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Atomic-Scale Chemical Imaging of Interdiffusion and Defects in $(La_{0.7}Sr_{0.3}MnO_3)_5/(SrTiO_3)_5$ Multilayers by Aberration Corrected Microscopy

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In electron microscopy it is often difficult to distinguish between true intermixing and the apparent broadening of an interface due to the shape of the electron probe or its tails. However, with recent advances in aberration correction, full 2D spectroscopic images (SI) can now be recorded at atomic resolution in under a minute[1], and the additional chemical information can be used to directly detect chemical intermixing and its impact on electronic structure of an interface. Using spectroscopic imaging, performed on a 5^{th} order aberration corrected NION UltraSTEM 100, we show here that the degradation of the magnetic properties of $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3$ (LSMO/STO) multilayers correlates with a higher degree of intermixing at the interfaces and the presence of extended defects in the $La_{0.7}Sr_{0.3}MnO_3$ layers.

During the growth of LSMO/STO multilayers by pulsed laser deposition, the micro-structure not only depends on the careful choice of growth temperature and oxygen partial pressure, but also on the laser spot size, which can influence the cation stoichiometry and energetics of the plume. For comparison, Fig. 1 shows SIs of two manganite/ titanate multilayers grown at a laser spot size of 7.5 and 1.6×10^{-2} cm^2, respectively. La elemental maps (a, c) as well as red-green false color images (b, d), obtained by combining the Ti and Mn maps show a different degree of intermixing. The multilayer grown with a smaller laser spot size shows less abrupt interfaces and an extended defect, marked by a white arrow in Fig. 1(d).

From the 2D Ti/Mn map shown in Fig. 2(d) enhanced intermixing can be directly inferred due to the observed variation of the Ti concentration in the LSMO layer showing weak Ti sublattice in some areas of the LSMO layers (probe tails would only cause a broadening of the interfaces). Note that sample drift (~1.0Å/min) during the acquisition causes the lattice to appear distorted.

Extended defects found in the multilayer grown at a laser spot size of 1.6×10^{-2} cm^2 were further studied using spectroscopic imaging. One type of defect, which has formed in the upper part of the LSMO layer, is shown in Fig. 2. Because of the character of the Sr EELS edges, it is more difficult to obtain a strontium elemental map at the same acquisition time as the Ti, Mn and La maps; however, information about the position of the Sr atoms is contained in the simultaneously recorded ADF image. Here, the La map (Fig. 2 (c)) shows missing La columns at the center of the defect, but from the ADF image these sites are clearly occupied by some other atomic species. Combined with the information from the Mn and Ti elemental maps we can conclude that Sr atoms fill the positions of the La sites, which suggests local off-stoichiometry. [2]

[1] D. A. Muller, L. F. Kourkoutis, M. Murfitt, J. H. Song, H. Y. Hwang, J. Silcox, N. Dellby, O.

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[2] The authors gratefully acknowledge technical support by Niklas Dellby (Nion Co.). LFK and DM supported by the ONR EMMA MURI and the Cornell Center for Materials Research, an NSF MRSEC. JHS, HYH supported by a Grant-in-Aid for Scientific Research on Priority Areas.

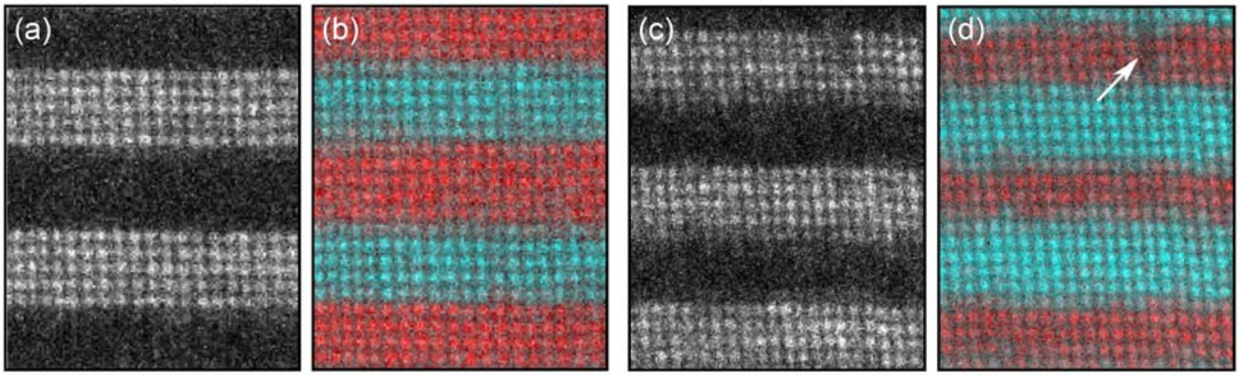

FIG. 1. Spectroscopic-imaging of $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3$ multilayers grown at a laser spot size of (a, b) 7.5 and (c, d) $1.6x10^{-2}$ cm^2. (a, c) La elemental maps and (b, d) red-green false color images, obtained by combining the Ti and Mn maps extracted from the spectrum images. The multilayer grown with a smaller laser spot size shows less abrupt interfaces and an extended defect, marked by a white arrow in (d). The growth direction is from bottom to top.

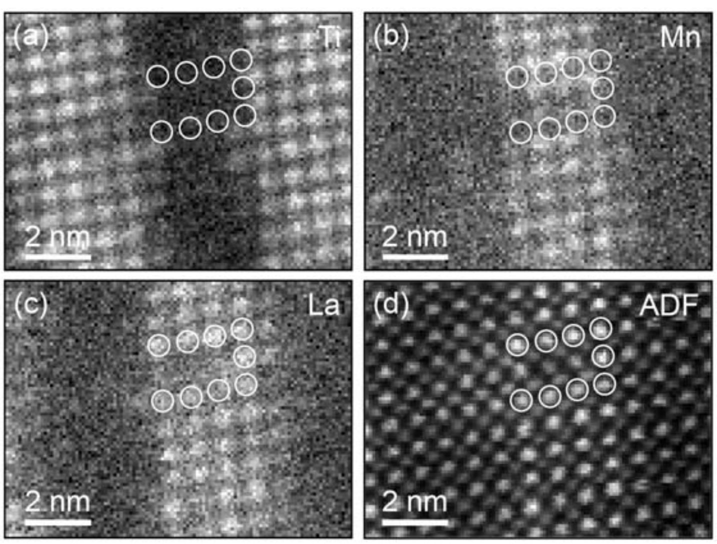

FIG. 2. Spectroscopic-imaging of an extended defect in a $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3$ multilayer grown at a laser spot size of $1.6x10^{-2}$ cm^2. (a) Ti, (b) Mn, (c) La elemental maps extracted from an 109x80 pixel spectrum image and (d) the simultaneously recorded ADF image. The white open circles indicate the position of the La columns around the defect. The ADF image, which also tracks the position of the Sr atoms, shows clear atomic columns in the center of the defect, while the La concentration is low, suggesting that the defect was formed to accommodate for access Sr in the LSMO layer.

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