Thickness and Oxidation Time Dependence of Tunnel Magnetoresistance in Ni-Fe/Co/Al-O/Co Junctions

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Ni-Fe/Co/Al-O/Co tunnel junctions with small active areas down to $3 \times 3 \mu\text{m}^2$ were fabricated by the micro-fabrication technique. The Al-O insulating layer was prepared by plasma oxidation of a thin sputtered Al film. The dependences of the tunnel magnetoresistance on the thickness of the Al film and the oxidation time were investigated. The interface structure of the junction was observed by using high-resolution electron microscopy. The relationship between the magnetoresistive properties and the interface structure is discussed.

Key words: ferromagnet, spin tunnel junction, magnetoresistance, ICP, plasma oxidation

1. Introduction

Since the discovery of large magnetoresistance effect in Fe/Al-O/Fe¹⁾ and Co/Al-O/Ni-Fe²⁾ junctions, tunnel magnetoresistance effect (TMR) has been studied intensively not only from the view point of fundamental physics but also device applications. For the application of the junctions to in magnetic reading heads or magnetic random access memory, the junction with low resistance is required because of some technical reasons: signal-to-noise ratio, quick response, and conformity to impedance of amplifier used in the products.

Recently, very large tunnel magnetoresistance (TMR) ratios up to 40 % are reported by several groups³⁻⁵⁾, but the resistance values are much higher than the values required for device application (about $10^4 \Omega$ at the junction area less than $1 \mu\text{m}^2$). In order to reduce the resistance, we have to make a very thin and continuous insulating layer. The structure and electric property of the insulating layer is strongly related to the fabrication method and experimental conditions. However, no one has studied systematically the relation among them. For the preparation of the insulating layer, some methods such as natural^{1,6)} or plasma oxidation^{2,4)} of metallic Al, direct sputtering using Al₂O₃ target⁷⁾, and reactive evaporation⁸⁾ can be employed. Plasma oxidation using inductively coupled plasma (ICP) may be useful for preparing the thin Al-O layer because the ions do not strike strongly against the thin Al surface. It is interesting to study the magnetoresistance properties with very thin Al layer oxidized by the same method. In this report the dependences of the junction resistance and the TMR ratio on the thickness

of metallic Al layer and oxidation time using ICP method were investigated. The relationship between the interface structure and magnetoresistance properties will be discussed.

2. Experimental procedure

2.1 Thin film fabrication

The following three types of junctions were prepared. Type X: Sub./Ni-Fe(19 nm)/Co(4 nm)/Al-O/Co(4 nm)/Ni-Fe(10 nm)/FeMn(50 nm)/Ta(5 nm). Type Y: Sub./Ni-Fe(19 nm)/Co(4 nm)/Al-O/Co(15-30 nm). Type Z: Sub./Ta(2 nm)/Al(30 nm)/Ta(3 nm)/Ni-Fe(3 nm)/Ir-Mn(10 nm)/Co(5 nm)/Al-O/Co(4 nm)/Ni-Fe(20 nm)/Ta(5 nm).

The layers were deposited by rf magnetron sputtering at an argon pressure of 0.08 Pa on a thermally oxidized Si substrate. The deposition rate was about 0.06 nm/s for every metal. The thickness of Al layer varies from 0.5 nm to 1.6 nm. The surface of Al film was oxidized for 40 to 360 s by ICP method in an oxygen-argon mixture at a pressure of 1 Pa. 70-100 W rf power was put in a coil mounted above the cathode as shown in Fig. 1. The entire growth of all the layers was done in-situ. Some junctions were annealed at 10^{-4} Pa for 1 hour in an external magnetic field of 330 Oe.

2.2 Micro-fabrication process

The tunnel junction films were patterned using lithographic process, as schematically illustrated in Fig. 2. Firstly the bottom electrode geometry was defined by using a metal mask. Then photoresist pattern was formed by photolithography. The thickness of the resist is 0.5 -1.5 μm . The shape of the resist pattern is shown in Fig. 2 (a). After the Ar ion milling

Fig. 1 Schematic illustration of ICP oxidation.

Fig. 2 Schematic illustrations of the micro-fabrication process used.

step, the active junction area was defined. The junction area has a square geometry, ranging from 100×100 to $3 \times 3 \mu\text{m}^2$. In this process, metallic short-circuiting is a serious problem. Currents flow through the materials that are redeposited on the side wall of the junction area, resulting in low MR ratio.

If the larger area around junction is sputtered by Ar ion, many atoms may be deposited on the side wall. Then, we defined small area with $10 \mu\text{m}$ width around the junction area for Ar ion etching. The effect of the short-circuiting on the incident angle of Ar ion and the etching depth were investigated⁹⁾. The optimum conditions obtained were using the acceleration voltage at 400 V and incident angle at 15 degree from the film normal. The etching was timed to stop at several nanometer below the surface of the insulating layer. After the Ar ion etching, the sample was exposed to oxygen plasma for a few minutes. After that, the photoresist on the film can be removed easily in an acetone bath with ultra sonic wave. The junctions were passivated with 250 nm of SiO_2 by rf sputtering. The contact hole was opened in SiO_2 by another lithographic process and CF_4 plasma etching. The bottom surface of the hole was cleaned by Ar ion etching. Finally 250 nm-thick-Cu was sputtered as a top electrode.

2.3 Measurement

The magnetoresistance measurement was performed by the dc four probe method at room temperature. The resistance and TMR ratio were measured by applying 1 mV to the junction. Tunnel barrier height and width were obtained from the current-voltage characteristics and fitting to Simmons' theory of tunneling¹⁰⁾. The layer structure of the junction was observed by cross-sectional high resolution electron microscopy (HREM).

Fig. 3 Junction area dependence of the resistance and MR ratio for the junction with 1.3-nm Al.

3. Results

Figure 3 shows the junction area dependence of the tunnel resistance for as-prepared and annealed junctions (Type Z). Thickness of the Al sputtered was 1.3 nm. The resistance increases in inversely proportion to the junction area. This means that the effect of electrical short-circuiting through the materials redeposited on the side wall is negligible and that the microstructured junctions are successfully made by using the process described above. The required resistance for MRAM or magnetic reading heads were also shown in Fig. 3 for comparison. Our experimental values are very close to the required value for MRAM¹¹⁾ but much higher than that for heads. TMR ratios are about 20% before annealing, which is independent of the junction area. The values increase after annealing where maximum values of 35% was obtained after annealing at 275°C. The dependences of the TMR ratio on annealing temperature are reported in elsewhere¹²⁾. The resistance value at $1 \mu\text{m}^2$ corresponds to the resistance-area products (R_{RAP}) of the junctions. The value is expected to be smaller with decreasing Al thickness. However, the result does not agree with our expectation.

Figures 4(a) and 4(b) show the contour of the R_{RAP} and MR ratio, as functions of Al thickness and oxidation time, respectively (Type X and Y). R_{RAP} is almost constant around the junctions with Al thickness of 1-1.3 nm oxidized for 120 s. The values increase rapidly with both increasing and decreasing Al thickness for oxidation time more than 120 s. R_{RAP} depend mainly on the oxidation time for the junctions with shorter oxidation time. The minimum values of the R_{RAP} obtained for the junctions with Al oxidized for 60 s and 120 s, which were $5 \text{ k}\Omega \cdot \mu\text{m}^2$ and $50 \text{ k}\Omega \cdot \mu\text{m}^2$ at 0.7 nm and 1.3 nm, respectively. The TMR ratio is largest in the junctions with 1.3 nm Al oxidized for more than 240 s. Hereafter we call this region (B). The values decrease with both in-

Fig. 4 Contour of resistance (a) and MR ratio (b) as functions of the Al thickness and oxidation time.

creasing and decreasing Al thickness (regions (A) and (C)). The TMR ratio of the junction which shows the minimum R_{RAP} (40-60 s and 0.7 nm) is 15 %.

Figures 5(a) and 5(b) show the contour of the barrier width and height, respectively, as functions of Al thickness and oxidation time. The barrier width is almost constant at Al thickness of 1-1.3 nm. The values increase rapidly with decreasing Al thickness. This corresponds to the rapid increase of the resistance of the junctions as shown in Fig. 4(a). The barrier height depends mainly on Al thickness and increases gradually with increasing Al thickness and oxidation time.

Figure 6 shows the HREM cross-section image of the junction with 0.7 nm Al oxidized for 60 s. From the image the crystalline texture of top and bottom electrodes and the amorphous nature of Al-O layer is confirmed. The observed thickness of Al-O layer varies from 0.8 to 1.3 nm. The right hand side of the image is very clear, but it is not clear on the left hand side. This corresponds to change of the thickness in the beam direction of the sample: The thickness increases from right to left. Superposition of the two or three interfaces are visible on the left hand side of the image (not clear). It is suggested that the roughness of the interface exists in the junction. Another images of the junction with 1.3 nm Al oxidized for 210 s showed very flat and clear interface between the insulator and magnetic electrodes.

4. Discussion

The variations of the resistance and TMR ratio depended on fabrication condition. This suggests that tunnel magnetoresistance is very sensitive to the interface structure. We

Fig. 5 Contours of (a) barrier width and (b) the height as functions of the Al thickness and oxidation time.

Fig. 6 HREM cross-section image of a tunnel junction with 0.7-nm Al oxidized for 60 s by the ICP method.

Fig. 7 Schematic representation of the interface structure of the tunnel junctions.

try to explain those variations using the model^{13,15)} as shown in Figs. 7(a)-(c). The regions (A), (B) and (C) in Fig. 4(b) correspond to the three regions where the interface structure of the junction should be different. In region (A), the barrier width is thinner than the thickness of bare Al. The unoxidized Al should remain under the oxidized Al (Fig. 7(a)). This causes the smaller MR ratios for the junctions in this region. In region (B), the Al is almost completely oxidized and the bottom Co surface is not oxidized (Fig. 7(b)). The HREM images of the junction with 1.3 nm Al oxidized for 210 s showed flat and clear interface between the insulator and magnetic electrodes. The sharp interface results in large TMR ratio for the junction in this region. In region (C) the barrier width is large and the barrier height is small as shown in Fig. 5. These results should originate from the oxidation of the bottom Co surface (Fig. 7(c)). The barrier height of Co-oxide could be lower than that of Al-oxide because the band gap of the Co-O is smaller than that of Al₂O₃. The HREM image (Fig. 6) suggests that there exist some pin holes in the junction with Al thickness less than 0.7 nm. Recently, Ando et al. studied the local current distribution of the tunnel junctions of very thin Al-O insulator¹⁶⁾. They confirmed very small pin holes from the current images. In order to obtain small R_{RAP} value with large TMR ratio, flat and thin Al-O layer without pin hole must be obtained.

The resistance obtained in this experiment is the lowest value among the reported values for plasma oxidized junctions. For applying the junctions to decives, the junctions with lower resistance is required. Recently very low resistance is reported by several groups in naturally oxidized junctions^{3, 17, 18)}. Because the ICP oxidation method has some advantages, it is also possible to obtain lower resistance junctions by optimizing the fabrication conditions.

5. Summary

Ni-Fe/Co/Al-O/Co tunnel junctions were fabricated by using magnetron sputtering system, ICP oxidation method, and the combination of photolithography and Ar ion etching technique. The tunnel junctions with small active area down to $3 \times 3 \mu\text{m}^2$ were successfully fabricated. The effects of micro fabrication process on magnetoresistance properties were examined. The dependences of the R_{RAP} , TMR ratio, barrier width, and barrier height on the thickness of Al film and oxidation time have been mapped out. More than 20% and 35% TMR ratio are obtained before and after annealing in the junctions with thick Al layer. The R_{RAP} value was also large in the junction. Low R_{RAP} value was obtained in the junctions with thin Al, which showed small TMR ratio. In order to apply the junctions to magnetic device such as a reading head, it is important to improve quality of the thin insulating layer.

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