Giant Magnetoresistance in Ferromagnet/Superconductor Superlattices

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We show magnetoresistance in excess of 1000% in trilayers containing highly spin-polarized $La_{0.7}Ca_{0.3}MnO_3$ and high-T_c superconducting $YBa_2Cu_3O_7$. This large magnetoresistance is reminiscent of the giant magnetoresistance (GMR) in metallic superlattices but with much larger values, and originates at spin imbalance due to the injection of spin-polarized carriers. Furthermore, in contrast to ordinary GMR, the magnetoresistance is intimately related to the superconductivity in the $YBa_2Cu_3O_7$ layer and vanishes in the normal state. This result, aside from its fundamental importance, may be of interest for the design of novel spintronic devices based on ferromagnet/superconductor structures.

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The need for smaller and faster electronics has motivated increasing interest in spintronic devices based on magnetoresistance (MR) effects associated to the accumulation and transport of spin-polarized electrons [1]. Extensive work is being conducted in F/N/F structures consisting of ferromagnetic (F) layers separated by nonmagnetic (N) spacers, in which current flow is controlled by the relative orientation of the magnetization in the F layers [2–6]. In F/S/F double junctions with a superconducting (S) spacer, proximity, or spin imbalance effects have been theoretically proposed to suppress superconductivity to different extents depending on the relative orientation of the magnetizations [7,8]. These effects might thus provide new sources of MR, which are expected to increase with the degree of spin polarization of the magnetic electrodes [9]. Here we consider F/S structures containing highly spin-polarized $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) and high-T_c superconducting $YBa_2Cu_3O_7$ (YBCO) showing large magnetoresistance, which is reminiscent of the giant magnetoresistance (GMR) in magnetic superlattices [10]. However, unlike in traditional GMR, the magnetoresistance occurs only when the spacer layer becomes superconducting and vanishes in the normal state.

In F/S/F junctions the interplay between magnetism and superconductivity may depress the superconductivity in a manner controlled by the relative orientation of the magnetization of the ferromagnetic layers. At a F/S interface, various processes may occur: electrons with energies below the superconducting gap may be Andreev reflected [11] giving rise to the proximity effect, while electrons with energies larger than the superconducting gap are injected into the superconductor. Superconductivity depression in F/S/F structures due to proximity effect is larger with a parallel than with an antiparallel orientation of the F layers. Therefore, with a proper choice of the operation temperature the F/S/F structure can be switched from superconducting to normal by applying a magnetic field, thus valving the current flow through the superconductor [7,12]. In the spin injection scenario, on the other hand, when the magnetizations of the ferromagnetic layers are in opposite directions, injected electrons encounter a large resistance to tunnel out resulting in a spin imbalance [8] above the superconducting gap. This nonequilibrium spin density causes a difference between the chemical potentials for spin-up and spin-down electrons in the superconductor which has a pair breaking effect depressing superconductivity in the same way that the Zeeman effect does in the paramagnetic limit [8].

An interesting scenario arises if the ferromagnets are fully spin polarized. Andreev reflection will be suppressed since it requires electrons with both spin orientations at the Fermi level [13,14], although crossed Andreev reflection might occur [15]. However, spin imbalance effects are expected to be enhanced, eventually yielding magnetoresistance. We have investigated this issue using a high temperature superconductor (YBCO) and a highly spin-polarized ferromagnet (LCMO). Heterostructures combining these materials are experiencing increasing interest [16–18]. We have synthesized F/S/F trilayers and F/S . . . S/F superlattices on (100) oriented $SrTiO_3$, using a high pressure (3.4 mbar) pure oxygen sputtering technique at high growth temperature (900 °C). Samples were epitaxial and interfaces were atomically flat with negligible interdiffusion and little structural disorder [18,19]. The LCMO thickness was fixed at 40 unit cells (u.c.) (16 nm) and the YBCO thickness was varied between 7 and 15 unit cells (8–18 nm). The large LCMO thickness ensures that there is no possible coupling between the superconducting layers [19]. Samples were magnetic and superconducting. Saturation magnetizations were in excess of 300 $\mathrm{emu/cm^3}$, as typically found in thicker manganite samples. Superconducting T_c was depressed down to 50–60 K as a result of a strong interplay between magnetism and superconductivity [19], and should not be ascribed to the small thickness of the YBCO layers, since similar YBCO films in heterostructures with nonmagnetic spacers have T_c values close to 90 K [20].

We have measured the magnetoresistance with the magnetic field applied parallel to the layers. Current contacts were in the plane of the layers (current in-plane geometry) and aligned perpendicular to the magnetic field direction. The field was swept between 1 and -1 T at temperatures fixed along the superconducting resistive transition [$R(H)$ loop]. Figure 1(a) shows $R(H)$ loops at various temperatures for a trilayer sample with 15-unit cells thick YBCO layer. Large magnetoresistance peaks are observed whose relative height decrease when temperature is increased. Figure 1(b) shows the resistive transition (dotted line) together with data at MR maxima (solid squares) and minima (down-pointing triangles). Note from the inset of Fig. 1(b) that MR calculated as $\Delta R/R = (R_{\max} - R_{\min})/R_{\min}$ decreases exponentially with increasing temperature along the resistive transition and it is abruptly suppressed at the resistive onset of the superconducting transition. This is evidence that superconductivity plays a key role in the occurrence of this MR phenomenon and rules out a conventional GMR effect as observed in magnetic superlattices. The relative height of the peaks with respect to background depends critically on the parallel orientation (adjusted within 0.1°) of the magnetic field to the plane of the layers. Figure 2 shows an enlarged view of the 55 K $R(H)$ loop together with the hysteresis loop measured at 58 K, just at the superconducting onset. The hysteresis loop did not change appreciably when the measuring temperature was at the zero resistance value (50 K). The steplike hysteresis loops points to some degree of antiferromagnetic (AF) alignment between the two F layers, resulting from different switching fields of bottom and top layer layers as shown by polarized neutron reflectometry (PNR); see Fig. 3. The PNR was measured on the POSYI reflectometer of the Intense Pulsed Neutron Source. The measured reflectivity for incident neutrons polarized parallel (I^+) and antiparallel (I^-) to the applied field can be fit to a model that takes into account *depth-dependent* variations in the chemical (nuclear) and magnetic scattering length density (SLD) of the film [21]. The magnetic SLD is directly proportional to the magnetization. Hence, it is possible to determine separately the

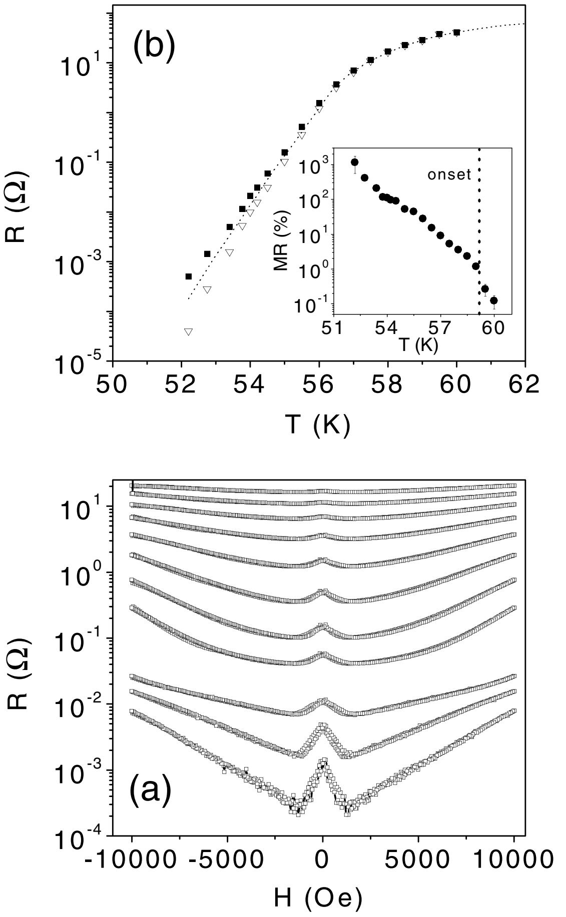

FIG. 1. (a) Resistance as a function of magnetic field, $R(H)$ loops, of a F/S/F trilayer LCMO (40 u.c.)/YBCO (15 u.c.)/LCMO (40 u.c.) at different temperatures along the resistive transition. Magnetic field, applied parallel to the layers, was swept between -1 and 1 T fields in an hysteresis loop sequence. Temperatures are 52.75, 53.4, 53.77, 54.5, 55, 55.5, 56, 56.5, 57, 57.5, and 58 K from bottom to top. (b) Resistive transition in zero magnetic field (dotted line) for the same sample. Symbols mark the temperatures for the field sweeps of the lower panel. Solid squares correspond to the MR maxima ($R_{\max}$) and open down-pointing triangles to MR minima ($R_{\min}$). Inset: Magnetoresistance calculated as $\Delta R/R$ versus temperature.

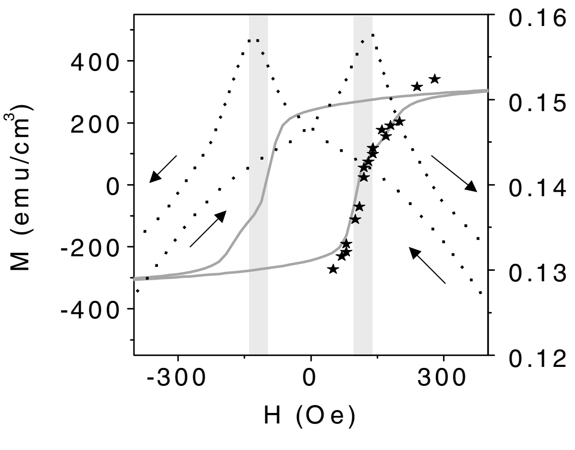

FIG. 2. Solid circles: Low field zoom of the $R(H)$ loop at 55 K of the same sample as in Fig. 1. Solid line: Hysteresis loop at 58 K (just at the superconducting onset). Stars are magnetization values obtained from polarized neutron reflectometry. Grey bars indicate the field region where the analysis of the neutron data shows AF alignment between the magnetizations of the LCMO layers (see text and Fig. 3).

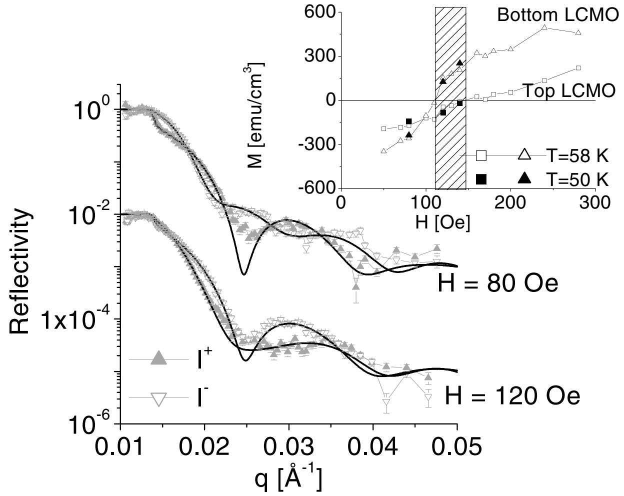

FIG. 3. : The measured (triangles) and fitted (lines) polarized neutron reflectivity as a function of momentum transfer q at 50 K for the sample in Fig. 2. Closed triangles refer to parallel neutron polarization, while open triangles refer to antiparallel neutron polarization. Inset: The magnetization of each individual LCMO layer as a function of applied field at 50 and 58 K. Data were collected after zero field cooling and for ascending fields after saturating at $H = -5$ kOe.

magnetization of the two F layers. As shown in Fig. 3, a good fit to the data can be obtained by assuming that the magnetization is along the field. The most important difference between the two data sets in Fig. 3 is the result that at $H = 80$ Oe there is ferromagnetic alignment, while at $H = 120$ Oe the layers are AF aligned (note that data were collected for ascending fields after saturating at $H = -5$ kOe). The inset of Fig. 3, showing the field dependent magnetization of the two layers, illustrates how the layers are reversing at different fields, thereby creating a field region with AF alignment. This behavior (observed at both 50 and 58 K) is probably due to the larger magnetization of the bottom F layer with respect to that of the top layer, as determined at saturation (5 kOe). Grey colored vertical bands in Fig. 2 also mark this field interval where PNR detects antiferromagnetic alignment. Interestingly, the MR peaks occur exactly in this field interval.

The question arises whether the MR is due to the magnetization switching itself or determined by the relative orientation of the magnetization in the ferromagnetic layers. The latter was found to be the case since experiments conducted on LCMO (top)/YBCO (bottom) bilayers did not show MR peaks (not shown). We can thus discard MR to originate at electromagnetic induction effects related to magnetic flux changes through the superconductor due to out-of-plane magnetization rotation at domain walls. In addition, the size of the MR peaks was independent on whether the in-plane current was parallel or perpendicular to the magnetic field. This also rules out explanations related to the anisotropic magnetoresistance (AMR) of the *single* ferromagnetic layers which, in fact, show up when the temperature is raised above the superconducting onset. It turns out that the magnetoresistance in the LCMO/YBCO/LCMO trilayers has many of the ingredients of the GMR in magnetic superlattices: it has actually giant values, it is independent of the current direction, and it depends on the relative orientation of the F layers. However, the magnetoresistance is not due to ordinary GMR since it is absent in the normal state of the YBCO layer and occurs only with the onset of superconductivity. Given that the resistance is increased with antiferromagnetic alignment of the F layers, this suggests that for the antiferromagnetically aligned magnetic layers the zero resistance critical temperature of the YBCO is reduced [see Fig. 1(b)]. Notice that this effect is opposite to the result observed by Gu *et al.* in F/S/F trilayers based on conventional low-T_c superconductors and transition metal ferromagnets, where proximity effect yields higher T_c values when magnetic layers are AF aligned [12].

We have found this magnetoresistance effect also in superlattices, though with smaller MR values. Figure 4 shows hysteresis loops above the superconducting onset of samples with seven and with 10-unit cell thick YBCO with three superconducting layers and four magnetic layers. While the sample with 7-unit cell thick YBCO layers shows a clear step in the hysteresis loop denoting AF alignment, the sample with 10-unit cell thick YBCO layers has a genuine ferromagnetic loop. The insets of Fig. 4 show MR curves at temperatures with comparable

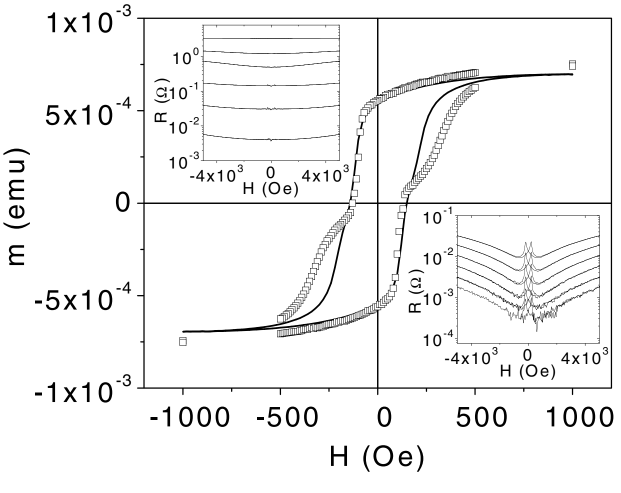

FIG. 4. Main panel: Hysteresis loops of a superlattice LCMO (40 u.c.)/YBCO (7 u.c.) showing AF alignment (open squares) and of a LCMO (40 u.c.)/YBCO (10 u.c.) superlattice not showing AF alignment (continuous line). Upper inset: $R(H)$ loops at selected temperatures of the superlattice LCMO (40 u.c.)/YBCO (10 u.c.). Temperatures are 47.5, 48.5, 49.5 50.5, 51.5, 53.5, and 60 K from bottom to top. No magnetoresistance peaks are observed for this YBCO thickness. Lower inset: $R(H)$ loops of the superlattice LCMO (40 u.c.)/YBCO (7 u.c.) showing AF alignment. Note that MR peaks are observed. Temperatures are 41, 41.5, 42, 42.5, 43, and 45.5 K from bottom to top.

resistance drop in the resistive transition. Notice that the sample not showing antiferromagnetic alignment has essentially zero magnetoresistance (upper inset), while clear magnetoresistance peaks are observed in the sample with 7 unit cells YBCO showing a step in the hysteresis loop (lower inset). The results of Fig. 4 provide further evidence that AF alignment is a key ingredient for the occurrence of the giant MR in F/S/F structures.

We now discuss the origin of this MR in the frame of the spin imbalance theory of Takahashi, Imamura, and Maekawa [8]. If the magnetizations of the F layers are antiparallel aligned, a nonequilibrium spin density is induced in the superconductor for currents perpendicular to the interface due to the imbalance of the currents carried by spin-up and spin-down electrons. In fact, assuming a nearly full spin polarization for the manganite layers, majority spins injected into the superconductor will find large resistance values to exit through the adjacent F layer. This will cause a spin density to buildup in the superconductor and, as a result, the superconducting gap is reduced yielding increased resistances for a given temperature in the AF configuration. Under ferromagnetic alignment, on the other hand, there will be no spin accumulation in the superconductor. Within this picture one would expect magnetoresistance if the thickness of the superconducting layer is shorter than the spin diffusion length. Spin relaxation in the normal state is determined by the spin flip scattering time, τ_{sf}, although when the temperature is lowered below T_c the spin relaxation time, τ_s, can be much longer [9]. In fact, the MR effect disappears in trilayers with YBCO layer thickness larger than 30 nm, suggesting that this distance might be an upper limit for the spin diffusion length.

The picture of the spin-polarized transport is more complicated in the current-in-plane (CIP) than in the current-perpendicular-to-plane geometry, but spin accumulation effects in antiparallel-aligned layers are still observed in CIP F/S structures as in other GMR or spin valve systems [1]. In the CIP geometry, equipotential planes are perpendicular to the layers, and current flows essentially parallel to the layers. However, since along the resistive transition (where our MR effect is observed) the resistance is finite, part of the current will be carried through the magnetic layers, where carriers will be frequently scattered into the superconducting layer. These electrons will cross the superconductor and will be strongly scattered at the interface with the other ferromagnetic layer if the alignment is AF. AF alignment therefore results in spin imbalance in the S layer at the F/S interface and thus in reduced T_c values (higher resistance values at a given temperature). Below the zero resistance critical temperature, current will be entirely channeled through the superconducting layer and the MR effect is not observed. Finally, we would like to point that the large MR might be related to the recently proposed inverse proximity effect in F/S structures [22].

In summary, we have found large MR (up to 1600%) in F/S LCMO/YBCO structures. This novel MR is reminiscent of the GMR in metallic superlattices insofar as it depends on the relative orientation of the magnetic layers and is independent on the relative direction of current and field, but with much larger values. However, in contrast to traditional GMR, the magnetoresistance vanishes in the normal state of the YBCO and occurs only in the superconduting state. Furthermore, the MR is opposite in sign to that observed in heterostructures 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 a spin imbalance resulting from an antiparallel alignment of the ferromagnetic layers. Apart from its fundamental interest, this large magnetoresistance in F/S/F structures may motivate the development of novel spintronic devices.

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