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Magnetotransport properties of $La_{2/3}Ca_{1/3}MnO_3$/$YBa_2Cu_4O_8$/$La_{2/3}Ca_{1/3}MnO_3$ sandwiches*

Zhao Kun(赵 昆)[a)b)†], Feng Jia-Feng(丰家峰)[c)],
Huang Yan-Hong(黄延红)[a)], and Wong Hong-Kuen(黄康权)[d)]

[a)] *Laboratory of Optical Physics, Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing* 100080, *China*

[b)] *International Center for Materials Physics, Chinese Academy of Sciences, Shenyang* 110016, *China*

[c)] *State Key Laboratory for Magnetism, Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing* 100080, *China*

[d)] *Department of Physics, The Chinese University of Hong Kong, Hong Kong, China*

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$La_{2/3}Ca_{1/3}MnO_3$ (LCMO)/$YBa_2Cu_4O_8$ (YBCO)/LCMO sandwiches are prepared by the facing-target sputtering technique. The superconducting transition temperature as a function of spacer layer thickness is described by the magnetic proximity effect in the superconductor/ferromagnet structure. The metal–semiconductor transition temperature exhibits a nonmonotonic behaviour which is strongly dependent upon the YBCO layer thickness, and is also shown from the measurement of resistance and magnetoresistance. These results imply that the magnetic spin interaction between the two LCMO layers may occur through the normal-conductive YBCO layer.

Keywords: colossal magnetoresistance, high temperature superconductivity, magnetic properties
PACC: 7130, 7470V, 7530E

1. Introduction

Superconductivity (SC) and ferromagnetism (FM) are antagonistic orderings, and the investigation of their possible coexistence and mutual influence in tailor-made SC/FM multilayer structures has been an active area of theoretical as well as experimental research. The colossal magnetoresistive (CMR) materials, i.e. $R_{1-x}A_xMnO_3$ (R = trivalent rare-earth ion, e.g. La, Nd, etc, and A=divalent element, e.g. Ba, Sr, Ca, Pb, etc), due to their half-metallic character, contain spin-polarized carriers in the ferromagnetic state.[1−3] On the other hand, in high T_C superconductors (HTS), e.g. $YBa_2Cu_3O_{7-\delta}$, carriers are paired in the form of Cooper pairs.[4] Therefore, HTS/CMR multilayer structures are an ideal model for studying the interplay between the superconductivity and magnetism.

A few works of SC/FM oxide layered films have been reported. Previously, Kasai *et al* reported on a novel proximity effect that supercurrents could pass through a La–Ca–Mn–O magnetic barrier as thick as 500 nm in $YBa_2Cu_3O_7$/La–Ca–Mn–O/$YBa_2Cu_3O_7$ trilayer junctions.[5] Furthermore, it was reported that the characteristics of $YBa_2Cu_3O_7$ / $La_{1-x}Sr_xMnO_z$ (LSMO) / $YBa_2Cu_3O_7$ trilayer junctions changed on varying the magnetic properties of the LSMO layer.[6] Ferromagnetic resonance measurements have indicated that the dynamic properties of the spin system in LSMO might be changed when the LSMO layer is sandwiched between $YBa_2Cu_3O_7$ layers.[7] In $La_{0.72}Ca_{0.25}MnO_z$ / $YBa_2Cu_3O_y$ / $La_{0.72}Ca_{0.25}MnO_z$ trilayered films, magnetoresistance (MR) is enhanced and is more than 1.5 times as large as that for $La_{0.72}Ca_{0.25}MnO_z$ films when the thickness of $YBa_2Cu_3O_y$ layer is less than 250 nm.[8] From these results, the novel proximity effects may be closely related to the spin states in La–Ca–Mn–O, La–Sr–Mn–O, and $YBa_2Cu_3O_7$.

It is well known that the CMR effect can coexist with high-T_C superconductivity in CMR/HTS layered films.[9−14] To the best of our knowledge, little attention has been paid to the variation of char-

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†E-mail: kzhao@aphy.iphy.ac.cn

acteristics of the ferromagnetic transition with the SC layer thickness. In this paper we investigate the magnetotransport properties of $La_{2/3}Ca_{1/3}MnO_3$ (LCMO) / $YBa_2Cu_4O_8$ (YBCO) / LCMO sandwiched structures. We choose YBCO as the superconducting spacer layer because it is much more stable than $YBa_2Cu_3O_7$ films in different oxygen environments. The strong dependence of the metal–semiconductor transition temperature (T_{MS}) and MR on the YBCO layer thickness is studied.

2. Experimental details

All samples were fabricated by using the facing-target sputtering (FTS) technique on $SrTiO_3$ (001) (STO) single-crystal substrates from pure LCMO and YBCO targets under different conditions.[15–17] The substrate temperature was kept at 740° C. A mixture of high-purity Ar and O_2, total pressure 10 Pa, was used as the discharge gases with the flow rates adjusted by two mass flow controllers. Prior to film deposition, the vacuum chamber was pumped down to 1×10^{-4} Pa, and then the targets were presputtered for 15 min. Immediately after each deposition, the vacuum chamber was back-filled with 100 kPa oxygen gas. The deposited film was then cooled to room temperature with the substrate heater power cut off. Based on our previous work, we prepared the YBCO films at r=5.5 and r=1 for the growth of LCMO layer, where r is the ratio of Ar and O_2 flow rates. 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 YBCO layer). In LCMO/YBCO/LCMO films, the thickness of the top and bottom LCMO layers, d_L, was fixed at 15, 50 and 100 nm. The thickness of YBCO layer, d_Y, was varied from 0 to 200 nm to examine the relationship between the superconductivity and ferromagnetism. The resistance as a function of temperature was measured by the standard four-probe technique. The film structure was examined by the $\theta/2\theta$ x-ray diffraction (XRD) method.[16] The magnetic moment of the samples as a function of temperature and applied magnetic field was measured by utilizing a vibrating sample magnetometer (VSM).

3. Results and discussion

The as-deposited films exhibit a mirror-like surface and have good adherence to all the substrates employed. From the z-profile, the roughness in the z-direction is less than 5 nm.[18] The surface morphology reveals the symmetry of the underlying lattice plane and shows the layered square islands with the same orientation and about the same size.[18]

The temperature dependence of the resistance of the LCMO/YBCO/LCMO sandwiches is shown in Fig.1. $R_\square$, the sheet resistance, is equal to ρ/d, with $d=2d_L+d_Y$. Since YBCO has a much higher conductivity than LCMO, the YBCO layer could be expected to short-circuit the LCMO layers. T_{MS} is a characteristic of the LCMO layers at high temperatures in Figs.1(a) and (b), which is defined as the peak point in $d\rho/dT$ curve shown in the inset in Fig.1 (a). The rapid drop in resistance at low temperatures is due to the superconducting transition of the YBCO layer.[18] The transition temperature $T_{C,ON}$ is defined as the onset of crossover resistance. When d_Y is depressed to 24 nm for LCMO(15 nm)/YBCO/LCMO(15 nm) trilayer films (Fig.1 (c)), clear metallic behaviour disappears although two crossover points, T_{MS} and $T_{C,ON}$, exist in the system. For the ultrathin YBCO layer, $d_Y < 10$ nm, no transition can be observed and the films are semiconductor-like in the whole temperature range studied.

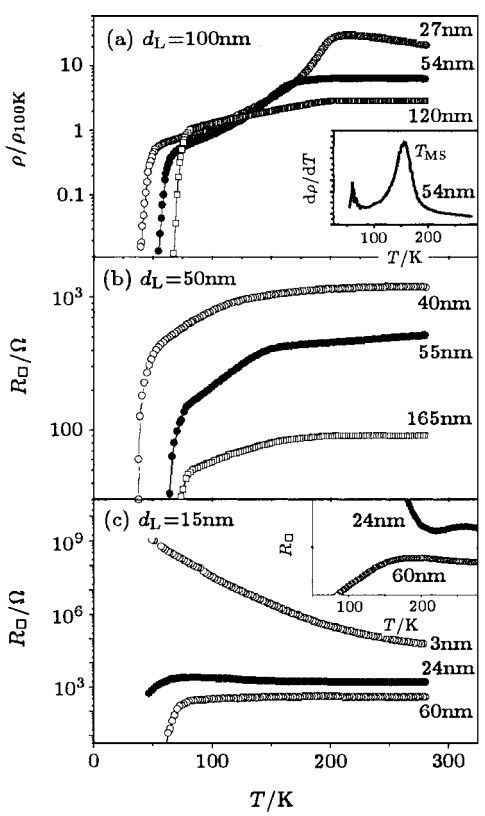


Fig.1. Temperature dependence of the resistivity of (a) LCMO (100 nm) / YBCO / LCMO (100 nm), (b) LCMO (50 nm) / YBCO / LCMO (50 nm), and (c) LCMO (15 nm) / YBCO / LCMO (15 nm) sandwiches. The inset in Figs.1(a) and 1(c) show the definition of T_{MS} and the T_{MS} for d_Y=24 and 60 nm. Numbers for the curves denote the d_Y values.

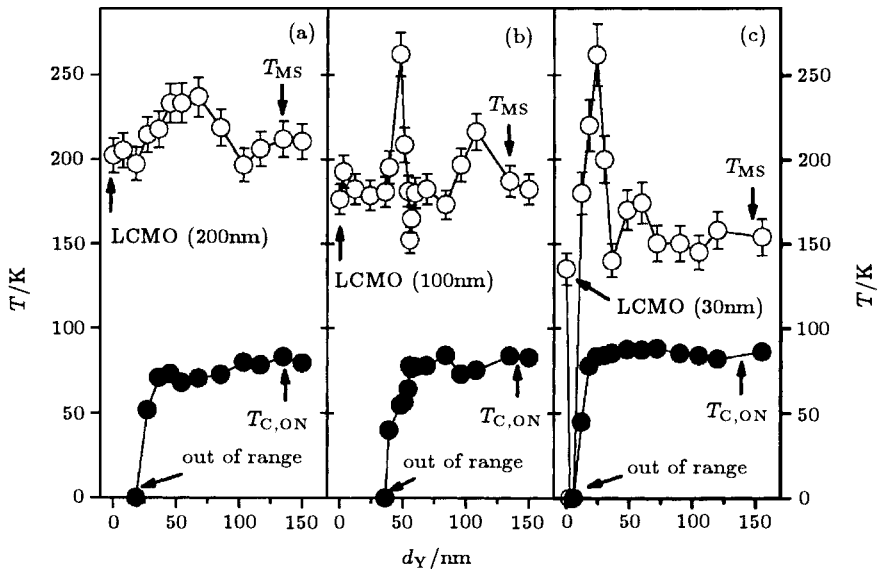


Fig.2. T_{MS} (○) and $T_{C,ON}$ (•) of (a) LCMO (100 nm)/YBCO/LCMO (100 nm), (b) LCMO (50 nm)/YBCO/LCMO (50 nm) and (c) LCMO (15 nm)/YBCO/LCMO (15 nm) as functions of d_Y. The lines are for a guide to the eyes only.

$T_{C,ON}$ and T_{MS} of the as-deposited samples are plotted against d_Y in Fig.2. For films with thick spacer, $T_{C,ON}$ is independent of its thickness. Degradation of $T_{C,ON}$ has been found for films with ultrathin YBCO layer. Extrapolation gives d_{cr}, below which the superconductivity is lost, and we have d_{cr}=16, 35 and 11.5 nm for the three series of d_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.[18] T_{MS} is close to the one of the individual LCMO layer for samples with thick YBCO layer, and shows a nonmonotonic behaviour with decreasing spacer layer thickness. When $d_Y > d_{cr}$, the peak values of T_{MS} are 262, 262 and 240 K for d_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,[19] where the T_{MS} is enhanced. The striking discovery in T_{MS} of our work shows that the magnetic interaction between the two LCMO layers is still survived when $d_Y > 20$ nm. Furthermore, the loss of T_{MS} for the series of d_L=15 nm with ultrathin YBCO layer in Fig.2 (c) may result from the interfacial roughness.

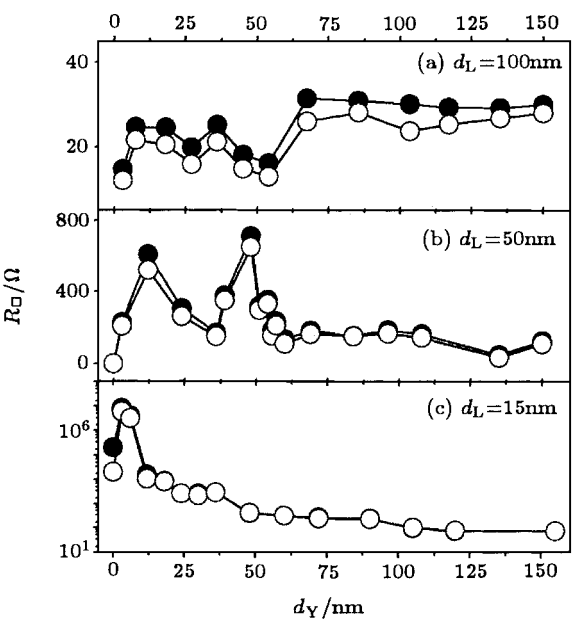


Fig.3. Sheet resistance of (a) LCMO (100 nm) / YBCO / LCMO (100 nm), (b) LCMO(50 nm) / YBCO / LCMO (50 nm) and (c) LCMO (15 nm) / YBCO / LCMO(15 nm) at 90 K as functions of YBCO layer thickness d_Y with the applied fields of H=0 (•) and 8×10^5 A/m (○).

Figure 3 shows the thickness dependence of $R_{\square}$ at 90 K, slightly above $T_{C,ON}$. In our set-up, the resistivity of the typical LCMO film deposited on STO substrate has the same order of magnitude as that of a thick YBCO film at the temperature slightly above $T_{C,ON}$, indicating that thick YBCO layer short-circuits the LCMO layers in the sandwich. This can be seen in the series of LCMO/YBCO/LCMO sandwiches with d_L=100 nm when d_Y >50 nm (Fig.3 (a)). Furthermore, a novel behaviour is observed in LCMO/YBCO/LCMO sandwiches with d_L=50 nm (Fig.3 (b)). $R_{\square}$ is shown in the same graph with the applied field along the film plane. The magnetization measurement suggested that 8×10^5A/m is enough to saturate the magnetic moment in LCMO layers, which indicates that the moments of top and bottom layers are parallel. In the GMR (giant magnetoresistance) case, the resistance depends only on the mutual alignments of the magnetic layers, which is smaller for parallel alignment than for anti-parallel alignment, and the interlayer coupling serves only as the convenient way to obtain anti-parallel alignment at zero fields in multilayer samples and has no direct influence on the resistance. The same oscillatory behaviour of the resistance at an applied field of up to 8×10^5 A/m as that at zero field indicates that the magnetic interaction exists in our samples. Therefore, a mechanism different from the standard model of GMR is responsible for the oscillatory behaviour in the LCMO/YBCO systems. As for the series of d_L=15 nm, the LCMO layers are so thin that the interaction between them is very weak, and no clear oscillation can be seen. The maximum resistance, $\sim 10^7\Omega$, of the sample with ultra-thin YBCO layer may also result from the interface roughness.

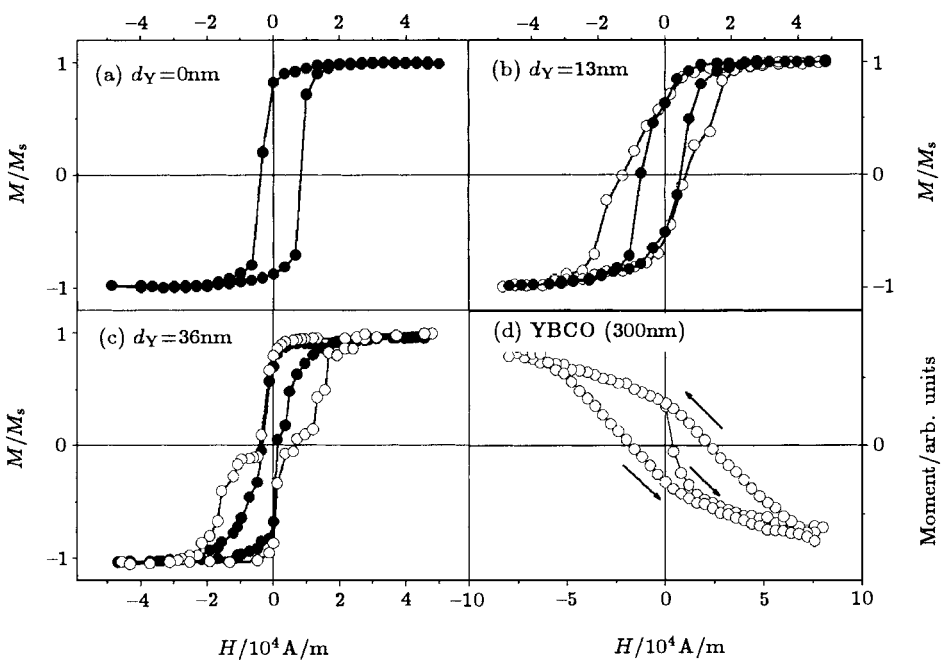


Fig.4. Hysteresis loops of (a) LCMO (100 nm), (b) LCMO (50 nm) / YBCO (13 nm) / LCMO (50 nm), (c) LCMO (50 nm) / YBCO (36 nm) / LCMO (50 nm) and (d) YBCO (300 nm) films at 15 K (○) and 90 K (●).

The magnetic properties of the samples were investigated using a VSM. A magnetic field (H) was applied parallel to the film surface during the measurements. A small nonhysteretic contribution from the STO substrate was eliminated by separately measuring its diamagnetic response. The hysteresis loops at 90 K are shown in Fig.4 corresponding to those of the samples represented by the valley and peaks of the $R_{\square}$-d_Y curve in Fig.3(b). The somewhat rounded rectangular shape and relatively high remnant magnetization ratio for the trilayer LCMO(50 nm)/YBCO (36 nm)/LCMO(50 nm) resembles that of a single LCMO film, suggesting that the ferromagnetic layers in this structure are either magnetically uncoupled or coupled ferromagnetically. Note that the sample has the first minimum resistance shown in Fig.3(b). On the other hand, the saturation field of LCMO(50 nm)/YBCO(13 nm)/LCMO(50 nm) in-

creases and the remnant magnetization decreases, indicating the appearance of some magnetic interaction between LCMO layers.

Figure 4 also shows the hysteresis loops at 15 K. When the YBCO layer is in its superconductive state, the loops of trilayers are broadened (Fig.4(b) and (c)). Especially, the LCMO(50 nm)/YBCO(36 nm)/LCMO(50 nm) sample shows two distinct steps in sharp contrast to that in its normal-conductive state. Przyslupski *et al* also found in $[La_{0.67}Sr_{0.33}MnO_3/YBa_2Cu_3O_7]_n$ multilayers that the magnetic hysteresis below the superconducting transition temperature was different from that above the superconducting transition and explained this in terms of the superposition of superconducting and ferromagnetic hysteresis curves.[14] In contrast, our data indicate that such different behaviour at different temperature is not only a simple superposition of Figs.4(a) and (d). In addition, alternating hard and soft magnetic phases can lead to either a broad or a narrow waist loop. Although further experiments need to be performed in order to clarify the phase-separation mechanism below $T_{C,ON}$, it is quite reasonable that the interplay of ferromagnetic and superconducting couplings in SC/FM systems would bring about the distinct step loops.

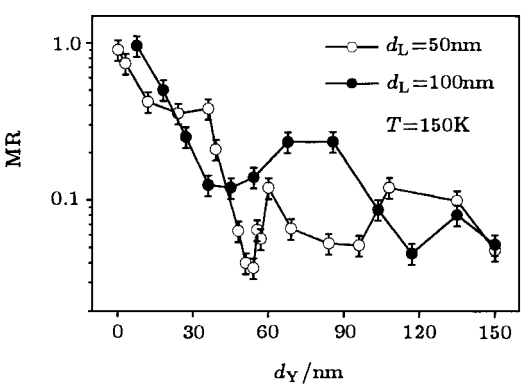


Fig.5. MR as a function of spacer layer thickness at 150 K in LCMO/YBCO/LCMO sandwiches with d_L=50 nm (○) and 100 nm (●). The lines are for a guide to the eyes.

MR was examined with an applied field up to H=8×10^{5}A/m at 150 K. At this temperature, the YBCO layer is in the normal-conductive state and a larger value of MR ratio can be measured easily because the maximum MR ratio occurs at temperatures slightly lower than T_{MS}. The field is applied in the film plane and perpendicular to the current direction (also in the film plane). The MR ratios are estimated using the expression MR = $[R(0)-R(H)]/R(0)$, where $R(H)$ and $R(0)$ are resistances with and without the applied field, respectively. $R(0)$ is taken as the standard value because the MR effects on LCMO are not saturated under an applied field up to 8×10^{5}A/m, while the magnetization of LCMO are saturated under an applied field about 3×10^{4}A/m in our experiment. Figure 5 shows the MR ratio for LCMO/YBCO/LCMO. The ratio is dependent on the spacer thickness and oscillates when the YBCO layer thickness decreases. A long period (l_p) is found, l_p ~30 and 80 nm for d_L=50 and 100 nm, suggesting that LCMO/YBCO/LCMO are not simple parallel electrical circuits, but indirectly coupled between the two LCMO layers across the YBCO layer. For d_L=15 nm, the MR ratio drop rapidly from 70% at d_Y=0 nm to 0.5% at d_Y=24 nm. When d_Y >25 nm, MR cannot be achieved because of the short-circuit effect of YBCO layer.

In the HTS/CMR structure, YBCO has a high critical temperature and short coherence length, while LCMO is metallic with not so large pair breaking effects due to its weak magnetism. The well-matched lattice between YBCO and LCMO weakens the effects of roughness and strain. In addition, the oxygen diffusion between LCMO and YBCO layers is negligible in our samples.[18] On the other hand, we also found the oscillatory T_{MS} in LCMO/YBCO multilayers (to be published elsewhere) resembling that in LCMO/YBCO/LCMO sandwiches. However, the origin of the long period is not clear. Bruno[20] has theoretically shown that the oscillation is a general behaviour. In the asymptotic limit $k_{Fs}d_s \gg 1$, the oscillation period $l_p = \pi/k_{Fs}$ is about 1 nm,[21] which is much smaller than our period. In addition, in our case the period is dependent on d_L, indicating that the pseudo-Fermi momentum of the superconductor (YBCO) k_{Fs} cannot entirely control the period of oscillation of the magnetic coupling across the superconductor.

A possible type of long-range coupling is dipolar coupling due to the roughness of the interface. From the previous results,[22–24] the dipolar magnetic interaction between finite magnetic films separated by a nonmagnetic slab can lead to ferro- or antiferromagnetic in-plane coupling. 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 cause a non-negligible coupling.[24]

Also the dipolar interaction, under particular circumstances, can lead to non-in-plane magnetizations. The roughness of the samples is less than 5 nm.[18] We did not have the expression of dipolar interaction energy between the two LCMO layers 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 $d_Y < d_{cr}$, T_{MS} is not enhanced but is the same as that for LCMO films, indicating that the magnetic shunt due to pinholes is ruled out.

Another possible mechanism may be related to the spin fluctuations in YBCO and LCMO layers. A sandwich can be considered as a layered structure with Cu–O and Mn–O conduction planes stacked apart from each other along the c-axis direction. Spin fluctuations in the LCMO layer may propagate in the c-axis direction from layer to layer and the adjacent LCMO layers may be coupled magnetically through the YBCO layer. To date, we do not have a complete microscopic understanding of these features. Further studies are in process.

4. Conclusion

In LCMO/YBCO/LCMO sandwiches, the superconducting transition temperature as a function of spacer layer thickness is described by the magnetic proximity effect in a superconductor/ferromagnet structure. The nonmonotonic behaviour of T_{MS}, $R_{\square}$ and MR are related to the spacer layer thickness. A clear oscillation of large period has been observed. These results imply that the magnetic spin interaction between the two LCMO layers may exist through the normal-conductive YBCO layer.

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