# 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 $\mathrm{Nb(35\ nm)/PdNi}(d_F\ \mathrm{nm})$ 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, $\pi$-Josephson junction ($\pi$-JJs) behaviors have been predicted due to exchange field-induced oscillations of the superconducting order parameter over the coherence length $\xi_F$ 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 order of 1 eV, and therefore, $\xi_F$ may be $0.1 \sim 1$ 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. $\mathrm{Cu}_{1-x}\mathrm{Ni}_x(x \sim 0.5)$ and $\mathrm{Pd}_{1-x}\mathrm{Ni}_x(0 < x < 0.3)$ are the cases, where $E_{ex}$ can be varied in meV range by changing Ni concentration. In these systems, $\xi_F$ may be of the order of$5 \sim 10$ 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.

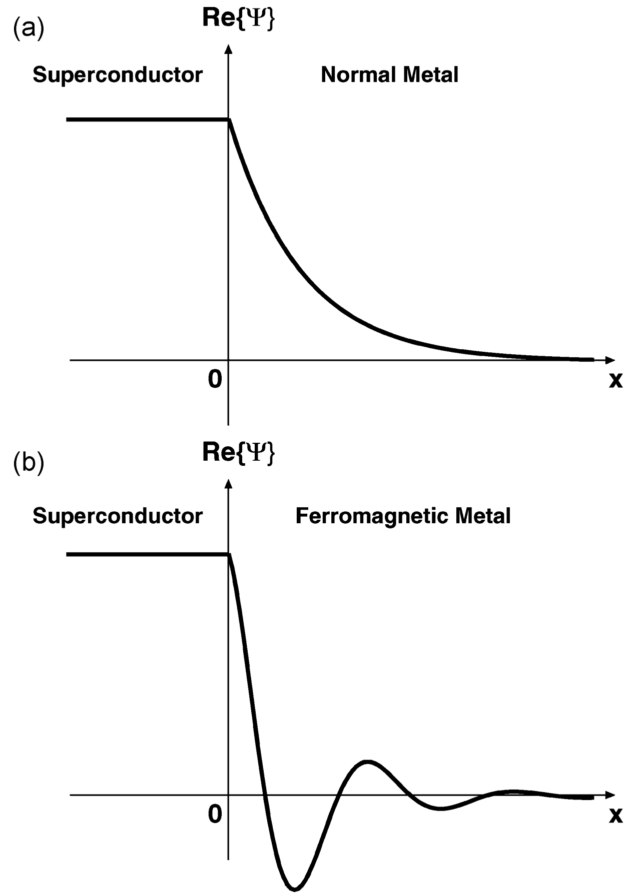


Fig. 1. Schematic view of spatial distribution of superconducting order parameter on (a) Superconductor-Normal metal (SN) interface and (b) Superconductor-Ferromagnet (SF) interface.

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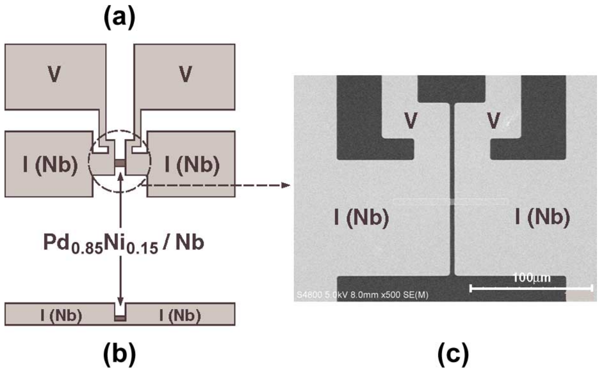


Fig. 2. (a) Schematic top view and (b) cross-sectional view of the VTB structure; (c) a SEM image of the VTB.

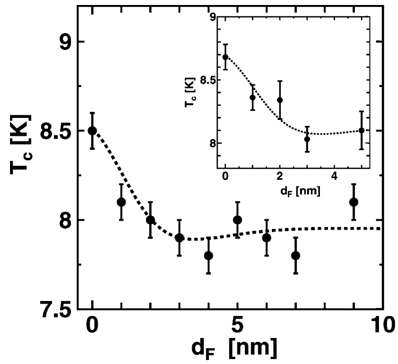


Fig. 3. The PdNi thickness dependence of the superconducting transition temperature for $\mathrm{Nb}(35\ \mathrm{nm})/\mathrm{PdNi}(d_F\ \mathrm{nm})$ bilayer. Data were extracted from the temperature dependence of the magnetic susceptibility measured by SQUID magnetometer. The inset is the result obtained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

In this paper, we investigated magnetic and transport properties in two different types of SF bilayer: (i) $\mathrm{Nb}(35\ \mathrm{nm})/\mathrm{Pd}_{0.85}\mathrm{Ni}_{0.15}(d_F\ \mathrm{nm})$ bilayer and (ii) $\mathrm{Nb}(100\ \mathrm{nm}) - \mathrm{Nb}(10\ \mathrm{nm})/\mathrm{Pd}_{0.85}\mathrm{Ni}_{0.15}(d_F\ \mathrm{nm}) - \mathrm{Nb}(100\ \mathrm{nm})$ (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 $\mathrm{Nb}/\mathrm{Pd}_{0.85}\mathrm{Ni}_{0.15}$ 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) $\mathrm{Nb}/\mathrm{Pd}_{0.85}\mathrm{Ni}_{0.15}$ (SF) bilayers and (ii) $\mathrm{Nb} - \mathrm{Nb}/\mathrm{Pd}_{0.85}\mathrm{Ni}_{0.15} - \mathrm{Nb}$ (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, $\mathrm{Pd}_{0.85}\mathrm{Ni}_{0.15}$ (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 $5 \times 5\ \mathrm{mm}^2$.

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 $\mathrm{Nb}(10\ \mathrm{nm})/\mathrm{PdNi}(d_F\ \mathrm{nm})$ bridge. The lateral size of the bridge is 5 $\mu\mathrm{m} \times 5\ \mu\mathrm{m}$.

## III. Results and Discussions

### A. $\mathrm{Nb}(35\ \mathrm{nm})/\mathrm{PdNi}(d_F\ \mathrm{nm})$ *Bilayer*

Fig. 3 shows the PdNi thickness dependence of the superconducting transition temperature for $\mathrm{Nb}(35\ \mathrm{nm})/\mathrm{PdNi}(d_F\ \mathrm{nm})$ 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 $d_F \sim 3\ \mathrm{nm}$ and has a minimum value at around $d_F = 3.5\ \mathrm{nm}$. 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 layer by proximity effect [11], [12]. Due to energy conservation, a Cooper pair entering into ferromagnet acquires a finite center-of-mass momentum, $\Delta p$, from the spin splitting of the up and down bands. In the dirty limit, $\Delta p$ can be expressed as

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

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 $d_F = 3.5\ \mathrm{nm}$.

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, $\rho_F$, of PdNi through the electronic mean free path, $l_F$, by

$$D_F = \frac{v_F l_F}{3} \tag{2}$$

in which

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

where $\gamma_F$ 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 $\mu\Omega\mathrm{cm}$. If we adopt the values of $\gamma_F = 1.12\ \mathrm{mJ/K^2cm^3}$ and $v_F = 2 \times 10^7\ \mathrm{cm/sec}$ for pure Pd instead of PdNi, $l_F$ can be calculated as $l_F = 1.48\ \mathrm{nm}$, which results in $D_F = 9.9 \times 10^{-5}\ \mathrm{m^2/sec}$. Using these parameters, we can estimate the effective exchange energy to be $E_{ex} \sim 150$ K. This result is in good agreement with the result reported by C. Cirillo *et al.* [14].

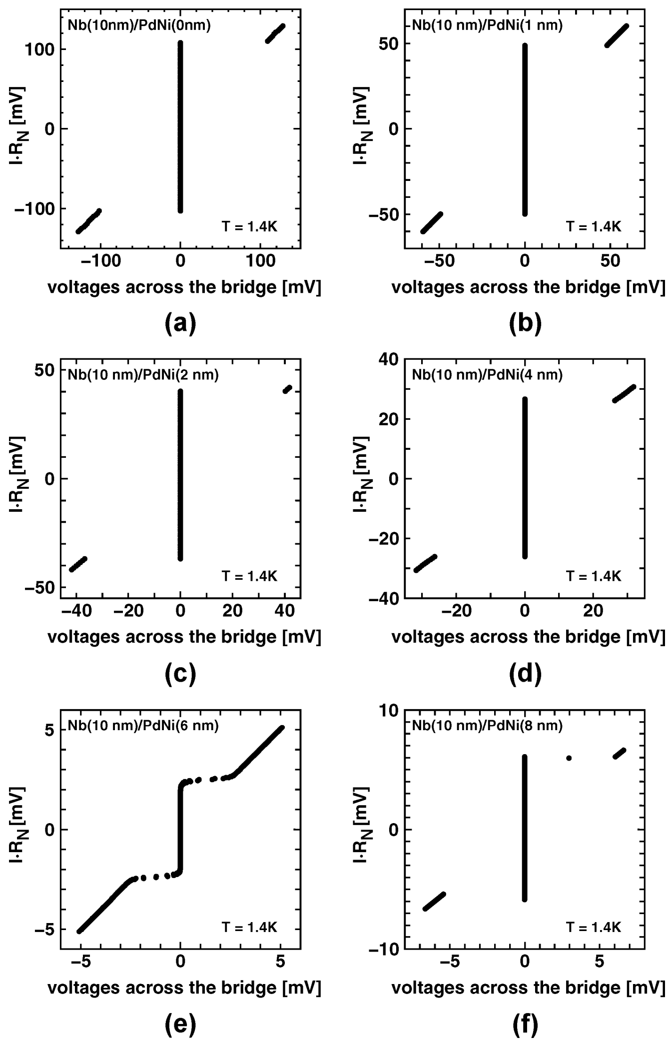


Fig. 4. Current-voltage curves for $\mathrm{Nb}-\mathrm{Nb}(10\ \mathrm{nm})/\mathrm{PdNi}(d_F\ \mathrm{nm})-\mathrm{Nb}$ VTBs at 1.4 K with (a) $d_F = 0\ \mathrm{nm}$, (b) $d_F = 1\ \mathrm{nm}$, (c) $d_F = 2\ \mathrm{nm}$, (d) $d_F = 4\ \mathrm{nm}$, (e) $d_F = 6\ \mathrm{nm}$, and (f) $d_F = 8\ \mathrm{nm}$.

## *B.* $\mathrm{Nb}-\mathrm{Nb}(10\ \mathrm{nm})/\mathrm{PdNi}(d_F\ \mathrm{nm})-\mathrm{Nb}$ *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 50 Ω 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

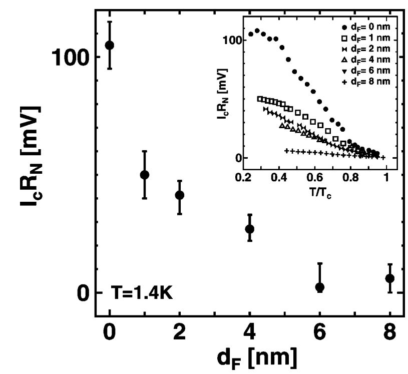


Fig. 5. F-layer thickness $d_F$ dependence of the superconducting critical current $I_c$. Typical $R_N$ value for 10 nm-thick Nb bridge was 50 Ω at 10 K. Inset: Temperature dependence of $I_c$ for VTBs with various $d_F$.

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 $\mathrm{Nb}(35\ \mathrm{nm})/\mathrm{PdNi}(d_F\ \mathrm{nm})$ 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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