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Dielectric breakdown along c-axis boundaries in magnetoelectric Cr_2O_3 for spintronic devices

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Voltage controlled boundary magnetism is crucial for spintronic devices with reduced power consumption. Magnetoelectric and antiferromagnetic Cr_2O_3 is an ideal material due to its electric field switching of nonvolatile boundary magnetism [1]. The boundary magnetization can switch an adjacent soft ferromagnetic layer via voltage-controlled exchange bias. Bulk Cr_2O_3 has a high dielectric breakdown field of 1000 $KVmm^{-1}$ and a large bandgap of 3.4 eV. However, even with a small electrode size of 0.04 mm^2 and thick Cr_2O_3 film of 0.5 μm, the highest reported dielectric breakdown field of Cr_2O_3 films is only 200 $KVmm^{-1}$ [2]. The breakdown field drops rapidly to 8 $KVmm^{-1}$ if electrode size increases to 35 mm^2 and film thickness decreases to 250 nm [2], making electric field induced switching of Cr_2O_3 based heterostructures very difficult. Here, we combine aberration corrected STEM characterization and spin polarized density functional theory (DFT) calculations to elucidate the structure, electronic properties, and magnetic properties of a new type of interface-stabilized planar crystallographic defect in Cr_2O_3 thin films that explains the structural origin of dielectric breakdown.

Figure 1(a) is a low-angle annular dark field (LAADF, 35-150 mrad) STEM image of a Cr_2O_3 film on Al_2O_3 substrate. LAADF is sensitive to strain fields [3], so the image emphasizes the columnar boundaries along the c-axis. The inset in Figure 1(a) is a high-angle ADF (HAADF, 84-165 mrad) STEM image along one of the boundaries. The HAADF signal scales as Z^a, so this image is dominated by Cr sites, and the oxygen atoms are invisible. Figure 1(b) is a plan view HAADF STEM (54-160 mrad) image. This intermediate range of scattering angles was used to mix sensitivity to both boundary strain field and Z-contrast of Cr sites on the boundary, so along the plan view axis the boundaries are imaged edge-on as narrow bright lines superimposed on the Cr_2O_3 lattice. The inset in Figure 1(b) shows the magnified boundary area with a boundary model constructed based on the plan view and cross section STEM images. Cr has an ABC staking order along the c-axis and is marked by solid symbols with a different color for each layer. The boundary structure can be described as a 60˚ rotation about the c-axis, plus a shift of $^1/_3[10\bar{1}0]$ parallel to the basal plan to match the Cr plan view symmetry. The boundary model matches with the DFT calculation of the Cr_2O_3/Al_2O_3 interface defect grains. Figure 1(c) is the relaxed boundary model from spin polarized DFT calculations. A multi-slice STEM simulation from the model is superimposed on the inset of Figure 1(a). In DFT, the boundary model shows continuous boundary spins that do not interrupt the surface magnetization utilized for exchange bias switching in devices.

Figure 2(a) is O K edge extracted from EEL spectrum images on the boundary. It shows an O pre edge feature that only exists only locally on the boundary. Multiple scattering EELS simulation from the DFT model with FEFF9.6 reproduces the pre-edge. As the O-K edge reflects the portion of the conduction band projected onto O atoms, the additional peak is an indication of a reduced bandgap on the boundary arising from O unoccupied 2p states. The reduced bandgap energy is also confirmed by density of states

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calculations, as shown in Figure 2(b). Breakdown voltage is roughly proportional to bandgap energy raised to a characteristic power, so, the c-axis boundaries with reduced bandgap energy in the Cr_2O_3/Al_2O_3 thin film systems are likely to create local easy breakdown paths.

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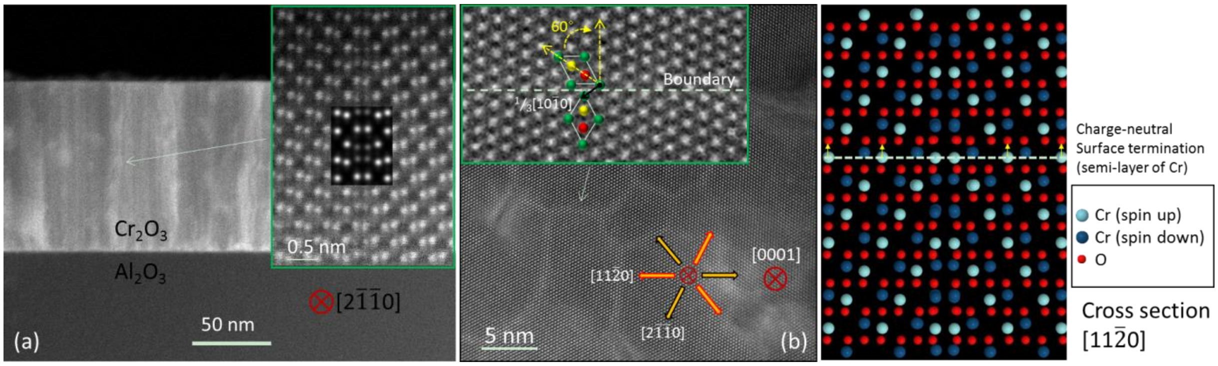

Figure 1. (a) Cross-section LAADF STEM image of Cr_2O_3/Al_2O_3 showing vertically aligned grain boundaries. The inset is HAADF STEM image of one of the boundaries, with multi-slice STEM simulation superimposed. (b) Plan-view HAADF STEM image. Inset is the magnified boundary image with structure model superimposed. Different color of balls represent the ABC ordered staking layer of Cr along the c-axis. (c) Relaxed boundary model from spin polarized DFT calculations. The white dashed line is one of the stable charge-neutral surface terminations, showing continuous spin across the boundary.

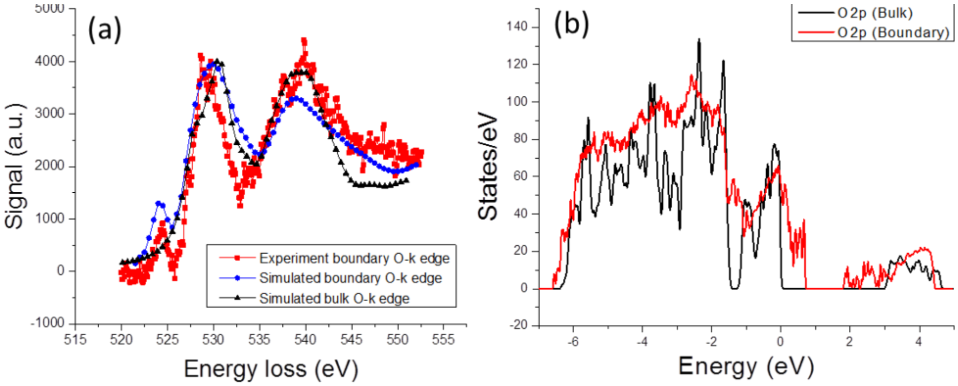

Figure 2. (a) Boundary O K edge showing a pre-edge and multiple scattering EELS simulations. (b) Density of states calculations showing that the pre-edge is inside the bandgap, reducing bandgap energy on the boundaries.

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