Spin-dependent magnetoresistance of ferromagnet/superconductor/ferromagnet La$_{0.7}$Ca$_{0.3}$MnO$_3$/YBa$_2$Cu$_3$O$_{7-\delta}$/La$_{0.7}$Ca$_{0.3}$MnO$_3$ trilayers

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We report on large magnetoresistance in ferromagnet/superconductor/ferromagnet trilayer structures made of La$_{0.7}$Ca$_{0.3}$MnO$_3$ and YBa$_2$Cu$_3$O$_7$. We find that the shape and height of the magnetoresistance peaks are not modified when the relative orientation of current and magnetic field is changed from parallel to perpendicular. Furthermore, we find that the temperature shift of the resistance curves is independent of current and of the sweep rate of the magnetic field. These observations favor the view that the magnetoresistance phenomenon originates in the spin dependent transport of quasiparticles transmitted from the ferromagnetic electrodes into the superconductor, and rule out interpretations in terms of spontaneous vortices or anisotropic magnetoresistance of the ferromagnetic layers.

INTRODUCTION

Thin film heterostructures combining ferromagnets (F) and superconductors (S) are ideally suited to study the interplay between both long range orderings.[1,2] When a superconductor is placed in contact with a ferromagnet both long range phenomena may compete at the interface, which gives rise to a variety of exotic phenomena like π junctions, a spatially modulated order parameter, etc.[3,4] Interesting effects occur at the interface between a superconductor and a ferromagnet. Due to the F/S proximity effect the pairing amplitude penetrates into the F side and the order parameter is also depressed in the S material due to the effect of the exchange field.[1,2] When a thin superconductor is brought in contact with an inhomogeneous ferromagnet the cancellation of the exchange field over the coherent volume results in novel effects. Examples are domain wall superconductivity[5,6] or the F/S/F superconducting spin switch.[7–9] In the first case, oppositely directed magnetizations at both sides of a domain wall promote nucleation of superconductivity if the coherence length is larger than the width of the domain wall.[5,6] In the second case, antiferromagnetic alignment of the magnetizations of the F layers results in larger T_c values than does the ferromagnetic alignment, due to the averaging out of the exchange field over the coherent volume.[7–9] In recent years there has been an increasing interest in structures combining oxide ferromagnets and oxide superconductors.[10–14] In particular, the combination of high-T_c superconductors (HTS) and colossal magnetoresistance materials (CMR), gives rise to a number of new properties and behaviors, which considerably enrich the study of F/S interplay.[15,16] The unconventional pairing symmetry (d-wave) of the superconductor with an anisotropic gap exhibiting nodes in [110] directions, affords quasiparticles to be incorporated at zero energy cost. On the other hand, the highly spin polarized conduction band of the manganites will

cause the suppression of Andreev reflection, and thus conventional proximity effect, at F/S interfaces. Thus d-wave superconductivity and half metallicity will conspire to enhance the relative importance of phenomena originating at the transfer of (spin polarized) quasiparticles. Perovskite HTS and CMR materials, chosen with good lattice matching, can be grown epitaxially one on top of the other with atomically flat interfaces exhibiting no interdiffusion.[17,18] In spite of the well defined chemical interface structure the electronic and magnetic structure at the interface can be significantly more complex due to charge transfer or other interface processes.[19]

In conventional F/S/F junctions the critical temperature may be modulated by the relative orientation of the magnetization in the two ferromagnetic layers. This T_c modulation results from a compensation of the exchange field over the coherent volume in the antiferromagnetic configuration if the thickness of the superconductor is comparable to the coherence length.[7–9] Since at the interface between a half metal and a superconductor proximity effect is suppressed,[21] T_c is affected distinctly in F/S/F structures with highly spin polarized carriers. In a previous letter 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$_2$Cu$_3$O$_7$(YBCO).[20] This MR originates from a larger resistance in the antiferromagnetic (AF) configuration of the F layers, as opposed to conventional proximity coupled F/S/F structures, where the larger resistance occurs in the F alignment.[7–9]

In this paper we explore different mechanisms as possible origins of this magnetoresistance. In particular we have analyzed the relative importance of vortex dissipation, anisotropic magnetoresistance (AMR) and giant magnetoresistance (GMR) like spin dependent effects related to the transmissions of (spin polarized) quasiparticles from the fer-

romagnet into the superconductor. We emphasize especially the current dependence since a markedly different current dependence is expected for each of the three proposed scenarios. In detail, vortex dissipation is zero when the current is applied parallel to the magnetic field, the AMR is maximized when the current is applied parallel to the magnetic field, and spin dependent transport should be current independent. We show that magnetoresistance measurements are independent of the (in plane) orientation of the applied magnetic field with respect to current, and to the magnitude of the electrical current itself. Furthermore, we show that the MR is also independent of the sweep rate of the magnetic field. This rules out explanations in terms of vortex dissipation or anisotropic magnetoresistance and constitutes a strong indication that spin dependent transport of (spin polarized) quasiparticles diffusing from the ferromagnet plays a major role in the MR phenomenon. Scattering at both F/S interfaces in the AF configuration has a pair breaking effect and depresses the critical temperature compared to the F case where this scattering process is absent.

EXPERIMENT

Samples were grown on (100) 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 and that of the YBCO at 13 (15 nm) and 15 unit cells (18 nm). The structure was analyzed using x-ray diffraction and transmission electron microscopy. Further details about growth and structure can be found elsewhere.[22,23] A x-ray refinement technique using the SUPREX 9.0 software was used to obtain quantitative information about the interface roughness.[24] The T_c was determined from four-contact resistance measurements as the zero resistance temperature. The magnetization was measured using a Quantum Design superconducting quantum interference device (SQUID) magnetometer. Magnetotransport measurements were performed in a cryostat equipped with a 9 T magnet (Quantum Design PPMS-9T) and an automatically controlled sample rotator. The samples were rectangles with a 10×5 mm^2 area and contact pads fabricated by silver evaporation. The plane of the film was aligned with magnetic field within 0.1°. The magnetic field was swept between ±1 T at fixed temperatures above and below the onset of the superconductivity.

RESULTS AND DISCUSSION

We have measured magnetoresistance at selected temperatures along the resistive transition with the magnetic field applied parallel to the layers. Figure 1 shows $R(H)$ loops at various temperatures for a trilayer sample with a 13-unit cells thick YBCO layer. Current flows in the plane of the layers (current in plane geometry), perpendicular to the magnetic field direction. The magnetic field was swept be-
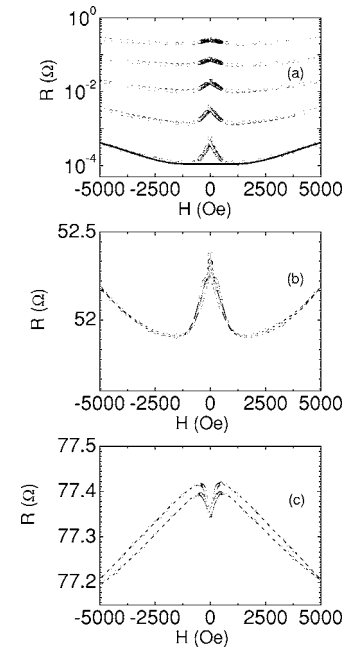

FIG. 1. (a) Resistance as a function of magnetic field, $R(H)$ loops, of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (13 u.c.)/ LCMO (40 u.c.)] at different temperatures along the resistive transition. The magnetic field, applied parallel to the layers and perpendicular to electric current, was swept between −0.5 and 0.5 T fields in an hysteresis loop sequence. The temperatures are 49, 49.5, 50, 50.5, and 51 K from bottom to top. The line in Fig. 1(a) is a fit to a thermally activated resistance with the activation energy depending on the field as $1/H^{0.5}$. (b) $R(H)$ loop of the same sample at 57 K (just below the superconducting onset). (c) $R(H)$ loop of the same sample at 61 K (just above the superconducting onset).

tween 0.5 and −0.5 T in an hysteresis loop sequence.

Large MR peaks are observed whose relative height decreases when the temperature is increased [see Fig. 1(a)]. We have previously shown that these peaks occur in a magnetic field region where polarized neutron reflectometry and SQUID magnetometry show an AF alignment between the LCMO layers[20] (not shown here). Probably, AF alignment results from the top layer having a larger coercivity than the bottom layer due to the different epitaxial strain in each layer.[20] Figure 1(a) shows that the MR peaks are superimposed on a resistance background which increases with magnetic field. Most likely this background is due to vortex dissipation since it is known that vortex motion in the liquid state is thermally activated with an activation energy depending on field as $1/H^{0.5}$. In fact, the line in Fig. 1(a) is a fit to a thermally activated resistance with the activation energy depending on field as $1/H^{0.5}$. It is worth mentioning that in samples with thicker YBCO (above 15 unit cells) such a thermally activated description of the background is hampered by the appearance of glassy properties in the vortex system at low temperatures and low fields. Figure 1 also shows that the MR peaks decrease when temperature is increased and they vanish abruptly at the superconducting onset. Figure 1(b) displays a positive MR peak at 57 K just below the superconducting onset, while Fig. 1(c) shows a much smaller negative peak at 61 K, just above the super-
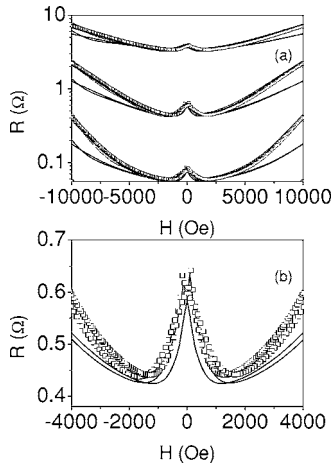
FIG. 2. (a) Resistance as a function of magnetic field, $R(H)$ loops, of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (15 u.c.)/LCMO (40 u.c.)] measured at 54.5, 55.5, and 56.5 K (from bottom to top) with the current in the plane of the layers, and applied parallel (line) and perpendicular (open symbols) to the magnetic field. (b) Enlarged view of the $R(H)$ loop measured at 55.5 K with the current applied parallel (line) and perpendicular (open symbols) to the magnetic field.

conducting onset. This proves that the superconductivity is an essential ingredient for the large MR seen in Fig. 1(a) (Ref. 20) (see the discussion below). Note also that the 61 K curve shows a decrease of the resistance when the magnetic field increases, which is characteristic of the colossal magnetoresistance of the manganite layers.

We consider three different scenarios to explain this MR phenomenon at the superconducting transition: (a) Vortex dissipation (including vortices due to stray fields of domains or domain walls), (b) anisotropic magnetoresistance (AMR), which in manganites is known to be large due to strong spin orbit scattering, and finally (c) GMR like dissipation originating at spin dependent transport. Each of these mechanisms have a very characteristic current-field dependence. Vortex dissipation is zero when the current is parallel to the field, AMR is maximized when the current is parallel to the field, and GMR is independent of both the current value and of the relative orientation of current and field. Experiments changing the current values and the direction between current and field are thus useful to explore the origin of the magnetoresistance.

Figure 2(a) shows the MR peaks of a trilayer sample with a YBCO thickness of 15 unit cells measured at 54.5, 55.5, and 56.5 K (from bottom to top) with the current in the plane of the layers, and directed parallel (line) and perpendicular (open symbols) to the magnetic field. As for the background, the high field dissipation increases substantially when the current is perpendicular to the field. For current parallel to the field the Lorentz force density on vortex lines ($J \times \phi_0$, where J is the current density and ϕ_0 is the flux quantum) vanishes and consequently the vortex dissipation due to vortices parallel to the external magnetic field vanishes as well. 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, re-
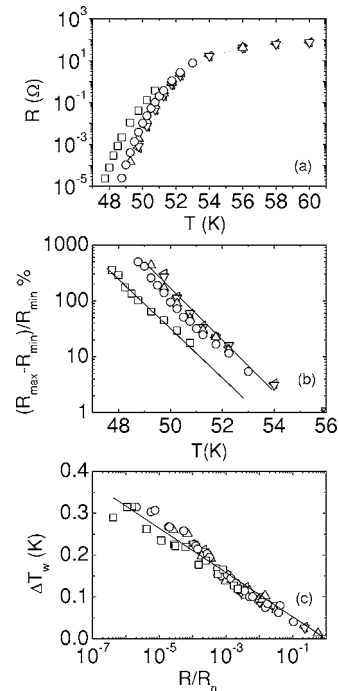

FIG. 3. (a) Resistance vs temperature curves for different current values of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (13 u.c.)/LCMO (40 u.c.)] at zero magnetic field [open squares (5 mA), open circles (1 mA), up triangles (0.5 mA), down triangles (0.1 mA), and left-facing triangles (0.05 mA)]. (b) Temperature dependence of the magnetoresistance computed from the maxima and minima in the $R(H)$ plots, using the same symbols as in (a). (c) Temperature shift of the resistance curves from parallel to antiparallel alignment of the magnetizations of the LCMO layers as a function of the resistance drop relative to the onset, using the same symbols as in (a). The solid lines are guides for the eye.

sponsible for the high field dissipation in this current-field configuration. Figure 2(b) shows an enlarged view of the MR peaks of the same sample as in Fig. 2(a), measured at 55.5 K with the current in the plane of the layers, and directed parallel (line) and perpendicular (open symbols) to the magnetic field. We want to emphasize that 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 (AMR) of the individual ferromagnetic layers, known to be important in manganite single-layer films. Nevertheless, the 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[25] [see Fig. 1(c)].

We have also performed current-dependent measurements for currents directed perpendicular to magnetic field. Increasing the current increases the transition width even at zero magnetic field as shown in Fig. 3(a). This occurs due to the increased vortex dissipation which adds a magnetic field dependent background to $R(H)$ curves (see Fig. 1). The magnetoresistance is calculated from resistance maxima and minima of the $R(H)$ loops as a resistance change relative to the background $\Delta R/R_b$ (where ΔR is the resistance value
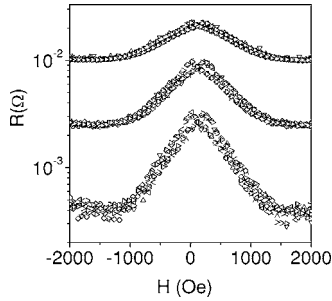
FIG. 4. Resistance as a function of magnetic field, $R(H)$ loops, of a F/S/F trilayer [LCMO (40 u.c.)/YBCO (15 u.c.)/LCMO (40 u.c.)] at 52, 52.5, and 53 K (from bottom to top) along the resistive transition. The sweep rates of the magnetic field are 50 Oe/s (up triangles), 25 Oe/s (down triangles), 5 Oe/s (diamonds), 1 Oe/s (left-facing triangles), and 0.1 Oe/s (right-facing triangles).

with respect to the background and R_b is the background resistance). An increasing current results trivially in smaller MR values as a result of dividing by the larger background resistance R_b as shown in Fig. 3(b). However by looking at the temperature shift of the resistance curve at the corresponding resistance value, instead of looking at MR (resistance shift at a given temperature), when magnetic alignment changes from parallel to antiparallel, a completely different picture emerges. Figure 3(c) shows the temperature shift, ΔT_w, for different current values as a function of resistance normalized to the onset values. The first observation is that there is a logarithmic dependence of the temperature shift as a function of resistance. Secondly, it is clear that the temperature shift is independent of current, evidencing that smaller magnetoresistance is caused solely by the increased background resistance. This provides further evidence for excluding vortex dissipation causing the MR peaks, and points strongly towards spin dependent effects on transport.

Further information about the origin of MR can be obtained by measuring the dependence of the magnetoresistance on the sweep rate of the magnetic field. On one hand, spin valve effects have been shown to be sweep rate independent,[26] while on the other hand spontaneous vortices (with a component perpendicular to the layers) induced in the superconductor due to the stray field of domains or domain walls may yield sweep rate dependent effects. Vortices created by domain walls would in principle also show a larger dissipation around the coercive field where the density of domains is maximized. One expects dissipation associated to the motion of domain walls, and thus a voltage should build up proportional to the domain wall velocity. To explore these possibilities we have done experiments changing the sweep rate of the magnetic field between 0.1 and 50 Oe/s (Fig. 4) and did not see any measurable change in peak shape or height. The explored time scale in the range of seconds is too slow for the magnetic relaxation or switching of the LCMO electrodes, where the characteristic time scale is set by the ferromagnetic resonance frequency in the GHz range. On the other hand it is a more realistic time scale for vortex relaxation phenomena (triggered by thermal activation over intersite barriers). HTS films are known to exhibit strong (logarithmic) relaxation, with a rate diverging at low temperatures.[27] The independence of the MR peaks on the sweep rate excludes interpretations in terms of spontaneous vortices or anisotropic magnetoresistance of the ferromagnetic layers and supports the view that the magnetoresistance phenomenon originates at the spin dependent transport of quasiparticles transmitted from the ferromagnetic electrodes into the superconductor.

In fact our MR phenomenon has many of the ingredients of the GMR in metallic superlattices in so far as it is independent of the current and of its direction relative to the field and depends solely on the orientation of the magnetization of the LCMO layers. Accordingly, we propose an explanation in terms of spin dependent scattering of spin polarized quasiparticles diffusing thermally from one ferromagnetic layer to the other. Although in our case the transport takes place parallel to the layers, normal electrons may diffuse from one ferromagnet to the other, keeping memory of their spin orientation if the superconductor is thin enough. In the AF configuration (for half metals) transport between the ferromagnets is not possible since there are no vacant states at the Fermi level with the right spin orientation.[28] Strong scattering occurs then at both interfaces under AF alignment, while it is absent when the magnetizations of the LCMO layers point to the same direction. The increased interface scattering in the AF configuration brings about an effective increase of the number of quasiparticles in the superconductor, which self-consistently reduces the critical temperature, thus providing a basis for the increased (magneto)resistance in the AP configuration. Recent reports have shown similar magnetoresistance on permalloy/Nb/permalloy trilayer structures,[29] suggesting that a high degree of spin polarization plays an important role in the occurrence of this phenomenon. For the YBCO thickness of this work the coupling of the F layers through normal electrons with subgap energy transmitted into the superconductor in the form of evanescent waves[30,31] is not possible. The length scale of this process is close to the Ginzburg Landau coherence length,[32] which is much shorter than the thickness of the superconducting spacer (16–18 nm) used in this work. Hopefully these results will open new theoretical avenues in the study of junctions between unconventional superconductors and spin polarized ferromagnets.[33–35]

In summary, we have found a large MR in F/S heterostructures made of highly spin polarized LCMO and high-T_c superconducting YBCO. This MR is reminiscent of the GMR in metallic superlattices as it depends on the relative orientation of the magnetic layers and is independent of the relative direction of current and field. Neither does the MR peak depend on the current values or on the sweep rate of the magnetic field. These results rule out vortex dissipation or AMR as sources of our MR phenomenon and point to a spin dependent transport as its more probable origin. However, in contrast to traditional GMR, the MR vanishes in the normal state of the YBCO and only occurs in the superconducting state. Furthermore, the MR is opposite in sign to MR effects observed in F/S/F heterostructures (superconducting spin switch) based on low-T_c superconductors and transition metal ferromagnets. The possible origin of this MR is the depressed order parameter in the superconductor due to strong interface scattering at the F/S interface in the AF configuration.

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