Critical Current Suppression in a Superconductor by Injection of Spin-Polarized Carriers from a Ferromagnet

V. A. Vas'ko, V. A. Larkin, P. A. Kraus, K. R. Nikolaev, D. E. Grupp, C. A. Nordman, and A. M. Goldman
School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455
(Received 12 September 1996)

The critical current of a thin film of the high-T_c superconductor $DyBa_2Cu_3O_7$, when incorporated in a heterostructure in which it is separated from a ferromagnetic underlayer of $La_{2/3}Sr_{1/3}MnO_3$ by an undoped ultrathin La_2CuO_4 film, has been found to be strongly suppressed by current flowing in the ferromagnetic film. A control experiment with a Au film replacing the ferromagnet did not show such a response, and resistive heating was ruled out by additional measurements. The effect would appear to be caused by pair breaking associated with spin-polarized carriers being injected into the superconductor. [S0031-9007(96)02287-9]

PACS numbers: 74.80.Dm, 74.50.+r, 74.76.Bz

The investigation of spin-polarized transport and spin-polarized tunneling in superconductors can provide useful information on spin-dependent electronic properties and spin relaxation, and may also lead to new superconducting devices. Pioneering experiments on spin-polarized tunneling in superconductors were carried out in the seventies by Tedrow and Meservey [1]. The injection of spin-polarized carriers into superconductors was first described theoretically by Aronov [2]. Johnson and Silsbee conducted the first studies of spin injection in metals [3,4]. The subject of spin-polarized transport was recently reviewed by Prinz [5]. With the rediscovery of the doped lanthanum manganite compounds [6,7] in which electrical transport involves spin-polarized carriers, interest in the subject of spin-polarized transport has been further revitalized. Lanthanum manganite, when doped with divalent metal ions of Ba, Ca, Sr, or Pb, exhibits a strong magnetoresistance which has been called "colossal" magnetoresistance (CMR). This is usually attributed to a double exchange interaction of manganese ions [8,9]. Magnetoresistance, defined as $[R(H)\text{-}R(0)/R(H)]$, can be as high as 10^3 to 10^6 percent at magnetic fields of a few Tesla at temperatures in the vicinity of the metal-insulator transition [10]. The double exchange picture implies that the degree of polarization of the charge carriers is very close to unity, unlike the case of classical metallic ferromagnets and ferromagnetic alloys [1,5]. This means that in principle CMR materials could be used as a source of spin-polarized charges for carrier injection studies. Furthermore, they have a perovskite crystal structure and therefore are compatible in the sense of epitaxial growth with high-T_c superconductors.

In this Letter we report on investigations of epitaxially grown heterostructures consisting of layers of $La_{2/3}Sr_{1/3}MnO_3$, La_2CuO_4, and $DyBa_2Cu_3O_7$ which are ferromagnetic, insulating, and superconducting, respectively. We find that current flowing in the ferromagnet produces a decrease in the critical current of the superconducting layer which we attribute to pair breaking by spin-polarized carriers crossing the boundary between the ferromagnet and the superconductor. Two samples of very similar quality and properties have been investigated. Here we report the results of detailed measurements on one of the samples.

Samples were grown by ozone-assisted, block-by-block molecular beam epitaxy (BBD MBE), a technique that has been shown to be capable of producing high-quality epitaxial films of the cuprates [11] and the manganites [12]. Details of the apparatus and procedures used can be found elsewhere [13]. The substrates used were $6.0 \times 6.0 \times 0.5$ mm (001) oriented $SrTiO_3$ single crystals. The heterostructures were prepared by growing consecutively layers of $La_{2/3}Sr_{1/3}MnO_3$, La_2CuO_4, and $DyBa_2Cu_3O_7$ with thicknesses of 400, 24, and 600 Å, respectively. A two-unit-cell-thick buffer layer of the insulator La_2CuO_4 was incorporated into the heterostructure to improve the crystal structure and properties of the $DyBa_2Cu_3O_7$ layer. Reflection high-energy electron diffraction (RHEED) patterns observed during growth suggested a high degree of both in-plane and out-of-plane orientation. The absence of spots in the RHEED patterns was evidence of planarity of the layers as they formed.

The crystal structure of the samples was characterized using a high-resolution four-circle x-ray diffractometer. The c axis of the crystal lattice of the layers was perpendicular to the (001) plane of the substrate. The c-axis lattice parameters were 3.845 Å for the $La_{2/3}Sr_{1/3}MnO_3$ layer and 11.69 Å for the $DyBa_2Cu_3O_7$ layer. The rocking curve width of the (002) diffraction peak of $La_{2/3}Sr_{1/3}MnO_3$ was 0.084°. For the (001) peak of $DyBa_2Cu_3O_7$, the corresponding value of 0.47° was higher, but still characteristic of high-quality epitaxial growth. For $DyBa_2Cu_3O_7$ films grown directly on $SrTiO_3$ substrates rocking curve widths for the (001) peak of 0.064° were typical.

The heterostructures, as shown in Fig. 1(a), were patterned for measurements using conventional photolithography. Spin-polarized carriers were injected into the superconductor by running a current parallel to the

 0031-9007/97/78(6)/1134(4)$10.00

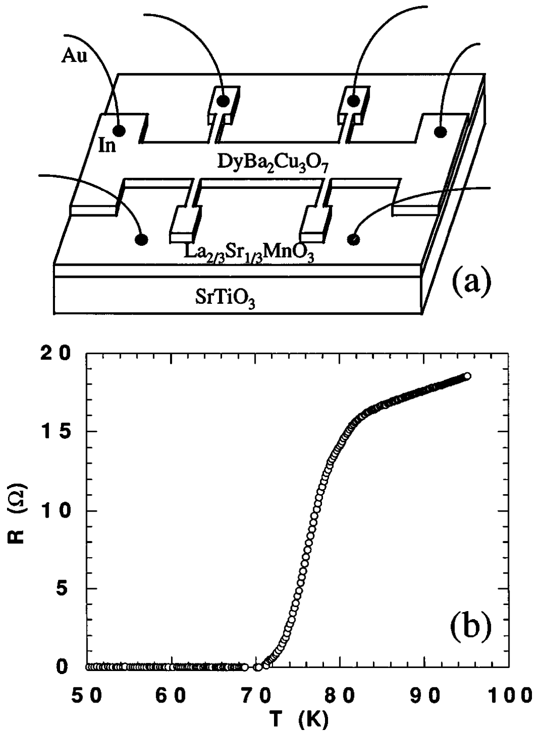

FIG. 1. (a) Geometry of the ferromagnet-superconductor sample used for measurements. The width of the $DyBa_2Cu_3O_7$ bridge was 300 μm, and the distance between the voltage leads was 3 mm. The substrate was 6×6 mm in area. (b) Temperature dependence of the resistance near the superconducting transition of $La_{2/3}Sr_{1/3}MnO_3/La_2CuO_4/DyBa_2Cu_3O_7$ heterostructure after lithographic patterning was carried out.

The temperature dependence of the resistance of the superconducting layer of one of the heterostructures is shown in Fig. 1(b). The transition temperature (zero resistance) is 71 K, which is lower than the usual transition temperature ($T_{c0} = 89$ K) of thick $DyBa_2Cu_3O_7$ films deposited on $SrTiO_3$ substrates. This may be due to some combination of the mechanical and physical influence of the underlying $La_{2/3}Sr_{1/3}MnO_3$ layer resulting in increased disorder of the cuprate layer, a direct effect of the magnetic properties of the underlayer on the superconductivity, or some degradation of the superconductor due to the photolithographic processing.

Some of the V-I characteristics of the superconducting strip obtained at a temperature of 50 K are shown in Fig. 2(a). These are shown for one direction of the current in the ferromagnet. Their asymmetry switches direction with respect to the zero current axis with a change in the direction of the current in the ferromagnet. As a function of that current, both a shift of the curves along the current axis and a reduction of the width of the plateau corresponding to zero voltage drop can be noted. The shift is a consequence of part of the current flowing in superconducting strip through the ferromagnetic layer. The superconductor effectively shorts out part of the ferromagnetic film resulting in some of the current supplied to the ferromagnet being injected into the superconductor. This geometry was chosen over direct injection to minimize lithographic processing of the heterostructure.

We characterized the $La_{2/3}Sr_{1/3}MnO_3$ underlayer by measuring the temperature dependence of its magnetization and the hysteresis in its variation with field applied in the film plane. Its Curie temperature was 330 K, and its saturation magnetization was 78 emu/g. The magnetization measured with about 0.5 G applied in the plane was a significant fraction (about 1/3) of the magnetization measured in more substantial fields with the same orientation. Indeed the effects on the I-V characteristics of currents injected into the superconductor from the ferromagnet, which will be described below, were independent of magnetic field from 10 to 500 G, suggesting that the magnetic domains were either large or substantially oriented, i.e., not randomly polarized. This is probably due to the low defect concentration and hence low coercivity of the films. Finally, the $La_{2/3}Sr_{1/3}MnO_3$ layers of the heterostructures had resistivities on the order of 100 $\mu\Omega$ cm at 100 K, which were comparable to the resistivities of the cuprate layers.

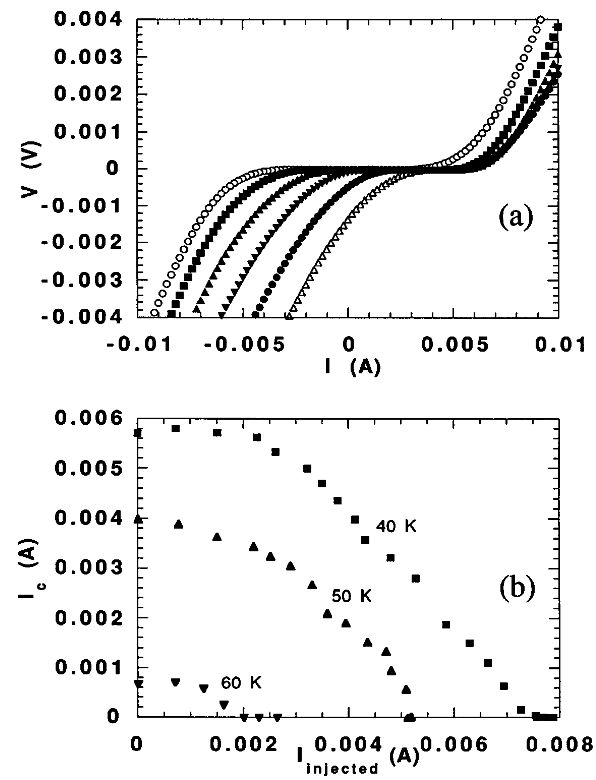

FIG. 2. (a) Voltage-current characteristics of $DyBa_2Cu_3O_7$ layer of $La_{2/3}Sr_{1/3}MnO_3/La_2CuO_4/DyBa_2Cu_3O_7$ heterostructure at $T = 50$ K when the parallel current in the ferromagnet equals 0 mA (open circles), 2 mA (squares), 4 mA (solid triangles), 6 mA (inverted triangles), 8 mA (solid circles), and 10 mA (open triangles). (b) Dependence of the critical current of the $DyBa_2Cu_3O_7$ layer of $La_{2/3}Sr_{1/3}MnO_3/La_2CuO_4/DyBa_2Cu_3O_7$ heterostructure on the current injected from the $La_{2/3}Sr_{1/3}MnO_3$ layer at temperatures of 40, 50, and 60 K.

the ferromagnetic layer bypassing it by going through the superconducting layer with zero resistance. It adds to the current supplied to the superconductor for one direction of the current in the film, and subtracts for the other. The magnitude of this effect is a measure of the current injected into the superconductor, which is equal to the shift of the curves along the current axis. About one third of the current flowing in the ferromagnet is injected into the superconductor in this geometry.

The critical current of a superconductor is usually taken to be the value of current at which 1 μV appears between the voltage probes in a four-probe dc measurement of its V-I characteristic. Because of the asymmetry in the V-I characteristic for this geometry the critical current is taken as the half-width of the plateau of the V-I characteristic over which $V < 1$ μV. Figure 2(b) shows the dependence of the critical current on the value of the current injected from the ferromagnet into the superconductor for three different temperatures well below the superconducting transition temperature. In our geometry, a current of 1 mA corresponds to the current density of 5.6×10^4 A/cm^2 which is lower than that of single film. It is not known whether this is an intrinsic effect of the ferromagnetic underlayer or a consequence of the increased disorder of these films relative to those grown directly on the substrate. Finally, the value of the injected current required to reduce the critical current to zero is comparable to the value at zero injection current.

The above observations lead us to assert that the injection into the superconductor of polarized carriers from the ferromagnet causes the reduction of the critical current of the superconductor. An unpolarized injected current would not be expected to reduce the value of the critical current significantly while it would produce a shift in the V-I characteristics if it were coupled to the superconductor in the geometry we have used. We have carried out several tests to be certain that the above assertion is correct and that our observations are not a consequence of processes other than the injection of spin-polarized carriers. First we eliminated resistive heating as a cause of the effect. It was observed that a current of 25 mA flowing in a plain ferromagnetic film of identical geometry and nearly identical resistivity was required to increase its temperature by 1 K at temperatures in the range of 40–80 K. A 1 K temperature rise would affect the critical current of the superconductor minimally. The maximum injection current in the suppression measurements of the order of 8 mA would then heat the heterostructure by not more than 1 K. Thus resistive heating cannot account for the observed reduction of the critical current of the superconducting layer. The ferromagnet-insulator-superconductor contact resistance was determined to be less than 0.1 Ω, out of a total resistance along the current path of 10 Ω, so that dissipation in the contact resistance between the two layers (across the La_2CuO_4 layer) under the conditions of the measurements should not have a significant effect on the temperature of the heterostructure.

We then eliminated the possibility that the effects were caused by a nonuniform or asymmetric distribution of current in the superconducting layer by altering the relative position of the leads connected to the ferromagnetic layer with respect to the midline of the patterned superconducting layer. We found that the critical current was reduced by a similar magnitude at the same transport current level in the ferromagnet using a variety of symmetric and asymmetric geometries. Suppression of the critical current was also observed with current flowing in the ferromagnet in a direction perpendicular to the direction of current flow in the superconducting strip [Fig. 1(a)]. In this case there was no shift in the V-I characteristic along the current axis.

Finally, a control experiment was performed to exclude the possibility that the observed critical current reduction could be produced by the injection of unpolarized carriers. An effect of this sort was found a number of years ago for metallic superconducting weak links near T_c [14]. In our control experiment the ferromagnetic film was replaced by a Au film grown on top of a patterned $DyBa_2Cu_3O_7$ film ($T_{c0} \approx 80$ K). The Au film covered the entire patterned superconducting film except for its contact pads on one side. The suppression of the critical current by current flowing in the Au film is seen in Fig. 3 to be much less than that for the case of the heterostructure containing a ferromagnetic layer. The shift of the V-I curves along the current axis demonstrates that current is indeed flowing into the superconductor from the Au film.

Although we have no detailed model of the effects we observe, they may be related to phenomena already discussed in the literature. The mechanisms determining the critical current of a film are complex, and even depend on how the critical current is defined. However, using any definition, critical current suppression should be related to order parameter suppression. There is a similarity, at least qualitatively, between the injection of spin-polarized carriers into a superconductor and the associated pair breaking, and the pair breaking resulting from spin-flip scattering when a superconductor is doped with magnetic impurities [15]. The out-of-equilibrium configuration resulting from the injection process was treated a number of years ago by Aronov [2], who showed that spin polarized carriers injected into a metallic conductor will lead to a shift $\Delta\zeta$ between the chemical potentials of carriers with opposite spins. As a result of $\Delta\zeta$, there will be a large population of polarized high-energy quasiparticle states bringing about a reduction of the superconducting order parameter. This is limited to distances the order of a spin diffusion length from the superconductor-ferromagnet boundary, which may be much greater than the corresponding length for the relaxation of unpolarized quasiparticles. This shift in $\Delta\zeta$ is on the order of $\alpha e E L_s$, where α is the degree of polarization of the current from the ferromagnet injected into the superconductor, e is the electron charge, E is the electric field intensity, and L_s is the spin diffusion length. It was already mentioned above that the polarization of the carriers in the ferromagnet may be close to

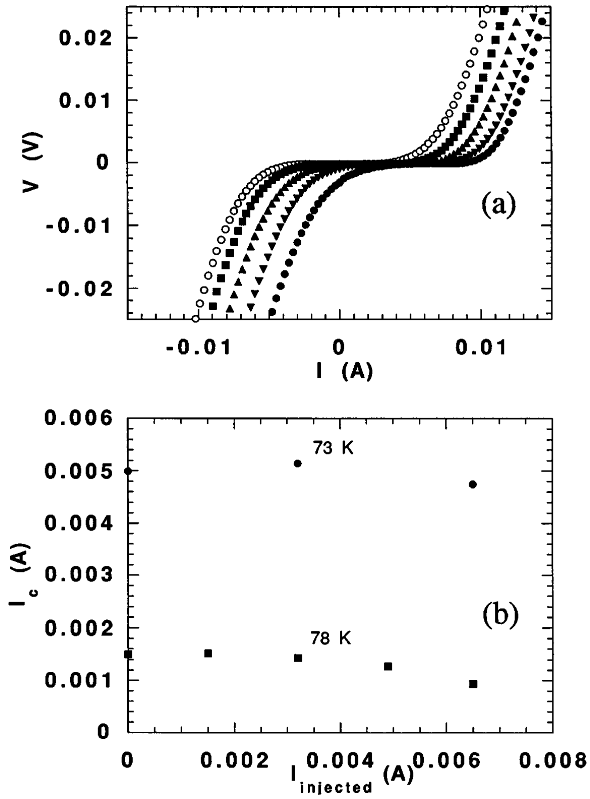

FIG. 3. (a) Voltage-current characteristics of the $DyBa_2Cu_3O_7$ layer of $DyBa_2Cu_3O_7/Au$ heterostructure at $T = 78$ K when the parallel current in the Au layer equals 0 mA (open circles), 5 mA (squares), 10 mA (triangles), 15 mA (inverted triangles), and 20 mA (solid circles). (b) Dependence of the critical current of the $DyBa_2Cu_3O_7$ layer of $DyBa_2Cu_3O_7/Au$ heterostructure on the current injected from the Au layer at temperatures of 73 and 78 K.

unity. If there is no spin-flip scattering at the boundary α may also be unity. One can estimate E to be 150 mV/cm at a current of 5 mA using the resistivity of $DyBa_2Cu_3O_7$ in the normal state and the geometry of the heterostructure. Then, if we have a spin diffusion length of a few hundred microns [16], $\Delta\zeta$ near T_c can be on the order of a few meV which would be a strong pair breaking effect.

The actual analysis of this problem is certainly more complicated, as it is necessary to take into account other phenomena associated with the boundary between the ferromagnet and the superconductor. For example, if strongly polarized carriers are injected with energies below that of the gap of the superconductor, modified Andreev scattering should prevent most carriers from entering the superconductor [17]. As a consequence, one would expect injected carriers to have energies above the gap. On the other hand, one might expect the pair potential at the surface of the superconductor to be reduced by the pair-breaking effect of carriers scattering off of the ferromagnetic boundary so that a large voltage drop might not be necessary for spin-polarized carriers to penetrate the superconductor [18].

In conclusion, we have fabricated and characterized epitaxial heterostructures of a ferromagnet $La_{2/3}Sr_{1/3}MnO_3$, an insulator La_2CuO_4, and a high-T_c superconductor $DyBa_2Cu_3O_7$. We have shown that current injected from the ferromagnetic layer reduces the critical current of the superconductor significantly. This result would appear to be due to weakening of superconductivity by the injection of spin-polarized carriers into the superconductor. There is no detailed theoretical model describing the observed effects, although it is probably associated with some out-of-equilibrium pair breaking. It is possible that fast switching devices based on this phenomenon could be fabricated. The study of the angular and geometrical dependence of the effect could also be useful in probing the symmetry of the pairing state of high-T_c superconductors.

The authors would like to thank A. I. Larkin, L. Glazman, Mark Johnson, and D. Van Vechten for useful discussions. This work was supported in part by the AFOSR under Grant No. F49620-96-1-0043, by the Office of Naval Research under Grant No. N00014-92-1368, and by the National Science Foundation under Grant No. NSF/DMR-9421941.

[1] P. M. Tedrow and R. Meservey, Phys. Rev. Lett. **26**, 192 (1971); Phys. Rev. B **7**, 318 (1973).
[2] A. G. Aronov, JETP Lett. **24**, 32 (1976); Sov. Phys. JETP **44**, 193 (1976).
[3] M. Johnson and R. H. Silsbee, Phys. Rev. B **37**, 5326 (1988).
[4] M. Johnson, Appl. Phys. Lett. **65**, 1460 (1994).
[5] G. A. Prinz, Phys. Today **48**, No. 4, 58 (1995).
[6] K. Chahara, T. Ohno, M. Kasai, and Y. Kozono, Appl. Phys. Lett. **63**, 1990 (1993).
[7] R. Von Helmolt, J. Weckerg, B. Holzapfel, L. Schultz, and K. Samwer, Phys. Rev. Lett. **71**, 2331 (1993).
[8] C. Zener, Phys. Rev. **82**, 403 (1951).
[9] P. G. de Gennes, Phys. Rev. **118**, 141 (1960).
[10] S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **264**, 413 (1994).
[11] Jean-Pierre Locquet, André Catan, Erich Mächler, Christoph Gerber, and J. Georg Bednorz, Appl. Phys. Lett. **64**, 372 (1994).
[12] V. A. Vas'ko, C. A. Nordman, P. A. Kraus, V. S. Achutharaman, A. R. Ruosi, and A. M. Goldman, Appl. Phys. Lett. **68**, 2571 (1996).
[13] V. S. Achutharaman, K. M. Beauchamp, N. Chandrasekhar, G. C. Spalding, B. R. Johnson, and A. M. Goldman, Thin Solid Films **216**, 14 (1992).
[14] T.-W. Wong, J. T. C. Yeh, and D. N. Langenberg, Phys. Rev. Lett. **37**, 150 (1976).
[15] A. A. Abrikosov and L. P. Gor'kov, Sov. Phys. JETP **12**, 1243 (1961).
[16] M. Johnson and R. H. Silsbee, Phys. Rev. Lett. **55**, 1790 (1985).
[17] M. J. M. DeJong and C. W. J. Beenakker, Phys. Rev. Lett. **74**, 1657 (1995).
[18] M. J. DeWeert and G. B. Arnold, Phys. Rev. B **39**, 11 307 (1989) have shown this to be the case for an insulating ferromagnet.