Microsc. Microanal. 21 (Suppl 3), 2015
doi:10.1017/S1431927615007308 Paper No. 0651

Imaging Local Polarization and Domain Boundaries in Multiferroic $(LuFeO_3)_m/(LuFe_2O_4)_n$ Superlattices

Megan E. Holtz[1], Julia A. Mundy[1], Jarrett A. Moyer[2], Charles M. Brooks[3], Hena Das[1], Alejandro F. Rebola[1], Robert Hovden[1], Craig J. Fennie[1], Peter Schiffer[2], Darrell G. Schlom[3,4], David A. Muller[1,4]

[1.] School of Applied and Engineering Physics, Cornell University, Ithaca, NY, USA
[2.] Department of Physics and Materials Research Institute, University of Illinois at Urbana-Champaign, Urbana, IL, USA
[3.] Department of Materials Science and Engineering, Cornell University, Ithaca, NY, USA
[4.] Kavli Institute at Cornell for Nanoscale Science, Ithaca, NY, USA

Materials that couple strong ferroelectric and ferromagnetic order hold tremendous promise for next-generation memory devices. However, many so-called multiferroics have properties that are either weak, emerge well below room temperature, and/or lack coupling between the electric and magnetic domains, stymieing technological applications. The atomic-scale design of new multiferroics, usually realized as heterostructures or interface phases, requires a local probe of physical properties and structure inside the material. This atomic-scale feedback on ferroelectric polarization and domain structure has helped lead us to a new strong ferrimagnet-ferroelectric with the highest known simultaneous transition temperatures. These $(LuFeO_3)_m(LuFe_2O_4)_n$ superlattices (Fig 1) are constructed by integrating the ferroelectric, antiferromagnetic $LuFeO_3$ and paraelectric, ferrimagnetic $LuFe_2O_4$. The hexagonal $LuFeO_3$ is an improper ferroelectric where the Lu-O buckles into a polar structure [1]. We quantify the polar structure—manifest as a displacement of the lutetium atoms—with atomic precision for different superlattice layerings. Our ferroelectric domain measurements show large polarization and regular domain walls correlate with improved magnetic moment and critical temperature, T_C.

Aberration-corrected scanning transmission electron microscopy (STEM) can provide atomic column positions with sub-picometer precision [2]. Here we apply atomic mapping with picometer precision to find local polarization and domain structure in $(LuFeO_3)_m(LuFe_2O_4)_n$ as a function of m,n in a 100 keV NION Ultra-STEM. Each image was formed from many (>20) cross-correlated fast acquisitions. 2D Gaussian fitting located the center of the lutetium columns, from which the displacement, and thereby polarization, was measured (Fig 2ab). After analyzing over 100 images, or > 4000 nm^2 of the material, we find that the $(LuFeO_3)_m(LuFe_2O_4)_1$ superlattices with $m \geq 2$ display a ferroelectric structure. The polarization is damped for lutetium rows adjacent to the $LuFe_2O_4$ layers for $2 \leq m \leq 5$, thus we would expect net polarization to grow with layer width. Indeed, the polarization of the parent-compound $LuFeO_3$ is reached for $m \geq 3$ multilayers and surprisingly is exceeded for m = 7,8 (Fig 2c).

We collected domain sizes and boundary type statistics from the local polarization data. We found that domain size grows with m, increasing the width in the growth plane but limited in height by the $LuFe_2O_4$ layers, for up to m=7. At m=7, the domain height stabilizes at half the height of the $(LuFeO_3)_7$ layer, and forms regular domains with tail-to-tail walls at the $LuFe_2O_4$ layer and head-to-head walls in the middle of the $(LuFeO_3)_7$ layer. First-principle calculations suggest that the doping associated with this particular domain configuration leads to the increase in magnetic moment, which we have observed (Fig 3). T_C is also seen to increase, approaching room temperature. STEM analysis combined with first-principles results suggests pursuing strongly ferroelectric domains with regular domain boundaries will lead to enhanced magnetization and T_C in these multiferroics. [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).

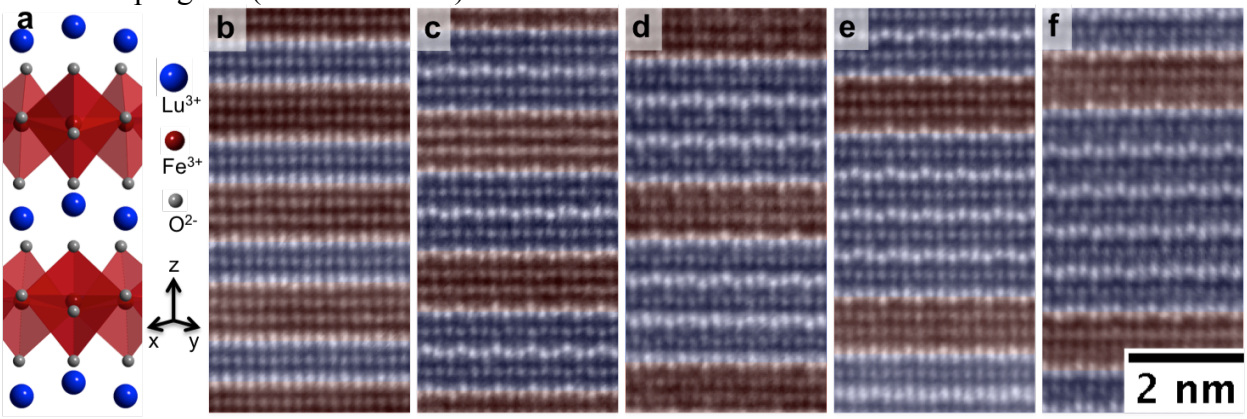

Figure 1. Diagram of the Lu-O buckling in $LuFeO_3$ (a), which leads to the ferroelectric structure [1]. ADF-STEM of the $(LuFeO_3)_m$ $(LuFe_2O_4)_n$ viewed along the [110] zone axis, the $(LuFeO_3)_n$ blocks highlighted in blue, for the *m, n* = 1, 1 structure (b), 2, 1 (c), 3, 1 (d), 4, 1 (e) and 5, 1 (f).

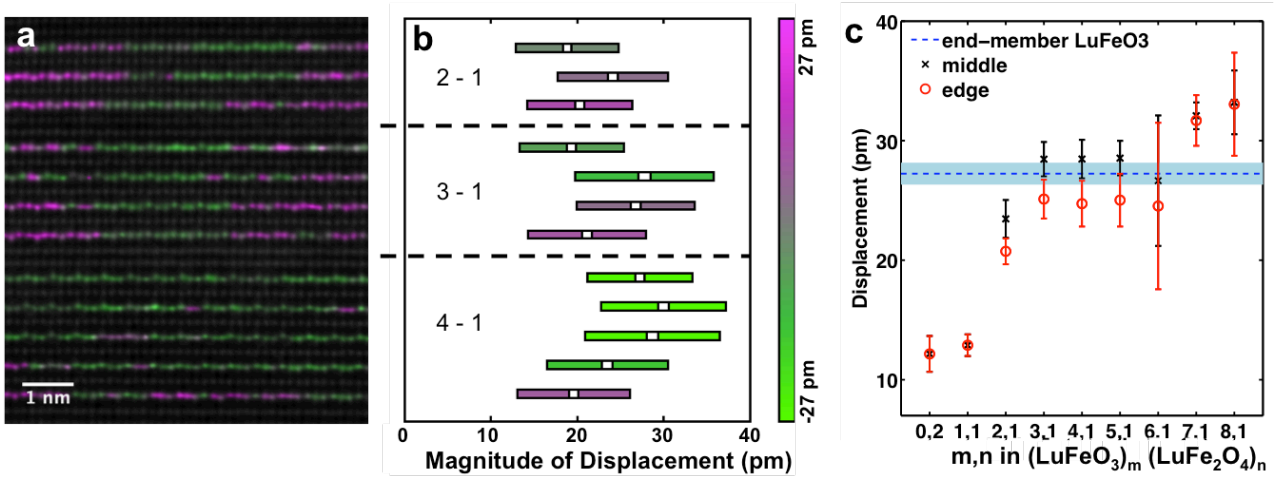

Figure 2. (a) STEM image of $(LuFeO_3)_m$ $(LuFe_2O_4)_n$ with color overlays for the magnitude and direction of the local ferroelectric displacements. (b) r.m.s. displacement (square) for each lutetium row. Color indicates mean polarization, with lengths corresponding to 20-80% of the distribution. (c) r.m.s displacement averaged over many images for varying (m,n), distinguishing between layers on the edge bordering the double iron layer and in the middle. Blue line is for $LuFeO_3$.

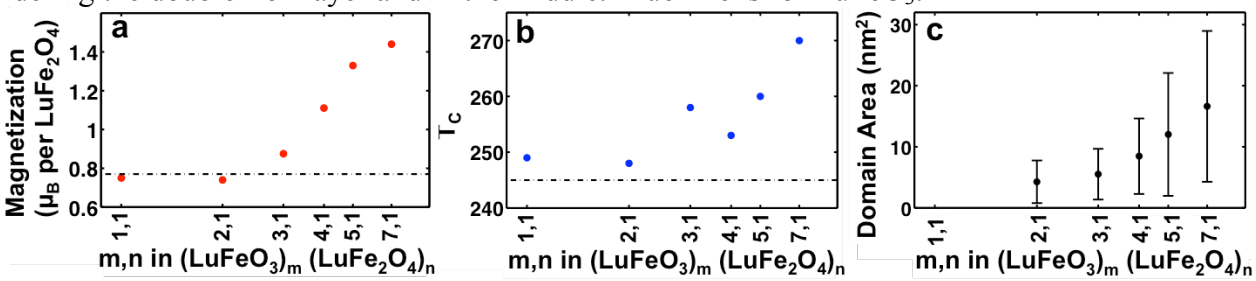

Figure 3. Magnetization and critical temperature for the $(LuFeO_3)_m$ $(LuFe_2O_4)_n$, plotted alongside ferroelectric domain area. Larger domain area correlates to higher magnetization.