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Observation of the intense de-pairing effect in $YBa_2Cu_3O_{7-\delta}$ due to the spin injection from $La_{0.5}Sr_{0.5}CoO_3$

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Abstract
We report experimental evidence for a huge pair breaking effect induced by spin polarized quasiparticles in a $YBa_2Cu_3O_{7-\delta}/La_{0.5}Sr_{0.5}CoO_3$ bi-layer fabricated by pulsed laser deposition. The temperature dependent magnetization measurements show evidence for the presence of both ferromagnetic and diamagnetic phases in the bi-layer. The current dependent electrical transport studies in the bi-layer exhibit a significant reduction in the superconducting transition temperature with the increase in applied current as compared to a single $YBa_2Cu_3O_{7-\delta}$ layer and it follows a $I^{2/3}$ dependence in accordance with the pair breaking effect. Here, we find that the current driven from a ferromagnetic electrode with low spin polarization, such as $La_{0.5}Sr_{0.5}CoO_3$ (-11%), into the superconductor can act as a strong pair breaker. This indicates that the spin polarization of the injecting electrode is not the only criterion in determining the pair breaking effect, rather the transparency of the interface for the spin polarization may also be significant. More interestingly, the spin diffusion length for $YBa_2Cu_3O_{7-\delta}$ has a much longer length scale than that reported earlier in the study of ferromagnetic/superconducting heterostructures.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The co-existence of singlet superconductivity and ferromagnetism in bulk materials is highly improbable due to their incompatible nature. However, it is possible to artificially fabricate superconductor (S)/ferromagnet (F) hybrid heterostructures using various thin film deposition techniques. In the last few years, S/F hybrid heterostructures have attracted considerable theoretical and experimental attention [1, 2] due to the co-existence of two antagonistic quantum phases, i.e. parallelly aligned spins in the ferromagnet and Cooper pairs with oppositely aligned spins in the superconductor. The mutual interaction between the two competing order parameters in hybrid S/F heterostructure gives rise to a variety of novel physical phenomena such as domain wall superconductivity [3–8], superconducting spin switch effect [9–11], and inverse spin switch effect [12–14], depression of superconducting T_c [15–18], oscillating superconducting T_c [19–21] due to 0 and π-phase coupling, etc. Basically there are two main mechanisms responsible for the interaction of the superconducting order parameter with the magnetic order parameter in S/F hybrid heterostructures: (1) an electromagnetic mechanism (interaction of Cooper pairs with magnetic field induced by magnetic moments); (2) exchange interaction of magnetic moments with the electrons in the Cooper pair. The latter comes into play when Cooper pairs enter a ferromagnet to induce superconductivity in the magnetic material and it is well known as the proximity effect. The exchange interaction always favors spin polarization and thus causes the superconducting order parameter to decay faster in the F layer than in

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normal metal. The characteristic decay length scale $\xi_F = \hbar v_f/\Delta E_{ex}$ is found to be <1 nm for typical ferromagnets where $\Delta E_{ex} = 1$–3 eV.

It is also known that the superconducting state in a material is principally governed by a competition between four energies: condensation, magnetic field expulsion, thermal, and kinetic. The strength of the superconducting state gets reduced with the increase in temperature (T), magnetic field (H), and current density (J). With the ease of fabricating S/F hybrid heterostructures by thin film deposition techniques, it has become feasible to study the effect of spin polarized (SP) transport in the superconducting state in S/F hybrid heterostructures. The SP transport and tunneling experiments in S/F hybrid heterostructures seem to be very useful for providing important information on spin dependent electronic properties in superconductors. The injection of SP carriers into superconductors was first experimentally studied by Johnson [22] in permalloy/niobium/permalloy tri-layers. Soon after the discovery of high T_c superconductors (HTS) in cuprates by Bednorz and Muller [23] in 1986, a plethora of research activities has been carried out all over the world to understand the formation of the superconducting state in these materials. But up to now no clear unified answer has emerged. In this regard, the SP transport and tunneling experiments in HTS/ferromagnet hybrid model systems would be highly promising to provide new insights into the direction of spin dependent electronic properties of HTSs. It has been regarded that the electrons injected from hole doped rare earth manganites are suitable probes for possible spin–charge separation in HTSs. In the past few years much attention has been paid to the S/F heterostructures consisting of half-metallic manganite ferromagnets ($La_{0.7}Ca_{0.3}MnO_3$, $La_{0.7}Sr_{0.3}MnO_3$) [16–18, 24–30], which have a nearly total intrinsic spin polarization at the Fermi level [31–33]. However, relatively far less attention has been given to ferromagnets with low spin polarization in the S/F heterostructures, e.g. cobaltites such as $La_{0.5}Sr_{0.5}CoO_3$. $La_{0.5}Sr_{0.5}CoO_3$ are reported to have low spin polarization (∼11%) as compared to that of $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) or $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) due to a difference in the electronic density of states at the Fermi level [34]. It has to be noted that one does not always need a half-metallic material to study the SP transport in S/F heterostructures; rather it is very important to choose suitable combinations of S/F heterostructures which are more robust in terms of spin polarization at the superconductor–ferromagnet interface. The interface can play a key role for determining the spin polarization of electrons that need to be injected into a superconductor from a ferromagnet. So, in this regard it is necessary to study various combinations of S/F heterostructures that can retain high spin polarization at the interface. In particular the ferromagnetic perovskites/HTS systems are more interesting because of their ability to form high quality heterostructures with sharp interfaces. It has also to be mentioned here that the S/F hybrid systems are very important not only for the understanding of the fundamental physics buried in it, but also for the possibility of numerous potential applications in the emerging field of spintronics [35, 36]. In this context, it is very important to know the spin diffusion length across different HTSs. This is because the spin diffusion length across a superconductor plays a key role in transferring the spin information from one ferromagnet to another ferromagnet in typical F/S/F oxide spin valve structures. Recent developments in magnetic thin film technology have triggered such renewed interest in this direction. In this paper, we report on the investigation of the SP carrier driven pair breaking effect in the S/F bi-layer. The bi-layer consists of a top metallic ferromagnet layer ($La_{0.5}Sr_{0.5}CoO_3$) and a bottom superconducting ($YBa_2Cu_3O_{7-\delta}$) layer.

2. Experiment

The $YBa_2Cu_3O_{7-\delta}$(YBCO)/$La_{0.5}Sr_{0.5}CoO_3$(LSCO) bi-layer as well as the YBCO single layer were grown on (100) $LaAlO_3$ (LAO) single-crystal substrates by pulsed laser ablation (frequency = 5 Hz, fluence = 5 J cm^{-2}) of ceramic targets prepared by the standard solid state reaction method [37]. The constituent S and F layers in the bi-layer were grown by sequential deposition at 0.2 m bar of pure oxygen pressure and at temperatures of 785 °C and 765 °C, respectively. In addition, all the deposited films were *in situ annealed* at 500 °C for about one hour in oxygen background and then cooled down slowly to room temperature. The thicknesses of individual layers were determined from the deposition time and were found to be YBCO(∼150 nm)/LSCO(∼35 nm) in the bi-layer. The phase purity and the oriented growth of the films were analyzed by x-ray diffraction (XRD) using an X'pert PANalytical machine. The in-plane and the out-of-plane dc magnetization studies were carried out using a PPMS-VSM. The electrical transport properties were carried out in a closed cycle refrigerator system using a standard four-probe configuration.

3. Results and discussion

Structural characterization of the films was carried out using conventional XRD. A typical θ–2θ XRD scan is shown in figure 1 for a YBCO/LSCO bi-layer grown on the LAO (100) substrate. It clearly shows the (00l) type YBCO peaks only indicating that the YBCO layer is c-axis oriented, i.e. the YBCO c-axis lies perpendicular to the substrate plane. However, it is very hard to distinguish between the substrate and the LSCO peaks for an oriented growth of LSCO film in the bi-layer since the lattice parameter of LSCO (0.383 nm) is very close to the lattice parameter of LAO (0.379 nm). Figure 2 shows an enlarged view of the diffraction pattern of the LSCO/YBCO bi-layer around the (200) peak of LAO. It is observed that the diffraction spectrum shows a shoulder positioned very close to the (200) LAO peak. This shoulder position corresponds to the (200) LSCO film peak. Besides, the XRD pattern indicates no signature of any peak around 33° (the most intense peak for bulk LSCO), which rules out the possibility of any polycrystalline nature of the growth of LSCO in the bi-layer. Thus, the XRD spectrum clearly reveals the oriented growth of both the layers on LAO. It has to be noted that our earlier studies on tri-layer structures grown

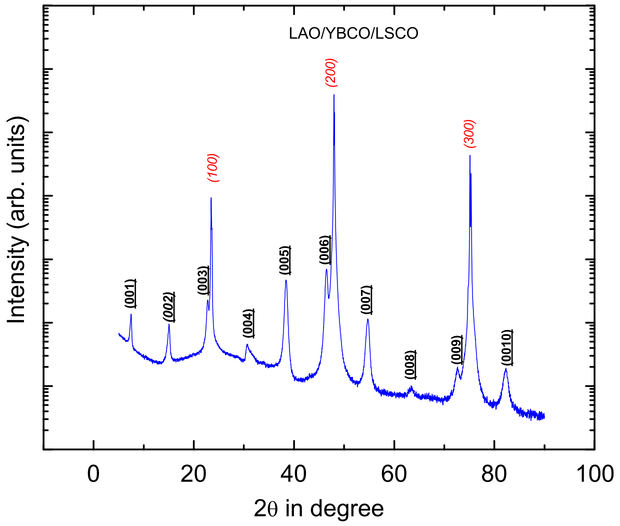

Figure 1. θ–2θ XRD spectrum for a YBCO/LSMO bi-layer grown on LAO (100). Peaks indexed by bold underlines and italics correspond to YBCO and LAO, respectively. The LSCO peaks largely overlap with LAO peaks.

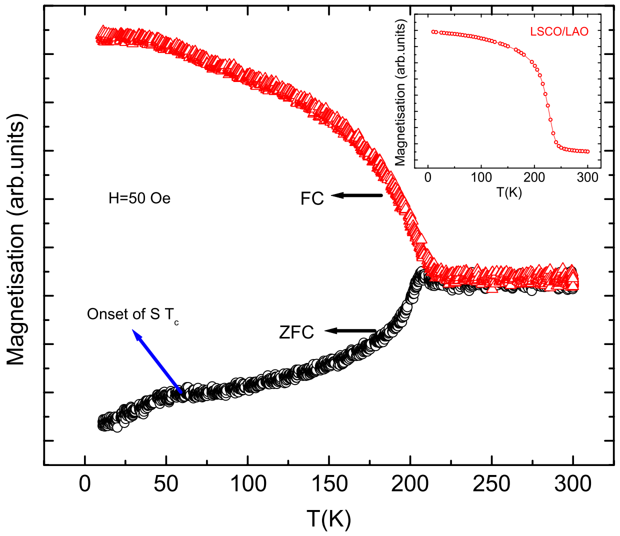

Figure 3. The temperature dependent FC and ZFC dc magnetization of the YBCO/LSCO bi-layer with a field of 50 Oe applied parallel to the film plane. The inset shows the temperature dependent FC for the single LSCO layer.

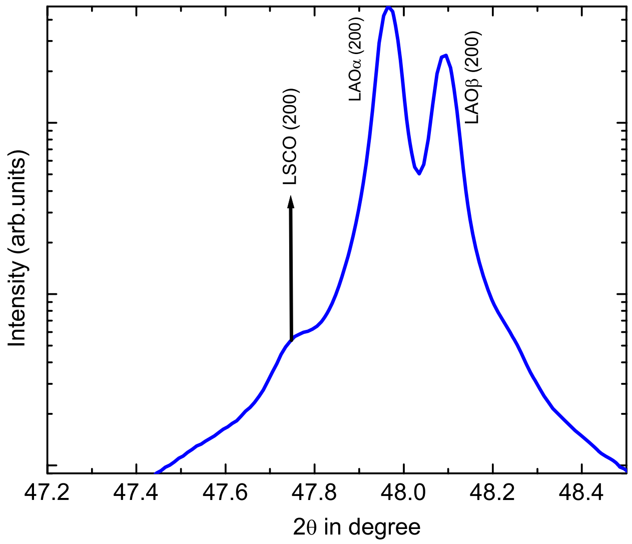

Figure 2. Zoom in view of the part of the θ–2θ XRD spectrum containing the (200) LSCO and LAO peaks.

under identical conditions showed the epitaxial nature of the film [15].

In addition, the presence of both ferromagnetic as well as superconducting layers in the bi-layer is characterized by the temperature dependent magnetization (M–T) as well as the magnetic hysteresis (M–H) studies. Figure 3 shows the field cooled (FC) as well as zero field cooled (ZFC) magnetization studies at 50 Oe applied along the film plane for the bi-layer. Firstly, it is very clear from the FC magnetization curve that the bi-layer shows a typical ferromagnetic-like behavior with a Curie temperature of ~220 K whereas the pure LSCO film of nearly the same thickness grown under identical conditions shows a Curie temperature of ~245 K, as shown in the inset of figure 3. The reduction of the ferromagnetic ordering temperature in the bi-layer may be due to charge transfer from the ferromagnetic layer to the superconductor or the difference in the growth of the LSCO layer on top of the YBCO layer compared to the growth on the LAO. The ZFC magnetization curve features a slope change below 70 K and this is strongly connected to the existence of the superconducting state of YBCO that is present in the bi-layer. Figure 4 displays the magnetic hysteresis (M–H) loops measured on the S/F bi-layer at 10 K with a magnetic field up to 70 kOe applied parallel and perpendicular to the film plane. It is clearly seen from figure 4 that the out-of-plane M–H loop of the bi-layer exhibits a well defined hysteresis curve which is associated with the diamagnetic behavior of the superconducting layer. In turn, the in-plane M–H plot for the bi-layer displays a characteristic ferromagnetic layer-like hysteresis curve. However, a careful look at the in-plane hysteresis curve reveals a two-step-like feature. The two-step-like feature is believed to arise from the existence of different coercive fields associated with different magnetic domains, as observed earlier by Luo *et al* [38] in $La_{0.5}Sr_{0.5}CoO_3$ thin films. Thus, a clear observation of magnetic anisotropies is found in the bi-layer and this experimental result is similar to the findings by Moran *et al* [39] in the $La_{2/3}Ca_{1/3}MnO_3/YBa_2Cu_3O_{7-\delta}/La_{2/3}Ca_{1/3}MnO_3$ tri-layer structure. Supporting the conclusions drawn from the XRD study, the presence of magnetic anisotropies further ensures the high quality epitaxial growth of bi-layer.

In addition to the magnetization measurements, we have performed extensive linear four-probe electrical transport measurements for a YBCO single layer as well as a YBCO/LSCO bi-layer. Figures 5 and 6 display the normalized resistance versus temperature curves ($R(T)/R(100)$) at different applied electric currents for a YBCO single layer as well as a LSCO/YBCO bi-layer (with YBCO having approximately the same thickness as single-layer YBCO), respectively. The inset in figure 6 shows the schematics of

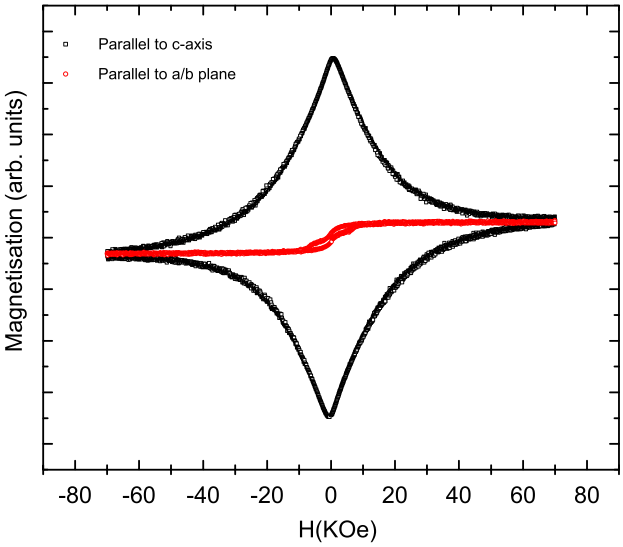

Figure 4. M–H hysteresis loop of the YBCO/LSCO bi-layer recorded at 10 K with the magnetic field applied parallel and perpendicular to the structure.

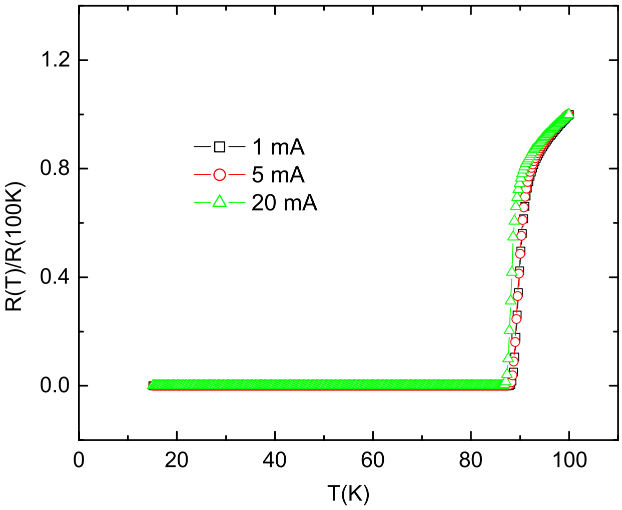

Figure 5. The in-plane temperature dependent normalized resistance of the YBCO layer with different applied currents from 1 to 20 mA.

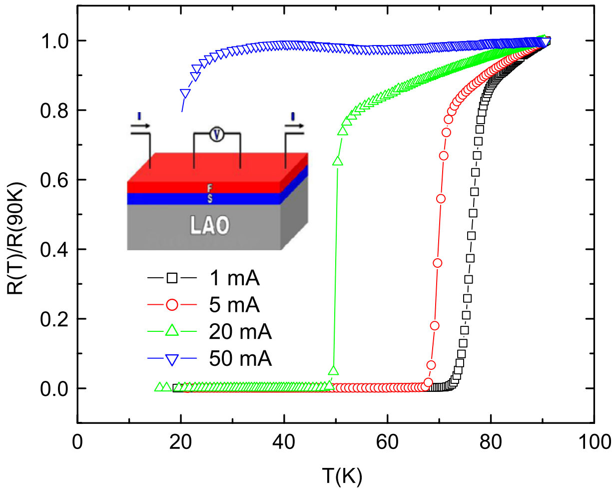

Figure 6. The in-plane temperature dependent normalized resistance of the YBCO/LSCO bi-layer with different applied current from 1 to 50 mA. The inset in it shows the schematic of transport measurement geometry.

transport measurement geometry. It is observed from figure 5 that the onset of superconducting T_c for the YBCO single layer is ~91 K for an applied current of 1 mA whereas the same for the LSCO/YBCO bi-layer is ~80 K, as seen from figure 6. The decrease in superconducting T_c in the bi-layer as compared to the single YBCO layer could be due to various possible reasons as explained below. First, the pair breaking effect due to (a) the magnetization of the ferromagnetic layer, (b) SP carrier injection (which will be described in the later part of our discussion). Second, the difference in the chemical potential between the top ferromagnetic oxide layer and the bottom superconducting layer may lead to oxygen diffusion from the superconducting layer to the magnetic layer at the annealing temperature, giving rise to an oxygen deficient YBCO layer. Third is the leakage of the Cooper pairs near the interface from the superconductor to the ferromagnetic layer (inverse proximity effect).

Further, it is very interesting to observe from figure 6 that the superconducting T_c of YBCO in the YBCO/LSCO bi-layer decreases drastically with the increase in the applied current values. It has to be noted here that we observe the onset of superconducting T_c in the bi-layer to be ~80 K when a current of 1 mA passes through it and it gets reduced to 73 K, 62 K, and 25 K with applied currents of 5 mA, 20 mA, and 50 mA, respectively. In order to understand the possible origin of such a drastic effect in the YBCO/LSCO bi-layer with increase in applied current; we have seriously looked into the resistive transition curves of a pure YBCO single layer with approximately the same thickness as in the bi-layer at different applied electrical currents. It is observed from figure 5 that for a single YBCO layer the onset of superconducting transition changes by ~2 K when the applied current changes from 1 to 20 mA and this phenomenon is well attributed to the pair breaking effect due to perturbation of the superconducting order parameter by ordinary quasiparticles. Thus the present experimental observation conspicuously hints that the shift of superconducting T_c with increasing applied current in the S/F bi-layer is huge as compared to that in a single YBCO bi-layer. The reason for such an observation can be explained by enhanced pair breaking effect due to the increase in transport current that induces the diffusion of non-equilibrium SP charge carriers from the ferromagnetic layer to the superconducting layer [40]. Here, the top ferromagnetic layer acts as a source for the SP carriers driven into the bottom superconducting layer. The driven SP carriers cause a shift in the chemical potential between spin up, $\mu_\uparrow$, and spin down, $\mu_\downarrow$, electrons in the superconductor and this gives rise to a non-equilibrium spin density. This non-equilibrium spin density leads to suppression of the superconducting order parameter by the pair breaking effect [41]. In the simplest picture, this phenomenon can be explained as [42] $\frac{\Delta(n_{qp})}{\Delta(0)} \approx 1 - \frac{2n_{qp}}{4N(0)\Delta(0)}$ where $\Delta(n_{qp})$ is the energy required to suppress the order parameter of the superconductor due to the density of SP quasiparticles n_{qp}. $N(0)$ and $\Delta(0)$ give the density of states and order parameter at $T = 0$ K respectively. Thus the injection of SP carriers over the superconducting gap depresses the order parameter monotonically with increase in n_{qp} and this results in the reduction of the superconducting T_c. Recent observations of the inverse spin switch effect [12–14] in F/S/F spin valves firmly reveals and supports the idea of a spin polarization driven pair breaking effect in superconductors as well. According to this, the F/S/F spin valve structure can have higher resistance in the antiparallel (AP) state of two ferromagnetic layers than in the parallel (P) state. This phenomenon is ascribed to the enhanced accumulation of SP quasiparticles in the superconductor due to the enhanced spin dependent reflection at the S/F interface in the AP state and thus it leads to the suppression of the superconducting gap. Though this results is different from what is expected by proximity effect theory, for P and AP configurations of magnetic layers, still it is experimentally observed that magnetic layers with high spin polarization lead to an inverse spin switch effect owing to large spin dependent reflection at the S/F interface. The spin polarization driven pair braking is very likely to occur when the spin diffusion length (l_{sd}) is comparable to the thickness of the superconducting layer. The l_{sd} can be regarded as a length scale over which the non-equilibrium spin distribution relaxes to equilibrium and it can be estimated by the well known relation $l_{sd} = (l_o v_f \tau_s)^{0.5}$ where τ_s is the SP electron diffusion time for relaxing to equilibrium, v_f is the Fermi velocity, and l_o is the electron mean free path. The relaxation phenomenon of SP quasiparticles only occurs through the spin exchange interactions like spin–orbit or magnetic impurity scattering that allow the spin flip process. This is unlike the relaxation phenomenon of simple quasiparticles via inelastic electron–phonon scattering where the spin flip does not occur. The relaxation diffusion time τ_s may be estimated by the relation; $\tau_s \sim 3.7\tau_{ex}k_B T_c/\langle\Delta(T)\rangle$, where τ_{ex} is the energy relaxation time for SP quasiparticles and $\langle\Delta(T)\rangle$ is the average energy gap [43]. The τ_{ex} is basically determined by the spin exchange interaction, i.e. $\tau_{ex} \sim h/E_{ex}$, where E_{ex} is the onsite spin exchange interaction in the superconductor. It has to be mentioned here that the l_{sd} in superconductors can be very high, as much as 1 cm, as found for the case of Al. However, for YBCO, the l_{sd} in the direction of the c-axis is approximately estimated to be 80 or 90 nm [43, 44]. According to the previously estimated values of l_{sd} in YBCO, the pair breaking by injected SP carriers should have been less effective in the bi-layer presented here due to the presence of YBCO layers with thicknesses of ~150 nm. This is because the higher the thickness of superconducting layer, the larger is the probability of recombination of injected SP carriers. Recent work by Singh *et al* [14] observed that with the increase in Nb thickness in Co/Pt–Nb–Co/Pt multilayers, the spin polarized carriers are not effective in breaking of Cooper pairs. In spite of all these observations, we clearly observe a huge reduction of the superconducting T_c in the YBCO/LSCO bi-layer with increase in the applied current and this in fact supports the work by Singh *et al* [14], where they observed a dependence of T_c suppression as a function of spin polarized current in Co/Pt–Nb–Co/Pt multilayers. Thus the observed experimental results suggests that SP carriers could play the role of strong pair breakers even up to a thickness of 150 nm YBCO layers. Thus the present result suggests that one needs to re-look at and re-estimate the value of l_{sd} in YBCO superconductor. Apart from the spin polarization driven pair breaking effect, the stray field originating from the domain walls within the F layer can also affect the superconducting properties in the S/F bi-layer [45, 46]. If the magnetic stray field exceeds the lower critical field, then it will drive the superconductor into the vortex state. Thus with increase in transport current the Lorentz force ($J \times B$) exerted on vortices will increase and consequently it will provoke the movement of vortices that will give rise to dissipation. Hence, with the increase in transport current, the superconductivity in the S/F bi-layer can be suppressed both by pair breaking due to SP carriers and dissipation by vortices.

In order to rule out the other possible reasons as a cause of this effect, we have investigated the resistive heating effect in pure LSCO film. It was observed that a current of 20 mA is required to increase its temperature by 2–3 K for a single-layer LSCO film of identical geometry at room temperature. In addition, it has also to be pointed out here that similar work by Vas'ko *et al* [47] showed that a current of 25 mA flowing in a ferromagnetic LSMO film increases its temperature by 1 K at a temperature in the range of 60–80 K. So, it is very clear from the above experimental observations that the resistive heating could not account for such a high reduction of T_c observed in the YBCO/LSCO bi-layer. Besides, it is very easy to distinguish between the pair breaking T_c shift and the resistive heating T_c shift by studying their functional dependence on the applied current (I) [48]. It is known that the pair breaking effect produces an apparent T_c shift proportional to $I^{2/3}$ whereas the resistive heating produces an apparent T_c shift proportional to I^2. Figure 7 shows the onset of superconducting T_c with the corresponding currents plotted as $I^{2/3}$ expected for the pair breaking effect. The solid line in figure 7 shows the best linear fit to the $I^{2/3}$ plot. Thus, this study clearly demonstrates that the shift of superconducting T_c in the bi-layer with applied current adheres to the $I^{2/3}$ dependence for pair breaking rather than the I^2 dependence for resistive heating. The inset in figure 7 shows the expected $I^{2/3}$ dependent onset T_c shift for a single YBCO layer. It is interesting to note from the extrapolation of the result in figure 7 that at the limit $I \to 0$, $T_c \sim 85$ K. This points to the fact that there is still a reduction of T_c by ~5–6 K as compared to the parent YBCO films. This may be attributed to the other possible earlier-described reasons such as (a) the inverse proximity effect, (b) an oxygen deficient YBCO layer due to interlayer diffusion at the interface, or (c) the effect of a magnetic layer, etc.

In general, the SP quasiparticle induced superconducting T_c suppression in S/F heterostructures should be proportional to the degree of spin polarization. But it is very interesting to note here that even though the LSCO has very low spin polarization compared to $La_{0.7}Sr_{0.3}MnO_3$ or $La_{0.7}Ca_{0.3}MnO_3$ as reported earlier, the SP quasiparticle induced T_c suppression in the YBCO/LSCO bi-layer is huge. This possibly suggests that the spin polarization at the YBCO–LSCO interface

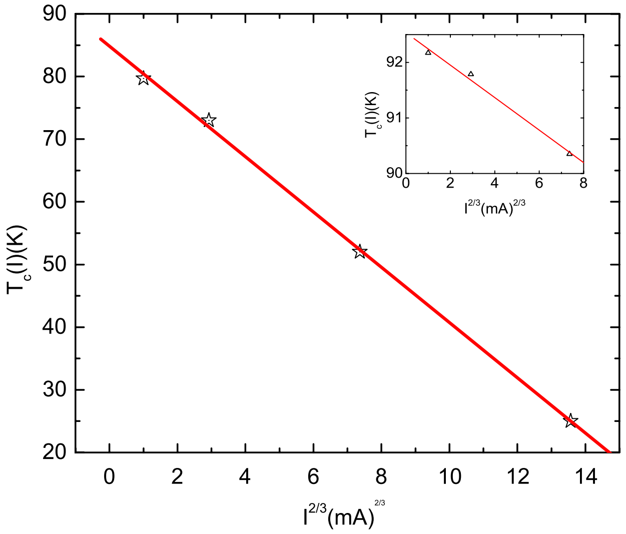

Figure 7. The shifted superconducting T_c in the YBCO/LSCO bi-layer at different currents plotted versus $I^{2/3}$. The solid line shows the best linear fit to the $I^{2/3}$ plot. The inset in it shows the same for a YBCO single layer.

remains intact. This might even suggest that the transparency for the SP carriers at this interface may be high compared to the other systems. Further, the spin polarization in LSCO is dominated by t_{2g} electrons near E_f whereas the same is dominated by e_g electrons for LSMO. So, it is also important to investigate whether the t_{2g} electrons could play a different role for such a huge reduction of T_c in the YBCO/LSCO bi-layer. Further experiments are required to elucidate these aspects.

4. Conclusion

In summary, we have grown a fully oriented YBCO/LSCO bi-layer on $LaAlO_3$ single-crystal substrates. We find that the bi-layer exhibits magnetic anisotropic behavior from the M–H measurements, i.e. it shows superconducting-like hysteresis when the field was applied perpendicular to the film plane and a typical ferromagnetic one when the field was applied parallel to the film plane. From electrical transport studies, we find the suppression of superconductivity is very intense in the YBCO/LSCO bi-layer as compared to the single YBCO layer with increase in applied current. This result appears to be due to the pair breaking effect associated with SP carriers being injected into the superconductor but not due to resistive heating. Our experimental result finds that the SP quasiparticle induced pair breaking effect can extend over ~150 nm of YBCO thickness in the YBCO/LSCO bi-layer and thus it suggests the spin diffusion length of YBCO could be longer than the earlier reported values. More interestingly, we realize that even the current driven from a material with low spin polarization such as $La_{0.5}Sr_{0.5}CoO_3$ (-11%) can also lead to a high suppression of superconductivity by the pair breaking effect. This suggests that the spin polarization of the ferromagnetic electrode is not the only criterion in reducing the superconducting T_c. Other effects such as the quality of the interface, the magnetization of the ferromagnet, etc may also play a significant role and these aspects need to be investigated further in detail.

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