## Superconducting proximity effect in a mesoscopic ferromagnetic wire

M. Giroud, H. Courtois, and K. Hasselbach

Centre de Recherches sur les Très Basses Températures-C.N.R.S. associated to Université Joseph Fourier, 25 avenue des Martyrs, 38042 Grenoble, France

D. Mailly

Laboratoire de Microstructures et de Microélectronique-C.N.R.S. 196 avenue H. Ravera, 92220 Bagneux, France

B. Pannetier

Centre de Recherches sur les Très Basses Températures-C.N.R.S. associated to Université Joseph Fourier,

25 avenue des Martyrs, 38042 Grenoble, France

(Received 15 June 1998; revised manuscript received 21 August 1998)

We present an experimental study of the transport properties of a ferromagnetic metallic wire (Co) in metallic contact with a superconductor (Al). As the temperature is decreased below the Al superconducting transition, the Co resistance exhibits a significant dependence on both temperature and voltage. The differential resistance data show that the decay length for the proximity effect is much larger than we would simply expect from the exchange field of the ferromagnet. [S0163-1829(98)51442-7]

Superconducting proximity effect consists in inducing superconductive properties in a nonsuperconducting metal. Although this effect has been studied for a long time,<sup>1</sup> it has gained some renewed interest due to recent experiments performed on samples of mesoscopic size. In such samples, the electron phase-breaking length  $L_{\omega}$  is larger than the sample length L. One can thus probe experimentally the characteristic energy scale of the proximity effect  $\epsilon_c = \hbar D/L^2$ , which is the Thouless energy related to the sample length. This has led for instance to the observation of large magnetoresistance oscillations in normal metal (N) loops in contact with a superconducting (S) island.<sup>2-4</sup> These oscillations provide a direct evidence for the long-range (up to  $L_{\omega}$ ) nature of the proximity effect. Another recent and striking result is the reentrant behavior. The excess conductance induced by proximity effect is maximum at a temperature or a bias voltage equivalent to the sample Thouless energy,<sup>5</sup> but the normal state conductance reappears at lower energy.

Most experiments were performed in noble metals or semiconductor two-dimensional (2D) electron gas, where electron interactions are negligible. In a free electron model, the zero-temperature, zero-bias resistance of a mesoscopic metallic wire is predicted to recover the normal state value.<sup>6–8</sup> In the presence of interactions, theoretical studies<sup>7,9</sup> predict a severe modification of the transport properties. Attractive (respectively, repulsive) electron-electron interactions are believed to result in a resistance lower (respectively, higher) than the normal-state one.<sup>7</sup> This could provide a probe for interactions in normal metals like Au, Ag, etc.<sup>10</sup>

In this article, we present an experimental study of the superconducting proximity effect in a ferromagnetic metal (F). Magnetic metals are in the strong interaction limit. Exchange interactions between electrons in a ferromagnet usually lead to efficient Cooper-pair breaking in *F-S* structures. However, it is worthwhile re-examining the actual proximity effect in a small ferromagnetic wire.<sup>11</sup> Some experiments<sup>12,13</sup> suggested long-range coherence effects, but without any clear conclusion.

Samples (see Fig. 1) were fabricated using a two-step lift-off process. The 50 nm thick Co layer was e beam evaporated on the patterned resist that was subsequently lifted off. The 100 nm thick Al islands were deposited after a soft in-situ ion-milling of the Co surface. The in-situ cleaning is a crucial step to achieve the desired high transparency of the Co-Al interface. In order to generate interferences, the Co conductor included a 0.5  $\mu$ m square loop. The distance between the Co reservoirs is 2  $\mu$ m. Many samples were patterned on the same substrate, with zero, one, or two Al islands. In the last two cases, one Al island was also linked to four contacts, in order to measure the Al wire and the Co/Al junction resistances. The width of the Co and Al wires were 100 and 140 nm, respectively. Here we will focus on two typical samples labeled 1 and 2, with one and two Al islands, respectively. The behavior of each of these two samples is representative of the properties of four samples we measured.

Figure 2 shows the temperature dependence of the resistance of samples 1 and 2. The normal-state resistance of samples 1 and 2 is 96.09  $\Omega$  and 98.35  $\Omega$ , respectively. With a Fermi velocity of 1.9.10<sup>6</sup> m/s in Co, we get an elas-



FIG. 1. Micrograph of sample 1. The Co wire and loop is in metallic contact with one Al island. Thin residual Al strips appear on the sides of the Al wire. The side length of the Co loop is 500 nm. The four Co contact pads are labelled I+, I-, V+, V-. The Al island is patterned with four contacts i+, i-, v, +v-, as indicated. In sample 2 (not illustrated), a second Al island is patterned on the left-hand side on the Co loop.

```
R11 872
```





FIG. 2. Temperature dependence of the resistance of samples 1 and 2. Sample 1 has one Al island in contact with the Co loop, sample 2 has two. The normal-state resistance, respectively, 96.09  $\Omega$  and 98.35  $\Omega$ , has been subtracted. Bias current 0.1  $\mu$ A.

tic mean free path  $l_e$  of 1.1 nm and a diffusion coefficient  $D = v_F l_e/3$  of 6.9 cm<sup>2</sup>/s. As the temperature is decreased below the Al superconducting transition, the resistance of both samples decreases, reaches a minimum around 0.9 K, and then increases. The temperature for the resistance minima is slightly different in the two samples, we do not have a simple explanation for that. The total amplitude of the variations is about 0.3% in sample 1 and 0.8% in sample 2. In both cases, the low-temperature saturation value of the resistance is *larger* than the normal-state resistance.

Figure 3 shows the same data for sample 2, together with the resistance of its Al wire and of the Co/Al junction. The Al wire becomes superconducting at 1.34 K, with a transition width of about 10 mK. The superconducting properties of the Al wire are not strongly depressed by the proximity of the Co interface.

We measured the junction resistance by injecting current from one side (I+) of the Co wire to one side of the Al wire (i-) and measuring the voltage drop between the opposite sides of the Co and Al wires (V- and v+). The small negative offset above 1.34 K, when the Al wire is normal, stems from a three-dimensional spreading of the current lines in the metallic electrodes of the junction. Such a sign reversal in a crossed-shaped junction only occurs when the resistance of the junction is significantly smaller than the electrodes resistances (here, 10  $\Omega$  and 0.4  $\Omega$ , respectively for



FIG. 4. Magnetoresistance of sample 1 Co wire and Al wire. The magnetic field is in the substrate plane, parallel to the Al wire. The Co resistance is hysteretic. The superconducting critical field of Al is close to 140 mT. Bias current 0.1  $\mu$ A.

Co and Al). This argument together with the measured resistance of 0.1  $\Omega$  at the lowest temperature when the Al wire is superconducting confirms that our junction is metallic. This order of magnitude is consistent with a transparency t of a few % after the relation  $R_t^{-1} = 2N(E_F)v_FSe^2t$ . We believe that the junction resistance peak below 1.34 K is related to charge-imbalance effects in the Al island, when the gap is small compared to the injected quasiparticle energy. The Co resistance change is not much larger than the Al junction one, but we stress that the Co resistance varies significantly below 0.5 K, whereas the junction resistance does not vary anymore. This clearly shows that the variation of Co resistance is not due to a current redistribution effect induced by variations of the junction resistance.<sup>14</sup>

The magnetoresistance of sample 1 and 2 was studied in magnetic fields applied perpendicular or parallel to the substrate. Figure 4 shows the magnetoresistance of sample 1 in parallel field. From the Al resistance, we measure a critical field of 140 mT for the Al wire. The Co wire has a small (less than 1% at 140 mT) and hysteretic magnetoresistance. No saturation is visible up to 170 mT. From this measurement, we can assert that our Co wire is ferromagnetic, but that all our experiments were performed in a regime where the Co magnetization is not saturated.

Figure 5 shows the magnetoresistance of the Co wire in perpendicular field, above the superconducting transition of Al at 1.5 K, and well below at 0.29 K. There is no magnetoresistance at 1.5 K, indicating that the Co magnetization is



FIG. 3. Temperature dependence of the resistance of the Al wire (right-hand scale), the Co wire and the Co/Al junction (left-hand scale) of sample 2. Bias current 0.1  $\mu$ A.



FIG. 5. Magnetoresistance of sample 1 at T=0.29 K and T=1.5 K in perpendicular field. Field scan from -200 to 200, then back to -200 mT. No periodic oscillations of the resistance were visible above the experimental noise. Bias current 0.1  $\mu$ A.



FIG. 6. Differential resistance of sample 2 measured in different conditions. Upper curve : T=32 mK, H=0. Middle curve : T=0.8 K, H=0. Lower curve : T=32 mK, H=130 mT (parallel to the substrate). The latter two curves have been offset by 0.5 and 1  $\Omega$ , respectively, for clarity.

in plane. The perpendicular negative magnetoresistance only appears when Al becomes superconducting, and is about eight times smaller than in parallel field. We searched for periodic oscillations of magnetoresistance as a function of perpendicular field. We achieved a resistance resolution better than  $10^{-5}$ , corresponding to  $10^{-3}e^2/h$ , by averaging over a large number of scans. The absence of Aharonov-Bohm oscillations at this level is a strong indication that the phase-breaking length in Co is smaller than 0.3  $\mu$ m. This casts some doubts on the possible observation of weaklocalization-like effects in ferromagnetic films.<sup>13</sup>

We measured the differential resistance as a function of the bias current. Figure 6 shows three such curves recorded in different conditions. The upper curve was recorded at the lowest temperature 32 mK. The differential resistance exhibits a peak at zero bias, a minimum in the 1.7  $\mu$ A range, and returns to the normal state value at high bias. This is strongly reminiscent of the re-entrance effect.<sup>5</sup> We estimate that Joule heating did not exceed 0.3 K in the range below 5  $\mu$ A, and we checked that the junction resistance did not vary in this bias range.

The lower two curves of Fig. 6 have been shifted for clarity. The middle curve was recorded at a temperature of 0.8 K. This curve looks very similar to the first one, with the remarkable exception that the zero-bias maximum is absent. In contrast, the high-bias features are unchanged. The lowest curve was recorded at very low temperature, in a parallel field of 130 mT, just below the Al critical field of 140 mT. This last curve shows a clear re-entrance peak of differential resistance below 1.0  $\mu$ A, but the high-bias peaks are no longer present.

Let us compare these results with the ones previously reported for Cu.<sup>5</sup> On increasing current, and thus voltage, we increase the energy of the electrons injected in the *F*-*S* mesoscopic sample. We can thus probe the energy dependence of the proximity-induced excess conductance. As in a "free

electron'' metal, we observe a resistance minimum both as a function of temperature and bias current. The differential resistance is maximum at zero temperature and voltage : this is the re-entrance effect for the metallic conductance of the normal metal.

The high-bias peaks are related to the Al superconducting gap and/or critical current. At  $T=32\,$  mK, the 130 mT magnetic field strongly depresses the Al gap, but does not affect the characteristic energies of the electrons injected in the F-S sample. Consequently, the re-entrance peak of the differential resistance at zero energy is still visible, but the high bias peaks disappear. On the other hand, at 0.8 K and zero field, most of the electrons have an energy above the characteristic energy of the re-entrance effect, but the Al gap is not yet depressed, so that only the zero bias maximum disappears. This is consistent with our picture of the proximity effect.

In the quasiclassical theory, the temperature of the resistance minimum is  $5 \epsilon_c / k_B$ . The temperature dependence data gives us a Thouless energy of 14  $\mu$ eV for sample 2. With our estimate of the diffusion coefficient in Co, this energy would give a characteristic length of 180 nm, much shorter than the total sample length of 2  $\mu$ m. A simple interpretation of this result is that the Co electrons reflected on the Al island keep their phase coherence only on this shorter length scale. The effective mesoscopic sample length we are probing is only 180 nm. It is also the order of magnitude of magnetic domain sizes for Co samples deposited in similar conditions.<sup>15</sup> Indeed, it has been suggested that domain walls could contribute to decoherence of electrons.<sup>16</sup>

This decay length is much larger than the "exchange length":  $L_{\text{exch}} = \sqrt{\hbar D/k_B T_{\text{Curie}}} = 2$  nm in the dirty limit  $(L_{\text{exch}} > l_p)$ , with  $T_{\text{Curie}} = 1388$  K being the Curie temperature of Co. This length scale arises from the magnetic energy splitting between the incident electron and the Andreev reflected hole in the exchange field of F.<sup>17,18</sup> In the Andreev reflection process, the electron spin is reversed. In consequence, the reflected hole has a different energy and momentum than the incident electron. This results in a finite decay length  $L_{\text{exch}}$ .

If we take sample 2 normal state resistance (98.35  $\Omega$ ) to convert the current bias into a voltage corresponding to the minimum differential resistance, we get 170  $\mu$ eV. This is about 2.4 times larger than the thermal energy  $k_BT$  at the resistance minimum in Fig. 2, and even larger than the Al gap. This confirms that the coherence effects only occur on a length scale shorter than the total wire length. In comparison, 180 nm of our Co wire would have a resistance of about 8.9  $\Omega$ . At the current bias 1.7  $\mu$ A of the minimum differential resistance, the voltage drop along this 180 nm coherence length is 15  $\mu$ V. This latter value is close to the Thouless energy derived from Fig. 2.

If we want to carry out a thorough quantitative analysis of our results, we encounter several difficulties. (i) A part of the resistance drop below the Al superconducting transition should originate from the local short circuit by the Al island. A complete description would require taking into account the current redistribution in the Co thickness beneath the Al wire. (ii) The amplitude of the resistance drop is relatively small in comparison with the expected 15% variation for the resistance of the regions affected by the proximity effect. This could be related to the fact that we do not have good

R11 875

reservoirs injecting at a given energy from a well-defined distance, but diffusively distributed phase breaking and inelastic events along the F wire.

Let us discuss the possible origin of the excess resistance, above the normal state residual resistance, at low temperature. This result is in agreement with the theoretical prediction of Stoof and Nazarov, that the zero-temperature and zero-voltage resistance of a *N*-*S* structure should exceed the normal-state resistance if repulsive e-e interactions are present. Following this viewpoint, our experiment would reflect a direct influence of the e-e interaction on a metallic resistance. From our data, we could extract a value for the electron-electron interaction parameter. Another quite different explanation for the excess resistance at low temperature could be the screening of magnetic field by the superconductor in contact with the ferromagnet. The modification of the magnetic domain configuration near the *F*-*S* interface<sup>19</sup> could enhance the resistivity in this region.

In conclusion, we have observed a proximity effect on the dissipative transport in a ferromagnetic metal in contact with a superconductor. Our results are in agreement with the early works of Petrashov<sup>12</sup> and Lawrence and Giordano.<sup>13</sup> From this work, we can assert that the behavior described in Ref. 13 is due to a superconducting proximity effect in the ferromagnetic metal. The energy dependence of the effect has been probed through the temperature and voltage dependence of the resistance. The decay length for the coherence effect appears to be about 180 nm in the Co film. This value is of the order of the expected size of the magnetic domains in such films. The excess resistance, above the normal-state value, at zero voltage and temperature, could be explained by interaction effects.

We thank B. Spivak, P. Butaud, and J. Caulet for stimulating discussions, D. Lafont, D. Mariolle (CEA-LETI) and Th. Fournier for the SEM micrograph of the samples, and J. Gilchrist for proofreading the manuscript.

- <sup>1</sup>P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1964).
- <sup>2</sup>V. T. Petrashov, V. N. Antonov, P. Delsing, and T. Claeson, Phys. Rev. Lett. **70**, 347 (1993); **74**, 5268 (1995).
- <sup>3</sup>A. Dimoulas, J. P. Heida, B. J. van Wees, T. M. Klapwijk, W. v. d. Graaf, and G. Borghs, Phys. Rev. Lett. **74**, 602 (1995).
- <sup>4</sup>H. Courtois, Ph. Gandit, D. Mailly, and B. Pannetier, Phys. Rev. Lett. **76**, 130 (1996).
- <sup>5</sup> P. Charlat, H. Courtois, Ph. Gandit, D. Mailly, A. Volkov, and B. Pannetier, Phys. Rev. Lett. **77**, 4950 (1996); Czech. J. Phys. **46**, 3107 (1996).
- <sup>6</sup>S. N. Artemenko, A. F. Volkov, and A. V. Zaitsev, Solid State Commun. **30**, 771 (1979).
- <sup>7</sup>T. H. Stoof and Y. Nazarov, Phys. Rev. Lett. **76**, 823 (1996).
- <sup>8</sup>A. A. Golubov, F. K. Wilhelm, and A. D. Zaikin, Phys. Rev. B **55**, 1123 (1997).
- <sup>9</sup>F. Zhou, B. Spivak, and A. Zyuzin, Phys. Rev. B 52, 4467 (1995).
- <sup>10</sup>R. F. Hoyt and A. C. Mota, Solid State Commun. **18**, 139 (1976).

- <sup>11</sup>B. Spivak and A. Zyuzin, cond-mat/9602005, Phys. Usp. (to be published).
- <sup>12</sup> V. T. Petrashov, V. N. Antonov, S. V. Maksimov, and S. R. Shaikhadarov, JETP Lett. **59**, 551 (1994).
- <sup>13</sup>M. D. Lawrence and N. Giordano, J. Phys.: Condens. Matter 8, L563 (1996).
- <sup>14</sup>F. K. Wilhelm, A. D. Zaikin, and H. Courtois, Phys. Rev. Lett. 80, 4289 (1998).
- <sup>15</sup>W. Wernsdorfer, K. Hasselbach, D. Mailly, B. Barbara, A. Benoit, L. Thomas, and G. Suran, J. Magn. Magn. Mater. **145**, 33 (1995).
- <sup>16</sup>G. Tatara and H. Fukuyama, Phys. Rev. Lett. 78, 3773 (1997).
- <sup>17</sup>E. A. Demler, G. B. Arnold, and M. R. Beasley, Phys. Rev. B 55, 15 174 (1997).
- <sup>18</sup>M. J. M. de Jong and C. W. J. Beenakker, Phys. Rev. Lett. 74, 1657 (1995).
- <sup>19</sup>A. I. Buzdin and L. N. Bulaevski, Sov. Phys. JETP **67**, 576 (1988).