Magnetoresistance of Single and Double Tunnel Junctions Formed by Direct Sputtering Using Al_2O_3 Target

H. Kubota, T. Watabe, Y. Fukumoto and T. Miyazaki

Department of Applied Physics, Graduate School of Engineering, Tohoku University, Sendai 980-8579, Japan

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Tunnel magnetoresistance (TMR) has been investigated in single ($\text{Ni-Fe}/\text{Al}_2\text{O}_3/\text{Co}$) and double ($\text{Ni-Fe}/\text{Al}_2\text{O}_3/\text{Co}/\text{Al}_2\text{O}_3/\text{Co}$) tunnel junctions. The Al_2O_3 layer was formed by a direct sputtering method with an Al_2O_3 target. Metal masks were used to form a cross pattern geometry. In the single junctions the dependence of the tunnel resistance and the TMR ratio on the Al_2O_3 thickness was investigated. The tunnel resistance increased exponentially with increasing the Al_2O_3 thickness. A maximum TMR ratio was 4-5 %. In the double junctions the dependence of the tunnel resistance and the TMR ratio on the thickness of central Co layer was investigated. The tunnel resistance decreased with increasing the Co thickness. The maximum TMR ratio was 2%, which was smaller than the value of the single junctions.

Key words: ferromagnetic tunnel junction, magnetoresistance, Al_2O_3 , double tunnel junction

1. Introduction

For the study of tunnel magnetoresistance (TMR) effect in FM/I/FM junctions the insulator has been formed by natural¹⁾⁻³⁾ or plasma oxidation^{4),5)} of the metallic aluminum. When the aluminum film is thick or oxidized incompletely, the metallic aluminum would exist under the aluminum-oxide layer. In such cases, the dependence of the MR ratio and the tunnel resistance on the nominal thickness of the insulator has not been clear. We have tried a direct sputtering method with an Al_2O_3 target to make the insulator. In this method it is possible to control directly the thickness of the insulator and to make double or multilayered tunnel junctions by sequential sputtering. The double tunnel junction is very interesting because of the enhancement of TMR by the resonant tunneling effect^{6),7)} or by Coulomb blockade⁸⁾. A little the experimental data on TMR effect in the double junction has been reported⁹⁾. We describe the dependence of TMR in the single junctions on the Al_2O_3 thickness and the dependence of TMR in the double tunnel junctions on the central magnetic layer thickness. Also the relation between TMR effect and the film structure will be described.

2. Experimental Procedure

Samples were prepared by rf magnetron sputtering at an argon pressure of 0.2 Pa. The deposition rates were 0.8, 0.3 and 0.1 nm/s for $\text{Ni}_{80}\text{Fe}_{20}$, Co, and Al_2O_3 , respectively. The purity of the targets are 99.9 and 99.99 % for magnetic metals and Al_2O_3 , respectively. Metal masks were used to form a cross pattern geometry. The area of the junction was varied between 100×100 and $500 \times 500 \mu\text{m}^2$. The layer structure of the single junction fabricated was {bottom electrode : $\text{Ni}_{80}\text{Fe}_{20}$ (20 nm)}/ Al_2O_3 ($d_{\text{Al-O}}=1-3$ nm)}/{top electrode : Co (50 nm)}. That of the double junction was {bottom electrode : $\text{Ni}_{80}\text{Fe}_{20}$ (20 nm)}/ Al_2O_3 (1.5 nm)}/{central Co ($d_{\text{Co}}=0-5$ nm)}/ Al_2O_3 (1.5 nm)}/{top electrode : Co (50 nm)}. All the

junctions were prepared on a thermally oxidized Si wafer. The magnetoresistance curve was measured by the dc four terminal method at room temperature with magnetic fields of 130 Oe applied in the film plane. The magnetization curves of the junctions were measured at room temperature. The trilayers Al_2O_3 (1.5 nm)/Co ($d_{\text{Co}}=0-5$ nm)/ Al_2O_3 (1.5 nm) were also fabricated on the same condition. Their magnetization curves and the resistivity with a sense current flowed in the plane were measured at room temperature.

3. Results and Discussion

3.1 Single tunnel junction

Figures 1 show the examples of magnetoresistance curves in the single tunnel junctions with the different value of $d_{\text{Al-O}}$. TMR is observed in the junctions with $d_{\text{Al-O}}=1$ and 1.5 nm. The MR ratio is 4-5 % except the junctions which showed a small tunnel resistance and a large MR ratio up to about 20 % due to the geometrical enhancement. TMR is not observed in the junction with $d_{\text{Al-O}}=3$ nm. The junction shows a very high tunnel resistance.

Figure 2 shows the dependence of the normalized tunnel resistance of the junctions on $d_{\text{Al-O}}$. In the figure, the data of the junctions fabricated by natural¹⁰⁾⁻¹²⁾ or plasma⁴⁾ oxidation are also plotted for comparison. For the present case, the normalized tunnel resistance increases exponentially with increasing $d_{\text{Al-O}}$. On the other hand, the resistance of the junctions prepared by other methods scatters and no systematic $d_{\text{Al-O}}$ dependence is observed. In the ideal case the tunnel resistance is expected to increase exponentially with increasing the thickness of the insulator. Therefore, the electrical properties of the Al_2O_3 fabricated by the direct sputtering method is better than the one fabricated by any other method^{3),10)-12)}. The barrier height and width were evaluated by fitting Simmons' relation to I-V curve measured at room temperature¹³⁾. The values obtained were about 0.6 eV and 2 nm, respectively, for the junctions with $d_{\text{Al-O}}=1$ and 1.5 nm. The barrier height is smaller than that reported^{3),14),15)}. The lower barrier height and larger width suggest the mixing at the interface. The small value of the MR ratio would be related to the interface structure.

3.2 Double tunnel junctions

Figures 3 show the magnetoresistance curves of the double tunnel junctions. The value of $d_{\text{Al-O}}$ is fixed at 1.5 nm and d_{Co} is varied from 0 to 5 nm. The junction with $d_{\text{Co}}=0$ corresponds to the single junction with $d_{\text{Al-O}}=3$ nm. In this junction, the tunnel resistance is very high and TMR is not observed. The junction with $d_{\text{Co}}=0.5$ nm, the tunnel resistance decreases rapidly and a small TMR is observed. TMR is observed clearly in the junctions with $d_{\text{Co}} \geq 1$ nm. The magnetoresistance curves of the junctions with $d_{\text{Co}}=2$ and 3 nm show the rapid decrease of the tunnel resistance near 20 Oe, which corresponds to the change of alignment of magneti-

Fig. 1 Magnetoresistance curves in $\text{Ni}_{80}\text{Fe}_{20}(20 \text{ nm})/\text{Al}_2\text{O}_3/\text{Co}(50 \text{ nm})$ single tunnel junctions.

zation of the top and the bottom electrodes from antiparallel to parallel. Above 20 Oe, the curves show the gradual decrease, which correspond to the rotation of the magnetization of the central Co layer.

Figure 4 shows the MR ratio versus the normalized tunnel resistance in the double junctions with various d_{Co} . In the figure the data of the single junctions are also plotted. The MR ratio increases up to 2 % for $0 < d_{\text{Co}} < 1 \text{ nm}$ and almost constant for $d_{\text{Co}} \geq 1 \text{ nm}$. The MR ratio of the double junctions with $d_{\text{Co}} \geq 1 \text{ nm}$ is about half as large as the value of the single junction with $d_{\text{Al}_2\text{O}_3} = 1.5 \text{ nm}$. The normalized tunnel resistance decreases with increasing d_{Co} . This reduction of the resistance and the MR ratio would be closely related to the change of the microstructure of the central Co and Al_2O_3 layers.

Figures 5 show the schematic illustrations of the cross section of the junctions. Figure 5 (a) and (b) correspond to the junctions with $d_{\text{Co}} = 0$ and 0.5 nm, respectively. The reduction of the tunnel resistance for $d_{\text{Co}} \leq 1 \text{ nm}$ is due to the decrease of the width of the tunnel barrier from 3 to 1.5 nm. The magnetization curve of the $\text{Al}_2\text{O}_3(1.5 \text{ nm})/\text{Co}(1 \text{ nm})/$

Fig. 2 The dependence of the normalized tunnel resistance on the thickness of Al_2O_3 in $\text{Ni}_{80}\text{Fe}_{20}(20 \text{ nm})/\text{Al}_2\text{O}_3/\text{Co}(50 \text{ nm})$ single tunnel junctions.

Fig. 3 Magnetoresistance curves in $\text{Ni}_{80}\text{Fe}_{20}(20 \text{ nm})/\text{Al}_2\text{O}_3(1.5 \text{ nm})/\text{Co}(d_{\text{Co}})/\text{Al}_2\text{O}_3(1.5 \text{ nm})/\text{Co}(50 \text{ nm})$ double tunnel junctions.

$\text{Al}_2\text{O}_3(1.5 \text{ nm})$ trilayer thin film showed a superparamagnetic curve at room temperature. Therefore, the central Co for $d_{\text{Co}} \leq 1 \text{ nm}$ would be small particles embedded in Al_2O_3 . Actually the resistivity of $\text{Al}_2\text{O}_3(1.5 \text{ nm})/\text{Co}(0.5 \text{ nm})/\text{Al}_2\text{O}_3(1.5 \text{ nm})$ trilayer thin film measured with a sense current flowed in the film plane was larger than $10^5 \mu\Omega \cdot \text{cm}$. The small MR ratios for $d_{\text{Co}} < 1 \text{ nm}$ is due to a random distribution of the direction of the magnetization of the small Co particles.

Figures 5 (c) and (d) correspond to the junctions with $d_{\text{Co}} = 2$ and 5 nm, respectively. In these junctions, the central Co layers are ferromagnet and continuous. If the structure of the Al_2O_3 layers were the same, the tunnel resistance should be constant independently of Co thickness for $d_{\text{Co}} \geq 2 \text{ nm}$. In practice, Co atoms grow partially like an island and the roughness of the surface of the central Co layer would be larger with increasing d_{Co} . Therefore, the Al_2O_3 around Co islands may be thinner for $d_{\text{Co}} = 5 \text{ nm}$ (Fig. 5 (d)) than for $d_{\text{Co}} = 2 \text{ nm}$ (Fig. 5 (c)), leading to the reduction of the tunnel resistance. The arrows in the figure indicate the direction of the magnetization in the magnetic field of between 20 and 40 Oe. The coercive field of the $\text{Al}_2\text{O}_3(1.5 \text{ nm})/\text{Co}(2 \text{ nm})/\text{Al}_2\text{O}_3(1.5 \text{ nm})$ trilayer thin film was about 40 Oe at room temperature. In the ideal case the magnetizations of the top and the bottom electrodes are aligned parallel and that of the central Co antiparallel in this field range. If the magnetizations of the layers were aligned as expected, the magnetoresistance curve would show the highest resistance between 20 and 40 Oe. In practice, the parts of the central Co should connect magnetically to the top Co layer and their magnetizations should rotate together at lower field. This may be the reason of the small MR ratios in the double junctions. The mechanism of the magnetic coupling is not clear but

Fig. 4 The MR ratio versus the normalized tunnel resistance in the double junctions.

the dipole-dipole interaction through the very thin insulator may be considered. The structural analysis of the surface of the central Co and the upper Al_2O_3 layer using AFM is important to obtain more information about the structure of the junctions.

4. Summary

Ni-Fe/ Al_2O_3 /Co and Ni-Fe/ Al_2O_3 /Co/ Al_2O_3 /Co tunnel junctions have been fabricated by a direct sputtering method using an Al_2O_3 target. The insulator can be formed by this method conveniently. The MR ratio in the single junction was 4-5 %. The tunnel resistance increased exponentially with increasing Al_2O_3 thickness. This indicates that the Al_2O_3 fabricated by this method has a good property of the insulator. But further examinations of sputtering conditions are necessary to obtain a larger MR ratio. In the double junctions the MR ratio and the normalized tunnel resistance depend on the thickness of the central Co layer. The values were about 2 % for the thickness of the central Co layer more than 1 nm, which is half as large as the one observed in the single junctions. The reason of the smaller MR ratio and the decrease of the tunnel resistance would be related to the microstructure of the central Co and the Al_2O_3 layers.

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Fig. 5 The schematic illustrations of the cross section of the double tunnel junctions.

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