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Growth and Structural Characterization of Multiferroic Thin Films and Nanopatterns

S. Xie, * Z. Pan, * J. Cheng, * G.E. Sterbinsky, * B.W. Wessels, * and V.P. Dravid*
* Department of Materials Science and Engineering, Northwestern University, 2200 Campus Drive, Evanston, Illinois 60208-3108

Multiferroic materials that couple ferroelectric order with ferromagnetic order have attracted increasing interest in recent years due to their potential applications in devices such as filters, oscillators, magnetic field sensors [1]. However, compounds that co-exist ferromagnetism and ferroelectricity are limited [2]. The availability of high-quality ferroelectric/magnetic multilayer thin films with precisely controlled composition, atomic arrangements and interfaces is a prerequisite for their incorporation into practical devices [3]. The integration of magnetoelectric heterostructures into nanodevice architectures requires the formation of patterned microstructures with feature sizes on the nanometer order.

We have grown multiple promising magnetoelectric systems such as $CoFe_2O_4/SrTiO_3$, $Fe_3O_4/SrTiO_3$ and $Fe_3O_4/BaTiO_3$ thin films by molecular beam epitaxy. The interfacial structure and chemistry of epitaxial $CoFe_2O_4$ and Fe_3O_4 thin films on (001) $SrTiO_3$ and $BaTiO_3$ substrates were investigated using various advanced S/TEM techniques such as high-resolution TEM (HRTEM), and atomic-resolution high angle annular dark field (HAADF) imaging combined with electron energy loss spectra (EELS) analyses at a sub-nanometer scale. The results show that the $CoFe_2O_4/SrTiO_3$, $Fe_3O_4/SrTiO_3$ and $Fe_3O_4/BaTiO_3$ interfaces are semi-coherent with requisite dislocation density to accommodate lattice mismatch. EELS analyses indicate that Fe valence is nominally +3, with no measurable change across the interfaces. The representative HAADF and EELS of $CoFe_2O_4/SrTiO_3$ (CFO/STO) system are shown in Fig. 1.

The same interfaces were fabricated in nanoscale patterned structures in order to investigate fundamental questions such as the pattern edge, shape and geometry effects, compared to their thin film counterparts. Ferromagnetic $CoFe_2O_4$ and ferroelectric $BaTiO_3$ nanopatterns were fabricated on MgO and $SrTiO_3$ substrates, respectively, by soft electron beam lithography (soft-eBL) approach developed in our group. The microstructural characterization reveals that $CoFe_2O_4$ and $BaTiO_3$ are epitaxially grown on MgO and $SrTiO_3$ substrates, with a simple cube-on-cube orientation relationship. Crystal orientation, shape development, and edge definition during fabrication are able to be directly controlled by adjusting the patterning parameters such as resist thickness and pattern directions with respect to underlying substrate. An example for $CoFe_2O_4$/MgO (CFO/MgO) system is shown in Fig. 2 [4]. The presentation will cover the comparison and differences in interfacial phenomena in nanopatterned structures compared to corresponding thin film forms.

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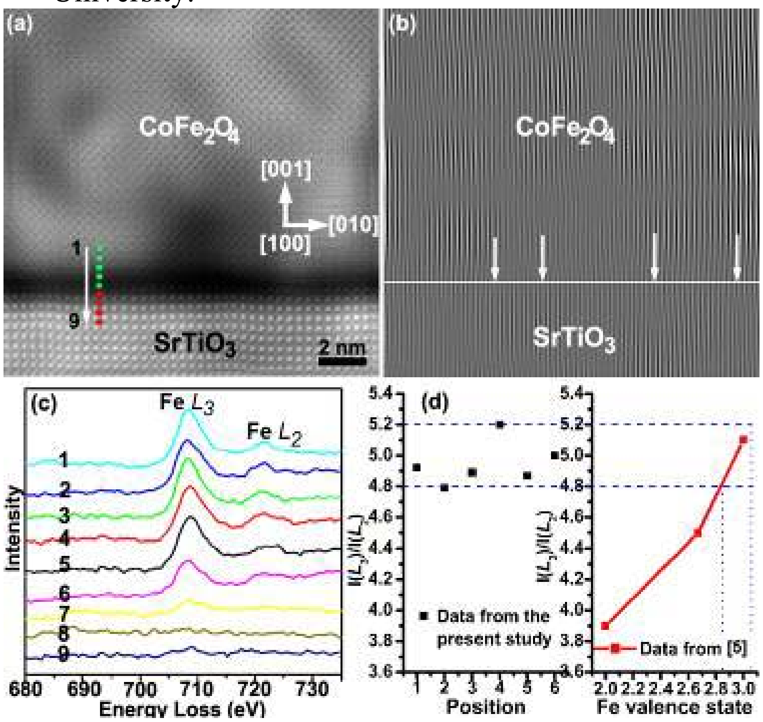

FIG. 1. (a) Atomic-scale HAADF image showing the epitaxial growth of CFO on STO. (b) The filtered image of (a) using (020) reflections showing the presence of interfacial mismatch dislocations. (c) Fe-L edges EELS profiles across the CFO/STO interface [denoted by solid circles in (a)]. (d) Intensity ratio I(L_3)/I(L_2) of Fe EELS profiles from position 1 to 6 and intensity ratio vs. Fe oxidation state relation reveal that Fe across the interface is primary of +3.

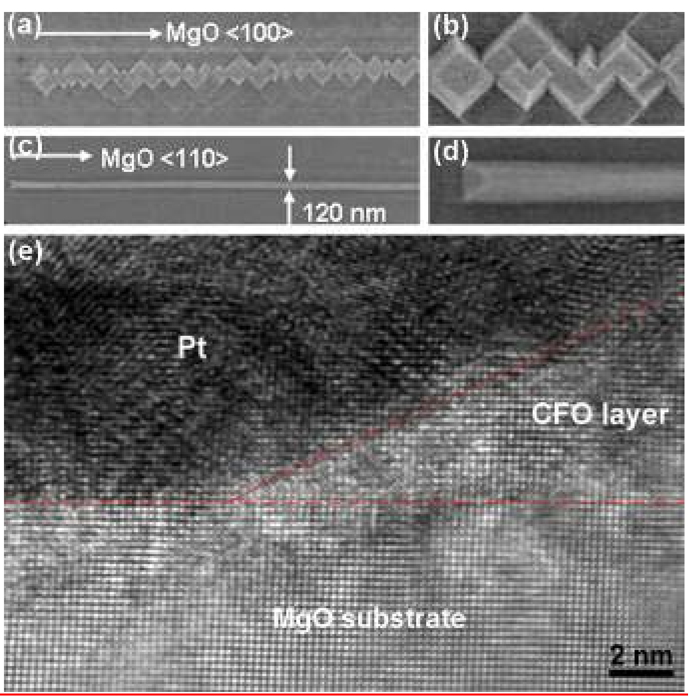

FIG. 2. (a) ESEM image of CFO line patterned along (100) MgO. (b) shows detail of the Zig-zag edge. (c) ESEM images of CFO line patterned along (110) MgO. (d) Image of one end of the line showing the faceting detail. (e) HRTEM image showing the epitaxial growth of CFO on MgO. The Pt in the right-up corner is introduced during TEM sample preparation using FIB.

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