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Author(s)	Matsuda, Ken-ichi; Niwa, Hirotaka; Akimoto, Yosuke; Uemura, Tetsuya; Yamamoto, Masafumi
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Magnetic and Transport Properties of Nb/PdNi Bilayers

Ken-ichi Matsuda, Hirotaka Niwa, Yosuke Akimoto, Tetsuya Uemura, and Masafumi Yamamoto

Abstract—We investigated the superconducting proximity effect in both Nb(35 nm)/PdNi superconductor (S)-ferromagnet (F) bilayer and Nb–Nb(10 nm)/PdNi–Nb (S–SF–S) variable-thickness bridges (VTBs). In Nb(35 nm)/PdNi(d_F nm) SF bilayer, non-monotonic behavior of the superconducting transition temperature T_c was observed as a function of PdNi thickness, d_F . This non-monotonic behavior of T_c can be interpreted from the view point of damped oscillatory superconducting order parameter induced in F-layer, and the effective exchange energy E_{ex} extracted from the data was approximately 150 K. On the other hand, in VTBs, it was found that the superconducting critical current I_c was monotonically decreased with increasing d_F .

Index Terms—Proximity effect, superconducting films, superconductor-ferromagnet heterostructures.

I. INTRODUCTION

THE interplay between ferromagnetism and superconductivity in thin film superconductor-ferromagnet (SF) heterostructures has attracted considerable interest in both theoretically and experimentally in the field of electronic devices [1], [2]. SFS Josephson junctions (JJs) [3]–[5] and FSF spin valves [6] are examples of such SF heterostructure devices.

Especially, SFS JJs have great advantages for manipulating a spin degree of freedom in Josephson-junction-based electronics, for example, JJ-based qubits [7]. In SFS JJs, π -Josephson junction (π -JJs) behaviors have been predicted due to exchange field-induced oscillations of the superconducting order parameter over the coherence length ξ_F in the ferromagnetic layer (see Fig. 1), i.e., the coupling of the magnetic field to the quasiparticle spins dominates over the orbital coupling, leading to pairing between exchange-split Fermi surface, and hence to oscillatory behavior of superconducting order parameter. This spatially non-uniform superconducting order parameter in F-layer is a sort of Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state [8], [9].

Experimentally, such oscillations of the order parameter have been observed in FSF trilayer systems involving Fe or Co ferromagnetic layer [10]. In these systems with such strong ferromagnets, however, exchange energies E_{ex} are typically of the

Fig. 1. Schematic view of spatial distribution of superconducting order parameter on (a) Superconductor-Normal metal (SN) interface and (b) Superconductor-Ferromagnet (SF) interface.

order of 1 eV, and therefore, ξ_F may be 0.1 ~ 1 nm even at sufficiently low temperatures. In general, it is difficult to fabricate such a homogeneous thin film using conventional deposition techniques. This difficulty is one of the reason why experimental studies on the superconducting proximity effect in SF heterostructures are still in their infancy.

For this reason, weak ferromagnetic alloys, whose E_{ex} can be controlled by changing the amount of magnetic element, are of great interest. $\text{Cu}_{1-x}\text{Ni}_x$ ($x \sim 0.5$) and $\text{Pd}_{1-x}\text{Ni}_x$ ($0 < x < 0.3$) are the cases, where E_{ex} can be varied in meV range by changing Ni concentration. In these systems, ξ_F may be of the order of 5 ~ 10 nm. Moreover, Nb/PdNi heterostructure has an advantage that the interdiffusion between the two layers will be limited by the difference of the crystal structures between BCC and FCC.

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The authors are with the Division of Electronics for Informatics, Hokkaido University, Sapporo 060-0814, Japan (e-mail: matsuda@ist.hokudai.ac.jp).

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Fig. 2. (a) Schematic top view and (b) cross-sectional view of the VTB structure; (c) a SEM image of the VTB.

In this paper, we investigated magnetic and transport properties in two different types of SF bilayer: (i) Nb(35 nm)/Pd_{0.85}Ni_{0.15}(d_F nm) bilayer and (ii) Nb(100 nm) – Nb(10 nm)/Pd_{0.85}Ni_{0.15}(d_F nm) – Nb(100 nm) (S-SF-S) variable-thickness bridge (VTB). While the non-monotonic behavior of the superconducting critical temperature, T_c , was observed as a function of d_F in Nb/Pd_{0.85}Ni_{0.15} bilayer, the critical current, I_c , of VTB monotonically decreased with increasing d_F .

II. EXPERIMENTALS

In this study, we prepared two different samples: (i) Nb/Pd_{0.85}Ni_{0.15} (SF) bilayers and (ii) Nb – Nb/Pd_{0.85}Ni_{0.15} – Nb (S-SF-S) variable-thickness bridges (VTBs) (see Fig. 2). All samples were deposited on oxidized Si substrates using multisource RF magnetron sputtering system at room temperature. Nb thin films were deposited using ultra pure Ar gas at 2.5 mTorr from a solid target at a rate of 3.28 nm/min. After Nb deposition, Pd_{0.85}Ni_{0.15} (hereafter PdNi) were successively deposited at 4.6 mTorr from a stoichiometric ingot target at a rate of 1.36 nm/min. The Curie temperature of the PdNi alloy used in this study was approximately 260 K. The lateral size of the bilayer samples is $5 \times 5 \text{ mm}^2$.

VTB samples were fabricated by using conventional photolithographic technique. Schematic view of VTB structure and a SEM image of the bridge part are shown in Fig. 2. The VTB sample consists of 100 nm-thick Nb electrode and Nb(10 nm)/PdNi(d_F nm) bridge. The lateral size of the bridge is $5 \mu\text{m} \times 5 \mu\text{m}$.

III. RESULTS AND DISCUSSIONS

A. Nb(35 nm)/PdNi(d_F nm) Bilayer

Fig. 3 shows the PdNi thickness dependence of the superconducting transition temperature for Nb(35 nm)/PdNi(d_F nm) bilayer. Data were extracted from the temperature dependence of the magnetic susceptibility measured by SQUID magnetometer. It is clearly shown that T_c rapidly decreases with increasing d_F until $d_F \sim 3 \text{ nm}$ and has a minimum value at around $d_F = 3.5 \text{ nm}$. Physically, this non-monotonic behavior of T_c can be interpreted in the framework of damped oscillatory behavior of superconducting order parameter induced in F

Fig. 3. The PdNi thickness dependence of the superconducting transition temperature for Nb(35 nm)/PdNi(d_F nm) bilayer. Data were extracted from the temperature dependence of the magnetic susceptibility measured by SQUID magnetometer. The inset is the result obtained from the temperature dependence of resistivity. The dotted line is the guide for the eyes.

layer by proximity effect [11], [12]. Due to energy conservation, a Cooper pair entering into ferromagnet acquires a finite center-of-mass momentum, Δp , from the spin splitting of the up and down bands. In the dirty limit, Δp can be expressed as

$$\Delta p = \frac{1}{\xi_F} = \sqrt{\frac{E_{ex}}{\hbar D_F}}, \quad (1)$$

where D_F is the diffusion coefficient in F-layer and E_{ex} is an exchange energy [3]. This additional momentum modifies the phase of the pair wave function, $\theta = \Delta p \cdot x$, that increases with the distance, x , from the S/F interface. Therefore, Cooper pairs that experience normal reflections at the vacuum interface of F layer causes the interference, which can lead to oscillations of T_c as a function of d_F . When $\theta = \pi$, the interference effect should be destructive, and therefore $T_c(d_F)$ takes minimum value. In our experimental data, $T_c(d_F)$ shows minimum at around $d_F = 3.5 \text{ nm}$.

According to (1), the effective exchange energy E_{ex} can be estimated from d_F at which $T_c(d_F)$ takes minimum value. The diffusion constant D_F in F-layer is related to the residual resistivity, ρ_F , of PdNi through the electronic mean free path, l_F , by

$$D_F = \frac{v_F l_F}{3} \quad (2)$$

in which

$$l_F = \frac{1}{v_F \gamma_F \rho_F} \left(\frac{\pi k_B}{e} \right)^2, \quad (3)$$

where γ_F is the electronic specific heat coefficient and v_F is the Fermi velocity [13]. The residual resistivity of 100 nm-thick PdNi obtained from the preceding experiment was approximately $22 \mu\Omega\text{cm}$. If we adopt the values of $\gamma_F = 1.12 \text{ mJ/K}^2\text{cm}^3$ and $v_F = 2 \times 10^7 \text{ cm/sec}$ for pure Pd instead of PdNi, l_F can be calculated as $l_F = 1.48 \text{ nm}$, which results in $D_F = 9.9 \times 10^{-5} \text{ m}^2/\text{sec}$. Using these parameters, we can estimate the effective exchange energy to be $E_{ex} \sim 150 \text{ K}$. This result is in good agreement with the result reported by C. Cirillo *et al.* [14].

Fig. 4. Current-voltage curves for Nb – Nb(10 nm)/PdNi(d_F nm) – Nb VTBs at 1.4 K with (a) $d_F = 0$ nm, (b) $d_F = 1$ nm, (c) $d_F = 2$ nm, (d) $d_F = 4$ nm, (e) $d_F = 6$ nm, and (f) $d_F = 8$ nm.

B. Nb – Nb(10 nm)/PdNi(d_F nm) – Nb VTB

Fig. 4 shows current-voltage curves for VTBs with various F-layer thicknesses at 1.4 K. Current data are plotted as IR_N product, and typical R_N value for 10 nm-thick Nb bridge was 50Ω at 10 K. A clear transition between superconducting state and resistive state with small hysteresis was observed in each sample. While the shape of the curve was not so affected by the existence of F-layer, the magnitude of the critical current, I_c , was strongly suppressed with increasing d_F .

The F-layer thickness dependence of the critical current is clearly shown in Fig. 5. As can be seen, I_c decreases monotonically with increasing d_F . In these VTBs, F-layer thickness dependence of T_c indicated a similar behavior. From the view point of the proximity effect in SF bilayer, it is expected that the effect of the non-uniform superconducting order parameter on I_c can be observed as a function of d_F . In our results, any clear evidence of such an effect cannot be observed in the transport properties of VTBs, and it seems that there may be a discrepancy between the non-monotonic behavior of T_c in Nb/PdNi bilayer and the monotonic behavior of I_c in VTBs. These results

Fig. 5. F-layer thickness d_F dependence of the superconducting critical current I_c . Typical R_N value for 10 nm-thick Nb bridge was 50Ω at 10 K. Inset: Temperature dependence of I_c for VTBs with various d_F .

imply that there is another effect behind the transport properties of VTBs that we do not take into account.

IV. CONCLUSION

The superconducting proximity effect was examined in both Nb(35 nm)/PdNi superconductor-ferromagnet (SF) bilayer and Nb–Nb(10 nm)/PdNi–Nb (S–SF–S) variable-thickness bridges (VTBs). In Nb(35 nm)/PdNi(d_F nm) SF bilayer, non-monotonic behavior of the superconducting transition temperature T_c was observed as a function of PdNi thickness, d_F . This non-monotonic behavior of T_c can be interpreted from the view point of damped oscillatory superconducting order parameter induced in F-layer, and the effective exchange energy E_{ex} extracted from the data was approximately 150 K. On the other hand, in VTBs, it was found that the superconducting critical current I_c was monotonically decreased with increasing d_F .

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