Evidence for Zero- and π-Phase Order Parameters of Superconducting Nb/Co Tri- and Pentalayers from the Oscillatory Behavior of the Transition Temperature

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The oscillation behavior of the superconducting transition temperature T_c as a function of the ferromagnetic Co layer thickness (d_{Co}) has been examined for Nb/Co superconductor(S)/ferromagnetic(F) trilayer series (F/S/F) and pentalayer series (F/S/F/S/F). T_c of the pentalayer series takes a local maximum between $d_{\mathrm{Co}} = 2.0$–3.2 nm, where T_c of the trilayer shows a local minimum. This difference in the T_c versus d_{Co} curves provides a clear evidence for the occurrence of the π phase in the pentalayers, which has been theoretically predicted by Buzdin *et al.*, Radović *et al.*, and Tagirov.

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For several decades, the superconducting properties of superconductor(S)/ferromagnet(F) layered materials have attracted much interest due to the peculiar oscillation behavior of the superconducting transition temperature T_c when plotted as a function of the ferromagnetic layer thickness d_F. This phenomenon has not been observed in S/nonmagnetic material(N) layered systems, where T_c monotonically decreases as a function of the N-layer thickness d_N. In S/F layers the superconducting pair distribution function $F(z)$ is modulated by the exchange potential E_{ex} in the ferromagnetic layer. At the boundary of S and F layers, $F(z)$ penetrating into the F layer interferes with that reflected from the opposite boundary of the F layer. As a result, the modulation of $F(z)$ in the F layer depends on d_F, yielding the oscillation of T_c [1]. Buzdin *et al.* [2] predicted the appearance of the π-phase superconducting state for the S/F multilayer; the pairing function $F(z)$ can stably take a phase factor difference of π between the neighboring superconducting layers (we call this state "π-phase state" or "π-phase scheme," hereafter), that is,

$$F(z + \lambda) = F(z)\exp(-i\pi), \tag{1}$$

z being the coordination vector directed perpendicular to the layer plane and $\lambda = d_S + d_F$.

Several experimental works on the T_c oscillation effect of S/F layers have been reported so far. These experiments can be classified into two categories on the basis of the number of the constituent S layers. One is the case in which only a single S layer exists and the other is the case in which two or more S layers are included. The bilayers or trilayers (F/S/F) such as Nb/Fe [3,4], Nb/Ni [1], Nb/(Fe/Cu) [5], V/Fe [6], Pb/Fe [7] correspond to the former case, while the trilayers (S/F/S) with surface S layers or multilayers such as Nb/Gd [8,9], Nb/CuMn [10,11], Nb/Co [12,13], V/Co [12] correspond to the latter case. All the S/F layers listed above show the oscillation phenomenon with at least one minimum and one peak, except for the case of V/Fe which shows the reentrant behavior.

As for the Nb/Fe bilayer belonging to the former type, Mühge *et al.* attributed the origin of the T_c oscillation to the change of the magnetic nature of the Fe sublayer correlated with the sublayer thickness. Sidorenko *et al.* [1] analyzed the oscillation of the Nb/Ni bilayer based on the inhomogeneous superconducting pairing like the Fulde-Ferrell-Larkin-Ovchenko state, where the proximity effect [14,15] was assumed to be essential for the oscillation. In the Nb/(Fe/Cu) bilayer, Vélez *et al.* [5] reported that, by changing d_F, Fe structure in the (Fe/Cu) sublayer transforms from fcc to bcc, which results in the oscillation of T_c due to the change in the spin flip scattering strength. For the V/Fe bilayer [6], the reentrant behavior was also explained by the proximity effect. For the Fe/Pb/Fe trilayer [7], Tagirov's proximity theory was applied.

As for the latter type systems with two or more S layers, Jiang *et al.* [8] analyzed their experimental curve of Nb/Gd based on the Radović's proximity effect theory taking account of the π-phase scheme. They did not fully agree with the π-phase scheme, emphasizing the importance of the magnetic fluctuations. Mercaldo *et al.* [10] and Attanasio *et al.* [11] found the T_c oscillation for Nb/CuMn with the CuMn sublayer being in a spin glass state and supported the π-phase scheme. In our previous paper [12], we observed the double minimum in T_c versus d_{Co} for the Nb/Co and V/Co multilayers, where experimental results were explained to come from the following three different type origins: (i) inelastic electron scattering by the virtual level in intermixed nonferromagnetic NbCo alloy layers, (ii) magnetic spin flip scattering by ferromagnetic Co layers which are not in so rigidly ferromagnetic due to the two-dimensional character of magnetization, and (iii) the strong pair breaking mechanism due to the mean-field exchange potential. In the paper, we concluded that the first minimum may originate from the mechanisms (i) and (ii), and the second minimum may be due to the mechanism (iii). More recently, Bagrets *et al.* [16] reinterpreted T_c oscillation of the Nb/Co multilayers [12] based on the Gor'kov equation without taking account of the π phase.

Theoretically, Buzdin [2], Radović [17,18], and also Tagirov [19] have led the T_c oscillation behavior as a function of d_F, taking account of the possibility of the π phase. The basic theoretical approach is that in S/F layers the ferromagnetic spin of the F layer yields the pair breaking effect on the Cooper pairs which penetrate into the F layer. The pair breaking parameter ρ is introduced into the equation, from which T_c is determined [2,17,19],

$$\ln(t_c) = \Psi(1/2) - \mathrm{Re}\Psi(1/2 + \rho/t_c), \tag{2}$$

where t_c is the reduced superconducting transition temperature ($= T_c/T_{c0}$) with T_{c0} being the transition temperature for the isolated S layer, Ψ the digamma function, $\mathrm{Re}\Psi$ means the real part of Ψ, and $\rho = k_S^2\xi_S^2/2$ with k_S being the propagation momentum of the pairing wave function in the superconducting layer, and ξ_S is the superconducting coherence length ($= (\hbar D_S/2\pi k_B T_{c0})^{1/2}$ with D_S being the electron diffusion constant of the S layer). The k_S value is determined from the matching condition for the pairing functions between S and F layers at the interface [1,19] as

$$k_S d_S \tan(k_S d_S) = \left(\frac{3d_S}{l_S}\right)\left(\frac{N_F v_F}{N_S v_S}\right) \times \frac{\tanh(k_F d_F)}{i\xi_F k_F + (2/T_M)\tanh(k_F d_F)} \tag{3}$$

for the 0 phase, where l_S is the electron mean free path in the S layer, N_F (N_S) the density of state of the conduction electron at the Fermi level in the F (S) layer, v_F (v_S) the Fermi velocity in the F (S) layer, $k_F = \sqrt{i\xi_F/l_F - 1}/\xi_F$ is the propagation momentum of the pairing wave function in the F layer with l_F being the electron mean free path in the F layer, $\xi_F = \hbar v_F/E_{ex}$ the magnetic stiffness length in the F layer, and T_M is the interface transparency parameter. For the π phase, the matching condition is also obtained by using the relation of Eq. (1) [18,20]. For the T_c versus d_F curve, the calculation predicts the different maximum position of T_c between the 0-phase (where the neighboring S layer's pair functions take an identical phase) and the π-phase schemes. In the π-phase scheme, the T_c maximum occurs at $d_F/\xi_F \simeq 1$, while in the 0-phase scheme the T_c minimum takes place at $d_F/\xi_F \simeq 1$ and the T_c maximum locates at much larger d_F ($d_F/\xi_F \simeq 3$, see Fig. 1) [21]. If the T_c maximum of the π-phase scheme is higher than the T_c minimum of the 0-phase scheme, the π phase is to be actualized. It is difficult, however, to draw theoretically T_c versus d_F curve without ambiguity because of several necessary material parameters which are not easy to estimate experimentally.

The purpose of the present study is to find the difference in the T_c behavior between the zero and π phases. The guideline is given in Fig. 1. This figure presents the experimental results of the Nb/Co multilayer by our previous study [12] together with Tagirov's fitting curves [14] based on the parameter values listed in the figure caption. It can be seen that our data for the Nb/Co multilayer with the Nb sublayer thickness $d_{Nb} = 40$ nm are satisfactorily fitted on the π-phase line (thin broken line) in the region of x ($= d_{Co}$ for the multilayer) about 1.2–3.1 nm. Accordingly, we should be able to make a clear distinction between the two schemes from the T_c versus x curves for the trilayer (F/S/F) with one S layer and the pentalayer (F/S/F/S/F) with two S layers. We have chosen $d_{Nb} = 30$ nm as the S-layer thickness, because for $d_{Nb} = 30$ nm, a clearer difference between the 0 and π phase is expected than for $d_{Nb} = 40$ nm, as shown by the thick lines in the figure.

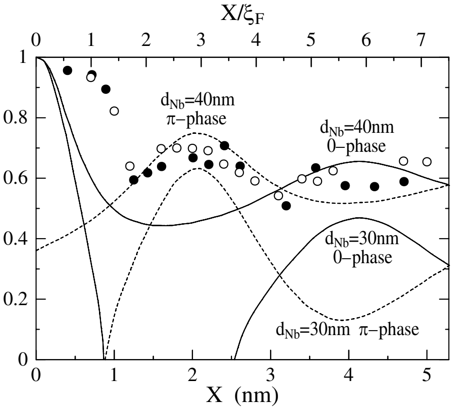

FIG. 1. T_c oscillation in the Nb/Co multilayer [12] plotted as T_c/T_{c0} versus x, together with the several fitting curves of Tagirov's calculation [14]. x is equal to the Co sublayer thickness d_{Co} for the multiplayer system. The fitting is based on the available parameters, $T_{c0} = 9.0$ K, $N_F v_F/N_S v_S = 0.28$, $\xi_S/\xi_0 = 0.182$, $T_M = 2.0$, and $l_F/\xi_F = 2.0$ for $d_S = 40$ and 30 nm, where ξ_0 is the BCS coherence length ($= 0.18\hbar v_S/k_B T_{c0}$). The thin full line and the thin broken line stand for $d_{Nb} = 40$ nm with the 0 and π phase, respectively. The thick full line and the thick broken line stand for $d_{Nb} = 30$ nm with the 0 and π phase, respectively.

The prepared layered specimens are Co(x/2 nm)/Nb(30 nm)/Co(x/2 nm) trilayers and Co(x/2 nm)/Nb(30 nm)/Co(x nm)/Nb(30 nm)/Co(x/2 nm) pentalayers where x was varied from about 0.8 nm to 5.2 nm. The thickness of the Co-edge layer was taken as $x/2$ so as to satisfy the boundary condition for the pair function [14]. (Note that $d_{Co} = x$ in the pentalayer, while $d_{Co} = x/2$ in the trilayer) The layered specimens were fabricated by rf-sputtering onto quartz-substrate with a very smooth surface. Target materials were Nb (purity is 99.95%) and Co (purity is 99.9%). After sputtered to the designed sequences, the samples were coated by Nb(2 nm) in order to protect the surface layer from oxidation. This Nb layer is not superconductive down to 2 K. The base pressures were under 2×10^{-7} Torr and sputtering was carried out in 20 mTorr Ar-pressure. The layer structure was confirmed by the low angle x-ray diffraction. The typical diffraction

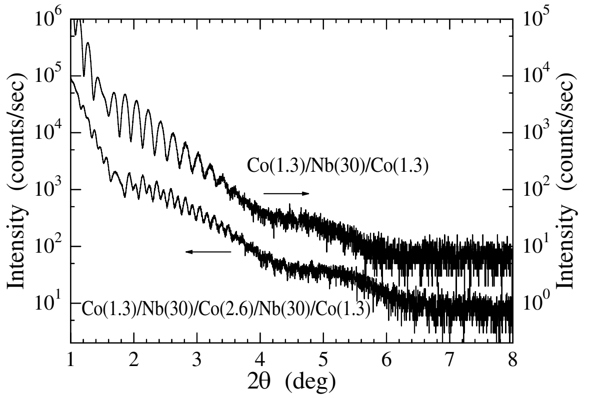

FIG. 2. The low angle x-ray diffraction profiles for the Co(1.3)/Nb(30)/Co(1.3) trilayer and Co(1.3)/Nb(30)/Co(2.6)/Nb(30)/Co(1.3) pentalayer.

patterns are shown in Fig. 2 for Co(1.3 nm)/Nb(30 nm)/Co(1.3 nm) trilayer and Co(1.3 nm)/Nb(30 nm)/Co(2.6 nm)/Nb(30 nm)/Co(1.3 nm) pentalayer. The satellite peaks characteristic of the layer structure can be seen. Analyses of the x-ray diffraction confirmed that deviation of the layer thickness from the designed one is less than 5%. T_c was decided as a middle point between 10% and 90% of the resistive transition measured by the four terminal method.

Figure 3 presents the typical results of the reduced electrical resistance R/R_n versus temperature T curve for one of the present pentalayer samples Co($x/2$)/Nb(30)/Co(x)/Nb(30)/Co($x/2$), where R_n is resistance in the normal state just above the transition temperature. The superconducting transitions take place within the width of 0.2 K between 10% and 90% change of R/R_n for most of the samples, suggesting the good sample quality. The data exhibit a clear nonmonotonic dependence of T_c on x.

Figure 4 shows the magnetic moment at 10 K as a function of x for the present tri- and pentalayers. In the figure, each symbol represents the layer samples sputtered in the same run. The magnetization for both series becomes zero around $x = 1.4$ nm. In the pentalayer series, the central Co layer is subject to the mixing or alloying with the both side Nb layers. Therefore, both tri- and pentalayer series may have a magnetically dead layer of about 0.7 nm. Usually thin Co layer retains the magnetic moment down to $1 \sim 2$ monolayer, as reported, for example, for the Cu/Co multilayer [22]. The large lattice mismatch between Nb⟨110⟩ and Co⟨0002⟩ of about 17% and/or forming of a mixture region may result in quenching of the magnetic moment up to as large $d_{\mathrm{Co}} \sim 0.7$ nm in the present layer samples.

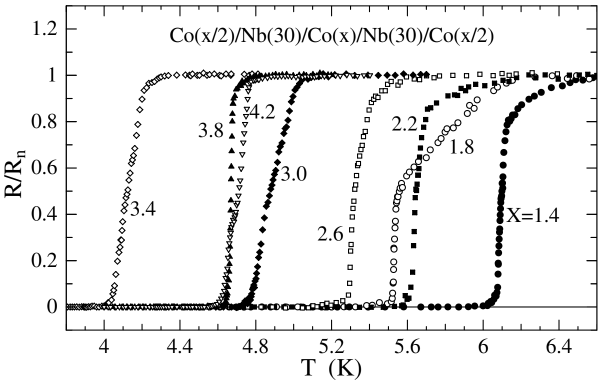

FIG. 3. The reduced resistance R/R_n versus T curve for one of the pentalayer series Co($x/2$)/Nb(30)/Co(x)/Nb(30)/Co($x/2$).

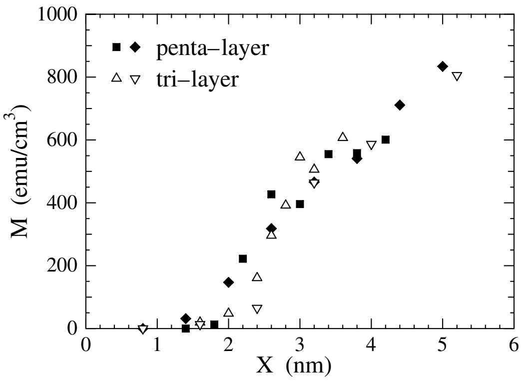

FIG. 4. The magnetic moment at 10 K versus x for the trilayers Co($x/2$)/Nb(30)/Co($x/2$) and the pentalayers Co($x/2$)/Nb(30)/Co(x)/Nb(30)/Co($x/2$). Each symbol represents the specimens sputtered in the same run.

Figure 5 shows T_c as a function of x for the tri- and pentalayers. The symbols are the same as Fig. 4. The curves of each two series of the trilayer and the pentalayer are slightly scattered, but several characteristic points can be seen. (i) In the trilayers, in which only the 0 phase is possible, T_c takes a local broad minimum around $x \sim 2.6$ nm. Nearly at the same position ($x \sim 2.3$ nm), the pentalayers take a local maximum, taking local minima

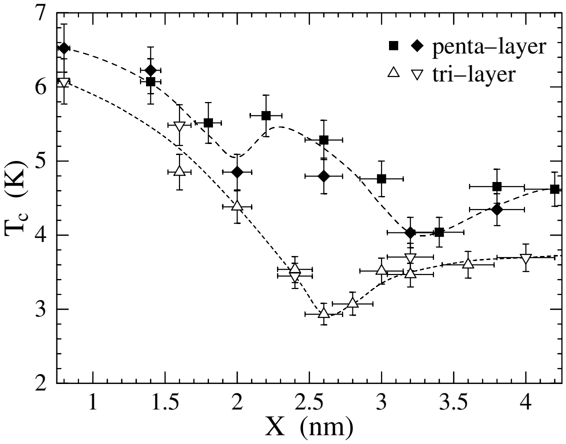

FIG. 5. T_c as a function of x for the trilayers Co($x/2$)/Nb(30)/Co($x/2$) and the pentalayers Co($x/2$)/Nb(30)/Co(x)/Nb(30)/Co($x/2$). The respective symbols stand for the data points of the samples sputtered in the same run as in Fig. 4. Lines are guides to the eyes.

at around $x = 2.0$ nm and 3.2 nm. (ii) In the region between the two minimum points, the π-phase superconductor is to be realized for the pentalayers, as predicted by Radović *et al.* (see Fig. 3 of Ref. [18]) and Tagirov (see Fig. 1). The different behavior between the tri- and pentalayer in the region between $x \sim 2.0$ and 3.2 nm provides a clear evidence for the occurrence of the π phase in the pentalayer. (iii) As mentioned in Fig. 4, the magnetic moment disappears nearly at about $x \sim 1.4$ nm for both tri- and pentalayers. This means that the T_c anomaly due to the spin flip scattering (which was attributed to the origin of the T_c oscillation of Nb/Fe case [3]) may be excluded for the origin of the present different oscillation behavior between the tri- and pentalayers.

According to Tagirov's calculation for our multiplayer data on the basis of parameters listed in the caption of Fig. 1, T_c of the 0 phase should degrade below 0 K for the trilayer for $d_{\mathrm{Nb}} = 30$ nm, and the reentrant behavior of T_c should be realized. The observed T_c depression for the present trilayers is not so drastic. This may come from the several factors which restrain the T_c depression. Garifullin *et al.* [6] suggested that interface roughness introduces random phase shifts for the interfering pair wave function, smearing out the interference pattern and the oscillation amplitude. On the basis of the Tagirov's formalism [19], the T_c oscillation amplitude is also controlled by the interface transparency parameter T_{M} in Eq. (3). T_{M} prescribes the mixing of the Cooper pairs with the ferromagnetic electron system and the smaller T_{M} value results in the smaller T_c oscillation amplitude. According to Garifullin *et al.*, the interface roughness should reduce the mixing of the Cooper pairs with the ferromagnetic electron system. Tagirov introduced the numerical value of $T_{\mathrm{M}} = 2$ for the fitting of the multiplayer data in Fig. 1. The present Nb/Co trilayer may actually have a T_{M} value smaller than 2 because of the enhanced interface roughness compared to the multiplayer system.

In conclusion, we have studied in detail the T_c oscillation behavior for Co/Nb/Co trilayers and Co/Nb/Co/Nb/Co pentalayers with the superconducting Nb layer thickness $d_{\mathrm{Nb}} = 30$ nm. The behavior of the T_c versus x curve is different between the tri- and pentalayer constructions; at around $x \sim 2.6$ nm the trilayer has a local minimum in T_c versus x, while the pentalayer has a local maximum around $x \sim 2.3$ nm. This difference should come from the different superconducting phase factor of the pair function $F(z)$. $F(z)$ of the trilayer consists of a single phase over the entire Co layer thickness d_{Co}, while in the pentalayer $F(z)$ of neighboring superconducting layers take a π-phase shift between $x \sim 2.0$ and 3.2 nm. From the appearance of the different T_c behavior between the trilayer and pentalayer Nb/Co systems, a clear evidence of the π phase of superconducting phase of superconductor/ferromagnet layered systems has been provided experimentally.

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