Appearance of an inhomogeneous superconducting state in La$_{0.67}$Sr$_{0.33}$MnO$_3$-YBa$_2$Cu$_3$O$_7$-La$_{0.67}$Sr$_{0.33}$MnO$_3$ trilayers

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An experimental study of proximity effect in La$_{0.67}$Sr$_{0.33}$MnO$_3$ - YBa$_2$Cu$_3$O$_7$ - 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 $\sim$ 50 Å to $\sim$ 1100 Å. In the light of existing theories of ferromagnet-superconductor (FM-SC) heterostructures, this observation suggests a long range proximity effect in the manganese, modulated by its weak exchange energy ($\sim$ 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.

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The interplay between superconductivity (SC) and ferromagnetism (FM) in FM-SC heterostructures leads to some interesting physical phenomena, one of which is the observed non-monotonic dependence of transition temperature $T_c$ and critical current $I_c$ on the thickness of ferromagnetic layer ($d_F$). Theoretically, such systems are treated as a boundary value problem, solving the Usadel equations for anomalous Green’s functions on both sides of the FM-SC interface. The pioneering work of Radovic et al. has successfully reproduced the non-monotonic behavior of $T_c$ in FM-SC multilayers and SC-FM-SC trilayers. Similar calculations by Buzdin et al. have revealed an oscillatory nature of Josephson current across a ferromagnetic spacer. However, the Radovic-Buzdin (RB) theory, which relies on competing “0” and “$\pi$” 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. In general, the microscopic basis of these theories is the formation of a Larkin-Ovchinnikov-Fulde-Ferrel (LOFF) state at the FM-SC boundary.

On the experimental scenario, there are increasing number of reports on the oscillating nature of $T_c(d_F)$ although negative results and different interpretations 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 FM-SC interfaces. Such studies have unambiguously established the existence of an oscillating order parameter. However, these results are explained on the basis of $\pi$-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 $\pi$-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.

We have studied a series of high quality trilayer structures in FM-SC-FM geometry with a $\sim$100 Å superconducting YBa$_2$Cu$_3$O$_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 $\sim$50 Å to $\sim$1100 Å. Details of film growth are described elsewhere. The suitability of the CMR manganite-high $T_c$ superconductor combination for epitaxial growth is also well-established in the literature.

FIG. 1: (a) Resistivity ($\rho(T)$) of LSMO films deposited on STO in the temperature range of 2 K - 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 ranging 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.
The magnetic nature of the LSMO layers as a function of thickness was established from transport and magnetization measurements. Fig. 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 ($\sim 2 \text{ m}\Omega\text{cm}$), and remains metallic down to 2 K. The paramagnetic metallic phase above the Curie temperature ($T_{\text{Curie}}$) which transitions to a ferromagnetic metallic phase at $T < T_{\text{Curie}}$, is clearly identifiable in all films. The ordering temperature acquires the near bulk value ($\sim 350 \text{ K}$) in films thicker than 200 Å, while thinner films show a slight drop in $T_{\text{Curie}}$, consistent with earlier measurements on ultrathin LSMO films. We have estimated the ferromagnetic exchange energy by fitting the 1000 Oe field-cooled $M_s(T)$ measurements (shown in Fig. 1) to the Bloch relation $M_s(T)/M_s(0) = 1 - AT^{3/2}$. Here $A = (C/S)(k_B/2E_{xx}S^{3/2})$, 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. All trilayer samples were rigorously checked for simultaneous occurrence of superconductivity and magnetism, using magnetization and transport measurements.

The superconducting transitions as seen in $\rho(T)$ measurements on various trilayers are presented in the inset of Fig. 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 Fig. 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. Fig. 2 also shows the variation of exchange energy ($E_{xx}$) extracted from the $M(T)$ data of Fig. 1 with $d_F$. The calculated value of $E_{xx}$ in the thick limit ($\sim 2 \text{ meV}$) is in good agreement with the results obtained directly from ferromagnetic resonance measurements on similar films. The decay of $T_c$ with $d_F$ in Fig. 2 is primarily monotonic except for the appearance of a plateau in the neighborhood of $d_F \sim 450 \text{ Å}$. 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.

The critical current $I_c(d_F)$ was measured in a standard four-probe geometry, as shown in a sketch in Fig. 3(a). Although in the normal state both LSMO and YBCO layers act as parallel conducting channels for the current, in the superconducting state current is preferen-
tially 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 Fig. 3 (a)) 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. The critical current $I_c$ reflects the relative amplitude of the pair-wave function.

The critical current $I_c$ has been extracted from the measurements of current-voltage characteristics, as shown in the inset of Fig. 3 (b) for a trilayer with $d_F \sim 350$ Å. In Fig. 3(a) 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 Fig. 3(b). The behavior of $I_c$ is most certainly oscillatory with an average period of $\sim 250$ 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 $\pi$ according to the relation $(\pi \hbar \nu_F / E_{ex})$. 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/sec, which is somewhat different from the value ($\sim 7.4 \times 10^7$ cm/sec) 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.

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 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. Fig. 4(a) shows the last two quadrants of the hysteresis loops measured at 100 K, where the YBCO is still in the normal state. The antiferromagnetically 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 Fig. 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 Fig. 4(a). 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 $\sim 200$ Å, which is close to the period ($\sim 250$ Å) obtained earlier from transport $I_c(d_F)$. 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.

As already mentioned, the current observations can not be explained on the basis of $\pi$-phase coupling, since here we have only one superconducting layer. This difficulty has been addressed by more recent theories [12,13,14], 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 \[ 21 \] 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 \( T_c(d_F) \) and \( L_c(d_F) \) curves.

The exact mechanism by which the supercurrent is continued as a quasiparticle current in an adjacent ferromagnetic layer is not known yet. 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. \[ 32 \] 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 ab-plane Andreev bound states \[ 34 \].

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 (\( \sim 200 \) \( \AA \)). 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 (\( E_{xx} \ll k_BT_c \)). 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 \( d_F \) in a manganite-cuprate heterostructure.

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