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Long range supercurrents in ferromagnetic CrO_2 using a multilayer contact structure

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We report measurements of long ranged supercurrents through ferromagnetic and fully spin-polarized CrO_2 deposited on TiO_2 substrates. In earlier work, we found supercurrents in films grown on sapphire but not on TiO_2. Here, we employed a special contact arrangement, consisting of a Ni/Cu sandwich between the film and the superconducting amorphous $Mo_{70}Ge_{30}$ electrodes. The distance between the contacts was almost a micrometer, and we find the critical current density to be significantly higher than found in the films deposited on sapphire. We argue this is due to spin mixing in the Ni/Cu/CrO_2 layer structure, which is helpful in the generation of the odd-frequency spin triplet correlations needed to carry the supercurrent. © *2012 American Institute of Physics*. [doi:10.1063/1.3681138]

Conventional spin-singlet Cooper pairs from a superconductor (S) dephase over a coherence length $\xi_F = \sqrt{\hbar D_F/h_{ex}}$ (dirty limit) in a ferromagnet (F) under the influence of its exchange field h_{ex} (and D_F the diffusion constant in the F-metal). Even for weak ferromagnets, ξ_F is only a few nm. Such dephasing would not occur with equal-spin triplet Cooper pairs, leading to a long range proximity (LRP) effect in the ferromagnet. It was predicted that triplet correlations can be induced at an S/F interface when h_{ex} is inhomogeneous,[1–3] for instance from domain walls or unaligned magnetic moments. This should also allow a Josephson current in an S/F/S geometry. To observe this, both interfaces are required to show similar inhomogeneities[4] as for instance in an S/F_1/F/F_2/S trilayer in which the magnetizations of the $F_{1,2}$ layers are non-collinear with the central F layer.

Early work on CrO_2 (Ref. 5) and Holmium[6] gave the first indications for such LRP effects in ferromagnets. In the first case, a supercurrent was measured in devices where superconducting electrodes of NbTiN with separations up to 1 μm were placed on unstructured 100 nm thick films of CrO_2 (a half metallic ferromagnet or HMF) which were grown on TiO_2 substrates. In the second case, the LRP effect was observed in ferromagnetic Ho wires of lengths up to 150 nm using an Andreev interferometer geometry. More recently, LRP effect were reported using Josephson junctions where a Co central layer was used in combination with PdNi, CuNi, or Ni layers;[7,8] and where a Co layer was used together with Ho layers to provide magnetic inhomogeneity.[9] Signatures of LRP effect were also observed with the Heusler Compound Cu_2MnAl (Ref. 10) and in Co nanowires.[11] At the same time, the observation of supercurrents over a length of 700 nm through CrO_2 deposited on sapphire substrates was reported.[12,13]

The experiments with Co junctions were up to Co thicknesses of 50 nm. Since Co is not fully spin polarized, the triplet decay is mainly set by the spin diffusion length and can be expected to be of the order of 100 nm. That makes the CrO_2 case with its significantly larger decay length of special interest, but in the previous experiments, the reproducibility was an issue. In particular, it was not clear where the inhomogeneous magnetization resides which is needed for the triplet generation. Also, in our previous work, we did not succeed in finding supercurrents in films deposited on TiO_2. Here, we report on observing long ranged supercurrents in CrO_2 grown on TiO_2, using 2 nm Ni as an extra layer in the contact geometry to induce an artificial magnetic inhomogeneity and 5 nm Cu to magnetically decouple the Ni and the CrO_2. We find much stronger supercurrents than in the case of sapphire, indicating that with the Ni/Cu sandwich we have a good generator for triplet Cooper pairs.

The devices were fabricated in a lateral geometry using 60 nm thick a-$Mo_{70}Ge_{30}$ superconducting contacts (transition temperature $T_c = 6$ K) deposited on unstructured 100 nm thick CrO_2 films grown on TiO_2 substrates. We made the devices through a lift-off mask using a bilayer resist. Ar-ion etching to remove Cr_2O_3 on the film surface was applied immediately prior to deposition, and the Cu/Ni/$Mo_{70}Ge_{30}$ sandwiches were sputtered *in situ*. Two junctions were made on each sample, perpendicular to each other, and both junctions were measured independently. More details are found in Refs. 12 and 13.

A supercurrent was measured in three devices out of five, named A_T, B_T, and C_T. On A_T (30 μm wide leads) both junctions showed a supercurrent. We call them A_T-a (600 nm gap) and A_T-b (800 nm gap). Samples B_T and C_T were prepared with 5 μm wide leads, in order to lower the absolute value of the currents, and a gap of 700 nm. Here, only one junction was showing a measurable critical current on each sample. Sample C_T was measured twice, in two different cryostats, one with extra filtering to minimize to amplifier contribution to the data in the zero-voltage branch. A drawback still is the limited lifetime of the samples. The supercurrent disappears

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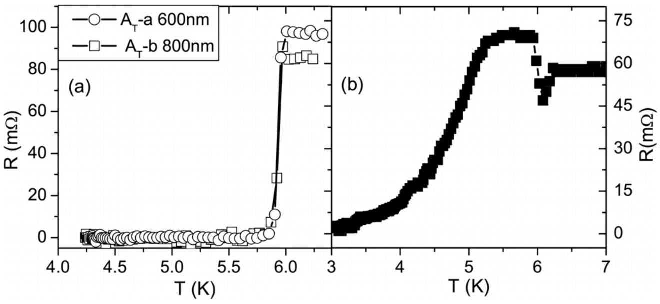

FIG. 1. Resistance R vs. Temperature T, (a) for junctions A_T-a (gap 600 nm; electrode width 30 μm) and A_T-b (gap 800 nm); (b) for B_T (gap 700 nm, electrode width 5 μm).

after a few cool-downs, possibly due to the effect of thermal cycling on the films.

For sample A_T, the resistance R as function of temperature T is given in Fig. 1(a) and shows a sharp down-jump at T_c. For junction B_T (Fig. 1(b)), $R(T)$ shows a small dip at 6 K, followed by an up-jump, a flat part, and then a slow decrease. For junction C_T, the behavior is similar but with a larger up-jump to 0.7 Ω, similar to our sapphire-based devices.[13]

Figure 2(a) shows an I-V characteristic for sample A_T-b, measured at 4.2 K. There is a zero-resistance branch up to a well-defined current of about 3 mA at which a finite voltage develops. On larger scales a bend in the curve is seen, followed by another transition at 15 mA to Ohmic behavior with $R_N = 100$ mΩ. Figure 2(b) shows I-V data measured on sample B_T at 3 K. The value for I_c is 1.2 mA, and the resistive branch has a value of 80 mΩ in very reasonable agreement with the normal state resistance. The residual resistance below I_c is a few mΩ. Sample C_T was first measured at 4.2 K in a cryostat with well-filtered leads. Here, the I-V characteristic showed sharp switching and some hysteretic behavior, with I_c of the order of 0.5 mA. The residual resistance below I_c is 3 mΩ. $I_c(T)$ was defined by a 1 μV criterion and measured for junction B_T and C_T in the temperature range of 2.5 K to 6 K. As shown in Fig. 3 for sample B_T, the behavior is almost linear. For sample A_T, we first measured the field dependence of I_c at 4.2 K, but we did not measure $I_c(T)$ because the supercurrent disappeared after the third cool-down. The measurement on sample C_T is also shown in Fig. 3. In a subsequent measurement, I_c had gone down to 70 μA, illustrating the fragility of the sample, but $I_c(T)$ also showed a linear increase. Figure 4(a) illustrates the effect of a magnetic field H_a on I_c at 4.2 K for both junctions A_T-a and A_T-b, with H_a in the plane of the junction and ⊥ I. It shows that I_c in the case of A_T-a is quite sensitive to H_a, with an initial fast decrease below 60 mT, but less so in the case of A_T-b. Figure 4(b) presents $I_c(H_a)$ at 3 K for junction B_T in three different configurations, H_a in-plane and $\parallel$, $\perp I$, and H_a out-of-plane. Here, the field-in-plane data show a relatively slow decrease, while the field-out-of-plane data show a small sharp peak, followed by a shoulder around 100 mT. Neither for A_T-a,b nor for B_T there is evidence for a Fraunhofer pattern.

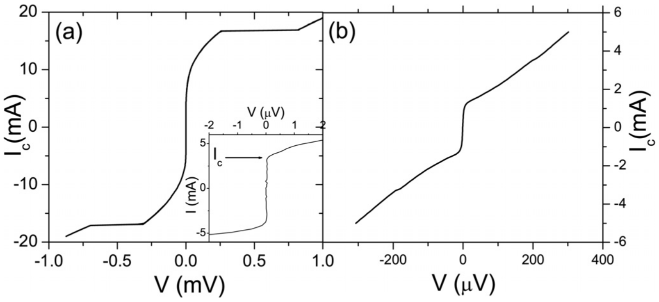

FIG. 2. Current I versus voltage V measured (a) for junction A_T-b at 4.2 K and (b) for junction B_T at 3 K. The inset show a blowup of the zero-resistance branch. The critical current at 3 mA is indicated with an arrow.

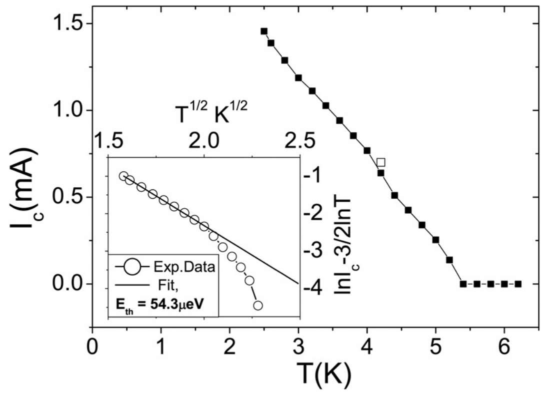

FIG. 3. $I_c(T)$ for junction B_T. The open symbol is I_c at 4.2 K for junction C_T, as follows from Fig. 2 inset: plot of $(\ln(I_c) - (3/2\,\ln(T))$ versus $\sqrt{T}$ to determine the Thouless energy E_{th}.

The claim from the measurements is that large supercurrents are now flowing through the CrO_2 bridge. In discussing these results, we address the following issues. We compare the residual resistance in the supercurrent measurements with the normal state resistance of the bridge; we discuss the possibility of depairing currents in the superconducting leads; a Thouless analysis is performed; and we discuss the effects of applying a magnetic field. The I_cs measured here can be compared with our previous measurements[12,13] on sapphire-based junctions. The current density at 4.2 K, ($d_{CrO_2} \approx 100$ nm, junction width 30 μm and 5 μm, current ≈ 3 mA and 0.5 mA, respectively) is of the order of 1×10^9 A/m^2 for A_T, B_T, as well as C_T. In all cases, it is 100 times larger than that of sapphire-based junctions and of similar magnitude as in the earlier observations of Keizer *et al.*[5] This suggests that a uniform spin active interface is present at the interface, due to the additional 2 nm Ni layer.

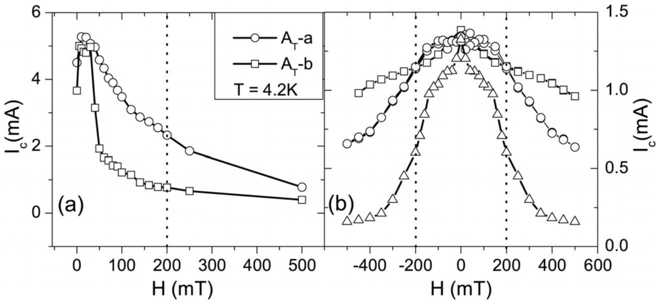

FIG. 4. Critical current I_c versus applied field (H_a) (a) at 4.2 K for junctions A_T-a (◯) and A_T-b (□) with H_a in-plane and ⊥ current I; (b) for junction B_T at 3 K in three different configurations, in-plane $H_a \parallel I$ (□), $H_a \perp I$ (◯), and out-of-plane $H_a \perp I$ (△). The vertical dotted lines indicate the field at 200 mT for reference purposes.

An important question is whether the $I-V$ characteristics such as shown in Fig. 2 are truly from the CrO_2 bridge and not just the superconducting contacts. For this, we take another look at the normal resistance of the bridge. Taking $\rho_{CrO_2} = 10\,\mu\Omega$cm, a film thickness of 100 nm, a bridge width of 5 μm, and a junction length of 700 nm, R_N comes out to be 140 mΩ (25 mΩ for the 30 μm wide contacts). This is significantly higher than what is measured in the zero-voltage branch of the I-V characteristics, where it is not more than a few mΩ. Note that the measured resistance above T_c is higher than the above estimate. This is because, when the superconducting leads become normal, the geometry of the sample is a very different one, with both high resistance MoGe and low-resistance CrO_2 contributing.

Another issue is how close I_c comes to the depairing current I_{dp} of the superconducting leads. For the sapphire-based junctions with their low I_c values, this was not relevant. For a-MoGe, J_{dp} is about 4×10^{10} A/m^2 at 4.2 K (Ref. 14). The thickness of the lead (40 nm) is smaller than that of the bridge, so its current density at the measured I_c for all junctions is about 2.5×10^9 A/m^2, still an order of magnitude smaller than J_{dp}. This probably explains, however, the second transition seen in Fig. 2(a), which takes place at a 5 times higher current density.

A further point is whether the value of the critical current is physically reasonable. This we estimate from the Thouless energy E_{Th} of the junction, which is a measure for its dephasing characteristics. For diffusive junctions, I_c is proportional to $T^{3/2}$ exp $\sqrt{2\pi k_B T/E_{Th}}$ (and $E_{Th} = (\hbar D_F)/L^2$, with L the length of the junction). We plot (ln(I_c)-(3/2 ln(T)) versus $\sqrt{T}$ (inset of Fig. 3). For junction B_T, we find $E_{th} = 54\,\mu$eV, similar to that of sapphire based junctions.[12,13] From E_{Th} we can estimate I_c at 4.2 K using theoretical results for a long junction.[15] For $(k_B T/E_{Th}) \approx 7.6$, we find from Ref. 15 that $I_c R_N \approx E_{Th} \approx 54\,\mu$V, which with $R_N = 60$ mΩ leads to $I_c = 0.9$ mA, quite close to the measured value.

The magnetic field effects are complicated. For $H_a \parallel I$, the junctions A_T-a,b are more sensitive to the field than B_T. For A_T-a,b, the first sharp decrease at 60 mT might correspond to the first flux quantum, which is a reasonable value according to the dimensions of the junctions, but no such behavior is seen for B_T. The suppression of I_c is stronger than in the earlier work. Taking 200 mT as reference, the suppression is over 70% for A_T, and still almost 30% for B_T, compared to 10% in the sapphire-based junctions. This points to a diminishing effectiveness of the Ni/Cu layer, although it might be argued that the effect should be even stronger: in 200 mT, both the CrO_2 and Ni magnetization should be saturated and aligned, removing a possible source of magnetic inhomogeneity. Instead, the supercurrents were not even quenched in 500 mT. It suggests that there is a residual magnetic inhomogeneity residing in the Ni/Cu/CrO_2 sandwich, which is not removed by the magnetic field. In conclusion, a Ni/Cu sandwich on top of ferromagnetic CrO_2 deposited on TiO_2 substrates leads to strong supercurrents over a distance of almost 1 μm. The Ni/Cu sandwich appears to furnish spin mixing and triplet generation similar to what was found in Co-based junctions.

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