High-temperature interface superconductivity in bilayer copper oxide films by pulsed laser deposition

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ABSTRACT In a seminal work, Gozar *et al.* reported on the high-temperature interface superconductivity in bilayers of insulating La_2CuO_4 and metallic $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.45$). An interesting question to address is how general and robust this interface superconductivity is. In the past, the cuprate bilayers were grown in a unique atomic-layer molecular beam epitaxy system, with a Sr doping range of $x \leq 0.47$, and the atomically flat interface was thought to be indispensable. Here, we have fabricated bilayers of La_2CuO_4 and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ by pulsed laser deposition. We have tried to extend the nominal doping range of Sr from the previous maximum of 0.47 to the present 1.70 (the nominal Sr content in the targets). X-ray diffraction result indicates that our $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ films with $x \leq 0.60$ have very high crystalline quality; but the film crystalline structure degrades gradually with further increasing x , and finally the structure is fully lost when x reaches 1.40 and higher. Although the film quality scatters dramatically, our experiments show that there exists superconductivity for bilayers in nearly the entire over-doped Sr range, except for a non-superconducting region at $x \sim 0.80$. These observations demonstrate that the interface superconductivity in copper oxides is very general and robust.

Keywords: cuprate, superconductivity, interface, pulsed laser deposition, high temperature superconductivity

INTRODUCTION

The effect of interface on superconductivity has received much attention. It was demonstrated that superconductivity can occur at the interface of two non-superconducting materials [1–6], and the critical temperature (T_c) can even be significantly enhanced com-

pared with the corresponding single-phase samples [1,7–9]. One of the most intriguing interfaces is the bilayer of La_2CuO_4 and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ [LSCO(x)] films. In a seminal work, Gozar *et al.* [1] reported on the existence of high- T_c superconductivity with an enhanced T_c at the interface between insulating La_2CuO_4 and metallic $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$. A following work showed that the high- T_c superconductivity could reside in a single CuO_2 plane on the La_2CuO_4 side [7]. The Sr interdiffusion across the interface was restricted [1], and the superconductivity was attributed mainly to the interface electronic redistribution [10]. An anomalous independence of interface superconductivity from carrier density was observed [11]. These discoveries make the bilayer of La_2CuO_4 and LSCO(x) a base for studying the origin of high-temperature superconductivity and interface emergent phenomena [12].

LSCO(x) is the simplest superconducting cuprate system that has single CuO_2 conducting layers whose hole concentration is determined by the Sr and/or oxygen concentration. It is insulating for $x < 0.06$, superconducting for $0.06 \leq x \leq 0.30$, metallic for $0.30 < x < 0.80$, and insulating again for $x \geq 0.80$ [13,14]. Previous studies on bilayers of La_2CuO_4 and LSCO(x) showed that the bilayers sustained superconductivity when the LSCO(x) layer was superconducting ($0.06 \leq x \leq 0.30$) [11,15], and created interface superconductivity when the LSCO(x) layer was metallic [1], with a maximum x of 0.47 [11]. Will the interface superconductivity persist with further increasing x , particularly for $x \geq 0.80$ where the LSCO(x) layer becomes insulating again? Unfortunately, the answer to it is unknown because the wide doping range for

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$x > 0.47$ is unexplored yet. The lacking of relevant studies might be in part because the heavily Sr-riched LSCO(x) films are not stable due to oxygen vacancies [13,16]. In previous studies [1,5–7,11] of interface superconductivity, the bilayers were grown in a unique atomic-layer-by-layer molecular beam epitaxy (MBE) system that was fantastic in making atomically flat high-quality samples, but might not be suitable in making the heavily Sr-riched LSCO(x) films. In contrast, due to its thermodynamically non-equilibrium nature, pulsed laser deposition (PLD) can be used to produce metastable films far from chemical equilibrium [17]. In this work, using PLD, we extensively explore the interface superconductivity of the bilayers of La_2CuO_4 and LSCO(x), with the nominal doping level x spanning from 0.00 to 1.70 (the nominal Sr content in the PLD targets). Our result shows that, although the film quality scatters, the high- T_c interface superconductivity is very general and can be achieved in a wide range of Sr doping levels.

EXPERIMENTAL SECTION

Target fabrication

The LSCO(x) targets for PLD growth were prepared by conventional solid-state reaction using La_2O_3 (99.9%), SrCO_3 (99.95%), and CuO (99%) powders. These powders were weighted with a ratio of the nominal composition of LSCO(x), mixed, and pre-heated at 900°C (except 1050°C for La_2CuO_4) for 24 h in air. After that, they were pressed into 1 inch-diameter pellets and sintered at 1100°C for 24 h or longer in air.

Film growth

All the films were grown on (001) LaSrAlO_4 single-crystalline substrates. Before growth, the substrates were pre-annealed *in situ* at 800°C under 1×10^{-4} mbar oxygen (O_2) for 20 min. The films grew at 730°C, under a mixed gas atmosphere of 0.033 mbar nitrous oxide (N_2O) and 0.077 mbar O_2 , followed by an *in-situ* post-annealing at 550°C, under 200 mbar O_2 , for 1 h, and then were cooled to room temperature under the same gas condition. A KrF excimer laser (248 nm) was used. The laser fluence was 0.95 J cm^{-2} . The laser spot size on target was 3.0 mm^2 . The repetition rate was 2 Hz. The sample-target distance was 55 mm. The nominal x for all the LSCO(x) films simply refers to x in the corresponding LSCO(x) targets.

Thickness control

The film thickness was determined by counting the growth laser pulses. The growth rate was about 1 nm per

100 pulses, which was calibrated by measuring low-angle X-ray reflectance (XRR) on a few LSCO(x) single-phase films (e.g., Supplementary information, Fig. S1). We deposited all the LSCO(x) single films with 4000 pulses, and all the bilayers with 4000 pulses for the bottom layer and 1000 pulses for the top layer.

Transport measurement

The typical sample size was 3.0 mm in length and 2.0 mm in width. Four electrodes (0.5 mm in length, and 2.0 mm in width) distributed evenly along the long side by evaporating silver on the top of the single-phase and bilayer films. These electrodes were connected by Al wire bonder, and used to measure the samples' resistances in a conventional four-probe method.

X-ray characterization

The X-ray diffraction (XRD) was performed using Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) on a Bruker AXS D8-Discover.

RESULTS

Crystalline structure of single LSCO(x) films

We have grown single and bilayer films with nominal composition LSCO(x) on (001) LaSrAlO_4 single-crystalline substrates by PLD. First, we characterized the structure of the single LSCO(x) films by XRD. Fig. 1 shows XRD data of the samples. The presence of the characteristic K_2NiF_4 structural peaks of (0 0 2n) indicates epitaxial growth of the films on the LaSrAlO_4 substrates. A close look of the XRD data shows clear finite-thickness oscillations around the film peaks for samples of $0.00 \leq x \leq 0.60$ (see Fig. 2), indicating the high quality of the films [14,18] in this range. However, peaks from impurity phase were observed for $x = 0.80$ and 0.90 (see Fig. 1). The (0 0 2n) film peaks are visible until the Sr content reaches $x = 1.10$, but become invisible for $x \geq 1.40$, indicating a full loss of epitaxial growth (see Fig. 1). We point out that although LSCO(x) has attracted huge research interests for years, very few reports [13] have been released on the heavily overdoped LSCO(x), partly because of the instability of the structure due to the loss of oxygen [19]. Heavily overdoped LSCO(x) films of reasonable quality were only reported by reactive coevaporation for $0.00 \leq x \leq 2.00$ [13], and by oxide molecular beam epitaxy for $0.00 \leq x \leq 1.00$ [14]. The x dependence of the c -axis lattice parameter calculated from the data shown in Fig. 1 (see Fig. 3) is consistent with the corresponding results in the two previous studies [13,14], implying that our film quality in the whole x range is

Figure 1 XRD of the LSCO(x) thin films with different nominal Sr contents (x). Peak indices are shown at the bottom and correspond to both film (the left) and LaSrAlO₄ substrate (the right) peaks. The black dots for $x=0.80$ and 0.90 indicate peaks from unspecified impurity phase. No epitaxial peaks were observed for $x=1.40$ and 1.70 , indicating a full loss of epitaxial film structure. The thickness for all the films is around 40 nm.

reasonable. Atomic force microscopy (AFM) images (see Fig. S2) show that the surface morphology is relatively smooth when x is small but becomes rough for the heavily overdoped samples. In short, these data confirm that high-quality LSCO(x) films can be obtained by PLD for $0.00 \leq x \leq 0.60$, while the film quality gradually degrades with further increasing x ($0.80 \leq x \leq 1.10$), and finally the epitaxial crystalline structure is fully lost for $x \geq 1.40$. Here we caution that the nominal x for $x \geq 1.40$ is immaterial because the films are likely not in a single-phase. The use of x in that range is only for simplicity.

Transport properties of single LSCO(x) films

As shown in Fig. 4, the undoped La₂CuO₄ film ($x=0.00$) shows a highly insulating behavior. The $x=0.05$ film still shows an insulating behavior but its overall resistance is much lower than that of $x=0.00$ film. Superconducting behavior was found for the $x=0.10$, 0.15 , and 0.25 samples, and the $x=0.15$ sample has the best superconducting properties. Further increase in doping level changes the

Figure 2 Enlarged view of the XRD data of the LSCO(0.35) film shown in Fig. 1 demonstrates clear finite-thickness oscillations in the vicinity of the (0 0 4) reflection, confirming the high-quality of the film. Similar oscillations were observed for the LSCO(x) films with x varying from 0.00 to 0.60.

Figure 3 Out-of-plane c -axis lattice parameter of films as a function of nominal Sr content (x). Derived from the XRD data shown in Fig. 1. The $x=1.40$, 1.70 , and 2.00 data are regenerated from previous studies as noted. The dashed line indicates the axis c value of (001) LaSrAlO₄ substrate.

transport into a metallic behavior ($0.35 \leq x \leq 0.60$, inset of Fig. 4). These behaviors agree well with the well-known LSCO(x) phase diagram, implying that the real Sr content of our samples is very close to the nominal x value. With further increasing x to above 0.80 , however, the films become insulating again, and the overall resistance increases with x until $x=1.10$. For $x=1.40$ and 1.70 , the films are still highly insulating but show a weaker temperature dependence that might be attributed to the collapse of their crystalline structures. Overall, the single LSCO(x) films prepared by PLD show comparable transport properties with the films prepared by reactive coeva-

Figure 4 The dependence of resistance on temperature for single films of different nominal Sr contents (x). The Sr content x of each film is as labeled in the figure. The top inset shows partial data in linear coordinate, in order to better identify their transport behaviors. The undoped La_2CuO_4 ($x=0.00$) is shown in thicker black symbols. The thickness for all the films is around 40 nm. Double logarithmic coordinates are used for clarity.

poration [13] or oxide molecular beam epitaxy [14].

Transport properties of bilayers

Previously, Gozar *et al.* [1] showed that the interface between the insulating ($x=0.00$) and the metallic ($x=0.45$) $\text{LSCO}(x)$ films was superconducting with high T_c , and the deposition sequence mattered. Here we first show that this seminal work can be well reproduced by PLD deposition, as demonstrated in Fig. 5. In order to explore thoroughly the possible interface superconductivity, we further extensively combined La_2CuO_4 and $\text{LSCO}(x)$, for

$0.05 \leq x \leq 1.70$, with both sequences. Fig. 6 shows the temperature-dependent resistance of bilayers with La_2CuO_4 on the bottom [hereafter labeled as $\text{La}_2\text{CuO}_4 + \text{LSCO}(x)$]; Fig. 7 shows the temperature-dependent resistance of bilayers with $\text{LSCO}(x)$ on the bottom [hereafter labeled as $\text{LSCO}(x) + \text{La}_2\text{CuO}_4$]. In each sequence of bilayers, the thicknesses of the bottom layer and the top layer are around 40 and 10 nm, respectively. The most important observation is that the bilayers exhibit superconductivity for a very wide x range.

The bilayers of both sequences are still insulating for $x=0.05$, and superconducting for $0.10 \leq x \leq 0.25$, similar to the single $\text{LSCO}(x)$ films. For $x \geq 0.35$, the overdoped range, while the single $\text{LSCO}(x)$ films are either metallic ($0.35 \leq x \leq 0.60$) or insulating ($x \geq 0.80$), the bilayers of both sequences are nearly always superconducting, except for an abnormality around $x=0.80$ ($x=0.60$ and 0.80 for $\text{La}_2\text{CuO}_4 + \text{LSCO}(x)$, Fig. 6; $x=0.80$ and 0.90 for $\text{LSCO}(x) + \text{La}_2\text{CuO}_4$, Fig. 7). Therefore, our present experiments indicate that the interface superconductivity is very general in the bilayers of $\text{LSCO}(x)$ and La_2CuO_4 . It is quite striking to see that, although the bilayers are of poor crystalline structure for $x \geq 1.40$ (see Fig. 1 and Fig. S2), they still exhibit good superconducting behaviors, suggesting that the interface superconductivity is a very robust phenomenon and is insensitive to the interface quality. Similar observation was also found recently in a $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3/\text{La}_2\text{CuO}_4$ heterointerface [20].

To compare different samples in a more objective way, we define T_c by the maximal slope of $R(T)$ that can be determined unambiguously as the temperature of the maximum of the derivative curve, $dR(T)/dT$ [11]. In Fig. 8, based on the transport data shown in Figs 6 and 7, and the T_c derived thereafter, we constructed phase diagrams for the bilayers of both sequence. “S”, “M”, and “I” represent the superconducting, metallic, and insulating

Figure 5 The dependence of resistance on temperature for single and bilayer films for $x=0.45$. (a) La_2CuO_4 , (b) $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$, and (c) the bilayer of La_2CuO_4 and $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$. The LaSrAlO_4 is the substrate. In each sequence the thicknesses of the bottom layer and top layer are around 40 and 10 nm, respectively.

Figure 6 The dependence of resistance on temperature for La_2CuO_4 + $\text{LSCO}(x)$ bilayers. The nominal Sr content x of each sample is as labeled in the figure. The top inset shows the schematic of the bilayers. The thicknesses of La_2CuO_4 and $\text{LSCO}(x)$ films are around 40 and 10 nm, respectively. Double logarithmic coordinates are used for clarity.

regions, respectively. “T” represents the region where the transport behavior involves an insulating-to-metallic or metallic-to-insulating transition (see Fig. S3 for an example). The T_c of single $\text{LSCO}(x)$ films (small circles and dashed line) and the bilayer data from the previous studies of Bozovic group [1,11] (the red stars and dots) are also included for comparison. Firstly, one can see that, for each sequence, the T_c for bilayers of high x value is comparable to that of low x value. This result agrees with the previous observation that the T_c of interface superconductivity is less dependent on the doping level of x [11]. Secondly, consistent with the previous study on the bilayers of $x=0.45$ [1], for almost all the x values, the interface superconductivity depends on the sequence, and the overall T_c of $\text{LSCO}(x)+\text{La}_2\text{CuO}_4$ is higher than that of the $\text{La}_2\text{CuO}_4+\text{LSCO}(x)$ [1]. The appearance of these two features suggests that the superconductivity observed in the bilayers of the present study is likely of the same nature as that in the previous studies [1,10,21]. We note that the T_c obtained in our study is comparable or slightly inferior to that of the as-grown bilayers from Bozovic’s group (the red empty stars in Fig. 8), but much less than that of the bilayers that had been optimally annealed in ozone (the red closed star and the red dotted line in Fig. 8b). This difference might originate from oxygen stoichiometry because all the samples used in our studies had not been treated by ozone. In addition, as shown in

Figure 7 The dependence of resistance on temperature for $\text{LSCO}(x)+\text{La}_2\text{CuO}_4$ bilayers. The nominal Sr content x of each sample is as labeled in the figure. The bottom inset shows the schematic of the bilayers. The thicknesses of $\text{LSCO}(x)$ and La_2CuO_4 films are around 40 and 10 nm, respectively. Double logarithmic coordinates are used for clarity.

Figs 4 and 8 (the circles and dashed line), even for the single-phase $\text{LSCO}(x)$, the T_c is lower compared with that grown by oxide MBE, indicating that the film quality of samples prepared by PLD is somehow lower than that of samples prepared by oxide MBE.

DISCUSSION

Previously, two mechanisms, cation interdiffusion and electronic redistribution, have been considered as the possible causes for the high- T_c interface superconductivity, and the electronic redistribution has been suggested to be the dominant one [1,6,10]. Similarly, our present observations can be explained better by electronic redistribution rather than cation interdiffusion. If cation interdiffusion is the dominant mechanism, one would expect that the bilayers for x around 0.80 are superconducting since there is no simple reason that the interdiffusion of Sr should drop at this particular content. In the electronic redistribution scenario, the mobile charge carriers (holes) are depleted from the overdoped $\text{LSCO}(x)$ and accumulated in La_2CuO_4 , according to their chemical potential difference. The chemical potential in $\text{LSCO}(x)$ inferred from X-ray photoemission data decreases with increasing x [22]. This explains the interface electronic redistribution. Unfortunately, the chemical potential data are not available for $x>0.30$. If it continues to decrease with increasing x in the full range up to 1.70,

Figure 8 Phase diagrams for (a) $\text{La}_2\text{CuO}_4+\text{LSCO}(x)$ and (b) $\text{LSCO}(x)+\text{La}_2\text{CuO}_4$ bilayers, derived from the data shown in Figs 6 and 7. T_c is defined as the transition onset that is determined as the position of the maximum of the derivative curve, $dR(T)/dT$. The cyan circles in (a) and the blue squares in (b) represent our experimental data. The empty stars in (a) and (b) represent the corresponding bilayers reported by Bozovic's group [1]; the red closed star in (b) represents the maximum T_c reached by Bozovic's group [1] by thoroughly annealing the bilayer in ozone. The red dotted line in (b) represents the Sr-doping range that has been studied by Bozovic's group [11] and the corresponding T_c . S, M, I and T denote superconducting, metallic, insulating, and transitional between metallic and insulating, respectively. T is exemplified in Fig. S3. For comparison, the T_c vs. x of the single $\text{LSCO}(x)$ films is shown in both panels with small circles and dashed line.

one would again expect that all the bilayers for $x>0.30$ are superconducting due to electronic redistribution, inconsistent with the experimental data. This inconsistency might be reconciled by considering the issue of oxygen non-stoichiometry. It is well known that $\text{LSCO}(x)$ tends to lose oxygen with increasing x [18]. So, the effect of increasing x is two-fold: while the Sr substitution for La can dope holes into the CuO_2 planes and decrease the chemical potential, the increased oxygen vacancies will reduce the amount of holes and increase the chemical potential. In this consideration, the abnormality around $x=0.80$, might be explained by that in this region the

effect of oxygen vacancies surpasses the effect of doping holes. This interpretation is partially supported by that around $x=0.80$, the $R(T)$ curve of $\text{LSCO}(x)$ changes dramatically from metallic to insulating (inset of Fig. 4). An alternative explanation is that the electronic redistribution is induced by oxygen diffusion [23].

Although electronic redistribution is a sound mechanism, it is still quite interesting and puzzling that, for $x\geq 1.00$, the bilayers of both sequences become superconducting again. In this case, the interface superconductivity is actually formed between two insulating copper oxides. One possibility is that in this range the effect of doping holes is stronger than the effect of oxygen vacancies, and thus the corresponding $\text{LSCO}(x)$ layer has a low chemical potential that causes electronic redistribution. Another possibility is that there are inhomogeneous impurity phases that are of low chemical potential, and thus can transfer holes to La_2CuO_4 . In addition, because of the poor crystalline structure in the high x range, the corresponding bilayers might have much severer cation interdiffusion than that observed in the previous studies [1,7]. At present, we are unable to distinguish all these possibilities yet. However, no matter what is the real underneath mechanism, it is clearly that the interface superconductivity is very general and not restricted to perfect crystalline structure. This point could be important for practical applications of interface superconductivity because it lowers the threshold to fabricate such devices.

Finally, we comment that there is actually one more intriguing possibility for the superconducting phase observed in the high x range, that is, the superconductivity in the heavily over-doped cuprate. A few recent experimental [24,25] and theoretical [26,27] studies demonstrated that exotic superconductivity can exist in heavily overdoped cuprates. One case is the nodeless pairing in superconducting CuO_2 monolayer grown on $\text{Ba}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ [24,26]. Another case is the superconductivity in $\text{Ba}_2\text{CuO}_{4-y}$ in which the octahedron was found in an exceptionally compressed version [25,27]. Interestingly, as shown in Fig. 3, the c -axis lattice constant of $\text{LSCO}(x)$ indeed decreases with increasing x in the heavily overdoped range. Its value becomes close to that of $\text{Ba}_2\text{CuO}_{4-y}$ (the red star in Fig. 3) for $x>1.0$, suggesting that in this range the octahedral might be compressed as well. Further studies are needed to examine this possibility.

CONCLUSIONS

In summary, we have demonstrated for the first time the

high- T_c interface superconductivity in the bilayers of La_2CuO_4 and overdoped $\text{LSCO}(x)$ by PLD. The nominal Sr content x has been significantly extended from the previous studies. Our result demonstrates that the superconductivity at this interface is robust and general. Since PLD is versatile in growing a large number of materials with reasonable quality, it provides the opportunity to combine more materials efficiently and thus may serve as a good tool to search for superconductivity in new interfaces.

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Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information The data of XRR and AFM, and an example for defining “T”, are available in the online version of the paper.

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脉冲激光沉积法制备双层结构铜氧化物界面高温超导

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摘要 在2008年发表的一篇经典工作中, Gozar等人报道了由绝缘性的 La_2CuO_4 和金属性的 $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$ 双层薄膜构成的体系存在界面超导. 一个重要的有待回答的问题是界面超导是否强健以及具有普遍性. 在Gozar等人的工作中, 铜氧化物双层结构是利用特制的氧化物分子束外延设备生长的, 最大Sr掺杂量仅为0.47. 在本工作中, 我们首次利用脉冲激光沉积法制备了铜氧化物双层结构, 并重复出了上述界面超导工作. 在此基础上, 我们将Sr的掺杂范围大幅扩展到1.70, 结果表明在由 La_2CuO_4 和过掺杂的 $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ 构成的双层结构中界面超导非常强健和普遍. 值得一提的是, 我们发现在 $x>0.8$ 范围内存在一个新的界面超导区间.