Giant Magnetoresistance Effect in Grain-Type Alloy Thin Films: II.

—Annealing and Double-Layering—

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An annealing study suggested that the internal diffusion and interfacial diffusion of Ag atoms in ferromagnetic FeCo grains made negative and positive contributions, respectively, to obtaining a large magnetoresistance (MR) ratio. The magnetic saturation field (H_s) associated with MR characteristics decreased in proportion to the decrease in resistivity as the annealing temperature and/or annealing time increased. The coercive force in double-layered films with constant Co-Ag thickness decreased with increasing permalloy layer thickness. This indicates that the two layers are magnetically coupled. However, the H_s was maintained to be extremely large. The MR ratio in the double-layered films was occasionally larger than that in a Co-Ag single layer.

Key words: grain, magnetoresistance, magnetic saturation field, annealing, double-layered structure

1. Introduction

Recently, it has been observed that a giant magnetoresistance (GMR) effect is present in 'grain-type' alloy thin films comprising mutually insoluble magnetic and nonmagnetic metals and/or alloys.^{1)~9)} These results demonstrated that the GMR effect is not restricted to multilayered structures and they showed that additional opportunities exist for technological applications. However, the magnetic saturation field (H_s) associated with the MR characteristics, which is important in the application in MR head devices, was too large in such grain systems. Here, one of the reasons for the large H_s is considered to be the existence of a Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between ferromagnetic grains through the mediation of the polarization wave in conduction band. It is extremely important that the magnetic coupling energy is weak in practice because the RKKY interaction has the spatial dependence $\cos(2\kappa_F D_{nm})/D_{nm}^3$, in which κ_F is the Fermi

wave vector and D_{nm} the distance between the magnetic spins. In addition, the dilution effect arising from the diffusion of nonmagnetic atoms in the ferromagnetic grains, which has been observed by X-ray diffraction,⁹⁾ further weakens the RKKY interaction. Therefore, a large magnetic field is necessary for complete alignment of the magnetization vectors of all the ferromagnetic grains. This magnetic feature is reflected in the large H_s in the grain-type alloy thin films.

From the above discussion, annealing of the grain-type alloy thin films should reduce the H_s . This is because the mutual insolubility of magnetic and nonmagnetic components effectively eliminates the interdiffusion between grains. In the present study, therefore, annealing under various conditions was explored to decrease the H_s in the grain-type alloy thin films. In addition, permalloy is found to possess soft magnetic characteristics. This means that it quickly responds to external magnetic field. Therefore, double-layered structure which is composed of the permalloy and grain-type alloy layers was also fabricated to improve the H_s . When both layers are magnetically coupled with each other, the H_s is expected to become small for the entire system. We report here the results of structural, magnetic, and MR studies on the annealed and double-layered films.

2. Experiment

Deposited $(\text{Fe}_{100-x}\text{Co}_x)_{20}\text{Ag}_{80}$ films ($x=20$ and 60 wt%) with 300 nm thicknesses were annealed at various temperatures and/or various time under a vacuum of 1×10^{-5} Torr. The samples were heated to set temperatures (T_s) at a rate of $10^\circ\text{C}/\text{min}$, and were then held at the T_s for set time (P_s). They were then cooled back to room temperature. In the system

Fig. 1 Structural characteristics of $(\text{Fe}_{80}\text{Co}_{20})_{20}\text{Ag}_{80}$ thin films annealed under various conditions.

used for double-layered structure preparation, the sputter sources were widely separated and the substrates moved past the sources, a single layer being deposited on each pass by a source. First, Co-Ag composite target (99.9% purity) was sputtered using RF method. The permalloy layer, $\text{Ni}_{80}\text{Fe}_{20}$ (99.9% purity), was continuously formed onto the Co-Ag layer by dc magnetron sputtering. Other experimental methods have been described previously.¹⁰⁾

3. Annealing Effects

By carefully controlling the temperature, the separation between the FeCo and Ag grains was found to sharply increase at about 400°C, as shown in Fig. 1. After the drastic structural change at 400°C, the Ag(111) lattice further extended with increasing annealing time, indicating that the substitution of atoms occurs continuously in the Ag-rich grains. In contrast, the lattice parameter of the FeCo grains changed little under these annealing conditions. This suggests that the Fe and/or Co atoms, which were eliminated from the Ag-rich phases, are absorbed by the pure FeCo grains that formed. Based on these experimental results, the structural changes that occur with annealing is expected to progress in two steps. Namely, the structure of the FeCo grains initially becomes distinct, and then they are gradually grown in the second step.

Accompanied with the improvement of grain structure, the MR and magnetic characteristics of FeCo-Ag thin films changed as shown in Figs. 2 and 3, respectively. Here the MR ratio is displayed as follows:

$$\Delta R/R_{\min} = (R(H) - R_{\min})/R_{\min}.$$

where a minimum in resistance, $R_{\min}$, was observed under a magnetic field (H) of ± 1 T. Since saturation of the MR and the magnetization was not observed in the measuring magnetic field range, the intrinsic H_s and saturated magnetization (M_s) values cannot be estimated. In this report, therefore, the H_s is defined as the value of the magnetic field corresponding to the MR ratio at a value 80% below each MR ratio, as shown in Fig. 4. The M_s is estimated from the magnetization value at $H=1$ T. In addition, coercive force (H_c^*) associated with the MR characteristics is evaluated from the value of the magnetic field corresponding to the maximum in the MR (see Fig. 4). The H_c^* is directly proportional to the H_c , which is obtained from the magnetic hysteresis loop.

The MR ratio increased until an annealing temperature of 400°C was reached. However, when the annealing temperature became higher and/or the annealing time became longer, the MR ratio decreased. The increase and decrease in the MR ratio correlate with the increase in M_s and $H_c (=H_c^*)$, respectively. The M_s drastically increased when the structure of the FeCo grains became clear due to the annealing, as shown in Figs. 1 and 3. This occurs because the dilution effect arising from the internal diffusion of the Ag atoms disappeared. Accordingly, the spin-dependent scattering for conduction electrons is enhanced, resulting in the increase in the MR ratio. However, the M_s did not change with further annealing. Under these annealing conditions, therefore, the contribution of the H_c is noticeable in the change in the MR ratio. The increase in the H_c value is due to the increase in the FeCo grain size, and indicates that the ferromagnetic grains possess a

Fig. 2 MR characteristics of $(\text{Fe}_{80}\text{Co}_{20})_{20}\text{Ag}_{80}$ thin films annealed under various conditions.

Fig. 3 Magnetic characteristics of $(\text{Fe}_{80}\text{Co}_{20})_{20}\text{Ag}_{80}$ thin films annealed under various conditions.

single-domain structure. Here it is extremely important that such growth in the magnetic grains is equivalent to the progress in the separation between the FeCo and Ag components as discussed for the structural characteristics. That is, appropriate diffusion between the magnetic and nonmagnetic atoms at the grain boundary is expected to be necessary for the appearance of the GMR effect in the grain-type alloy thin films. In summary, though the internal diffusion of the nonmagnetic atoms in the ferromagnetic grains negatively contributes to the realization of the large MR ratio, the interfacial diffusion between the magnetic and nonmagnetic grains possesses an opposite effect.

As shown in Fig. 4, annealing at an appropriate time and temperature is an effective way to decrease the H_s while increasing the MR ratio. Here the change in the H_s possesses a strong correlation with the decrease in the resistivity (ρ) arising from the improvement in the crystallinity (see Figs. 1 and 2). This result supports our idea that the RKKY interaction between the ferromagnetic grains dominates the H_s in the grain-type alloy thin films. Specifically,

since the decrease in ρ induces an increase in the mobility of the conduction electrons, the RKKY interaction between the ferromagnetic grains becomes stronger, resulting in a rise in the magnetic response for the entire system. However, the complete saturation of the MR could not be achieved at magnetic fields less than 1 T, even when the annealing conditions were fully optimized. This suggests the presence of free magnetic grains that are not in the RKKY interaction's network.

4. Double-Layering Effects

In the Co-Ag system, the best result in the MR characteristics has been obtained in the $\text{Co}_{19}\text{Ag}_{81}$ film with a maximum MR ratio as high as 19.4%.¹⁰⁾ However, as shown in Fig. 5(a), when the $\text{Co}_{19}\text{Ag}_{81}$ was selected as the composition in the grain-layer, the magnetic moments of both layers in the double-layered films were individually magnetized, indicating the absence of interlayer magnetic coupling. This occurs because the Co concentration is low. Therefore, in order to enhance the magnetic interaction with permalloy layer, the Co concentration in

Fig. 4 MR curves of (a) as-deposited and (b) annealed $(\text{Fe}_{40}\text{Co}_{60})_{20}\text{Ag}_{80}$ thin films.

the grain-layer was dared to be large (40 wt%), resulting in the somewhat small MR ratio (about 7%). Figs. 5(b)~(d) show the M - H curves of the double-layered films with various permalloy layer thicknesses, in which the $\text{Co}_{40}\text{Ag}_{60}$ layers were held constant at 68 nm. The H_c was found to decrease as the permalloy layer thickness increased. Since there was no structural change for the Co-Ag and permalloy layers were magnetically coupled with each other. The $\text{Co}_{40}\text{Ag}_{60}:\text{Ni}_{80}\text{Fe}_{20}$ thickness ratio dependence in the H_c is shown in Fig. 6. The critical layer thickness ratio, above which the H_c drastically decreases, is seen to be present in the samples with the 68 nm-thick Co-Ag layer. Though the H_c changed little in the Co-Ag : Ni-Fe thickness ratio smaller than 2, the M - H curves indicated the presence of the interlayer magnetic coupling. Namely, in this range, the Co-Ag layer magnetically dragged the permalloy one. On the other hand, in the materials containing 39 nm-thick Co-Ag layer, the H_c decreased as soon as the permalloy layer was added. In addition, even if the thickness ratio was the same, the samples containing the thinner Co-Ag layer possessed the lower

Fig. 5 M - H curves at room temperature of (a) $\text{Co}_{19}\text{Ag}_{81}$ (68 nm)/ $\text{Ni}_{80}\text{Fe}_{20}$ and (b)~(e) $\text{Co}_{40}\text{Ag}_{60}$ (68 nm)/ $\text{Ni}_{80}\text{Fe}_{20}$ double-layered films with various $\text{Ni}_{80}\text{Fe}_{20}$ layer thicknesses ($t_{\text{Ni-Fe}}$). $t_{\text{Ni-Fe}}$: (a) 501 nm; (b) 0 nm; (c) 170 nm; (d) 446 nm.

H_c . These indicate that the magnetic coupling energy between the Co-Ag and permalloy layers decreases on increasing the distance from the interface.

Figure 7(b) shows the MR curve of 50 nm-thick Co-Ag single-layer film. When the permalloy layer was formed onto this grain-layer under the same condition, the MR ratios in the double-layered films prepared were larger or smaller than that in the single layer, as shown in Figs. 7(a) and (c). In the grain-layer, the RKKY interaction is present between ferromagnetic grains through the mediation of the polarization wave in conduction band. Since the sign of the RKKY interaction alternates with the change in the distance between the magnetic grains, a random magnetic state may be produced by a proper

Fig. 6 Coercive force in $\text{Co}_{40}\text{Ag}_{60}/\text{Ni}_{80}\text{Fe}_{20}$ double-layered films in relation to the Co-Ag:permalloy thickness ratio. $\text{Co}_{40}\text{Ag}_{60}$ layer thickness: 39 nm ($\square$); 68 nm ($\circ$).

combination of ferromagnetic and antiferromagnetic RKKY interactions under zero-field. The MR is expected to decrease when the random orientation changes to a ferromagnetic arrangement as the external magnetic field increases. To explain the increase of the MR ratio in the double-layered films, therefore, we note the magnetic domain structure in the permalloy layer. Namely, under zero-field, the complexity in the domain structure further increases the randomness in the magnetic state in the grain-layer. On the other hand, the decrease in the MR ratio was quantitatively explained by the model that the double-layered structure is equivalent to a parallel circuit. Specifically, the magnetic field dependence of the resistance of the double-layered films, $R_{\text{NF/CA}}(H)$, can be displayed as follows:

$$R_{\text{NF/CA}}(H) = R_{\text{NF}}(H) \cdot R_{\text{CA}}(H) / (R_{\text{NF}}(H) + R_{\text{CA}}(H)),$$

where $R_{\text{NF}}(H)$ and $R_{\text{CA}}(H)$ were experimentally estimated in each single-layer film.

5. Summary

The internal diffusion of the nonmagnetic atoms in the ferromagnetic grains, which induced the decrease in the M_s , negatively contributed to the realization of the large MR ratio. The interfacial diffusion between the magnetic and nonmagnetic grains, which is related to the spin-dependent scattering for

Fig. 7 Typical MR curves of (a), (c) Co-Ag/Ni-Fe double-layered films and (b) a Co-Ag single-layer film.

the conduction electrons at the grain boundary, possessed an opposite contribution. The H_s associated with the MR characteristics was strongly correlated with ρ , suggesting that the RKKY interaction between the ferromagnetic grains dominates the H_s in the grain-type alloy thin films. The MR ratio in the double-layered films was occasionally larger than that in the Co-Ag single layer. Though the H_c decreased with increasing permalloy layer thickness, indicating the presence of the interlayer magnetic coupling, the H_s was maintained to be large.

From the application point of view, it is important that annealing at an appropriate time and temperature is effective for decreasing the H_s with increasing MR ratio. It is also extremely important that the increase in the MR ratio in the double-layered structure indirectly reduces the H_s .

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