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**PAPER**

**Suppressed \( B_{c2} \) in a superconducting/ferromagnetic bilayer**

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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\(_2\)Cu\(_3\)O\(_{7−δ}\) superconducting layer in a superconductor/ferromagnet (SC/FM) hybrid structure. The YBa\(_2\)Cu\(_3\)O\(_{7−δ}\)/La\(_{0.7}\)Sr\(_{0.3}\)MnO\(_3\) (YBCO/LSMO) hybrid bilayers as well as 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 \( 0−\pi \) 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
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 scantily explored. Here, we report on the influence of the upper critical field of YBCO in YBCO/LSMO bilayer with varying ferromagnetic 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.
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 TC 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,

\[ R(B, T) = R_0 e^{U_{BT} / k_B T} \]

where \( U_{BT} \) is the activation energy for the vortex motion. Indeed our earlier study [35, 36] revealed that \( U_{BT} \) 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 (\( R_\Phi \)) 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:
\[
\ln \left( \frac{1}{t} \right) = \psi \left( \frac{1}{2} + \frac{h}{2t} \right) - \psi \left( \frac{1}{2} \right),
\]
(1)

where \( \psi \) is the Digamma function and \( t = T/T_c \) is the reduced temperature and \( h \) is given by the following relation:

\[
h = \frac{4B_{c2}}{\pi^2(-dR_{c2}/dT)_{T=T_c}}.
\]
(2)

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

\[
B_{c2}(0) = -0.693T_c \left( \frac{dR_{c2}}{dT} \right)_{T=T_c}.
\]
(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 \( \left( \frac{dR_{c2}(T)}{dT} \right)_{T=T_c} \), which is a crucial quantity for the determination of \( B_{c2} \). The slope \( \left( \frac{dR_{c2}(T)}{dT} \right)_{T=T_c} \) under a particular \% of \( R_c \) can be calculated.

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.
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 $\frac{dB}{dT}$, $T_c$ 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 $\left(\frac{dB_c(T)}{dT}\right)_{T=T_c}$ 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:
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 \( \sim 91 \) 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 \( \sim 76 \) T and 68 T for SC/FM bilayers BL1 and BL2 respectively. The corresponding values in the GL formalism are \( \sim 104 \) 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 \( \sim 46 \) 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]:

\[
\frac{\Delta(n_{qp})}{\Delta(0)} \approx 1 - \frac{2n_{qp}}{4N(0)\Delta(0)},
\]

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 \( \Delta(n_{qp}) \) 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

### 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) | \( \frac{\Delta B(T)}{\Delta T} \) \( T = T_c \) | \( B_{c2\text{WHH}}(0) \) (x 10^7 T) | \( B_{c2\text{GL}}(0) \) (x 10^7 T) | \( \xi_{\text{WHH}(0)} \) (\AA) | \( \xi_{\text{GL}(0)} \) (\AA) |
|--------|-------|--------------|-----------------------------------|----------------------------------|----------------------------------|------------------|------------------|
| 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 \( \frac{\Delta B(T)}{\Delta T} \) \( T = T_c \) with % \( R_f \) for YBCO thin film.
YBCO layer. This thickness is comparable to the spin diffusion length \( l_{sd} = (\hbar v_F T_F)^{0.5} \sim 10 \text{ nm} (T \rightarrow 0) \) and 80 nm \((T \rightarrow T_c)\) respectively in YBCO as calculated by Yeh et al [42] and others [43, 44]. Here, \( \hbar \) is the mean free path in YBCO, \( v_F \) is the Fermi velocity and \( T_F \) 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 \( d_S \gg l_{sd} \) [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 \( \xi_S = \left( \frac{\hbar v_F}{k_B T_c} \right)^{0.5} \) and \( \xi_F = \left( \frac{\hbar v_F}{\Delta E_F} \right) \) where \( \xi_S \) and \( \xi_F \) 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 \( \Delta E_F \) 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 \( \Delta E_F \sim (1-3) \text{ eV} \) and \( v_F \sim 10^7 \text{ cm s}^{-1} \), \( \xi_S \) is estimated to be \( \sim (0.2-0.7) \text{ nm} \) whereas \( \xi_F \) is reported to be \( \sim 1 \text{ nm} \). We estimate the in plane GL coherence length \( \xi_{co} \) to be of the order of \( \sim 2 \text{ nm} \) for YBCO and bilayers under 50% \( R_F \) criterion. These lengths (listed in table 1) are calculated using the GL relation \( B_{2d}(0) = \phi_0 / 2\pi \xi_{co}^2 \), where \( \phi_0 \) 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 \( \xi_S \) suggests that only a small thickness \( \sim (1-2) \text{ nm} \) 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 \( \mu_B \) to 2.9 \( \mu_B \) when its thickness increases approximately from 5 to 30 nm [47]. Therefore, we conjecture the effective magnetic field experienced by 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 \( B_{c2} \) 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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