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Probing Jahn–Teller Distortions at Superconducting La_2CuO_4 Interfaces

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A wide range of intriguing functionalities can emerge at oxide heterostructure interfaces depending on the choice of the constituents. One of the most exciting phenomena is the discovery of high temperature interfacial superconductivity (HT-IS) at the interface between two non-superconducting, i.e. metallic (overdoped) and insulating (undoped), layers [1]. It is well-established that the properties of complex perovskite-based oxide structures are strongly influenced by small structural changes of the BO_6 octahedral network, and one of the interface-mediated phenomena occurring at such interfaces is the Jahn–Teller (JT) effect [2]. For instance, in native La_2CuO_4 (LCO), the CuO_6 octahedron is elongated along the "apical direction" by the JT effect and exhibits two long and four short Cu-O bonds [3].

In the present contribution, we report on the local octahedral distortions linked with dopant distributions at high-temperature superconducting LCO bilayer interfaces grown by atomic layer-by-layer oxide molecular-beam epitaxy. The bilayers consist of six unit cells in total: A three unit cell thick overdoped metallic $La_{1.6}A_{0.4}CuO_4$ layer is followed by an undoped insulating La_2CuO_4 layer on top [4], where A refers to a substitution of La with the dopants Ba^{2+}, Sr^{2+}, and Ca^{2+}. The interfaces are probed by aberration-corrected scanning transmission electron microscopy techniques. 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. The collection angles for high-angle annular dark-field (HAADF) and annular dark-field (ABF) images were 75-310 mrad and 11-23 mrad, respectively. For the measurement of the interatomic distances the O-O picker tool software has been used [5].

The high epitaxial quality and the coherent interfaces are revealed via HAADF imaging. The elemental distribution across the interfaces are determined by atomically-resolved EELS spectrum images indicating the obvious differences in cationic distribution at the interfaces depending on the cation size as well as exhibiting mild differences compared to 2D doping with the same cations [6]. For determining the atomic column positions and quantitatively investigating the octahedral distortions and interatomic distances (e.g. O-O and La-La), atomically resolved HAADF and ABF images were acquired simultaneously. An ABF image taken from the Ba-doped bilayer is presented as an example in Figure 1, which also represents the elemental redistribution lengths of each dopant marked with color-coded arrows, namely ~1.4 nm for Ca-doped, ~1.6 nm for Sr-doped and ~2.6 nm for Ba-doped bilayer. Results will be presented for each dopant and the correlations of JT distortions with HT-IS will be discussed.

In conclusion, the interrelation between the cationic size mismatch between dopant (A^{2+}) and host La^{3+} ions and the local structure are reported, and a relation between the nature of superconductivity (bulk vs interface) and JT distortions is suggested [7]. This work underlines the crucial effect of the dopant distribution on the octahedral network and, in turn, on the final superconducting properties, and highlights the importance of advanced STEM techniques for the study of oxide interfaces.

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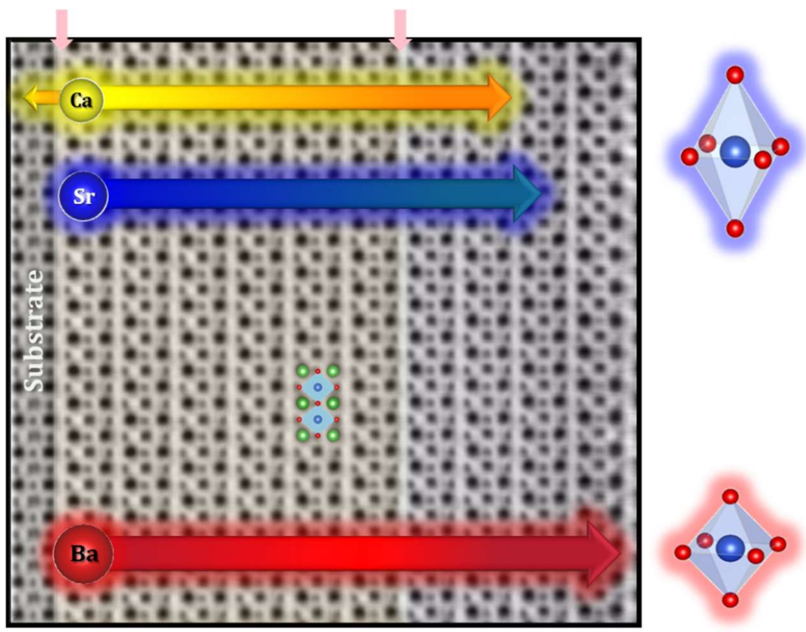

Figure 1. STEM-ABF image of the Ba-doped La_2CuO_4 bilayer displaying all atomic positions. The color-coded arrows represent the redistribution lengths of each dopant: Red for Ba, blue for Sr, and yellow for Ca. Expanded (blue) and contracted (red) CuO_6 octahedrons represent Jahn-Teller and anti-Jahn-Teller distortions. The pink arrows displayed on the top of the image indicate the nominal interfaces.

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