Area-dependence of spin-triplet supercurrent in ferromagnetic Josephson junctions

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Abstract. In 2010, several experimental groups obtained compelling evidence for spin-triplet supercurrent in Josephson junctions containing strong ferromagnetic materials. Our own best results were obtained from large-area junctions containing a thick central Co/Ru/Co “synthetic antiferromagnet” and two thin outer layers made of Ni or PdNi alloy. Because the ferromagnetic layers in our samples are multi-domain, one would expect the sign of the local current-phase relation inside the junctions to vary randomly as a function of lateral position. Here we report measurements of the area dependence of the critical current in several samples, where we find some evidence for those random sign variations. When the samples are magnetized, however, the critical current becomes clearly proportional to the area, indicating that the current-phase relation has the same sign across the entire area of the junctions.

The interplay between magnetism and superconductivity has been a fertile ground for new physics over several decades. On the one hand, bulk materials exhibiting both forms of order are rare, although there is recent progress in discovering new materials with exotic properties [1, 2, 3]. On the other hand, combining conventional superconducting (S) and ferromagnetic (F) materials relaxes the constraints, leading to a wide variety of possibilities. The interplay between the two is governed either by the magnetic field [4] or the exchange field [5]; in this work we restrict our attention to the latter. The essential physics arises from a modification of Andreev reflection – the microscopic process by which electrons enter or leave S. A conceptually simple picture is to consider two electrons belonging to a Cooper pair, which happen to cross the S/F interface. Because they enter opposite spin bands in F, they have different Fermi wavevectors, hence the pair acquires a center-of-mass momentum perpendicular to the S/F interface [6]. That, in turn, leads to oscillations in the pair correlation function, which manifest themselves as oscillations in the $T_c$ of S/F bilayers [7], inversion in the proximity-induced density of states on the F side of S/F bilayers [8], and oscillations between “0” and “$\pi$” states in S/F/S Josephson junctions [9, 10]. The oscillatory phenomena associated with S/F hybrids are fascinating and have potential for some applications; on the other hand, they are very short-range [5], decaying on a length scale governed by the exchange energy or by the mean free path in F.

A new chapter in S/F physics was opened in 2001 with the prediction that spin-triplet pair correlations can be induced in S/F hybrids made from conventional spin-singlet superconductors, in the presence of specific types of magnetic inhomogeneity that include non-collinear magnetizations [11, 12, 13, 14]. Since the two electrons from a spin-triplet pair with $m = \pm 1$ (where $m$ is the component of spin angular momentum along the magnetization direction of F) live in the same spin band in F, they are not subject to the exchange field. As a result,
such spin-triplet pair correlations are long-range in F, decaying on a length scale governed by
the temperature or by spin-flip or spin-orbit scattering. The experimental search for spin-triplet
pairs produced a few promising hints a few years ago [15, 16], but conclusive evidence for their
existence came only last year when several groups reported long-range supercurrents in S/F/S
Josephson junctions [17, 18, 19, 20]. Our own results [17] were based on junctions of the form
S/N/F′/N/SAF/N/F′/N/S (see Fig. 1), where SAF is a Co/Ru/Co “synthetic antiferromagnet”
[21], N is Cu, and F′ and F″ are thin ferromagnetic layers (Ni in this work). Conversion of spin-
singlet to spin-triplet pairs in our samples can be viewed as taking place in two steps [14]: 1) The
two electrons of a spin-singlet Cooper pair leaving the bottom S layer acquire different
phase factors in the F′ layer, which generates the \( m = 0 \) triplet component; 2) Rotation of the
\( m = 0 \) triplet component into the basis defined by the SAF magnetization generates the \( m = 1 \)
and \( m = -1 \) triplet components in the new basis, as long as it is non-collinear with the F′
basis. Conversion back to spin-singlets occurs between the SAF and F″ layers as the inverse
process. This type of junction, with three ferromagnetic layers, had been previously proposed
theoretically by Houzet and Buzdin [22], but with a simple F layer in place of the SAF, and
without the crucial inner two N layers which serve to magnetically decouple the F′ and F″ layers
from the Co layers. The critical supercurrent in these samples hardly decays as the total Co
thickness varies from 12 to 30 nm [17], in sharp contrast to similar samples without F′ layers
where the supercurrent decays very rapidly with increasing Co thickness [23].

The results described above [17] immediately raise several questions, only some of which
have been addressed by subsequent work [24]. One of the most important outstanding questions
concerns the current-phase relation of the junctions, or more precisely, the current-phase relation
as a function of position across the lateral dimensions of the junction. At issue is the theoretical
prediction that S/F′/F/F″/S or S/F′/SAF/F″/S junctions (we omit explicit reference to the N
layers for simplicity) can be either “0” or “\( \pi \)” junctions depending on the relative orientations of the
magnetizations in the three ferromagnetic layers [22, 25, 26, 27]. (In the weak-coupling limit,
a 0-junction has the current-phase relation \( I_s = I_c \sin(\phi) \), whereas a \( \pi \)-junction has the current-
phase relation \( I_s = I_c \sin(\phi + \pi) = -I_c \sin(\phi) \), where \( \phi \) is the gauge-invariant superconducting
phase difference between the two S electrodes.) Looking through the junction in the direction of current
flow, the three magnetizations can either rotate in the same direction from one to the
next (with either chirality), or the rotation from the F′ layer to F can be in the opposite
sense with respect to the rotation from F to the F″ layer. (We define the direction of rotation
using the angle that is less than \( \pi \).) In the former case, the junction is a 0-junction, whereas in
the latter case it is a \( \pi \)-junction [27]. Since our samples have relatively large lateral dimensions
(10, 20, or 40 µm in our published work [17]), and since a typical Co domain size is only on the
order of 3µm in our sputtered films in the relevant thickness range [28], we expect our samples to contain many domains which produce a random pattern of 0 and π junctions. In that case one would expect the typical critical current of a sample to be proportional to the square root of the number of domains, hence proportional to the square root of the junction area. This is quite different from the usual case where the current-phase relation is uniform across the entire area of the junction, and the critical current is proportional to the junction area. The main goal of this paper is to determine experimentally which type of area scaling occurs in our samples.

Our sample fabrication procedure is described in detail in our previous publications [29, 23, 17]. All of the samples discussed in the present work contain two 1.5-nm-thick Ni layers as the F' and F" layers as shown in Figure 1. Photolithography and ion milling are used to define Josephson junctions with circular cross-section. Most of our published data were obtained from junctions with diameters of 10 or 20 µm. Taking into account sample-to-sample fluctuations, we were not able to determine the area scaling law from our previous data sets [17, 24]. Since the goal of the present work is to measure how the critical current scales with the junction area, we fabricated junctions with diameters of 3, 6, 12, and 24 µm on each substrate. Although the success rate of the largest junctions is low, we obtained data from 3, 6, and 12 µm diameter junctions on several different substrates. All of the measured junctions display characteristics obeying the standard overdamped form: \( V = R_N(I^2 - I_c^2)^{1/2} \), where \( R_N \) is the normal state resistance.

For every Josephson junction we fabricate, we measure the dependence of \( I_c \) with magnetic field applied perpendicular to the current direction, i.e. in the plane of the substrate. The left panels of Figure 2 show the resulting “Fraunhofer patterns” of a typical set of junctions in the virgin state, fabricated on a single substrate, with diameters of 3, 6, and 12 µm. The quality of the virgin-state Fraunhofer patterns varies, which is not surprising given the amount of magnetic material contained in the junctions [30, 29]. If the SAF is working effectively, then the Co layers produce zero net magnetic flux in the junctions, because a given domain in one Co layer is antiparallel to that in the other Co layer [23]. Deviations from ideal Fraunhofer patterns are most probably due to the two Ni layers. Figure 2 also shows Fraunhofer patterns of the same
three junctions after they have been subjected to a large in-plane magnetic field of 1800 Oe. (The large field is then removed to obtain the low-field Fraunhofer pattern.) One immediately notices three features in the data: 1) the quality of the Fraunhofer patterns is improved; 2) the central peak of the pattern is shifted to a small negative field; and 3) the maximum critical current determined from the largest peak in the pattern is much larger than in the virgin state data. All three features can be explained by two hypotheses: i) the Ni layers become fully magnetized in the direction of the applied field; and ii) the SAF undergoes a "spin-flop" transition, whereby the two Co layers end up perpendicular to the direction of the applied field [31, 32]. The net result is that the Ni and Co magnetizations end up perpendicular to each other, which optimizes generation of the spin-triplet pairs [22, 26, 27]. Meanwhile, the central peak of the Fraunhofer pattern is shifted to a small negative field where the magnetic flux due to the external field exactly cancels the magnetic flux due to the Ni magnetization [33, 29]. Further evidence for the occurrence of the spin-flop transition in our samples will be provided elsewhere [34].

The evolution of the $I_c$ enhancement as the samples are magnetized is shown in Figure 3. The figure shows $I_cR_N$ as a function of $H_{app}$, the magnitude of the applied field, as that field is increased in discrete steps. After each step, the full Fraunhofer pattern is measured near zero field; the quantity plotted is from the value of $I_c$ obtained from the peak of the resulting Fraunhofer pattern. For all three samples, $I_c$ starts to increase when $H_{app}$ exceeds about 800 Oe, and attains a plateau when $H_{app}$ reaches about 1800 Oe. Data from samples with different Ni thicknesses [34] show that the values of $H_{app}$ where $I_c$ increases vary with Ni thickness in a manner consistent with the coercive field of thin Ni films (larger coercive field for thinner films). The field at which the spin-flop transition occurs cannot be determined from these data; all we know is that it is less than 500 Oe – the coercive field of our thickest Ni layers measured.

Returning to the question of how $I_c$ scales with junction area, Figure 4 shows a plot summarizing data from 18 Josephson junctions on 6 separate substrates. We plot $I_cR_N$ in both the virgin state and after the samples are fully magnetized, as a function of junction diameter. Several systematic features of the data stand out. First, the data obtained from the magnetized samples are remarkably consistent over all junction sizes and all substrates. Since $R_N$ is inversely proportional to the junction area $A$, the fact that $I_cR_N$ is independent of area implies that $I_c$ scales linearly with junction area in the magnetized state, i.e. $I_c \propto A$. This means that the supercurrent is adding coherently across the area of the junction. That is expected when the two Ni layers are magnetized in the same direction across the whole junction area. For example,
if we define the direction of $H_{app}$ as $\theta = 0$, so that the two Ni magnetizations point along $\theta = 0$, then the lower and upper Co layers should point either along $\theta = \pi/2$ and $-\pi/2$, respectively, or the converse, due to the spin-flop. In either case, the sense of rotation from the bottom Ni layer to the lower Co layer will be the same as the sense of rotation from the upper Co layer to the top Ni layer. This would be true even if the Co magnetizations chose an angle different from $\pm \pi/2$, as long as that angle is not 0 and the two Co magnetizations point in opposite directions.

The virgin state data shown in Figure 4 show more variation from substrate to substrate. Before discussing those data, we first mention some variations in the ion milling process we used to fabricate the samples. To produce Josephson junctions with a well-defined lateral area, it is only necessary to ion mill through the upper 25-nm-thick Nb layer (see Fig. 1). We generally continue milling through the two Cu layers, the F’ layer, and part-way through the upper Co layer to ensure that there is no Nb left outside of the junction area. Of the samples reported in this study, samples #9, #12 and #14 were ion milled part-way through the upper Co layer. In these samples the lower Co layer is unaltered by the ion milling, and should maintain the domain structure of the continuous film. For these samples, panel (a) of Figure 4 shows that the virgin-state $I_cR_N$ decreases with increasing junction diameter $D$, with an almost inverse linear dependence (except for the 12µm pillar of sample #12). That means that $I_cR_N$ varies nearly inversely with the square root of the junction area, $I_cR_N \propto A^{-1/2}$, which means that $I_c$ is proportional to the square root of the area, $I_c \propto A^{1/2}$. That is what one would expect if the junctions consist of multiple domains contributing a random distribution of 0 and $\pi$ junctions. In this scenario, the large increase in $I_c$ that occurs when the samples are magnetized is largely due to a transition from $I_c \propto A^{1/2}$ scaling to $I_c \propto A$ scaling. Optimization of the angle between the Ni and Co magnetizations to $\pi/2$ also plays a factor [34]; we cannot determine from our data which factor plays the larger role.

![Figure 4](image_url)

**Figure 4.** Summary of $I_cR_N$ vs. Josephson junction lateral diameter for all samples measured, in both the virgin state (solid symbols) and after magnetization (open symbols). Each different symbol shape and color refer to a different substrate.

Panel (b) in Figure 4 shows data from several substrates that were ion milled further – either part-way through the lower Co layer (samples #15 and #16) or in one case all the way through the lower Co layer (sample #7). In those samples the scaling of the virgin state data is less clear, and may even be consistent with $I_c \propto A$ scaling. It is possible that further ion milling of the central Co/Ru/Co SAF and even the bottom Ni layer (in the case of sample #7) imposes constraints on the domain structure of the ferromagnetic layers, so that the domain magnetizations can no longer be described as pointing in random directions.

The large sample-to-sample fluctuations in $I_cR_N$ seen in the virgin state data in Figure 4
are strongly correlated with the quality of the Fraunhofer patterns. For example, the 12 μm junction of sample #12, which clearly deviates from the $I_c \propto A^{1/2}$ scaling proposed above, had a particularly nice Fraunhofer pattern – much nicer than those from the 3 and 6 μm junctions on the same substrate. Similar correlations were observed in other samples: e.g. the 6 μm junction of sample #16 had a nicer Fraunhofer pattern than did the 3 or 12 μm junctions on that substrate.

In conclusion, we have observed strong $I_c \propto A$ scaling in S/F'/SAF/F”/S Josephson junctions after they are magnetized, demonstrating coherent superposition of supercurrent over the whole junction area. In the virgin state, there is evidence for $I_c \propto A^{1/2}$ scaling in some samples, but not in others. Large variations in the quality of the Fraunhofer patterns acquired from the virgin-state samples partially mask the area scaling. We plan to repeat this study using Pd$_{1-x}$Ni$_x$ alloy in place of Ni for the F’ and F” layers, since our previous work showed that PdNi layers do not distort the virgin-state Fraunhofer patterns [29].

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References

[1] Niewa R, Shlyk L, Blaschkowski B 2011 Z. Kristallogr. 226 352
[2] Shermadini Z et al. 2011 Phys. Rev. Lett. 106 117602
[3] Jeevan H S, Khasawneh M A, Pratt Jr W P and Birge N O 2010 Phys. Rev. Lett. 105 150502
[4] Luyskaytov I F and Pokrovsky V L 2005 Adv. Phys. 54 67
[5] Buzdin A I 2005 Rev. Mod. Phys. 77 935
[6] Demler E A, Arnold G B and Beasley M R 1997 Phys. Rev. B 55 15174
[7] Jiang J S, Davidovic D, Reich D H and Chien C L 1995 Phys. Rev. Lett. 74 314
[8] Khasawneh M A, Aprili M, Lesueur J and Grison X 2001 Phys. Rev. Lett. 86 304
[9] Ryazanov V V, Oboznov V A, Rusanov A Yu, Veretennikov A V, Golubov A A and Aarts J 2001 Phys. Rev. Lett. 86 2427
[10] Kontos T, Aprili M, Lesueur J, Genet F, Stephanidis B, Boursier R 2002 Phys. Rev. Lett. 89 137007
[11] Bergeret F R, Volkov A F and Efetov E B 2001 Phys. Rev. Lett. 86 4096
[12] Kadigrobov A, Sheldrer I R and Jonson M 2001 Europhys. Lett. 54 394
[13] Eschrig M, Kopul J, Cuevas J C and Gerd Schm 2003 Phys. Rev. Lett. 90 137003
[14] Eschrig M 2011 Physics Today 64, No. 1, 43
[15] Keizer R S, Goennenwein S T B, Klapwijk T M, Xiao G and Gupta A 2006 Nature (London) 439 825
[16] Sosnin I, Cho H, Petroshav V V and Volkov A F 2006 Phys. Rev. Lett. 96 157002
[17] Khaire T S, Khasawneh M A, Pratt Jr W P and Birge N O 2010 Phys. Rev. Lett. 104 137002
[18] Robinson J W A, Witt J D S and Blamire M G 2010 Science 329 59
[19] Sprungmann D, Westerholt K, Zabel H, Weiodes M and Kohlstedt H 2010 Phys. Rev. B 82 060505
[20] Anwar M S, Czeschka F, Hesselberth M, Porcu M and Aarts J 2010 Phys. Rev. B 82 100501
[21] Parkin S S P, More N and Roche K P 1990 Phys. Rev. Lett. 64 2304
[22] Houzet M and Buzdin A I 2007 Phys. Rev. B 75 060504(R)
[23] Khasawneh M A, Pratt Jr W P and Birge N O 2009 Phys. Rev. B 80 020506(R)
[24] Khasawneh M A, Khaire T S, Klose C, Pratt Jr W P and Birge N O 2011 Supercond. Sci. Technol. 24 024005
[25] Volkov A F, Bergeret F S and Efetov K B 2003 Phys. Rev. Lett. 90 117006
[26] Volkov A F and Efetov K B 2010 Phys. Rev. B 81 144522
[27] Trifunovic L and Radovic Z 2010 Phys. Rev. B 82 020505(R)
[28] Borchers J A, et al. 1999 Phys. Rev. Lett. 82 2796
[29] Khaire T S, Pratt Jr W P and Birge N O 2009 Phys. Rev. B 79 094523
[30] Bourgeois O, Gandit P, Lesueur J, Sulpice A, Grison X and Chaussy J 2001 Eur. Phys. J. B 21 75
[31] Zhu J G and Zheng Y 1998 IEEE Trans. Mag. 34 1063
[32] Tong H C, Qian C, Miloslavsky L, Funada S, Shi X, Liu F and Dey S 2000 J. Appl. Phys. 87 5055
[33] Ryazanov V V 1999 Physics-Uspekhi 42 825
[34] Klise C, Khaire T S, Wang Y, Pratt Jr W P, Birge N O, McMorran B J, Ginley T P, Borchers J A, Kirby B J, Maranville B B, Unguris J 2011 Preprint arXiv:1108.5666