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# Direct Observation of Asymmetric Sr Diffusion in Sr- δ-Doped $La_2CuO_4$

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Superconductivity in copper oxides arises when a parent insulator compound is doped beyond some critical concentration [1]. In the case of $La_2CuO_4$ (LCO), high-$T_c$ superconductivity is obtained either by substituting $La^{3+}$ with $Sr^{2+}$ or by inserting interstitial $O^{2-}$ [2]. Latest advance of the unique capabilities of layer-by-layer oxide molecular beam epitaxy (MBE) allows controlling the structural composition down to the single atomic layer. Recently, we have fabricated Sr-δ-doped $La_2CuO_4$ multilayered structures in which some atomic layers of LaO have been fully substituted by SrO layers. By varying the spacing between the LCO and SrO layers high-$T_c$ superconductivity (~ 40 K) has been obtained.

For the present contribution, the structure and cation redistribution in the Sr-δ-doped LCO multilayers on $LaSrAlO_4$ (LSAO) substrate was investigated using a JEOL ARM 200CF scanning transmission electron microscope (STEM) equipped with a cold field-emission electron source, a probe corrector, a large solid angle SDD-type EDX detector, and a Gatan GIF Quantum ERS spectrometer. The microscope was operated at 200 kV with a probe semi-convergence angle of 30 mrad. Collection angles for high-angle annular dark-field (HAADF) and annular bright-field (ABF) images were 90 - 370 mrad and 11-23 mrad, respectively.

LCO exhibits an orthorhombic structure with lattice parameters $a$ = 0.5335 nm, $b$ = 0.5415 nm, and $c$ = 1.3117 nm. Neglecting the small orthorhombic distortion of the LCO lattice, it can be regarded as pseudo-tetragonal with lattice parameters of $a$ = 0.3801 nm and $c$ = 1.3117 nm (Fig. 1a). LSAO has a tetragonal structure with lattice parameters $a$ = 0.3754 nm and $c$ = 1.2635 nm (Fig. 1b). The HAADF and ABF images in Figure 1c and 1d show the orientation relationship between LCO and LSAO. Figure 2a represents a typical cross-sectional HAADF image showing no structural defects. Atomically resolved HAADF and ABF images of the Sr-δ-doped region, which were simultaneously acquired, are presented in Figure 2b and 2c.

A detailed study of the Sr redistribution at the interface was performed by atomic resolution HAADF in combination with EDX and EELS, as shown in Figure 3a-c. Due to the difference in atomic number ($Z_{Sr}$ = 38, $Z_{La}$ = 57), the atomic columns dominated either by La or Sr give rise to different contrast in the HAADF image (Fig. 3a). In the Sr-δ-doped region the atomic column intensity is significantly lower than in pure LCO. An average image intensity profile along the growth direction is shown in Figure 3b, where for the Sr-δ-doped region the image intensity exhibits a relatively sharp intensity drop followed by a slowly increasing intensity pointing at an asymmetric distribution of Sr along the growth direction. The Sr-L EDX and Sr-$L_{2,3}$ EELS (insert in Fig. 3a) line-scan profiles provide a robust proof that the Sr concentration in LCO is (i) distributed across a few layers and (ii) has an asymmetric profile [3].

References:

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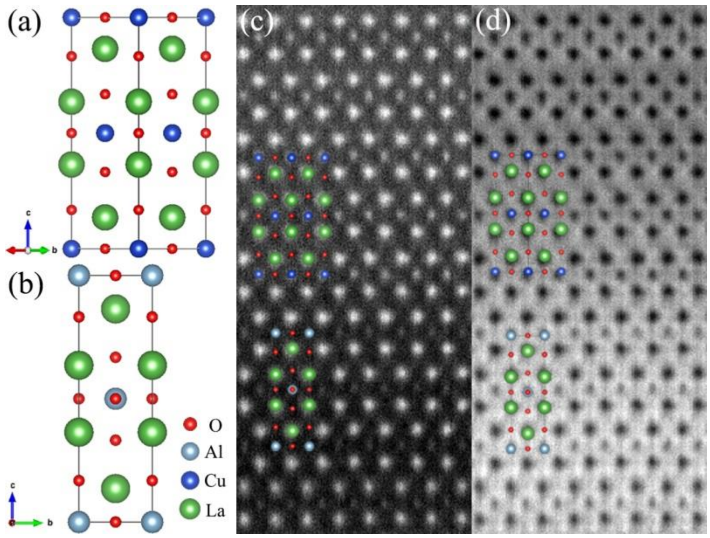


**Figure 1.** The projected crystal structure of (a) LCO and (b) LSAO. (c) Atomic-resolution HAADF and (d) ABF images showing the orientation relationship between LCO and LSAO.

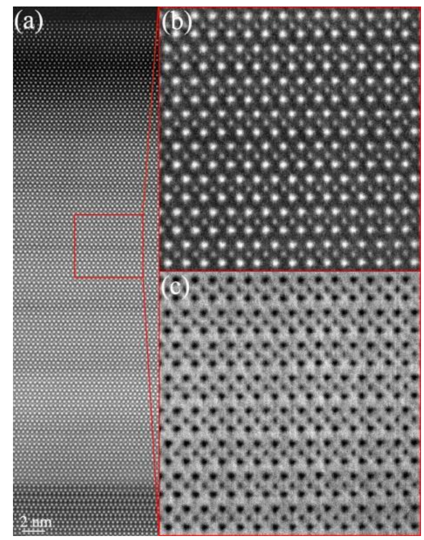


**Figure 2.** (a) HAADF image of nominally 1 ML Sr-δ-doped LCO on LSAO. Enlargement of simultaneously acquired (b) HAADF and (c) ABF images of the Sr-δ-doped area.

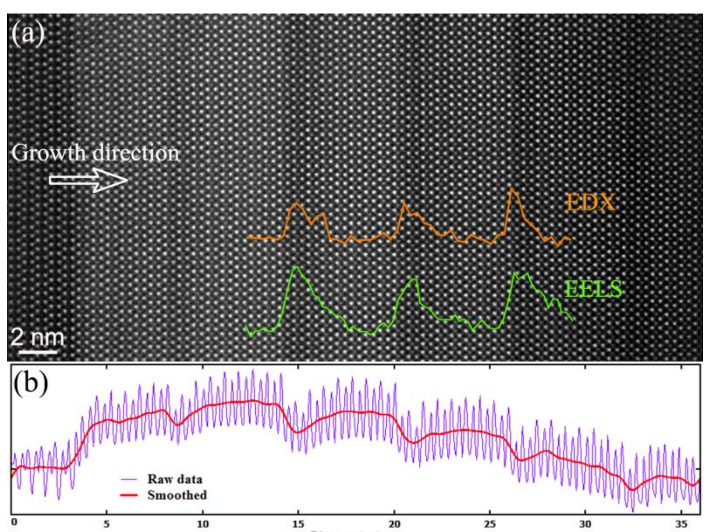


**Figure 3.** (a) HAADF image of Sr-δ-doped LCO. The LSAO substrate is located on the left side, the top of the multilayer on the right side. The inset shows the integrated Sr-L EDX and Sr-$L_{2,3}$ EELS line profiles. (b) HAADF image intensity profile along the growth direction integrated perpendicular to the growth direction.

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