Linking cationic distribution and growth kinetics in δ-doped superconducting La_2CuO_4 heterostructures

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The ability of improving interface sharpness to limit cationic intermixing is a step forward to the discovery on novel interface functionalities in oxide heterostructures. One of the most important growth techniques for interface engineering is atomic-layer-by-layer oxide molecular-beam epitaxy (ALL-oxide MBE), which enables e.g. the synthesis of La_2CuO_4 (LCO)-based high-temperature superconductor structures [1]. For characterising the interfacial structure of such systems, in which the interface coherency and chemistry play a key role for the physical properties [2,3,4], a sub-Å probe of a spherical aberration-corrected scanning transmission electron microscopy (AC-STEM) is indispensable.

In the present study, we use AC-STEM to investigate δ-doped superconducting La_2CuO_4 heterostructures, in which single LaO layers were substituted by MO layers of different ionic radius and charge, namely M = Ba^{2+}, Sr^{2+}, Ca^{2+} and Dy^{3+}as substituting cations for La [5]. A JEOL JEM-ARM200F equipped with a cold field-emission electron source, a probe C_s-corrector (DCOR, CEOS GmbH), a Gatan GIF Quantum ERS spectrometer is used for STEM investigations. STEM imaging and electron energy-loss spectroscopy (EELS) were performed at probe semi-convergence angles of 20 mrad and 28 mrad, respectively; the collection angle range for high-angle annular dark-field (HAADF) images was 75 - 310 mrad.

STEM-HAADF imaging demonstrates the high epitaxial quality, but also indicates asymmetric dopant distribution profiles (Figure 1a, b). In order to examine the cation distribution and concentration, detailed EELS (Figure 1b) and EDXS analyses were performed. Atomically-resolved EELS investigations substantiate the asymmetric dopant distribution profiles. Most importantly, the profiles are asymmetric for all dopants though with different dopant redistribution lengths. Such distribution is correlated with a qualitative model based on thermodynamic considerations and growth kinetics. As far as the dopant redistribution mechanism is concerned, the main factor leading to intermixing in the substrate direction is thermal diffusion. On the other hand, the wider distribution of the dopant (~2 unit cells) in growth direction is a consequence of the high lateral mobility of the atoms, which triggers the tendency to cationic intermixing at the surface at high temperatures during the growth process.

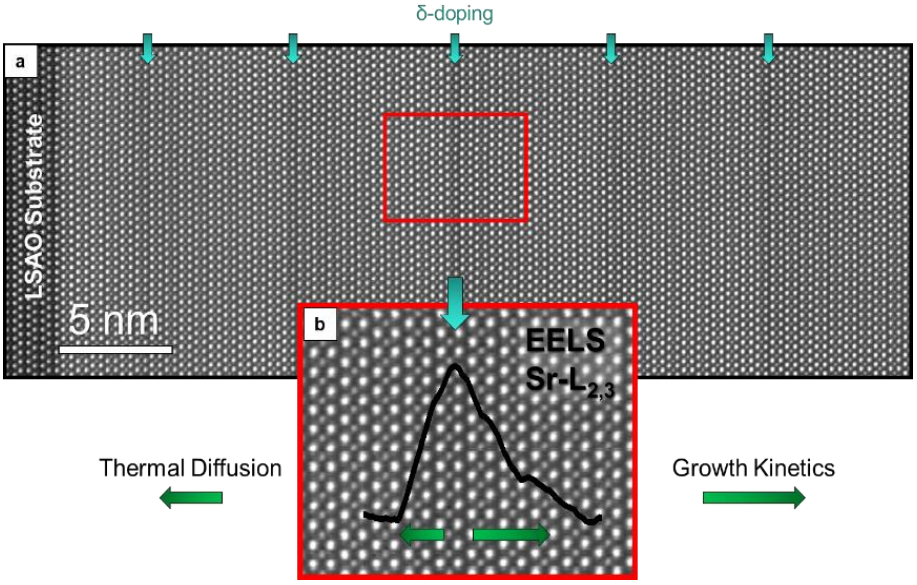

Figure 1. (a) STEM-HAADF image of a Sr δ-doped heterostructure exhibiting high structural quality. (b) Larger magnification of the area highlighted by the red rectangle in (a). The inset shows the integrated Sr-$L_{2,3}$ EELS line profile presenting an asymmetric dopant distribution.

References

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