Magnetization-Dependent T_c Shift in Ferromagnet/Superconductor/Ferromagnet Trilayers with a Strong Ferromagnet

Ion C. Moraru, W. P. Pratt, Jr., and Norman O. Birge*

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

(Received 20 June 2005; published 27 January 2006)

We have measured the superconducting transition temperature T_c of Ni/Nb/Ni trilayers when the magnetizations of the two outer Ni layers are parallel (P) and antiparallel (AP). The largest difference in T_c occurs when the Nb thickness is just above the critical thickness at which superconductivity disappears completely. We have observed a difference in T_c between the P and AP states as large as 41 mK—a significant increase over earlier results in samples with higher T_c and with a CuNi alloy in place of the Ni. Our result also demonstrates that strong elemental ferromagnets are promising candidates for future investigations of ferromagnet/superconductor heterostructures.

DOI: 10.1103/PhysRevLett.96.037004 PACS numbers: 74.45.+c, 73.43.Qt, 85.25.−j, 85.75.−d

Heterostructures composed of ferromagnetic (F) and superconducting (S) materials have attracted much theoretical and experimental attention due to the rich physics produced by the interplay between competing symmetries of the order parameters [1]. In an S/F bilayer the exchange field of the ferromagnet modulates the superconducting order parameter as it decays inside the ferromagnet over a very short distance. Kontos *et al.* [2] used tunneling spectroscopy to observe the damped oscillations of the order parameter by measuring the density of states (DOS) for different thickness ferromagnets. Ryazanov *et al.* [3] observed π-state Josephson coupling in an S/F/S trilayer first by varying temperature, then later by varying the thickness of the ferromagnet [4]. Earlier, several groups [5–7] had observed oscillations in the critical temperature T_c of S/F bilayers as a function of the ferromagnet thickness d_F. Under ideal conditions T_c oscillations arise from interference between the transmitted superconducting wave function through the S/F interface and the wave reflected from the opposite surface of the ferromagnet, although in some cases alternative explanations have been proposed [8]. In many experiments, weakly ferromagnetic alloys were used in order to reduce the size of the exchange splitting in the conduction band, E_{ex}, and thus increase the penetration length ξ_F for Cooper pairs, where $\xi_F = \hbar v_F/2E_{ex}$ in the clean limit and v_F is the Fermi velocity of the ferromagnet [9].

An alternative way to probe the influence of a ferromagnet on a superconductor is to look for T_c variations in an F/S/F trilayer structure based on the mutual orientation of the two ferromagnet magnetizations [10,11]. This effect was observed [12] and later reproduced [13] in a $Cu_{1-x}Ni_x$/Nb/$Cu_{1-x}Ni_x$ system, where a weak ferromagnet was used because it is "less devastating to superconductivity." The largest difference in T_c observed between the antiparallel (AP) and parallel (P) states of the F-layer mutual magnetizations was only 6 mK when T_c was 2.8 K. Unlike other experiments [2–7] that require the ferromagnet thickness to be comparable to ξ_F, however, a positive feature of this experiment is that the T_c difference is predicted to persist even for thick F layers [10,11]. Thus it proves advantageous in studying systems with strong elemental ferromagnets, which have extremely short values of ξ_F.

Experimental studies of F/S systems with strong ferromagnets are of interest because they provide new challenges to theory, which does not yet address the full complexity of the ferromagnetic state with its different DOS and v_F of the majority and minority spin bands. Furthermore, pure elemental ferromagnets are in the clean limit, $\xi_F < l_F$ where l_F is the mean free path; this complicates use of the popular Usadel equations normally applied to the dirty limit. We are motivated to work in the limit of thick ferromagnetic layers, in anticipation of future situations where superconducting order with spin-triplet symmetry is induced in a superconductor surrounded by ferromagnets with noncollinear magnetizations [14]. (When $d_F \gg \xi_F$, the singlet component of the order parameter is completely damped.) Lastly, we wish to understand whether, in an F/S/F system with a strong ferromagnet, a large difference in T_c between the P and AP states can be achieved, as envisioned in the proposals for a superconducting spin switch [10,11]. The Ni/Nb system has been shown to be a viable candidate for experiments on F/S systems [15,16]. In this Letter we will show that Ni/Nb/Ni trilayers exhibit a significant T_c shift depending on the mutual orientation of the magnetizations of the two Ni layers.

Sets of Ni(7)/Nb(d_s)/Ni(7)/$Fe_{50}Mn_{50}$(8)/Nb(2) multilayers (all thicknesses are in nm) were directly deposited onto Si substrates by magnetically enhanced triode dc sputtering in a high vacuum chamber with a base pressure in the low 10^{-8} Torr and an Ar pressure of 2.0×10^{-3} Torr. The Ni thickness of 7 nm was chosen to be much longer than ξ_F, which we estimate to be 0.8 nm using $2E_{ex} = 0.23$ eV and $v_F = 0.28 \times 10^6$ m/s for the majority band [17]. The purpose of the FeMn is to pin the magnetization direction of the top Ni layer by exchange bias [18]. The nonsuperconducting Nb capping layer protects the FeMn from oxidation. After deposition, the samples were heated to 180 °C under vacuum, just above the blocking temperature of FeMn, and cooled in an applied field of 200 Oe in the plane of the multilayer. This procedure pins the top Ni layer while leaving the bottom Ni layer free to rotate in a small applied magnetic field.

Four-probe resistance measurements with the current in the plane of the multilayer were performed to determine T_c. Samples had lateral dimensions 4.3 mm × 1.6 mm. The T_c of each sample was defined to be the temperature at which the resistance dropped to half its normal state value. Figure 1 shows the results for T_c measurements for samples from several sputtering runs, where d_s was varied between 16–52 nm. T_c shows a strong dependence on the superconductor thickness close to a critical thickness, d_s^{cr}, where the sensitivity to ferromagnetism is enhanced. There is no superconductivity above 36 mK for $d_s < d_s^{cr} \approx 16.5$ nm.

The magnetic configuration of our structures was verified on simultaneously sputtered samples of larger lateral size, in a SQUID magnetometer. Figure 2 shows a plot of magnetization vs applied field H for a sample with $d_s =$ 18 nm taken at 100 K. The narrow hysteresis loop near $H = 0$ shows the switching behavior of the free Ni layer with a coercive field $H_c = 35$ Oe. The wider loop shows switching of the pinned layer and is shifted to nonzero H due to the exchange bias between the top Ni layer and the FeMn. Applied fields of ± 100 Oe switch the spin valve between the P and AP configurations. The nearly zero net magnetization observed at -100 Oe indicates very good AP alignment between the pinned and free Ni layers, while the nearly saturated magnetization observed at $+100$ Oe indicates good P alignment. Similarly good alignment of the P and AP states can be achieved at low temperature. The inset to Fig. 2 shows a minor hysteresis loop with $H_c \approx 50$ Oe taken at 2.29 K, which corresponds to the middle of the superconducting transition for this sample. We obtain the same behavior for temperatures above and below the transition temperature.

Measurements of T_c^{P} and T_c^{AP} were performed by alternating the applied field between $+100$ and -100 Oe, as the temperature was slowly decreased through the transition region. The largest shift in critical temperature, $\Delta T_c \equiv T_c^{\mathrm{AP}} - T_c^{\mathrm{P}}$, should occur in samples with the Nb thickness close to d_s^{cr}. Figure 3 shows a plot of R vs T for a sample with $d_s = 17$ nm, measured in a dilution refrigerator. Two distinct transitions are observed for P and AP alignment, with a separation in temperature $\Delta T_c \approx 28$ mK. A second sample with $d_s = 17$ nm showed a $\Delta T_c \approx 41$ mK, but with a slightly broader transition centered at 0.34 K. Samples with $d_s = 18$ nm and T_c between 2 and 3 K exhibit values of ΔT_c of only a few mK, similar to the CuNi/Nb/CuNi samples measured previously [12,13].

The inset to Fig. 3 shows a plot of R vs H for the first $d_s = 17$ nm sample at a temperature in the middle of the transition (0.51 K). The data clearly show well-established P and AP states at ± 100 Oe, respectively, with a difference in resistance of 1.5 Ω. Above the transition the resistance does not change perceptibly when switching from P to AP alignment. An interesting feature of the R vs H curve is the behavior of the resistance as the field is swept down from $+150$ Oe towards -50 Oe and as the field is swept up from -150 Oe towards $+50$ Oe. In both cases the resist-

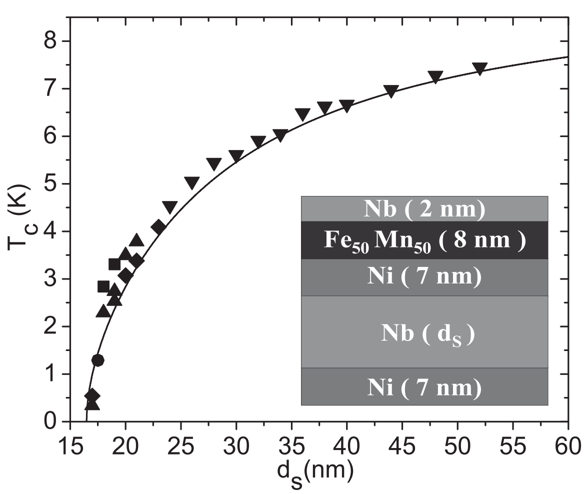

FIG. 1. Critical temperature vs Nb thickness for Ni(7)/Nb(d_s)/Ni(7)/$Fe_{50}Mn_{50}$(8)/Nb(2) samples (all thicknesses are in nm). Different symbols represent different sputtering runs. The solid line represents the theoretical fit. Inset: Schematic cross section of the samples.

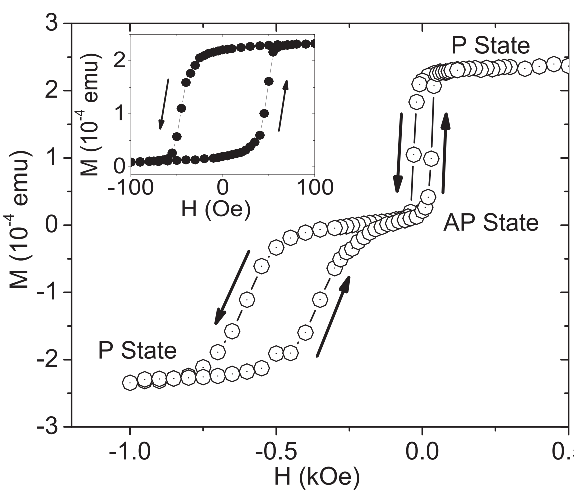

FIG. 2. Magnetization vs applied field for a $d_s = 18$ nm sample measured at $T = 100$ K. At ± 50 Oe the free bottom Ni layer switches while the pinned top Ni layer switches at -500 Oe. Inset: minor loop measured at $T = 2.29$ K showing the switching of the free Ni layer.

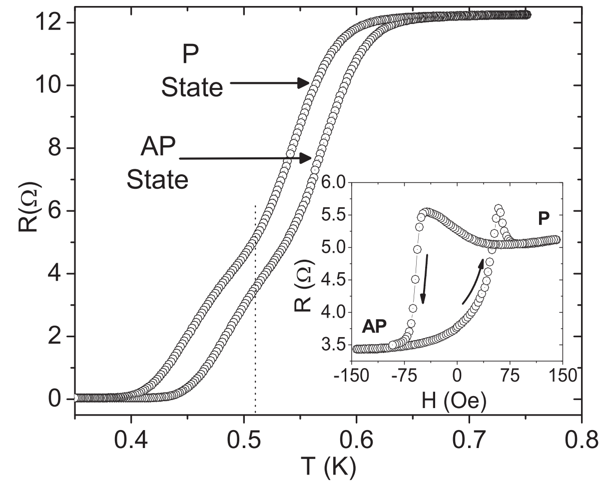

FIG. 3. Resistance vs temperature for the P and AP states of a $d_s = 17$ nm sample measured in ± 100 Oe. Two distinct transitions are observed, with $\Delta T_c = 28$ mK. Inset: Resistance vs applied field at $T = 0.51$ K (dotted line in main graph).

ance increases to a value higher than that of the P state after the field passes through zero. We believe this behavior involves the breaking of the free ferromagnetic layer into domains when $H \approx H_c$. The domain-wall fringe fields penetrate the superconductor, thus suppressing T_c slightly and producing a higher resistance. Note that this effect is *opposite* to that observed by other groups [19,20], where inhomogeneous magnetization led to enhanced superconductivity in F/S bilayers. In those experiments, the domain size must be smaller than the superconducting coherence length so that the Cooper pairs sample multiple domains [21], and the magnetic field penetrating into the superconductor must be small.

The critical temperature of F/S/F trilayers in the P and AP states has been calculated theoretically by several groups [10,11,22–24]. Since many experiments employ ferromagnetic alloys, the usual approach involves solving the Usadel equations in the dirty limit for both the superconductor and the ferromagnet. (The dirty limit applies to S when $l_S < \xi_{BCS} = \hbar v_S \gamma / \pi^2 k_B T_{c0}$, and to F when $l_F < \xi_F$, where l_S and l_F are the electron mean free paths in S and F, and T_{c0} is the transition temperature of the bulk superconductor.) In our case, however, the ferromagnetic metal is both pure and strong, thus in the clean limit $l_F > \xi_F$. Hence we use the theory of [11] as modified in section 3.2 of [25] to make it more appropriate for the clean limit. This theory does not, however, incorporate a full description of the majority and minority spin bands of a strong ferromagnet, with different DOS, v_F, and transmission coefficients. The expression for the normalized critical temperature of the trilayer is

$$\ln t_c + \mathrm{Re}\Psi\left(\frac{1}{2} + \frac{2\phi^2}{t_c (d_s/\xi_S)^2}\right) - \Psi\left(\frac{1}{2}\right) = 0, \qquad (1)$$

where $t_c \equiv T_c/T_{c0}$ and T_{c0} is the critical temperature of an isolated Nb film of the same thickness as the one in the trilayer. The function ϕ is determined from the condition $\phi \tan\phi = R$ for the P state or $(\phi \tan\phi - R')(R' \tan\phi + \phi) - (R'')^2 \tan\phi = 0$ for the AP state, where the complex function $R = R' + iR''$ is given by

$$R = \frac{d_s}{\xi_S} \frac{N_F v_F \xi_S}{2N_S D_S} \frac{1}{\sqrt{1 - i\xi_F/l_F} + 2/T_F}. \qquad (2)$$

Equation (2) is valid when the ferromagnets are thick enough so that the tanh functions in [25] can be set to 1. This assumption is validated by data on Nb/Ni bilayers [16] where oscillations in $T_c(d_F)$ are completely damped for $d_F > 4$ nm. The dimensionless parameters that enter into this theory are the ratios d_s/ξ_S, ξ_F/l_F, the S/F interface transparency T_F, and the combination $N_F v_F \xi_S / 2N_S D_S$. N_F and N_S are the densities of states at the Fermi energy of the F and S layers, v_F is the Fermi velocity of the ferromagnet, and D_S is the diffusion constant of the superconductor.

To avoid fitting the data with four free parameters, we follow the strategy outlined by Lazar *et al.* [8] and by Sidorenko *et al.* [16]. We determine the superconducting coherence length, ξ_S, from measurements of the critical field vs temperature of isolated Nb films, with the magnetic field applied perpendicular to the film plane. For films in the thickness range 20–50 nm, the values of ξ_S are close to 6 nm, which we use for our fits [26]. From the asymptotic form of Eq. (1) as $t_c \to 0$, one finds $2\phi^2/(d_s^{cr}/\xi_S)^2 = 1/4\gamma$, where $\gamma = 1.781$. Substituting $d_s^{cr} \approx 16.5$ nm and using Eq. (2) (while ignoring the small imaginary term), we obtain the constraint

$$\frac{N_F v_F \xi_S}{2N_S D_S (d_s^{cr})} \frac{1}{1 + 2/T_F} \approx \frac{\phi^{cr} \tan\phi^{cr}}{(d_s^{cr}/\xi_s)} = 0.24. \qquad (3)$$

Estimates of the product $N_F v_F$ vary substantially in the literature. From [17,27], we obtain respectively $N_F = 1.77 \times 10^{48}\ \mathrm{J^{-1}\,m^{-3}}$ and $v_F = 0.28 \times 10^6$ m/s. Fierz *et al.* [28], however, quote $\rho_F l_F = 0.7$–$2.3\ \mathrm{f\Omega\,m^2}$ for Ni, which when combined with the Einstein relation $1/\rho_F l_F = N_F v_F e^2/3$, implies values 3–10 times smaller for $N_F v_F$. Combining these values with $N_S = 5.31 \times 10^{47}\ \mathrm{J^{-1}\,m^{-3}}$ [29] and using our measured $D_S(d_s^{cr}) = 2.8 \times 10^{-4}\ \mathrm{m^2/s}$, we obtain $T_F = 0.05$–0.6. The bulk resistivity of our sputtered Ni films at 4.2 K is $\rho_F = 33\ \mathrm{n\Omega\,m}$, which leads to values of l_F between 7 and 70 nm, given the range in $\rho_F l_F$ quoted above. Since the Ni used in our trilayers is thin, l_F is probably limited by surface scattering, so we use the lower estimate $l_F = 7$ nm, hence $\xi_F/l_F \approx 0.1$. In fact, the fit to $T_c(d_s)$ is quite insensitive to the values of T_F and ξ_F/l_F. We used $\xi_F/l_F = 0.1$ and $T_F = 0.3$ to obtain the curve shown in Fig. 1, which fits the data remarkably well.

A more stringent test of the theory is the prediction of ΔT_c, which depends sensitively on both T_F and ξ_F/l_F. Thickness deviations from nominal values produce scatter

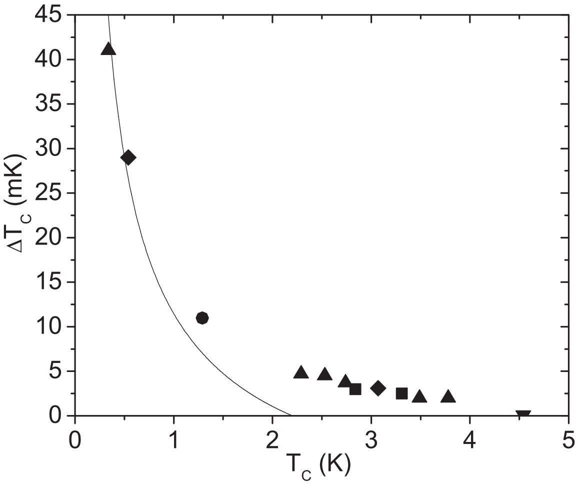

FIG. 4. Symbols: ΔT_c vs T_c for our 11 thinnest samples. The line represents a fit using $\xi_F/l_F = 0.7$ and $T_F = 1.0$, values larger than our best estimates.

in plots of T_c or ΔT_c vs d_s, therefore Fig. 4 shows a plot of ΔT_c versus T_c. If we calculate ΔT_c using our best estimate of ξ_F/l_F and the upper limit of T_F given above, the maximum value of ΔT_c is only a few mK when T_c is well below 1 K—hardly visible on Fig. 4. If we relax the constraints we have placed on the parameters, and instead try to produce the best fit to the $\Delta T_c(d_s)$ data, we find that a reasonable fit can be obtained when ξ_F/l_F is allowed to be much larger than our original estimate. Figure 4 shows a fit using $\xi_F/l_F = 0.7$ and $T_F = 1.0$. Similar curves can be produced by simultaneously varying ξ_F/l_F and T_F while keeping their product nearly constant. Fitting the ΔT_c data requires letting ξ_F/l_F exceed our estimate substantially. Our l_F estimate may be too large, because the resistivity is dominated by the longer of the majority or minority band l_F, whereas the F/S proximity effect depends on the shorter of the two [8]. A shorter l_F is also implied by the observation of complete damping of T_c oscillations in Nb/Ni bilayers for $d_F > 4$ nm [16]. Nevertheless, producing a reasonable fit to our ΔT_c data entails either increasing ξ_F/l_F beyond the clean limit, or increasing T_F beyond our original estimate.

In conclusion, we have observed a large difference in T_c between the P and AP magnetic states of Ni/Nb/Ni trilayers, with $T_c^P < T_c^{AP}$. Recently, Ruzanov *et al.* [30] reported a T_c difference between the P and AP states of a $Ni_{0.8}Fe_{0.2}/Nb/Ni_{0.8}Fe_{0.2}$ trilayer, but with $T_c^P > T_c^{AP}$. Understanding these opposing behaviors in F/S systems with strong ferromagnets will require further experiments, as well as theoretical models able to account for the complexity of real ferromagnets [31].

We are grateful to R. Loloee and J. Bass for fruitful discussions. This work was supported by NSF Grants No. DMR 9809688, No. 0405238, and by the Keck Microfabrication Facility.

*Electronic address: birge@pa.msu.edu

[1] For a review, see Yu. A. Izyumov, Yu. N. Proshin, and M. G. Khusainov, Phys. Usp. **45**, 109 (2002).
[2] T. Kontos, M. Aprili, J. Lesueur, and X. Grison, Phys. Rev. Lett. **86**, 304 (2001).
[3] V. V. Ryazanov *et al.*, Phys. Rev. Lett. **86**, 2427 (2001).
[4] V. V. Ryazanov *et al.*, J. Low Temp. Phys. **136**, 385 (2004).
[5] J. S. Jiang, D. Davidovic, D. H. Reich, and C. L. Chien, Phys. Rev. Lett. **74**, 314 (1995).
[6] L. V. Mercaldo *et al.*, Phys. Rev. B **53**, 14 040 (1996).
[7] Th. Mühge *et al.*, Phys. Rev. Lett. **77**, 1857 (1996).
[8] L. Lazar *et al.*, Phys. Rev. B **61**, 3711 (2000).
[9] The experiments in Refs. [2–4] were in the dirty limit, so $\xi_F = \sqrt{\hbar D_F/2E_{ex}}$, where D_F is the diffusion constant.
[10] A. I. Buzdin, A. V. Vedyayev, and N. V. Ryzhanova, Europhys. Lett. **48**, 686 (1999).
[11] L. R. Tagirov, Phys. Rev. Lett. **83**, 2058 (1999).
[12] J. Y. Gu *et al.*, Phys. Rev. Lett. **89**, 267001 (2002).
[13] A. Potenza and C. H. Marrows, Phys. Rev. B **71**, 180503(R) (2005).
[14] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. B **69**, 174504 (2004).
[15] Y. Blum, A. Tsukernik, M. Karpovski, and A. Palevski, Phys. Rev. Lett. **89**, 187004 (2002).
[16] A. S. Sidorenko *et al.*, Ann. Phys. (N.Y.) **12**, 37 (2003).
[17] D. Y. Petrovykh *et al.*, Appl. Phys. Lett. **73**, 3459 (1998).
[18] J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).
[19] A. Yu. Rusanov, M. Hesselberth, J. Aarts, and A. I. Buzdin, Phys. Rev. Lett. **93**, 057002 (2004).
[20] R. J. Kinsey, G. Burnell, and M. G. Blamire, IEEE Trans. Appl. Supercond. **11**, 904 (2001).
[21] T. Champel and M. Eschrig, Phys. Rev. B **71**, 220506(R) (2005).
[22] C.-Y. You *et al.*, Phys. Rev. B **70**, 014505 (2004).
[23] Ya. V. Fominov, A. A. Golubov, and M. Yu. Kupriyanov, JETP Lett. **77**, 510 (2003).
[24] I. Baladié and A. Buzdin, Phys. Rev. B **67**, 014523 (2003).
[25] L. R. Tagirov, Physica C (Amsterdam) **307**, 145 (1998).
[26] From $\xi_S = \sqrt{\hbar D_S/2\pi k_B T_{c0}}$, we obtain $\xi_S = 7$–8 nm for $d_s = 20$–50 nm, where D_S is determined from resistivity measurements and T_{c0} is the critical temperature of isolated Nb films. We find D_S $(\mathrm{m^2/s}) = 1.8 \times 10^{-4} + 5.9 \times 10^{-6} d_s$ and $T_{c0}(\mathrm{K}) = 9.1$–$43/d_s$, with d_s in nm.
[27] J. W. D. Connolly, Phys. Rev. **159**, 415 (1967).
[28] C. Fierz *et al.*, J. Phys. Condens. Matter **2**, 9701 (1990).
[29] A. R. Jani, N. E. Brener, and J. Callaway, Phys. Rev. B **38**, 9425 (1988).
[30] A. Yu. Rusanov, S. Habraken, and J. Aarts, cond-mat/0509156.
[31] B. P. Vodopyanov and L. R. Tagirov, JETP Lett. **78**, 555 (2003).