Eur. Phys. J. B **35**, 331–337 (2003)
DOI: 10.1140/epjb/e2003-00285-1

THE EUROPEAN PHYSICAL JOURNAL B

New insight on spin polarized current injection in high-T_c cuprate/manganite devices

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Received 27 June 2002 / Received in final form 3 July 2003
Published online 15 October 2003 –

Abstract. Recently, numerous experiments have been reported on critical current reductions in thin films cuprates as being due to spin injection from ferromagnetic manganites. However, little theoretical justification for these very strong effects exists, and the necessary spin relaxation length is always larger than predictions. In the present work, we investigate the possibility that these effects are due to a different origin and we report on devices designed such that the temperature of the layer itself can be measured in situ. Our data show that similar reductions of the critical current are quantitatively correlated to heating in the manganite electrode due to dissipation of the polarization current.

PACS. 72.25.Hg Electrical injection of spin polarized carriers – 74.76.Bz High-Tc films – 74.80.Dm Superconducting layer structures: superlattices, heterojunctions, and multilayers

1 Introduction

Making a superconducting three terminal device with a decent gain has been a long standing research effort for many years. Among other possibilities, it has been proposed that the critical current of a superconducting strip can be efficiently modulated by injection of spin polarized electrons. Such a device would be part of the growing spin-based electronics often named "spintronics" [1]. The key ingredient for reliable and predictible operation is that the spin effect dominates all the other possible sources of reduction of the critical current.

High-T_c superconducting devices look particularly attractive in this field. The discovery of ferromagnetic manganites boosted research, due to their almost 100% carrier polarization, together with their perovskite structure which provides excellent lattice matching, and good interfaces with superconducting cuprates. Numerous experiments [2–6] report critical current (I_C) reductions of a high T_c superconducting bar (S) when a spin-polarized current (I_{pol}) is applied from a ferromagnet (F).

The spin-injection mechanism relies on the fact that the large spin relaxation time expected in such materials would prevent polarized quasiparticles from recombining into pairs, therefore depressing the order parameter. In other words, the quasiparticle lifetime would be enhanced due to the large number of polarized quasiparticles. This mechanism is basically a non-equilibrium effect where the spin polarization simply increases the relaxation time. The subsequent decrease of the order parameter also affects the critical current. It can also be seen as a magnetic effect, where the shift between the chemical potentials of the up and down spin populations is analogous to an exchange field [7].

Battacharjee and coworkers [8] have calculated the reduction of the normalized gap for a given excess of quasiparticles in both *s*-wave and *d*-wave cases as a function of n, where n is the number of excess excitations in the spin-injection volume. According to their work, the size of the experimental effects reported in references [2–6] would then require values of $\frac{n}{4N(\varepsilon_F)\Delta(0)}$ of about 10^{-1} where $N(\varepsilon_F)$ is the density of states at the Fermi level and $\Delta(0)$ the zero temperature gap. In the framework of the interlayer tunneling model (see Ref. [8] and references therein), the variations of Ic would be about 5% for $\frac{n}{4N(\varepsilon_F)\Delta(0)} = 10^{-2}$. This seems quite difficult to achieve in practice. Another question is how the decrease of the order parameter affects the critical current which is dominated by vortex pinning, given that the depairing critical current is never attained in real experiments.

Takahashi and coworkers [7] gave a different calculation in the case (quite different from the experimental situation described below) where S is sandwiched between two F layers, and where the spin relaxation length is larger than the thickness of S.

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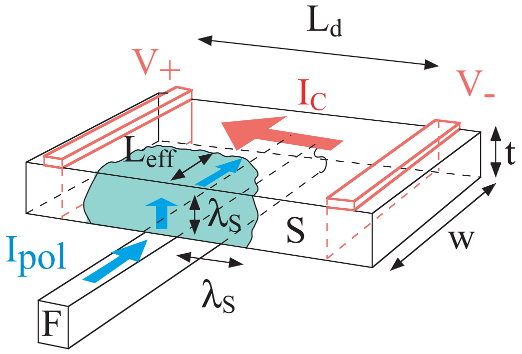

Fig. 1. Sketch of an experiment of current injection from a ferromagnetic bar into a superconducting bar, and consequent measurement of the critical current. The injection volume V_i is shown by the shaded area, and the detection volume V_d is the part of the sample situated between the two voltage pads. In such a configuration, $V_i = L_{eff} \times \lambda_s \times t$, since λ_s is generally larger than the superconductor thickness t. The out-of-equilibrium volume is limited along the direction of I_{pol} by L_{eff}, the maximum value of L_i and λ_s, and by λ_s perpendicular to the current.

In order to properly analyze a "spin-injection" experiment, two elements must be considered and compared: firstly, the spin-injection volume where the non-equilibrium effects are generated, and secondly the detection volume. Let L_i be the injection length, *i.e.* the length along the interface over which the currents are injected from the ferromagnetic material to the superconductor. L_i will depend on the interface resistance as well as on the ferromagnetic resistance. Let us define L_{eff} as the maximum length between the injection length L_i and the spin relaxation length λ_s. The spin-injection volume will therefore be defined by the spin-injection area (L_{eff}^2 or $L_{eff} \times \lambda_s$ depending on the geometry of the injection electrode) times the spin relaxation length λ_s or the superconductor thickness t, when $\lambda_s > t$ (which is most often the case). In Figure 1, the injection volume is sketched in grey. Actually, if I_{pol} is extracted through another ferromagnetic electrode, a similar volume can be defined at the extraction (not shown for the sake of clarity). The detection volume V_d will correspond to the part of the superconductor where a non-zero voltage can be measured. In Figure 1, V_d is represented by the volume of the sample included between the two voltage pads.

If the detection volume is external to or too large compared to the spin-injection volume, then no effect can be measured. The ideal experiment is to have the detection volume included in the spin-injection volume. In practice, this condition is very difficult to meet for the type of experiments discussed here, however, the condition that only the section of V_d perpendicular to the current be entirely included within V_i turns out to be sufficient. This condition is also necessary. As represented schematically in Figure 1, if L_{eff} is smaller than the width of the superconducting strip w, then the order parameter (and therefore the critical current density) will only be depressed over a small volume, and the critical current will short-circuit this region. Therefore, only a very weak effect will be detected due to the fact that the effective width of the superconducting bar is only slightly reduced. This will never lead to a complete suppression of the critical current as reported by many groups. In conclusion, to measure such an effect, L_{eff} has to be of the order of magnitude of w.

The argument will be similar for the vertical direction: in this case, the spin relaxation length λ_s will have to be the same order of magnitude as the superconducting layer thickness t, which is probably met in most of the experiments reported to date.

As we will see in the next section, due to the above considerations and because of the geometry of the injection, most of the experiments reported to date would actually imply a spin-relaxation length of hundreds of microns or even millimeters... However, a theoretical estimate in the normal state of cuprates by *Q*. Si predicts about 0.1 μm [9].

When comparing the huge field of applications that could be opened by a truly "spintronic" high-T_c device, and the lack of theoretical justification for such important effects (in particular such large spin relaxation lengths), it is very important to carefully test all spurious effects that could account for the experimental data reported to date.

The aim of the present work is to analyze these data with great care, and to present the results of an experiment performed to probe the physical origin of the critical current reductions. We also propose a more suitable experiment for measuring critical current reductions due to spin injection.

2 Brief review of the experiments

The first experiment was reported by Vas'ko *el al.* [2] in 1997 in an underdoped (T_c = 71 K) $DyBa_2Cu_3O_7$ (DBCO) bar patterned on a $La_{0.66}Sr_{0.33}MnO_3$ (LSMO) layer. A buffer layer of 2.4 nm La_2CuO_4 (LCO) was deposited in between. The critical current of the bar I_c was measured as a function of the polarized current I_{pol}, which was applied between two contacts directly on the LSMO layer. Important reductions of I_c were reported at all temperatures until complete suppression of the superconductivity ($I_c = 0$). When injecting from a gold electrode I_c was found to only slightly decrease with I_{pol}. The authors therefore concluded in favor of a pair-breaking effect due to massive injection of spin-polarized carriers. As discussed in the introduction of this work, L_{eff} is required to be at least 300 μm (the width of the $DyBa_2Cu_3O_7$ bar) or more likely about 1 mm (according to Fig. 1a in Ref. [2]) in order to depress the order parameter over the whole width of the bar, and thereby allow measurements for the complete reduction of the critical current. Taking an upper limit of 1 kΩ cm for the LCO resistivity at 100 K (which is highly over-estimated for a 2.4 nm thick layer which has little chance of being continuous), and a value of 100 $\mu\Omega$ cm for the resistivity of LSMO given by Vas'ko and coworkers, a very simple calculation gives an injection length L_i of 30 μm. This value is at least an order of magnitude too small to explain the observed effect. As far as the spin relaxation length itself is concerned, 1 mm is not physically reasonable. This argument also applies to hot normal quasiparticle effect since in this case the quasiparticle lifetime and the mean free path should be even smaller.

The reason given for excluding thermal effects was that in a similar LSMO film the rise of temperature due to a current was measured and estimated to be no more than 1 K. Measuring the overall increase in temperature of the substrate is meaningless, since even a very localized hot spot on the superconducting bar is enough to significantly decrease the critical current, while leading to only a moderate heating of the substrate itself, as we will demonstrate further. Moreover, the heating will depend on the effective resistance encountered by the polarization current, therefore will be very different between a plain LSMO film and the experimental configuration. The only way to properly address this problem would be to measure the temperature locally.

In a similar experiment, Dong *et al.* [3] tried to separate purely magnetic effects from heating effects by comparing injection in $YBa_2Cu_3O_7$ (YBCO) from $Nd_{0.66}Sr_{0.33}Mn0_3$ (ferromagnet with a resistivity of 1000 $\mu\Omega$ cm) to from a paramagnetic material $LaNi0_3$ (with a resistivity of 300 $\mu\Omega$ cm), whose cristallographic structure are very similar. In both cases, a reduction of I_c was observed, smaller for $LaNi0_3$ injection. This clearly points towards a heating effect, since it scales inversely with the resistivity of the injection electrodes. Surprisingly enough, the authors concluded to magnetic effects and excluded thermal effects.

Again, the injection length can be calculated in this configuration which yields a value of 40 μm. If the contact used to inject the current is separated by more of 40 μm to the nearest voltage pad used to measure the critical current, then the effect has no chance of being observed. (In this case the spin-injection volume will be external to the detection volume.) This information is not given in reference [3]. Again, if the width of the device is larger than 40 μm (which is the case for many of the samples), varying the widths from 20 to 250 μm should have a strong effect, which was not reported.

Following a different approach, Yeh *et al.* [4] used the same type of comparison with pulsed measurements technique in order to track heating effects. The effect was found to depend on the measuring rate, and the reduction of I_c was found to be considerably lowered as compared to the dc measurements for the same configuration. This is "smoking gun" evidence for heating. Nevertheless, the authors entirely attribute the critical current variations measured when using pulses of 200 μs width to spin-injection. In order to draw such a conclusion, one would expect to see a saturation of the effect when decreasing the pulse length, which was not reported. As a matter of fact, there is no indication in this work of an effect other than heating. Until recently, other experimental papers have been published using various configurations, all of which report that similar effects can be entirely attributed to spin injection, while none are able to rule out heating effects [5,6,10]. Koller *et al.* [11] attributed the effect to heating due to the high resistance of the F/S interface. Mikheenko *et al.* [12] used a SQUID experiment to show evidence that, at least for current pulses longer than 100 ms, the effect is dominated by heating.

A review paper from Gim and coworkers [13] gave similar conclusions on some of the experiments described above. Their approach relies mainly on the fact that no suitable comparison is made with a proper non-magnetic reference material with comparable resistivity and interface resistance. In their own experiment, they do not see any effect when the injected current is lower than the critical current. However, their analysis consists in separating spin effects from "hot" quasiparticle effects, which they claim might be responsible for the observed phenomena. However, it seems to us that this effect is even less plausible than the spin-polarization effect, given the argument above that the recombination time should be even smaller for normal quasiparticles than for spin-polarized quasiparticles. Simple heating cannot be ruled out. The estimations of the power dissipated per cm^2 are not reliable since there is no reason to believe that this power is homogeneously distributed over the sample; depending on the current paths, it might be substantially higher locally.

To summarize, careful analysis of the most referenced experiments which report a lowering of the critical current due spin-injection effects shows that their experimental configurations are far from being optimal. As is most often the case, either the injection volume is not included within the detection volume or it is to small to produce sizeable effects on the critical current unless the spin relaxation length is $\sim$100 μms or even mms. However, even in ultrapure Al single crystal at very low temperature, λ_s reaches only 100 μm [13]. It is quite hard to imagine that it could be an order of magnitude larger in oxides at about 100 K. Moreover, in many experiments, a heating effect is clearly evidenced and therefore cannot be ruled out.

In all the measurements described previously, no independent measurement of the *in situ* film temperature was reported, which should be the proper test for ruling out heating effects. We therefore designed two suitable devices using the LSMO/YBCO system.

3 Results and discussion

c-axis LSMO/YBCO bilayers were grown using the pulsed laser deposition technique on a $SrTi0_3$ substrate, as described elsewhere [14]. The thicknesses were 200 nm and 50 nm respectively. A superconducting bar 400 μm wide with a micro-bridge 40 μm wide and 140 μm long was designed, using either the Ar milling technique, or ion irradiation (Aramis – Orsay) which made the material insulating out of the region of interest. On the injection electrodes, YBCO was removed using dilute H_3PO_4

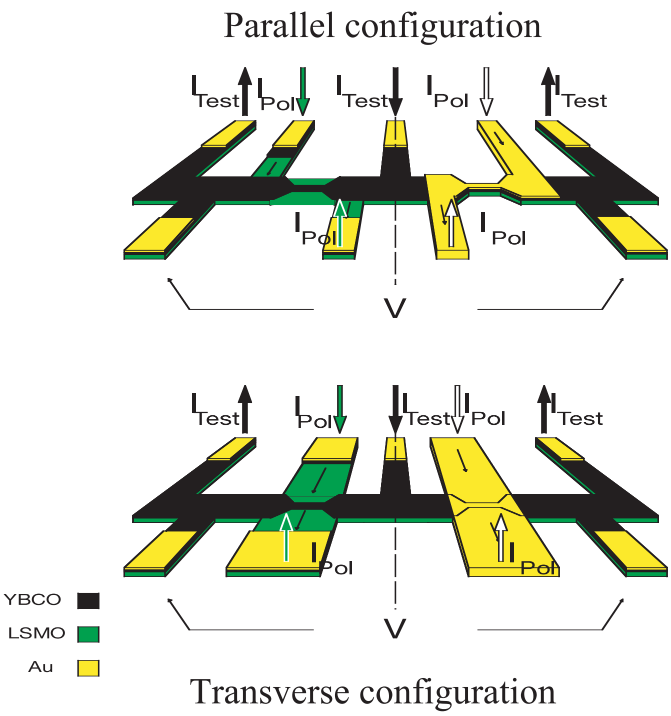

Fig. 2. Schematics of the injection geometry on our samples. Black: $YBa_2Cu_3O_{7-d}$, dark grey: $La_{0.66}Sr_{0.33}MnO_3$, light grey: gold. a) Parallel injection geometry; b) Transverse injection geometry. The length of the microbridge is 140 μm, and the width is 40 μm.

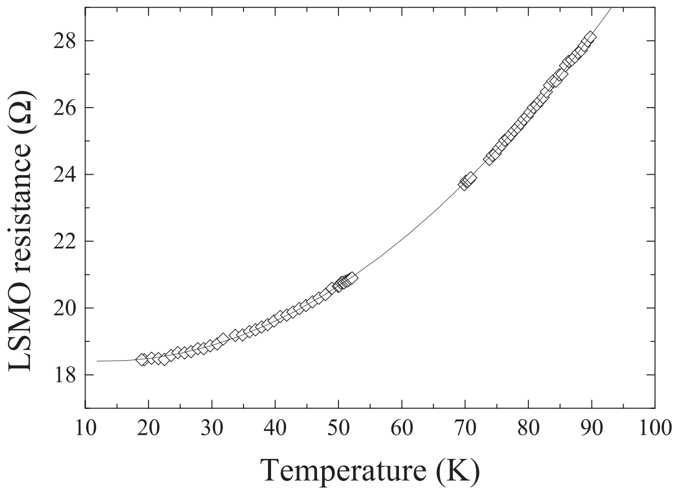

Fig. 3. Resistance as a function of temperature for the $La_{0.66}Sr_{0.33}MnO_3$ electrode.

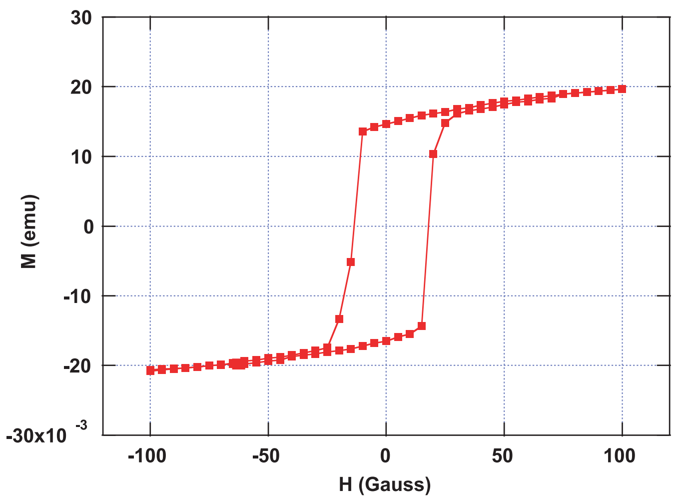

Fig. 4. Magnetic properties of the ferromagnetic electrode: magnetization curve at 35 K.

wet etching. The configuration schemes are presented in Figures 2a and 2b for parallel and transverse injection configurations respectively. They have two main advantages: first, since the injection circuit is totally under control and based on the known resistivity of the material, one knows how much current is injected from the ferromagnetic material and in which part of the sample. The current effectively injected can be measured by the displacement of the current/voltage curves of the superconductor in the parallel configuration. It was verified that the current flowing through the LSMO electrode was totally injected in $YBa_2Cu_3O_{7-d}$. Second, the resistivity of the LSMO electrode as a function of temperature can be measured independently, which provides an accurate thermometer within the sample (see Fig. 3). A comparative device with injection from a gold electrode was also designed on the same sample.

Magnetization measurements were performed on a similar LSMO layer, which showed excellent ferromagnetic properties with coercitive fields of about 20 Gauss, and a Curie temperature of more than 320 K, as expected (see Fig. 4).

Sample #AL1840 had a T_c of about 86 K and a parallel injection configuration. Sample #AL1784 was underdoped, with a T_c of about 51 K and a perpendicular injection configuration. The I/V curves for different values of I_{pol} are shown in Figures 5 and 6, for sample #AL1840 and #AL1784, respectively. A reduction of I_c with I_{pol} is clearly evidenced in both cases. For the parallel configuration, the curves were corrected using the current displacement (equal to I_{pol}) due to the superposition of the "spin-polarized" current and the probe current. The curves were nearly identical with no applied magnetic field and in magnetic fields of 1600, 3300 or 4100 G, applied either in the plane of the layer or perpendicular to it. The effect was therefore independent of the ferromagnetic domains orientation.

For both samples, we first measured the resistance R of the LSMO electrode as a function of temperature (Fig. 3). The tangent slope $\gamma = \Delta R/\Delta T$ in the vicinity of the sample temperature T was then determined. Next, the variation of I_c as a function of temperature was measured and its slope $\alpha = -\Delta I_c/\Delta T$ was calculated in the vicinity of T. Finally, I_c and R were measured for different values of I_{pol}, and both happened to be linear in I_{pol}^2 ($\Delta I_c = -\eta I_{pol}^2$ and

Table 1. Parameters giving the tangent slope of R; $\gamma = \Delta R/\Delta T$ and $\beta = \Delta R/I_{pol}^2$, and the tangent slope of I_c; $\alpha = -\Delta I_c/\Delta T$ and $\eta = -\Delta I_c/I_{pol}^2$.

Parameters		γ (mΩ/K)	α (mA/K)	β (Ω/A^2)	$\alpha\beta/\gamma$ (A^{-1})	η (A^{-1})
AL1840	$T = 70$ K	187.14	1.1	4387	25.8	26.5
AL1840	$T = 78$ K	213.15	0.28	5396	7.1	7.1
AL1840	$T = 80$ K	219.84	0.17	5509	4.2	4.0
AL1784	$T = 30$ K	42.3	0.116	246.7	0.68	0.45

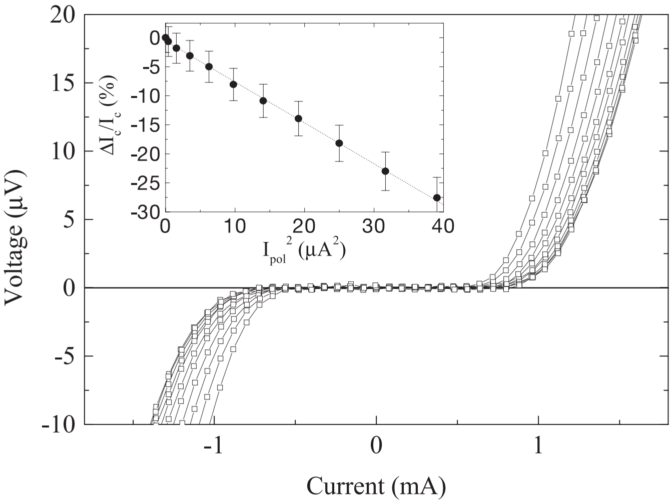

Fig. 5. $I(V)$ characteristics at 78 K for sample AL1840 (parallel configuration). I_{pol} varies from 0 mA to 6.25 mA in steps of 0.625 mA. The $I(V)$ curves originally appear displaced from I_{pol} since I_{pol} and I_{test} add to each other. Corrections from this displacement have been made. The inset shows the quadratic decrease of I_c with I_{pol} due to heating. The dotted line is the calculation based on measured sample heating.

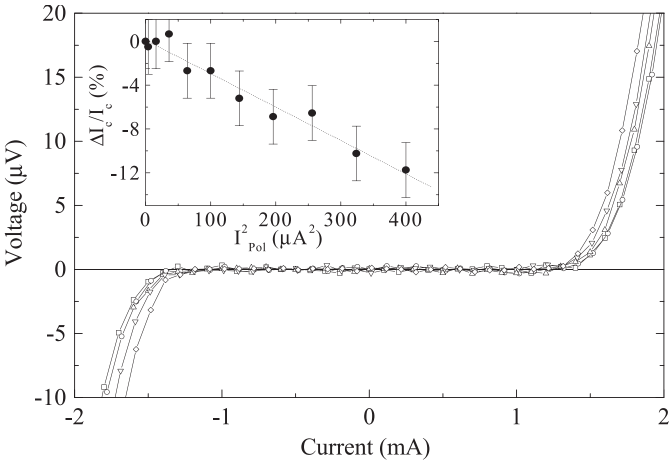

Fig. 6. $I(V)$ characteristics at 30 K for sample AL1784 (transverse configuration). No displacement was observed as expected for a purely transverse configuration. I_{pol} varies from 0 mA to 16 mA in steps of 4 mA. The inset shows the quadratic decrease of I_c with I_{pol} due to heating. The dotted line is the calculation based on measured sample heating.

$\Delta R = \beta I_{pol}^2$). The fact that R increases with I_{pol} is experimental proof that heating occurs during the injection of current through the ferromagnetic electrode.

The experimental values of η were then compared to the values corresponding to a heating effect of the LSMO electrode due to the current flowing through it, estimated using $\Delta I_c = -(\alpha\beta/\lambda)I_{pol}^2$ (see Tab. 1) As can be seen in the inset of Figures 5 and 6, the agreement between ΔI_c inferred from heating effects and the measured ΔI_c is extremely convincing, and the effect can be unambiguously attributed to heating in the ferromagnetic electrode, for both samples.

During the experiment, the sample was thermally coupled to a copper block and the temperature was measured with a thermometer very close (2 mm) to the substrate. The temperature rise on this probe was never more than 1 K, which proves that this test is not concluding and that only a local probe (the LSMO electrode resistance itself) can rule out heating effects.

Could spin-injection effects be masked behind heating effects? Of course, spin-polarized carriers do enter the superconductor and live for a time τ_s before relaxing. This should certainly be the case in this system due to the high polarization rate of LSMO, and due to the *d*-wave character of the superconductivity of $YBa_2Cu_3O_{7-d}$ where low energy excitations (and therefore spin-polarized carriers) can enter the superconductor much more easily than in *s*-wave superconductors for instance. However, the large effects reported by the groups referenced above and attributed to spin injection were not present in our samples. This is attested by the results in Table 1. It is also to be noted that the variations of I_c reported here are lower than those reported in the literature, therefore an extra amount of heating cannot be the cause. As a matter of fact, the interface resistance in our device is very low (typically 10 $\mu\Omega\,cm^2$) since there is no artificial barrier between the SC and F materials, therefore the Joule heating is also low there. Of course, the question of the injection length (which is smaller in our case, estimated to be 8 μm) remains, but it is comparable to the width of the microbridge (40 μm), and thus more favorable than in most other experiments. Moreover, the spin injection volume is automatically fully included in the detection volume for our measuring configuration.

Lastly, in order to reconcile our results with the conclusions in previous papers, we would need to assume that the effective polarization rate of the current is very low in our samples. We first rule out the possibility that the magnetic ordering at the top surface of LSMO is somewhat degraded. The polarization rate and the magnetic quality of LSMO are attested by numerous experiments performed on films grown in the same deposition chamber by the Thales-CNRS group on magnetism, where an effective polarization rate of about 80% is found [16,17]. Moreover, the SPES work done by Park *et al.* on LSMO films shows that within the temperature range of our measurements, the surface polarization is of the order of magnitude of the bulk polarization [18].

The remaining possibility would be that the spin relaxation rate at the interface is very large: this is connected to the role played by the interface barrier and the need for an artificial one. Let us first recall that other groups have reported "spin-injection" effects without any artificial barriers [5,12]. We do believe that at least at the injection point, the current in our experiment is spin-polarized even without the presence of a barrier at the interface. The mechanisms of depolarization at the interface are twofold: firstly, some disorder at the interface can produce spin-orbit coupling which flips the spin; and secondly, the electronic mismatch between the two materials may increase the time needed for an electron from the F material to enter the SC which may then decrease the polarization rate in the SC [15]. As far as the first point is concerned, the presence of a barrier plays a depolarizing role, by increasing the number of interfaces, the level of defects and impurities, and therefore the level of spin-orbit scattering. For the second point, Fert and Jaffrès [15] have established the following criterion: in order to inject a current from a ferromagnet into a metal or a semiconductor with a reasonable polarization rate, one needs to ensure that the products of the resistivity times the spin diffusion length are comparable in both materials. In fact, if this product is lower in the metal than in the ferromagnet, then the current is correctly polarized. If it is higher, then the presence of a resistive barrier is needed to ensure proper polarization. In our case, the electrons entering the SC experience a metallic DOS with a quasiparticle resistivity (extrapolated from 100 K) of about 30 $\mu\Omega\,$cm at 30 K. The spin diffusion length in YBCO is taken to be 0.1 μm according to estimates by Si [9]. The product in YBCO is then 3×10^{-4} $\mu\Omega\,\text{cm}^2$. In LSMO, the resistivity is about 100 $\mu\Omega\,$cm at the same temperature. Therefore, the LSMO spin relaxation length (SRL) has to be about (or superior) to 30 nm in order that the injected current polarization rate be close to the LSMO polarization rate. This seems attainable in practice, since in a typical ferromagnetic material such as Co, the spin relaxation length is 59 nm. If for any reason the spin relaxation length in YBCO is reduced, the condition for the SRL in LSMO will be relaxed. The native interface resistance will relax this condition too. Therefore, as the conditions for satisfactory spin-polarized injection are fulfilled without an insulating layer, the role of a barrier would only be to add spin scattering.

4 Conclusion

We investigated the possibility that the critical current reduction experiments reported by many groups could be due to heating instead of spin-injection. Our work was motivated by the lack of theoretical explanation for the huge spin relaxation length deduced form the experiments and the observed strength of the effects, which were not consistent with the occurrence of an out-of-equilibrium effect taking place over a very limited volume.

A specific device was designed, where the injection currents are not distributed uncontrollably over a plane, but instead over well-defined ferromagnetic electrodes which carry the injection current. The interest lies in the possibility of measuring the ferromagnetic electrode resistance as a function of the injected current, and therefore be able to locally probe possible rises in temperature.

We observe a local rise in temperature in the presence of a polarization current, despite all the care to ensure optimal thermalization of the sample. This temperature rise was quantitatively correlated to the variation of the critical current by independently measuring the critical current variations and the LSMO resistance variations as a function of temperature.

We estimate that "spin-injection" experiments should be revisited and examined more critically in light of the above considerations and results. At least the existence of critical current reductions due to "spin-injection effects" in high-T_c superconductors should not be systematically taken for granted, especially as far as potential applications are concerned, and the configuration of the measurement should be carefully examined. For every new experiment the constraint over λ_s should be evaluated and compared with the theoretical predictions.

Indications were given above of the characteristics necessary to properly measure such an effect. In particular, the dimensions of the superconducting bar and the distance between the electrodes should obey certain constraints in order to properly probe the out-of-equilibrium volume.

The experiment proposed by Takahashi and coworkers [7] offers a more straightforward way of putting into evidence the effect. More recently, an experiment directly probing the order parameter (and not the critical current) was performed, which is extremely promising [19].

The authors gratefully acknowledge E. Jacquet, F.R. Ladan, C. Martinet, P. Monod, F. Lalu and S. Gautrot.

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