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Imaging Local Polarization and Domain Boundaries with Picometer-Precision Scanning Transmission Electron Microscopy

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Domain walls and their motion play an essential role in ferroelectric and ferromagnetic materials. The motion of the walls provides key functionalities, such as reversible computer memory. Emergent phenomena, such as enhanced conductance, permittivity and permeability, have also been stabilized at domain walls. Also, the elusive coupling of ferroelectricity and ferromagnetism is promising for next-generation memory devices. In order to fully harness domain walls, it is critical to understand their atomic scale structure and properties. Here we use atomic mapping to probe ferroelectricity in hexagonal $LuFeO_3$ and $ErMnO_3$ (Fig 1a) at all relevant length scales. At the picometer scale, we find the local polarization by mapping the Lu (Er) buckling, which drives the improper ferroelectricity [1]. At the nanoscale, we characterize the domain architecture. Atomic-scale feedback on ferroelectric polarization and domain structure has helped lead us to a new ferrimagnet-ferroelectric just below room temperature.

To create a high-temperature multiferroic, we integrate the ferroelectric $LuFeO_3$ and the ferrimagnetic $LuFe_2O_4$ (Fig 1a) into superlattices that exceed both end-members in critical temperature (T_C), moment, and polarization. We investigate the polarization in $(LuFeO_3)_m(LuFe_2O_4)_1$ as a function of *m* in a 100 keV NION Ultra-STEM. Each image was formed by many fast acquisitions, with a large beam current and stable stage enabling rapid, minimally distorted images. 2D Gaussian fitting located the center of the lutetium columns with 1 pm precision [2]. With the Lu displacements, color overlays and data from each Lu row illustrate the domain structure and polarization (Fig 1b). By analyzing over 90,000 Lu columns, we collect statistics as a function of composition (Fig 1c). As *m* increases, the polarization increases, with enhanced distortions neighboring the $LuFe_2O_4$ layers for $m>5$. The distortions in the $LuFe_2O_4$ reduces magnetic frustration and increases T_C up to 281 K for $m=9$. The domain walls become regular with increasing *m*, with tail-to-tail walls aligning on the $LuFe_2O_4$ layers. At these charged domain walls, DFT indicates that hole doping on the $LuFe_2O_4$ layers boosts the magnetic moment (Fig 1c).

Removing the constraints of the multilayer geometry increases the diversity of domains in this class of improper ferroelectrics, including polarization vortices. In bulk $ErMnO_3$ the polarization is damped more strongly near charged domain walls. At the vortex center, where all six domains meet (Fig 2), we see the amplitude of the polarization gradually vanishes, in contrast to conventional ferroelectrics. [3]

References:
[1] H Das, *et al*, Nat. Comm. **5** (2014) 2889.
[2] A Yankovich *et al*, Nat. Comm. **5** (2014) 4155.
[3] Work supported by the U.S. DOE BES, Award #DE-SC0002334. EM Facility support from the NSF MRSEC program (DMR 1120296).

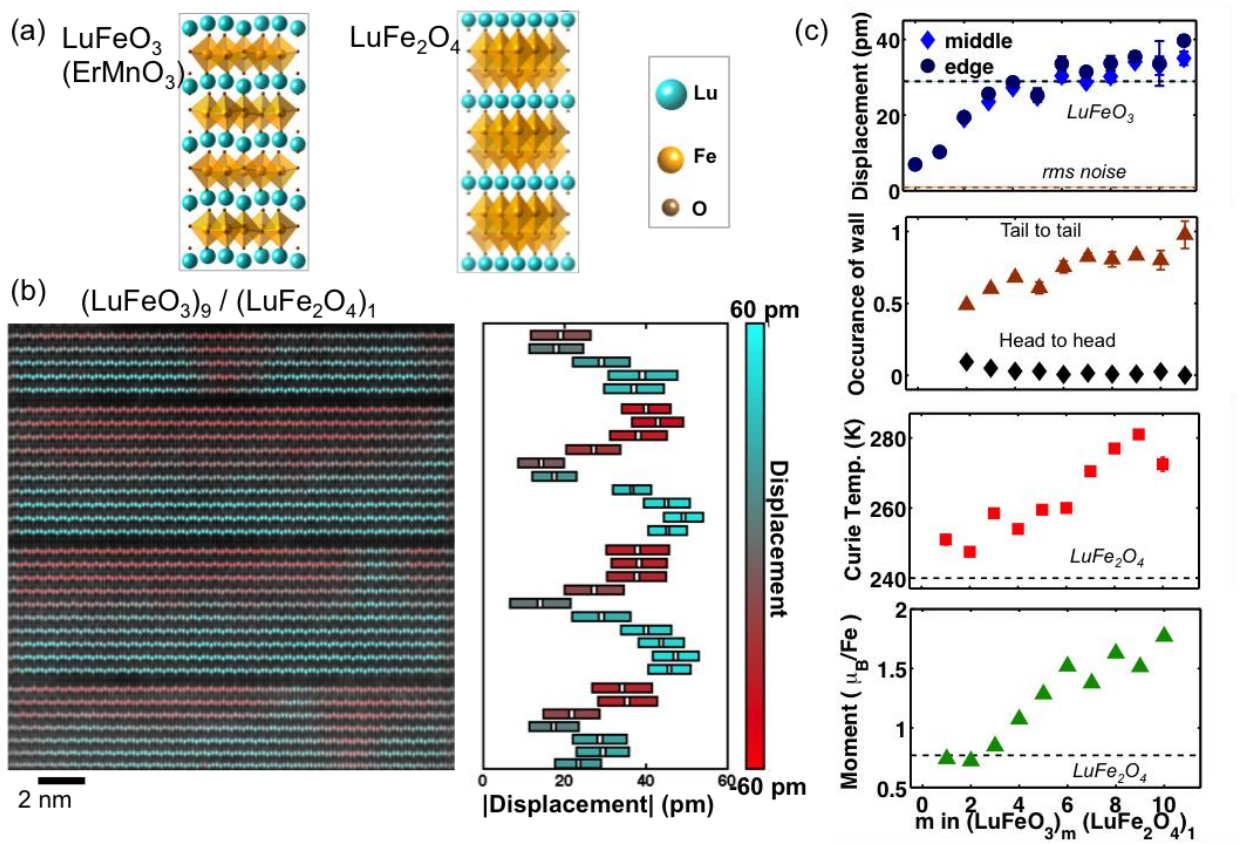

Figure 1. (a) schematic of $LuFeO_3$ and $LuFe_2O_4$, with Lu distortions driving the polarization in the $LuFeO_3$. (b) STEM image of the $(LuFeO_3)_9$ / $(LuFe_2O_4)_1$ with a color overlay corresponding to the Lu displacement, see colorbar. For each Lu row, the mean displacement is plotted with 20 – 80 % spread. (c) Plots of the Lu displacement, occurrence of domain walls neighboring the $LuFe_2O_4$ layer, T_C, and M for the superlattices, with enhanced properties for large *m* in $(LuFeO_3)_m$ / $(LuFe_2O_4)_1$.

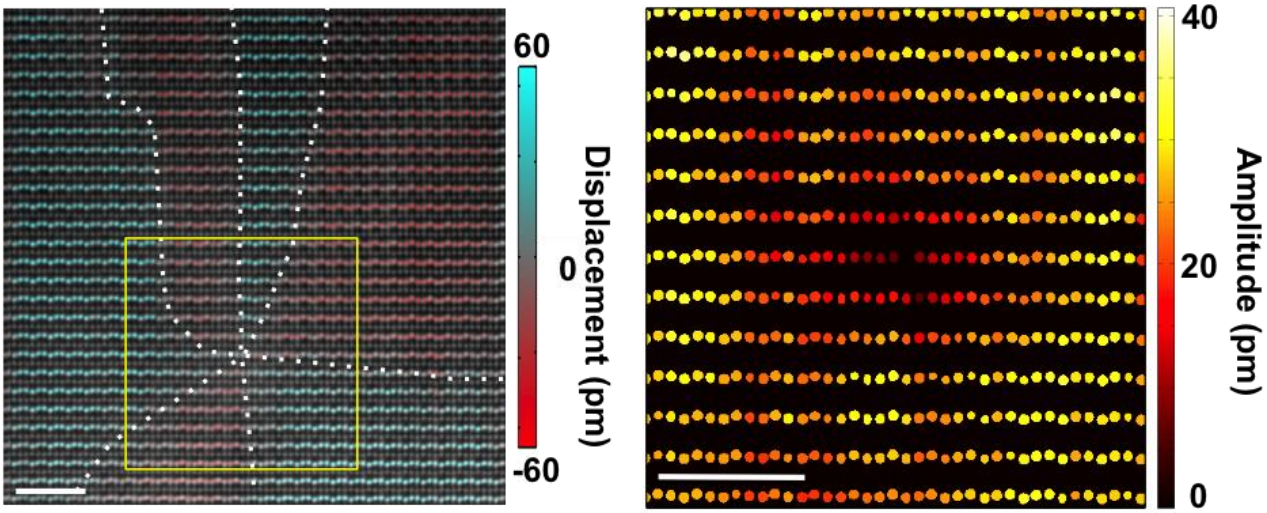

Figure 2. (a) STEM image of Zr-doped $ErMnO_3$ with a color overlay corresponding to the Er displacement, with turquoise indicating a positive polarization and red indicating negative. A vortex center is observed where 6 domains meet. (b) Amplitude of the ferroelectric displacement of the Er atoms shown in the yellow box in (a), showing vanishing amplitudes near the vortex center. Scale: 2nm.

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