RAPID COMMUNICATIONS

PHYSICAL REVIEW B **80**, 020506(R) (2009)

Josephson junctions with a synthetic antiferromagnetic interlayer

Mazin A. Khasawneh, W. P. Pratt, Jr., and Norman O. Birge*

Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824-2320, USA

(Received 12 May 2009; published 31 July 2009)

We report measurements of the critical current vs Co thickness in Nb/Cu/Co/Ru/Co/Cu/Nb Josephson junctions, where the inner Co/Ru/Co trilayer is a "synthetic antiferromagnet" with the magnetizations of the two Co layers coupled antiparallel to each other via the 0.6-nm-thick Ru layer. Due to the antiparallel magnetization alignment, the net intrinsic magnetic flux in the junction is nearly zero, and such junctions exhibit excellent Fraunhofer patterns in the critical current vs applied magnetic field even with total Co thicknesses as large as 23 nm. There are no apparent oscillations in the critical current vs Co thickness, consistent with theoretical expectations for this situation. The critical current of the junctions decays over four orders of magnitude as the total Co thickness increases from 3 to 23 nm. These junctions may serve as useful templates for future explorations of spin-triplet superconducting correlations, which are predicted to occur in superconducting/ferromagnetic hybrid systems in the presence of certain types of magnetic inhomogeneity.

DOI: 10.1103/PhysRevB.80.020506 PACS number(s): 74.50.+r, 74.45.+c, 75.70.Cn

Superconducting/ferromagnetic (S/F) hybrid systems have received much attention in the past decade.[1] When a conventional spin-singlet Cooper pair crosses the S/F interface, the two electrons enter different spin bands, hence the pair picks up a momentum shift proportional to the exchange energy.[2] This physical process leads to a number of oscillatory phenomena in S/F systems, including oscillations in the T_c of S/F bilayers and in the critical current of S/F/S Josephson junctions as a function of F-layer thickness.[1] There are proposals to use S/F/S π junctions as components in superconducting circuits or in various quantum computing schemes.

A recent development is the prediction of a new kind of spin-triplet pair correlations in S/F induced in conventional S/F systems by the presence of certain forms of magnetic inhomogeneity.[3–5] Unlike spin-singlet pairs, spin-triplet pairs are not subject to the exchange field, hence they should propagate long distances in a ferromagnetic material—limited only by the temperature or by spin-flip or spin-orbit scattering. One place to search for spin-triplet correlations is in thick S/F/S Josephson junctions, where the spin-singlet supercurrent is exponentially suppressed by the exchange field.[6] Depending on their geometry and the type of F material, however, thick S/F/S junctions may contain a large amount of intrinsic magnetic flux, which distorts the "Fraunhofer" pattern of the critical current I_c vs applied magnetic field and reduces the reliability of I_c measurements. Pseudospin valves of the type Co/Cu/Permalloy sandwiched between Nb layers have been used to control I_c by switching the relative orientation of magnetizations of the F layers in weak magnetic fields.[7] Maximum I_c is obtained for antiparallel orientations. Our method utilizes antiparallel orientation of identical Co layers.

We report here measurements of Nb/Cu/Co/Ru/Co/Cu/Nb Josephson junctions, where the central Co/Ru/Co trilayer is a synthetic antiferromagnet with the magnetizations of the two Co layers exchange-coupled antiparallel (AP) to each other via the 0.6-nm-thick Ru layer.[8] The total Co thickness was varied between 3 and 23 nm—much thicker than in previous studies of S/F/S junctions using Co with thicknesses up to 5 nm.[9] Over our range of Co thicknesses, I_c drops by more than four orders of magnitude while exhibiting a nearly perfect Fraunhofer pattern over the entire range. We do not observe any signature of spin-triplet correlations in these samples, but we suggest that they may serve as a useful platform for future searches for triplet correlations, perhaps by adding additional magnetic layers with inhomogeneous magnetization adjacent to the Nb layers.

Multilayer samples of the form Nb(150)/Cu(5)/Co(x)/Ru(0.6)/Co(x)/Cu(5)/Nb(25)/Au(15), with all thicknesses in nm, were grown by dc triode sputtering in an Ar pressure of 2.5 mTorr, in a system with base pressure of 2×10^{-8} Torr. The thin Cu layers change the growth characteristics of the Co layers, and result in larger I_c of the junctions for thick Co layers. (Results for samples with and without the Cu layers will be shown below.) The total Co thickness, $d_{Co}=2x$, was varied between 3 and 23 nm. The multilayers were patterned into circular pillars of diameters 10, 20, 40, and 80 μm using an image reversal photolithographic process and Ar ion milling. The milling was followed immediately by deposition of 160 nm of SiO_x, then lift off of the photoresist mask. Top Nb electrodes of thickness 200 nm were deposited by sputtering. A schematic diagram of the sample geometry is shown in Fig. 1. All I_c measurements were performed at 4.2 K with the samples inside a Cryoperm magnetic shield, using a superconducting quantum interference device (SQUID)-based current comparator method.[10] Current-voltage characteristics of all samples fol-

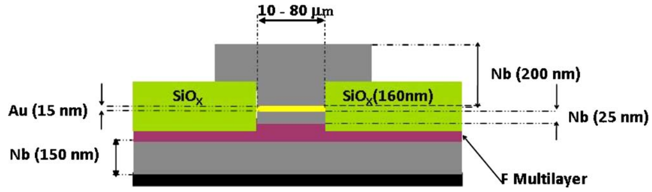

FIG. 1. (Color online) Schematic diagram of S/F/S Josephson junction cross section, where the "F multilayer" refers to the Co/Ru/Co trilayer, with or without additional Cu buffer layers adjacent to the Nb electrodes. Current flow is in the vertical direction. The magnetic field is applied in the plane of the layers, i.e., perpendicular to the current direction.

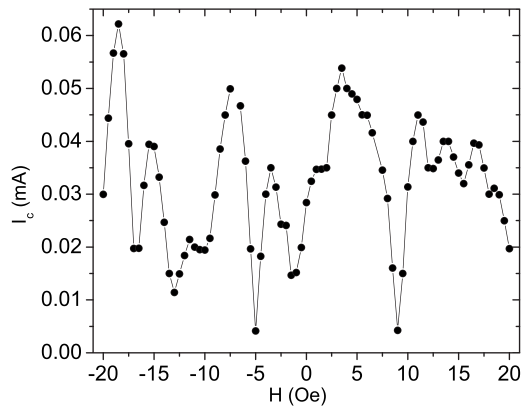

FIG. 2. Critical current vs magnetic field applied in the film plane (perpendicular to the current direction) for a Nb/Co/Nb circular Josephson junction of diameter 40 μm and $d_{Co}=5$ nm.

lowed the standard form for overdamped Josephson junctions.

A valuable tool for characterizing the quality of Josephson junctions is the measurement of I_c vs magnetic field applied perpendicular to the current direction. Observation of a good Fraunhofer pattern for junctions guarantees that the current flow is uniform across the junction and that there are no shorts in the surrounding insulator. Observation of the Fraunhofer pattern in S/F/S junctions with strong ferromagnets, however, can be problematic due to the intrinsic magnetic flux of the ferromagnetic domains. For sufficiently thin F layers, the Fraunhofer patterns can be extremely good.[11] In junctions with extremely small lateral dimensions, good Fraunhofer patterns can be obtained over a larger range of F-layer thickness.[12] But for sufficiently thick F layers, the Fraunhofer pattern becomes random, with no clear central maximum. An example for a circular junction of diameter 40 μm, with a single Co layer 5 nm thick, is shown in Fig. 2. The deep minima in I_c at $H=-5$ and $+8$ Oe demonstrate that there are no shorts in the oxide surrounding the junction. The overall pattern, however, is quite random due to the magnetic domain structure of the Co film. Similar random Fraunhofer patterns have been seen previously in S/F/S junctions containing other strong ferromagnetic materials: Gd (Ref. 13) and Ni.[14]

Fabrication of Josephson junctions containing the synthetic antiferromagnetic trilayer, Co(x)/Ru(0.6)/Co(x), circumvents this problem. Figure 3 shows Fraunhofer patterns for four samples with total Co thicknesses varying from 6.1 to 23 nm. The first three patterns are nearly perfect, while the last one is very good. The maximum field shift of the patterns is a few Oe, which indicates a very strong antiferromagnetic coupling between the top and bottom Co layers. Solid lines are fits to the theoretical Airy formula for junctions with circular cross section:

$$I_c(\Phi) = I_c(0)\frac{2J_1\left(\frac{\pi\Phi}{\Phi_0}\right)}{\left(\frac{\pi\Phi}{\Phi_0}\right)}, \qquad (1)$$

where J_1 is the Bessel function of the first kind, $\Phi_0=h/2e$ is the flux quantum, $\Phi=H_{ext}(2\lambda_L+d)w$ is the magnetic flux penetrating the junction with λ_L as the London penetration depth, w as the junction diameter, and d as the barrier thickness. Figure 4 shows M vs H for a sample with $d_{Co}=4$ nm. M does not saturate until H is at least 5 kOe, and there is very little hysteresis between curves with H increasing and decreasing, consistent with strong AP coupling of the two Co layers.

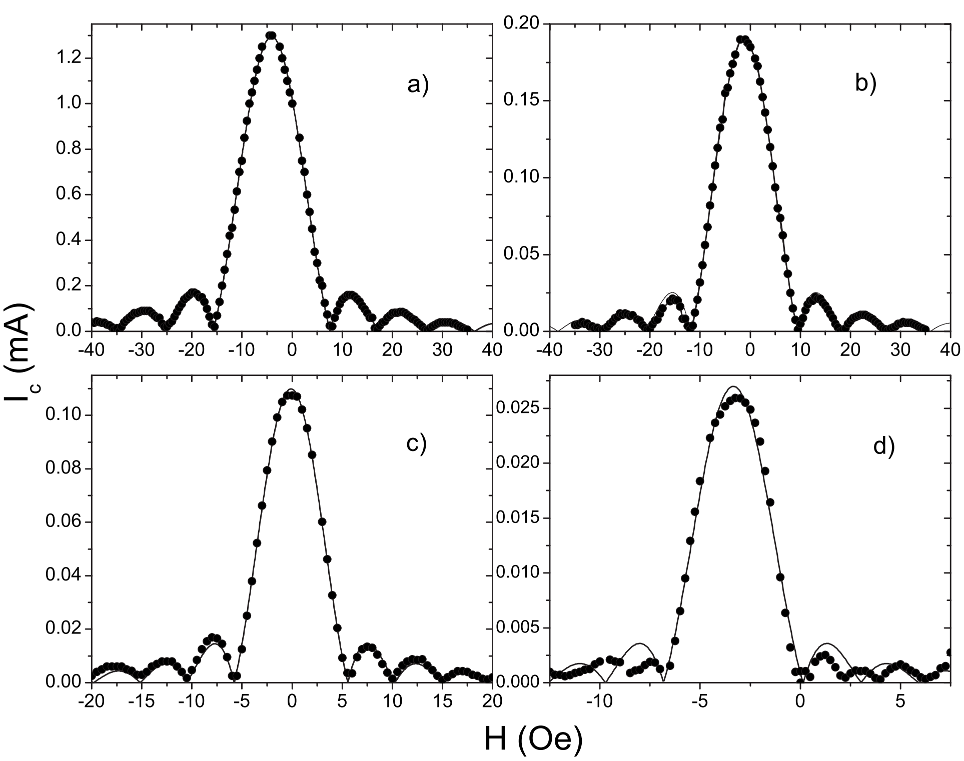

FIG. 3. Critical current vs applied magnetic field obtained for Nb/Cu/Co/Ru/Co/Cu/Nb circular Josephson junctions with total Co layers thickness of: (a) 6.1, (b) 11, (c) 18, and (d) 23 nm. The pillar diameters w are 10, 10, 20, and 40 μm, respectively. The solid lines are fits to Eq. (1).

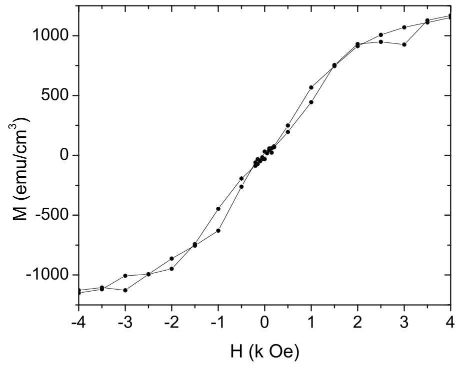

FIG. 4. Magnetization vs applied field at $T=10$ K for a Co(4)/Ru(0.6)/Co(4) trilayer grown on 150 nm of Nb.

We subjected one Josephson junction sample to a series of large in-plane magnetic fields and then remeasured I_c vs H at low field. The resulting Fraunhofer patterns showed only slight distortion after applying fields as large as 5 kOe. After applying 10 kOe the central peak in the Fraunhofer pattern split into two peaks of about half the original magnitude. After warming the sample to room temperature and cooling back to 4.2 K, an excellent Fraunhofer pattern was obtained once again.

The dependence of I_c on total Co thickness (d_{Co}) is summarized in Fig. 5. The figure shows two sets of data: black circles represent samples fabricated with Cu buffer layers, while red triangles represent samples fabricated without. In

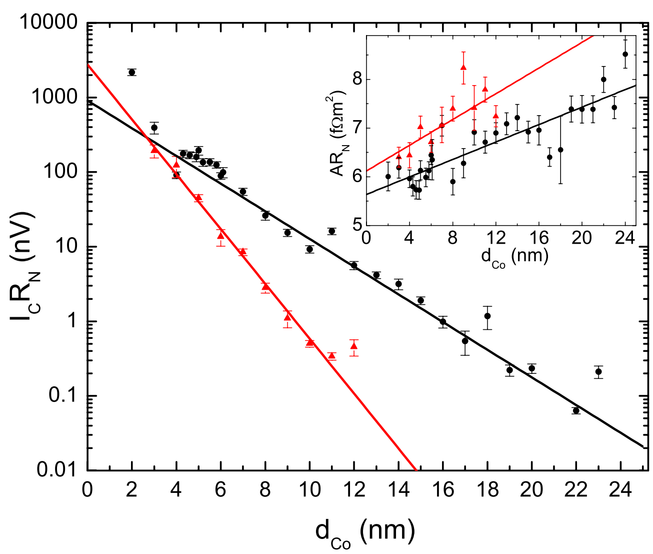

FIG. 5. (Color online) Product of critical current times normal-state resistance vs total Co thickness for all of our SAF Josephson junctions. Red points (triangles) are data for samples without Cu buffer layers, while black points (circles) are data for samples with Cu buffer layers. Error bars represent the standard deviation of measurements taken on more than one pillar on the same substrate, with the minimum uncertainty chosen to be 10%. The solid lines are fits to a simple exponential decay, with decay lengths of 1.18 ± 0.05 and 2.34 ± 0.08 nm, respectively. Inset: AR_N vs d_{Co}, where the data point scheme is the same as in the main figure and A is area of the Josephson junctions. The lines are fits to the data whose slopes give the effective resistivities of the Co layers.

both cases I_c decays exponentially with d_{Co}. In samples without the Cu, the decay is faster than in samples with Cu. We focus on the larger data set—the samples with Cu. An immediate question is whether these samples can be π junctions; i.e., does I_c oscillate with d_{Co}? While the data do not display convincing oscillations, there are a few data points (e.g., for $d_{Co}=4.0$, 18, and 23 nm) that are substantially above or below their neighbors. To address the question of oscillations, we fabricated a set of samples in one sputtering run with closely spaced Co layer thicknesses in the range of 4.3–6.1 nm. Those samples do not exhibit any local minima in I_c, whereas Robinson *et al.*[9] observed a spacing of 1.0 nm between local minima for Nb/Co/Nb junctions containing a single Co layer.

Several theoretical papers address the expected behavior of I_c vs d_{Co} for Josephson junctions containing two magnetic layers with noncollinear magnetizations.[15–17] We discuss only the situation relevant to our experiments, where the two ferromagnetic layers have equal thickness and antiparallel magnetizations. In the ballistic limit, such S/F/F/S junctions are predicted to behave similarly to S/N/S junctions—with a slow algebraic decay and no oscillations in I_c—because the relative phase shift acquired by the two electrons of a Cooper pair as they travel through the first F layer is exactly cancelled by the phase shift they acquire through the second F layer.[15] In the presence of disorder I_c decays exponentially with F layer thickness but still without any oscillations of the kind associated with S/F/S junctions. Our data are consistent with this picture.

To extract quantitative information from our data, we must go a step further with the theory. The theoretical works cited above calculate the exact form of the I_c decay only in certain limits, e.g., for the pure ballistic case with no elastic scattering and for the diffusive limit with $E_{ex}\tau \ll \hbar$, where τ is the mean free time between collisions. Josephson junctions with Co, however, fall into an intermediate limit, where $E_{ex}\tau > \hbar$, but $\Delta\tau \ll \hbar$, with Δ as the superconducting gap. Although theories in the intermediate limit do not exist for S/F/F/S junctions, they do exist for S/F/S junctions[18,19] and predict exponential decay of I_c with a decay constant equal to the mean free path in the F material. (Theories for S/F/S junctions also predict oscillations, which are not present in the S/F/F/S case studied here.) The solid lines in Fig. 5 are least-squares fits of an exponential decay to our two data sets, with decay lengths 2.34 ± 0.08 nm for the samples with Cu buffer layers and 1.18 ± 0.05 nm for the samples without Cu. The ratio of these with-Cu to without-Cu decay lengths is 2.0 ± 0.1. Since the effective resistivities of the Co determined from the slopes in the inset to Fig. 5 are inversely proportional to the mean free paths, we determine the ratio of the mean free paths in the Co with Cu to without Cu to be 1.5 ± 0.5. This mean-free-path ratio is consistent with the ratio for the decay lengths, in accord with predictions for S/F/S junctions[18,19] in the intermediate limit.

It is instructive to compare our results with those of Robinson *et al.*,[9,12] who studied S/F/S junctions made with the strong ferromagnets Co, Ni, Fe, and Py, all of which are believed to lie in the intermediate limit defined above. Those workers found that, for Ni and Py, the I_c vs d_F data followed an algebraic decay for small d_F and an exponential decay for larger d_F, with the crossover interpreted as occurring when d_F surpasses the mean free path, l_e. For Co, the data could be fit with either an algebraic or exponential decay over the thickness range studied (0.8–5 nm). As shown in Fig. 5, our I_c data decay exponentially over the entire range of $d_{Co}=2-23$ nm, with the possible exception of our first data point. Given the extra scattering in our samples from the two Co/Ru interfaces,[20] it is not surprising that $l_e<d_{Co}$ over the entire range of Co thicknesses we measured. What is surprising is that, if we were to plot the data of Robinson *et al.*[12] (ignoring the oscillations) in Fig. 5, they would lie a factor of 100 higher than our data over the narrow thickness range covered by both experiments. This suggests that the thin Ru layer severely suppresses I_c, possibly due to spin memory loss at the Co/Ru interfaces.

The single exponential decay of I_c vs d_{Co} shown in Fig. 5 indicates a lack of spin-triplet superconducting correlations in these samples, which would manifest themselves as a crossover to a slower decay with increasing d_{Co}. (The point at $d_{Co}=23$ nm might seem promising, but a sample with $d_{Co}=24$ nm exhibited a very small supercurrent and no Fraunhofer pattern, hence it was excluded from the figure.) There are several possible reasons why we do not observe the long-range triplet correlations (LRTCs). First, there could be substantial spin memory loss at the Co/Ru interfaces—an issue we intend to clarify in the near future using giant magnetoresistance techniques. Second, the amplitude of the LRTC generated at the S/F interfaces may be too small to measure. This could occur either if the domain structure in the Co films contains mostly domains aligned along a single directions in space (the LRTC requires noncollinear magnetizations) or if the LRTC component has random phases at adjacent Co domain walls and hence averages to zero over the lateral dimensions of the samples.[21] The latter situation could be ameliorated by fabricating samples with smaller lateral dimensions, while both issues could be addressed by utilizing a magnetic material with a well-characterized form of magnetic inhomogeneity, such as the spiral magnetic structure occurring in materials such as Ho.[22–24]

In this context, we note that optimizing the *generation* of the LRTC at the S/F interface may involve a choice of materials that does not optimize *propagation* of the LRTC through the subsequent ferromagnetic materials. It is here where we believe the Josephson junctions reported in this Rapid Communication may hold the most promise. One could produce samples of the form S/X/SAF/X/S, where X is a magnetic material chosen to optimize LRTC generation, while SAF is a suitable synthetic antiferromagnet with little spin memory loss, either the Co/Ru/Co trilayer studied here or a weaker SAF such as Co/Cu/Co.[25] Once the SAF layer becomes sufficiently thick (greater than about 23 nm for the case of the Co SAF studied here), the singlet supercurrent is suppressed by over four orders of magnitude. Generation of the LRTC at the S/X interfaces would then be manifested as a long-range spin-triplet supercurrent that persists out to SAF thicknesses far beyond what has been measured here.

We are grateful to S. Bergeret, Y. Blanter, D. van Harlingen, A. Volkov, and K. Efetov for helpful discussions and to B. Bi, T. Khaire, R. Loloee, and Y. Wang for technical assistance. This work was supported by the Department of Energy under Grant No. DE-FG02-06ER46341.

*birge@pa.msu.edu

[1] A. I. Buzdin, Rev. Mod. Phys. **77**, 935 (2005).
[2] E. A. Demler, G. B. Arnold, and M. R. Beasley, Phys. Rev. B **55**, 15174 (1997).
[3] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. Lett. **86**, 4096 (2001).
[4] A. Kadigrobov, R. I. Shekhter, and M. Jonson, Europhys. Lett. **54**, 394 (2001).
[5] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Rev. Mod. Phys. **77**, 1321 (2005).
[6] R. S. Keizer, S. T. B. Goennenwein, T. M. Klapwijk, G. Xiao, and A. Gupta, Nature (London) **439**, 825 (2006).
[7] C. Bell, G. Burnell, C. W. Leung, E. J. Tarte, D.-J. Kang, and M. G. Blamire, Appl. Phys. Lett. **84**, 1153 (2004).
[8] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).
[9] J. W. A. Robinson, S. Piano, G. Burnell, C. Bell, and M. G. Blamire, Phys. Rev. Lett. **97**, 177003 (2006).
[10] D. Edmunds, W. Pratt, and J. Rowlands, Rev. Sci. Instrum. **51**, 1516 (1980).
[11] D. Sprungmann, K. Westerholt, H. Zabel, M. Weides, and H. Kohlstedt, J. Phys. D **42**, 075005 (2009).
[12] J. W. A. Robinson, S. Piano, G. Burnell, C. Bell, and M. G. Blamire, Phys. Rev. B **76**, 094522 (2007).
[13] O. Bourgeois, P. Gandit, J. Lesueur, A. Sulpice, X. Grison, and J. Chaussy, Eur. Phys. J. B **21**, 75 (2001).
[14] T. S. Khaire, W. P. Pratt, Jr., and N. O. Birge, Phys. Rev. B **79**, 094523 (2009).
[15] Ya. M. Blanter and F. W. J. Hekking, Phys. Rev. B **69**, 024525 (2004).
[16] A. Vedyayev, C. Lacroix, N. Pugach, and N. Ryzhanova, Europhys. Lett. **71**, 679 (2005).
[17] B. Crouzy, S. Tollis, and D. A. Ivanov, Phys. Rev. B **75**, 054503 (2007).
[18] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. B **64**, 134506 (2001).
[19] O. Kashuba, Y. M. Blanter, and V. I. Falko, Phys. Rev. B **75**, 132502 (2007).
[20] C. Ahn, K.-H. Shin, and W. P. Pratt, Jr., Appl. Phys. Lett. **92**, 102509 (2008).
[21] S. Bergeret (private communication).
[22] I. Sosnin, H. Cho, V. T. Petrashov, and A. F. Volkov, Phys. Rev. Lett. **96**, 157002 (2006).
[23] A. F. Volkov, A. Anishchanka, and K. B. Efetov, Phys. Rev. B **73**, 104412 (2006).
[24] G. B. Halasz, J. W. A. Robinson, J. F. Annett, and M. G. Blamire, Phys. Rev. B **79**, 224505 (2009).
[25] S. S. P. Parkin, R. Bhadra, and K. P. Roche, Phys. Rev. Lett. **66**, 2152 (1991).