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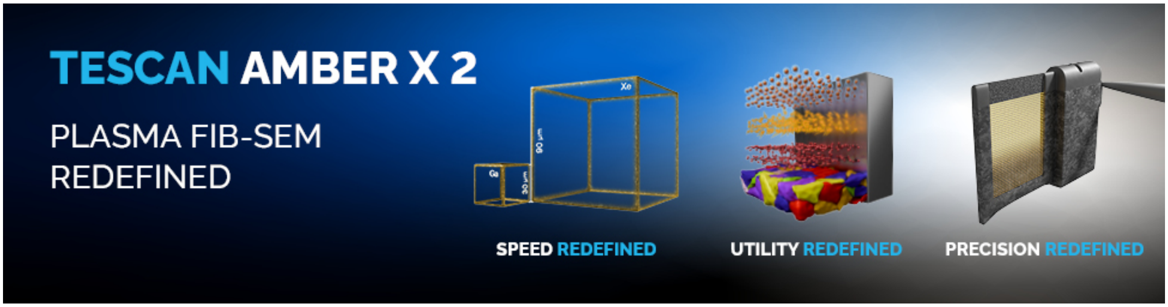

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Meeting-report

Nano-PUND and STEM EBIC Imaging for Ferroelectric Polarization Mapping

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In any material, electric charges shift in response to applied fields. For well over a century now, we have had a theoretical framework – Maxwell's equations – to describe the shifting charge and the applied fields. The task still at hand, however, is to measure, describe, and understand how real materials behave within this framework.

The displacement field $\boldsymbol{D}$ is defined in terms of its relationship to the electric field $\boldsymbol{E}$ and the polarization $\boldsymbol{P}$ via $\boldsymbol{D} \equiv \epsilon_0 \boldsymbol{E} + \boldsymbol{P}$. The displacement field's great theoretical advantage is that, via the first Maxwell equation $\nabla \cdot \boldsymbol{D} = \rho_f$, it depends only on the free charge density ρ_f [1]. Motion of the free charge density generates the free currents ($\partial \rho_f / \partial t + \nabla \cdot \mathbf{J}_f = 0$), which can be measured and controlled using standard electrical transport methods. Thus information on the displacement field $\boldsymbol{D}$ is experimentally accessible in microfabricated electronic circuits, where conducting leads make connections to some material or device of interest: electrical currents in the leads can be measured. The electric field $\boldsymbol{E}$ is also experimentally accessible in a device with leads, as a potential V applied to the leads produces $\boldsymbol{E} = -\nabla V$. However, the polarization $\boldsymbol{P}$ in some insulating material can be difficult to determine, as it is not directly measurable. Moreover, polarization depends on materials' properties that can be difficult to calculate, even in the ideal case. And in real materials the problems are truly forbidding, as composition inhomogeneities, defects, and other non-idealities make characterizing even the basic physical structure challenging.

For many technological applications, however, the polarization is of paramount interest. Ferroelectric materials, for instance, exhibit remanent polarization that, properly harnessed, could form the basis for a next-generation computer memory technology [2]. The combination of nano-PUND and STEM EBIC imaging can measure the displacement field $\boldsymbol{D}$ and the electric field $\boldsymbol{E}$ separately, which determines the polarization $\boldsymbol{P}$.

In the positive-up, negative-down (PUND) technique, two identical voltage pulses of the same, say, positive polarity are applied to a ferroelectric material in the polarized down state while the current is measured. The polarization switches only during the first pulse, so subtracting the second pulse's current from the first isolates the ferroelectric switching current. Nano-PUND improves this technique's dynamic range by injecting a current calibrated to cancel linear charging currents due to, for example, stray capacitance [3].

However, transport techniques such as PUND alone cannot determine the switching displacement field $\boldsymbol{D}$ if the material under test is not completely homogeneous. For instance, in the case of hafnium zirconium oxide ($\mathrm{Hf_{0.5}Zr_{0.5}O_2}$, HZO), our demonstration material [4], many crystalline phases of similar free energies compete with the desired ferroelectric phase. Thus the HZO between two electrodes might not all be ferroelectric. And even if it is, it might not all switch at the same coercive field. To connect the measured switching current I with the current density $\boldsymbol{J}$ that relates to $\boldsymbol{D}$, a technique capable of determining the cross-sectional area of the switching domains is required.

Scanning transmission electron microscope (STEM) electron beam-induced current (EBIC) imaging can map which domains switch, and when. In STEM EBIC imaging, the focused STEM beam is rastered over the sample while the beam-induced currents in the sample are collected and digitized. The EBIC contrast mechanism that is often the strongest arises because the primary electron beam is energetic enough to create electron-hole pairs in the sample [5]. In regions where there is a local electric field, these electron-hole pairs are more likely to separate and contribute to the EBIC. A STEM EBIC image is thus a map of the local $\boldsymbol{E}$-fields. This map can even be calibrated by applying a known $\boldsymbol{E}$-field and measuring the resulting EBIC. Thus STEM EBIC imaging assists PUND in the determination of $\boldsymbol{D}$ (by determining the switching area), and measures local $\boldsymbol{E}$-fields itself directly. This powerful combination thus determines the polarization $\boldsymbol{P}$ as well, providing a complete picture of the fields in a ferroelectric sample [6].

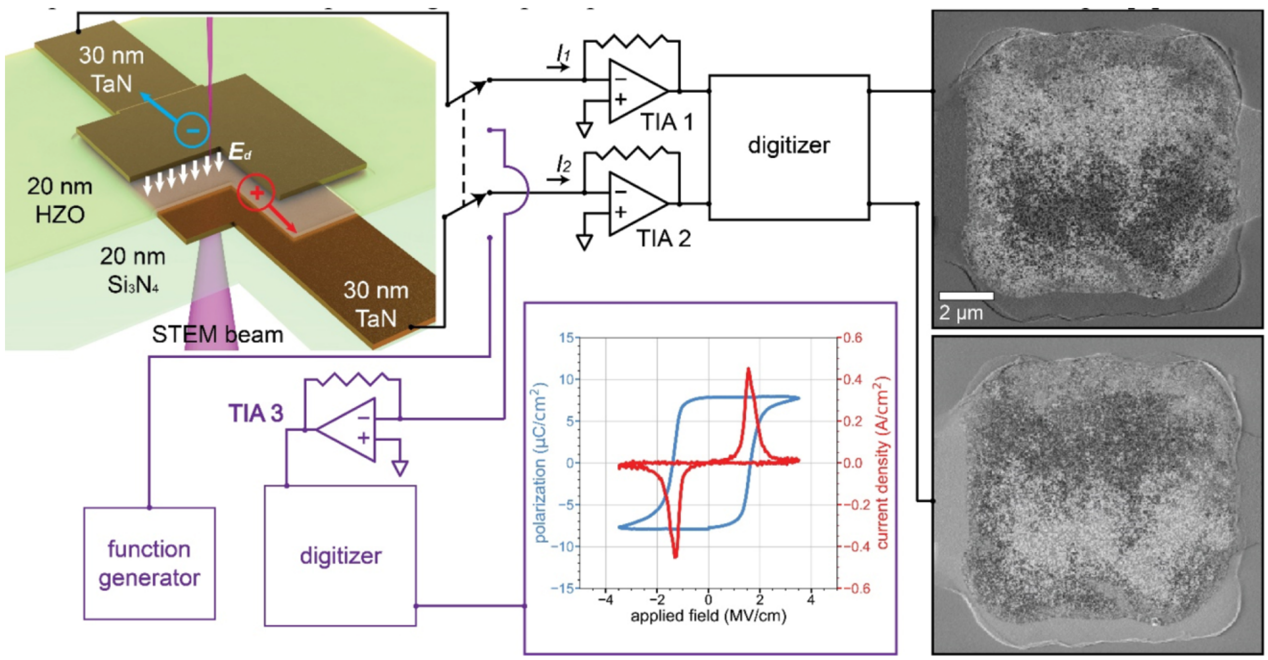

Fig. 1. Overview of the experimental arrangement for in-situ STEM EBIC and PUND measurements. A cutaway view of a polarized TaN/HZO/TaN capacitor showing separation of electron-hole pairs due to remanent electric fields. The PUND setup (purple) polarizes the film and measures the sample's global hysteresis loop. The "polarization" measured by PUND alone is more accurately termed the displacement, and it is calculated assuming that the whole capacitor switches (see Fig. 2). The EBIC setup (black) maps the device's local polarization state immediately after each PUND sequence. Positive (hole) current appears as bright contrast and negative (electron) current appears as dark contrast in the digitized images on the right.

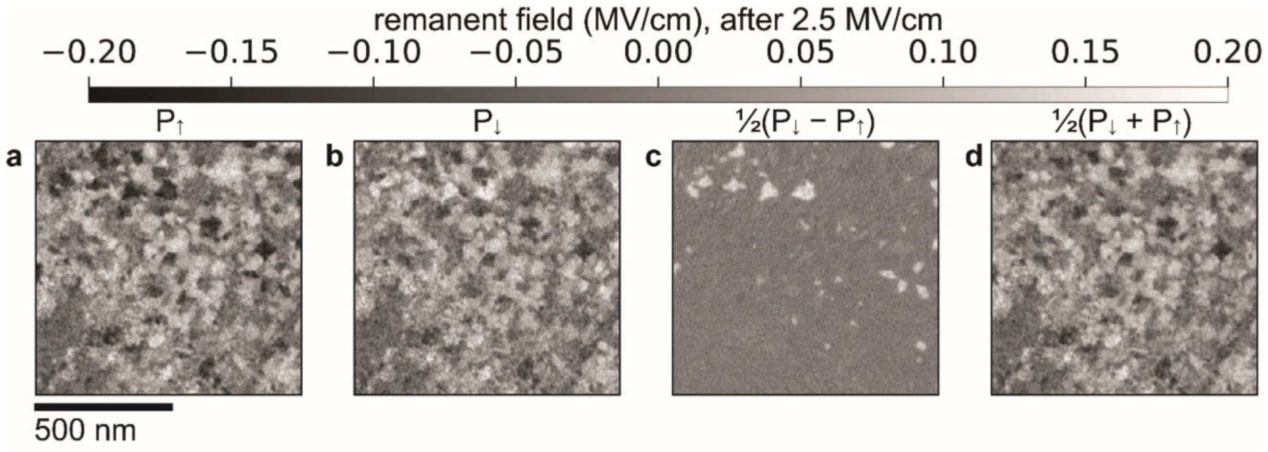

Fig. 2. Remanent field maps (a - d) show the local electric fields in small portion of the TaN/HZO/TaN capacitor shown in Fig. 1. These images, which are constructed by subtracting the EBIC collected from the bottom electrode from that of the top electrode, are calibrated in **E**-field units by applying a known voltage to the electrodes separately. At this peak PUND applied field strength magnitude of 2.5 MV/cm, only a portion of the capacitor switches from the nominally-$P_\uparrow$ (a) to the nominally-$P_\downarrow$ (b) state. Polarization state difference (c) and sum (d) image highlight the switching and non-switching domains, respectively. Transport methods such as PUND only see the switching domains (c), and cannot determine the switching area without the assistance STEM EBIC imaging.

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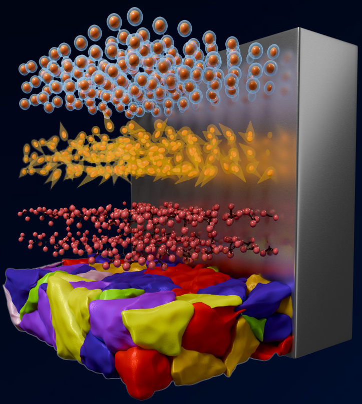

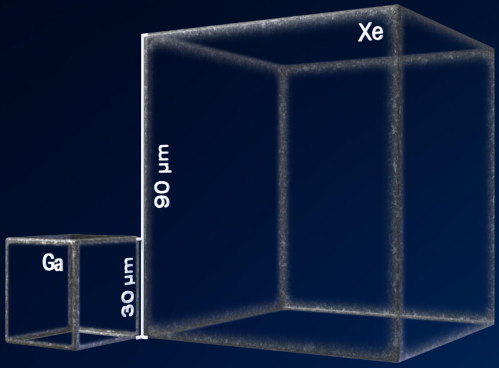
Xe
90 µm
Ga
30 µm