Magnetotransport study on $La_{0.5}Sr_{0.5}CoO_{3-\delta}$/$YBa_2Cu_3O_{7-\delta}$/$La_{0.7}Ca_{0.3}MnO_{3-\delta}$ trilayer system

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Epitaxial ferromagnetic ($La_{0.5}Sr_{0.5}CoO_{3-\delta}$)/superconducting ($YBa_2Cu_3O_{7-\delta}$)/ferromagnetic ($La_{0.7}Ca_{0.3}MnO_{3-\delta}$) trilayers are grown by pulsed laser deposition. The magnetotransport studies reveal that a much lower magnetic field is required to suppress the superconductivity in the trilayer. Moreover, the superconducting T_c is markedly different for the trilayer system when the magnetic field is applied parallel to the c axis compared to that perpendicular to the c axis ($T_{c\parallel c\text{ axis}} < T_{c\perp c\text{ axis}}$). The trilayer system also exhibits a huge +*ve* magnetoresistance below superconducting T_c, which could arise due to vortex dissipation in the liquid state of a superconductor in the trilayer structure.

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Heterostructures composed of ferromagnetic (F) and superconducting (S) materials have attracted a lot of theoretical and experimental attention due to the rich physics produced by competing effects of superconductivity and magnetism (two antagonistic quantum phenomena) especially when the thickness of individual layers are on the scale of their characteristic lengths.[1] The mutual interaction between F and S materials give rise to some new exciting physical effects such as shift of superconducting transition temperature[2] (T_c), dramatic shrinkage of ferromagnetic domains and domain walls below superconducting T_c,[3] and the existence of π-phase coupling.[4,5] The interplay between superconductivity and spin polarized systems also has potential application in the emerging field of spintronics.[6,7] Recently, an excess of 1000% magnetoresistance has been reported in a F/S/F trilayer below the superconducting T_c.[8]

In high T_c superconductors (HTSs), the vortex motion and the energy dissipation in mixed state have gathered enough fundamental and technological importance. The reason for this interest is the existence of several vortex phases in the H-T phase diagram which are separated by different kinds of phase transitions. At higher temperatures, the vortices remain in a fluid state until they decay above H_{c2} (upper critical field). Extensive works have been performed in this regard in single crystals compared to thin films. Thin films are particularly interesting systems to study the effect of disorder on the vortex phase diagram. One of the interesting issue is also to look at the behavior and dynamics of vortices in the presence of ferromagnetic layers in F/S hybrid structures. In this connection, the multilayers composed of S/F materials are also ideal candidates to study the vortex dynamics[9,10] in superconductors as well as many peculiar properties of HTSs.

In addition to vortex dynamics, interaction across a S/F interface between Cooper pairs and spin polarized electrons could lead to changes in the properties of both materials within the proximity length on either side of the interface. At the F/S interface, when Cooper pairs enter the ferromagnet from the superconductor, they experience an exchange interaction, which favors spin polarization. This causes the superconducting order parameter to decay faster in the F layer than in normal metal, within a length scale $\xi_F = \hbar v_F / \Delta E_{ex}$, where v_F is the Fermi velocity and ΔE_{ex} is the exchange splitting.[11] In typical ferromagnets, where $\Delta E_{ex} = 1-3$ eV, the ξ_F is <1 nm, which is much smaller than the superconducting coherence length ($\geqslant 10$ nm) for low temperature superconductors. On the contrary, HTS/ferromagnet contacts can show pronounced proximity effect as the HTS has a shorter coherence length (~ 0.2 nm).[12] The superconductivity is also suppressed near the interface of the superconductor within a characteristic length scale ξ_S given by $(\hbar D_S / 2k_B T_c)^{0.5}$, where D_S is the electron diffusion coefficient for the superconductor[13] and ξ_S is of the order of the superconductor coherence length. In this regard, the ferromagnetic perosvkite oxide/HTS systems are particularly interesting because of their ability to form high quality multilayers.[14–20]

Another interesting aspect which has not been investigated thoroughly is the effect of ferromagnetic layers on the critical field of superconductor in F/S heterostructures. Theoretically, it has been predicted that the effect of I_m (mean field exchange potential of a magnetic layer) on a superconductor in a F/S supperlattice can reduce the critical field.[21] This becomes more interesting because of the unusual electronic properties of the superconductor unlike the normal Fermi liquid. In order to investigate the unusual electronic properties and the mechanism of high T_c superconductivity, one needs to investigate the normal state properties at very low temperature, i.e., $T \ll T_c$. In the case of $YBa_2Cu_3O_7$ (YBCO), we need to apply a magnetic field above the upper critical field of this material, which is quiet high, to make a transition from the superconducting state to the normal state. It has to be noted that one requires a magnetic field of the order of 240 T applied $\parallel$ to CuO_2 plane or 120 T applied $\perp$ to the CuO_2 plane to destroy the superconductivity in an optimally doped YBCO sample.[22] This high magnetic field requirement occurs as the YBCO has shorter coherence length ($H_{c2} = \Phi_0 / 2\pi\xi^2$) and since $\xi_{ab} > \xi_c$, $H_{c2\parallel ab} > H_{c2\parallel c}$. So it is very hard to explore the normal state properties of high T_c superconductors (cuprates) at $T \ll T_c$, since the upper critical field of these materials is very high and becomes intangible using laboratory magnets. If the F/S heterostructure could reduce the critical field associated with the superconducting layer in the heterostructure by a considerable amount, then it could be a suitable candidate for such studies. In order to understand the effect of a magnetic layer on the critical field and vortex dynamics of a superconductor,

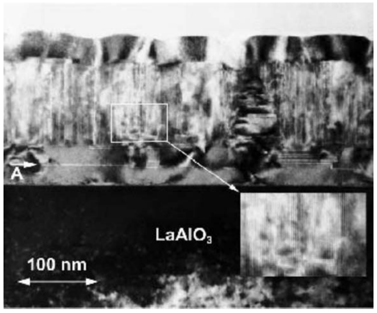

FIG. 1. Cross-sectional TEM image of the LSCO (45 nm)/YBCO (100 nm)/LCMO (30 nm) trilayer structure.

we have carried out experiments on a trilayer system where the superconducting layer is sandwiched between two ferromagnetic layers.

In this work, we investigate the magnetotransport properties of the $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ (LSCO)/$YBa_2Cu_3O_{7-\delta}$ (YBCO)/$La_{0.7}Ca_{0.3}MnO_{3-\delta}$ (LCMO) trilayer structures. We find that a threshold thickness of YBCO is required for the onset of superconductivity in the trilayer structure. We have also noticed that a strikingly low magnetic field is required for the suppression of the superconductivity of YBCO in the trilayer structure. Interestingly, the shift in T_c for the trilayer due to the application of the magnetic field is anisotropic.

The F/S/F trilayers and F/S bilayer are grown on $LaAlO_3$ single crystal substrates by pulsed laser ablation of ceramic targets at a frequency of 5 Hz and with a fluence of 5 J/cm^2. All the ceramic targets are prepared by the solid state reaction method. The F(LSCO), S(YBCO), and F(LCMO) layers in the trilayer structures are grown by sequential deposition at 0.2 mbar of pure oxygen pressure and at temperatures of 765, 785, and 755 °C, respectively. The thickness of YBCO and LCMO is varied by keeping a constant LSCO bottom layer thickness. Electrical and magnetotransport properties are measured using a standard four-probe configuration in a magnetic field strength up to 11 T and a temperature down to 4.2 K.

In order to confirm the layered structure, we have performed cross-sectional transmission electron microscopy (TEM) studies. Figure 1 shows a cross-sectional TEM image of a LSCO (45 nm)/YBCO (100 nm)/LCMO (30 nm) trilayer structure grown on a single crystalline $LaAlO_3$ substrate. The region of the YBCO layer, marked by a rectangle in Fig. 1, is magnified by a factor of 2 as an inset, clearly shows vertically running (001) planes with a distance of 1.2 nm, i.e., c-axis lattice parameter. This reveals that the YBCO layer has the [001] axis in-plane, i.e., it is a-b oriented. We believe that the bottom layer LSCO is responsible for the a-b orientation growth of the YBCO layer.

Figure 2 shows the zero field normalized resistance versus temperature curves of LSCO/YBCO/LCMO trilayers for different thicknesses of the YBCO and LCMO layers. We find that the superconducting T_c of YBCO in the trilayers is lower than the bulk superconducting T_c of YBCO. The possible explanations for such lowering of superconducting T_c in the trilayer structures are explained in the later part of our discussion. Again it is very clear from Fig. 2 that no onset of superconducting T_c is found down to 4.2 K for LYL1, which has the smallest thickness of YBCO among all the trilayer structures. It has been reported[23] that the lowering of superconducting T_c becomes less with the decrease in magnetic layer thickness in the F/S heterostructure. Even though the top ferromagnetic layer in LYL1 has the lowest thickness, still no onset of superconductivity is found in it. So, we find that a threshold YBCO thickness is required for the onset of superconductivity in trilayer structures and that the onset of superconducting T_c increases with the increase in YBCO thickness.

Figure 3 shows the in-plane, temperature dependent normalized resistance in zero field and in the presence of an

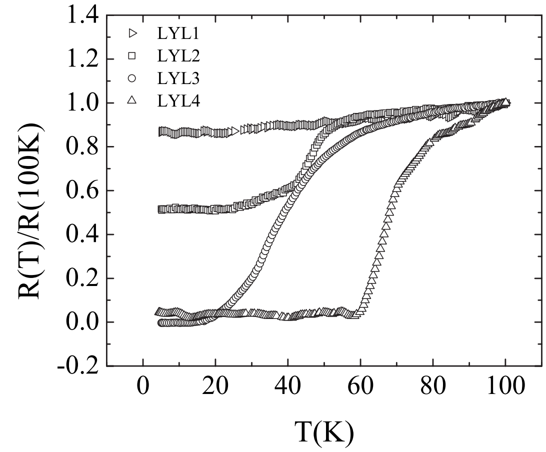

FIG. 2. In-plane temperature dependent zero field normalized resistance of trilayers with different thicknesses. The individual layer thickness in the trilayers is given inside brackets, [LYL1: LSCO(45 nm)/YBCO(25 nm)/LCMO(8 nm), LYL2: LSCO(45 nm)/YBCO(50 nm)/LCMO(15 nm), LYL3: LSCO(45 nm)/YBCO(100 nm)/LCMO(30 nm), and LYL4: LSCO(45 nm)/YBCO(150 nm)/LCMO(15 nm)].

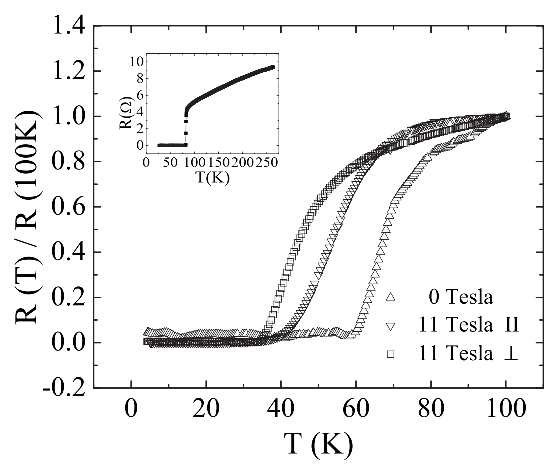

FIG. 3. In-plane temperature dependent normalized resistance of the $La_{0.5}Sr_{0.5}CoO_{3-\delta}$/$YBa_2Cu_3O_{7-\delta}$/$La_{0.7}Ca_{0.3}MnO_{3-\delta}$ (LYL4) trilayer in the presence of 0 and 11 T magnetic field (applied parallel and perpendicular to the sample surface). The inset shows the resistance versus temperature curve in zero field for $La_{0.5}Sr_{0.5}CoO_{3-\delta}$/$YBa_2Cu_3O_{7-\delta}$ bilayer.

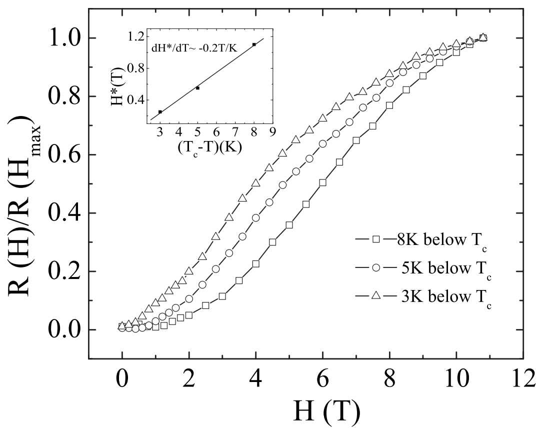

FIG. 4. Normalized resistance of the LYL4 trilayer as a function of applied magnetic field at three temperatures below the superconducting T_c. The inset shows the slope in the H^*-T phase diagram in the vicinity of T_c.

applied field for the LYL4 trilayer system, which has the highest T_c among all the samples that we have investigated. We find the onset of superconducting T_c in zero field for the trilayer system to be ~70 K. Note from Fig. 3 that when we apply a field of 11 T perpendicular to the plane of the sample (i.e., perpendicular to the c axis of YBCO in the trilayer structure), the superconducting zero resistance T_c of YBCO gets reduced by ~20 K, whereas when we apply the same field parallel to the sample plane (i.e., parallel to the c axis of YBCO in the trilayer structure), the superconducting zero resistance T_c of YBCO gets reduced by ~15 K. This allows us to study the vortex motion caused by the applied magnetic field, which is reflected as the appearance of a small resistance in the mixed state of YBCO in the trilayer structure. Thus, one can approximately estimate the magnetic field required to suppress the superconducting T_c (dH/dT) in the vicinity of T_c to be <1 T/K for either orientation of the field with respect to the c axis. Nevertheless, it is very clear from the above discussion that $(dH/dT)_{H\|c\text{-axis}} < (dH/dT)_{H\perp c\text{-axis}}$ in the vicinity of T_c. An exact estimation for the value of dH/dT in the vicinity of T_c has been provided in Fig. 4.

This strikingly lower value of dH/dT near T_c for YBCO in the trilayer cannot be attributed due to oxygen deficiency (underdoping) of the YBCO layer. This is because strong evidence have been provided recently by observing the temperature dependence of H_c' (the magnetic field at which the normal state transport is fully restored) in $YBa_2Cu_3O_{6.6}$ crystals.[24] They find that even when the T_c of $YBa_2Cu_3O_{6.6}$ is reduced down to 3.5 K from 60 K by electron irradiation, the value of H_c' is only reduced by a factor of 2. This experimental result clearly reveals that though the T_c changes drastically, the variation of critical field is moderate. Hence, even for underdoped samples which have relatively low T_c, we require a very high field to study the normal state properties at $T \ll T_c$. In this regard, the trilayer structure seems to be more efficient since the destruction of superconductivity in it is very intense, which could be due to the presence of magnetic layers. Hence, these studies become feasible using laboratory magnets.

It is very interesting to see from Fig. 3 that when the magnetic field of 11 T is applied parallel to the sample plane (i.e., parallel to the c axis of YBCO in the trilayer), the T_c is shifted by ~15 K, whereas when the magnetic field is applied perpendicular to the c axis, the T_c is shifted by ~20 K. So, it clearly reveals that the T_c shift is less when the magnetic field is applied along the c axis compared to the T_c shift when the magnetic field is applied perpendicular to the c axis of YBCO in the trilayer. This is quite different from what is observed in conventional single layer YBCO thin films as well as single crystals.[25,26] At this point, a more detailed study is required to reveal the underlying physics behind such peculiar observation. Note that in both orientations of the trilayer structure, the superconductivity is suppressed intensely.

The decrease in superconducting T_c in trilayers could be assigned due to various reasons. First, the YBCO layer might not have attained enough oxygen to restore its original bulk stoichometry since annealing in oxygen background for the trilayer is done only after the LCMO layer is grown on top of the YBCO layer. Second, the injection of spin polarized carriers from the top of the LCMO layer into the YBCO layer leads to the breaking of Cooper pairs inside the superconductor. The spin polarized current driven from a ferromagnet to a superconductor gives rise to a nonequilibrium spin density in the superconductor. This nonequilibrium spin density causes a difference between the chemical potential for spin up and spin down electrons in the superconductor, which leads to the suppression of superconducting order parameter by the pair breaking effect.[27] This pair breaking effect extends over the spin diffusion length (l_s) in the superconductor. The spin diffusion length in YBCO can be known by using the relation $l_s = (l_o v_F \tau_s)^{0.5}$ where τ_s is the spin polarized electron diffusion time, v_F is the Fermi velocity, and l_o is the electron mean free path.[28] Assuming proper values of l_o, v_F, and τ_s, it can be shown that l_s extends up to a few nanometers in YBCO. This phenomenon can be written as[29] $\Delta(n_{qp})/\Delta(0) \approx 1 - [2n_{qp}/4N(0)\Delta(0)]$, where $\Delta(n_{qp})$ is the energy required to suppress the order parameter of a superconductor due to the density of spin polarized quasiparticles n_{qp}. $N(0)$ and $\Delta(0)$ give the density of states and order parameter at $T=0$ K, respectively. The pair breaking due to the injection of spin polarized carriers has been taken into account in many magnetic oxide/superconductor heterostructures,[23,30] but, as it is mentioned that the spin diffusion length exists up to a few nanometers into YBCO, the pair breaking by injected spin polarized carriers could not play a major role in the suppression of superconductivity in trilayers which have YBCO layer thicknesses in the range from 25 to 150 nm. It has already been reported[31,32] that even with 14 nm thickness of YBCO in LCMO/YBCO superlattices, the interplay between superconductivity and magnetism is negligible. Third, the difference in chemical potential between the ferromagnetic oxide layers and superconducting layer may lead to oxygen diffusion from the superconducting layer to magnetic layers at the annealing temperature, giving rise to an oxygen deficient YBCO layer. In order to rule out the possibility of poor growth quality of YBCO on LSCO, we have grown bilayers consisting of LSCO/YBCO with the same thickness as the LYL4 trilayer system. The resistance versus temperature curve for a bilayer is given in the inset of Fig. 3. It is clear from the inset that the onset of superconducting transition temperature in a bilayer is ~85 K, which is much higher than the superconducting transition temperature in trilayers. So, we conclude that the lowering of T_c in the trilayers compared to the bilayer may not be due to the growth problem, but may be due to underdoping, arising out of oxygen deficiency in it.

We have also measured resistance as a function of the magnetic field applied perpendicular to the sample surface (along the *a*-*b* plane) in current-in-plane (CIP) geometry. The field was swept between 0 and 10.8 T at three different temperatures below superconducting T_c, and the reduced resistance is plotted as a function of applied field for three different temperatures as indicated in Fig. 4 for the LYL4 sample. The inset in it shows H^* versus T_c-T curve for three different temperatures below superconducting T_c for the trilayer system, where H^* is the magnetic field corresponding to the onset of vortex dissipation occurring in the mixed state of YBCO in the trilayer structure at different temperatures. The values of H^* corresponding to different temperatures below the superconducting T_c is found from the data in Fig. 4. One important aspect of our experimental result is that it allows us to study the influence of magnetic layers on vortex dissipation in the liquid state of YBCO present in the trilayer system. Here, we describe the behavior of dH^*/dT near T_c for YBCO in the trilayer obtained from the inset given in Fig. 4. Our calculated value of dH^*/dT (from the inset) near T_c for the trilayer system turns out to be -0.2 T/K. From the magnetoresistance curves for YBCO epitaxial thin films as well as YBCO single crystals presented by Xiaowen *et al.*[25] and Shibata *et al.*,[33] one can estimate the dH^*/dT to be $\geqslant -0.6$ T/K, which is higher than the value that we have observed in our trilayer system. It has been reported by Sekitani *et al.*[22] that the value of dH_{c2}/dT of YBCO near T_c is about -10 and -1.9 T/K for a field applied ∥ and ⊥ to CuO_2 planes. They show that their experimental results in the H_{c2}-T phase diagram for a field applied ∥ CuO_2 planes can be well explained when they incorporate both spin-orbit (SO) effect ($\lambda_{SO}=5.1$) and spin-Zeeman effect ($\alpha=1.5$) in the Werthamer–Helfand–Hohenberg formalism, which otherwise predicts a transition at around 600 T for ($\lambda_{SO}=0$, $\alpha=0$). The difference in critical field arises because the spin-orbit effect weakens the degree of spin pairing in the superconductor and the spin-Zeeman effect increases the Pauli susceptibility of the superconductor. Thus, both effects limit the critical field. The drastic reduction of dH^*/dT near T_c in our case cannot be attributed to the increase in spin-orbit effect in the trilayer system because the presence of magnetic layers may not influence the spin-orbit interaction within the superconductor. At this point in time, the most feasible explanation is the exchange potential coupled with spin-Zemaan effect (due to the applied field as well as the internal field of magnetic layers), which could lead to such a strikingly low value of dH^*/dT near T_c. This lower value of dH^*/dT near T_c ascertains that the suppression of superconductivity in the YBCO layer is intense when it is sandwiched between two ferromagnetic layers compared to what is observed in an optimally doped YBCO sample.

It is also seen from Fig. 4 that very close to T_c (3 K below T_c), a very small magnetic field is sufficient to change the resistance drastically, whereas for two other temperatures (5 and 8 K below T_c), the change in resistance with magnetic field is comparatively less. This could be due to the fact that the suppression of the superconducting order parameter by the application of a magnetic field becomes weaker as we go toward lower temperature. The increase in resistance with applied magnetic field is most likely due to vortex dissipation in the liquid state of YBCO in the trilayer. The resistance in a vortex liquid state can be described by the thermally activated form $R(H,T)=R_0\exp[-U(H,T)/k_BT]$, where $U(H,T)$ is the activation energy for vortex motion. Since it is reported[9,34–36] that the vortex motion in the liquid state is thermally activated with the activation energy dependent on the field as an inverse power law, we expect an increase in resistance with increase in magnetic field. It has already been pointed out that as we measure the resistance in CIP geometry with the magnetic field applied perpendicular to current direction, the Lorentz force (F_L) on a unit length of vortex line is $J\times\Phi_0$ (where J is the current density and Φ_0 is the flux quantum) and is nonzero, and which consequently provokes the movement of flux lines or flux bundles, giving rise to dissipation. In addition to the vortex motion caused by the application of a magnetic field perpendicular to current direction, we also speculate at this point in time that the presence of two ferromagnetic layers at the bottom and at the top of the YBCO layer might have a drastic effect on the vortex dynamics in the vortex liquid state. The role of exchange potential coupled with spin-Zeeman effect on the vortex melting in this system needs to be studied. Detailed theoretical modeling needs to be carried out to understand these phenomena.

In summary, we have grown epitaxial LSCO/YBCO/LCMO trilayer structures on $LaAlO_3$ single crystal substrates. We find that the onset of superconducting T_c increases with the increase of YBCO thickness in the trilayer structure. We observe a remarkable anisotropic superconducting T_c ($T_{cH\parallel c\text{-axis}}<T_{cH\perp c\text{-axis}}$) of YBCO in the trilayer for the magnetic field applied parallel and perpendicular to the *c* axis. This anisotropy of the superconducting transition temperature in the trilayer is quite opposite to what is observed in YBCO single crystals as well as in thin films.[25,26] In conventional YBCO thin films as well as single crystals, the lowering of superconducting T_c is greater when the field is applied parallel to the *c* axis as compared to that applied perpendicular to the *c* axis. Our results also reveal that there occurs an intense suppression of superconductivity of YBCO in the LSCO/YBCO/LCMO trilayer structure compared to a single YBCO film as well as a single crystal. Apart from its fundamental interest, the system also shows huge magnetoresistance.

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