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Physica C 418 (2005) 138-143



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Oscillatory superconducting transition temperature in YBa₂Cu₄O₈/La_{2/3}Ca_{1/3}MnO₃/YBa₂Cu₄O₈ heterostructures

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Received 2 August 2004; received in revised form 12 November 2004; accepted 23 November 2004 Available online 15 December 2004

Abstract

YBa₂Cu₄O₈/La_{2/3}Ca_{1/3}MnO₃/YBa₂Cu₄O₈ (Y-124/LCMO/Y-124) heterostructures were prepared by facing-target sputtering technique. The oscillatory behavior of superconducting transition temperature ($T_{\rm C}$) with the thickness of LCMO ($d_{\rm L}$) has been observed. The strongest nonmonotonic information in $T_{\rm C}$ - $d_{\rm L}$ curves appears clearly when $d_{\rm L}$ is larger than the critical thickness $d_{\rm L}^{\rm CR}$. The metal-insulator transition temperature ($T_{\rm MI}$) can only be detected at $d_{\rm L} > d_{\rm L}^{\rm CR}$. For LCMO/Y-124/LCMO trilayer films, the $T_{\rm MI}$ has also shown an oscillation with decreasing spacer layer thickness. These kinds of symmetric dependence on the spacer layer in LCMO/Y-124 systems suggest strongly the interplay of ferromagnetic and superconducting couplings.

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PACS: 74.78.Bz; 75.47.Gk; 74.78.Fk; 71.70.Gm *Keywords:* High- T_C films; Colossal magnetoresistance; Heterostructure; Interlayer coupling

1. Introduction

Since the advent of improved growth capabilities, the possibility to study proximity effects between layers with dissimilar intrinsic properties, such as superconductivity and ferromagnetism, has initiated much research activity [1]. For ferromagnetic (F) layers sandwiched between superconducting (S) layers it is expected that the superconducting transition temperature (T_C) decrease monotonically with increasing magnetic layer thickness. In fact interest in this topic increased considerably after Wong et al. [2] showed

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^{0921-4534/\$ -} see front matter @ 2004 Elsevier B.V. All rights reserved. doi:10.1016/j.physc.2004.11.019

a nonmonotonic dependence of $T_{\rm C}$ as a function of the Fe layer thickness in V/Fe superlattice with fixed V thickness. Shortly afterwards, the possibility for oscillation of $T_{\rm C}$ as a function of the ferromagnetic layer thickness in S/F multilayers was demonstrated theoretically [3]. It was shown that for specific F layer thickness the Josephson coupling between two S layers can lead to a junction with an intrinsic phase difference $\Delta \phi = \pi$ which, in turn, exhibits a higher $T_{\rm C}$.

Theoretical treatments of superconductorferromagnet-superconductor junctions have been made over the years. A critical thickness of a ferromagnetic barrier, above which there is no overlap of superconducting wave functions and hence zero Josephson current, is predicted by Kuplevakhskii and Fal'ko [4]. On the other hand, Buzdin et al. [5] reported an oscillation of the critical current of the junction with the ferromagnetic barrier thickness. Experiments on low $T_{\rm C}$ Pb–Fe– Pb junctions by Claeson [6] indicated a critical thickness of 0.5 nm. This small value may be due to efficient pair breaking by spin flip scattering at the Pb-Fe interfaces. Radovic et al. [3] have suggested that the unusual effects predicated, e.g., the oscillatory $T_{\rm C}$ with changing the thickness of ferromagnetic layer should be much more pronounced in S/F superlattices with high $T_{\rm C}$ and short coherence length superconductor. Since the discovery of the high $T_{\rm C}$ superconductivity, tremendous experimental results have been reported. Due to the structural similarity of the two classes of perovskite compounds, high $T_{\rm C}$ superconductor (HTS) and colossal magnetoresistance (CMR) ferromagnet, the unique combination of the two oxide layers is possible. Kasai et al. [7] reported the observation of supercurrents in trilayer junctions with a 500 nm thick $La_{0.7}Ca_{0.3}MnO_z$ ferromagnetic barrier. Recently, Holden et al. [8] investigated ellipsometric measurements of the far-infrared dielectric properties of YBa₂Cu₃O₇/ La_{2/3}Ca_{1/3}MnO₃ superlattices. Their results provided clear evidence that the free-carrier response is strongly suppressed in these structures as compared to the one in pure YBa₂Cu₃O₇ and La_{2/3}Ca_{1/3}MnO₃ films. Pena et al. [9] found experimental evidence for coupling between superconducting layers through ferromagnetic spacer in

YBa₂Cu₃O₇/La_{0.7}Ca_{0.3}MnO₃ superlattices consistent with a long-range F/S proximity effect. In our previous work, we found that the $T_{\rm C}$ of YBa₂Cu₄O₈ layer was strongly suppressed by ferromagnetic La_{2/3}Ca_{1/3}MnO₃ layers due to the magnetic proximity effect in La_{2/3}Ca_{1/3}MnO₃/YBa₂Cu₄O₈/La_{2/3}Ca_{1/3}MnO₃ sandwiches [10].

In this paper we report a complex $T_{\rm C}$ oscillation phenomena for YBa₂Cu₄O₈ (Y-124)/La_{2/3}Ca_{1/3} MnO₃ (LCMO)/Y-124 heterostructures. Here, we choose Y-124 only because it is much more stable than YBa₂Cu₃O₇ in different oxygen environment. The strong dependence of the $T_{\rm C}$ on the LCMO layer thickness is studied. We find a long-range proximity effect which yields coupling between superconducting Y-124 layers through the ferromagnetic spacer layer.

2. Experimental

All samples were fabricated on $10 \text{ mm} \times 3 \text{ mm}$ (001) SrTiO₃ (STO) single crystal substrates from pure LCMO and Y-124 targets under a range of conditions by facing-target sputtering technique [11,12]. The film thickness was controlled by sputtering time with the deposition rate being carefully calibrated (0.05 nm/s for LCMO layer, 0.075 nm/s for Y-124 layer). The as-deposited films exhibited a mirror-like surface and were adherent well on all substrates employed. From the z-profile, the roughness in z-direction is less than 5 nm [10]. The surface morphology reveals the symmetry of the underlying lattice surface and shows oriented, layered, square islands of about the same size [10]. The resistance as a function of temperature was measured by the standard four-probe technique with CIP (current in plane) geometry and the distance between voltage contacts was fixed at 6 mm. The magnetic moment of the samples as functions of temperature and applied magnetic field was measured by utilizing a vibrating sample magnetometer. During the measurements, a magnetic field was applied parallel to the film surface. A small nonhysteretic contribution from the STO substrate was eliminated by separately measuring its diamagnetic response.

3. Results and discussion

Fig. 1 shows the temperature dependence of the relative resistivity $(\rho/\rho_{100 \text{ K}})$ and its temperature coefficient $(d(\rho/\rho_{100 \text{ K}})/dT)$ of Y-124 (75 nm)/LCMO (75 nm)/Y-124 (75 nm) film. The resistivity curve shows the metal-insulator transition of LCMO layer at high temperature, and the cross point at low temperature arises from the superconducting transition of Y-124 layers. The metal-to-insulator transition temperature, T_{MI} and T_{C} are defined as the peak points in the differential curve as shown in Fig. 1.

The hysteresis loops of Y-124 (75 nm)/LCMO (75 nm)/Y-124 (75 nm) film are shown in Fig. 2 with the magnetic field applied parallel to the layers. Magnetization loops at 120 K shows a saturation field of 1000 Oe, and resembles that of a single LCMO film [13]. The sample displays a characteristic superconducting like hysteresis loop below the $T_{\rm C}$ and the flattened slop probably results from a small stray field due to the magnetic layer when H > 0 Oe. Measurement of the zero field cooling (ZFC) magnetic moment as a function of temperature (see the inset) shows a clear ferromagnetic transition at 170 K. At lower temperature (below 70 K), the signature of the diamagnetic response of the Y-124 layer shows up, reducing the total magnetic moment of the sample, which becomes negative at lower temperatures.



Fig. 1. The temperature dependence of the resistivity $(\rho/\rho_{100 \text{ K}})$ and its temperature coefficient $(d(\rho/\rho_{100 \text{ K}})/dT)$ of Y-124 (75 nm)/LCMO (75 nm)/Y-124 (75 nm) film. T_{MI} and T_{C} are defined as the peak points in the differential curve as shown in the inset.



Fig. 2. Magnetic hysteresis loops of Y-124 (75 nm)/LCMO (75 nm)/Y-124 (75 nm) film with magnetic field parallel to the film plane at 15, 40 and 120 K, respectively. The inset displays the temperature dependence of ZFC magnetization with an applied field of 1000 Oe.

 $T_{\rm MI}$ of as-deposited Y-124/LCMO/Y-124 sandwiches are plotted against the thickness of LCMO layer $d_{\rm L}$ in Fig. 3. $T_{\rm MI}$ is independent of its thickness for films with thick spacer layer. Degradation of $T_{\rm MI}$ has been found on films with thin LCMO layer. The extrapolation gives critical thickness $d_{\rm L}^{\rm CR}$ below which the $T_{\rm MI}$ can not be detected. $d_{\rm L}^{\rm CR}$ is about 5.5, 7.0 and 9.0 nm for $d_{\rm Y} = 7.5$, 45 and 75 nm respectively, here, $d_{\rm Y}$ is the thickness of YBCO layer. Jin et al. studied the effects of film thickness for the La_{0.67}Ca_{0.33}-MnO₃ film as early as 1995, and concluded that 100 nm is a promising thickness for CMR effect



Fig. 3. $T_{\rm C}$ (\bigcirc) and $T_{\rm MI}$ (\bullet) of (a) Y-124 (7.5 nm)/LCMO ($d_{\rm L}$)/Y-124 (7.5 nm), (b) Y-124 (45 nm)/LCMO ($d_{\rm L}$)/Y-124 (45 nm) and (c) Y-124 (75 nm)/LCMO ($d_{\rm L}$)/Y-124 (75 nm) as functions of $d_{\rm L}$. The lines are for a guide to the eyes.

[14]. Rao et al. accessed similar issue, and observed a significant enhancement of strain and an accompanied weakening of ferromagnetic order in ultrathin film [15]. In a recent paper, Sun et al. found a sharp drop of $T_{\rm MI}$ in LCMO film if $d_{\rm L} < 40$ nm, and even without MR transition for $d_{\rm L} \leq 3$ nm [16]. In our case, the larger threshold value $d_{\rm L}^{\rm CR}$ may be corresponding to the nonmagnetic (or magnetically 'dead') interface layer resulted from the roughness at the interface [17].

Fig. 3 also shows the $d_{\rm L}$ dependence of $T_{\rm C}$ for Y-124/LCMO/Y-124 trilayer films. For $d_{\rm Y}$ = 7.5 nm, $T_{\rm C}$ exhibits a rapid drop with increasing $d_{\rm L}$ and is not detected when $d_{\rm L} > 6$ nm in our setup. In the other two series, $T_{\rm C}$ exhibits a minimum with increasing $d_{\rm L}$ up to $d_{\rm 1}$ for the ultra-thin LCMO layer. As $d_{\rm L}$ is further increased from $d_1T_{\rm C}$ rises to a maximum at $d_2 \sim 30$ nm. The $T_{\rm C}$ is essentially independent of $d_{\rm L}$ for $d_{\rm L} \gg 50$ nm and is ~ 65 and 70 K for $d_{\rm Y} = 45$ and 75 nm when $d_{\rm L}$ = 80 nm. In a study of Fe/Nb system for different magnetic layer thickness, $T_{\rm C}$ reaches the first minimum $d_{\rm Fe}$ and ferromagnetism appears at almost the same Fe thickness as d_{Fe} [18,19]. In the Gd/Nb system the first minimum of $T_{\rm C}$ also correlates with the ferromagnetic ordering of the Gd layer [20,21]. In contrast, it should be pointed out that $T_{\rm MI}$ is smaller than Curie temperature T_f for our thin samples with low T_f and it is not sure that the magnetic proximity effect is still effective in the range, i.e., $d_{\rm L} < d_{\rm L}^{\rm CR}$ since T_f of these samples can not be determined. So, d_1 cannot be directly connected to the onset of ferromagnetism in the LCMO layers.

The thickness dependence of resistivity at 90 K is shown in Fig. 4. It is interesting to note that the resistivity increases rapidly when $d_{\rm L}$ is increased from 0 to about 5 nm, a value close to d_1 above which the roughness at the interface becomes stable and independent of the increased layer thickness. d_1 may be partially resulted from the interface roughness effect. Although no evidence existed for nonmagnetic interfacial layers in Y-124/LCMO/Y-124 sandwiches, this mechanism, inelastic electron scattering in intermixed nonmagnetic layers at the interface may induce strong repulsive interaction on the Cooper pair, may play a role for the minimum of $T_{\rm C}$ at d_1 [18,19]. Fur-



Fig. 4. Dependence of resistivity of Y-124/LCMO/Y-124 sandwiches at 90 K on spacer layer thickness $d_{\rm L}$. The lines are for a guide to the eyes.

thermore, the maximum in $T_{\rm C}$ occurs at $d_2 > d_{\rm L}^{\rm CR}$. It is inferred that magnetic spin-flip scattering mechanism may be dominant with increase of $d_{\rm L}$ in an intermediate $d_{\rm L}$ region where ferromagnetic spin configuration is not so rigid due to twodimensional character of magnetism of the LCMO layers, and play an important role in causing the maximum of $T_{\rm C}$ at d_2 [22].

 $T_{\rm C}$ and $T_{\rm MI}$ of the as-deposited LCMO/Y-124/ LCMO films are plotted against $d_{\rm Y}$ in Fig. 5. For films with thick spacer, $T_{\rm C}$ is independent of its thickness. Degradation of $T_{\rm C}$ has been found for films with ultrathin Y-124 layer. Extrapolation gives $d_{\rm Y}^{\rm CR}$ below which the superconductivity is lost, and we have $d_{\rm Y}^{\rm CR} = 16$, 35 and 11.5 nm for



Fig. 5. $T_{\rm C}$ (\bigcirc) and $T_{\rm MI}$ (\bullet) of (a) LCMO (15 nm)/Y-124 ($d_{\rm Y}$)/LCMO (15 nm), (b) LCMO (50 nm)/Y-124 ($d_{\rm Y}$)/LCMO (50 nm) and (c) LCMO (100 nm)/Y-124 ($d_{\rm Y}$)/LCMO (100 nm) as functions of $d_{\rm Y}$. The lines are for a guide to the eyes. $T_{\rm MI}$ of LCMO single layer films (\Box) are shown in the figure.

the three series of $d_{\rm L} = 100$, 50 and 15 nm, respectively. The results indicate that the superconducting properties are strongly affected by the ferromagnetic layers due to the magnetic proximity effect [10]. $T_{\rm MI}$ is close to the one of the individual LCMO layer for samples with thick Y-124 layer, and shows a nonmonotonic behaviour with decreasing spacer layer thickness. When $d_{\rm Y} > d_{\rm Y}^{\rm CR}$ the peak values of $T_{\rm MI}$ are 257, 256 and 235 K for $d_{\rm L} = 15$, 50 and 100 nm respectively, much higher than those for 30, 100 and 200 nm LCMO films, indicating that the LCMO layers are strongly coupled in those samples with thin YBCO spacer. The exchange coupling between the LCMO layers is also found in [LCMO (10 nm)/ $Pr_{0.67}Ca_{0.33}MnO_3(PCMO)]_n$ superlattices when PCMO thickness is less than 2 nm, where the $T_{\rm MI}$ is enhanced [23]. The striking discovery in $T_{\rm MI}$ of our work shows that the magnetic interaction between the two LCMO layers is still survived when $d_{\rm Y} > 20$ nm.

The strongest nonmonotonic behaviors in $T_{\rm C}$ - $d_{\rm L}$ (Fig. 3) and $T_{\rm MI}$ - $d_{\rm Y}$ (Fig. 5) curves appear clearly at the region $d_{\rm L} > d_{\rm L}^{\rm CR}$ and $d_{\rm Y} > d_{\rm Y}^{\rm CR}$. In the HTS/CMR structure, Y-124 has high critical temperature and short coherence length, while LCMO is metallic with not so large pair breaking effects due to its weak magnetism. The wellmatched lattice between Y-124 and LCMO weakens the effects of roughness and strain. In addition, the oxygen diffusion between LCMO and Y-124 layers is negligible in our samples [10]. It is speculated that some coupling between the top and bottom layers, through the spacer layer, plays an important role.

A possible type of long-rang coupling is dipolar coupling due to the roughness of the interface [24–26]. The dipolar coupling between two perfectly flat infinite planes will vanish in the absence of roughness while in the presence of topographical inhomogeneities the magnetostatic coupling arising from surface magnetic dipoles created by roughness may originate a non-negligible coupling [26]. Also the dipolar interaction, under particular circumstances, can originate non-in-plane magnetizations. The roughness of samples is less than 5 nm [10]. We did not have the expression of dipolar interaction energy as a function of the distance between them, but the dipolar coupling may play a role in the oscillation with the spacer thickness. Furthermore, although pinholes may be present in LCMO/YBCO/LCMO films when $d_{\rm Y} < d_{\rm Y}^{\rm CR}$ $T_{\rm MI}$ is not enhanced and is the same as that for LCMO films, indicating that the magnetic short due to pinholes is ruled out.

Another possible mechanism may be related to the spin fluctuations in Y-124 and LCMO layers. The sandwiches can be considered to be a layered structure with Mn–O and Cu–O conduction planes which are stacked apart from each other. Chahara et al. think that spin fluctuations may be a key to understand the phenomena in HTS/CMR system [27]. In contrast, Holden et al. reported recently the long-range charge transfer from the HTS layer into the CMR layer for HTS/CMR superlattices from optical measurements, and concluded that spin diffusion (driven by the gradient in spin polarization between CMR and HTS layers) can not lead to a long-range spin polarization of the charge carriers deep inside the HTS layers [8].

In a word, our results provide evidence for interplay of ferromagnetic and superconducting couplings in HTS/CMR layered systems. To date, we do not have a complete microscopic understanding of these features yet. Further studies are required before one can distinguish between these equally fascinating possibilities. These results may be stimulating for the development of a theory of the HTS/CMR proximity effect.

4. Conclusion

The interlayer coupling of superconducting Y-124 thin films through ferromagnetic LCMO layer has been investigated from the transport properties of Y-124/LCMO/Y-124 sandwiches. With the increase of the LCMO layer thickness, the $T_{\rm C}$ exhibited an oscillation with a local minimum and a maximum. The strongest nonmonotonic information in $T_{\rm C}$ - $d_{\rm L}$ curves appears clearly at the region $d_{\rm L} > d_{\rm L}^{\rm CR}$. In LCMO/Y-124/LCMO trilayer films, with decreasing spacer layer thickness, the $T_{\rm MI}$ also shows an oscillatory behavior. The kind of symmetric dependence on the spacer layer suggests strongly the interplay of ferromagnetic and superconducting couplings in HTS/CMR layered systems.

Acknowledgements

This work was supported by the Research Foundation of Shandong Provincial Education Department of China (No. 03A05), the National natural Science Foundation of China (Nos. 50371102 and 10334070), and the Hong Kong Research Grant Council.

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