Atomic-Scale Quantitative and Analytical STEM Investigation of Sr-δ-Doped La2CuO4 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 La2CuO4 (LCO), high-Tc superconductivity is obtained either by substituting La3+ with Sr2+ or by inserting interstitial O2− [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 LaSrAlO4 (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-Tc 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-Tc 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 100mm2 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 (ZSr=38, ZLa=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-L2,3 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-$\delta$-doped LCO multilayers epitaxially grown on a LSAO substrate. Simultaneously acquired (b) HAADF and (c) ABF images of one Sr-$\delta$-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-$\delta$-doped regions. (b) O-$K$ edge from a Sr-$\delta$-doped region and from LCO. (c) Integrated O-$K$ pre-edge intensity profile across 4 Sr-$\delta$-doped regions.