Dopant distribution and Jahn–Teller distortions at superconducting La_2CuO_4 interfaces

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Epitaxial interfaces in oxide systems have produced many novel functionalities such as ferroelectricity, magnetism and superconductivity. One of the most prominent effects is high-temperature interfacial superconductivity occurring at the interface of two non-superconducting La_2CuO_4-based layers [1]. In this work, utilizing the unique capabilities of atomic-layer-by-layer oxide molecular-beam epitaxy (ALL-oxide MBE), $La_{1.6}A_{0.4}CuO_4/La_2CuO_4$ (i.e. metal/insulating) bilayers, in which *A* refers to the dopant, i.e. Ca^{2+}, Sr^{2+}, and Ba^{2+}, were grown. All bilayer systems were found to be superconducting with critical transition temperatures up to ≈40 K, despite none of the layers is superconducting on its own [2].

To assess the role of the dopant size on the interface superconductivity, each bilayer was investigated by aberration-corrected scanning transmission electron microscopy (STEM) [3]. 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 large solid-angle JEOL Centurio SDD-type energy-dispersive X-ray spectroscopy (EDXS) detector, and a Gatan GIF Quantum ERS spectrometer was used. STEM imaging and electron energy-loss spectroscopy (EELS) were performed at probe semi-convergence angles of 20 mrad and 28 mrad, respectively. Collection angles for high-angle annular dark-field (HAADF) and annular bright-field (ABF) images were 75 - 310 mrad and 11 - 23 mrad, respectively.

The high-structural quality and perfect coherent interfaces were revealed via HAADF-STEM imaging (Fig. 1a). To study dopant distribution, detailed EELS analyses were performed (Fig. 1b) and the dopants were found to be inhomogeneously distributed in the metallic layer revealing distinct differences among the three dopant species. Furthermore, ABF-STEM imaging provided quantitative information on interatomic distances and octahedral distortions yielding different Jahn - Teller (JT) and anti-JT effects. A series of exciting findings are highlighted: (i) the *c*-lattice parameter of the bilayers is linearly dependent on the ionic size while dopant-specific redistribution is found at the interfaces, (ii) superconductivity is highly dependent on the dopant of choice, (iii) larger dopant (i.e. Ba^{2+}) hinders interfacial superconductivity due to a wide distribution of the dopant, and (iv) dopant distribution has a remarkable effect on the JT and anti-JT distortions.

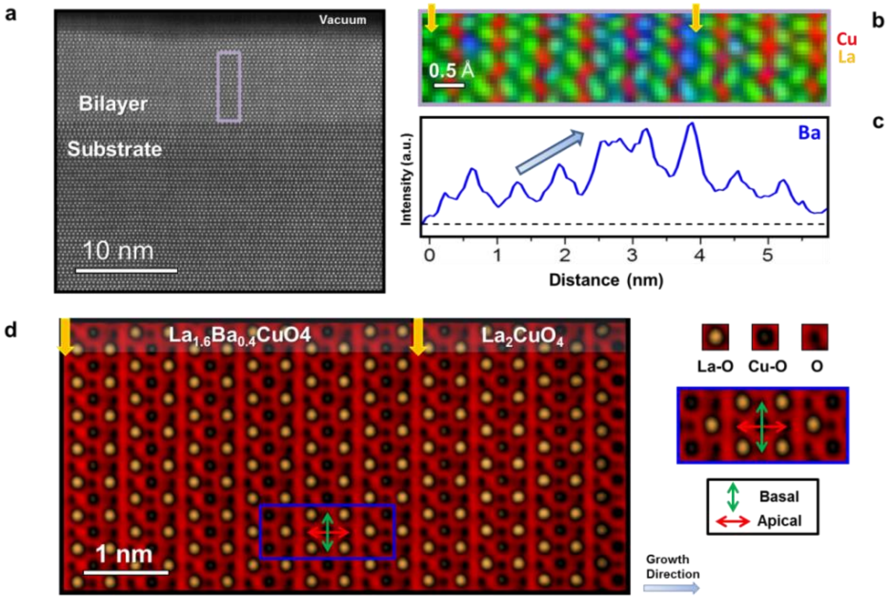

Figure 1. (a) STEM-HAADF image of a Ba-doped bilayer presents high structural quality, (b) atomically resolved EEL SI showing the elemental distribution of a Ba-doped bilayer, (Red: Cu, Green: La, and Blue: Ba) (c) the average profile of the Ba-distribution obtained from (b) and (d) superimposed simultaneously acquired HAADF (dark yellow) and ABF (red) images taken from a $La_{1.6}Ba_{0.4}CuO_4/La_2CuO_4$ bilayer sample showing all atomic columns in one image. Orange arrows on the images indicate the nominal interfaces.

References

[1] Gozar, A. et al., *Nature*, **455**, 782 (2008).
[2] Suyolcu, Y. E. et al., *Scientific Reports,* **7**(1), 453 (2017).
[3] Suyolcu, Y. E. et al., *Advanced Materials Interfaces*, **4,** 1700737 (2017).