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Appearance of an inhomogeneous superconducting state in $La_{0.67}Sr_{0.33}MnO_3$–$YBa_2Cu_3O_7$–$La_{0.67}Sr_{0.33}MnO_3$ trilayers

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Abstract. An experimental study of proximity effect in $La_{0.67}Sr_{0.33}MnO_3$–$YBa_2Cu_3O_7$–$La_{0.67}Sr_{0.33}MnO_3$ trilayers is reported. Transport measurements on these samples show clear oscillations in critical current (I_c) as the thickness of $La_{0.67}Sr_{0.33}MnO_3$ layers (d_F) is scanned from ∼50 Å to ∼1100 Å. In the light of existing theories of ferromagnet–superconductor (FM–SC) heterostructures, this observation suggests a long range proximity effect in the manganite, modulated by its weak exchange energy (∼2 meV). The observed modulation of the magnetic coupling between the ferromagnetic LSMO layers as a function of d_F, also suggests an oscillatory behavior of the SC order parameter near the FM–SC interface.

Keywords. Proximity effect; inhomogeneous superconductivity.

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1. Introduction

The interplay between superconductivity (SC) and ferromgnetism (FM) in FM–SC heterostructures leads to some interesting physical phenomena, one of which is the observed non-monotonic dependence of transition temperature T_c [1–6] and critical current I_c [7,8] on the thickness of ferromagnetic layer (d_F). Theoretically, such systems are treated as a boundary value problem, solving the Usadel equations [9] for anomalous Green's functions on both sides of the FM–SC interface. The pioneering work of Radovic *et al* [10] using this formalism, has successfully reproduced the non-monotonic behavior of T_c in FM–SC multilayers [1–3] and SC–FM–SC trilayers [2]. Similar calculations by Buzdin *et al* [11] have revealed an oscillatory nature of Josephson current across a ferromagnetic spacer. However, the Radovic–Buzdin (RB) theory, which relies on competing '0' and 'π' phase coupling between adjacent superconducting layers to explain the non-monotonic nature of T_c and I_c, cannot be applied to systems where there is only one SC layer in contact with a ferromagnetic film such as the FM–SC–FM trilayer and FM–SC bilayer structures. Another restricted point of the RB theory is the assumption of perfect transparency of the FM–SC interface. Recently, these issues have been addressed using more realistic boundary conditions [12–14]. In general, the microscopic basis of these theories is the formation of a Larkin–Ovchinnikov–Fulde–Ferrel (LOFF)-like [15,16] inhomogeneous SC-state at the FM–SC boundary [17].

On the experimental scenario, there are increasing number of reports [1–6] on the oscillating nature of $T_c(d_F)$ although negative results [18] and different interpretations [19] have also been reported in some cases. A more sensitive way of addressing this issue is the measurement of critical current $I_c(d_F)$ through the FM–SC interface. Such studies [7,8] have unambiguously established the existence of an oscillating order parameter. However, these results are explained on the basis of π-phase coupling between two superconducting layers. In this paper, we report the observation of oscillating critical current in FM–SC–FM trilayer structures where the concept of π-coupling does not apply altogether. Unlike the itinerant ferromagnet-weak coupling BCS superconductor-based structures, the constituents in the present case are exotic, showing localized spin ferromagnetism and a highly anisotropic superconducting order parameter. This first-time observation of an oscillating I_c in such a system is remarkable.

2. Experimental results

2.1 *Magnetic behavior of the LSMO boundary*

We have studied a series of high quality trilayer structures in FM–SC–FM geometry with a ∼100 Å superconducting $YBa_2Cu_3O_7$ (YBCO) layer sandwiched between ferromagnetic $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) layers, prepared by pulsed laser ablation on single crystal $SrTiO_3$ substrates. Thickness of the LSMO layers (d_F) were varied from ∼50 Å to ∼1100 Å. Details of film growth are described elsewhere [20]. The suitability of the CMR manganite high-T_c superconductor combination for epitaxial growth is also well-established in the literature [21–24].

The magnetic nature of the LSMO layers as a function of thickness was established from transport and magnetization measurements. figure 1 shows the resistivity ($\rho(T)$) of few representative LSMO thin films in the temperature range of 2 K and 370 K. Resistivity of these films at room temperature is low (∼2 mΩ cm), and remains metallic down to 2 K. The paramagnetic metallic phase above the Curie temperature (T_{Curie}) [25] which transits to a ferromagnetic metallic phase at $T < T_{Curie}$, is clearly identifiable in all films. The ordering temperature acquires the near bulk value (∼350 K) in films thicker than 200 Å, while thinner films show a slight drop in T_{Curie}, consistent with earlier measurements on ultra-thin LSMO films [26]. We have estimated the ferromagnetic exchange energy by fitting the 1000 Oe field-cooled $M_s(T)$ measurements (shown in figure 1) to the Bloch relation $M_s(T)/M_s(0) = 1 - AT^{3/2}$. Here $A = (C/S)(k_B/2E_{ex}S)^{3/2}$, where S is the total spin per Mn ion in LSMO and C is the Bloch constant with a value 0.059 for a cubic lattice [27]. All trilayer samples were rigorously checked for simultaneous occurrence of superconductivity and magnetism, using magnetization and transport measurements [20].

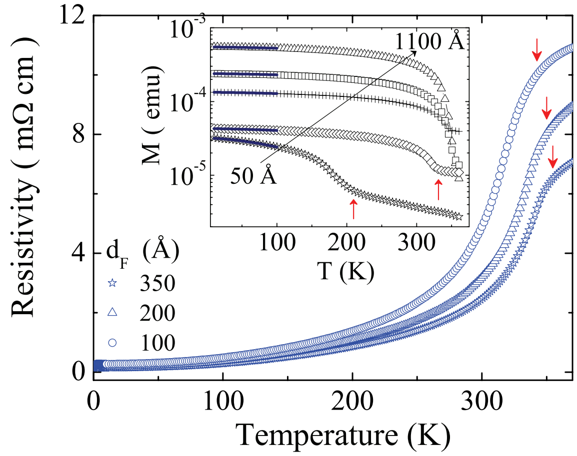

Figure 1. Resistivity ($\rho\,(T)$) of LSMO films deposited on STO in the temperature range of 2–370 K. Thickness of the films varies from 100 Å to 350 Å. Inset: The 1000 Oe field-cooled magnetization of single layer LSMO films of thickness ranges from 50 Å to 1100 Å. In all cases the magnetic field was applied in the plane of the film. The solid lines are fits to the Bloch relation (see text for details). Curie temperatures have been marked by the arrows.

2.2 *Transition temperature*

The superconducting transitions as seen in $\rho(T)$ measurements on various trilayers are presented in the inset of figure 2. The one-step transitions seen in this inset exclude the possibility of any metallurgical activities between LSMO and YBCO, which would otherwise lead to the formation of a degraded phase of YBCO at the interface, with lower T_c. The transition temperature $T_c(d_F)$ of the trilayers normalized with respect to the T_c of a trilayer with only 50 Å LSMO on both sides of YBCO is plotted in figure 2 as a function of d_F. The $T_c(d_F)$ has been defined as the temperature at which the sample resistance reaches half the extrapolated normal state resistance. Figure 2 also shows the variation of exchange energy (E_{ex}) extracted from the $M(T)$ data of figure 1 with d_F. The calculated value of E_{ex} in the thick limit (∼2 meV) is in good agreement with the results obtained directly from ferromagnetic resonance measurements on similar films [28]. The decay of T_c with d_F in figure 2 is primarily monotonic except for the appearance of a plateau in the neighborhood of $d_F \sim 450$ Å. The absence of oscillations in the $T_c(d_F)$ curve suggests a limited transparency of the FM–SC interface. In spite of the near perfect lattice matching between LSMO and YBCO some uncontrollable factors, like the Fermi-velocity mismatch between the two materials in contact may lead to a smearing of the $T_c(d_F)$ oscillations [13].

2.3 *Critical current*

The critical current $I_c(d_F)$ was measured in a standard four-probe geometry, as shown in a sketch in figure 3a. Although in the normal state both LSMO and

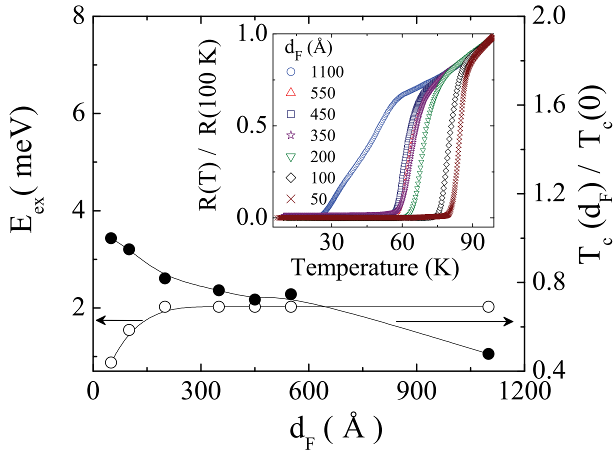

Figure 2. Superconducting transition temperature (T_c) of trilayers, normalized with respect to T_c of a 100 Å YBCO in LSMO–YBCO–LSMO trilayer, is plotted (on right y-axis) with thickness (d_F) of LSMO boundaries. The exchange energies of the corresponding single layer LSMO films (calculated from the fittings in the inset of figure 1) is plotted on the left y-axis. The solid lines are only guides to the eyes. Inset shows the resistive transitions of the trilayers into the superconducting state as a function of temperature.

YBCO layers act as parallel conducting channels for the current, in the superconducting state current is preferentially directed into the YBCO. However, owing to the small thickness of the superconducting channel in our trilayers and the induced superconducting order at the boundary, the proximally important interface region of the FM layers (shaded portion at the LSMO–YBCO interface, shown in the sketch of figure 3a) also contributes to the flow of supercurrent. Clearly, as the YBCO thickness is fixed in all cases, magnitude of I_c is expected to reflect the relative amplitude of the pair-wave function in different samples. The critical current I_c has been extracted from the measurements of current–voltage characteristics, as shown in the inset of figure 3b for a trilayer with $d_F \sim 350$ Å. In figure 3a, we show the I_c of all trilayers as a function of temperature. The behavior of I_c is clearly non-trivial as the thickness of LSMO boundaries in these heterostructures is varied. The same data have been plotted as isothermal curves at several temperatures as a function of d_F in figure 3b. The behavior of I_c is most certainly oscillatory with an average period of ∼250 Å and more than an order of magnitude change in current between the maxima and minima. Theoretically, this period corresponds to the distance over which the induced pair wave function changes its phase by π according to the relation ($\pi\hbar v_F/E_{ex}$) [17]. Assuming a LOFF-like picture for the current situation and using the measured exchange energy (2 meV), we obtain a Fermi velocity $v_F \sim 2.4 \times 10^6$ cm/s, which is somewhat different from the value ($\sim 7.4\times10^7$ cm/s) derived from band structure calculations [29]. This discrepancy might be a reflection of the large uncertainty involved in determining the value of v_F for CMR manganites from band structure calculations, due to strong hybridization effects of Mn-d and O-p bands.

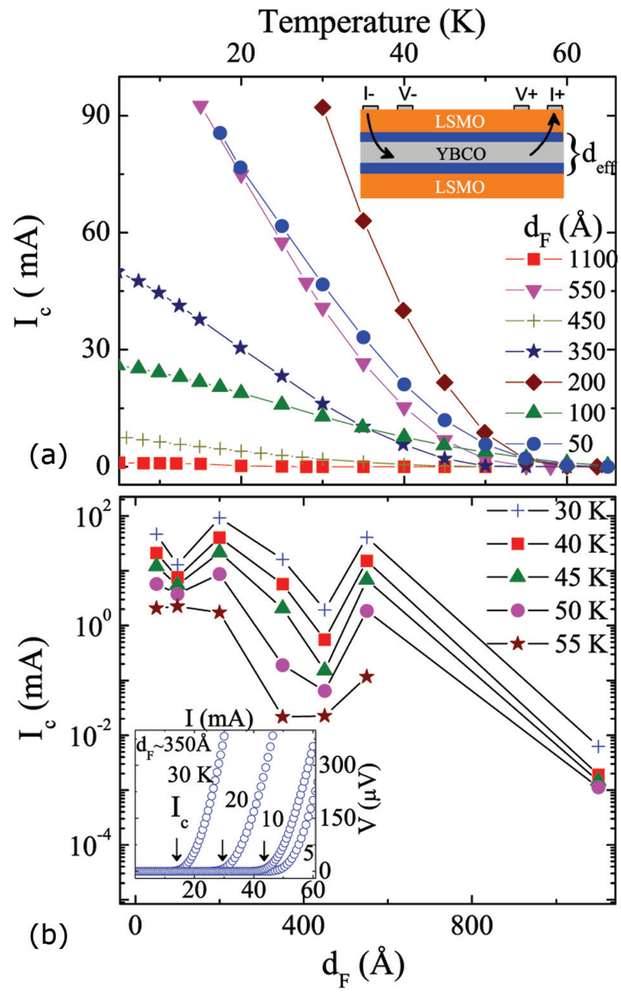

Figure 3. (**a**) In-plane critical current (I_c) of LSMO–YBCO–LSMO trilayers plotted as a function of temperature. Inset is a sketch of the measurement geometry. The shaded portions at the YBCO–LSMO interfaces are the inhomogeneous superconducting regions which contribute to the overall critical current of the system. (**b**) The data of panel (a) have been re-plotted as $I_c(d_F)$ isotherms at several temperatures. Inset shows the IV curves of a trilayer with $d_F \sim 350$ Å at temperatures 5, 10, 20 and 30 K. Arrows indicate the critical current I_c.

2.4 *Magnetic coupling*

To further verify the oscillating nature of $I_c(d_F)$, we conducted DC-magnetization measurements where diamagnetic supercurrents are intrinsically generated inside the YBCO layer in response to the applied magnetic field. These measurements were performed with an in-plane field geometry which produces the screening

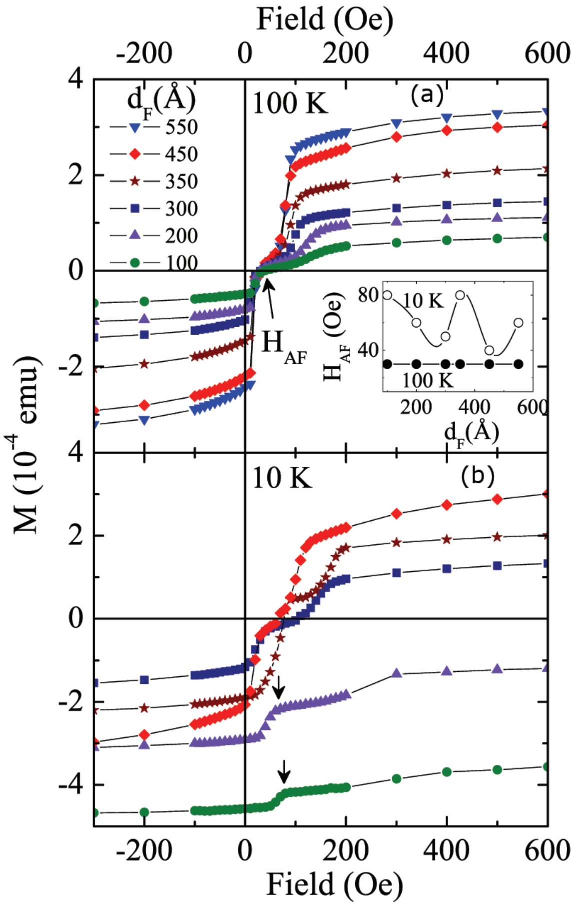

Figure 4. Panels (**a**) and (**b**) show the last two quadrants of zero-field-cooled hysteresis curves at 100 K and 10 K respectively. All measurements were carried out with a field-in-plane geometry. The inset in panel (a) compares the antiferromagnetic coupling field H_{AF} (indicated by arrows in some cases) at temperatures 10 K and 100 K, as a function of the thickness of LSMO boundary (d_{F}). The solid lines in the inset are only guides to the eyes.

currents along the cross-section of the trilayers. The diamagnetic moment of this induced current acts as an opposing field which suppresses the effective magnetic field felt by the LSMO layers. Therefore, a change in the induced current (equivalently the diamagnetic moment) should be detectible from the magnetic coupling behavior of the LSMO boundaries. Zero-field-cooled magnetization measurements on our trilayer samples revealed a clear region of antiferromagnetic coupling between the moments of the top and the bottom LSMO layers at low fields (<200 Oe), as manifested by a plateau in the magnetization curve. Figure 4a shows the last two quadrants of the hysteresis loops measured at 100 K, where the YBCO is still in the normal state. The antiferromagnetic coupling field (H_{AF}) extracted from the

M–H loops at 100 K is found to be the same (30±5 Oe) for all samples. Panel (b) of figure 4 shows the magnetization measured at 10 K. Here the ferromagnetic contribution of the LSMO layers is superimposed on the strong diamagnetic moment of YBCO. However, the plateau arising from antiferromagnetic coupling between the LSMO layers is still observable. Furthermore, in clear contrast to the data at 100 K, the coupling field H_{AF} in this case is oscillatory with d_{F}, as shown in the inset of figure 4a. The oscillatory behavior appears to be a signature of the modulation of screening critical currents. Most interestingly, the period of oscillation in this case is found to be ∼200 Å, which is close to the period (∼250 Å) obtained earlier from transport $I_{\mathrm{c}}(d_{\mathrm{F}})$. The large range of proximity effect seen here is consistent with the results of Kasai *et al* [30], who have reported a measurable supercurrent across YBCO–LSMO–YBCO trilayer junctions with LSMO spacers of the order of 1000 Å.

3. Discussion

As already mentioned, the current observations can not be explained on the basis of π-phase coupling, since here we have only one superconducting layer. This difficulty has been addressed by more recent theories [12–14], predicting similar oscillations in heterostructures consisting of a single superconducting layer. We, however, realize the difficulty in mapping the current situation onto these theories which have been developed assuming the s-wave symmetry of the superconductors order parameter. On the other hand, there is overwhelming experimental evidence for a d-wave pairing symmetry in YBCO, with pair transport along the c-axis occurring only via Josephson tunneling. However, a few points independent of the symmetry of the order parameter can be picked up for a qualitative analysis. The non-monotonic changes in the superconducting properties with d_{F} can be understood from the predicted [31] non-monotonic drop in the pair-amplitude at the FM–SC interface, constrained by a maximum at the outer boundary of the ferromagnet. When a node (minimum) of the pair wave function appears at the FM–SC interface, the Cooper pairs entering the ferromagnet die quickly. On the other hand, an antinode at the interface provides better chances of survival for the Cooper pairs. Thus, the appearance of nodes and antinodes at the interface should manifest as a minimum and maximum in $T_{\mathrm{c}}(d_{\mathrm{F}})$ and $I_{\mathrm{c}}(d_{\mathrm{F}})$ curves.

The exact mechanism by which the supercurrent is continued as a quasiparticle current in an adjacent ferromagnetic layer is not yet known. However, the zero energy Andreev bound states, believed to be the origin of zero-bias conductance peaks (ZBCP) observed in HTSC, might play a role here. Kasiwaya *et al* [32,33] have shown that such bound states may lead to a spontaneous quasiparticle current across a ferromagnet $d_{x^2-y^2}$-wave superconductor junction depending on the phase of order parameter at the interface, when the interface is perpendicular to the ab-plane. Interestingly, the ZBCP is also seen in LSMO–YBCO junctions where the granularity of the c-axis oriented YBCO leads to sampling of the ab-plane Andreev bound states [34].

4. Conclusions

In conclusion, we have observed clear oscillations in critical current of LSMO–YBCO–LSMO trilayers as a function of the LSMO thickness. The period of oscillation was found to be large (∼200 Å). This non-monotonic behavior appears to be a manifestation of the LOFF-like oscillatory superconducting order parameter near the FM–SC interface in the limit of weak exchange energy ($E_{\rm ex} \ll k_{\rm B}T_{\rm c}$). The magnetic coupling behavior of the LSMO boundaries also points towards similar results. To our knowledge, this is the first observation of oscillatory critical current as a function of $d_{\rm F}$ in a manganite–cuprate heterostructure.

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