Possibility of Cooper-pair formation controlled by multi-terminal spin injection

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\textbf{Abstract.} A multi-terminal lateral spin valve consisting of three ferromagnetic nanopillars on a Cu/Nb bilayer has been fabricated. We investigated the influence of the spin injection on the superconducting properties at the Cu/Nb interface. The non-local spin valve signal exhibits a clear spin insulation signature due to the superconducting gap of the Nb. The magnitude of the spin signal is found to show the probe configuration dependence. From the careful analysis of the bias current dependence, we found the suppression of the superconductivity due to the exchange interaction between the Cooper pair and accumulated spin plays an important role in the multi-terminal spin injections. We also discuss about the possibility of the Cooper-pair formation due to the spin injection from the two injectors with the anti-parallel alignment.

1. Introduction

Ferromagnet/superconductor hybrid structure is a fantastic and attractive system because the combination between the spin and superconducting transports is expected to produce various intriguing phenomena\cite{1–7}. However, superconducting properties are, in general, easily destroyed by the direct contact with the ferromagnetic material\cite{8–10}. This is because the Cooper pairs are deformed by the strong exchange interaction from the ferromagnet and/or other extrinsic effects such as the interface mixing and the diffusion of the magnetic impurities. These serious obstacles can be solved by introducing a normal metal spacer between the ferromagnetic metal and superconductor. Indeed, a conventional lateral spin valve combined with a superconductor enable us to inject the pure spin current into the superconductor\cite{11, 12}. The advantage of the electrical spin injection is its precise tunability of the magnitude of the spin accumulation. However the relatively large electrical resistivity of the ferromagnetic injector produces non-negligible Joule heating, resulting in the suppression of the superconductivity at the interface\cite{11}. To reduce the Joule heating from the ferromagnet, we have developed a lateral spin valve consisting of a nano-pillar spin injector/detector and the Cu/Nb bilayer channel. By using this novel device structure, we showed that the superconducting Nb acts as an insulator for a pure spin current and that the superconducting proximity effect is induced also in the Cu layer with the non-equilibrium spin accumulation\cite{13}. Thus, a nano-pillar-based lateral spin valve with a normal metal/superconductor bilayer provides an ideal platform for investigating the interplay between superconductivity and ferromagnetism. In this paper, by extending the nano-pillar spin injector to the multi-terminal spin injectors as shown in Fig. 1, we control the
Figure 1. Schematic illustration of multi-terminal spin injection into a Cu/Nb bilayer. (a) Suppression of the superconductivity under parallel spin injection and (b) formation of the Cooper pairs under anti-parallel spin injection.

Figure 2. (a) Scanning electron microscope image of the measured sample. (b) Anisotropic magnetoresistance of each injectors. The recovery of the resistance to the high resistance state indicates that the flip of the magnetization.

spin current in a Cu/Nb bilayer channel by the magnetic alignment of the injectors and seek a novel control method of the superconducting properties using the spin injection.

2. Experimental
We have fabricated a nanopillar based lateral spin valve consisting of three Ni-Fe nanopillars and Cu/Nb bilayer channel. First, a Ni-Fe/Cu/Nb trilayer film as a ferromagnet/normal metal/superconductor by using a high vacuum thin-film growth system consisting of two magnetron sputterings and a Joule evaporator with the base pressure less than $5 \times 10^{-5}$ Pa. Here, the Ni-Fe and Nb are grown by the magnetron sputterings while the Cu is deposited by a Joule evaporation. The thicknesses for the Ni-Fe, Cu and Nb are, respectively, 20 nm, 100 nm and 45 nm. Then, the trilayer film was patterned into the nanopillar structure by using electron-beam lithography and Ar ion milling. Figure 2(a) is the scanning electron microscope image of the measured sample. The lateral dimensions for two ferromagnetic nanopillars are designed as $320 \times 60 \text{nm}^2$, $210 \times 50 \text{nm}^2$ and $210 \times 80 \text{nm}^2$, and the center-center distance between the neighboring ferromagnetic nanopillars is 500 nm. Here, the superconducting transition temperature of the fabricated Nb was 6.8 K.
Figure 3. Schematic illustration of the spin accumulation in the Cu/Nb bilayer and magnetic field dependence of the non-local resistance at (a) 10 K and (b) 2.2 K. The black and gray arrows indicate the magnetization of the detector and the injector, respectively. The spin signal at 2.2 K is much larger than that at 10 K, which indicates the spin current absorption is effectively protected by the superconducting gap.

We investigate the spin transport properties of the Cu/Nb bilayer by measuring the non-local spin valve signals [14–16]. Here, the left- and right-hand-side Ni-Fe nanopillars NiFe1 and NiFe3 were used as the spin injectors and the middle Ni-Fe nanopillar NiFe2 was used as the spin detector. There are several advantages in this device structure. First, the interfaces have ideal conditions because the trilayer was prepared by in-situ multi-layered film grown system. As second advantage, a large spin accumulation can be generated by flowing a large current because the quasi-two-dimensional Cu heat sink strongly suppresses the temperature increase due to the spin injection. Third advantage is the variety of the magnetic configuration owing to the combination of triple ferromagnetic nanopillars with different switching fields.

3. Results and Discussion

First, we confirmed that three Ni-Fe nanopillars have different switching fields by measuring the anisotropic magnetoresistance (AMR) of each nanopillar with the probe configurations shown in the inset of Fig. 2(b). Figure 2(b) shows the AMR curves for three Ni-Fe nanopillars measured at 10 K under the magnetic field along the $y$ axis. Since the magnetization is laying in $xy$ plane, the magneto-resistance measured by the current along the $z$ axis should show no field dependence of the resistance. However, the inhomogeneous current distribution produces a small in-plane component of the current, resulting in a small field dependence of the resistance. As can be seen in Fig. 2(b), the magnetizations of three Ni-Fe nanopillars showed the different switching fields depending on their geometrical shape. Thus, the magnetization configuration can be controlled by the external magnetic field.

Next, we evaluate the spin transport properties at the Cu/Nb interface by measuring the non-local spin valve signals with the different temperature. Figures 3(a) and 3(b) show the non-local spin signals measured at 10 K and 2.2 K, respectively. Although the spin signal at 10 K
Figure 4. Electrochemical potentials in the Cu layer and the measured non-local resistance under (a) single- and (b) two-terminal spin injection. The black and gray arrows indicate the magnetization of the detectors and the injectors, respectively. The spin signal $\Delta R_S$ is defined by the difference of the resistance at the zero magnetic field. $R_{BG}$ is the background resistance, which indicates the non-local resistance without spin accumulation.

was as small as 0.18 m$\Omega$ because of the strong spin absorption effect of the Nb metal, the spin signal at 2.2 K becomes 6 times larger than that at 10 K. This significant increase of the spin signal at lower temperature clearly indicates that the spin current absorption was effectively protected by the superconducting gap of the Nb at the interface. Here, the non-local spin signal at 2.2 K shows unconventional broad field dependence, where the resistance change between the parallel and anti-parallel states decreases with increasing the magnitude of the magnetic field. This can be understood by the reduction of the spin signal because the superconducting gap at the Nb/Cu interface decreases by the application of the magnetic field, leading to the less protection of the spin absorption. We also point out the relatively large background resistance in Fig. 3(a) because of the spreading of the injecting current in the two-dimensional Cu film. This background resistance is significantly suppressed by the superconductivity, as shown in Fig. 3(b).

To investigate the influence of the spin accumulation on the superconducting properties, we perform two kinds of the spin injection. One is a conventional single spin injection using a NiFe3 injector and NiFe2 detector, as schematically shown in Fig. 4(a). The other one is a dual spin injection (two-terminal injection) by using NiFe1 and NiFe3 injectors with NiFe2 detector, as schematically shown in Fig. 4(b). Figures 4(a) and 4(b) show the representative spin signals measured at 2.2 K with the low bias current of 0.26 mA. To evaluate the magnitude of the spin accumulation precisely, we measured the minor loops. For the two-terminal measurement, the vertical axis is the voltage divided by the current for each electrode. Therefore, the overall
resistance change for the two-terminal spin injection is approximately twice of the single-terminal one.

We then investigate the bias current dependence of the spin transport properties. Since the heating effect under the electrical spin injection is mainly generated from the Ni-Fe nanopillar, we assume that the influence of the Joule heating can be characterized by the local current of each electrode[17]. Therefore, we expect that the influence of the Joule heating under the two-terminal spin injection is almost same as that for the single-terminal one when the current flowing in each nanopillar is the same value. On the other hand, the influence of the spin accumulation for the two-terminal spin injection should be two times larger than that for the single-terminal one. So, these two effects should reduce the spin signal because both the temperature increase and the magnetic field suppress the superconductivity. However, the current dependence should show the different signature. Namely, the influence of the Joule heating is proportional to $I^2$ while that of the spin accumulation should be proportional to $I$. Therefore, by carefully analyzing the bias current dependence of the spin signals both for the single- and two-terminal spin injections, we can clarify the influence of the spin accumulation on the superconducting property.

Figure 5(a) shows the bias current dependences of the spin signals for the single- and two-terminal spin injections. Here, we plot the spin signal at the zero magnetic field $\Delta R_S$, in order to remove the extrinsic effect of the spin accumulation. As can be seen in the plot, the reduction of the spin signal for the two-terminal spin injection was found to be larger than that for the single-terminal spin injection. This is consistent with our expectation that the spin accumulation destroys the superconductivity. To analyze this more quantitatively, the bias current dependence was fitted with the following equation.

$$\Delta R_S = \Delta R_{S0} - aI^2 - bI,$$

where $\Delta R_{S0}$ is the spin signal at zero bias, $a$ and $b$ are the coefficients for the Joule heating and spin accumulation, respectively. From the fittings, for the single case, $a$ and $b$ are $1.01 \times 10^{-2}$ mΩ mA$^{-2}$ and 0.21 mΩ mA, respectively. For the two-terminal spin injection case, we obtained that $a$ and $b$ are $1.06 \times 10^{-2}$ mΩ mA$^{-2}$ and 0.30 mΩ mA, respectively. Thus, the influence of the Joule heatings for both configurations were almost same. On the other hand, the influence of the spin accumulation in two-terminal spin injection is larger than that of the single spin injection, by a factor of 1.4. This is a clear evidence that the spin accumulation effectively suppresses superconductivity. We believe that the exchange interaction between
the spin-polarized electron and the Cooper pair is a microscopic origin of this reduction, as schematically shown in Fig. 1(a).

Finally, we investigate the possibility of the Cooper-pair formation, by injecting the electrons with anti-parallel spins, as shown in Fig. 1(b). Since the two-terminal spin injection with the anti-parallel configuration does not induce the detectable spin signal, the spin signal does not provide any evidence of the Cooper-pair formation due to the anti-parallel spin injection. Therefore, we discuss about the bias current dependence of the background resistance $R_{\text{BG}}$, in order to pick up the signature of the Cooper-pair formation. As shown in Fig. 5(b), $R_{\text{BG}}$ gradually increases with increasing the bias current both for the single and two-terminal injections. However, for the two-terminal injection, in spite of the larger total current, the increase of $R_{\text{BG}}$ was smaller than that for the single terminal injection. This indicates that the suppression of the superconductivity was weaker in the two-terminal injection than that in the single-terminal injection. Since the effective conversion from the electrons to the Cooper pair tends to protect the spin absorption, the less increase of $R_{\text{BG}}$ suggests the effective Cooper-pair formation under the two-terminal spin injection with anti-parallel orientation. Additionally, as shown in Fig. 4(b), the broad field dependence of the non-local signal for the anti-parallel configuration seems to be smaller than that for the parallel configuration. Since the Cooper-pair formation effectively occurs under the anti-parallel configuration, the observed difference between two configurations can be explained by the difference in the formation rate. These results imply that the two-terminal spin injection with the anti-parallel orientation assists the Cooper-pair formation.

4. Summary
We have investigated the influence of the electrical spin injection from the multi-terminal Ni-Fe nanopillars on the superconducting properties in the Nb/Cu bilayer film. By comparing the non-local spin valve signal of the single-terminal spin injection with that of the two-terminal spin injection, we are able to analyze the influence of the spin injection and the Joule heating on the superconductivity separately. The non-equilibrium spin accumulation is found to suppress the superconductivity of the Nb. However, its magnitude is not simply proportional to the number of the injectors. This implies that the Cooper-pair formation process can be controlled by injecting the spin-polarized electrons.

Acknowledgments
This work was partially supported by Grant-in-Aid for Scientific Research on Innovative Area, "Nano Spin Conversion Science" (26103002), that for Scientific Research (S) (25220605), and that for Young Scientists (B) (17K14109).

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