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Suppressed B_{c2} in a superconducting/ferromagnetic bilayer

A Gaurav[1], B R Sekhar[1,2], P S Anil Kumar[3] and D Samal[1,2]

[1] Institute of Physics, Sachivalaya Marg, Bhubaneswar, 751005, India
[2] Homi Bhabha National Institute, Anushakti Nagar, Mumbai 400085, India
[3] Dept. of Physics, Indian Institute of Science, Bangalore, 560012, India

E-mail: dsamal@iopb.res.in

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Abstract

We investigate the influence of a ferromagnetic layer ($La_{0.7}Sr_{0.3}MnO_3$) on the upper critical field of $YBa_2Cu_3O_{7-\delta}$ superconducting layer in a superconductor/ferromagnet (SC/FM) hybrid structure. The $YBa_2Cu_3O_{7-\delta}/La_{0.7}Sr_{0.3}MnO_3$ (YBCO/LSMO) hybrid bilayers as well YBCO single layer were fabricated on (001) $LaAlO_3$ (LAO) substrate using pulsed laser deposition.The temperature dependent upper critical field $B_{c2}(T)$ of type-II superconductors are generally described in the frameworks laid down by Werthamer–Helfand–Hohenberg and Ginzburg–Landau. We employ both formalism to estimate $B_{c2}(T)$ of YBCO from magneto-transport data in SC/FM bilayers of varying ferromagnetic layer thickness as well as for YBCO single layer. We find that the $B_{c2}(T)$ of YBCO in YBCO/LSMO bilayer gets suppressed as compared to that of single YBCO layer. Further to this, we also observe that the $B_{c2}(T)$ of YBCO in the bilayer system decreases with increasing ferromagnetic LSMO layer thickness. These two results are discussed in the light of magnetic proximity effect, spin-diffusion induced pair breaking and enhanced effective magnetic field experienced by the YBCO layer with increasing ferromagnetic LSMO layer thickness.

Introduction

Singlet superconducting and ferromagnetic order are antagonistic to each other and generally they do not coexist in bulk materials. However, the fabrication of thin film heterostructures using thin film deposition techniques has made it possible to investigate the interplay between superconductivity and ferromagnetism in proximity. The control over layer thickness provides an added opportunity to change the relative strength of competing order parameters by varying the layer thickness. The mutual interaction between the two competing order parameters at the superconductor/ferromagnet (SC/FM) interface gives rise to a variety of novel electronic phenomena like 0–π phase coupling [1], spin-triplet superconductivity [2–7], possibility to observe majorana fermions [8], domain wall superconductivity [9], superconducting spintronics [10] etc which have led to a wide study of such systems over last few decades. When a superconducting layer is brought in contact with a ferromagnetic layer, the superconducting order can be suppressed due to magnetic proximity effect [11], which includes (a) electromagnetic interaction of Cooper pairs with the magnetic field induced by magnetic moments of the magnetic layer (b) exchange interaction of the magnetic moment with the electrons in cooper pair and (c) self-injection of the spin polarized quasiparticles from FM to SC.

High-T_c cuprate superconductors in comparison to low-T_c counterparts have several unique characteristics that makes it interesting to study. Some of them include two-dimensional layer structure, short coherence length and high upper critical field (B_{c2}), deep magnetic penetration depth, low superfluid density, competing unusual electronic states and several magnetic vortex phases in the $B-T$ phase diagram. In type-II superconductors, B_{c2} defines the magnetic field strength below which the vortices appear and above which they behave as normal metals. The direct and accurate measurement of B_{c2} provides a measure of superconducting strength. However, measuring the upper critical field in cuprates require intense magnetic fields, because of their

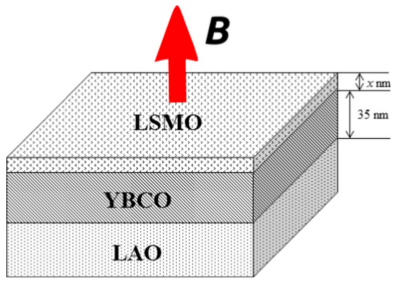

Figure 1. Schematic of the superconducting (YBCO)/ferromagnetic(LSMO) bilayer with varying LSMO thickness grown on LAO. $x = 0$, 16 and 28 for single layer YBCO, bilayer BL1 and bilayer BL2 respectively. The red arrow perpendicular to the *ab* plane of the sample shows the direction of the magnetic field applied in our experiment.

shorter coherence lengths ($B_{c2}(0) = \phi_0/2\pi\xi^2$). For example, in the first report on multi-phase Y–Ba–Cu–O, Chu and co-workers estimated $B_{c2}(0)$ to be in between 80 and 180 T [12]. The subsequent works on single-phase YBCO [13, 14] single crystals and thin films found $B_{c2}(0)$ within this range [15–18]. The requirement for high magnetic field in cuprates has limited the direct measurement of $B_{c2}(T)$ and the investigation of normal state properties at $T < T_c$ to a few laboratories which are capable of producing field in the excess of 100 T using pulsed magnets. Other methods to suppress superconductivity include perturbation by chemical doping [19–21], epitaxial strain/stress induced by substrates [22–25] etc. Extensive works have been performed in this regard on bulk materials. Besides the chemical doping and strain induced effects, the proximity of ferromagnetic layer to a superconducting layer in the designed SC/FM hybrids are found to perturb the superconducting properties substantially [11, 26–30]. A recent Nernst effect study by Matusiak *et al* on oxide ferromagnet ($La_{0.67}Sr_{0.33}MnO_3$)/superconductor (YBCO) bilayer system with varying superconductor thickness demonstrated that superconductivity in bilayer gets suppressed at lower applied magnetic field than in single YBCO layer, and therefore superconducting phase space in the B–T plane gets reduced [31]. SC/FM and in particular YBCO/LSMO hybrid structures with varying superconducting layer thickness have been studied widely. However, the hybrid structures of YBCO/LSMO with varying ferromanetic layer thickness remain scantly explored. Here, we report on the influence of the upper critical field of YBCO in YBCO/LSMO bilayer with varying ferromagetic LSMO layer thickness and attempt to understand the underlying physics.

In this work, we use a FM layer of LSMO with a varying thickness in bilayer geometry with YBCO to investigate the influence of the magnetic layer on the B_{c2} of YBCO in a bilayer system of YBCO/LSMO . Data points in ($B_{c2}(T)$, T) phase space were extracted from temperature dependent resistance plots in the presence of magnetic field up to 11 T. Building on the data points extracted close to the T_c, we incorporate the theory of Werthamer–Helfand–Hohenberg (WHH) and Ginzburg–Landau (GL) to span the $B-T$ phase space. We observe a suppression of $B_{c2}(T)$ in bilayers as compared to that of the bare YBCO layer. Moreover we find that the $B_{c2}(0)$ and in general $B_{c2}(T)$ of YBCO in YBCO/LSMO bilayers decrease with increasing LSMO layer thickness.

Experimental details

Superconducting thin film YBCO and the bilayer thin film consisting of YBCO and ferromagnetic LSMO were grown on single crystal LAO substrate using pulsed laser deposition at a fluence of 5 J cm^{-2} and a frequency of 5 Hz. The bilayer thin films were grown by sequential deposition of YBCO and LSMO at O_2 pressure of 0.2 mbar and temperature of 765 °C and 755 °C respectively. After growth the films were annealed for an hour at 500 °C in the presence of 500 mbar O_2 to compensate for oxygen deficiency they were then cooled down to room temperature in the oxygen ambience.

Thin film of 35 nm thick YBCO, namely YB and that of bilayers namely BL1 (35 nm YBCO/16 nm LSMO) and BL2 (35 nm YBCO/28 nm LSMO) were fabricated as indicated in figure 1. The structural characterization of the bilayer thin films was performed using x-ray diffraction and it reveals *c*-axis oriented growth as shown in figure 2. The ferromagnetic character of the LSMO single layer was established with SQUID measurement as shown in figure 2(b). The magneto-transport properties were measured down to 5 K and upto a magnetic field of 11 T in the standard four-probe geometry. The field was aligned parallel to the *c*-axis throughout the experiment.

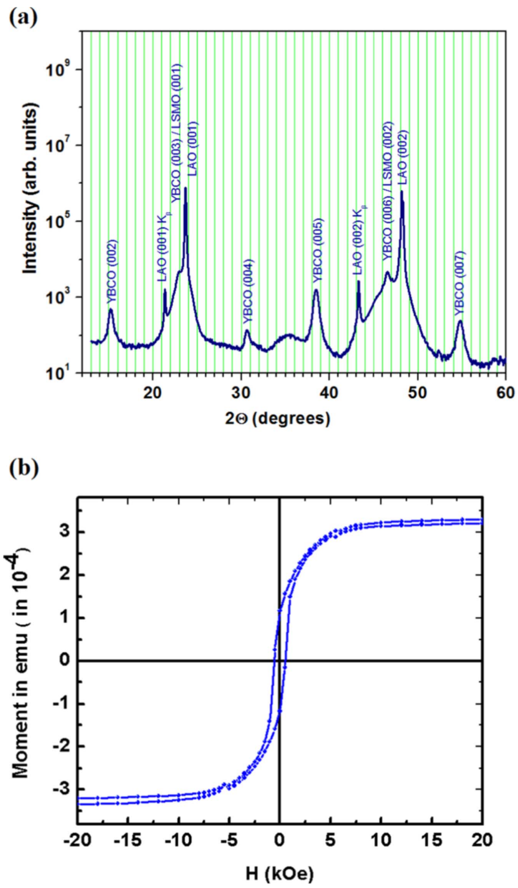

Figure 2. (a) θ–2θ x-ray diffraction pattern for YBCO (35 nm)/LSMO(28 nm) bilayer grown on LAO(001) substrate. The (001) and (002) reflections of LSMO overlap with (003) and (006) reflections of YBCO. (b) A representative *M*–*H* hysterisis loop at 10 K showing ferromagnetic character of LSMO layer grown on LAO.

Results and discussions

Magnetic field dependence of the resistance versus temperature for YBCO and YBCO/LSMO bilayers are shown in figure 3. The resistance of all the samples has been normalized to their respective values at 120 K for the sake of comparison. It is observed that the zero field superconducting T_C of YBCO decreases in the YBCO/LSMO bilayers as compared to that of single YBCO layer. This is attributed to spin polarized carrier driven pair breaking and magnetic proximity effect as has been discussed in detail in our earlier work [32–34]. With the application of the magnetic field the resistive transition gets broadened due to vortex dissipation and remarkably the broadening increases with the FM layer thickness. The shift in the superconducting transition measured between 0 T and 11 T are found to be ~1 K, 2 K and 3 K for YBCO, BL1 and BL2 respectively. It is known that the resistance in the vortex liquid state is described by a thermally activated form, $R(B,\ T) = R_0 e^{\frac{-U(B,T)}{k_B T}}$, where $U(B,\ T)$ is the activation energy for the vortex motion. Indeed our earlier study [35, 36] revealed that $U(B,\ T)$ decreases significantly in the SC/FM bilayers due to the magnetic proximity effect and thus consequently promotes the easy movement of the vortex giving rise to dissipation.

Field induced broadening of the resistive transitions in type-II superconducting materials makes the identification of the true value of T_c ambiguous from magneto-transport measurements. Therefore, the extraction of $B_{c2}(T)$ from magneto-transport measurements is debatable. We use a method to find T_C at different values (90%, 50% and 10%) of the full normal state resistance (R_F) as illustrated in figure 4. Consequently, we estimate the upper critical field $B_{c2}(0)$ at three different %s of the normal state resistance using two different formalisms that will follow in the coming paragraphs.

The dependence of the upper critical field on the temperature for type-II superconductors is well described by the theory of Werthamer–Helfand–Hohenberg or WHH [37]. We use a simplified WHH relation, which excludes spin-paramagnetic and spin–orbit effects to construct the *B*–*T* phase diagram as shown in figure 5(a). The WHH equation considering only the orbital effects, in the dirty limit is given by the following relation:

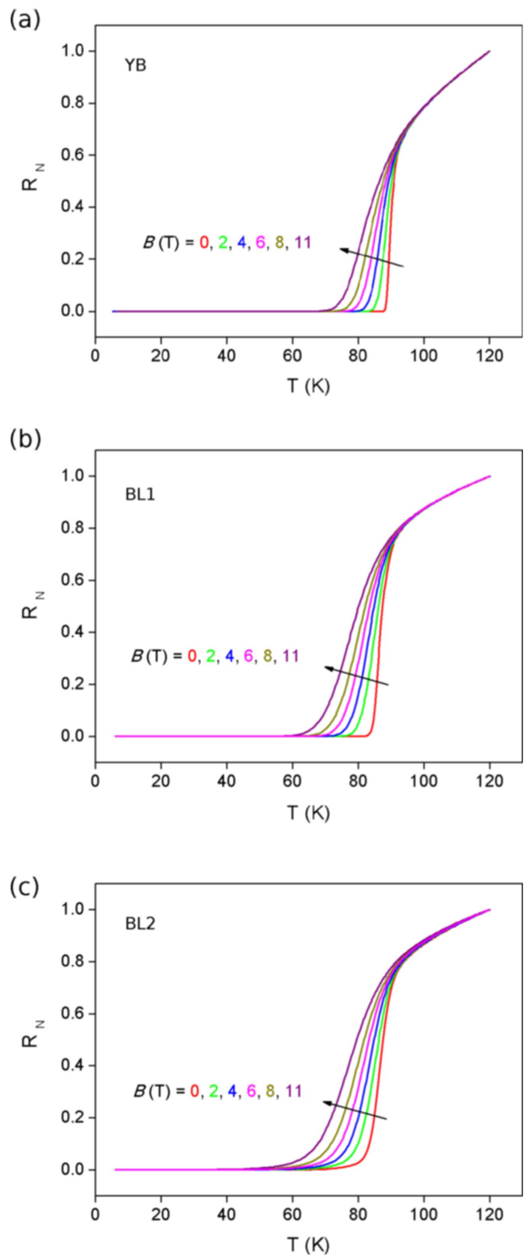

Figure 3. Normalized resistance versus temperature for (a) YBCO (YB) and bilayers (b) BL1 and (c) BL2 under external magnetic field applied perpendicular to the film plane. Figure (a) and (b) are reproduced from our earlier work [35]. Reprinted with permission, copyright 2010 IOP Publishing.

$$\ln\left(\frac{1}{t}\right) = \psi\left(\frac{1}{2} + \frac{h}{2t}\right) - \psi\left(\frac{1}{2}\right), \tag{1}$$

where ψ is the Digamma function and $t = T/T_c$ is the reduced temperature and h is given by the following relation:

$$h = \frac{4B_{c2}}{\pi^2(-\mathrm{d}B_{c2}/\mathrm{d}T)_{T=T_c}}. \tag{2}$$

As $T \to 0$, equation (1) reduces to

$$B_{c2}(0) = -0.693T_c\left(\frac{\mathrm{d}B_{c2}}{\mathrm{d}T}\right)_{T=T_c}. \tag{3}$$

The WHH relation given in equation (1) is a first order differential equation which is solved numerically to fit our data. The $B_{c2}(0)$ can also be found directly (without fitting) from equation (3) which is a solution to equation (1) in the limit $T \to 0$. We see that both the equations (1) and (3) contain the term $\frac{\mathrm{d}B_{c2}(T)}{\mathrm{d}T}$, which is a crucial quantity for the determination of B_{c2}. The slope $\left(\frac{\mathrm{d}B_{c2}(T)}{\mathrm{d}T}\right)_{T=T_c}$ under a particular % of R_F can be calculated

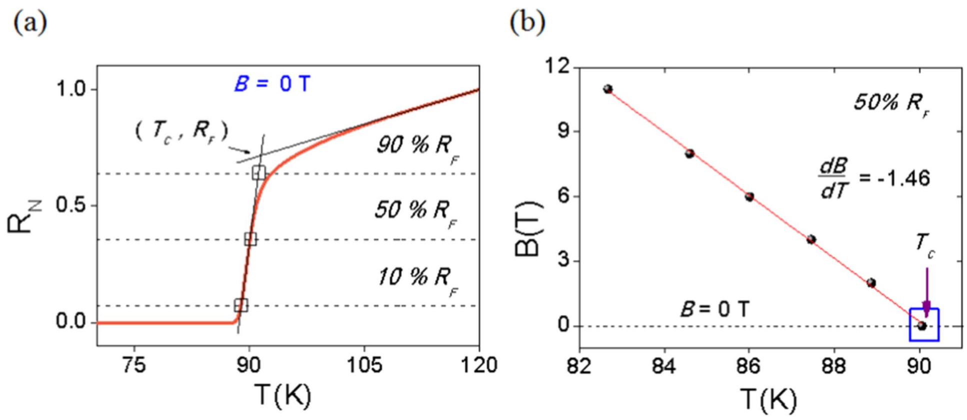

Figure 4. (a) Demonstrates a method for finding the T_c for YBCO. Two straight lines are drawn on the normal and transition region of the resistive curve (R–T) whose intersection gives the T_c and R_F (we call it full resistance). Lines are drawn parallel to the T-axis at 90%, 50% and 10% of R_F to get the corresponding T_c values at the intersection point as shown by the boxes. This method is repeated to determine T_c values under various applied fields. (b) $B-T$ phase space for YBCO under 50% R_F criterion. The extracted data are fitted with a straight line resulting in $\mathrm{d}B/\mathrm{d}T$ (slope) $= -1.46$ and T_c (x-intercept) $= 90.1$ K, indicated by the point enclosed in blue box.

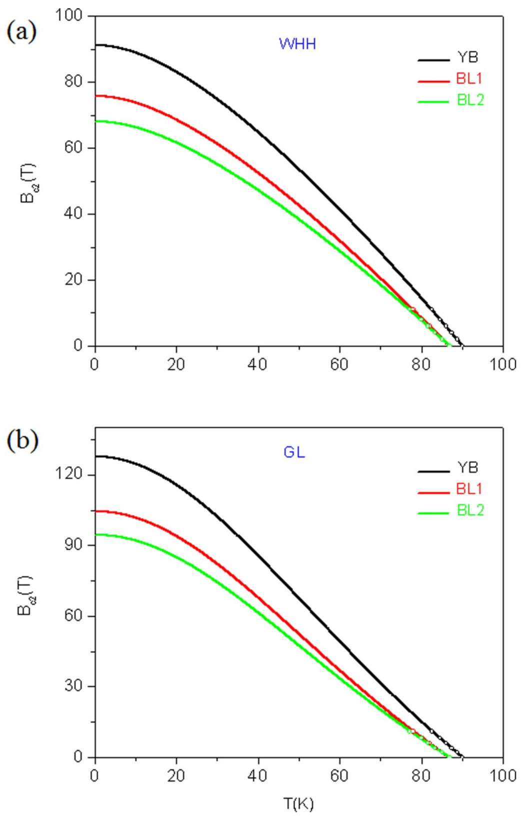

Figure 5. $B-T$ phase diagrams for YBCO (YB) and bilayers (BL1, BL2) constructed from magneto-transport data (under 50% R_F criterion) using (a) WHH and (b) GL formalism.

from the fitted straight line as shown in figure 4(b). The intercept of this line at $B = 0$ gives T_c, which is a small refinement to the originally extracted T_c from the R–T measurement at $B = 0$. The $\frac{\mathrm{d}B}{\mathrm{d}T}$, T_c and $B_{c2}(0)$ of the YBCO and the bilayers calculated for 90%, 50% and 10% of the R_F are listed in table 1. The evolution of $\left(\frac{\mathrm{d}B_{c2}(T)}{\mathrm{d}T}\right)_{T=T_c}$ with % R_F for the single YBCO layer is shown in figure 6. The table (table 1) also lists $B_{c2}(0)$ calculated using the theory of Ginzburg–Landau (GL) [38] given by the following relation:

Table 1. A list of parameters for YBCO (YB) and YBCO/LSMO bilayers (BL1, BL2) extracted (estimated) from the magneto-transport measurement.

System↓ Units	T_c (K)	$\left\lvert \frac{dB_{c2}(T)}{dT}\right\rvert_{T=T_c}$ (−)	$B_{c2\text{WHH}}(0)$ ($\times 10^2$ T)	$B_{c2\text{GL}}(0)$ ($\times 10^2$ T)	$\xi_{ab\text{WHH}}(0)$ (A^0)	$\xi_{ab\text{GL}}(0)$ (A^0)
90% YB	91.6 ± 2.9	3.85 ± 0.49	2.44 ± 0.32	3.48 ± 0.22	11.60 ± 0.76	9.72 ± 0.31
90% BL1	89.2 ± 1.4	3.28 ± 0.30	2.02 ± 0.19	2.88 ± 0.14	12.74 ± 0.60	10.69 ± 0.25
90% BL2	89.9 ± 0.1	2.71 ± 0.07	1.69 ± 0.04	2.39 ± 0.04	13.96 ± 0.18	11.72 ± 0.09
50% YB	90.1 ± 0.0	1.46 ± 0.02	0.91 ± 0.01	1.28 ± 0.01	18.96 ± 0.13	16.05 ± 0.06
50% BL1	86.8 ± 0.0	1.26 ± 0.01	0.76 ± 0.00	1.04 ± 0.00	20.83 ± 0.05	17.74 ± 0.04
50% BL2	87.0 ± 0.0	1.13 ± 0.01	0.68 ± 0.00	0.94 ± 0.01	21.93 ± 0.06	18.66 ± 0.07
10% YB	88.7 ± 0.0	0.89 ± 0.01	0.55 ± 0.01	0.75 ± 0.00	24.5 ± 0.2	20.94 ± 0.06
10% BL1	84.2 ± 0.1	0.80 ± 0.02	0.46 ± 0.01	0.63 ± 0.00	26.6 ± 0.3	22.82 ± 0.08
10% BL2	83.9 ± 0.0	0.73 ± 0.01	0.42 ± 0.01	0.57 ± 0.01	27.8 ± 0.2	23.96 ± 0.16

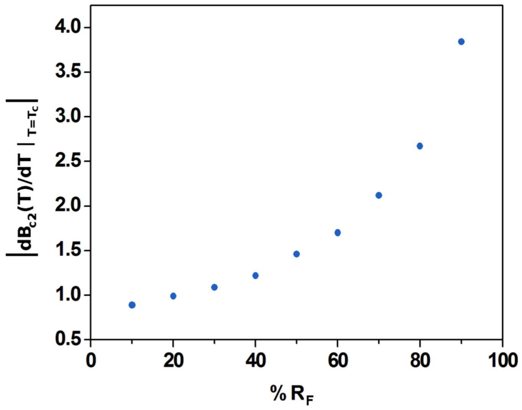

Figure 6. The evolution of $\left\lvert \frac{dB_{c2}(T)}{dT}\right\rvert_{T=T_c}$ with % R_F for YBCO thin film.

$$B_{c2}(T) = B_{c2}(0)\left[\frac{1-t^2}{1+t^2}\right], \tag{4}$$

where $t = T/T_c$ is the reduced temperature and $B_{c2}(0)$ is the upper critical field at 0 K. The $B_{c2}(0)$ values calculated using GL is found to be higher than the corresponding WHH values. The full B–T phase diagram constructed using GL is shown in the figure 5(b). The values of $B_{c2}(0)$ under 50% R_F criterion are estimated to be ~91 T and 128 T using WHH and GL formalism respectively. These values come close to the values estimated from previous works on YBCO thin films [15, 17, 18] and single crystals [16], which were determined by magnetization measurements in the pulsed magnetic fields. However, under 50% criterion we estimate WHH $B_{c2}(0)$ to be ~76 T and 68 T for SC/FM bilayers BL1 and BL2 respectively. The corresponding values in the GL formalism are ~104 T and 94 T respectively. Clearly it is found that the $B_{c2}(0)$ of the bilayer decreases (i) as compared to the single YBCO layer and (ii) with the increasing ferromagnetic LSMO thickness. Moreover, resistive measurements with % R_F as low as 10, provide WHH $B_{c2}(0)$ values of 55 T for YBCO and ~46 T and 42 T for BL1 and BL2 respectively. Nakao *et al* estimated $B_{c2}(0)$ to be 40 T from magnetization measurement performed using induction method in high magnetic field pulses on single crystal YBCO [39]. A value close to the estimations of Nakao *et al* has also been found by Smith *et al* from their onset flux flow data [15]. Considering the 10% R_F (nearly onset of flux flow), these results add to the consistency and validation of the method we use for the estimations of the $B_{c2}(0)$.

Irrespective of formalisms used for the estimation of $B_{c2}(0)$, we observe a decrease in the upper critical field of YBCO in YBCO/LSMO bilayer. LSMO is a highly spin polarized ferromagnet with spin polarization values ranging from 58% to 92% in the bulk [40]. The suppression therefore may be attributed to the injection of spin polarized carrier from the top LSMO layer to YBCO, resulting in non-equilibrium quasiparticles density of states in the YBCO layer which can be described by the following equation [41]:

$$\frac{\Delta(n_{\text{qp}})}{\Delta(0)} \approx 1 - \frac{2n_{\text{qp}}}{4N(0)\Delta(0)}, \tag{5}$$

where n_{qp} is the density of spin polarized quasiparticles, $N(0)$ is the density of states of the SC at $T = 0$ K, $\Delta(0)$ is the superconducting order parameter and $\Delta(n_{\text{qp}})$ is the energy required to suppress the order parameter of the superconductor, respectively. In our case the FM layers are grown on fixed thickness (35 nm) of superconducting

YBCO layer. This thickness is comparable to the spin diffusion length $l_{sd} = (l_0 v_F \tau_S)^{0.5} \sim 10$ nm ($T \rightarrow 0$) and 80 nm ($T \rightarrow T_c$) respectively in YBCO as calculated by Yeh *et al* [42] and others [43, 44]. Here, l_0 is the mean free path in YBCO, v_F is the Fermi velocity and τ_S is the spin diffusion time. Based on these assumptions it is expected that the YBCO/LSMO bilayer will show suppressed superconducting parameters as compared with the single YBCO layer. Generally, non uniform spin injection from a FM to a SC in SC/FM bilayer system leads to double transition in R–T plot when $d_S > l_{sd}$ [29]. In our case however, we observe a single superconducting transition in the $R-T$ plot for YBCO/LSMO bilayers. This can be understood as the thickness of the constituent YBCO layer and l_{sd} are in the similar range. The discussions so far successfully explains the suppression of B_{c2} in bilayer as compared with the single YBCO layer. However, it fails to explain the suppression of B_{c2} within the bilayer system of fixed SC layer thickness and varying FM layer thickness.

When a SC is sandwiched to a FM in a hybrid structure, the superconducting and ferromagnetic order compete on the length scales of $\xi_S = \left(\frac{\hbar D_S}{k_B T_c}\right)^{0.5}$ and $\xi_F = \left(\frac{\hbar v_F}{\Delta E_{ex}}\right)$ where ξ_S and ξ_F are decay lengths of the superconducting order in SC and in FM respectively, $\hbar$ is the reduced Planck's constant, D_S is the electron diffusion coefficient for the superconductor, k_B is the Boltzman's constant, T_c is the critical temperature of the superconductor, v_F is the Fermi velocity and ΔE_x is the exchange splitting energy of the ferromagnet [11]. These length scales correspond to a measure on which the superconducting order parameter decay in SC and FM layers respectively, due to proximity effect [11, 45]. For typical values of $\Delta E_{ex} \sim$ (1–3) eV and $v_F \sim 10^7$ cm s^{-1}, ξ_F is estimated to be ~(0.2–0.7) nm whereas ξ_s is reported to be ~1 nm. We estimate the in plane GL coherence length ξ_{ab} to be of the order of ~2 nm for YBCO and bilayers under 50% R_F criterion. These lengths (listed in table 1) are calculated using the GL relation $B_{c2\|c}(0) = \phi_0/2\pi\xi_{ab}{}^2$, where ϕ_0 is the magnetic flux quanta. Unlike BCS superconductors, high temperature superconductors have very small coherence length which lets the superconductivity survive down to a very small thickness [46]. The small value of ξ_S suggests that only a small thickness ~(1–2) nm of the YBCO layer gets affected due to proximity effect. These considerations establish that the superconducting parameters like T_c and $B_{c2}(0)$ should not be drastically affected for $d_S \gg \xi_S$, where d_S is the thickness of the SC layer in the SC/FM bilayer. Our bilayers satisfying these conditions are supposed to show identical T_c and $B_{c2}(0)$. On the contrary we see large differences in the estimated $B_{c2}(0)$ between the bilayers having different LSMO layer thickness. It has to be noted that magnetization in thin films is very different from their bulk counterpart and usually decreases with the decreasing FM thickness [47, 48]. The LSMO spin moment has been found to increase approximately from 0.4 μ_B to 2.9 μ_B when its thickness increases approximately from 5 to 30 nm [47]. Therefore, we conjecture the effective magnetic field experienced by the the YBCO layer in proximity to the LSMO layer increases with increasing LSMO layer thickness. This can be accounted for the relatively large suppression of the B_{c2} in BL2 as compared to BL1.

Conclusions

In conclusion, we have investigated the influence of ferromagnetic LSMO layer on the upper critical field $B_{c2}(0)$ of YBCO in YBCO/LSMO bilayer. From the WHH and GL analysis of magneto-transport data, we have estimated the upper critical field of YBCO in YBCO/LSMO bilayer system and have compared with the value obtained for the single YBCO layer. For example, using WHH analysis under 50% R_F criterion we have calculated $B_{c2}(0)$ of 91 T, 76 T and 68 T for YBCO, BL1 and BL2 respectively. We find that the upper critical of YBCO in YBCO/LSMO bilayer gets suppressed by a few tens of Tesla as compared to the single YBCO layer. Moreover, we also observe that the extent of suppression of $B_{c2}(0)$ increases with increasing LSMO layer thickness. We have provided a comprehensive discussion to account for the suppression based on spin polarized quasiparticle injection induced cooper-pair breaking and magnetic proximity effects. However, these phenomenon fell short to completely describe our result of enhanced suppression in the bilayer system as the LSMO thickness is increased. We attribute the observed effect to the local magnetic field emanating from the ferromagnetic LSMO layer, which increases with the increasing LSMO layer thickness.

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ORCID iDs

A Gaurav https://orcid.org/0000-0002-6335-4039

D Samal https://orcid.org/0000-0003-2618-4445

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