Observation of spin-triplet superconductivity in Co-based Josephson Junctions

Trupti S. Khaire, Mazin A. Khasawneh, W. P. Pratt, Jr., Norman O. Birge

Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824-2320, USA
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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 Figure 1a. 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$_2$ 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].

![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 (labelled F in a). The layers labelled 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.](http://example.com/figure1.png)
The layers labelled X represent either Pd$_{0.88}$Ni$_{0.12}$ or Cu$_{0.49}$Ni$_{0.52}$ ferromagnetic alloys. The purpose of the Co in the central Co/Ru/Co trilayer is to suppress the conventional spin-singlet Josephson supercurrent. The critical current of similar Josephson junctions containing Co/Ru/Co trilayers, but without the X layers, decays exponentially with increasing Co thickness with a decay constant of 2.34±0.08 nm [10]. The thin Ru layer induces antiparallel exchange coupling between the domains in the two Co layers [12], to produce nearly zero total magnetic flux in the junctions. As a result, the critical current vs. applied magnetic field data of these Josephson junctions exhibit nearly-ideal “Fraunhofer patterns,” as shown in Figure 2. These patterns give us reliable measurements of the maximum possible 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.

We discuss first the case where X = Pd$_{0.88}$Ni$_{0.12}$, a weakly-ferromagnetic alloy with a Curie temperature of 175 K [11]. Figure 3a shows the product of critical current times normal state resistance, $I_cR_N$, as a function of total Co thickness, $D_{Co} = 2d_{Co}$. Red circles represent junctions with X = PdNi and $d_{PdNi} = 4$ 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].) Error bars represent the standard deviation determined from measurements of two or more pillars on the same substrate, or are set to 10% in the few cases where data from only a single pillar were available. $I_cR_N$ product as a function of $d_{X}$ for two series of junctions with fixed $D_{Co} = 20$ nm. Red circles: X = PdNi; blue triangles: X = CuNi. (The square at $d_X = 0$ is 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.

The subtle role of the X layers in enhancing the supercurrent is illustrated in Figure 3b, 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
Figure 3a. When the X layer reaches a critical thickness, $I_c R_N$ increases rapidly, indicating that spin-triplet pair correlations are being produced in the Nb/Cu/X interface region. $I_c R_N$ reaches a maximum for $d_X$ values of a few nm, then decreases at larger values of $d_X$. We believe that the decrease in $I_c R_N$ at large $d_X$, visible for $X = \text{PdNi}$, is caused by the destruction of the spin-triplet correlations created at the Nb/Cu/X interface 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.

What essential properties give PdNi and CuNi their ability to produce spin-triplet pair correlations in these Josephson junctions? We speculate that the two crucial ingredients for generation of the triplet are domain size, which should be comparable to the superconducting coherence length in Nb, and out-of-plane magnetocrystalline anisotropy. While the domain size of PdNi is not known, the domain size in Cu$_{0.9}$.Ni$_{0.53}$ has recently been measured to be about 100 nm [13], which is not so different from the Nb coherence length $\xi_N = 14$ 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 [16] and thus to non-collinear magnetizations in neighboring domains—a key requirement for production of the triplet [8]. Both PdNi [11] and CuNi [17] are known to have out-of-plane magnetic anisotropy. In this context, it is interesting to note what happens when the Cu buffer layers between the X and Co layers are omitted. We have tried this for $X = \text{PdNi}$, and found that the supercurrent is much smaller than in samples with Cu buffer layers. Presumably the domain structure of the PdNi is changed by exchange-coupling to the Co, in a way that is detrimental to production of the triplet correlations. We suspect that the PdNi magnetization is forced to lie in-plane near the interface with Co, which leads to less non-collinear magnetization in neighboring domains [13].

It is natural to ask whether there are other materials besides PdNi or CuNi that can produce spin-triplet correlations. Clearly Co alone is not sufficient, as demonstrated by the samples without X layers [10]. 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 [18]. Non-collinear magnetization resides only in the domain walls, which is apparently not enough to produce a significant amount of spin-triplet. Aside from the presence of non-collinear magnetizations, theory suggests that any “spin-active” interface between a superconductor and a ferromagnet can produce spin-triplet correlations [19]. We have tried using X = Cu$_{0.9}$.Pt$_{0.06}$, an alloy with strong spin-orbit scattering, but preliminary data show very little, if any, signature of the triplet. We predict that PdCo, another weakly ferromagnetic alloy with properties similar to those of PdNi, will produce triplet correlations.

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, $l_c$, is the shortest relevant length scale in the problem, the spin-singlet supercurrent should decay on the length scale $\xi_F = \sqrt{D_F E_F}$, where $D_F$ and $E_F$ are the electron diffusion constant and exchange energy in the ferromagnet. Josephson junctions containing Co, however, are in the “intermediate” limit, with $l_c$ longer than $\xi_F$, but shorter than $\xi_S$, the superconducting coherence length. In that limit, the spin-singlet supercurrent decays on the length scale $l_c$, which is estimated to be 2.4 - 3.0 nm from previous studies [10, 20]. Spin-triplet supercurrent, in contrast, should decay over a much longer length scale given by the smaller of the normal metal coherence length, $\xi_N = \sqrt{D_N E_N}$, or the spin memory length, $L_{sf} = \sqrt{D_F / \tau_{sf}}$, where $\tau_{sf}$ 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 [21], we estimate $D_F = 5 \times 10^{-3}$ m$^2$/s and $5 \times 10^{-4}$ 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 [22, 23]. Unfortunately, sample-to-sample fluctuations in the experimental data in Figure 3a mask any discernible decay for $D_{Co}$ between 12 and 28 nm. Better statistics or data over a much larger range of $D_{Co}$ will be needed to extract a meaningful estimate of the decay length for the spin-triplet supercurrent. We have not yet attempted to increase $D_{Co}$ much more due to concerns about the efficacy of the Co/Ru/Co exchange-coupled trilayer.

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$_2$RuO$_4$ [1]. 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 or-
hital 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 [24], is counter-intuitive, as it implies that the equal-time pair correlation function vanishes. Theoretical guidance is needed to find an intuitive, as it implies that the equal-time pair correlation function vanishes. Theoretical guidance is needed to find such a direct method to probe the odd-frequency aspect of the pair correlations experimentally, rather than relying on the observed long-range supercurrent to deduce the spin-triplet character combined with robustness to disorder.

Looking back, there were hints of long-range proximity effects in superconducting/ferromagnetic hybrid systems as early as 10 years ago [25, 26, 27, 28], but there was no way to control the observed effects. More recently, Sosnin et al. [29] 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 authors did not observe a Josephson effect on PdNi or CuNi thickness, will pave the way to many new experiments [30, 31].

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* Electronic address: birge@pa.msu.edu

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