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Linking Dopant Distribution and Interatomic Distortions at $La_{1.6}M_{0.4}CuO_4/La_2CuO_4$ Superconducting Interfaces

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The crystal structures of complex oxide materials are highly adaptable to changes in composition and this compatibility provides the opportunity of synthesizing them in different forms, such as ultrathin films and/or heterostructures. Oxide heterostructures exhibit remarkable electronic properties which do not exist in bulk forms and the emergent phenomena occurring at the interfaces are impressive [1]. One of the exciting interface effect is high temperature interfacial superconductivity discovered in heterostructures consisting of an insulating La_2CuO_4 (LCO) and metallic $La_{1.55}Sr_{0.45}CuO_4$ (LSCO) layers, while neither of these layers is superconducting [2].

Using the unique capabilities of atomic-layer-by-layer oxide molecular-beam epitaxy (ALL-oxide MBE), we have synthesized bi-layers consisting of 3 unit cells of metallic $La_{1.6}M_{0.4}CuO_4$ layer and 3 unit cells of the undoped insulating (I) LCO layer neither of which is superconducting by its own, where M represents the dopant, namely, Sr^{2+}, Ca^{2+}, and Ba^{2+} which hold the same ionic charge but differ in atomic radii. The M-I bilayers were superconducting with a critical transition temperature of ~17 K, ~37 K, and ~38.5 K for M = Ca, Sr, and Ba, respectively.

In the present work, we report investigations on the local chemistry and crystal structure including oxygen positions across the interfaces of cuprate bi-layers using high-resolution analytical scanning transmission electron microscopy (STEM) techniques. For the analyses, a JEOL JEM-ARM200F STEM equipped with a cold field-emission electron source, a probe C_s-corrector (DCOR, CEOS GmbH), a large solid-angle JEOL Centurio SDD-type energy-dispersive X-ray spectroscopy (EDXS) detector, and a Gatan GIF Quantum ERS spectrometer was used. STEM imaging and electron energy-loss spectroscopy (EELS) were performed at probe semi-convergence angles of 20 mrad and 28 mrad, respectively. Collection angles for high-angle annular dark-field (HAADF) and annular bright-field (ABF) images were 75-310 mrad and 11-23 mrad, respectively.

HAADF images reveal high structural quality and perfect coherent interfaces (Fig. 1). In order to determine the elemental distribution across the interfaces, atomic resolution EELS and EDXS analyses were conducted. EDXS analyses and EEL spectrum images (SI) reveal different cation redistribution behaviors and lengths depending on the cation (i.e. La-dopant) size mismatch. Figure 2 shows the atomic arrangement at the interfaces of the Sr-doped bilayer sample, which shows a relatively homogeneous distribution of Sr.

Atomically resolved HAADF and ABF images were acquired simultaneously. Since ABF images provides oxygen positions, interatomic distortions (cation – cation and CuO_6 octahedra) were measured and correlated with the dopant distributions obtained via chemical analyses. In Fig. 3a two of the quantitatively studied images (i.e. HAADF and ABF), are presented as an overlay and the arrows indicate the nominal interfaces. Figure 3b shows the measured interatomic distances which are indicated with arrows. Results will be presented for the different dopants and correlations with high-temperature interfacial superconductivity will be discussed.

Since high temperature interface superconductivity in cuprates is sensitive to local chemistry and structural distortions, these results assist the basis for understanding the origin of cation size effects on the structure and superconductivity of doped LCO bi-layers.

References:

[1] Hwang, H. Y. et al., Nature Materials, **11** (2012) p.103.
[2] Gozar, A., et al., Nature, **455** (2008) p.782.
[3] The research leading to these results has received funding from the European Union Seventh Framework Programme [FP7/2007-2013] under grant agreement n°312483 (ESTEEM2).

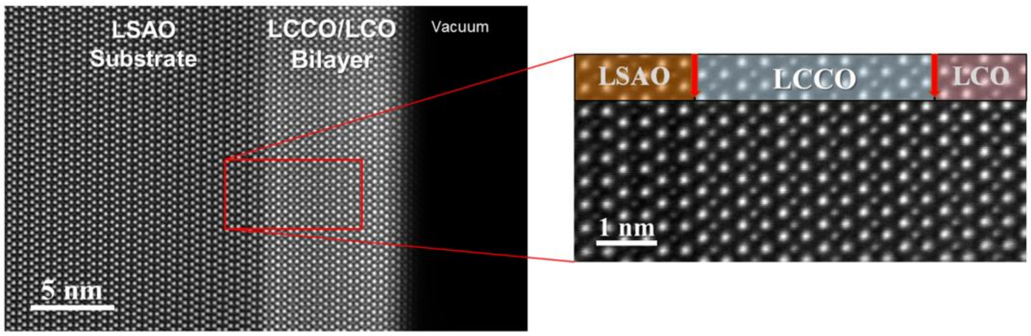

Figure 1. Low-magnification STEM-HAADF image of Ca-doped bi-layer showing a coherent interface and high structural quality.

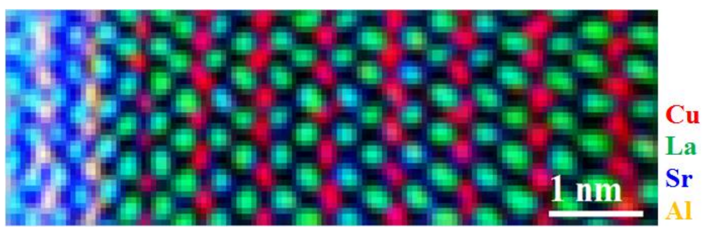

Figure 2. Atomically resolved EEL SI using showing the elemental distribution. (Red: Cu, Green: La, Blue: Sr, and Yellow: Al).

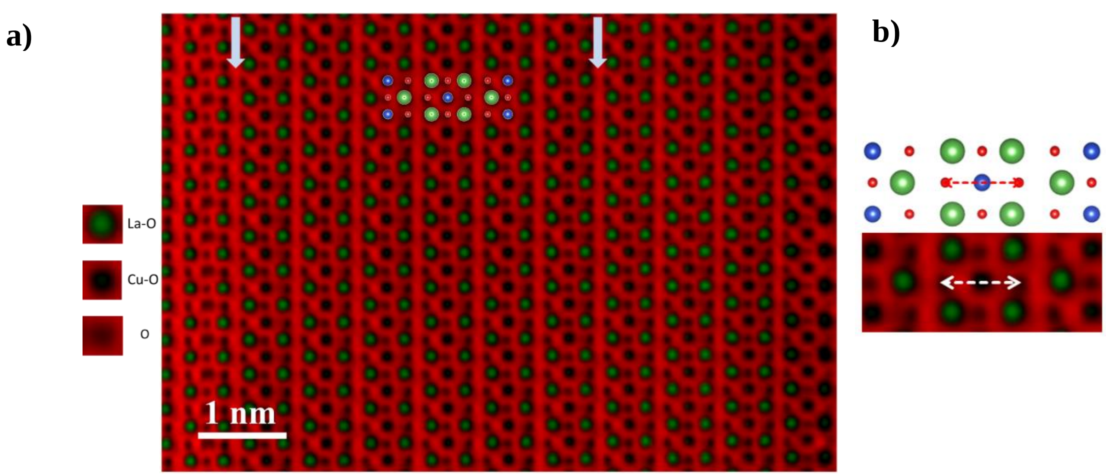

Figure 3. a) ABF-HAADF hybrid image of the Ba-doped bi-layer showing all atom positions in one image and in b) the measured interatomic distances.

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