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Observation of standard spin-switch effects in ferromagnet/superconductor/ferromagnet trilayers with a strong ferromagnet

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We have measured the superconducting transition temperature T_c of ferromagnet/superconductor/ferromagnet trilayers using Permalloy ($Py=Ni_{84}Fe_{16}$) as a strongly polarized ferromagnetic material. For a parallel (P) or antiparallel (AP) alignment of the magnetization directions of the outer ferromagnets, we observe a T_c difference as large as 20 mK, with a stronger suppression of superconductivity in the P state than in the AP state. This behavior is opposite to the recent observations of Rusanov *et al.* [Phys. Rev. B **73**, 060505 (2006)] in Py/Nb/Py trilayers, but is consistent with earlier results on trilayers with Ni or CuNi alloy as the ferromagnetic material.

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The presence of a ferromagnetic (F) material in contact with a conventional superconductor (S) results in a strong mutual influence.[1] The superconducting correlations penetrate into the ferromagnet and oscillate in sign over a very short distance, due to the large energy difference between the majority and minority spin bands in the ferromagnet. Bilayers, trilayers, and multilayers of S and F materials exhibit a wide variety of novel phenomena, including oscillations of the superconducting critical temperature[2–4] and density of states[5] and Josephson junctions with a π-shifted ground state.[6]

In this Rapid Communication we focus on the "superconducting spin switch" first discussed in 1966 by de Gennes[7] and rediscovered in 1999 by Tagirov[8] and by Buzdin, Vedyayev, and Ryzhanova.[9] Those authors predicted that the critical temperature T_c of a F/S/F trilayer should depend on the relative magnetization direction of the two F layers, with the smallest T_c occurring in the parallel (P) state and the largest T_c in the antiparallel (AP) state. Those predictions were verified long ago by Deutscher and Meunier[10] and more recently by Gu *et al.*,[11] Potenza and Marrows,[12] and Moraru *et al.*[13] in a variety of F/S/F systems. It came as a surprise, therefore, when Rusanov *et al.*[14] recently reported observation of the *inverse* spin-switch effect in a series of Py/Nb/Py (Py=Permalloy) trilayer samples. Although the difference in T_c between the P and AP magnetization configurations was small in that work, the data showed clearly that the resistance in the transition region was higher for the AP configuration than for the P one. In fact, similar behavior had previously been observed by Peña *et al.*[15] in F/S/F trilayers made from superconducting $YBa_2Cu_3O_4$ and ferromagnetic $La_{0.7}Ca_{0.3}MnO_3$, with a spin polarization expected to be close to 100%. Those authors interpreted their observations as arising from enhanced reflection of spin-polarized quasiparticles at the F/S interfaces in the AP state leading to a stronger suppression of superconductivity,[16] and Rusanov *et al.*[14] claimed that the inverse spin-switch behavior is generic for F/S/F trilayers with strong ferromagnets. We believe that the mechanism based on the reflection of quasiparticles at the S/F interface[16] can explain changes in resistance under nonequilibrium conditions, but cannot explain differences in the equilibrium T_c between the P and AP states. Given our earlier work showing standard spin-switch behavior in Ni/Nb/Ni trilayers,[13] we were motivated to carry out independent measurements of T_c in Py/Nb/Py trilayers.

A series of $Py(8)/Nb(d_s)/Py(8)/Fe_{50}Mn_{50}(8)/Nb(2)$ multilayers (all thicknesses are in nm) was fabricated, with superconducting layer thickness d_s varying between 20 and 150 nm. The samples were grown directly onto oxidized Si substrates by magnetically enhanced triode dc sputtering in a high-vacuum chamber with a base pressure in the low-10^{-8}-Torr range and an Ar pressure of 2.0×10^{-3} Torr. The FeMn layer fixes the magnetization direction of the top Py layer by exchange bias[17] after undergoing a brief annealing and in-field cooling process. The Nb capping layer protects the FeMn from oxidation and is not superconducting.

Samples were patterned by mechanical masks for four-terminal current-in-plane resistance measurements, with $4.3\ mm\times1.6\ mm$ lateral dimensions. The critical temperatures were determined by ac resistance measurements with current of 10 μA, corresponding to a current density less

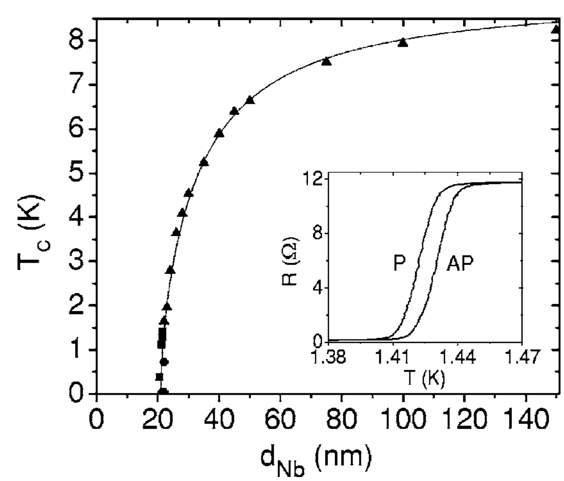

FIG. 1. Critical temperature vs Nb thickness for a series of $Py(8)/Nb(d_s)/Py(8)/Fe_{50}Mn_{50}(8)/Nb(2)$ samples (all thicknesses are in nm) from several sputtering runs. The solid line represents the theoretical fit as explained in the text. Inset: R vs T for a d_s=21.5 nm sample illustrating the difference between T_c for the P and AP states.

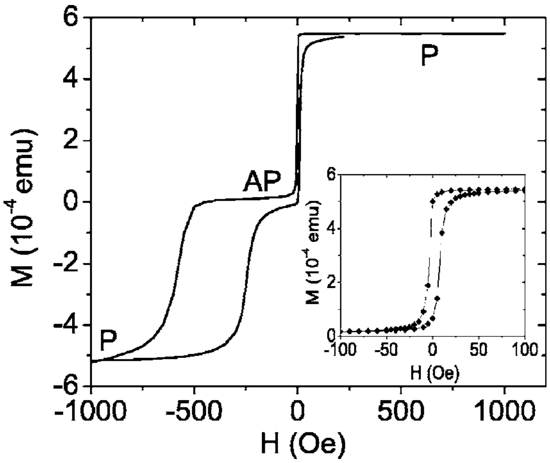

FIG. 2. Magnetization vs applied field for a d_s=23 nm sample measured at T=4.2 K. At $H \approx \pm 10$ Oe the free bottom Py layer switches while the pinned top Py layer switches at around −500 Oe. Inset: minor loop measured at T=4.2 K showing good switching of the free Py layer.

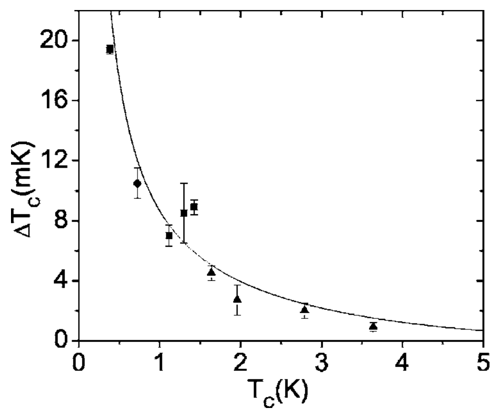

FIG. 3. ΔT_c vs critical temperature for a series of Py(8)/Nb(d_s)/Py(8)/$Fe_{50}Mn_{50}$(8)/Nb(2), where $\Delta T_c \equiv T_c^{AP} - T_c^P$. The fit to the data is obtained using the theory of Fominov *et al.* (Ref. 18) as outlined in the text.

than 3×10^5 A/m^2, low enough to be in the linear response regime. T_c was defined to be the temperature at which the resistance dropped to half its normal-state value. The results for the T_c measurements on our trilayers are given in Fig. 1, showing a strong dependence of T_c on the Nb thickness close to a critical thickness d_s^{cr}, where the sensitivity to ferromagnetism is enhanced. No superconductivity is observed above 36 mK for $d_s < d_s^{cr} \approx 20.5$ nm.

We have verified the magnetic configuration of our structures on simultaneously sputtered samples of larger lateral size using a superconducting quantum interference device (SQUID) magnetometer. Figure 2 shows a plot of M vs H for a sample with d_s=23 nm taken at 4.2 K, illustrating typical spin-valve behavior for the trilayer. The narrow hysteresis loop near H=0 shows the switching of the free Py layer with a coercive field H_c=5–10 Oe, while the wider loop shows switching of the pinned layer, shifted to nonzero H due to the exchange bias. The minor loop shown in the inset to Fig. 2 illustrates that fields of ±100 Oe switch the trilayer fully between the P and AP states. The nearly zero net magnetization observed at −100 Oe suggests very good AP alignment, while nearly saturated magnetization at +100 Oe indicates good P alignment. Similarly, well-defined P and AP states can be achieved at T_c and below.

Measurements of T_c^P and T_c^{AP} were obtained by alternating the applied field between the values +100 and −100 Oe and monitoring the two resistances as the temperature was slowly decreased through the transition. The largest shift in critical temperature, $\Delta T_c \equiv T_c^{AP} - T_c^P$, should occur in samples with d_s close to d_s^{cr}. The inset to Fig. 1 shows a plot of R vs T for a sample with a nominal thickness d_s=21.5 nm, measured in a dilution refrigerator. Two distinct transitions are observed for P and AP alignment close to 1.42 K, with a temperature separation of $\Delta T_c \approx 9$ mK. Samples with $d_s \approx 22$ nm have T_c's between 2 and 3 K and exhibit values for ΔT_c of only a few mK, similar to results obtained previously in other F/S/F systems.[11–13] For samples with $d_s > 26$ nm, no ΔT_c is observed. Magnetoresistance data, obtained for several samples with nonzero ΔT_c, were taken at temperatures in the middle of the superconducting transition and clearly showed the switching of the magnetic layers, with the P-state resistance always larger than the AP one. Figure 3 shows a plot of ΔT_c vs T_c for nine samples. The largest observed ΔT_c for our Py/Nb/Py trilayers is about 20 mK for a sample with d_s=20.5 nm and T_c=0.385 K. All our measurements show $T_c^P < T_c^{AP}$, a result opposite to what Rusanov *et al.*[14] observed in similar Py/Nb/Py trilayer systems.

The T_c of F/S/F trilayers in the P and AP states has been calculated theoretically by several groups.[8,9,18–20] The usual approach involves solving the Usadel equations in the dirty limit, which for the superconductor implies $l_S < \xi_{BCS} = \hbar v_S \gamma / \pi^2 k_B T_{c0}^{bulk}$ and for the ferromagnet $l_F < \hbar v_F / E_{ex}$, where l_S and l_F are the electron mean free paths in S and F. Here, T_{c0}^{bulk} is the bulk transition temperature of S, v_S and v_F are the Fermi velocities in the S and F materials, E_{ex} is the exchange energy of F, and γ=1.7811. For Py, the clean limit exchange length is $\hbar v_F / E_{ex}$=1.0 nm using E_{ex}=0.135 eV and majority Fermi velocity $v_F^{\uparrow}=0.22 \times 10^6$ m/s,[21] while the majority and minority band mean free paths are $l_F^{\uparrow}$=4 nm and $l_F^{\downarrow}$=0.6 nm, respectively.[22] Hence Py is on the border between the clean and dirty limits.[24]

We compare our data with the theory of Fominov *et al.*,[18] although it does not distinguish between the majority and minority spin bands of the F material. The following equations, which describe the critical temperatures T_c^P and T_c^{AP} for the P and AP cases, are obtained in the limit of a thin S layer with a constant superconducting gap Δ and a strong ferromagnet with $E_{ex} \gg \Delta$:

$$\ln \frac{T_{c0}}{T_c^P} - \mathrm{Re}\, \Psi\left(\frac{1}{2} + \frac{V_h}{2} \frac{\xi_S}{d_s} \frac{T_{c0}}{T_c^P}\right) + \Psi\left(\frac{1}{2}\right) = 0, \qquad (1)$$

$$\ln \frac{T_{c0}}{T_c^{AP}} - \Psi\left(\frac{1}{2} + \frac{W}{2} \frac{\xi_S}{d_s} \frac{T_{c0}}{T_c^{AP}}\right) + \Psi\left(\frac{1}{2}\right) = 0, \qquad (2)$$

where $\xi_S = \sqrt{\hbar D_S / 2\pi k_B T_{c0}}$, D_S is the diffusion constant in S, and T_{c0} is the critical temperature for an isolated S layer of thickness d_s. Fominov *et al.* emphasize that T_c varies with the relative magnetization angle even for $d_s > \xi_S$ because the critical temperature of the trilayer is suppressed as compared to that of the isolated Nb layer—i.e., $T_c \ll T_{c0}$. Consequently, the condition for which this theory is valid, $d_s \ll \xi = \sqrt{\hbar D_S/(2\pi k_B T_c)}$, is considerably weaker than the condition $d_s \ll \xi_S$, because $\xi \gg \xi_S$. In the limit of thick ferromagnets, the tanh functions in Ref. 18 are set to 1 and the functions V_h and W in Eqs. (1) and (2) become

$$V_h = \frac{\rho_S \xi_S}{(1-i)\rho_F/2k_h + R_B A}, \qquad W = \mathrm{Re}\{V_h\}, \tag{3}$$

where $k_h = \sqrt{E_{ex}/\hbar D_F}$ is the inverse dirty-limit exchange length in F and $R_B A$ is the boundary resistance times area of the S/F interface, a parameter that reflects both the interface quality and Fermi surface mismatch between the S and F materials. Equations (1)–(3) produced the fits to the Py/Nb/Py data shown in Figs. 1 and 3.

Estimates for the parameters appearing in the theory were obtained from additional measurements on bulk and thin film samples. Since the F-layer thickness is greater than $l_F^{\uparrow}$ and $l_F^{\downarrow}$ and remains fixed for all our samples, we have used a bulk value for the resistivity of Py: namely, $\rho_F = 123$ nΩ m.[25] By contrast, the thickness of the S layer in our trilayers changes and we have measured ρ_S as a function of the thickness on bare Nb thin films. In addition, the variation of T_{c0} with thickness was also measured on the same Nb films. The explicit dependencies of ρ_S and T_{c0} were taken into account in Eqs. (1) and (2).[26] The coherence length was obtained by performing perpendicular field measurements on the bare Nb films, giving $\xi_S \approx 6$ nm in the thickness range of our data. Taking the limit of $T_c \to 0$ in Eq. (1), for the behavior as d_s approaches d_s^{cr}, results in the relation $d_s^{cr}/\xi_S = 2e^C |V_h|$ where $C = 0.577$ is the Euler constant. Using this constraint and Eq. (3) one can obtain an estimate for the boundary resistance:

$$R_B A \approx 2e^C \rho_S(d_s^{cr}) \frac{\xi_S^2}{d_s^{cr}}, \tag{4}$$

where the value of ρ_S is taken at the critical thickness. After constraining $R_B A$ as shown above and using the measured values for the resistivities and ξ_S, k_h is the only remaining fit parameter.

Using Eq. (4) with $d_s^{cr} = 20.5$ nm and $\xi_S = 6$ nm gives the estimate $R_B A = 1.5$ fΩ m^2, which when utilized in Eq. (1) yields a fit that follows the T_c vs d_s data very well, as shown in Fig. 1. The fit is somewhat insensitive to the value of k_h. By contrast, k_h is tightly constrained by fitting to the ΔT_c vs T_c data. The results of that fit are illustrated in Fig. 3, showing good agreement with $k_h = 1.0$ nm^{-1}. Independent estimates of k_h using the values of v_F and l_F discussed earlier are 0.8 and 2.0 nm^{-1} for the majority and minority spin bands, respectively.[22]

The excellent fit shown in Fig. 3 motivated us to apply the theory of Fominov *et al.* to our previously reported data on Ni/Nb/Ni trilayers.[13] The results of the fit to the ΔT_c vs T_c data from the Ni/Nb/Ni trilayer are illustrated in Fig. 4 and also show excellent agreement. It is not not obvious *a priori* that the Ni layers in those samples are in the dirty limit,

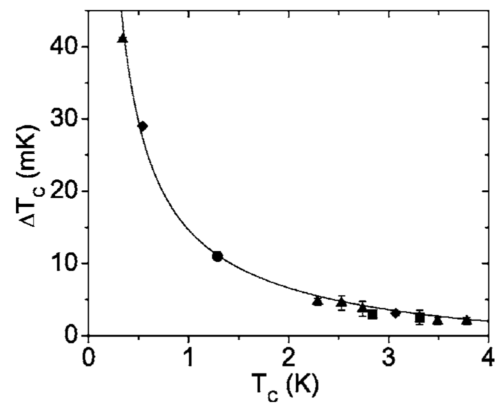

FIG. 4. ΔT_c vs critical temperature for a series of Ni(7)/Nb(d_s)/Ni(7)/$Fe_{50}Mn_{50}$(8)/Nb(2) (Ref. 13). The fit to the data is obtained using the theory of Fominov *et al.* (Ref. 18).

although recent experimental data on S/F/S systems[24] indicate a crossover to the dirty-limit behavior for Ni with layer thickness around 8 nm. The values $\rho_F = 33$ nΩ m, $\xi_S = 6$ nm, and $d_s^{cr} = 16.5$ nm were used in the fit, which gave $R_B A = 2.3$ fΩ m^2 for the Ni/Nb interface and $k_h = 0.5$ nm^{-1}. Independent measurements of the Nb/Ni interface resistance using current-perpendicular-to-plane resistance measurements of Nb/Ni multilayers yielded $R_B A = 2.35 \pm 0.25$ fΩ m^2, in excellent agreement with the value obtained from the fit to the T_c vs d_S data. Our independent estimate of k_h varies over a broad range due to uncertainty in determining the value of the diffusion constant (or mean free path) in Ni.[13] From the measured resistivity, we obtain values of l_F ranging between 7 and 70 nm, depending on the chosen value of the product $\rho_F l_F$ for Ni.[27] Combining that with the values for $E_{ex} = 0.115$ eV and $v_F = 0.28 \times 10^6$ m/s,[21] we obtain the range 0.16–0.5 nm^{-1} for k_h. The value corresponding to the shorter l_F agrees with the one from the fit to the data in Fig. 4.

It is unclear why Rusanov *et al.*[14] observe inverse spin switch behavior, $T_c^P > T_c^{AP}$, since we see the standard behavior, $T_c^P < T_c^{AP}$. Their samples were deposited under ultrahigh vacuum, but the coherence lengths of their thick Nb films are similar to ours, $\xi_S = (2/\pi)\xi_{GL} \approx 8$ nm. Also, we use exchange bias to pin the magnetization direction of one Py layer, whereas they rely on the different coercivities of the two layers. However, switching data in their micron-scale samples show a clear plateau, suggesting that a good AP state was achieved. In addition, they observe a difference between T_c^P and T_c^{AP} even when the Nb layer is very thick, 60 nm, whereas we measure a difference only for $d_s \lesssim 26$ nm. Variations in resistance or T_c have also been observed in F/S bilayers due to domain formation during magnetization switching.[28–30] But Rusanov *et al.* state that the features indicating the inverse spin-switch effect in their trilayers were not observed in bilayers. This, combined with their data on micron-scale samples that appear to be single-domain, argues against the dominance of domains in producing the inverse effect.

In summary, we observe similar spin-switch behavior in Py/Nb/Py and Ni/Nb/Ni trilayers—both S/F systems with strong ferromagnets. The results from both systems are fit well with the dirty-limit Usadel theory of Fominov *et al.*[18] This success is unexpected given that this theory assumes identical electronic properties (density of states, Fermi velocity, and mean free path) for the majority and minority spin bands of the ferromagnetic material, especially since Py is known to have a strong spin-scattering asymmetry.[23] The success of a dirty-limit theory in Ni is also surprising and may be due partly to strong diffusive scattering of electrons from the S/F interfaces.

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[1] For a review, see Yu. A. Izyumov, Yu. N. Proshin, and M. G. Khusainov, Phys. Usp. **45**, 109 (2002).

[2] J. S. Jiang, D. Davidovic, D. H. Reich, and C. L. Chien, Phys. Rev. Lett. **74**, 314 (1995).

[3] L. V. Mercaldo, C. Attanasio, C. Coccorese, L. Maritato, S. L. Prischepa, and M. Salvato, Phys. Rev. B **53**, 14040 (1996).

[4] Th. Mühge, N. N. Garif'yanov, Yu. V. Goryunov, G. G. Khaliullin, L. R. Tagirov, K. Westerholt, I. A. Garifullin, and H. Zabel, Phys. Rev. Lett. **77**, 1857 (1996).

[5] T. Kontos, M. Aprili, J. Lesueur, and X. Grison, Phys. Rev. Lett. **86**, 304 (2001).

[6] V. V. Ryazanov, V. A. Oboznov, A. Yu. Rusanov, A. V. Veretennikov, A. A. Golubov, and J. Aarts, Phys. Rev. Lett. **86**, 2427 (2001).

[7] P. G. de Gennes, Phys. Lett. **23**, 10 (1966).

[8] L. R. Tagirov, Phys. Rev. Lett. **83**, 2058 (1999).

[9] A. I. Buzdin, A. V. Vedyayev, and N. V. Ryzhanova, Europhys. Lett. **48**, 686 (1999).

[10] G. Deutscher and F. Meunier, Phys. Rev. Lett. **22**, 395 (1969).

[11] J. Y. Gu, C.-Y. You, J. S. Jiang, J. Pearson, Ya. B. Bazaliy, and S. D. Bader, Phys. Rev. Lett. **89**, 267001 (2002).

[12] A. Potenza and C. H. Marrows, Phys. Rev. B **71**, 180503(R) (2005).

[13] I. C. Moraru, W. P. Pratt, Jr., and N. O. Birge, Phys. Rev. Lett. **96**, 037004 (2006).

[14] A. Yu. Rusanov, S. Habraken, and J. Aarts, Phys. Rev. B **73**, 060505(R) (2006).

[15] V. Peña, Z. Sefrioui, D. Arias, C. Leon, J. Santamaria, J. L. Martinez, S. G. E. te Velthuis, and A. Hoffmann, Phys. Rev. Lett. **94**, 057002 (2005).

[16] S. Takahashi, H. Imamura, and S. Maekawa, Phys. Rev. Lett. **82**, 3911 (1999).

[17] J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).

[18] Ya. V. Fominov, A. A. Golubov, and M. Yu. Kupriyanov, JETP Lett. **77**, 510 (2003).

[19] I. Baladié and A. Buzdin, Phys. Rev. B **67**, 014523 (2003).

[20] C.-Y. You, Ya. B. Bazaliy, J. Y. Gu, S.-J. Oh, L. M. Litvak, and S. D. Bader, Phys. Rev. B **70**, 014505 (2004).

[21] D. Y. Petrovykh, K. N. Altmann, H. Höchst, M. Laubscher, S. Maat, G. J. Mankey, and F. J. Himpsel, Appl. Phys. Lett. **73**, 3459 (1998).

[22] K. N. Altmann *et al.*, Phys. Rev. Lett. **87**, 137201 (2001) report values for $l_F^{\uparrow}$ and $l_F^{\downarrow}$ in $Ni_{90}Fe_{10}$ and $Ni_{80}Fe_{20}$. Interpolating between these two alloys, one finds approximately $l_F^{\downarrow}=0.6$ nm and $l_F^{\uparrow}>3$ nm. From the spin-scattering asymmetry in Py, $\beta=0.73$, (Ref. 23), we expect $l_F^{\uparrow}/l_F^{\downarrow}=(1+\beta)/(1-\beta)=6.4$. Using $l_F^{\downarrow}=0.6$ nm gives $l_F^{\uparrow}=4$ nm, which is consistent with the measured lower limit.

[23] S. D. Steenwyk, S. Y. Hsu, R. Loloee, J. Bass, and W. P. Pratt, Jr., J. Magn. Magn. Mater. **170**, L1 (1997).

[24] J. W. A. Robinson, S. Piano, G. Burnell, C. Bell, and M. G. Blamire, Phys. Rev. Lett. **97**, 177003 (2006).

[25] W. P. Pratt *et al.*, J. Appl. Phys. **79**, 5811 (1996).

[26] Bare Nb films in the thickness range of our data result in ρ_S [nΩ m]$=99+3415/d_s$ [nm] and T_{c0} [K]$=9.1-43/d_s$ [nm].

[27] The Einstein relation with $v_F=0.28\times10^6$ m/s[17] and $n(E_F)=1.77\times10^{48}$ J^{-1} m^{-3} from J. W. D. Connolly, Phys. Rev. **159**, 415 (1967), gives $\rho_F l_F=0.24$ fΩ m^2. C. Fierz *et al.*, J. Phys.: Condens. Matter **2**, 9701 (1990), give $\rho_F l_F=0.7-2.3$ fΩ m^2.

[28] A. Yu. Rusanov, M. Hesselberth, J. Aarts, and A. I. Buzdin, Phys. Rev. Lett. **93**, 057002 (2004).

[29] R. J. Kinsey, G. Burnell, and M. G. Blamire, IEEE Trans. Appl. Supercond. **11**, 904 (2001).

[30] R. Steiner and P. Ziemann, Phys. Rev. B **74**, 094504 (2006).