Transport and Magnetic Properties of HTS/CMR Multilayers

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Abstract

We report magneto-transport and magnetization studies of heterostructures comprising of alternately stacked $YBa_2Cu_3O_{7-\delta}$ (YBCO) and $La_{1-x}Pb_xMnO_3$ (LPMO) layers. The c*-axis oriented bi-layer and four-layer YBCO/LPMO heterostructures were prepared* in-situ *on (100) $SrTiO_3$ substrates using pulsed laser deposition. Zero-field cooled (ZFC) and field-cooled (FC) magnetization data show that in a four-layer structure the LPMO layers get antiferromagnetically coupled when the thermodynamic-fluctuation induced Cooper pairing begins in the YBCO layers, which is well supported by the magnetization versus applied field data. Evidence of antiferromagnetic coupling is also reflected in the temperature dependence of the magnetoresistance.*

Introduction

SUPERCONDUCTIVITY (SC) AND ferromagnetism (FM) are antagonistic orderings and 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 for last four decades [1]. The antagonism between SC and FM is understandable from the microscopic theory: SC requires an attractive interaction between electron pairs with antiparallel spins (i.e. formation of Cooper pairs), whereas for same electrons participation in magnetic ordering demands a parallel alignment of electron spins through an exchange interaction. Therefore, in a SC/FM heterostructure if exchange field of FM exceeds the condensation energy of Cooper pairs (measured in terms of energy gap, Δ) then destruction of superconductivity can take place due to paramagnetic effect. However, in SC/FM heterostructures both superconductivity and ferromagnetism can coexist. This is because proximity effect can induce a superconducting order parameter in FM layer; on the other hand, the neighboring pair of FM layers can interact with each other via the SC layer.

An important question is how superconductivity of a layered SC/FM structure is influenced by the presence of magnetic layers. The theoretical work of Radovic et al [2] predicted a phase difference $0<\phi<\pi$ between neighboring SC layers and an unusual oscillatory dependence of superconducting transition temperature (T_c) on the FM layer thickness (d_{FM}). However, the experimental reports on the evidence of π-phase superconductivity and oscillations in T_c as a function of d_{FM} have been mixed [3-5]. For example, in V/Fe system some experiments showed oscillations in T_c-d_{FM} [3], while on same system other experiments showed a rapid decrease in T_c with increasing d_{FM} followed by a plateau [4]. Negative results were also published for Nb/Fe system [5].

Another important issue of SC/FM multilayers is how the magnetism of FM layers is influenced by the presence of SC layers. Theoretically it was shown that the singlet Cooper pairing of electrons in SC layers leads to a long-range antiferromagnetic interaction between FM layers due to Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange [1,6]. At present, however there is no experimentally known SC/FM heterostructure that shows magnetic coupling either above or below T_c. A recent experimental attempt in GdN/NbN/GdN trilayers failed to observe the magnetic coupling between the FM layers [7]. Sa de Melo theoretically showed that magnetic coupling in SC/FM heterostructures can be observed if the superconductor has higher T_c and ferromagnet has not so large pair breaking effect [8]. Thus, heterostructures comprising of high temperature oxide supercondcuctors and colossal magnetoresistance ferromagnets are considered to be good systems for such studies.

In this paper, we present some interesting experimental results of magneto-transport and magnetic studies carried out on $YBa_2Cu_3O_{7-\delta}/La_{1-x}Pb_xMnO_3$ (YBCO/LPMO) heterostructures fabricated using pulsed laser deposition technique. The results, for the first time, show that LPMO layers get antiferromagnetically coupled when the thermodynamic-fluctuation induced Cooper pairing begins in the YBCO layers.

Experimental

The bi-layer and four-layer YBCO/LPMO structures were fabricated on (100) $SrTiO_3$ (STO) single-crystal substrates by pulsed-laser ablation technique. The details of fabrication are described elsewhere [9]. Briefly, to fabricate YBCO/LPMO heterostructures a laser beam from a KrF excimer laser (wavelength 248 nm, pulse width 20 ns and repetition rate of 5 Hz) was alternately focused onto YBCO and LPMO targets. All the layers were deposited *in-situ* at substrate temperature of 730°C and oxygen pressure of 0.2 torr. In the present studies the thickness of YBCO layer was kept 100 nm, whereas the thickness of LPMO layer was varied between 25 and 100 nm.

X-ray diffraction (XRD) measurements were performed using CuK radiation to assess the orientation of the grown heterostructures. The surface morphology of samples was examined using atomic force microscopy (AFM). AFM measurements were carried out under ambient conditions using a scanning probe microscope (model-SPM Solver P47) in contact mode. Rectangular cantilevers of Si_3N_4 (length 200 μm and width 40 μm) having force constant of 3 N/m were employed for these measurements.

The electric resistance of the heterostructures was measured using a standard four-probe method in the temperature range between 50 and 300 K using a closed cycle helium cryostat (APD-make). The magnetoresistace of the heterostructures was determined by applying a field of 1T parallel to the plane of the substrates. The magnetic properties of the heterostructures were studied using a SQUID magnetometer (Quantum design MPMS model).

Results and Discussion

A typical XRD plot for [YBCO (100nm)/LPMO (50nm)]$_2$ heterostructure is shown in Fig. 1. The presence of only (00*l*) reflections suggests that both YBCO and LPMO layers have grown with *c*-axis perpendicular to substrate plane. In the expanded plot, as shown in the inset of Fig. 1, (002) LPMO, (200) STO and (006) YBCO peaks are clearly discernible. It is noted that the *c*-lattice parameter for LPMO in the heterostructure is 0.3909nm, which is same as that measured for a LPMO film directly grown on STO substrate. This suggests that crystallinity of LPMO layers is unaffected by the presence of YBCO layers. The *c*-lattice parameter of YBCO in

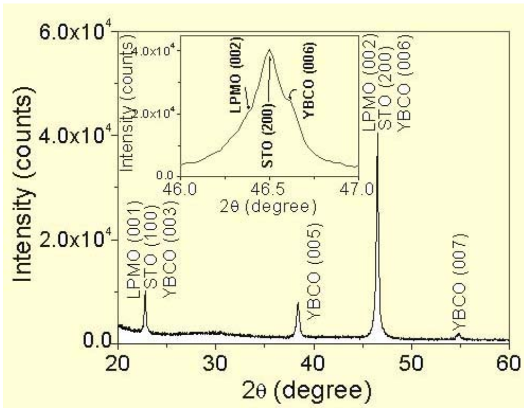

Fig. 1 XRD plot of a [YBCO(100nm)/LPMO(50nm)]$_2$ heterostructure. The inset shows the expanded plot in the 2θ range between 46 and 47°.

the heterostructure is determined to be 1.1673 nm. The oxygen content, (7-δ), of YBCO is computed using the empirical relation: (7-δ) = 74.49 – 57.87 *c*, where *c* is the *c*-lattice parameter in nm [10].The value of oxygen content, (7-δ), comes out to be 6.94, which suggests that YBCO layers are fully oxygenated.

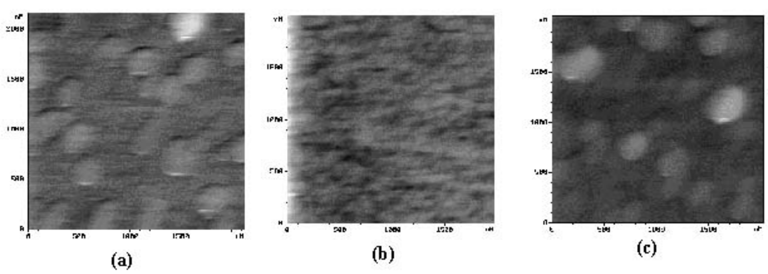

Fig. 2 2000 nm X 2000 nm AFM scans of (a) YBCO film on STO substrate, (b) LPMO film on STO substrate, and (c) YBCO(100nm)/ LPMO(50nm)]$_2$ heterostucture on STO substrate.

In Fig. 2 we show the AFM images of the top LPMO layer in [YBCO (100nm)/LPMO (50nm)]$_2$ heterostructure. For comparison, the surface morphologies of *c*-axis oriented YBCO and LPMO films grown on STO are also shown in Fig 2. YBCO film grows by 3D island growth, while LPMO film grows via 2D layer-by-layer growth. The surface morphology of top LPMO layer in a heterostructure exhibits a carpeting effect i.e. the morphology of LPMO is dictated by the morphology of the YBCO layer. These results along with XRD data suggest high quality of the grown heterostructures.

The temperature dependences of normalized resistance and magnetoresistance for a typical [YBCO(100nm)/LPMO(25nm)]$_2$ heterostructure are shown in Fig. 3. The magnetoresistance (MR) is expressed as $(R_0-R_H)/R_H$ X100, where R_0 and R_H are the resistance values under zero and 1 T magnetic fields. For comparison, the data of YBCO and LPMO films are also presented. From Fig. 3 we infer the following:

(i) For YBCO film, the resistance varies linearly between 140 and 300 K. Below 140 K (marked by T_{CP} in the Fig. 3 (a)), the resistance deviates from the linearity, which is well described in terms of thermodynamic-fluctuations induced formation of Cooper pairs [11]. Application of magnetic field did not yield any changes in the normal state resistivity, while T_c is suppressed marginally by ~1K.

(ii) The R(T) plot of LPMO film exhibits an insulator-to-metal transition (T_{IM}) at 235 K, which is a characteristic property of colossal magnetoresitive materials [12]. The T_{IM} corresponds to paramagnetic-ferromagnetic transition that occurs approximately simultaneously with insulator-to-metal transition. The temperature dependence of MR exhibits aximal (peak) near T_{IM}. A common explanation of MR is usually provided in the framework of double-exchange mechanism, which is based on the assumption of the appearance of Mn^{+4} with the substitution of La^{+3} by a divalent cation (Pb^{+2}). It is believed that in this case ferromagnetism results from the strong ferromagnetic exchange between Mn^{+3} and Mn^{+4}.

(iii) The normal state behavior of [YBCO(100nm)/LPMO (25nm)]$_2$ structure is clearly influenced by both insulator-to-metal transition of LPMO layers at ~235 K and fluctuation conductivity of YBCO layers at

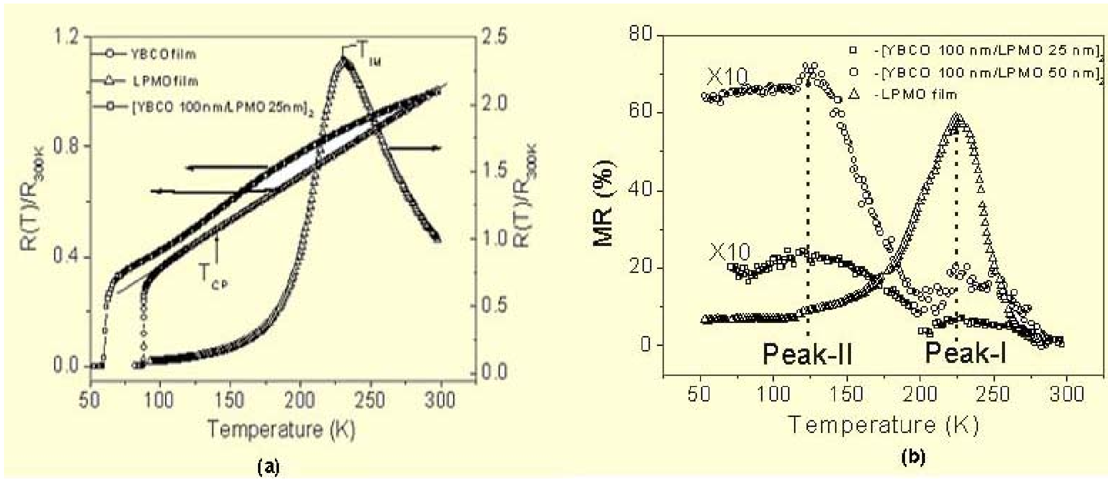

Fig. 3 (a) Normalized resistance as a function of temperature recorded for YBCO film. LPMO film and $[YBCO(100nm)/LPMO(50nm)]_2$ heterostructure. The straight line is linear fit to the normal state resistance data of YBCO film and T_{CP} corresponds to onset of fluctuation conductivity in YBCO film. T_{IM} represents the insulator-to-metal transition in LPMO film. (b) Temperature dependence of magnetoresistance (obtained at 1T magnetic field) for LPMO film, $[YBCO(100nm)/LPMO(25nm)]_2$ and $[YBCO(100nm)/LPMO(50nm)]_2$ heterostructures.

~140K. The heterostructure exhibits superconductivity at 59 K. The T_c of heterostructures was found to decrease monotonically with increasing the thickness of LPMO layers without any significant broadening of the transition width. This indicates that the suppression of T_c is not due to the chemical reaction between LPMO and YBCO. The suppressed superconductivity in the hetero-structure has been attributed to the self-injection of quasi-particles from YBCO to LPMO [13]. Due to d-wave symmetry of the order parameter in YBCO, there is a significant population of quasiparticle excitations. The ferromagnetic LPMO, having nearly perfect spin polarization, allows only those quasiparticles to diffuse across the YBCO/LPMO interface whose spins are parallel to those of LPMO. This induces further pair breaking in the YBCO and results in depressed T_c. Interestingly the temperature dependence of the MR exhibits two distinct peaks at temperatures 235 K (peak-I) and 130 K (peak-II), respectively. The magnitude of Peak-II being much larger as compared to that of Peak-I. The origin of Peak-I is obviously related to the CMR property of LPMO layers; while Peak-II appears to be correlated with T_{CP} and to investigate its origin we concentrate on the magnetic properties of the heterostructures.

In Fig. 4, we show the temperature dependence of the magnetization recorded in zero-field cooled (ZFC) and field cooled (FC) conditions. For a bi-layer structure two different transition regions can be identified: (i) a paramagnetic to ferromagnetic (FM) at ~240K and (ii) a ferromagnetic to diamagnetic transition at ~72 K. The ferromagnetic ordering at ~240 K corresponds to that observed for LPMO. It may be noted that in ferromagnetic state, the ZFC signal is lower than FC signal, and this indeed is the case with ferromagnetic materials. The diamagnetic transition at ~72 K indicates that the bi-layer structure has undergone a transition to superconducting state. In comparison, the four-layer structure shows two anomalous features: (i) antiferromagnetic (AF) ordering at 135 K (as revealed by FC data) and (ii) a larger ZFC signal as compared to FC signal in the temperature range 75-170 K. Since at 135 K the individual LPMO layers are expected to show FM ordering, therefore an AF ordering at this temperature in a four-layer structure can arise only if the spins of two LPMO layers, namely, F1

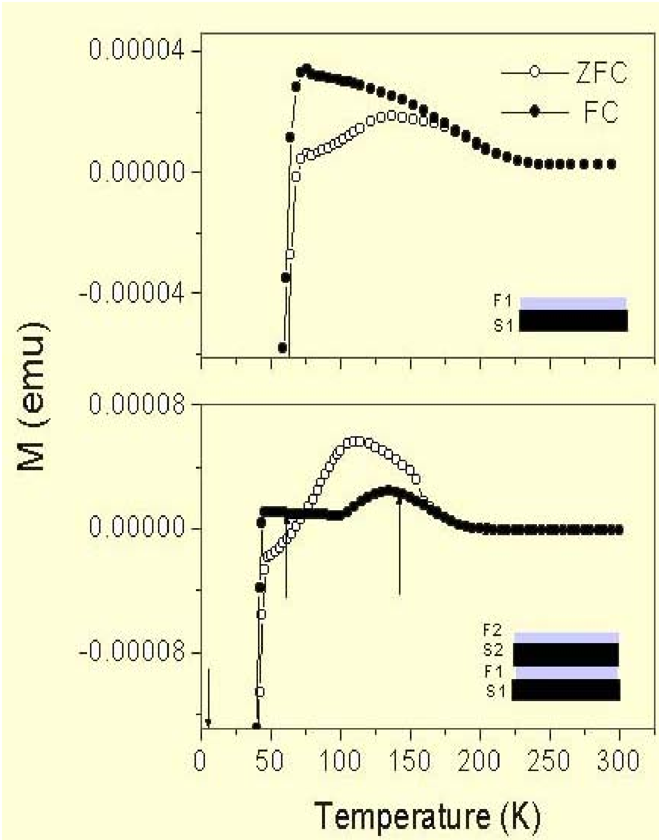

Fig. 4. Zero field cooled (ZFC) and field cooled (FC) magnetization as a function of temperature recorded for bi-layer YBCO(100nm)/LPMO(50nm) and four-layer [YBCO(100nm)/LPMO(50nm)]$_2$ heterostructures. The inset shows the schematic of the heterostructures: S1 and S2 are YBCO layers, and F1 and F2 are LPMO layers. Arrows marked at 140, 60 and 5K correspond to ferromagnetic, antiferromagnetic and diamagnetic states, respectively.

and F2, align in the opposite directions. It is interesting to note that AF ordering correlates with the formation of thermodynamic-fluctuations induced Cooper pairs in the YBCO layer. This suggests that AF coupling between CMR layers is prompted by the formation of Cooper pairs in the HTS layer.

A larger ZFC signal as compared to FC signal is a consequence of AF coupling between the two CMR layers i.e. F1 and F2, and is understood as follows. For the measurement of ZFC magnetization, the sample is first cooled to 5 K and then a field of 100 Gauss is applied parallel to the plane of the structure. The data is acquired during the warming cycle of the sample. At 5K, both YBCO layers i.e. S1 and S2 are in superconducting state and these layers would expel any applied field that is lower than lower critical field. Thus, magnetic layers F1 and F2, which are AF coupled, will have an additional magnetic flux that is expelled from S1 and S2 layers. The additional flux in F1 layer will have contributions from both S1 and S2 layers, while for F2 the additional flux would be only from S2. This would lead to a net positive magnetic moment as the temperature is increased above T_c. In the case of FC measurement, first a 100 G field is applied at room temperature and then magnetization is recorded during the cooling cycle of the sample. As field gets trapped at defects in S1 and S2 layers, the expelled field is almost negligible, which results in a temperature independent magnetization due to AF interaction between F1 and F2. However, at lower temperatures superconductivity dominates and the heterostructure shows a sharp diamagnetic transition.

In order to confirm the change in magnetic ordering i.e. ferromagnetic to antiferromagnetic to diamagnetic as a function of temperature in a four-layer structure, magnetization versus applied field (M-H) loops were recorded at three different temperatures belonging to different regimes of Fig. 4(b), which are marked by arrows. The obtained M-H loops are shown in Fig. 5. At 140 K the M-H loop is typical of a ferromagnetic state. However, at 60 K at low fields the magnetization varies less sharply with the field. In addition, the saturation magnetization decreases significantly, which confirms the occurring of antiferromagnetic coupling in the multilayer structure. At 5K, the M-H loop corresponds to a superconducting state.

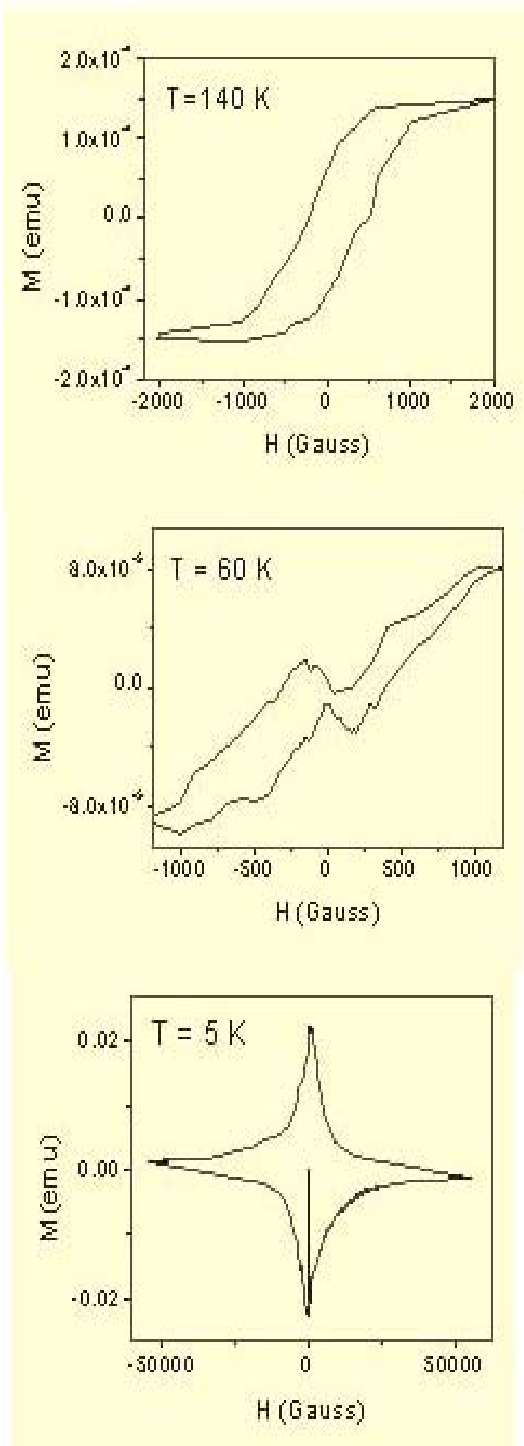

Fig. 5 Magnetization as a function of field applied parallel to the substrate-plane for [YBCO(100nm)/LPMO(50nm)]$_2$ heterostructure recorded at three different temperatures, namely 140 K, 60 K and 5K.

Interestingly the position of peak-II in temperature dependence of MR, Fig. 3(b), coincides with the AF coupling of CMR layers. This implies that the origin of peak-II is similar to that observed for magnetic/nonmagnetic GMR multilayers [14]. In the temperature region close to peak-II, F1 and F2 are AF coupled and therefore the resistance of the heterostructure is high due to spin-dependent scattering. On application of high magnetic field, F1 and F2 get ferromagnetically aligned resulting in the decrease of the resistance and hence a magnetorersistance.

The antiferromagnetic state in YBCO/LPMO heterostructures as revealed from magnetization and magnetoresistance data is therefore consistent with theoretical prediction whereby antiferromagnetic state arises due to long range RKKY exchange between neighboring FM layers through SC interlayer [1,6]. Recently, the coexistence of superconductivity and magnetisms has also been reported in a naturally layered $RuSr_2GdCu_2O_8$ compound [15,16] in which CuO layers are responsible for superconductivity while Gd layers are the magnetic layers. The Curie temperature and superconducting transition temperature for this compound are respectively 132 K and 46 K, respectively. This implies that superconducting state arises in a system that already has a magnetic ordering- a case similar to our YBCO/LPMO heterostructures. In $RuSr_2GdCu_2O_8$ compound, it has been experimentally shown that CuO layers show superconductivity, while in Gd layers a ferromagnetic ordering occurs in such a way that the magnetization of neighbouring layers are antiparallel i.e. the system exhibits a canted antiferromagnetim [16].

Conclusions

We have studied the magneto-transport and magnetic properties of YBCO/LPMO heterostructures. We have shown that these heterostrucutures exhibit both superconductivity and magnetism. Zero-field cooled (ZFC) and field-cooled (FC) magnetization data of a four-layered YBCO/LPMO structure is complex and exhibit different magnetic states as the temperature is lowered i.e. paramagnetic, ferromagnetic, antiferromagnetic and diamagnetic states. The antiferromagnetic state is found to be correlated with thermodynamic-fluctuations induced Cooper pairing in YBCO. This suggests that Cooper pairing in YBCO leads to antiferromagnetic interaction between LPMO layers through RKKY indirect exchange. The temperature dependence of the magnetoresistance exhibits two peaks at temperatures 235 K and 130 K respectively. The magnetoresistance peak at 235 K corresponds to colossal magnetoresisitve property of LPMO layer; while peak at 130 K is a consequence of antiferromagnetic coupling of LPMO layers.

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The simultaneous analysis of the magnetic properties and the transport behavior suggests a bimodal grain size distribution. A detailed quantitative description of the unusual features observed in the transport properties is given. PACS numbers: 75.47.De; 75.70.Cn; 75.20.En; 73.43.Qt. I. INTRODUCTION.Â In this paper we present a systematical study of the magnetic and magnetotransport properties of vac-uum evaporated granular Fe-Ag structures. The ob-served large, negative non-saturating magnetic eld de-pendence and the unusual sublinear temperature depen-dence (d2R/dT 2 < 0) of the resistivity have been an-alyzed simultaneously.