Magnetization-Orientation Dependence of the Superconducting Transition Temperature in the Ferromagnet-Superconductor-Ferromagnet System: CuNi/Nb/CuNi

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The superconducting critical temperature (T_c) of ferromagnet-superconductor-ferromagnet systems has been predicted to exhibit a dependence on the magnetization orientation of the ferromagnetic layers such that $T_c^{\rm AP} > T_c^P$ for parallel (P) and antiparallel (AP) configurations of the two ferromagnetic layers. We have grown CuNi/Nb/CuNi films via magnetron sputtering and confirmed the theoretical prediction by measuring the resistance of the system as a function of temperature and magnetic field. We find an ~25% resistance drop occurs near T_c in $Cu_{0.47}Ni_{0.53}$(5 nm)/Nb(18)/CuNi(5) when the two CuNi layers change their magnetization directions from parallel to antiparallel, whereas there is no corresponding resistance change in the normal state.

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Proximity effects between a superconductor (S) and a ferromagnet (F) have been intensively studied [1]. F/S artificial superlattices provide the possibility of controlled studies of the interplay between superconductivity and ferromagnetism [2]. One of the most unusual effects observed in F/S structures is the nonmonotonic dependence of the superconducting critical temperature, T_c, on the thickness of the ferromagnetic layer, t_F. This has been studied experimentally for various systems, such as Nb/Gd multilayers [3], Nb/CuNi bilayers [4], Fe/V/Fe trilayers [5], and Fe/Pb/Fe trilayers [6]. Recently structures with two F layers and one S layer have been discussed theoretically in connection with magnetoresistive memory elements [7] or superconductive spin switches [8,9]. An $F/F/S$ structure was proposed in Ref. [7] and $F/S/F$ structures in Refs. [8,9]. In both cases F layers suppress the T_c of the S layer, but the magnitude of the suppression depends on the relative magnetization orientation of the two F layers. The magnetization orientation of the $F/S/F$ structure can be controlled by a weak magnetic field (H) which by itself is insufficient to destroy superconductivity. Specifically it was proposed that the $F/S/F$ system can be switched between the superconducting and normal states at a temperature (T) between T_c^P and $T_c^{\rm AP}$, T_c for parallel (P) and antiparallel (AP) configurations of the two F layers, respectively. Efforts have been reported for the system $F/I/S/I/F$ with an insulating layer, I, between F and S layers [10]. In the present Letter, we report the first experimental observation of $T_c^{\rm AP} > T_c^P$ in an all-metallic $F/S/F$ system.

Recently, a number of approaches have been used to calculate the T_c value of S/F bilayer systems [4,11] especially to understand the nonmonotonic behavior of $T_c(t_F)$. $\Delta T_c = T_c^{\rm AP} - T_c^P$, in the $F/S/F$ sandwich system, has been predicted similarly using calculations based on the solution of the Usadel's equations under a strong exchange field in the two F layers for P or AP configurations [8,9,12]. ΔT_c can be affected by the transparency of the S/F interface [13] for Cooper pairs, the magnitude of the magnetic exchange energy in F layer [4], and the spin-orbit scattering [14]. ΔT_c between the AP and P states can be qualitatively understood utilizing the simple fact that Cooper pairs consist of two electrons with opposite spin directions. Pair-breaking effects due to the spin-polarized electrons extending into the S layer from F layers are weaker in the AP-aligned configuration since spin polarizations from both F layers are of opposite signs and cancel each other. On the other hand, pair-breaking effects are stronger when P-aligned because the spin polarizations from both F layers are of the same sign, so that the destruction of the superconductivity is enhanced. Therefore, for certain conditions, an $F/S/F$ structure can have either zero or a lower value of T_c for the P case than for the AP case. A schematic of such an $F/S/F$ proximity switch device is illustrated in Fig. 1(a).

We chose dilute ferromagnetic Cu_xNi_{1-x} alloys ($x \leq 0.6$) as the F layer. CuNi was similarly used previously for the study of $S/F/S$ junctions [15,16] and S/F bilayers [4], since its weak ferromagnetism is less devastating to superconductivity. Sputtering targets of $x = 0.4$ and 0.5 were made by pressing a mixture of Cu and Ni powders. The Curie temperature, T_C, of sputtered CuNi films is ~70 K and ~30 K for $x = 0.4$ and 0.5, respectively, based on the temperature dependence of the magnetization, $M(T)$. Magnetic hysteresis measurements at 5 K show a coercive field of ~100 Oe. According to the dependence of T_C on x [17] for Cu_xNi_{1-x} alloys, the Cu compositions of the thin films are expected to be 0.47 (instead of $x = 0.4$) and 0.49 ($x = 0.5$), which are slightly different from the target compositions. In order to get well-defined P and AP alignments between two CuNi layers, we initially employed an exchange-biased spin-valve stack of CuNi/Nb/CuNi/$Fe_{50}Mn_{50}$. The antiferromagnetic (AF) FeMn layer pins the magnetization of the adjacent CuNi layer via exchange bias so that it remains fixed in weak magnetic fields that can reorient

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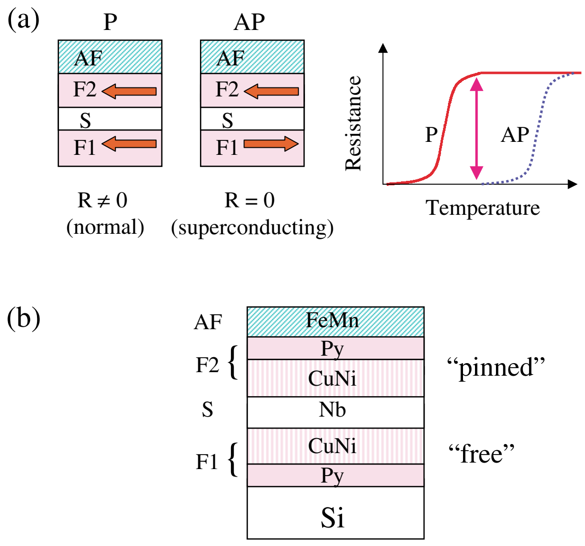

FIG. 1 (color online). (a) Schematic structure of an $F/S/F$/AF proximity switch device. (b) Sample structure of an exchange-biased spin valve, Py/CuNi/Nb/CuNi/Py/FeMn. As shown by the arrow in (a), resistance can change from a finite value to zero at $T_c^P < T < T_c^{\mathrm{AP}}$.

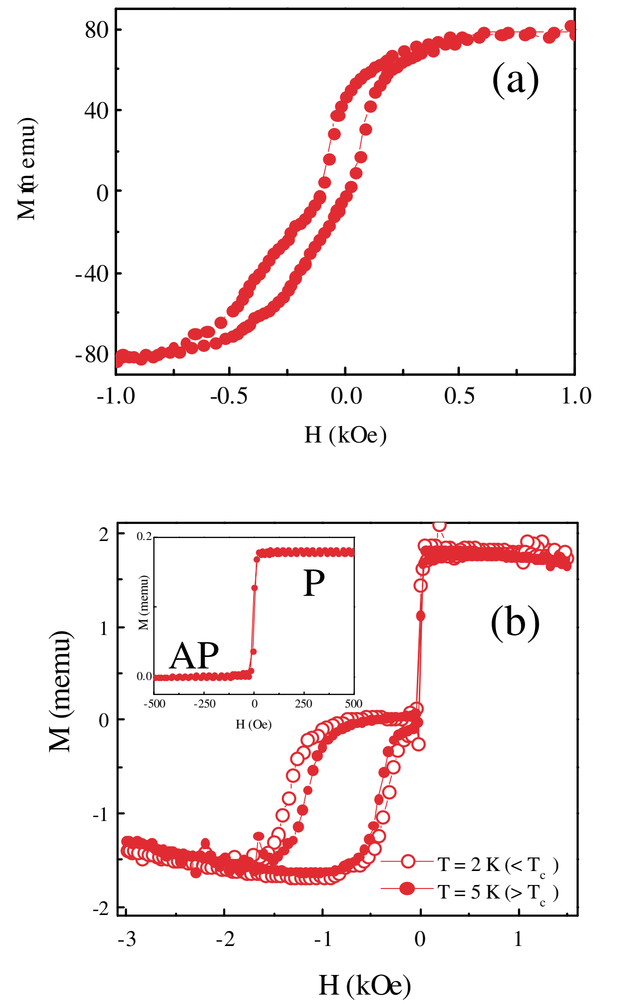

FIG. 2 (color online). (a) $M(H)$ of $Cu_{0.47}Ni_{0.53}$(20 nm)/Nb(20)/CuNi(20)/FeMn(8). (b) $M(H)$ of Py(4 nm)/$Cu_{0.47}Ni_{0.53}$(5)/Nb(18)/CuNi(5)/Py(4)/FeMn(6). Empty and filled circles in (b) denote data measured at $T = 2$ K ($< T_c$) and $T = 5$ K ($> T_c$), respectively. A minor loop measured between ± 500 Oe is shown in the inset of (b).

the magnetization of the "free" CuNi layer, allowing control of the P and AP configurations of the two F layers.

However, a problem we encountered was that we could not get a very well-defined AP region due to the rather large coercivity of the CuNi layer compared to the exchange-bias field, H_E, as shown in Fig. 2(a). The coercive field of the CuNi layer is about 100 Oe and the H_E is only about ~150–200 Oe. Therefore, the two hysteresis curves of the free and pinned CuNi layers overlap. To solve this problem, we inserted a soft ferromagnetic layer of permalloy (Py = $Ni_{82}Fe_{18}$) adjacent to the CuNi layers to yield Py/CuNi/Nb/CuNi/Py/$Fe_{50}Mn_{50}$ multilayers, as shown in Fig. 1(b). The Py layer decreases the coercivity of the CuNi layer and creates a range of fields where an AP alignment of the two CuNi layers is well defined. It is known that if both the Py and CuNi layer are thin, their magnetic moments will couple and reverse together under an external field due to the strong direct exchange interaction at the interface [18]. Therefore, coupling with the Py layer reduces the field required to switch the CuNi layer. A multilayer, starting with the bottom Py and ending with the FeMn layer, was deposited onto a Si substrate using a high vacuum chamber with a base pressure of 10^{-8} Torr and an Ar pressure of ~1.5–4 mTorr. The thicknesses of the CuNi, t_{CuNi}, and Nb, t_{Nb}, were varied in the range of $0 \leq t_{\mathrm{CuNi}} \leq 20$ nm and $18 \leq t_{\mathrm{Nb}} \leq 35$ nm. To set the exchange bias, the multilayers were heated to 370 K and cooled through the Neel temperature of the FeMn layer in a magnetic field of 1 T.

Figure 2(b) shows the $M(H)$ curves of Py(4 nm)/$Cu_{0.47}Ni_{0.53}$(5)/Nb(18)/CuNi(5)/Py(4)/FeMn(6) measured at 5 K ($> T_c \approx 2.81$ K) and 2 K ($< T_c$). The hysteresis loop of the top CuNi/Py layer is shifted due to the exchange bias between the Py and FeMn layers. The hysteresis loops measured in the normal and superconducting states do not show much difference and both have well-defined P and AP configurations. A minor loop measured between ± 500 Oe is shown in the inset, where P and AP states between two CuNi layers are well defined [19]. The fact that the net magnetization value at the AP configuration is zero also suggests that the top and bottom Py/CuNi layers are aligned in opposite directions and the magnetizations cancel. For the resistance (R) measurement, +300 and −300 Oe are used to establish the P and AP alignments, respectively. As shown in the $M(H)$ curve, ± 300 Oe is enough to create a single domain configuration, so that we can neglect the magnetic stray-field effect which exists only at the edge of the sample in our case.

Figure 3(a) shows the $R(H)$ data of the Py(4 nm)/$Cu_{0.47}Ni_{0.53}$(5)/Nb(18)/CuNi(5)/Py(4)/FeMn(6) measured both at $T > T_c$ (5 K) and $T \sim T_c$ (2.81 K) and

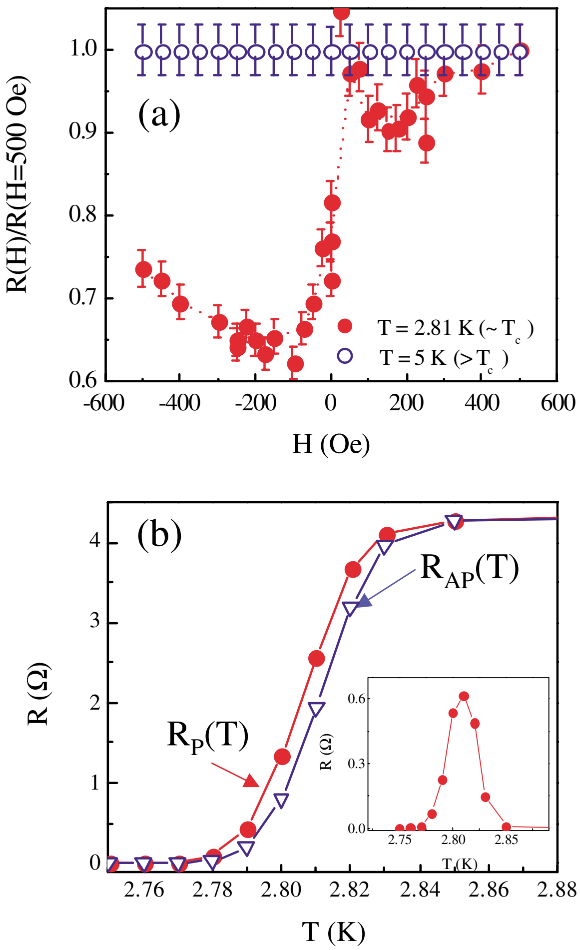

FIG. 3 (color online). (a) $R(H)/R(500\ \mathrm{Oe})$ curves at $T = 5$ K ($> T_c$) and $T = 2.81$ K ($\sim T_c$) and (b) $R_P(300\ \mathrm{Oe}, T)$ and $R_{\mathrm{AP}}(-300\ \mathrm{Oe}, T)$ for Py(4 nm)/$Cu_{0.47}Ni_{0.53}$(5)/Nb(18)/CuNi(5)/Py(4)/FeMn(6). $\Delta R(T) = R_P(T) - R_{\mathrm{AP}}(T)$ is shown in the inset of (b).

normalized at $R(H = 500\ \mathrm{Oe})$. For $T > T_c$, the S layer is in the normal state and $R(H)$ does not change between ± 500 Oe, indicating that R is not affected by whether the two CuNi layers are aligned P or AP. However, when the S layer enters into the superconducting state at $T \leq T_c$, $R(H)$ shows a dramatic change during alternating the configuration of the two CuNi layers between P and AP. We observe that R decreases when the field goes from positive (P configuration) to negative (AP configuration): $R_{\mathrm{AP}} < R_P$, where R_P and R_{AP} are the resistances measured at $H = 300$ and -300 Oe, respectively. The sign of this change is consistent with the fact that AP configuration has a higher T_c. Indeed, in the idealized situation of infinitely sharp superconducting transitions, theoretical calculations [8,9] predict that R would decrease from a finite value to zero in a temperature region between T_c^{AP} and T_c^P, as illustrated by an arrow in Fig. 1(a). However, only partial resistance reduction is observed on $R(T)$ in our experiment as shown in Fig. 3(b) since the transition width is finite and T_c shift is rather small. We find an $\sim$25% change of R ($\{[R(300\ \mathrm{Oe}) - R(-300\ \mathrm{Oe})]/R(500\ \mathrm{Oe})\} \times 100\%$) upon switching from P to AP alignment. We made measurements of $R_P(T) = R(300\ \mathrm{Oe}, T)$ and $R_{\mathrm{AP}}(T) = R(-300\ \mathrm{Oe}, T)$ values. It is important to note that the methodology of making measurements at fixed T while reversing the field ensures the accuracy of the ΔR values: there is virtually no T drift during the magnetization switch. The T dependence of $\Delta R = R_P - R_{\mathrm{AP}}$ is shown on the inset of Fig. 3(b). The curve is smooth and shows nonzero values within the T_c width (~ 0.09 K). Both characteristics of $R(H)$ and $\Delta R(T)$ are consistent with the picture that two $R(T)$ curves in different magnetization configurations shifted by a small ΔT value. The $R_P(T)$ and $R_{\mathrm{AP}}(T)$ curves themselves are shown in Fig. 3(b). From ΔR value at the middle of the transition and the slope of $R_P(T)$ [or $R_{\mathrm{AP}}(T)$], we find $\Delta T_c \approx 6$ mK for $t_{\mathrm{CuNi}} = 5$ nm and $t_{\mathrm{Nb}} = 18$ nm. Since the ΔT_c value is smaller than the width of T_c, we were not able to observe a situation where the system is normal in the P configuration and becomes fully superconducting in the AP configuration. Figure 4 shows the average $\overline{T_c} = (T_c^{\mathrm{AP}} + T_c^P)/2$ and ΔT_c measured for samples with various t_{CuNi} but fixed value of $t_{\mathrm{Nb}} = 19$ nm.

The proximity effect in $F/S/F$ systems is usually described by the Usadel equations [20] valid in the limit of small mean free path [4,8,9,21]. The strength of the superconducting interaction in the S layer is characterized by the T_c of the single S layer without F layers, T_{c0}. The ferromagnetic layers are modeled by introducing spin split bands described by the mean field exchange constant, I. Transport in the S and F layers is characterized by the electronic diffusion coefficients D_S and D_F, respectively. It is assumed that I is large enough to prevent any influence of superconductivity on magnetism (a theory for the opposite case was considered in Ref. [22]). The boundary conditions involve two

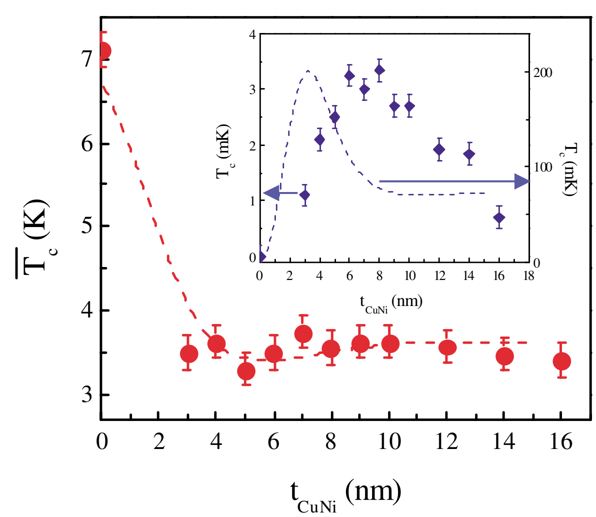

FIG. 4 (color online). $\overline{T_c}(t_{\mathrm{CuNi}})$ for Py(4 nm)/$Cu_{0.49}Ni_{0.51}$(t_{CuNi})/Nb(19)/CuNi(t_{CuNi})/Py(4)/FeMn(6). $\Delta T_c(t_{\mathrm{CuNi}})$ is shown in the inset. Experimental data and theoretical calculations are plotted as symbols and dashed lines, respectively.

parameters: the band structure mismatch, γ, and the barrier strength, γ_b (see notation in Ref. [4]). The T_c is found from the system of Usadel equations on the anomalous Green's function and a self-consistency condition on the order parameter. In some parts of the $\{t_S, t_F, D_S, D_F, T_{c0}, I, \gamma, \gamma_b\}$ parameter space, approximations (e.g., a "single mode approximation" [23]) are possible and analytic solutions for T_c can be found [8,9]. But, as noted in Ref. [4], experiments, including ours, usually do not correspond to any of the limiting cases and numeric solution is necessary. To analyze experiments we generalized the numeric solution procedure for the case of an F/S bilayer described in Ref. [4] to study the trilayer case without further approximations. We calculated $\overline{T_c}$ and ΔT_c of our $F/S/F$ system using a method similar to the one of Ref. [4] and the results are plotted in Fig. 4 as dashed curves along with the experimental data points [24]. We find good agreement between the experimental data and theoretical prediction for $\overline{T_c}(t_F)$. There is also qualitative agreement between the data and calculation for $\Delta T_c(t_F)$ [25]. The fact that the measured $\overline{T_c}(t_F)$ dependence agrees with the theory but $\Delta T_c(t_F)$ is off by $\sim 10^2$ might indicate that either we did not find a proper point in the parameter space of the theory, or that the theory does not capture all the features of the proximity effect. ΔT_c of the opposite sign was predicted in Ref. [26] which cannot be obtained in the present framework [8,9].

The small value of ΔT_c in our system compared to theory could be due to several reasons. First, there is only a small region in t_F giving a large value of ΔT_c, for example, $t_F \sim 0.5\sqrt{4\hbar D_F/I}$ [8,25]. Therefore, t_F and t_S should be optimized to get a large value of ΔT_c. Second, when the interface transparency decreases, ΔT_c becomes smaller [8,25]. The interface transparency is affected by the interface quality. Third, the two CuNi layers may not be identical. This asymmetry makes the value of ΔT_c smaller since the cancellation of the pair-breaking effect will not be perfect in AP alignment if the two CuNi layers are not the same. Fourth, the contribution from the Py layer adjacent to the CuNi layer should be taken into account especially for samples with small values of t_{CuNi}. Finally, local mechanical strain and compositional fluctuations can have an effect on the magnetic behavior of the CuNi layers, whose net composition is close to the onset of ferromagnetism.

In conclusion, we employed an exchange-biased spin valve, Py/CuNi/Nb/CuNi/Py/FeMn, to investigate the dependence of T_c on the magnetization orientation of the two ferromagnetic CuNi layers that sandwich a superconducting Nb layer. We observe experimentally that $T_c^{\mathrm{AP}} > T_c^{P}$. Optimization of the value of ΔT_c and a better theoretical understanding are warranted to obtain further insights and to explore practical applications.

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