Tunnel Conductance in Ni₈₀Fe₂₀/Al-oxide/Al Junctions below the Superconducting Temperature of Al Films

Mikihiko Oogane, Tadaomi Daibou, Hitoshi Kubota, Yasuo Ando and Terunobu Miyazaki Department of Applied Physics, Graduate School of Engineering, Tohoku University, Aoba-yama 05, Sendai 980-8579, Japan Fax: 81-022-217-7947, e-mail: oogane@mlab.apph.tohoku.ac.jp

Spin polarization of the Ni₈₀Fe₂₀/Al-oxide interface was measured using Superconductor/Insulator /Ferromagnet (S/I/F) structure. High quality SiO₂/Al/Al-oxide/Ni₈₀Fe₂₀ (A) and SiO₂/Ni₈₀Fe₂₀/Al-oxide/ Al (B) junctions were prepared using magnetron sputtering. Al films deposited in the mixture gas of Ar (0.08 Pa) and O₂ (0.004 Pa) were used as superconducting layers. The Al-oxide layers prepared by plasma oxidation were used as insulating layers. For the junction with an Al top electrode, annealing process at 250 °C for one hour was necessary to improve superconducting property of the Al film. The magnitudes of the spin polarization of the fabricated junctions were 38 % (A) and 21 % (B), respectively. It is considered that the spin polarization values are strongly influenced by the detailed structure of the Ni₈₀Fe₂₀/Al-oxide interface.

Key words: Spin polarization, Superconductor/Insulator/Ferromagnet structure, Al, Ni80Fe20

1. INTRODUCTION

Since magnetic tunnel junctions (MTJs) were first studied in 1975,¹ junctions with significant tunnel magneto-resistance (TMR) effect at room temperature have been developed.²⁻³ MTJs are greatly useful for various technological applications, especially for nonvolatile magnetic random access memories (MRAMs).⁴

It is expected from simple model that TMR ratio increases with increasing spin polarization of ferromagnet/insulator interface.¹ Therefore, materials with high spin polarization are necessary for the development of MTJ devices. However, the relationship between the magnitude and the sign of spin polarization and electronic structure of the ferromagnetic material is still a debatable subject.⁵⁻⁷ Spin polarized tunneling (SPT) technique using a superconducting film as a spin detector has played an important role in the investigation of the tunneling spin polarization.5-7 The maximum TMR ratio reported were almost explained by the spin polarization values obtained using this technique.^{2,7} The magnitudes of the spin polarization of the Ni based alloys varied among the past reports.⁶⁻⁷ This reason is considered that the magnetic property of the Ni based alloy is very sensitive to the interface state. In this work, the high quality SiO₂/Al(5nm)/Al-oxide/Ni₈₀Fe₂₀(20nm) (JunctionsA) and SiO₂/Ni₈₀Fe₂₀(20nm)/Al(2nm)-oxide/ Al(5nm) (Junctions B) junctions were fabricated for the spin polarization measurement.

2. Experimental Procedure

Cross type tunnel junctions (both A and B) were fabricated using magnetron sputtering with shadow masks. Junction area was $100 \times 100 \ \mu\text{m}^2$. The base pressure of sputtering system was lower than 2×10^{-7} Pa. The Al films were prepared in pure Ar gas and in Ar (0.08 Pa) and O₂ (0.004 Pa) mixed gas. The Al-oxide layers were prepared by inductively coupled plasma (ICP) oxidation of the metallic Al films. For Junctions A,

after deposition of the bottom 5-nm-thick Al films, the surfaces were oxidized without breaking a vacuum. For Junctions B, after deposition of the bottom $Ni_{80}Fe_{20}$ films, the 2-nm-thick Al films were deposited and the surfaces were oxidized. RF powers applied for plasma oxidation were 10 and 100 W, and oxidation time was varied from 30 to 600 sec. The mixture gas of O₂ (0.75 Pa) and Ar (0.25 Pa) was used for the plasma oxidation. Junctions B were annealed at 250 °C for one hour after the deposition of the top Al films.

The conductance-voltage (G-V) characteristics were measured using the conventional four point AC/DC technique. Superimposed on a very small AC current, a DC bias is swept from -1.5 to 1.5 mV. The output AC voltage was detected by lock-in amplifier. The measurement temperature was 0.4 K, and the maximum magnetic field applied was 4 Tesla. The measurement temperature was enough low to neglect the thermal excitation of quasiparticle. In addition, inelastic electron tunnel spectra (IETS) were measured using the same system at 10 K to investigate metal/insulator interface state.

3. Experimental Results and Discussion

3.1 Al/Al-oxide/Ni₈₀Fe₂₀ junction (Junctions A)

The superconducting transition temperature of the Al film deposited in pure Ar was 1.8 K, which was not enough high if thermal excitation of quasiparticle at measurement temperature of 0.4 K was considered. Therefore, the Al films were deposited in Ar (0.08 Pa) and O₂ (0.004 Pa) mixed gas. Fig. 1 shows the temperature dependence of the resistivity for the Al film prepared. The Al film showed superconducting transition temperature of 2.2 K and a critical field of nearly 4 T. In addition, the surface of the Al film prepared in O₂ mixed gas was relatively smooth. The average roughness of the Al film estimated from the AFM image was 0.5 nm.

The high quality junction is necessary for the spin



Fig. 1 ρ -*T* characteristic of the Al film for the spin polarization measurement.

polarization measurement using the SPT technique. When leakage conductance through a junction is large, it is impossible to evaluate spin polarization correctly. Therefore, as the first stage of the spin polarization measurement, we investigated oxidation process for the Al-oxide layers in order to obtain high quality junctions with low leakage conductance. Figs. 2 (a)-(b) show G-Vcharacteristics of the Junctions A measured at 0.4 K. The magnitudes of the conductance were normalized by the conductance at higher bias voltages more than 1mV, where the Al films were in normal state. The applied powers of plasma oxidation were (a) 10 W and (b) 100 W. The oxidation times were (a) 600 sec and (b) 45 sec. The resistance and area products (RA) of both junctions were nearly the same (about $2 \times 10^6 \ \Omega \mu m^2$). G-V characteristics of these junctions were quite different. In ideal, the normalized conductance should be zero at zero-bias, because superconductor has the energy gap at the Fermi level. In experiment, as shown in fig. 2(a), the conductance at zero-bias was very large (about 0.8), corresponding to large leakage conductance through this junction. On the other hand, in fig. 2 (b), normalized conductance at zero-bias was very small (below 0.1), and two peaks corresponding to the edges of the energy gap were clearly observed. The difference of the amount of the leakage conductance originates in the difference of the oxidation process. Details about the relation between the oxidation process and the leakage conductance will be reported in another publication.8

Fig. 3 shows G-V characteristics of the junction corresponding to fig. 2 (b) measured at 4 T. The four peaks splited by Zeeman effect were clearly observed. Spin polarization of Al-oxide/Ni₈₀Fe₂₀ interface was evaluated to 38 % using the following equation.

$$P = [(\sigma_1 - \sigma_3) - (\sigma_4 - \sigma_2)] / [(\sigma_1 - \sigma_3) + (\sigma_4 - \sigma_2)] \quad (1)$$

The magnitude of spin polarization obtained is slightly smaller than that of the recent report (45 %),⁷ and is larger than that of the old report (28 %).⁶

3.2 Ni₈₀Fe₂₀/Al-oxide/Al junction (Junctions B)

Figs. 4 (a)-(b) show G-V characteristics of Junctions B. Both Al and Al-oxide layers were prepared by the



Fig. 2 The Conductance-voltage characteristics in the Al/Al-oxide/Ni₈₀Fe₂₀ junctions at 0.4 K and at 0 T. Al-oxide layers were prepared at (a) RF power 10 W and 600 sec and (b) RF power 100 W and 45sec.



Fig. 3 The Conductance-voltage characteristics of the Al/Al-oxide/Ni $_{80}Fe_{20}$ junctions at 0.4 K and 4 Tesla



Fig. 4 The Conductance-voltage characteristics of the Al/Al-oxide/Ni₈₀Fe₂₀ junctions at 0.4 K and 0 T. The junction in (a) is as deposited sample, and the junction in (b) is annealed sample.

same conditions as those for the junction corresponding to fig. 2 (b). G-V characteristic in fig. 4 (a) was slightly different from that in fig. 2 (b). The conductance at zero-bias was almost the same, but the peaks of the conductance were lower and broader than those in fig. 2 (b). In this case (fig. 4 (a)), the split of the conductance peaks in the large magnetic field were not clear and the spin polarization can not be evaluated. This broadning of the peaks would be caused by the distribution of the superconducting transition temperature of the Al interface. Mixing at the Al-oxide/Al interface would occur heavily during the sputtering of the Al film, resulted in the distribution of superconducting transition temperature. The junction in fig. 4 (b) was annealed at 250 °C for one hour after deposition of the top Al film. The peaks of conductance were sharper than those in fig. 4 (a) and similar to those in fig. 2 (b). The annealing process improved distribution of the superconducting transition temperature at the Al/Al-oxide interface.

Figs. 5 (a)-(b) show IET spectra of Junctions A and B measured at 10 K and 0 T. For both the junctions, the top Ni₈₀Fe₂₀ electrodes were positively biased in the positive bias sides and the magnitudes of spectra were normalized by the each conductance. IET spectroscopy has been applied to the study of inelastic excitations owing to phonons, magnons and molecular vibrations at the interfaces of tunnel junctions.9-10 According to the previous reports, typical spectrum of junctions with bottom Al electrodes showed a peak around 30 mV corresponding to the excitation of the Al phonon.¹⁰ In fig. 5 (a), the peak around -30 mV was very broad. After annealing the sample, the peak became remarkable as shown in fig. 5 (b). This result suggested that the Al/Al-oxide interface became sharp by annealing, resulted in homogeneous energy dispersion of the Al phonon over the junction area.

Fig. 6 shows G-V characteristics of the junction corresponding to fig. 4 (b) at a magnetic field of 3 T. In this case, the applied magnetic field was slightly smaller than the one in fig. 3, because the critical field of the Al film deposited on the Al-oxide was smaller than that of the Al film on the SiO₂ substrate. The four peaks were clearly observed even in 3 T. The evaluated spin polarization from this spectrum was 21 %. The magnitude of the spin polarization was about half of that of Junctions A.

3.3 Comparison of IET spectra between Junctions A and B

The current-voltage (I-V) characteristics and IET spectra of Junctions A, B were measured at 10 K (above superconducting transition temperature of the Al films). The barrier heights and widths of these junctions were evaluated from I-V characteristics fitted by Simmon's equation.¹¹ For both the junctions, the asymmetries in positive and negative biases were small and the fitting curves agreed well with the experimental data. The barrier heights were about 2 eV and the barrier widths were about 1.2 nm for both the junctions. Consequently, the difference of the detailed interface structure did not affect the I-V characteristics. Figs. 7 (a)-(b) show IET spectra of Junctions A and B, respectively. For Junctions A, the top Ni₈₀Fe₂₀ electrode was positively biased and for Junctions B, the bottom Ni₈₀Fe₂₀ electrode was positively biased at positive bias side. If an insulating



Fig. 5 The IET spectra in the $Ni_{80}Fe_{20}/Al-oxide/Al$ junctions at 10 K. The junction in (a) was as deposited sample, and the junction in (b) was annealed sample.



Fig. 6 The Conductance-Voltage characteristics in the $Ni_{80}Fe_{20}$ /Al-oxide/Al junctions in the 3 Tesla magnetic field.



Fig. 7 The IET spectra in the (a) Junction A and (b) Junction B at 10 K. Junction B was annealed at 250 °C for one hour.

layer is Al-oxide, a peak at 110 - 120 mV corresponding to the excitation of the Al-oxide phonon can be seen in the typical spectra.¹⁰ In both the spectra in figs. 7 (a), (b), the peaks corresponding to the Al-oxide phonon were observed at the same bias of -110 mV. However, in the positive bias, Junctions A showed a peak at about 120 mV, which was about 20 mV larger than the peak in Junctions B. This peak shift would be due to compounds of a Al-Ni(Fe)-O at the Ni₈₀Fe₂₀/Al-oxide interface. Such a compound can also be a reason for the reduction of the spin polarization. Further investigation is necessary to make clear the relation between the interface structure and the spin polarization.

4. Summary

We succeeded to fabricate high quality SiO₂/Al/ Al-oxide/Ni₈₀Fe₂₀ (A) and SiO₂/Ni₈₀Fe₂₀/Al-oxide/Al (B) junctions. The spin polarization of both the junctions were evaluated from their *G-V* characteristics at 0.4 K with high magnetic fields. The magnitudes of the spin polarization were 38 % (A) and 21 % (B), respectively. IET spectrum suggested that the compound of Al-Ni (Fe)-O was formed at the Ni₈₀Fe₂₀/Al-oxide interface, caused the reduction of the spin polarization in Junctions B.

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