Magnetoresistance in Fe-Mn/Ni-Fe-Co/Co/Cu/Co/Ni-Fe-Co Sandwiches with NiO or NiO/Hf Buffer Layer

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Magnetoresistance effects and film structures have been investigated in [Fe-Mn/Ni-Fe-Co/Co/Cu/Co/Ni-Fe-Co/NiO/Si] sandwiches. The NiO buffer layers increase exchange bias fields applied to the Ni-Fe-Co/Co magnetic bilayers neighboring with the Fe-Mn layers. However, the NiO buffer layers decrease the magnetoresistance ratios of the sandwiches. Therefore, we also inserted Hf buffer layers between the NiO buffer layers and the Si substrates. The Hf buffer layer, 1.0 nm in thickness, increases the magnetoresistance ratios of the sandwiches.

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I. Introduction

Giant magnetoresistive effects have been investigated in many magnetic multilayers⁽¹⁾⁻⁽³⁾. The magnetoresistive effects of exchange-biased sandwiches⁽⁴⁾, which are also called "spin valves", have also been investigated because the sandwiches show high magnetoresistance ratios at relatively low magnetic fields.

In the Fe-Mn/Ni-Fe/Cu/Ni-Fe exchange-biased sandwiches, the buffer layers are formed between the sandwiches and the substrates, in order to form the Ni-Fe/Cu/Ni-Fe layers with the fcc-(111) textures which are observed by X-ray diffraction⁽⁵⁾. This is because when the Ni-Fe/Cu/Ni-Fe layers have the (111) textures, the Fe-Mn layers on the Ni-Fe/Cu/Ni-Fe layers become the γ -Mn structures which show antiferromagnetic properties at room temperature. The Fe-Mn antiferromagnetic layers apply the exchange bias fields to the Ni-Fe magnetic layers neighboring with the Fe-Mn layers. It has also been reported that the Nb, Ta, Ti, Zr and Hf (IVa and Va elements of the periodic table) buffer layers cause the strong (111) textures in the Ni-Fe/Cu/Ni-Fe layers⁽⁵⁾.

In this study, we investigated magnetoresistance effects and film structures of [Fe-Mn/Ni-Fe-Co/Co/Cu/Co/Ni-Fe-Co] sandwiches with NiO buffer layers. The NiO buffer layers do not show the antiferromagnetic properties when the thickness is thinner than 20 nm⁽⁶⁾. But, the NiO buffer layers make the Ni-Fe-Co/Co/Cu/Co/Ni-Fe-Co layers with the weak (111) textures. Moreover, the NiO layers can be exposed to the air, because the NiO is oxide. Therefore, we can form the exchange-biased sandwiches using sputtering apparatuses with fewer targets.

II. Experimental Procedures

The NiO buffer layers and the sandwiches were prepared using an ion beam sputtering apparatus. The incident angle of the Argon ion beams from the ion gun to the targets was 45 degrees from the target surfaces. The incident angle of the sputtered particles to the substrates was almost perpendicular to the substrate surfaces. The distance between the targets and substrates was 120 mm.

We used Si(100) wafers which were 7 mm square as the substrates. The Si substrates were water-cooled during sputtering. At first, we deposited the NiO buffer layers on the Si substrates. The thicknesses of the NiO buffer layers were 5, 10, 15 and 20 nm. This is because the NiO buffer layers do not show the antiferromagnetic properties when the thickness is thinner than 20 nm⁽⁶⁾. The acceleration voltage of the ion gun was 900 V with an ion current of 120 mA. The deposition rate was 0.10 nm/s. Argon pressure during sputtering was 0.02 Pa. After the deposition, we exposed the Si substrates with the NiO buffer layers to the air. We also prepared the NiO(10 nm)/Hf buffer layers for a part of this study. For the NiO/Hf buffer layers, we deposited the Hf layers before the deposition of the NiO layers. The acceleration voltage of the ion gun was 300 V with an ion current of 60 mA for the deposition of the Hf layers. The deposition rate was 0.025 nm/s. Argon pressure during sputtering was also 0.02 Pa.

The sandwiches were grown on the buffer layers with the Si substrates, or directly grown on the Si substrates. The acceleration voltage of the ion gun was 300 V with an ion current of 30 mA, when the Ni-16at.%Fe-18at.%Co, the Co and the Cu layers were deposited. The ion current was changed to 60 mA, when the Fe-40at.%Mn layers were deposited. The ion currents were

decided in order to obtain about the same deposition rates. The deposition rates were around 0.025 nm/s. The Argon pressure during sputtering was also 0.02 Pa.

The magnetization curves and the magnetoresistance ratios of the sandwiches were measured at room temperature. A vibrating sample magnetometer was used to measure the magnetization curves with a maximum magnetic field of 20 kA/m. The magnetoresistance ratios were measured using the 4-terminal method. The maximum magnetic field was 80 kA/m. The magnetic field was applied in-plane and perpendicular to the current. The magnetoresistance ratio was defined as the ratio of the total resistivity change to the resistivity at a magnetic field of 80 kA/m. The film structures were investigated using a diffractometer with Cu-K α X-rays.

III. Experimental Results

1. Magnetoresistance effects in sandwiches

The structures of the sandwiches are Cu(5.0 nm)/Fe-Mn(10.0 nm)/Ni-Fe-Co(3.0 nm)/Co(2.0 nm)/Cu(3.0 nm)/Co(1.0 nm)/Ni-Fe-Co(9.0 nm)/NiO(t nm)/Si substrates, where the thickness of the NiO buffer layer t is changed. The Cu layer, on the Fe-Mn layer, is a capping layer. Figure 1 shows the magnetization curves of the sandwiches. The figure indicates that the magnetization curve of the magnetic bilayer (Ni-Fe-Co/Co bilayer)

Fig. 1 Magnetization curves of Cu(5.0 nm)/Fe-Mn(10.0 nm)/Ni-Fe-Co(3.0 nm)/Co(2.0 nm)/Cu(3.0 nm)/Co(1.0 nm)/Ni-Fe-Co(9.0 nm)/NiO(t nm)/Si sandwiches with various NiO buffer layer thicknesses t .

neighboring with the Fe-Mn layer is shifted from zero field in each sandwich. It is understood that the magnetic bilayer neighboring with the Fe-Mn layer is exchange-biased by the antiferromagnetic Fe-Mn layer. The other magnetic bilayer (Co/Ni-Fe-Co bilayer) neighboring the NiO layer is not biased by the Fe-Mn layer, because the Cu layer magnetically separates two magnetic bilayers in each sandwich.

In the sandwich without the NiO buffer layer (NiO: 0 nm), the magnetizations of two magnetic bilayers are parallel at the applied fields just below 0 kA/m, when the applied magnetic field varied from the negative to the positive direction. The magnetizations change from parallel to antiparallel at the applied field near 0 kA/m. The magnetizations are antiparallel at the applied fields from about 0 to 5 kA/m. The magnetizations change from antiparallel to parallel at the applied field around 5 kA/m. The magnetizations of two magnetic bilayers do not become antiparallel sufficiently, when the applied field varies from positive to negative direction. This is because the exchange bias field which is applied to the magnetic bilayer by the antiferromagnetic Fe-Mn layer is low.

In the other sandwiches with the NiO buffer layers, the magnetization curves of the magnetic bilayers neighboring the Fe-Mn layers are strongly shifted. This is because the exchange bias fields are high. The exchange bias field slightly increases as the thickness of the NiO buffer layer increases from 5 nm to 20 nm. The magnetizations of two magnetic bilayers become antiparallel sufficiently, when the applied field varies both directions.

Figure 2 shows the relationship between the thickness of the NiO buffer layer and the coercive force of the magnetic bilayer which is not exchange-biased by the Fe-Mn layer. The coercive force is 0.26 kA/m in the sandwich without the NiO buffer layer. The coercive force slightly increases as the thickness of the NiO buffer layer in-

Fig. 2 Relationship between the thickness of the NiO buffer layer and the coercive force of the magnetic bilayer which is not exchange-biased by the Fe-Mn layer in Cu(5.0 nm)/Fe-Mn(10.0 nm)/Ni-Fe-Co(3.0 nm)/Co(2.0 nm)/Cu(3.0 nm)/Co(1.0 nm)/Ni-Fe-Co(9.0 nm)/NiO(t nm)/Si sandwiches with various NiO buffer layer thicknesses t .

creases from 0 to 10 nm. The coercive force becomes high when the thickness of the NiO buffer layer is above 15 nm.

Figure 3 shows the relationship between the thickness of the NiO buffer layer and the magnetoresistance ratio. The magnetoresistance ratio is 5.1% in the sandwich without the NiO buffer layer. As shown in Fig. 1, the magnetizations of two magnetic bilayers change between parallel and antiparallel sufficiently in the sandwiches with the NiO buffer layers. However, the magnetoresistance ratios of the sandwiches with the NiO buffer layers are lower than that of the sandwich without the NiO buffer layer. The resistivity of the sandwich is almost constant though the thickness of the NiO buffer layer changes, and is around $27 \times 10^{-8} \Omega \cdot \text{m}$. Therefore, the amounts of the resistivity changes of the sandwiches with the NiO buffer layers are smaller than that of the sandwich without the NiO buffer layer.

2. Film structures of sandwiches

The structures of the sandwiches were investigated by X-ray diffraction. Figure 4 shows X-ray diffraction patterns of the sandwiches. The figure also indicates the positions of the fcc-(111) diffraction peaks of the single-layered films of the Ni-Fe-Co, Co, Cu and Fe-Mn layers. As shown in the figure, we observed one broad diffraction peak around 44 degrees for each sandwich. It is thought that the each peak consists of the diffraction peaks of the Ni-Fe-Co, Co, Cu and Fe-Mn layers. In this study, the diffraction peaks can not be distinguished because they exist at about the same positions. The (111) diffraction peaks of the NiO layers exist at around 36.5 degrees. Therefore, the observed peaks around 44 degrees do not include the diffraction peaks of the NiO layers. As shown in the figure, the intensity of the (111) diffraction peak, which consists of the (111) diffraction peaks of the Ni-Fe-Co, Co, Cu and Fe-Mn layers, is weak in the sandwich without the NiO buffer. The intensi-

Fig. 3 Relationship between the thickness of the NiO buffer layer and the magnetoresistance ratio in Cu(5.0 nm)/Fe-Mn(10.0 nm)/Ni-Fe-Co(3.0 nm) / Co(2.0 nm) / Cu(3.0 nm) / Co(1.0 nm) / Ni-Fe-Co(9.0 nm)/NiO(t nm)/Si sandwiches with various NiO buffer layer thicknesses t .

Fig. 4 X-ray diffraction patterns of Cu(5.0 nm)/Fe-Mn(10.0 nm)/Ni-Fe-Co(3.0 nm) / Co(2.0 nm) / Cu(3.0 nm) / Co(1.0 nm) / Ni-Fe-Co(9.0 nm)/NiO(t nm)/Si sandwiches with various NiO buffer layer thicknesses t .

ties of the (111) diffraction peaks are relatively strong in the sandwiches on the NiO buffer layers. Figure 4 also shows the full width at half maximum (FWHM) of the (111) diffraction peaks. The FWHM of the (111) diffraction peak is 1.74 degrees in the sandwich without the NiO buffer. The FWHM becomes small when the sandwiches are formed on the NiO buffer layers. From the above results, we think that the (111) texture of Ni-Fe-Co, Co, Cu and Fe-Mn layers is relatively strong in the sandwiches formed on the NiO buffer layers.

We also made multilayers, which had the structures of [Ni-Fe-Co(1.5 nm)/Cu(2.2 nm)] $\times$ 10/NiO(0–20 nm)/Si(100), to observe the roughness of the interfaces among the layers in the multilayers with/without the NiO buffer layers. We changed the thickness of the NiO buffer layer formed between the multilayers and the Si(100) substrate. We measured low angle X-ray diffraction patterns of the multilayers, and observed the 1st and the 2nd order diffraction peaks due to the superlattice periodicity. We measured the intensity ratios I_{2nd}/I_{1st} , where I_{1st} and I_{2nd} are the intensities of the 1st order peak and the 2nd order peak, respectively.

Figure 5 shows the I_{1st} , the I_{2nd} and the intensity ratio I_{2nd}/I_{1st} as functions of the thickness of the NiO buffer layer in the multilayers. As shown in the figure, the I_{1st} decreases as the thickness of the NiO buffer layer increases below 10 nm, and slightly increases as the thickness of the NiO buffer layer increases above 10 nm. On the contrary, the I_{2nd} increases as the thickness of the NiO

Fig. 5 The intensity of the 1st order diffraction peak I_{1st} , the intensity of the 2nd order diffraction peak I_{2nd} and the intensity ratio I_{2nd}/I_{1st} as a function of the thickness of the NiO buffer layer in the multilayers in $[\text{Ni-Fe-Co}(1.5 \text{ nm})/\text{Cu}(2.2 \text{ nm})] \times 10/\text{NiO}(0-20 \text{ nm})/\text{Si}(100)$ multilayers.

buffer layer increases below 10 nm, and slightly decreases as the thickness of the NiO buffer layer increases above 10 nm. From the above results, the intensity ratio I_{2nd}/I_{1st} is low in the multilayer without the NiO buffer layer, and increases as the thickness of the NiO buffer layer increases below 10 nm. The intensity ratio decreases again as the thickness of the NiO buffer layer increases above 10 nm. It is thought that the NiO buffer layer decreases the roughnesses of the interfaces between the Ni-Fe-Co layers and the Cu layers in the multilayers.

IV. Discussion

1. Exchange bias fields applied to magnetic bilayers

As shown in Fig. 1, the NiO buffer layers increase the exchange bias fields applied to the magnetic bilayers in the sandwiches. In order to understand the increase of the exchange bias field, we observed the high angle X-ray diffraction patterns of the sandwiches. As shown in Fig. 4, the NiO buffer layers increase the intensities of the (111) diffraction peaks which consist of the diffraction peaks of the Ni-Fe-Co, Co, Cu and Fe-Mn layers, and decrease the FWHM of the (111) diffraction peaks. From the results, we think that the (111) textures of the Ni-Fe-Co, Co, Cu and Fe-Mn layers in the sandwiches with the NiO buffer layers are stronger than that for the sandwich without the NiO buffer layer.

It has been reported that the exchange bias field becomes high when the (111) texture of the Ni-Fe/Cu/Ni-Fe layers is strong in the Fe-Mn/Ni-Fe/Cu/Ni-Fe sandwiches with the various buffer layer materials⁽⁵⁾. This is because the (111) textures increase the Fe-Mn crystallites which have γ -Mn structures. The Fe-Mn which has the γ -Mn structure is antiferromagnetic at room temperature⁽⁷⁾⁽⁸⁾. Therefore, the magnetic layer is strongly biased by the Fe-Mn layer in the sandwiches with the strong (111) textures. On the other hand, the Fe-Mn crystallites

have α -Mn structures when the sandwich does not have the (111) texture. The Fe-Mn which has the α -Mn structure is not antiferromagnetic at room temperature⁽⁷⁾⁽⁸⁾. Therefore, the magnetic layer is not biased by the Fe-Mn layer in the sandwich without the (111) texture. From the above results for the reported sandwiches, it is also thought that the relatively strong (111) texture increases the Fe-Mn crystallites which have the γ -Mn structures, and thus increases the exchange bias field applied to the magnetic bilayer in this study.

2. Decrease in the magnetoresistance ratio in sandwiches with NiO buffer layers

Though the NiO buffer layers increase the exchange bias fields applied to the magnetic bilayers as shown in Fig. 1, the magnetoresistance ratios become low as shown in Fig. 3. In order to understand the decrease in the magnetoresistance ratio, we investigated the interface roughnesses of the multilayers which had the structures of $[\text{Ni-Fe-Co}(1.5 \text{ nm})/\text{Cu}(2.2 \text{ nm})] \times 10/\text{NiO}(0-20 \text{ nm})/\text{Si}(100)$. As shown in Fig. 5, the NiO buffer layers increase the intensity ratios I_{2nd}/I_{1st} of the multilayers. It is thought that the NiO buffer layers decrease the roughnesses of the interfaces between the Ni-Fe-Co layers and the Cu layers in the Ni-Fe-Co/Cu multilayers.

Of course, the multilayers do not have the Co layers, and their stacking structures are different from those of the sandwiches. However, the relatively flat interfaces were thought to be caused by the flat layers in the multilayers, and it is thought that the NiO layers made the relatively flat layers in the multilayers. Thus, we think that the NiO buffer layers may also make the relatively flat layers, and decrease the roughnesses of the interfaces between the Co layers and the Cu layers in the sandwiches.

It has been reported that the roughness determined by low angle X-ray diffraction has a good correlation with the magnetoresistance ratio⁽⁹⁾, and the increase in the interface roughness between the magnetic layers and the nonmagnetic layers increases the magnetoresistance ratio⁽¹⁰⁾⁻⁽¹²⁾. On the contrary, it has been also reported that the increase in the interface roughness decreases the magnetoresistance ratio⁽⁹⁾⁽¹³⁾⁻⁽¹⁴⁾. However, the decrease in the magnetoresistance ratio is accompanied by the increase in the remanent magnetization in the above reports⁽⁹⁾⁽¹⁴⁾. When the remanent magnetization is high, the magnetizations of the magnetic layers do not change between parallel and antiparallel sufficiently. It decreases the magnetoresistance ratios of the multilayers.

In this study, the magnetizations of the two magnetic bilayers of the sandwiches change between parallel and antiparallel sufficiently as shown in Fig. 1. Therefore, it is thought that the changes in the magnetoresistance ratio are not accompanied by the changes in the remanent magnetization in this study. Therefore, it is likely that the NiO buffer layers decrease the interface roughnesses between the Co layers and the Cu layers, and thus decrease the magnetoresistance ratios of the sandwiches.

In order to more understand the relationship between the interface roughness and the magnetoresistance ratio,

we investigated the multilayers which had the structures of $[\text{Ni-Fe-Co}(1.5 \text{ nm})/\text{Cu}(2.2 \text{ nm})] \times 10/\text{NiO}(10 \text{ nm})/\text{Hf}(0-10 \text{ nm})/\text{Si}(100)$. The Hf buffer layers were inserted to change the interface roughnesses between the Ni-Fe-Co and the Cu layers.

Figure 6 shows the I_{1st} , the I_{2nd} and the intensity ratio I_{2nd}/I_{1st} as functions of the thickness of the Hf buffer layer in the multilayers. As shown in the figure, the I_{1st} increases as the thickness of the Hf buffer layer increases below 1 nm, and decreases as the thickness of the Hf buffer layer increases above 1 nm. On the contrary, the I_{2nd} decreases as the thickness of the Hf buffer layer increases below 1 nm, slightly increases as the thickness of the Hf buffer layer increases between 1 and 5 nm, and slightly decreases as the thickness of the Hf buffer layer increases above 5 nm. From the above results, the intensity ratio I_{2nd}/I_{1st} is high in the multilayer without the Hf buffer layer, and low in the multilayer with the Hf buffer layer. It is thought that the Hf buffer layer increases the roughnesses of the interfaces between the Ni-Fe-Co layers and the Cu layers in the multilayers. The lowest intensity ratio is obtained when the thickness of the Hf buffer layer is 1 nm. The interfaces may be the roughest when the thickness of the Hf buffer layer is 1.0 nm.

The above results are obtained in the multilayers which do not have the Co layers. Their staking structures are differ from those of the sandwiches. However, we can expect that the Hf buffer layers cause the rough interfaces also in the sandwiches.

Therefore, we made the sandwiches which have the structures of $\text{Cu}(5.0 \text{ nm})/\text{Fe-Mn}(10.0 \text{ nm})/\text{Ni-Fe-Co}(3.0 \text{ nm})/\text{Co}(2.0 \text{ nm})/\text{Cu}(3.0 \text{ nm})/\text{Co}(1.0 \text{ nm})/\text{Ni-Fe-Co}(9.0 \text{ nm})/\text{NiO}(10.0 \text{ nm})/\text{Hf}(0-10 \text{ nm})/\text{Si}(100)$. Figure 7 shows the relationship between the thickness of the Hf buffer layer and the magnetoresistance ratio in the sandwiches. The Hf buffer layers increase the magnetoresistance ratios of the sandwiches. The highest mag-

Fig. 6 The intensity of the 1st order diffraction peak I_{1st} , the intensity of the 2nd order diffraction peak I_{2nd} and the intensity ratio I_{2nd}/I_{1st} as a function of the thickness of the Hf buffer layer in $[\text{Ni-Fe-Co}(1.5 \text{ nm})/\text{Cu}(2.2 \text{ nm})] \times 10/\text{NiO}(10.0 \text{ nm})/\text{Hf}(0-10 \text{ nm})/\text{Si}$ multilayers.

Fig. 7 Relationship between the thickness of the Hf buffer layer and the magnetoresistance ratio in $\text{Cu}(5.0 \text{ nm})/\text{Fe-Mn}(10.0 \text{ nm})/\text{Ni-Fe-Co}(3.0 \text{ nm})/\text{Co}(2.0 \text{ nm})/\text{Cu}(3.0 \text{ nm})/\text{Co}(1.0 \text{ nm})/\text{Ni-Fe-Co}(9.0 \text{ nm})/\text{NiO}(10.0 \text{ nm})/\text{Hf}(0-10 \text{ nm})/\text{Si}$ sandwiches.

netoresistance ratio is obtained when the thickness of the Hf buffer layer is 1.0 nm.

From the above results, we think that the Hf buffer layers may increase the roughnesses of the interfaces between the Co layers and the Cu layers, and thus increase the magnetoresistance ratios of the sandwiches in this study.

V. Conclusions

$[\text{Fe-Mn}/\text{Ni-Fe-Co}/\text{Co}/\text{Cu}/\text{Co}/\text{Ni-Fe-Co}/\text{NiO}/\text{Si}]$ and $[\text{Fe-Mn}/\text{Ni-Fe-Co}/\text{Co}/\text{Cu}/\text{Co}/\text{Ni-Fe-Co}/\text{NiO}/\text{Hf}/\text{Si}]$ sandwiches with various thicknesses of the NiO and the Hf buffer layers, respectively, were grown. Their magnetoresistance effects and the film structures have been investigated. The following results were obtained.

- (1) The NiO buffer layers make the relatively strong (111) textures in the $\text{Fe-Mn}/\text{Ni-Fe-Co}/\text{Co}/\text{Cu}/\text{Co}/\text{Ni-Fe-Co}$ layers, and increase the exchange bias fields applied to the magnetic bilayers.
- (2) The NiO buffer layers decrease the magnetoresistance ratios of the sandwiches.
- (3) The Hf buffer layers increase the magnetoresistance ratios of the sandwiches. The highest magnetoresistance ratio of 5.0% is obtained when the thickness of the Hf buffer layer is 1.0 nm.

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