Conventional proximity effect in bilayers of superconducting underdoped $La_{1.88}Sr_{0.12}CuO_4$ islands coated with nonsuperconducting overdoped $La_{1.65}Sr_{0.35}CuO_4$

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Following a recent study by our group in which a large T_c enhancement was reported in bilayers of the nonsuperconducting $La_{1.65}Sr_{0.35}CuO_4$ and superconducting $La_{1.88}Sr_{0.12}CuO_4$ films [Yuli *et al.*, Phys. Rev. Lett. **101**, 057005 (2008)], we checked if a similar effect occurs when superconducting $La_{1.88}Sr_{0.12}CuO_4$ islands are coated with a continuous layer of the nonsuperconducting $La_{1.65}Sr_{0.35}CuO_4$. We found that no such phenomenon is observed. The bare superconducting islands film behaves as an insulator where transport occurs via hopping or tunneling between islands but exhibits a weak signature of localized superconductivity at ~15–18 K. When overcoated with a $La_{1.65}Sr_{0.35}CuO_4$ film, it becomes superconducting with a T_c onset of 16.6 K, which is less than that of a thick $La_{1.88}Sr_{0.12}CuO_4$ film (25.7 K). We therefore conclude that the lower T_c in the bilayer is due to a conventional proximity effect.

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A recent paper by Yuli *et al.*[1] has reported the observation of a large T_c enhancement in bilayers of the nonsuperconducting, heavily overdoped (OD) $La_{1.65}Sr_{0.35}CuO_4$ (LSCO-35) and superconducting underdoped (UD) $La_{1.88}Sr_{0.12}CuO_4$ (LSCO-12), in comparison with the bare film of the latter. This study was based on the idea that in the UD regime of the high-temperature superconductors, T_c is determined by phase fluctuations, while pairing without phase coherence occurs in the pseudogap regime at considerably higher temperatures,[2–7] similar to the case of granular superconductors.[8] In the OD regime however, pairing and phase order occur simultaneously, with a robust phase stiffness. Therefore, in the interface of bilayers composed of UD and OD films, one can envision a scenario in which the high phase stiffness of the OD layer locks via Josephson coupling the phases of the preformed pairs in the UD layer. This together with the high pairing in the UD layer is expected to lead to a T_c enhancement effect above that of both components.[9,10] A large T_c enhancements effect was reported by Gozar *et al.* in a similar bilayer system of $La_{1.55}Sr_{0.45}CuO_4$ and La_2CuO_4.[11] It is unclear to us whether this observation is due to the aforementioned combination of UD and OD layers, as it is questionable whether the ozonized La_2CuO_4 which they used is actually underdoped. Motivated by these works, we turned to study bilayers consisting of an insulating LSCO-12 islands film (below the percolation threshold) coated by a continuous metallic LSCO-35 layer. The fundamental question here is whether a similar T_c enhancement effect takes place also in such a granular system. This question touches upon the wider issue of the connection between nominally granular and electronically disorder superconductors.[12] In contrast to the case of bilayers made of continuous LSCO-12 and LSCO-35 films,[1] no T_c enhancement effect was found here when the LSCO-12 film was granular. Although global phase coherence was achieved upon the deposition of LSCO-35 layer on top of the insulating LSCO-12 islands film and a resistive superconducting transition was obtained, the corresponding transition temperature was smaller than that of a continuous LSCO-12 film prepared under similar conditions (25.7 K) and was rather close to the apparent T_c of the localized superconductivity in the islands (~15–18 K). Our results are thus in accord with a conventional proximity effect where global phase coherence is achieved via Josephson coupling between the localized superconducting grains as discussed by Merchant *et al.*[8]

The bilayers and films in the present study were grown epitaxially on (100) $LaAlO_3$ (LAO) wafers by standard laser-ablation deposition using the third harmonic of a Nd-YAG laser with 10-ns laser-pulse duration, 355-nm wavelength, and 1.2 J/cm^2 laser fluence on the cuprate targets. The optically polished LAO wafers had 10×10 mm^2 area and 0.5-mm thickness. Deposition was done at 790 °C wafer temperature while the ambient oxygen pressure was maintained at 90 mTorr. Cooling was done in 0.75 atm of oxygen pressure with a dwell of 2 h at 450 °C. In order to reform the surface of our polished substrates and improve their surface quality, we always deposited on them a thin epitaxial LAO template layer of 10-nm thickness prior to the deposition of the other layers. Then the samples were prepared *in situ* as shown schematically in Fig. 1, where half of the wafer is coated with the reference islands film (nominal 5 nm LSCO-12), and the other half with the cap-layer film (50 nm LSCO-35) on top of the islands film, using a shadow mask.

Since the nominal 5-nm thick LSCO-12 islands film is very sensitive to humid air, its exposure to the atmosphere was kept to a minimum (a few minutes), until it was mounted in the measuring probe in He gas ambient. The resistance versus temperature [R(T)] was then measured using the standard four-probe dc technique and the results are shown in Fig. 2. The resistance of this film shows a metal to insulator transition at about 170 K, a signature of the superconducting transition with a maximum at 18 K and minimum at 11 K, and an insulating behavior at lower temperatures.

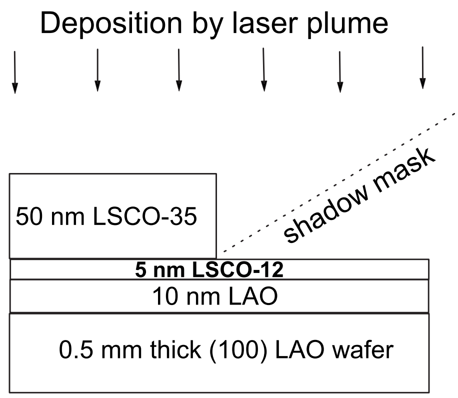

FIG. 1. The experimental setup for *in situ* deposition of a bilayer and its reference film on the same wafer.

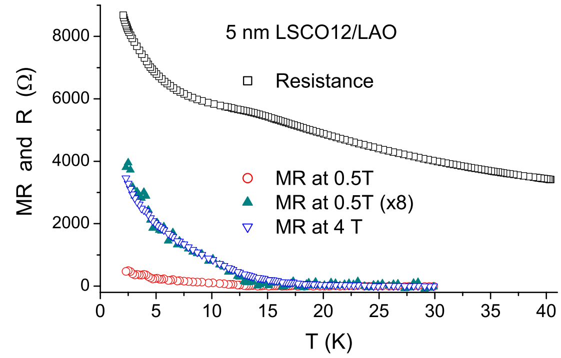

FIG. 3. (Color online) Resistance (top curve) and magnetoresistance (lower curves) versus temperature of another nominally 5-nm-thick LSCO-12 film deposited on a 10-nm-thick LAO layer coating a (100) LAO wafer.

The resistance and magnetoresistance [MR=R(H)−R(0)] versus temperature of a similar 5-nm-thick LSCO12 film is shown in Fig. 3. One can see that the onset of the magnetoresistance, which indicates the onset of localized superconductivity in this globally insulating film (see below), follows the flattening of the resistance curve in the temperature range of 13–16 K. Therefore, the decreased resistance of the LSCO-12 film of Fig. 2 in this temperature range can be safely related to superconductivity in that film. Such type of "reentrant superconductivity" behavior (shown in Figs. 2 and 3) is typical of superconductor island films at the insulating side of the superconductor to insulator transition and is attributed to the onset of localized superconductivity in the grains.[13] While this feature does not lend itself to an accurate determination of the corresponding T_c, the magnetoresistance data of Fig. 3 is sufficient for this purpose. We note in passing that the magnetoresistance in this regime may be either negative[14] or positive[15] as we observe here, possibly depending on the "distance" from the superconductor to insulator transition. We find that for a 5-nm-thick LSCO-12 film the onset of the magnetoresistance is at $T_c \approx 18$ K, which is still significantly lower than that of the continuous 50 nm thick LSCO-12 film (see Fig. 2). The conductance of this nominally 5-nm-thick film is therefore due to hopping and/or tunneling between superconducting LSCO-12 grains. Hence we can refer to this film as an "islands film," as opposed to a continuously superconducting film (generally, when the film is thicker) since no superconducting percolation path exists in it between the contacts in the R(T) measurement.

To further characterize the surface morphology of this islands film, we show in Fig. 4 an atomic force microscope (AFM) image of a 2×2 μm^2 area. A typical height profile Z (nm) along the line marked on the image is also shown. One can see that there are holes in this film which are at least 3-nm deep and possibly deeper since the microlever of the AFM (the tip) is too wide to penetrate them. The rougher area at the top-left side of the image, which has about twice the roughness of the smoother area, appears as isolated islands of a few μm in size and occupies only about 5% of the total area of the film. We can thus conclude from Figs. 2–4 that the connections between the grains or islands in the nominally 5-nm-thick LSCO-12 film are weak and that transport in this film via these weak links is controlled by hopping or tunneling.

We now turn to the main results of this study. After the nominally 5-nm-thick LSCO-12 islands film was deposited on the whole wafer, a 50-nm-thick LSCO-35 film was deposited on half of the wafer by the use of a shadow mask as shown in Fig. 1. Then the mask was removed and a nominally 3-nm-thick layer of gold was deposited on the whole wafer at the same deposition temperature (790 °C) and in 400 mTorr of oxygen gas pressure. This very thin gold layer,

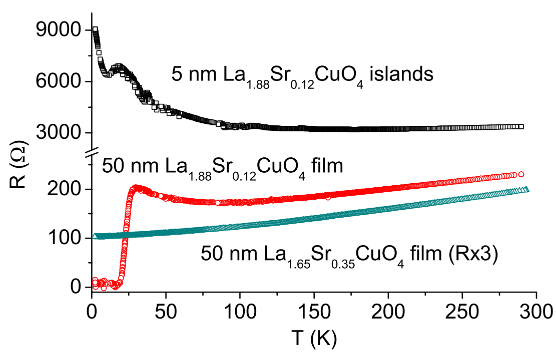

FIG. 2. (Color online) Resistance versus temperature of the three relevant reference films. The nominally 5-nm-thick LSCO-12 islands film, a 50-nm-thick LSCO-12 film, and a 50-nm-thick LSCO-35 film, all on 10 nm deposited LAO layer on the (100) LAO wafers.

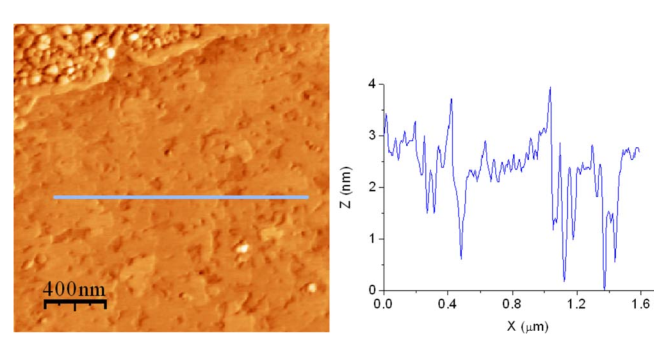

FIG. 4. (Color online) An AFM image of the nominally 5-nm-thick LSCO-12 film, together with a typical height profile along the line shown in the image.

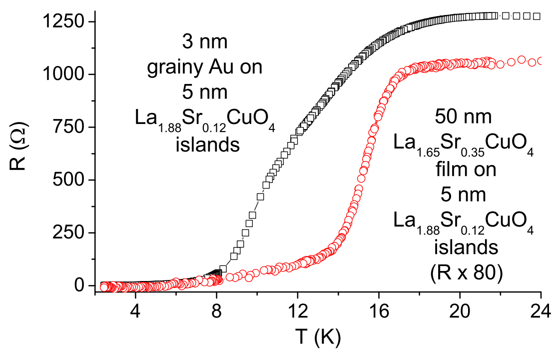

FIG. 5. (Color online) Resistance versus temperature of the 3-nm Au/50-nm LSCO-35/5-nm LSCO-12 trilayer and the 3-nm Au/5-nm LSCO-12 bilayer. (The top 3-nm Au layer in the trilayer is not marked in the figure).

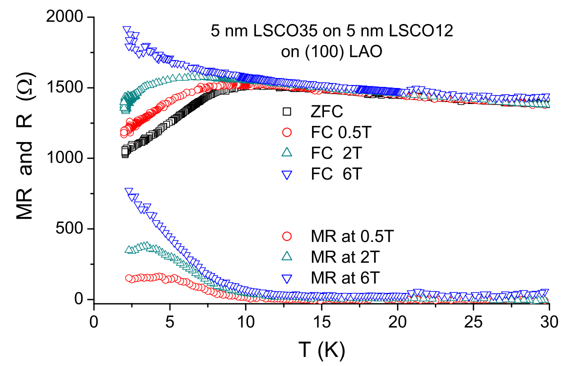

FIG. 6. (Color online) Resistance (upper curves) and magnetoresistance (lower curves) versus temperature under various field cooling conditions, of 5-nm-thick LSCO-35 on 5-nm-thick LSCO-12 bilayer deposited on a 10-nm LAO layer coating a (100) LAO wafer.

which was composed of loosely connected ball-like grains, was mainly used to improve the contacts in the transport measurement, but also induced a proximity effect in the LSCO-12 islands film. The wafer was then patterned by wet acid etch to separate its two halves by a $2\times 10\ \mathrm{mm}^2$ wide strip along the contact line of the mask with the wafer. The resistance versus temperature of the two halves of this wafer was measured in the same cooling run and is shown in Fig. 5. We note first that the resistance versus temperature data in the 24–300 K range (which is not shown here), behave as the 50-nm-thick LSCO-35 film of Fig. 2 in the trilayer case, and as six times the resistance of the 50-nm-thick LSCO-12 film of Fig. 2 in the bilayer case. The onset of the superconducting transition (defined here as the temperature of 10% drop in resistance) of the Au/LSCO-12 bilayer and the Au/LSCO-35/LSCO-12 trilayer are very similar, 15.6 and 16.6 K, respectively. We note here that using the same definition of T_c onset for the bare 5-nm-thick LSCO-12 islands film of Fig. 2, one also finds the same T_c of 16.6 K for the localized superconductivity in the loosely connected grains. The respective midpoint transitions in Fig. 5 are already quite different and equal to 11.6 and 15.3 K. The same behavior and even more so is true for the extrapolated to zero-resistance transition temperatures which are found at 8.3 and 13.7 K, respectively. At lower temperatures, there is a Josephson "tail" or "knee" of the resistance in both cases, which in the trilayer reaches zero at 5–6 K, and in the bilayer decreases exponentially and approaches zero at 2 K. The improved superconducting quality of the trilayer compared to that of the bilayer is due to both the thicker LSCO-35 film and the better Fermi wave-vector matching in the former system. Our results indicate a clear proximity effect and Josephson coupling in both cases, where, on the one hand global phase coherence was induced in the insulating 5-nm (base) LSCO-12 islands film, but, on the other hand, T_c was lower than that of the bare 50-nm-thick reference LSCO-12 film, as depicted in Fig. 2. More importantly, T_c onset in both bilayer and trilayer films are comparable to the location of the "reentrant" peak feature observed in the R(T) curve of the islands film (Fig. 2), which is associated with the onset of localized superconductivity in the LSCO-12 islands. Our data are similar and in agreement with a previous study by Nesher *et al.*[16] where a proximity effect was observed in a bilayer of $Y\mathrm{Ba_2Cu_3O_{7-\delta}}$ islands overcoated with a normal $Y\mathrm{Ba_2Cu_{2.7}Fe_{0.3}O}_y$ layer (above its superconducting transition temperature).

It may be instructive to compare our findings also with those of Merchant *et al.*[8] There, global phase coherence was induced in an insulating Pb islands film by overcoating with a normal Ag layer. The T_c of this bilayer first increased with increasing Ag thickness up to nearly the bulk T_c of lead but then decreased with further *in situ* Ag deposition. The initial increase in T_c was attributed to enhanced intergrain Josephson coupling and the subsequent decrease, to the Ag-Pb proximity effect. Our results are in partial agreement with these observations. On one hand we find emergence of global superconductivity upon overcoating the insulating LSCO-12 islands film by a metallic layer of either a thin (3 nm) Au or a thick (50 nm) LSCO-35, in agreement with.[8] On the other hand, the T_c-onset values in both cases were similar ($\sim$16 K) and well below that of a bare 50-nm-thick LSCO-12 film (25.7 K), which is in contrast with (Ref. 8). We therefore tested the case where a very thin metallic LSCO-35 ad layer of 5-nm thickness is used to overcoat a 5-nm-thick LSCO-12 film grown under the same conditions as that in Fig. 3. Figure 6 shows the measured transport results of the zero-field-cooled (ZFC) resistance, field-cooled (FC) resistance under various fields, and the corresponding magnetoresistance (MR) of this bilayer as a function of temperature. As one can see T_c of this bilayer at approximately 10–12 K is still much lower than that of the thick LSCO-12 film, unlike what Merchant *et al.* had found for bulk Pb. Possibly, this discrepancy is due to the much shorter coherence length of the present cuprate bilayer (3–4 nm) as compared to that of bulk Pb (about 100 nm at 5 K). In the cuprates therefore, any metallic ad layer such as the LSCO-35 film here, thicker than 5 nm will have no further significant effect on the proximity T_c of the bilayer. We thus conclude that the fact that the T_c onset in both our trilayer and bilayer films were close to the reentrant superconductivity feature of the LSCO-12 islands film (and the corresponding onset of magnetoresistance), indicates that it is determined by the T_c of the grains. It is important to note here that we have observed superconducting transition also in bilayers for which the corresponding insulating LSCO-12 islands film did not exhibit reentrance. T_c in this case, was determined by the onset of the magnetoresistance.

As we have seen in Figs. 2 and 3, the base LSCO-12 islands film has an insulating behavior at low temperatures with no T_c while the ten times thicker and clearly continuous reference LSCO-12 film has a T_c onset of 25.7 K and $T_c(R=0)$ of 19.6 K. For the sake of completeness, we also show in Fig. 2 the metallic but nonsuperconducting behavior of the overdoped LSCO-35 film. Note that due to the low resistance of this film, the resistance of the curve shown is multiplied by a factor of 3. Another point to note is that the normal resistance of the Au/LSCO-12 islands bilayer just above the superconducting transition is about 80 times larger than that of the Au/LSCO-35/LSCO-12 islands trilayer. This indicates that the resistance of the nominally 3-nm-thick Au layer is much higher than that of the 50-nm LSCO-35 film, and corroborate the islands nature of this layer too. The ball-like (or islands) surface morphology of the Au layer was also seen in several AFM images in this study and in other experiments where gold films were deposited under similar conditions. We also note that in the trilayer case, the top gold layer only reduces the contact resistance and does not contribute to the proximity effect which obviously occurs at the LSCO-12 and LSCO-35 interface, located 50 nm beneath the Au grains. And finally, the data of Fig. 6, which shows clear superconductivity and was taken on a bilayer without gold, proves that the gold layer has no effect on the trilayer data of Fig. 5.

To summarize our main finding, the transport results of Fig. 5 on the bilayer and trilayer together with the results of the bare reference layers of Fig. 2, demonstrate conventional proximity and Josephson-coupling effects between the LSCO-12 islands and either the gold or the LSCO-35 layers, where the bilayer or trilayer T_c values are found in between those of the bare reference films. Our findings here differ from our previous results, where a T_c enhancement with respect to that of a continuous LSCO-12 film, was found for both bilayers of 10-nm LSCO-12 on 90-nm LSCO-35, and 10-nm LSCO-35 on 90-nm LSCO-12.[1] It should be noted that in Ref. 1 all the layers were grown on (100) $SrTiO_3$ (STO) wafers. In this case, thicker base layers (either LSCO-35 or LSCO-12) were used in order to release tensile strains with the STO substrate so that the interface of the bilayers will be less affected by strains. In the preset study, we chose to work with an LAO substrate since its lattice match with the various LSCO-*x* cuprates is better (cubic a axes are 0.3788 nm for LAO versus 0.3905 nm of STO while LSCO-10 has a 0.3784-nm lattice constant[17]). The fact that no T_c enhancement was found here, in contrast to Ref. 1, may be due to the following reason. The enhanced T_c in Ref. 1 was found to be confined to a continuous two-dimensional interface layer between the LSCO-12 and LSCO-35 films. In the present study, a continuous two-dimensional interface layer cannot exist since the LSCO-12 islands film was below the percolation threshold. Nevertheless, Josephson coupling between islands induced by either the Au or LSCO-35 ad layers was sufficient for the actual emergence of global phase coherence in the bilayer and trilayer films in the present study.

In conclusion, conventional proximity effect and Josephson coupling were observed in the present study in bilayers of an insulating LSCO-12 islands film with either LSCO-35 or Au. Although global phase coherence developed in these films, the transition temperatures appeared to be limited by the proximity effect, showing no enhancement with respect to bare LSCO-12 films.

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