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Atomic-scale Identification of High-temperature Superconductivity at La_2CuO_4 Interfaces

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Complex oxide thin films and heterostructures host novel functionalities and have stimulated large interest recently due to the ability to tailor physical properties at the atomic scale. Intriguing effects are induced by complex interactions at the interfaces of the epitaxial oxide systems. A prominent phenomenon is the high-temperature interfacial superconductivity (HT-IS) at the interface between two non-superconducting, i.e. metallic (M) and insulating (I), La_2CuO_4 (LCO) layers [1].

In this work, we focus on high-temperature superconductivity (HTSC) of iso- and hetero-structural LCO-based interfaces [2,3]. We design (i) $La_{1.6}A_{0.4}CuO_4$–LCO (A = Ca, Sr, Ba) M–I bilayers [4,5] and (ii) La_2CuO_4–$La_{2-x}Sr_xNiO_4$ (LCO–LSNO) multilayers [6] using atomic-layer-by-layer oxide molecular beam epitaxy (ALL-oxide MBE). In order to correlate structural and functional properties, we extensively characterize the interfaces using aberration-corrected analytical scanning transmission electron microscopy (STEM) techniques, such as electron energy-loss spectroscopy (EELS), high-angle annular dark-field (HAADF), annular bright-field (ABF) imaging and energy-dispersive X-ray spectroscopy (EDXS). For the investigations, a JEOL JEM-ARM200F STEM equipped with a cold field-emission electron source, a probe C_s-corrector (DCOR, CEOS GmbH), a Gatan GIF Quantum ERS spectrometer and a large solid-angle JEOL Centurio SDD-type EDXS detector was used. STEM imaging and EELS were performed at probe semi-convergence angles of 20 mrad and 28 mrad, respectively. The collection angles for HAADF and ABF images were 75-310 mrad and 11-23 mrad, respectively. For the STEM analysis, the O-O picker tool [7] and STEM SI Warp [8] software have been utilized.

In the case of M–I bilayers, the choice of the dopants remarkably influences the superconducting mechanisms (i.e. bulk vs interface) as a consequence of dopant distribution near the interfaces independent of the high-quality coherent interfaces. For Ca- and Sr-doped bilayers, the interfaces are chemically sharp and thus present striking interface effects, i.e. electronic redistribution is predominant. In the case of Ba-doping, the HTS is rather ascribed to "classical" homogeneous doping determined by cationic intermixing and a wide redistribution of Ba [5]. We further demonstrate the occurrence of HT-IS by the heteroepitaxial contact of LCO with LSNO, where the HAADF images demonstrate the structural quality (Figure 1 and Figure 2a). The final superconducting properties of the LCO–LSNO interface can be tuned by changing the structural parameters and HTS is characterized by the accumulation of holes at the interfaces due to the local cationic intermixing and the presence of space-charges as determined by atomically resolved EELS elemental and fine structure mapping (Figure 2b) [6]. These findings highlight the crucial effect of the interface-sharpness and interface-design on the superconducting properties and underline the significance of combined advanced aberration-corrected STEM techniques for the study of functional oxide interfaces [9].

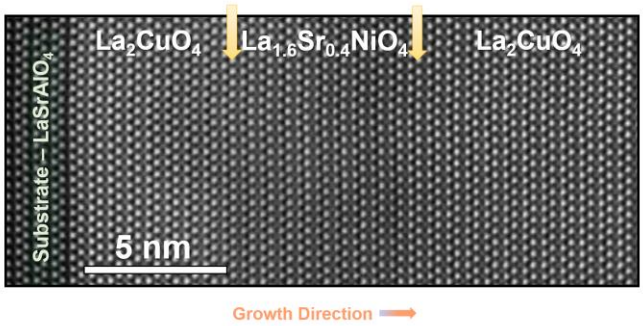

Figure 1. STEM-HAADF image of LCO–LSNO–LCO trilayer demonstrating the high epitaxial quality. The orange arrows indicate the nominal interfaces.

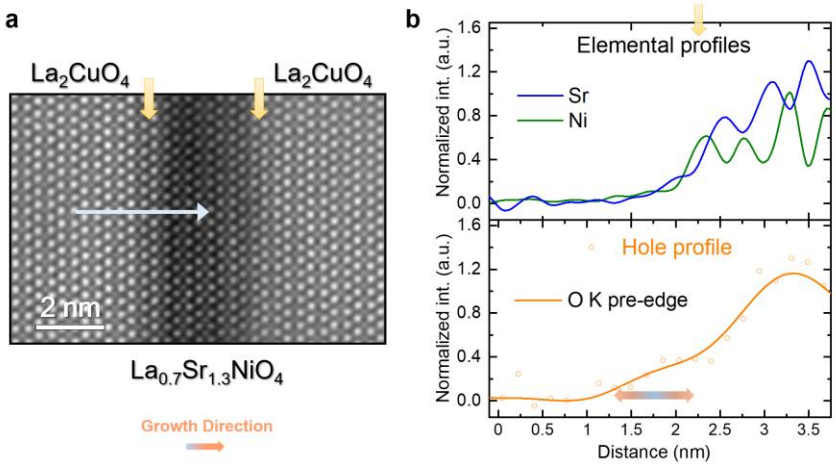

Figure 2. (a) STEM-HAADF image of LCO–LSNO–LCO multilayer demonstrating the epitaxial quality. The blue arrow represents the region of the acquired line scan profiles. (b) Sr (blue), Ni (green), and O K pre-edge (orange) profiles indicate the decoupling between the Sr and the hole profiles. The orange arrows indicate the nominal interfaces.

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