Evidence of Submicron-Scale Phase Separation in Pr-doped LCMO film

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A key feature of the CMR manganite family of materials is their tendency towards multiphase coexistence: they spontaneously form domains (of size 0.05-5 μ m) characterized by differing degrees of charge and magnetic order. The origin and effect on other properties of these domain structures is a fundamental issue in the materials science of these compounds. In this work, we show that both charge-order and magnetic domain structures can be detected by electric force and magnetic force microscopy operated at low temperatures, used to image 800 Å thick films of (La_{0.6}Pr_{0.4})_{0.67}Ca_{0.33}MnO₃ on LaAlO₃(100) prepared using PLD deposition.

Electrostatic force microscopy (EFM) can detect local variation in charge distributions at sample surface. Using phase detection technique, the contrast is given by

$$\Delta \boldsymbol{f} \propto \frac{\partial F_e}{\partial z},$$

where f is the oscillation phase of the sensing cantilever. In the case of conducting surfaces, the force is produced by image charges and is always attractive, i.e., the contrast is always dark. In the case of insulators with trapped charges, the force is given by $\vec{F} = \int \mathbf{r} \vec{E} dv$

and could be either attractive or repulsive depending on the polarity of the trapped charges. Thus, one can readily establish that the electrostatic contrast is due to trapped charges by observing a contrast reversal with opposite tip voltage polarity. Magnetic force microscopy operates in a similar manner, except that the contrast variation in proportional to the strength of the local magnetic poles on the surface.

Figure 1 shows high resolution $2x2 \ \mu m^2$ scans of the sample, obtained at 40 degrees below the Tc =187 K with the tip polarity reversed. The images on the left are the topography of the film and to the right are the corresponding the EFM of that surface. A striking result is the observation of bright and dark patches on the EFM images, and whose contrast (in some areas) reverses with tip polarity. These domains appear to be elongated strips or labyrinths of about 500 nm wide, and completely disappear above Tc. These observations are consistent with the schematic prediction based on electron microscopy measurements of Uehara *et al.*[1].



Figure 1. EFM images of the Pr-doped LCMO film on the LAO substrate. (a) and (c) show the topography of the same area of the sample surface, (b) and (d) are the corresponding electric structures. The sample temperature is 145 K, which is below Tc=187 K. (a)/(b) are $2\times2 \ \mu\text{m}^2$ images with V=+0.5 and (c)/(d) are with V=-0.8.



Figure 2. MFM images of the Pr-doped LCMO film on the LAO substrate. Left panels show the topography. Right panels are the corresponding magnetic tructures. The magnetic features are absent above Tc (top), and persist at 150K, below Tc (bottom). These are $10x10 \ \mu\text{m}^2$ images.

The magnetic force image of the same sample is shown in Figure 2. The magnetic features emerge when the sample goes below the transition temperature. The dark and bright contrast can be regarded as the magnetic poles. With close inspection and recalling that magnetic poles occur in pairs, one can discern that the low temperature surface is comprised of isolated ferromagnetic grains in a matrix of non-magnetic materials.

Having both the EFM and MFM images of the surface, we can compare the positions of the charge ordered and magnetic grains. More importantly, we can elucidate the theory that predicts that the change ordered regions are non-magnetic, while the conducting regions are magnetic. Figure 3 shows an analysis of the EFM images of opposite polarities. The images of opposite polarities were digitally added to each other. Consequently, regions whose polarities have reversed will show-up as gray in the composite image, while regions whose contrast was invariant will appear as very bright or very dark. Clearly, these polarity invariant regions, which we associate with the conducting areas of the surface, form isolated patches whose diameters vary from a fraction of a micron to several microns.



Figure 3. An analysis of the EFM images with opposite polarities. This image is formed by adding the EFM images of the same area. The bright and dark regions are areas with no contrast reversal and are associated with conducting regions.



Figure 4. High resolution MFM image of the sample at a low temperature, showing the isolated ferromagnetic grains.

It is interesting to compare this image with Figure 4, which directly show the ferromagnetic grains. We find qualitative agreement in the size and distribution of the conducting grains measured using EFM and the ferromagnetic grains measured using MFM. That the grains are somewhat larger in MFM is due to the fact that the magnetic images taken at some distance is larger than the source.

These results, while preliminary, conclusively demonstate the delineation of the phase separation into ferromagnetic and charge ordered components. That the charge ordering can be detected with EMF imaging and be directly correlated to magnetic structures is an intriguing new discovery; it opens up a whole range of experiments including comparison with MFM and NSOM experiments and study of effects of applied fields and currents.

References

[1] M. Uehara, S. Mori, C. H. Chen, and S.-W. Cheong, *Nature* 399,560 (1999)