Observation of Spin-Triplet Superconductivity in Co-Based Josephson Junctions

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We have measured a long-range supercurrent in Josephson junctions containing Co (a strong ferromagnetic material) when we insert thin layers of either PdNi or CuNi weakly ferromagnetic alloys between the Co and the two superconducting Nb electrodes. The critical current in such junctions hardly decays for Co thicknesses in the range of 12–28 nm, whereas it decays very steeply in similar junctions without the alloy layers. The long-range supercurrent is controllable by the thickness of the alloy layer, reaching a maximum for a thickness of a few nm. These experimental observations provide strong evidence for induced spin-triplet pair correlations, which have been predicted to occur in superconducting-ferromagnetic hybrid systems in the presence of certain types of magnetic inhomogeneity.

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When a conventional spin-singlet superconductor is brought into contact with a normal metal, superconducting pair correlations penetrate into the normal metal over distances as large as a micron at low temperature, creating the superconducting proximity effect [1]. If the normal metal is replaced by a ferromagnet, the pair correlations penetrate only a few nanometers, as the exchange field in the ferromagnet leads to a rapid loss of phase coherence between electrons with opposite-pointing spins [2,3]. This limitation would not arise if the Cooper pairs in the superconductor had spin-triplet symmetry, which occurs only rarely in nature [4,5]. It was predicted several years ago that spin-triplet superconducting correlations could be induced at the interface between a conventional spin-singlet superconductor and a ferromagnet with inhomogeneous magnetization [6,7]. Moreover, these pair correlations are in a new symmetry class: they have even relative orbital angular momentum but are odd in frequency or time [8]. A promising hint of spin-triplet correlations in half-metallic CrO_2 was reported in 2006 by Keizer *et al.* [9]; however, there has been no confirmation of that result in the intervening time. Here we present strong evidence for spin-triplet pair correlations in Josephson junctions fabricated from common metals: Nb and Co. The magnetic inhomogeneity is supplied by thin layers of a weakly ferromagnetic alloy—either PdNi or CuNi—inserted between the Co and Nb layers. As the Co thickness is increased, the maximum supercurrent in the Josephson junctions decays very slowly—in sharp contrast to the very fast decay observed in similar junctions without these alloy layers [10]. The strength of the triplet correlations can be controlled by the thickness of the alloy layer, reaching its maximum for a thickness of a few nm.

A schematic diagram of our Josephson junction samples is shown in Fig. 1(a). The entire multilayer structure up through the top Au layer is sputtered onto a Si substrate in a single run, without breaking vacuum between subsequent layers. The multilayers are subsequently patterned into circular pillars using photolithography and Ar ion milling, after which the SiO_x insulating layer is thermally evaporated to isolate the top Nb contact from the base. Finally, the top Nb contact is sputtered through a mechanical mask. The Au layer is fully superconducting due to the proximity effect with the surrounding Nb layers. The Nb superconducting layers have critical temperature near 9 K, which allows us to measure the Josephson critical supercurrent at 4.2 K with the samples dipped in liquid helium. Details of our fabrication and measurement procedures are given in our previous publications [10,11].

The detailed sequence of internal layers [labeled F for "ferromagnetic" in Fig. 1(a)] is shown in Fig. 1(b). The

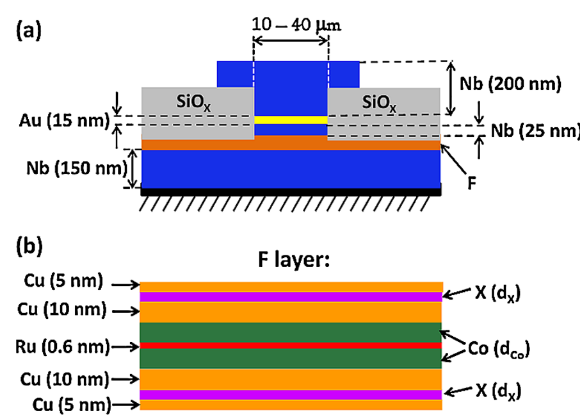

FIG. 1 (color online). (a) Schematic diagram of the Josephson junction samples, shown in cross-section. (b) Detailed sequence of the metal layers inside the Josephson junctions (labeled F in a). The layers labeled X are either PdNi or CuNi alloy. The functions of the various layers are described in the text. Only the thicknesses of the Co and X layers are varied in this work. The Cu buffer layers play no active role in the devices, but are important to isolate the X layers magnetically from the Co layers.

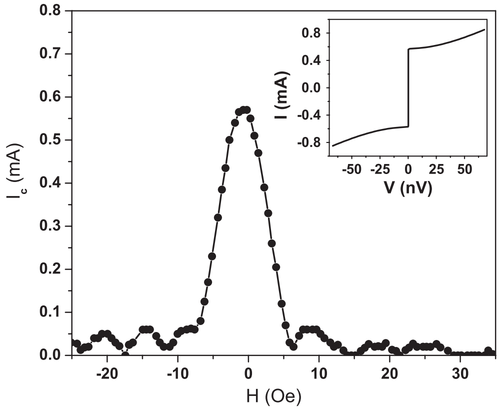

FIG. 2. Critical current (I_c) vs applied magnetic field (H) for a 10 μm diameter Josephson junction with $d_{\mathrm{Co}} = 13$ nm and $d_{\mathrm{PdNi}} = 4$ nm, measured at $T = 4.2$ K. The excellent "Fraunhofer pattern" results from cancellation of the intrinsic magnetic flux in the junction, due to antiparallel exchange coupling of the two Co layers via the thin Ru layer. (The lines are guides to the eye.) The inset shows the current-voltage (I-V) characteristic of the junction at $H = 0$.

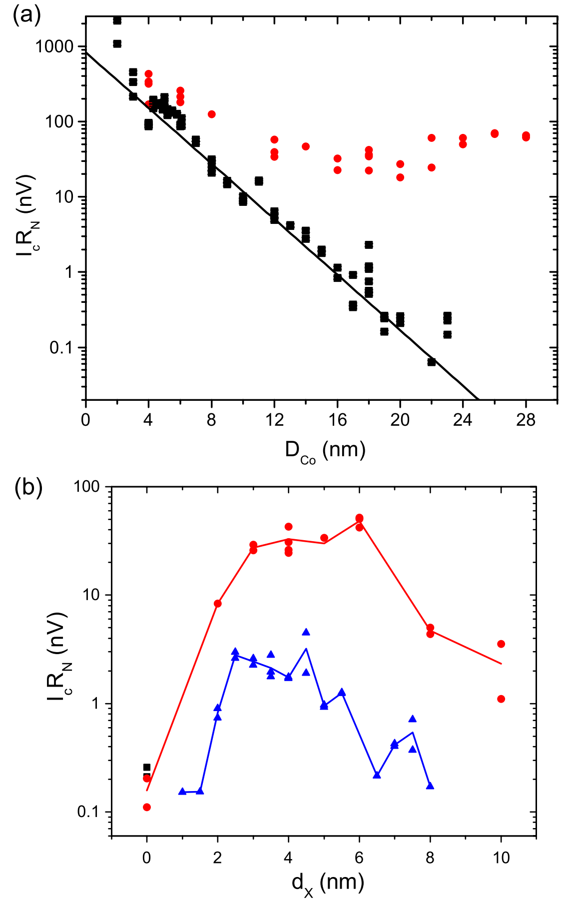

FIG. 3 (color online). (a) Product of critical current times normal state resistance, I_cR_N, as a function of total Co thickness, $D_{\mathrm{Co}} = 2d_{\mathrm{Co}}$. Red circles represent junctions with $X =$ PdNi and $d_{\mathrm{PdNi}} = 4$ nm, whereas black squares represent junctions with no X layer (taken from Ref. [10]). As D_{Co} increases above 12 nm, I_cR_N hardly drops in samples with PdNi, but drops very rapidly in samples without. (The solid line is a fit of the data without PdNi to a decaying exponential, also from Ref. [10]. In [10], data from multiple junctions with the same value of D_{Co} were represented by a single data point with an error bar; here, each device is represented by its own data point.) (b) I_cR_N product as a function of d_X for two series of junctions with fixed $D_{\mathrm{Co}} =$ 20 nm. Red circles: $X =$ PdNi; blue triangles: $X =$ CuNi. (The two squares at $d_X = 0$ are taken from Ref. [10].) In both cases, I_cR_N first increases, then eventually decreases with increasing d_X. Lines are guides to the eye.

purpose of the ferromagnetic Co is to suppress the conventional spin-singlet Josephson supercurrent. As explained in more detail in Ref. [10], we have inserted a thin Ru layer in the center of the Co layer, which induces antiparallel exchange coupling between the domains in the two Co layers [12], leaving nearly zero net magnetization in the junctions. As a result, the critical current vs applied magnetic field data exhibit nearly ideal "Fraunhofer patterns", as shown in Fig. 2. These patterns give us reliable measurements of the maximum critical current in each sample, while also indicating that the current flow in the junctions is uniform and that there are no shorts in the surrounding SiO insulator. (Without the Ru layers, the Fraunhofer patterns of Josephson junctions similar to the ones studied here are random, and the critical currents are very small [10].) The layers labeled X represent either $\mathrm{Pd}_{0.88}\mathrm{Ni}_{0.12}$ or $\mathrm{Cu}_{0.48}\mathrm{Ni}_{0.52}$ ferromagnetic alloys. The Cu layers between the X layers and the Co layers serve two purposes. First, they isolate the X and Co layers magnetically, so the magnetization of the X layers is not exchange coupled to that of the Co layers. Second, we have found in our earlier work that the quality of our sputtered Co is higher when sputtered on Cu than on some other materials (Nb in [10]).

We discuss first the case where $X = \mathrm{Pd}_{0.88}\mathrm{Ni}_{0.12}$, a weakly ferromagnetic alloy with a Curie temperature of 175 K [11]. Figure 3(a) shows the product of critical current and normal state resistance, I_cR_N, vs total cobalt thickness, $D_{\mathrm{Co}} \equiv 2d_{\mathrm{Co}}$, for a series of samples with fixed PdNi layer thickness, $d_{\mathrm{PdNi}} = 4$ nm. (The normal state resistance, R_N, is determined from the inverse slope of the I-V curve for $I \gg I_c$.) There is no discernible decay of I_cR_N for $D_{\mathrm{Co}} > 12$ nm. For comparison, Fig. 3(a) also shows data from Ref. [10] for junctions not containing PdNi. In those samples I_cR_N decays very rapidly with increasing D_{Co}, with a decay constant of 2.34 ± 0.08 nm [10]. When $D_{\mathrm{Co}} = 20$ nm, I_cR_N is over 100 times larger in the samples with PdNi than in the samples without PdNi. The long-range character of the Josephson current in samples with PdNi represents strong evidence for its spin-triplet nature.

The subtle role of the X layers in enhancing the supercurrent is illustrated in Fig. 3(b), which shows I_cR_N vs d_X with $X =$ PdNi or CuNi for two sets of samples with D_{Co} fixed at 20 nm. Without any X layer, I_cR_N is very small, consistent with the data shown in Fig. 3(a). When the X

layer reaches a critical thickness, I_cR_N increases rapidly, reaches a maximum for d_X values of a few nm, then decreases at larger values of d_X. The decrease in I_cR_N at large d_X signals the destruction of the spin-triplet correlations, perhaps due to spin memory loss in the bulk of the X layers. The spin memory lengths in these PdNi and CuNi alloys are very short—about 2.8 nm in PdNi [13] and 1.4 nm in CuNi [14]. This would explain why we found no evidence for spin-triplet supercurrent in our previous measurements of Josephson junctions containing only PdNi layers of thickness 30–100 nm [11]. Evidently, a thin PdNi or CuNi layer is essential to produce spin-triplet correlations, whereas a thick layer suppresses them.

Why don't samples without X layers exhibit spin-triplet supercurrent? According to theory [6–8], spin-triplet correlations are generated if the Cooper pairs from the superconductor experience regions of noncollinear magnetization within their coherence length, ξ_S. Scanning electron microscopy with polarization analysis (SEMPA) measurements on Co films grown under similar conditions as ours reveal magnetic domains with typical sizes of a few microns, but with the magnetization directions of neighboring domains largely antiparallel [15]. Noncollinear magnetization resides only in the domain walls, which is apparently not enough to produce a significant amount of spin triplet.

We discuss two possible sources of noncollinear magnetization in our samples. (i) If the Cooper pairs experience noncollinear magnetization between adjacent X-layer domains, then domain size and out-of-plane magnetocrystalline anisotropy are likely to be key ingredients. While the domain size of PdNi is not known, the domain size in $Cu_{0.47}Ni_{0.53}$ has recently been measured to be about 100 nm [16], which is not so different from the Nb coherence length $\xi_S = 14$ nm. Competition between out-of-plane magnetocrystalline anisotropy and the in-plane shape anisotropy of thin films can lead to stripe domains with canted magnetization [17] and thus to noncollinear magnetizations in neighboring domains. Both PdNi [11] and CuNi [18] are known to have out-of-plane magnetic anisotropy. (ii) If the Cooper pairs experience noncollinear magnetization between the X layers and the Co layers, then almost any ferromagnetic layer could work in place of the X layers, as long as the magnetization directions of the X and Co layers are independent. In this scenario, our samples can be viewed as realizations of the $S/F'/F/F''/S$ junctions studied theoretically by Houzet and Buzdin [19], but with Cu buffer layers separating the three ferromagnetic layers. The advantage of using weakly-ferromagnetic PdNi and CuNi alloys for the F' and F'' layers in this case is simply to preserve good Fraunhofer patterns [11], which would be destroyed by using strong ferromagnetic layers instead. We stress that the Cu buffer layers are essential in both scenarios. We have tried omitting the Cu buffer layers between the X and Co layers for $X =$ PdNi, and found that the supercurrent is much smaller than in samples with Cu buffer layers. Presumably the magnetization of the PdNi domains is forced by exchange coupling to lie parallel to that of the Co, which is detrimental to production of the triplet correlations by both sources (i) and (ii).

Aside from the presence of noncollinear magnetizations, theory suggests that any "spin-active" interface between a superconductor and a ferromagnet can produce spin-triplet correlations [20]. We have tried using $X = Cu_{0.94}Pt_{0.06}$, an alloy with strong spin-orbit scattering, but preliminary data show very little, if any, signature of the triplet.

Comparison of our results with theory is problematic. The magnitude of the spin-triplet supercurrent depends on the details of the PdNi or CuNi domain structure, while theoretical calculations exist only for idealized magnetic configurations. More useful is a discussion of the decay lengths of the spin-singlet and spin-triplet supercurrents. In the "dirty" limit, where the mean free path, l_e, is the shortest relevant length scale in the problem, the spin-singlet supercurrent should decay on the length scale $\xi_F = \sqrt{\hbar D_F/E_{\mathrm{ex}}}$, where D_F and E_{ex} are the electron diffusion constant and exchange energy in the ferromagnet. Josephson junctions containing Co, however, are in the "intermediate" limit, with l_e longer than ξ_F, but shorter than ξ_S, the superconducting coherence length. In that limit, the spin-singlet supercurrent decays on the length scale l_e, which is estimated to be 2.4–3.0 nm from previous studies [10,21]. Spin-triplet supercurrent, in contrast, should decay over a much longer length scale given by the smaller of the normal metal coherence length, $\xi_N = \sqrt{\hbar D_F/2\pi k_B T}$, or the spin memory length, $L_{\mathrm{sf}} = \sqrt{D_F \tau_{\mathrm{sf}}}$, where τ_{sf} is the mean time between spin-flip or spin-orbit scattering events. Estimation of D_F for Co is difficult due to its strong ferromagnetism and to the widely varying densities of states and Fermi velocities of the different bands. From our measured Co resistivity, the Einstein relation, and the densities of states of majority and minority electrons at the Fermi surface [22], we estimate $D_F = 5 \times 10^{-3}$ m^2/s and 5×10^{-4} m^2/s for the majority and minority electrons, respectively, which give $\xi_N = 40$ nm and 10 nm at $T = 4.2$ K. L_{sf} in Co has been measured to be about 60 nm, also with large uncertainty [23,24]. Unfortunately, sample-to-sample fluctuations in the experimental data in Fig. 3(a) mask any discernible decay for D_{Co} between 12 and 28 nm. We have fabricated and measured some samples with larger D_{Co}, but the Fraunhofer patterns are poor, most likely due to less effective antiparallel coupling of the Co layers.

The spin-triplet pair correlations observed here and discussed in Ref. [8] are quite different from those believed to occur in materials such as Sr_2RuO_4 [4]. The Cooper pairs in the latter satisfy the spin-statistics theorem of quantum mechanics by having odd relative orbital angular momentum (p wave). According to theory [8], the triplet pair correlations induced in superconductor/ferromagnet hybrid systems have even relative orbital angular momentum; in particular, they can be *s* wave, which implies that they are robust in the presence of disorder. Quantum mechanics is not violated because the correlations are odd in frequency, or equivalently odd under time reversal. This idea, first proposed in a model for liquid helium-3 by Berezinskii [25], is counterintuitive, as it implies that the equal-time pair correlation function vanishes. Further experiments will be required to confirm unambiguously the unusual symmetry of the pair correlations in our samples [26].

Looking back, there were hints of long-range proximity effects in superconducting-ferromagnetic hybrid systems as early as 10 years ago [27–30], but there was no way to control the observed effects. More recently, Sosnin *et al.* [31] observed phase-coherent oscillations in the resistance of a Ho wire connected to two superconductors, but the authors did not observe a Josephson supercurrent, nor did they comment on its absence. The observation by Keizer *et al.* [9] of a supercurrent through CrO_2 was an exciting advance, but the critical currents in those samples varied by 2 orders of magnitude in similar samples. We anticipate that our results, which exhibit systematic dependence of the spin-triplet supercurrent on PdNi or CuNi thickness, will pave the way to many new experiments [32,33].

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Note added in proof: A theoretical paper closely related to our experiment has appeared very recently [34].

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