Electronic reconstruction and enhanced superconductivity at La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$/La$_{1.55}$Sr$_{0.45}$CuO$_4$ bilayer interface

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Abstract – We report enhanced superconductivity in bilayer thin films consisting of superconducting La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ with $0.06 \leq x < 0.20$ and metallic but non-superconducting La$_{1.55}$Sr$_{0.45}$CuO$_4$. These bilayers show a maximum increase in superconducting transition temperature ($T_c$) of more than 200% for $x = 0.06$, while no change in $T_c$ is observed for the bilayers with $x > 0.20$. The analysis of the critical current and kinetic inductance data suggests 2–3 unit cells thick interfacial layer electronically perturbed to have a higher $T_c$. A simple charge transfer model with cation intermixing explains the observed $T_c$ in bilayers. Still the unusually large thickness of interfacial superconducting layers cannot be explained in terms of this model. We believe that the stripe relaxation as well as the proximity effect also influence the superconductivity of the interface.

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Introduction. – A number of recent experiments and theories have provided a new insight into remarkably different electronic properties of the interfaces between two correlated electron systems as compared to those of the parent compounds in their bulk form [1]. One of the unusual realizations of these effects is the superconductivity (SC) seen at the interface of a certain class of oxides [2-6]. A recent experiment by Gozar et al. shows the existence of superconductivity at the interface of a bilayer composed of La$_2$CuO$_4$ and La$_{1.55}$Sr$_{0.45}$CuO$_4$ films, with neither of them being a superconductor on its own [2]. Moreover, an enhancement of the superconducting transition temperature ($T_c$) has been reported for La$_{2-x}$Sr$_x$CuO$_4$ and La$_{1.18}$Sr$_{0.82}$CuO$_4$ thin films when capped by an overdoped metallic La$_{1.65}$Sr$_{0.35}$CuO$_4$ [3]. From a theoretical point of view, these results have been interpreted as a consequence of strong pairing interaction of the UD layer on the large phase stiffness present in the OD layer, which results in the formation of a more robust superconducting phase in the OD layer [10]. Also the theoretical approach which considers the delocalization of carriers at the interfaces due to the electrostatic potentials seems to be in agreement with many experimental results [11].

The Nd doping in La$_{2-x}$Sr$_x$CuO$_4$ introduces static charge stripe order and associated spin stripes, which appear to coexist with SC at very low temperatures [12]. However, in thin epitaxial films of these compounds, the lattice mismatch-induced strain can strengthen or weaken the stripe order [6,13,14]. In the case of films deposited on SrLaAlO$_4$ (SLAO), a weakening of the stripe pinning potential and thus enhanced stripe fluctuations leading to a $T_c$ larger than the value for bulk samples has been reported [6,13]. The overdoped La$_{2-x}$Sr$_x$CuO$_4$ is a conventional Fermi liquid in which the dynamic spin susceptibility scales with $T_c$ and eventually disappears at large $x$ where it ceases to be a superconductor [15]. Furthermore, it is believed that the metallic nature can originate due to the transverse fluctuation...
of the charged stripes in such compounds [16,17]. We have fabricated the bilayer thin films of superconducting La$_{1.55}$Sr$_{0.45}$CuO$_4$ (LSCO) and metallic but non-superconducting La$_{1.55}$Sr$_{0.45}$CuO$_4$ (LSCO). This system allows us to investigate how different pairing scales present in these two compounds as well as the potential imbalance across the interface modify the electronic properties of the interface. Moreover, it is still unknown how the overdoped material will influence the stripe order present in the Nd-doped compound when an interface of these two materials is created. Our study shows a large enhancement of $T_c$ (up to 18.5 K) in the bilayers from its value in the case of the bare Nd-doped films. Our analysis of the data strongly indicates that the enhanced $T_c$ is due to the presence of an interfacial superconducting layer of few unit cells thickness in the bilayer.

**Experimental details.** – The epitaxial thin films of LSCO and LNSCO along with LSCO/LNSCO bilayer as sketched in fig. 1(e) were fabricated on (001) oriented SLAO substrates by pulsed laser deposition at 800°C in 230 mTorr oxygen pressure. The samples were cooled in atmospheric pressure of oxygen to room temperature with one-hour annealing at 500°C to realize full oxygenation of the structure. The single layer films and each component of the bilayers were 50 nm thick. The present study includes a series of LNSCO and bilayer films with $x = 0.06, 0.12, 0.16, 0.20, \text{ and } 0.25$. The crystallographic structure and interface quality of the films were characterized in detail using X-ray diffraction (XRD) and atomic force microscopy (AFM). The 2θ-XRD scans clearly revealed the presence of both LSCO and LNSCO layers in the bilayer. In combination with ω and φ scans, we have established a high-quality epitaxial growth of these structures with c-axis along the normal to the plane of the substrate. The low-angle oscillations seen in X-ray reflectivity curves are used to determine the thickness of the films which are consistent with the nominal film thickness as determined from the growth rate. The AFM scans over an area of $2 \times 2 \mu \text{m}^2$ showed a typical root-mean-square roughness of 1.3 nm, which is about one unit cell thick ($c \approx 1.3 \text{ nm}$). The fitting to reflectivity curve using a genetic algorithm yields the interface roughness of (1.4 ± 0.1) nm and the surface roughness of (1.7 ± 0.1) nm [18]. The in-plane transport measurements were performed in four-probe geometry. The ac screening measurements were carried out with a two-coil mutual inductance method [19].

**Results and discussion.** – Figures 1(a)–(e) show the in-plane resistivity $\rho(T)$ of LNSCO/LSCO bilayer along with the $\rho(T)$ of LNSCO monolayer films for different values of $x$. We observe distinctly higher $T_c$ for the bilayers with $x = 0.06, 0.12, \text{ and } 0.16$ as compared to that for the corresponding bare LNSCO film (see fig. 1(a)–(c)). The maximum $T_c$ enhancement of $\approx 18.5 \text{ K}$ is observed for the bilayer with $x = 0.06$. Here $T_c$ is defined as the onset temperature of the superconducting transition. For the bilayers with $x = 0.20$ and 0.25, the LSCO capping on the LNSCO films does not affect the $T_c$ as seen in fig. 1(d), (e). Another interesting feature of our data is that the superconductivity in the bilayers can exist at a much higher temperature than in any LNSCO monolayer film (see fig. 2(a)). Although such kind of enhancement has been reported in LSCO based bilayers [5], the $T_c$ enhancement reported here is quite large (≈ 210%). The observed enhancement of $T_c$ in bilayers is not sensitive to the thickness of the individual layers or to the sequence of the layers. This suggests that the observed phenomenon is purely an interface effect. However, a natural question that emerges out is that concerning the thickness of the interface region that is being perturbed to have an enhanced $T_c$. We have...
Fig. 2: (Color online) (a) The $T_c$ values as a function of Sr doping level ($x$) for LNSCO film (circle), LSCO/LNSCO bilayer (square), bulk LNSCO [20] (dashed line) and the prediction for bilayer using eq. (1) (solid line). The $T_c$’s of LNSCO films are higher than that of their bulk counterparts as the films are under the compressive strain. (b) The critical current $I_c(T)$ for LNSCO (open circle) and bilayer films (filled circle) for $x = 0.12$. The contribution of the interfacial layer (square), extracted from the $I_c(T)$ data, is shown with its linear fit (dotted line).

Fig. 3: (Color online) (a) The Re $V(T)$ for LNSCO($x = 0.12$)/LSCO bilayer with the Gaussian fits showing loss peaks due to the interfacial layer (green line) and LNSCO layer (red line). The inset shows the magnified view of the smaller peak located at $\approx 13.7\, K$. (b) The Im $V$ vs. $T$ curve for the bilayer with the contributions from the interfacial layer (green) and LNSCO layer (red). The inset shows the $L_k^{-1}(T)$ for the interfacial layer (multiplied by 5) and LNSCO layer. The blue dashed lines show the extrapolation of $L_k^{-1}(T)$ towards $T = 0\, K$.

Here we have assumed that both superconducting layers have same critical current density and are confined within the LNSCO side of bilayer. We obtain similar values (2–3 unit cells) of the interfacial layer thickness for the bilayers with $x < 0.20$. On the other hand, the $I_c(T)$ data for both LNSCO and bilayer films are almost identical for $x = 0.20$ and 0.25. This indicates that the superconductivity in these bilayers is due to only LNSCO layer.

A clear and direct evidence for the enhancement of $T_c$ is given by the ac screening measurements of the bilayer and Nd-doped films. The screening voltage for the bilayer with $x = 0.12$ is shown in fig. 3. We observe two distinct loss peaks in the real part of the pick-up coil voltage ($V$) for the bilayer. A smaller peak is located at $\approx 13.7\, K$ in addition to the prominent loss peak at 10.3\, K. While latter is due to the superconducting transition of LNSCO layer, as confirmed by the presence of a loss peak occurring at the same temperature in a plain LNSCO film (not shown here), the small high temperature peak can be assigned as the superconducting transition of the interfacial layer. Similarly, the deconvolution of Im $V$ data (fig. 3(b)) of the bilayer shows two contributions corresponding to the LNSCO layer and the interfacial layer. We want to point out the fact that the enhancement of $T_c$ determined using the screening or $I_c(T)$ data is small compared to the enhancement of the onset temperature of the resistive transition. We have extracted the inverse sheet kinetic inductance $L_k^{-1}(T)$ of the superconducting condensate for each component from the deconvoluted data [6] and then have calculated the Kosterlitz-Thouless-Berezinskii transition temperature ($T_{KTB}$) of the quasi–two-dimensional

estimated the thickness from the temperature dependence of critical current $I_c(T)$ data. Figure 2(b) shows the typical $I_c(T)$ curves for both monolayer and bilayer film with $x = 0.12$. In the latter case, the $I_c(T)$ for $T \leq 9.0\, K$ draws contributions from both the LNSCO layer which has a $T_c$ of \approx 9.0\, K and the interfacial layer with $T_c \approx 14.0\, K$ while, for the temperature between 9.0\, K and 14.0\, K, the $I_c$ is only because of the interface. We have assumed the bilayer to be a combination of two parallel superconducting layers of well-defined thicknesses (say, $d_{\text{LNSCO}}$ and $d_{\text{Interface}}$) and $T_c$’s. The ratio between the critical currents of LNSCO and interfacial layer at $T = 0\, K$ is $I_c/\text{LNSCO}(0)/I_c/\text{Interface}(0) = d_{\text{LNSCO}}/d_{\text{Interface}}$. Since $d_{\text{LNSCO}} + d_{\text{Interface}} \approx 100\, nm$ for our bilayer, the thickness of the interfacial layer is \approx 2.70\, nm or 2 unit cells.
interface layer. The $T_{KTB}$, given by the expression
$$L_k^{-1}(T_{KTB}) = (8\pi k_B/\Phi_0^2)T_{KTB},$$
where $\Phi_0$ is the flux quantum, lies close to the $T_c$. This observation further
suggests the two-dimensional (2D) nature of the interface superconducting layer. In a 2D superconductor,
$L_k^{-1} \propto n_s d$, where $n_s$ is the superfluid density and $d$ is the layer thickness. Thus
$L_k^{LSCO} / L_k^{interfac} = d_{LSCO} / d_{Interface}$ under the assumption of a uniform
superfluid density in the bilayer. A rough estimate of $d_{interface}$ can be obtained considering the two-
superconducting-component model for the bilayer, as discussed before. Using the extrapolated values of $L_k^{-1}$ at 0 K for both the LNSCO and the interface layer (see the inset of fig. 3(b)), we have extracted $d_{interface}$ as 
approximately equal to the interface

A simple yet satisfactory explanation of enhancement in $T_c$ can be provided by taking into account the charge transfer as well as the diffusion of the ions at the interface. Firstly, we consider the latter phenomenon. The individual entities of the bilayers contain two types of dopants, viz. Sr$^{2+}$ and Nd$^{3+}$. The concentration of these ions on both sides of the interface is different and this inequality of the concentration can trigger the physical motion of the ions from higher to lower density side. Such effect ensures a smeared ion profile instead of a sharp one [2,3]. The ion profiles reported in refs. [2,3] follow an empirical relation for the ion density at $j$-th plane: 
$$n_j = n_1 + \Delta n e^{-j/\lambda},$$
where $\Delta n = n_1 - n_2$ with $n_1$ and $n_2$ as the densities on both sides of the interface ($n_1 > n_2$) and $\lambda$ is the ion diffusion length. Here $j \geq 0$ and $j = 0$ represents the starting point of the decaying ion profile from a level of $n_1$ and $j > 0$. The fits yield the value of $\lambda$ approximately equal to the interface roughness observed in these heterostructures. In our study, we have assumed that $\lambda = 1$ unit cell, which is the roughness value for our case. Using the above-mentioned relation, we have simulated the ion profile due to inter-diffusion from the nominal ion densities. Figures 4(a), (b) show the diffused ion profile for Nd$^{3+}$ and Sr$^{2+}$ in LNSCO ($x = 0.12$)/LSCO bilayer, respectively. Next we need to examine the charge transfer occurring at the interface.
The replacement of a La\(^{+3}\) by Sr\(^{+2}\) will introduce a hole in the CuO\(_2\) plane while the Nd\(^{+3}\) doping does not create any extra charge. Thus, there will be a varying charge (hole) profile created due to inhomogeneous Sr\(^{+2}\) doping across the interface (shown by open circles in fig. 4(b)). As a result, a non-uniform electric potential landscape is formed across the interface, which leads to a redistribution of electronic charges in different oxide planes. This has been modeled using Poisson’s equation in discrete form [11]. According to this model, the modified hole density \(p_i\) at any CuO\(_2\) plane (say, the \(i\)-th one) with initial hole density \(x_i\) is given by the relation [11]

\[ p_{i+1} + p_{i-1} - (2 + \alpha) p_i = -\alpha x_i \]

where \(\alpha\) represents the degree of charge localization in the system. We have performed the numerical simulation of above recurrence relation to obtain the actual hole doping profile at different CuO\(_2\) planes near the bilayer interface as shown in fig. 4(b).\(^1\)

Figure 4(c) shows the actual doping levels in various bilayers after taking into account cation intermixing and charge redistribution near the interface as mentioned above. Clearly we can see the oxide planes with enhanced \(T_c\) for the bilayers with \(x = 0.06, 0.12\) and 0.16 while the \(T_c\) remains unchanged for other bilayers. The \(T_c\) profile for LNSCO(\(x = 0.12\))/LSCO bilayer near the interface (inset of fig. 4(c)) shows a \(T_c\) of 24.7 K in the second LNSCO plane (\(i = -2\)) from the interface, which is close to the observed \(T_c\) in the bilayer. Thus, we may conclude that the enhanced SC is due to the second CuO\(_2\) plane from the LNSCO/LSCO interface on the LNSCO side. Similarly the \(i = -2\) layer is responsible for the enhanced \(T_c\) in the bilayer with \(x = 0.20\), while the \(i = -1\) layer for \(x = 0.06\). In the case of \(x = 0.20\) and 0.25, the SC is from the bulk part of LNSCO layer ( \(i\) values near \(-77\). The calculated values of \(T_c\) matches quite well with the experimental values as shown in fig. 2(a).

We have gone ahead to generate a generic map of \(T_c\) as a function of Sr doping \((x)\) and Nd doping \((y)\) for La\(_{2-x-y}\)Sr\(_x\)Nd\(_y\)CuO\(_4\)/La\(_{1.55}\)Sr\(_{0.45}\)CuO\(_4\) bilayer systems as shown in fig. 5. Only for a small portion of doping values with \(x < 0.005\) and \(y > 0.37\) (top left corner of fig. 5), a OD layer, namely \(i = +1\), contributes to enhanced SC. As we move away from this region, the enhanced SC layer goes deeper into LNSCO layer. After a certain level, the enhanced SC is lost and only bulk SC of LNSCO layer remains. Interestingly the CuO\(_2\) plane with highest \(T_c\) lies on the UD side for the majority of the phase space contrary to the other major explanation for \(T_c\) enhancement, viz. the phase fluctuation model, where the enhanced SC should lie on OD side [10].

Although the above-mentioned model satisfactorily explains the \(T_c\) enhancement seen in our bilayers, it also predicts that the enhanced SC is only confined within one CuO\(_2\) plane (or at most two). On the contrary, we observe an anomalously thick (2–3 unit cells) interfacial layer. We can notice that, near the layer with maximum \(T_c\), i.e., \(i = -2\), there are few other \(T_c\) values of 20.2 K and 15.6 K for \(i = -1\) and \(-3\) planes, respectively, as shown in the inset of fig. 4(c). Despite the fact that these values are lower than the eventual \(T_c\) of the bilayer (or \(i = -2\) layer), they are still larger than that of the bare LNSCO film. One would expect to see the presence of these superconducting planes as additional contributions in screening voltages. But instead we only observe the effect of the bulk LNSCO layer and the interfacial layer. We speculate that the \(T_{c}(s)\) of few layers close to the \(i = -2\) plane are also enhanced and thus an enhanced SC layer of few unit cells is formed. A potential explanation of this phenomenon involves the stripe fluctuations in the LNSCO layer. As shown in fig. 1, the lightly doped LNSCO films show an upturn in resistance at lower temperatures, which follows a log(1/\(T\))-dependence. The logarithmic insulating nature has been observed due to stripe ordering in La\(_{2-x-y}\)Nd\(_x\)Sr\(_2\)CuO\(_4\) [24]. With increasing doping level, the stripe fluctuation increases and ultimately two-dimensional metal is formed at overdoped side [13,17]. The presence of dynamically fluctuating stripes in LSCO can weaken the stripe order in few layers of LNSCO near the interface, if not more, and thus promotes enhancement in \(T_c\). Another

\(^1\)The numerical simulations are done with the value of \(i\) ranging from \(-77\) to \(77\), corresponding to 50 nm of LNSCO and 50 nm of LSCO layer thickness so that \(i < 0\) represents the CuO\(_2\) planes in LNSCO side, while \(i > 0\) in LSCO side. We have used an empirical value of \(\alpha = 0.5\). A finite stack of CuO\(_2\) layers in bilayers introduces the boundary conditions: \(x_i = 0 = p_i\) for \(i = \pm 78\) (substrate side) and \(78\) (free space side).
possibility is the proximity effect due to which the SC can be enhanced in few more interfacial layers. Gariglio et al. have suspected the existence of the proximity effect in La_{1.6-x}Nd_{x}Sr_{0.4}CuO_{4}/La_{2}CuO_{4} bilayer [25]. The superconductivity in the bilayer lies only at the second CuO_{2} plane in La_{2}CuO_{4} from the interface [3]. The Zn doping in this plane suppressed the $T_{c}$ considerably as expected. Surprisingly, the doping in few neighboring non-superconducting planes reduced the $T_{c}$ albeit by a smaller amount. This suggests some kind of coupling between the planes present near the superconducting plane.

**Conclusion.** — In summary, the bilayers consisting of superconducting La_{1.6-x}Nd_{0.4}Sr_{0.4}CuO_{4} capped by the overdoped metallic La_{1.55}Sr_{0.45}CuO_{4} have a superconducting transition at a higher temperature as compared to the $T_{c}$ of bare LNSCO films for $x < 0.20$. The ac screening measurements show two distinct superconducting transitions in such bilayers with the higher-$T_{c}$ phase corresponding to the interfacial layer. The maximum $T_{c}$ enhancement of $\approx 18.5$ K (or 210%) is seen for the bilayer with $x = 0.06$. On the contrary the $T_{c}$ of the bilayers with $x > 0.20$ remains unchanged. The enhancement of superconductivity is confined to $\approx 2$-3 unit cells thick interfacial layer, as evidenced by the temperature-dependent critical current and ac screening data. The enhancement observed in these bilayers can be explained by a model considering the charge redistribution due to the potential differences and the cation inter-diffusion across the interface. Still the reason of unusually large interfacial layer thickness is unclear. The relaxation of stripe order in LNSCO and the proximity effect are crucial factors for further understanding of the interface superconductivity in cuprate heterostructures.

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