Enhancement of the Superconducting Transition Temperature of $La_{2-x}Sr_xCuO_4$ Bilayers: Role of Pairing and Phase Stiffness

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The superconducting transition temperature T_c of bilayers comprising underdoped $La_{2-x}Sr_xCuO_4$ films capped by a thin heavily overdoped metallic $La_{1.65}Sr_{0.35}CuO_4$ layer, is found to increase with respect to T_c of the bare underdoped films. The highest T_c is achieved for $x = 0.12$, close to the "anomalous" 1/8 doping level, and exceeds that of the optimally doped bare film. Our data suggest that the enhanced superconductivity is confined to the interface between the layers. We attribute the effect to a combination of the high pairing scale in the underdoped layer with an enhanced phase stiffness induced by the overdoped film.

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There is considerable evidence that T_c in the underdoped (UD) regime of the cuprate high-temperature superconductors is governed by phase fluctuations while some sort of pairing occurs at considerably higher temperatures [1–6], akin to the case of granular superconductors [7]. In contrast, the overdoped (OD) region is more conventional in the sense that pairing and phase order take place simultaneously. Consequently, systems which are composed of layers of UD and OD cuprates constitute a unique laboratory for studying the interplay between superconductivity's two necessary ingredients: pairing and phase coherence. Such systems may also serve as models of the naturally occurring multilayered cuprate compounds, such as the Hg series, where measurements of the ^{63}Cu Knight shift have demonstrated that in every unit cell the outer planes tend to become OD, while the inner planes become UD [8,9]. From a practical point of view, the UD-OD multilayers offer the enticing prospect of raising T_c above that of both components, by combining the high pairing scale of the UD layers with the large phase stiffness of the OD layers [10,11].

In this Letter we present a systematic study of $La_{1.65}Sr_{0.35}CuO_4$-$La_{2-x}Sr_xCuO_4$ [LSCO(0.35)-LSCO(x)] bilayers, where x varies from the UD to the OD regime. Our most significant finding is an enhancement of T_c in bilayers containing an UD ($x < 0.15$) layer. The highest T_c, well above that of the optimally doped bare film, was achieved for bilayers with $x = 0.12$, close to the "anomalous" $x = 1/8$ doping level. T_c did not change when the bottom layer was overdoped. Our magnetization measurements, tunneling spectra, temperature-dependent resistance data and nonlinear $V(I)$ characteristics suggest that the enhanced superconductivity occurs at the interface between the layers. We attribute the T_c enhancement (beyond strain effects that cannot fully account for our observations), to an effective combination of the high pairing scale of the UD layer with an increased phase stiffness at the interface, induced by pair-propagation through the OD component. We also point out that the fact that the maximal T_c enhancement occurs at $x = 0.12$ may reflect on the role of stripes in the high-temperature superconductors.

LSCO(x) films and LSCO(0.35)-LSCO(x) bilayers with $x = 0.06$, 0.08, 0.10, 0.12 (UD), $x = 0.15$ (optimally doped) and $x = 0.18$ (OD) were epitaxially grown on (100) $SrTiO_3$ (STO) wafers by laser ablation deposition [see schematic illustration in Figs. 1(a) and 1(c)]. The LSCO(x) films were 90 nm thick, and the LSCO(0.35) overlayer, grown *in situ* without breaking the vacuum,

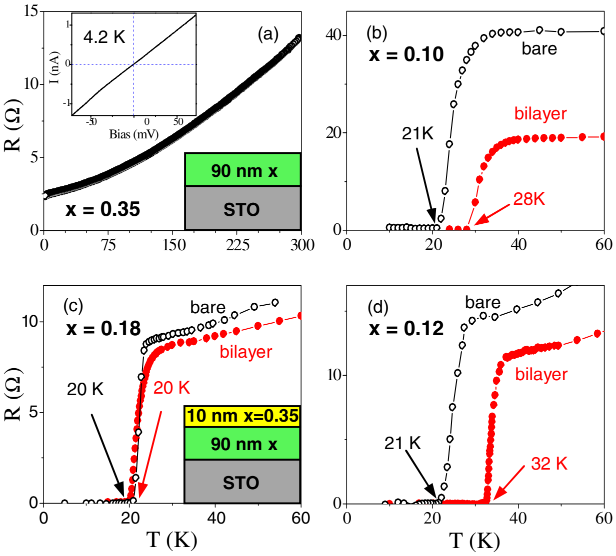

FIG. 1 (color online). (a) $R(T)$ of a bare LSCO(0.35) film. The inset depicts the I-V tunneling characteristic of the same film, taken by STM at 4.2 K. (b)–(d) $R(T)$ curves of the LSCO(0.35)-LSCO(x) bilayers with $x = 0.10$, 0.18, and 0.12, and of the corresponding bare films. The arrows mark the zero-resistance transition temperature.

was 10 nm thick. X-ray measurements confirmed a c-axis orientation perpendicular to the substrate. Temperature-dependent resistance, $R(T)$, measurements were performed using the standard 4-probe technique. Special care was taken to stabilize the temperature before each resistance measurement and to avoid sample heating. We have also measured the properties of a bare 90 nm LSCO(0.35) film, grown on STO, as presented in Fig. 1(a). The $R(T)$ data showed no sign of a superconducting transition down to a temperature of 2 K. Tunneling spectra taken at 4.2 K using a scanning tunneling microscope (STM) exhibited Ohmic behavior; see inset to Fig. 1(a). Therefore, we conclude that the $x = 0.35$ layer is metallic in the temperature range of our experiments ($T > 4.2$ K).

Typical $R(T)$ curves of $x = 0.10$, 0.12, and 0.18 bilayers are presented in Figs. 1(b)–1(d) along with the corresponding bare film data. The relatively low T_c values of the bare films agree with previous studies of LSCO films grown on STO [12–14], as also shown in Fig. 2(b). The tensile strain generated by the lattice-constant mismatch between the film and substrate causes the transition temperature to drop below the T_c of the corresponding bulk sample. As illustrated in Fig. 1(b), the transition temperature of an LSCO(0.35)-LSCO(0.10) bilayer was higher than the T_c of the LSCO(0.10) UD bare film. On the other hand, no effect on T_c was observed for bilayers with an LSCO(0.18) OD film, see Fig. 1(c). This contrast in the behavior of the UD and OD bilayers is clearly apparent in Fig. 2(a), which presents a compilation of the zero-resistance transition temperatures of all the films and bilayers measured by us. An enhancement of T_c was observed for all the UD bilayers studied, with a magnitude that decreased both towards the UD boundary of the superconducting phase and the optimal doping level of the bare films. Surprisingly, the largest enhancement, of 11 K, was found for the $x = 0.12$ bilayer. Moreover, the T_c of the LSCO(0.35)-LSCO(0.12) bilayer was higher than those of both the bare optimally doped LSCO(0.15) film and its bilayer. We have also measured a sequence of inverted bilayers, where a 10 nm LSCO(x) film was deposited on top of a 90 nm LSCO(0.35) layer, and have found essentially the same behavior. It is also worth noting that, in a control experiment, no T_c enhancement was observed in bilayers of gold and LSCO(0.12).

Establishment of bulk superconductivity is accompanied by a diamagnetic Meissner signal. With our SQUID magnetometer sensitivity we could not detect any such signal at the enhanced T_c of our bilayers. However, a clear diamagnetic response was observed when each bilayer was cooled through the transition temperature of the corresponding bare LSCO(x) film. This behavior points to the fact that the enhancement does not occur in the bulk of the sample, but is likely an interface phenomenon. We find further support for this conclusion in our STM data.

The tunneling spectra of our bilayers, measured by an STM on the surface of the LSCO(0.35) top layer, exhibited a predominantly Ohmic (gapless) behavior similar to that of the bare LSCO(0.35) film shown in Fig. 1(a). However, when the thickness of the top LSCO(0.35) layer was reduced from 10 to 5 nm, the differential conductance revealed a gap in the low-energy density of states over large parts of the sample surface, as depicted in Fig. 3. It is possible that the STM tip is coupled to a superconducting region at the interface [assuming that the LSCO(0.35) is in the ballistic regime], or alternatively, that the gap is a consequence of a proximity effect in the metallic layer due to such a region. The latter interpretation seems more convincing in light of the absence of coherence peaks from the bilayer data, and the fact that the zero-bias conductance is rather high, about 75% of its normal state value. This should be compared with the spectra measured on the bare LSCO(0.10) film, shown in the inset of Fig. 3, where the normalized zero-bias conductance is about 3 times smaller and the coherence peaks are well devel-

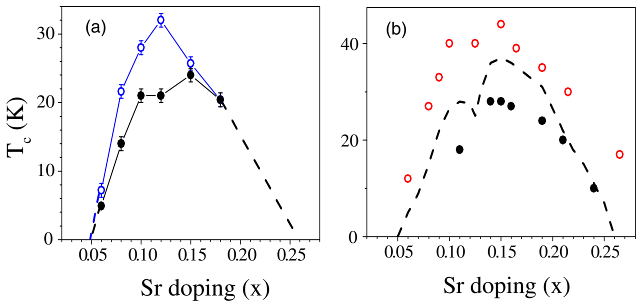

FIG. 2 (color online). (a) T_c vs x of the bilayers (open symbols) and bare films (solid symbols) measured in this work. (b) T_c of LSCO films grown on LaSrAlO$_4$ (open symbols), and on STO (solid symbols), as compiled from Refs. [13,19]. The dotted line depicts the T_c of bulk LSCO.

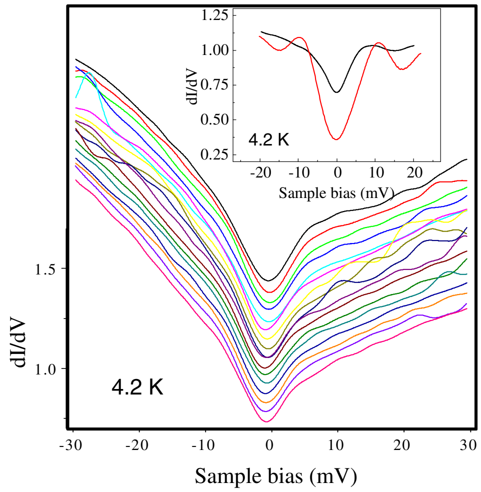

FIG. 3 (color online). Tunneling spectra of a bilayer composed of a 5 nm LSCO(0.35) film on top of a 90 nm LSCO(0.10) layer. The data were taken at 4.2 K and at equidistant steps along a 31 nm long line. Inset: A spectrum of the bare LSCO(0.10) film (red or gray curve) and of the bilayer (black curve).

oped. Regardless of the mechanism responsible for the appearance of the gap, this behavior further suggest that the T_c enhancement effect does not occur in the bulk of LSCO(0.35) layer, but is apparently confined to the interface region. We note that superconductivity in metal-insulator LSCO multilayers was also reported in Ref. [14], yet the doping dependence and corresponding theoretical implications were not addressed.

Superconductivity in a two-dimensional system disappears via a Berezinskii-Kosterlitz-Thouless (BKT) transition [16,17], where it is destroyed by phase fluctuations due to the unbinding of thermally excited vortex-antivortex pairs. Consequently, we have looked for the tell-tale signatures of a BKT transition in our data, and found them exclusively in bilayers showing enhancement of T_c, as demonstrated in Fig. 4 for the LSCO(0.35)-LSCO(0.12) bilayer. Specifically, we have fitted the measured temperature-dependent resistance to the predicted BKT form $R(T) = R_0 \exp(-bt^{-1/2})$, valid just above the transition temperature T_{BKT}. Here R_0 and b are material parameters and $t = T/T_{\mathrm{BKT}} - 1$. The best fit yields $T_{\mathrm{BKT}} \cong 32.2$ K, slightly below the value extracted from the resistance derivative, $T_{\mathrm{BKT}} \cong 32.6$ K, as shown in Fig. 4(a). We note that the fit is in very good agreement with data in the temperature range of the transition. At higher temperatures the fit deviates from the data since the resistance of the LSCO(x) layer exceeds that of the LSCO(0.35) layer and the current flows primarily through the latter. The $V(I)$ characteristics are consistent with a BKT transition as well, where one expects $V \propto I^a$, with $a = 3$ just below T_{BKT} and growing with decreasing temperature. Figure 4(b) exhibits such a behavior and provides the estimate $T_{\mathrm{BKT}} \cong 32.5$ K, close to the values stated above. Such signatures, indicative of a BKT transition, were not observed for the LSCO(0.35)-LSCO(0.18) bilayer (that did not exhibit a T_c enhancement), nor on the LSCO bare films.

What is the reason for the enhancement? Previous reports of T_c enhancement in LSCO thin films, attributed the effect either to epitaxial compressive strain exerted by the substrate [13,18,19], or to excess oxygenation of the film [19,20]. Our samples were annealed in standard oxygen environment at moderate temperatures which generally yield a stoichiometric oxygen content [19], thus making it highly unlikely that over-oxygenation plays a role in the enhancement reported here. The effect of compressive strain is depicted in Fig. 2(b), where we plot T_c data [19], for LSCO films grown on LaSrAlO$_4$, whose lattice-constant mismatch with our LSCO(x) layers is somewhat larger than that of LSCO(0.35) [18]. Apparently, compressive strain increases T_c for every x within the superconducting region of the phase diagram. Moreover, the original dome structure of this region is preserved, and, in particular, maintains its maximum at $x = 0.15$. The T_c enhancement in our bilayers presents a markedly different behavior, as seen in Fig. 2(a). First, it occurs only for UD bilayers. Second, the original peak in T_c is shifted from $x = 0.15$ to the vicinity of $x = 0.12$, where a dip or flattening occurs in the T_c curve of the bare films. Thus, strain alone cannot account for the enhancement found in the bilayer systems. Finally, since the maximal enhanced T_c is far larger than the optimal T_c of the bare films, we can rule out migration of cations across the interface as the source of the effect.

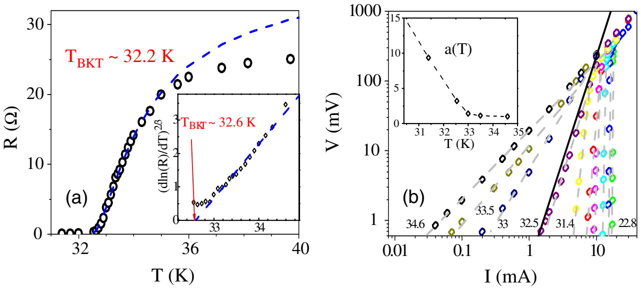

FIG. 4 (color online). (a) $R(T)$ of the bilayer with $x = 0.12$. The blue (dark gray) dashed line is a fit to the expected BKT behavior near the transition, yielding the estimate $T_{\mathrm{BKT}} \approx 32.2$ K. Inset: The same data plotted as $(\frac{d\ln R}{dT})^{-(2/3)}$ versus T, which fits to $T_{\mathrm{BKT}} \approx 32.6$ K. (b) $V(I)$ characteristics at various temperatures. The solid line corresponds to $V \propto I^3$. The inset shows the exponent a in $V \propto I^a$ as a function of T.

A previous study [7] of an analogous system to the bilayers discussed here, may shed light on our findings. There, T_c of a granular Pb film covered by a silver overlayer, was found to initially increase with Ag thickness. Despite being insulating, tunneling into the bare lead film demonstrated well-developed superconductivity on each grain below the bulk T_c of lead. Strong phase fluctuations between the grains denied the system of establishing global superconductivity. Apparently, the silver enhanced the intergrain Josephson coupling, leading to a larger phase stiffness and higher T_c. The parallels with our bilayers are compelling. Like in the granular lead film, T_c of UD cuprates is governed by their small superfluid stiffness, while there are indications for pairing above T_c (the analogy may go even further in view of the evidence for electronic inhomogeneities in these systems [21]). We suggest that pair tunneling through the metallic LSCO(0.35) overlayer strengthens the phase coupling between locally superconducting regions of the LSCO(x) layer in the vicinity of the interface, thereby enhancing T_c in this portion of the sample. Such coupling is possible since the coherence length in the LSCO(0.35) layer, at the relevant temperatures (estimated from data presented in Ref. [22]), is larger than the typical spatial scale, ~2–3 nm, of the superconducting-gap inhomogeneities in the cuprates [6,21]. When the bottom layer is overdoped, phase stiffness ceases to be a limiting factor and the enhancement disappears. On the other hand, the decrease in the enhancement towards the UD boundary of the superconducting region may reflect the reduction of the excitation gap in this limit, as measured by angle resolved photoemission spectroscopy (ARPES) [23], and by STM [24].

In view of this proposed scenario we need to recall that no enhancement of T_c was observed in our Au-LSCO(0.10) bilayer. Such a negative result may stem from the differences in both the Fermi wave vectors and lattice structures of the two layers, which could significantly reduce the tunneling amplitude through the interface. Additionally, since the induced phase couplings in the bottom layer depend on the pair-propagation amplitude through the top metallic film, it is possible that vestiges of pairing in the LSCO(0.35) layer play a role in establishing the enhancement in the LSCO(0.35)-LSCO(x) systems. Finally, we note that the lack of enhancement in the Au-LSCO(0.10) sample implies that screening due to the top metallic layer is not responsible for the effect which we measure, in contrast to Ref. [25].

Another distinctive feature of our data deserves attention. The maximal enhanced T_c is achieved when the UD layer is approximately $1/8$ doped. At the same doping level the lanthanum based cuprates exhibit an anomaly in the $T_c(x)$ curve, ranging from a local plateau, in the case of LSCO, to a substantial dip for $La_{2-x}Ba_xCuO_4$ [26]. This anomaly is commonly associated with the formation of robust static charge and spin stripe-order [27]. While there are many theoretical indications that the confinement of strongly interacting electrons to quasi-one-dimensional systems, typically leads to a large pairing scale [6], it is also clear that such confinement severely hampers the emergence of global phase coherence, and consequently lowers T_c. The notion that pairing attains a maximum at $x = 1/8$, together with a concomitant increase in phase fluctuations, gains support from experimental signatures as well. Specifically, ARPES experiments show that the single-particle gap in $La_{2-x}Ba_xCuO_4$ is largest for $x = 0.125$ [23], and measurements of the vortex-Nernst effect, which is indicative of a phase-disordered superconducting state, find that in LSCO the maximal signal is attained at the same doping level [4]. In light of these facts it appears that the LSCO(0.35)-LSCO(0.12) bilayer is a system that takes advantage of the significant pairing correlations of the $x = 1/8$ state, possibly due to stripes, while avoiding its limitations vis-à-vis phase coherence by tunneling between regions of local superconducting order (for which stripes are natural candidates) through the OD metallic layer.

In conclusion, we have found that the deposition of a thin, heavily OD (metallic) LSCO film on top of an UD LSCO layer can enhance its T_c by up to 50%, and presented evidence that the effect takes place at the interface between the UD and OD components. The enhancement does not occur when the bottom layer is OD. Our findings corroborate the thesis that superconductivity in the UD cuprates is controlled by the small phase stiffness in this regime. The fact that the maximal enhanced T_c occurs near $x = 1/8$ indicates that the optimal doping level, $x = 0.15$, in bare LSCO samples, may be a result of a suppression of the original peak at $x = 1/8$ due to phase fluctuations. It may also reflect on the role of charge inhomogeneities (such as stripes) in these systems, and demonstrates that once their suppressing effect on the phase coherence is alleviated, the predicted large pairing scale which they induce could increase T_c.

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