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# Spin dependent transport at oxide $La_{0.7}Ca_{0.3}MnO_3/YBa_2Cu_3O_7$ ferromagnet/superconductor interfaces

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## Abstract

We have found large magnetoresistance in ferromagnet/superconductor/ferromagnet heterostructures made of $La_{0.7}Ca_{0.3}MnO_3$ and $YBa_2Cu_3O_7$. It originates at an increase of the width of the resistive transition when the magnetizations of the ferromagnetic layers are aligned antiparallel. We find that the shape and height of the magnetoresistance peaks are not modified when the angle between current and magnetic field is changed from parallel or perpendicular. Furthermore, we find that the temperature shift of the resistance curves is independent of the current values. This favors the view that the magnetoresistance phenomenon originates at the spin dependent transport of quasiparticles transmitted from the ferromagnetic electrodes into the superconductor, and rules out interpretations in terms of spontaneous vortices or anisotropic magnetoresistance of the ferromagnetic layers.

*Keywords:* Superconductivity; Magnetic properties; Films; Interfaces

## 1. Introduction

Superconductivity and ferromagnetism are rarely found on the same material due to their mutually incompatible nature. Thin film heterostructures combining ferromagnets (F) and superconductors (S) are very amenable to study the interplay between both long range orders.[1,2] There has been increasing interest in recent years in structures combining cuprates and manganites,[3–7] which incorporate a variety of interesting new ingredients. First, it has been demonstrated that many of these perovskite oxides can be readily combined, thanks to their good lattice matching and chemical compatibility, yielding good quality heterostructures with smooth and well-defined interfaces.[8,9] The larger critical temperature, $T_C$, of the high temperature superconductors (HTS) (as compared to low $T_C$ superconductors) sets an energy scale for the condensation energy which is comparable to the exchange coupling of the ferromagnet (superconducting and magnetic critical temperatures are not that dissimilar), which favors the competition between both long range phenomena in F/S heterostructures. The short coherence length of the HTS in the *c*-direction (0.1–0.3 nm) enables superconductivity to survive even in very thin layers in the presence of a ferromagnet (FM). Furthermore, the unconventional d-wave symmetry of the superconducting order parameter may give rise to novel quantum phenomena, related to the occurrence of Andreev bound states at the interface with the ferromagnet. In addition, the high degree of spin polarization of the conduction band of the manganites enhances the F/S competition and may open the door to important spin dependent transport effects yielding (useful) magnetoresistance.

In a previous paper we have reported very large magnetoresistance, MR, (in excess of 1000%) in F/S/F structures made of $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) and $YBa_2Cu_3O_7$ (YBCO).[10] This MR originates at a larger resistance in the antiferromagnetic (AF) configuration of the F layers, as opposed to conventional F/S/F proximity coupled structures where the larger resistance occurs in the F alignment.[11–13] In conventional F/S/F junctions there is a modulation of the critical temperature by the relative orientation of the magnetization in the F layers, which results of a compensation of the exchange field over the coherent volume in the AF configuration if the thickness of the superconductor

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is comparable to the coherence length.[11–13] Since at the interface between a half metal and a superconductor proximity effect is suppressed, the mechanism ruling the $T_C$ in F/S/F structures with highly spin polarized carriers is different.

In this paper we show that magnetoresistance measurements are insensitive to the (in plane) orientation of the applied magnetic field and current, and to the magnitude of the electric current. This constitutes a strong indication that spin dependent transport of (spin polarized) quasiparticles diffusing from the ferromagnet may play a major role in the MR phenomenon and rules out explanations in terms of vortex dissipation or anisotropic magnetoresistance. In the AF configuration of the magnetizations of the manganite layers quasiparticles transmitted from one of the electrodes may not be able to enter into the other. This scattering at both interfaces has a pair breaking effect and depresses the critical temperature stronger than in the ferromagnetic configuration where this scattering process is absent.

## 2. Experimental procedure

Samples were grown on (1 0 0) oriented $SrTiO_3$ single crystals in a high pressure (3.4 mbar) dc sputtering apparatus at high growth temperature (900 °C). The high oxygen pressure and the high deposition temperature provide a very slow (1 nm/min) and highly thermalized growth which allows the control of the deposition rate down to the unit cell limit. For this study we grew F/S/F trilayers keeping the thickness of the LCMO fixed at 40 unit cells (15 nm) and the thickness of the YBCO at 13 (15 nm) and 15 unit cells (18 nm). Structure was analyzed using X-ray diffraction and transmission electron microscopy. Further details about growth and structure can be found elsewhere.[14,15] X-ray refinement technique using the SUPREX 9.0 software were used to obtain quantitative information about the interface roughness.[16] $T_C$ was obtained as the zero resistance temperature from 4 point contacts resistivity measurements performed in a PPMS Quantum Design apparatus. Zero field resistance curves showed $T_C$ vales ranging between 48 and 52 K in this YBCO thickness range.

## 3. Results and discussion

We have measured magnetoresistance at selected temperatures along the resistive transition with the magnetic field applied parallel to the layers. Fig. 1 shows $R(H)$ loops at various temperatures for a trilayer sample with 13 unit cells thick YBCO layer. Current contacts were in the plane of the layers (current in plane geometry) and aligned perpendicular to the magnetic field direction. Magnetic field was swept between 0.5 and −0.5 T in an hysteresis loop sequence.

Large MR peaks are observed whose relative heights decrease when temperature is increased (see Fig. 1(a)). Fig. 1(a) also shows that MR peaks are superimposed on a background in which resistance increases with field. Most likely this background resistance is due to vortex dissipation, since it is known that the activation energy for vortex motion decreases as field is increased. Fig. 1(a) also shows that MR peaks decrease with increasing temperature along the resistive transition and vanish at the onset of the superconducting transition. This evidences that superconductivity plays a key role in the occurrence of this MR phenomenon in contrast to the conventional giant magnetoresistance GMR effect observed in magnetic superlattices. Interestingly MR peaks occur in a magnetic field interval where polarized neutron reflectometry and SQUID magnetometry show AF alignment between the LCMO layers[10] (not shown). When temperature is increased above the superconducting onset the MR becomes very small and negative as expected from the anisotropic magnetoresistance (AMR) of the manganite (see Fig. 1(b) and the discussion below).

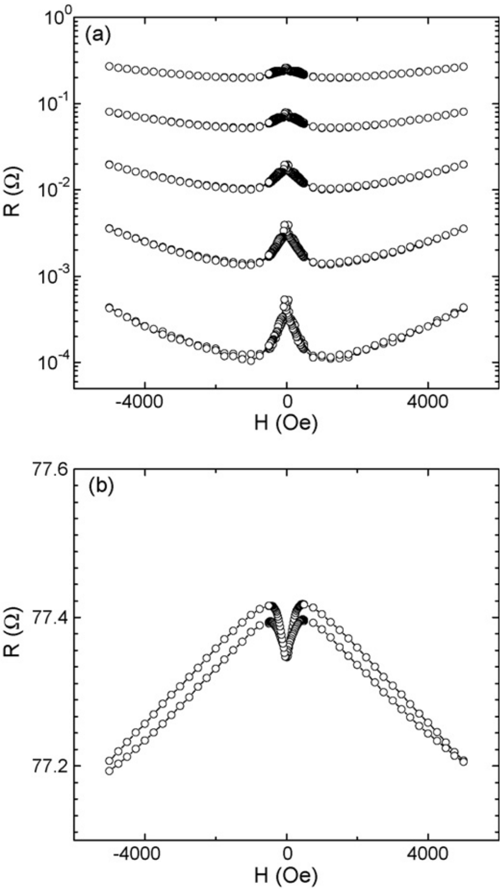

Fig. 1. (a) Resistance as a function of magnetic field, $R(H)$ loops, of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (13 u.c.)/LCMO (40 u.c.)] at different temperatures along the resistive transition. Magnetic field, applied parallel to the layers and perpendicular to electric current (0.1 mA), was swept between −0.5 and 0.5 T fields in an hysteresis loop sequence. Temperatures are 49, 49.5, 50, 50.5, and 51 K from bottom to top. (b) $R(H)$ loop of the same sample at 61 K (above the superconducting onset).

Three different mechanisms can be thought of to explain this MR phenomenon along the superconducting transition: vortex dissipation (including vortices due to stray fields of domains or domain walls), AMR, which in manganites is known to be large due to the strong spin orbit scattering and GMR like resistance originating at spin dependent transport. Each of these mechanisms have a very different current-field dependence. Vortex dissipation is zero when current is parallel to field, AMR is maximized when current is parallel to field and GMR is independent

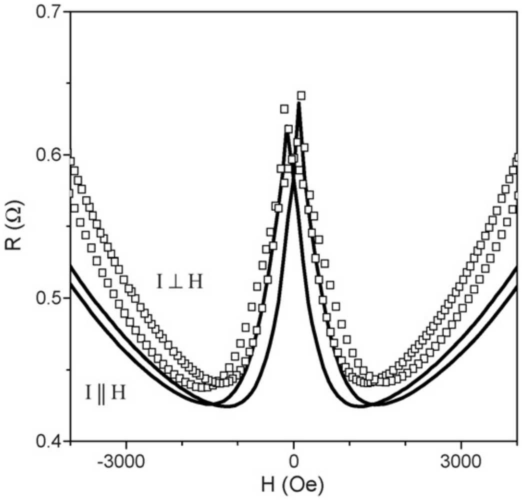

Fig. 2. Resistance as a function of magnetic field, $R(H)$ loop, of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (15 u.c.)/LCMO (40 u.c.)] measured at 55.5 K with current (0.1 mA) directed parallel (line) and perpendicular (open symbols) to magnetic field.

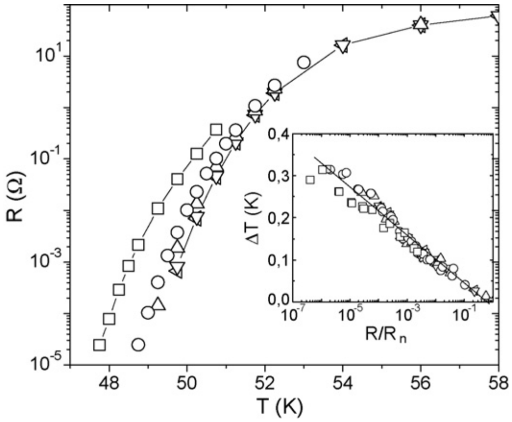

Fig. 3. Main panel: (zero magnetic field) resistance vs. temperature curves for different current values of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (13 u.c.)/LCMO (40 u.c.)]. Open squares (5 mA), open circles (1 mA), up triangles (0.5 mA), down triangles (0.1 mA), triangles facing left (0.05 mA). *Inset*: Temperature shift ($\Delta T$) as a function of resistance normalized to the onset resistance. Same symbol code as in main panel.

of current values and of direction between current and field. Experiments changing current values and the direction between current and field are thus useful to explore the origin of the magnetoresistance. The size of the MR peaks was independent on whether the in plane current was parallel or perpendicular to the magnetic field. Fig. 2 shows MR peaks of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (15 u.c.)/LCMO (40 u.c.)] measured at 55.5 K with current in the plane of the layers, and directed parallel (line) and perpendicular (open symbols) to magnetic field. It can be observed that the high field dissipation increases substantially when current is perpendicular to field. For current parallel to field the Lorentz force density on vortex lines ($\mathbf{J} \times \phi_0$, where $\mathbf{J}$ is the current density and $\phi_0$ is the flux quantum) vanishes and so does therefore the vortex dissipation due to vortices parallel to the external magnetic field. We cannot exclude additional vortices perpendicular to the layers due to a small misalignment of the magnetic field or spontaneous vortices due to the stray field of domains, responsible for the high field dissipation in this current-field configuration. But remarkably the size and shape of the peaks do not depend on the angle between magnetic field and current, ruling out explanations in terms of vortices parallel to the layers. This also discards the contribution of the anisotropic magnetoresistance of the single ferromagnetic layers. Actually, AMR shows up when the temperature is raised above the superconducting onset and it is in fact negative (larger dissipation when current is perpendicular to field) as previously found in manganite thin films[17] (see Fig. 1(b)).

We have also done measurements as a function of the current values for currents directed perpendicular to magnetic field (comprised between −0.5 and 0.5 T). Increasing current increases transition width as shown in Fig. 3. This occurs due to increased vortex dissipation which adds a magnetic field dependent background to the magnetoresistance plots of Fig. 1. Magnetoresistance $\Delta R/R_\mathrm{b}$ is calculated from resistance maxima and minima of $R(H)$ loops as a resistance change ($\Delta R$) relative to the minimum background level ($R_\mathrm{b}$). Increasing current results trivially in smaller MR values as a result of dividing by the larger background resistance $R_\mathrm{b}$. However, if instead of looking at magnetoresistance (resistance shift) one looks at the temperature shift of the resistance curve when magnetic alignment changes from parallel to antiparallel, a completely different picture is obtained. The inset of Fig. 3 shows the temperature shift ($\Delta T$) for different current levels as a function of resistance normalized to the onset value. The first observation is that there is a logarithmic dependence of the temperature shift as a function of the normalized resistance. In second term it is clear that the temperature shift is independent of current; i.e., smaller magnetoresistance results solely of the increased background resistance. This result provides further evidence against vortex dissipation originating the MR peaks and points to spin dependent effects in transport. In fact, this MR phenomenon has many of the ingredients of the GMR in metallic superlattices insofar it is independent on current and of its direction relative to field, and depends on the orientation of the magnetization of the LCMO layers. According to the spin imbalance theory of Takahashi, Imamura, and Maekawa,[18] which analyses spin transport in a F/S/F double junction depending on the orientation of the magnetization in the F layers, in the AF configuration (and for a half metal) transport is not possible since there are no vacant states at the Fermi level with the right spin orientation. This yields increased scattering at the interface in the AF configuration which does not occur if the LCMO layers have parallel magnetizations. Although in our case transport takes place parallel to the layers, normal electrons may diffuse from one ferromagnet to the other if the superconductor is thin enough. Strong scattering still occurs in the AF configuration which results in an effective increase of the number of quasiparticles, which self consistently reduces the critical temperature, thus providing a basis for the large magnetoresistance. Recent reports have shown similar magnetoresistance on permalloy/Nb/permalloy trilayer structures,[19] suggesting that a high degree of spin polarization plays an important role in the occurrence of the phenomenon.

## 4. Conclusions

We have shown that the large magnetoresistance shown in F/S/F trilayers made of highly spin polarized LCMO and high $T_C$ superconducting YBCO originates at an increase of the transition width when the magnetic alignment changes from parallel to antiparallel. At a given temperature the increase in transition width yields the magnetoresistance measured in $R(H)$ loops. It turns out that the increased transition width is logarithmic on resistance and independent on current. Furthermore magnetoresistance is independent on the direction between magnetic field and electric current. This results rule out vortex dissipation or AMR as sources of our MR phenomenon and point to spin dependent transport as its more probable origin.

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