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Atomic-Scale Quantitative and Analytical STEM Investigation of Sr-δ-Doped La_2CuO_4 Multilayers

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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]. At internal interfaces, the enhancement of the superconducting critical temperature is influenced by the interfacial structure [3]. Recently, by using atomic layer-by-layer oxide molecular beam epitaxy (MBE), we have fabricated Sr-δ-doped LCO multilayered structures on $LaSrAlO_4$ (LSAO) substrate, in which some atomic planes of LaO were intentionally substituted by SrO. By varying the spacing between the LCO and SrO layers high-T_c superconductivity (~ 40 *K*) was obtained [4]. Here we lay emphasis on the detailed and quantitative STEM analysis.

For the present contribution, we combine atomic-resolved quantitative STEM imaging with analytical STEM-EELS/EDX analysis to enhance understanding of high-T_c superconductivity at Sr-δ-doped LCO interfaces with respect to the local lattice and oxygen octahedral distortion, as well as cation and electron hole redistribution. STEM investigations were performed using a JEOL ARM 200CF scanning transmission electron microscope equipped with a cold field emission electron source, a D-COR probe corrector, a 100mm^2 Centurio EDX detector, and a Gatan GIF Quantum ERS spectrometer.

Figure 1 (a) shows a cross-sectional HAADF STEM image of Sr-δ-doped LCO multilayers, revealing that LCO and the LSAO substrate exhibit perfect epitaxy and show no local structural defects at the -δ-doped interfaces. 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. In the Sr-δ-doped region the atomic column intensity is significantly lower than in pure LCO. The average image intensity profile in growth direction shows that at the Sr-δ-doped region the image intensity has a relatively sharp drop of intensity followed by a slowly increasing intensity pointing to an asymmetric Sr distribution. Atomic-resolved HAADF and ABF images, which were simultaneously acquired at the Sr-δ-doped region, are presented in Fig. 1 (b) and (c). The local lattice and copper-apical-oxygen distortions were quantitatively evaluated by image analysis.

A detailed study on the redistribution of Sr and of electron holes at the interface was performed by a combination of atomic-resolved STEM-EELS/EDX. The Sr-*L* EDX and Sr-$L_{2,3}$ EELS (Fig.2 a) line-scan profiles show that Sr is redistributed a few layers in LCO and has an asymmetric concentration profile. The electron holes across the Sr-δ-doped interfaces were characterized by analysis of the O-*K* near-edge fine structure, as presented in Fig. 2 (b) and (c). These findings, suggesting a rather complex charge rearrangement mechanism, will be discussed. [5]

References:
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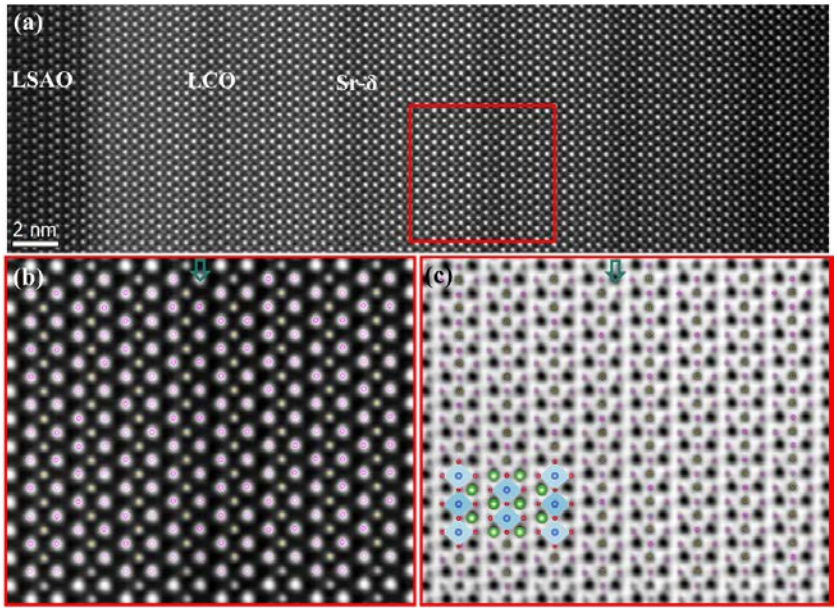

Figure 1. (a) HAADF STEM image of Sr-δ-doped LCO multilayers epitaxially grown on a LSAO substrate. Simultaneously acquired (b) HAADF and (c) ABF images of one Sr-δ-doped area, on which the atomic columns have been located for quantification of the lattice and copper-apical-oxygen distortions.

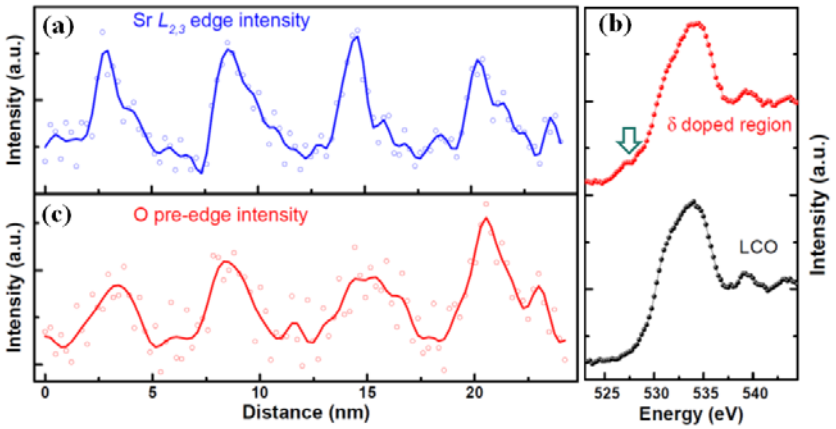

Figure 2. (a) Integrated Sr-$L_{2,3}$ EELS line profiles across 4 Sr-δ-doped regions. (b) O-*K* edge from a Sr-δ-doped region and from LCO. (c) Integrated O-*K* pre-edge intensity profile across 4 Sr-δ-doped regions.