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Oxygen Intercalation and Doping-Induced 2D High-T_c Superconductivity at the $CaCuO_2/SrTiO_3$ Interface

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Energy consumption in high-performance computing is nowadays a serious issue, with data centers consuming more than 2% of the world energy and growing. As semiconductor technology is reaching the critical integration density, supercoducting devices with high switching speed and lossless interconnects are regarded as a possible answer to this problem. At the same time, the interface between complex oxides (e.g., the LAO/STO interface) is emerging as a very promising electronic system because it hosts many fascinating electronic phenomena that are absent in its original constituents. Superconductivity is one of such phenomena but until now only with a very low critical temperature (T_c ≈ 100 mK). Here, by means of aberration-corrected STEM and EELS, we demonstrate doping-induced high-T_c superconductivity with T_c = 50 K at a single interface plane between the insulating oxides $CaCuO_2$ (CCO) and $SrTiO_3$ (STO). Our results show that it might be possible to isolate a single CuO_2 plane with high-T_c superconductivity for realization of 2D superconducting field effect devices operating at a temperature much higher than the LAO/STO interface.

$[(CaCuO_2)_n/(SrTiO_3)_m]_N$ superlattices made by N repetitions of the (CCO)/(STO) interface (n and m being the number of unit cells of CCO and STO, respectively), and STO/CCO/STO trilayers were grown by PLD using different oxidizing conditions on NdO terminated, (110) single crystal $NdGaO_3$ (NGO). The superlattices grown in highly oxidizing conditions showed T_c = 50 K [1]. Earlier studies had suggested a superconductivity mainly confined at the interface between CCO and STO [2]. Moreover, R(T) measurements on NGO/CCO/STO and NGO/STO/CCO samples had shown that only the NGO/CCO/STO sample, having the interface stacking CuO_2-Ca-TiO_2-SrO hosts superconductivity. By analogy with cuprate superconductors, we surmise that superconductivity arises as extra oxygen ions are incorporated in the interface Ca plane, acting as apical oxygen for Cu and providing holes to the CuO_2 planes. The fact that only in the CuO_2-Ca-TiO_2-SrO interface the Ca plane can host extra oxygen can be explained considering that 1) $CaTiO_3$ is a stable phase, and 2) in the non-superconducting interface TiO_2-SrO-CuO_2-Ca, the SrO plane is already stoichiometrically full of oxygen ions; therefore, no doping can occur. Here we prove this hypothesis by using annular bright field (ABF) imaging, a technique for imaging light elements, and atomically resolved EELS at the O-K edge. Figure 1a shows an ABF image of the CCO/STO interface with CuO_2-Ca-TiO_2-SrO stacking. The image has been inverted to display the atomic columns as bright on a dark background. The position of the interface was identified by simultaneously acquiring the high angular annual dark field (HAADF) image, in which the intensity depends nearly on Z^2 and thus can be used to identify cation positions. In addition, spectrum images were taken near the interface and the Ti-$L_{2,3}$, Ca-$L_{2,3}$, and Cu-$L_{2,3}$ integrated intensities plotted as shown in Fig. 1b.The upper arrows in Fig. 1a indicate the interfacial CaO_x plane, where O columns are clearly seen between the brighter spots indicating the Ca columns. This is also seen in the intensity profile for this plane shown in Fig. 1c (upper panel). The lower panel of Fig. 1c shows the intensity profile taken on

the second Ca plane from the interface. For this plane, both image and line profile show a dramatic reduction of O. The subsequent planes as well as the Ca plane at the STO/CCO interface with stacking TiO_2-SrO-CuO_2-Ca, do not show any detectable amount of O.

As documented by x-ray absorption spectroscopy (XAS), the doping holes in cuprate superconductors show a specific signature in the O-K edge. Upon doping the parent compound, a pre-edge feature (peak A) emerges in the O-K at around 529 eV, while the prepeak around 532 eV (peak B) decreases in amplitude. Peak A is associated to the low-energy quasi-particle band called Zhang-Rice singlet: a locally bound d^9 copper 3d hole hybridized with a doped ligand hole distributed on the planar oxygen 2p orbitals. Peak B instead, is associated with the upper Hubbard band and results from a $3d^9$->$\underline{1s}3d^{10}$ transition of the undoped material [3]. Figure 1d compares the EELS at the O-K edge acquired within the same spectrum image across the trilayer sketched in the inset. The curves are spectra well within the STO (green, dashed line), at the second Ca plane from the interface (blue, solid line), and well within the CCO layer (red, dotted line). We find that peak A (the doped hole concentration) decays with a characteristic length of 1-2 u.c. from the interface and is absent at the non-superconducting STO/CCO interface, for which ABF images do not show intercalated extra O [4].

References:
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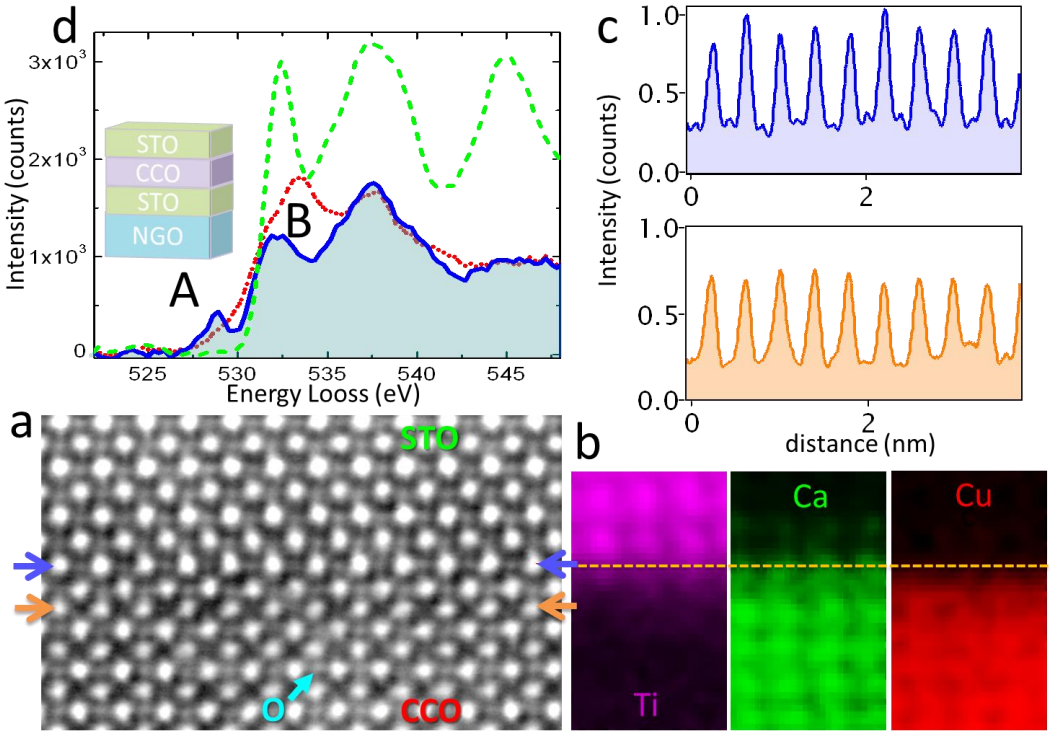

Figure 1. Imaging and spectroscopy of the CCO/STO interface. a) Inverted ABF image. b) Ti, Ca, and Cu EELS maps. c) Intensity profiles of the CaO_x plane at the location of the arrows in a). d) O-K edge of STO (dashed line), interface CaO_x plane (solid line), and bulk CCO (dotted line).