the DC voltage allowed them to tailor nanodomains with controlled polarization orientations, that, among others, introduced charged ferroelectric domain walls [9]. Charged domain walls occur, when positive ends of the polarization ("head") of two neighboring domains point towards each other or the negative ends ("tails"), respectively. This polarization orientation leads to an energetically instable configuration with an absolute charge at the domain walls. Charged domain walls can serve as two-dimensional electronic conduction pathways through otherwise insulating ferroelectric materials and carry

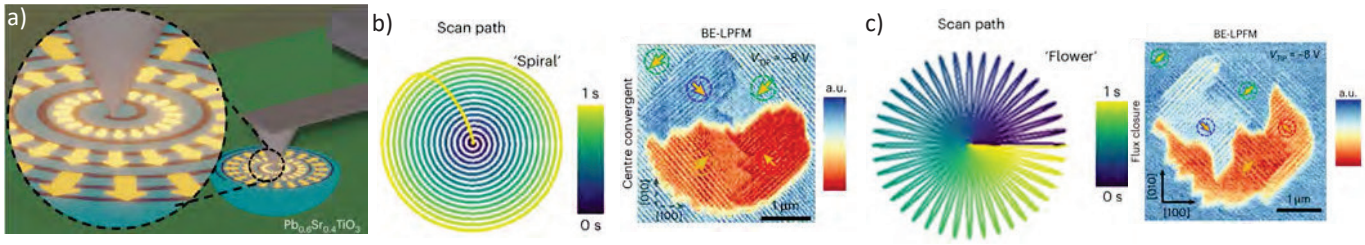

Fig. 3: a) Illustration of experimental PFM setup with customized scan trajectories. b) "Spiral" writing trajectory and corresponding in-plane PFM measurements after writing with +8 V demonstrating convergent polarization states in a head-to-head orientation. c) "Flower" writing trajectory and corresponding in-plane PFM measurements after writing with -8 V demonstrating flux-closure polarization states. The polarization orientations of the domains are shown with the orange arrows in b and c [9]. Copyright © 2024, UT-Battelle, LLC, 2024

a variety of other unique physical properties. In particular, the mobile nature of ferroelectric domain walls makes themselves intriguing candidates for nanoelectronic components such as artificial synapses for neuromorphic computing [10].

To summarize, PFM can further deepen the knowledge on ferroelectric properties such as domain formation, structure and switching behavior. Researchers have demonstrated that PFM can even be applied to customize domain patters that could find future application in novel computing concepts.

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Dr. Ilka Hermes

Ilka Hermes studied chemistry at the Johannes Gutenberg University in Mainz and began her work on scanning probe microscopy (SPM) in 2013 to resolve the arrangement of water molecules on mineral surfaces. For her dissertation, she moved to the Max Planck Institute for Polymer Research, where she used electrical and electromechanical SPM to understand the impact of different structural interfaces on charge carriers in perovskite solar cells. After a three-year stint in industry at the SPM manufacturer Park Systems Europe GmbH, Ilka started a junior research group for advanced correlative SPM at the Leibniz Institute of Polymer Research in Dresden, where her group is investigating the impact of the nanostructure on charge transport in perovskite solar cells and novel 2D materials.

The art of SPM

Artist: **Kim Noelle Dreier (Bielefeld University)**

SPM technique: **Dynamic Atomic Force Microscopy (amplitude modulation mode)**

Investigated System: **silver iodide AgI (0001) plane in n-heptane**

Title: ***"The organic-inorganic aquarium"***