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Annealing effects on the magnetoresistance characteristics of grain-type alloy thin films

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Abstract. Structural, magnetoresistance (MR), and magnetic characteristics were studied on FeCo–Ag grain-type alloy thin films annealed under various conditions. Internal diffusion and interfacial diffusion of the Ag atoms in the ferromagnetic FeCo grains possessed negative and positive contributions to obtaining a large MR ratio, respectively. The magnetic anisotropy field (H_k) associated with the MR characteristics decreased in accordance with the decrease in resistivity as the annealing temperature and/or annealing time increased. This suggests that the Ruderman–Kittel–Kasuya–Yosida interaction between the ferromagnetic grains dominates the H_k in such systems.

When two kinds of metal that are mutually insoluble are evaporated or sputtered at the same time, each metal is known to individually form fine grains in the prepared film [1]. Recently, it has been observed that a giant magnetoresistance (GMR) effect is present in such 'grain-type alloy thin films' comprising magnetic and non-magnetic metals [2–7]. 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 anisotropy field (H_k) associated with the MR characteristics, which is important in the application of MR head devices, was too large in such grain systems. From our studies on rare-earth superlattices [8–10], one of the reasons for the large H_k 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 the 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 wavevector and D_{nm} the distance between the magnetic spins. In addition, the dilution effect arising from the diffusion of non-magnetic atoms induces a decrease in magnetic moments in the ferromagnetic grains. This further weakens the RKKY interaction between the magnetic grains. 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_k in the grain-type alloy thin films.

As described above, the inter-grain diffusion is considered to be one of the causes of the large H_k. Here, annealing of the grain-type alloy thin films should improve (i.e. reduce) the H_k. This is because the mutual insolubility of magnetic and non-magnetic components effectively eliminates the interdiffusion between grains. In the present study, therefore, annealing under various conditions was explored to provide further insight into the MR characteristics of the grain-type alloy thin films. We report here the results of detailed structural, MR, and magnetic studies on the annealed films.

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The samples used were deposited $(Fe_{1-x}Co_x)_{0.2}Ag_{0.8}$ films (x = 0.2 and 0.6: weight fraction) with 3000 Å thicknesses, in which MR ratios larger than 10% at room temperature in the magnetic field range of ±1 T were obtained. The preparation procedure has been described previously [7]. The annealing was carried out at various temperatures and for various times under a vacuum of 1×10^{-5} Torr. The samples were heated to set temperatures (T_s) at a rate of 10 °C min^{-1}, and were then held at T_s for a set time (P_s). They were then cooled back to room temperature. The structure of each annealed sample was characterized by x-ray diffraction (XRD) with a Rigaku RAD-2C x-ray diffractometer using Cu Kα radiation. The MR was measured at room temperature, about 293 K, in a four-terminal geometry with an in-plane direct current of 1 mA. A magnetic field up to 1 T was applied parallel to the current. Magnetization and hysteresis loops were obtained at room temperature using a vibrating sample magnetometer (TOEI VSM-3S).

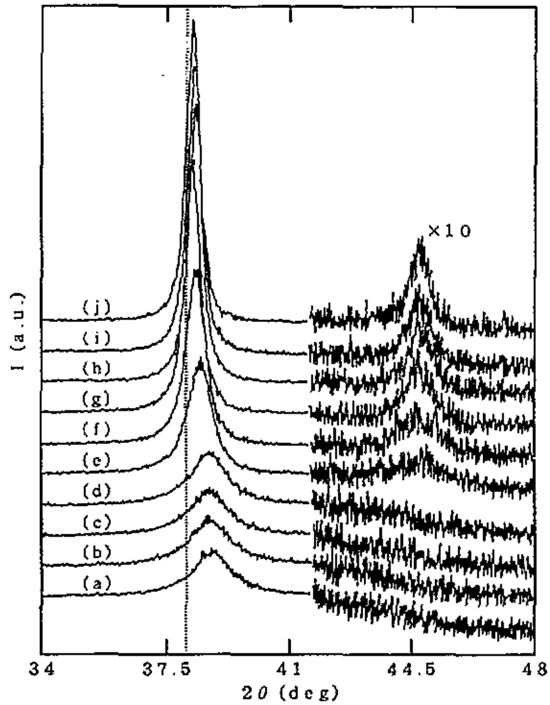

Figure 1. XRD patterns of $(Fe_{0.8}Co_{0.2})_{0.2}Ag_{0.8}$ thin films annealed under various conditions: (a) as-deposited; (b) T_s = 100 °C, P_s = 0 min; (c) 200, 0; (d) 300, 0; (e) 400, 0; (f) 400, 10; (g) 400, 40; (h) 400, 70; (i) 400, 100; (j) 400, 150. The broken line indicates the known position of the Ag(111) diffraction peak.

As shown in figure 1, the diffraction arising from the FeCo grains was not present in the XRD pattern of the as-deposited films. Instead, the Ag(111) diffraction was shifted to a higher angle than the characteristic position of the bulk material. Though the FeCo alloy and the Ag metal are mutually insoluble, the grain structure is concluded to be incomplete in the films deposited at room temperature, indicating that the Fe and/or Co atoms substitute for some of the Ag sites. This occurs because the migration effect of the atoms is low. As the annealing temperature and/or annealing time increased, the Ag(111) peak approached the usual position in accordance with the appearance of a new peak due to the [110]-oriented FeCo grains. This indicates that annealing makes the grain structure distinct. The preferred orientation of (111) and (110) planes observed in the Ag and FeCo grains, respectively, occurs because they are the most densely close-packed planes in the FCC-Ag and BCC-FeCo lattices. By carefully controlling the temperature, the separation between the FeCo and Ag grains was found to increase sharply at about 400 °C, as shown in figure 2. 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 are expected to progress in two steps: initially the structure of the FeCo grains becomes distinct, and then they gradually grow.

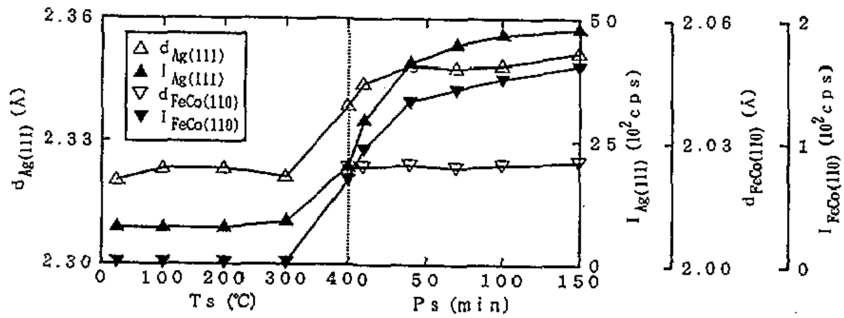

Figure 2. Structural characteristics of $(Fe_{0.8}Co_{0.2})_{0.2}Ag_{0.8}$ thin films annealed under various conditions.

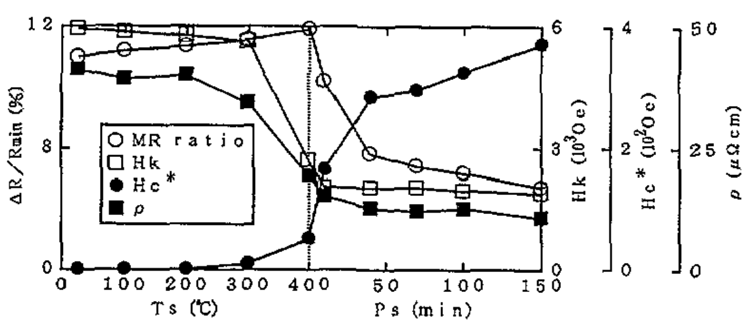

Figure 3. MR characteristics of $(Fe_{0.8}Co_{0.2})_{0.2}Ag_{0.8}$ thin films annealed under various conditions.

Together with the improvement of grain structure, the MR and magnetic characteristics of FeCo–Ag thin films change as shown in figures 3 and 4, respectively. Here the MR ratio is displayed as follows:

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

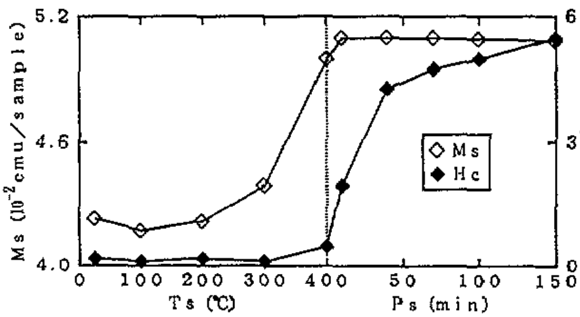

Figure 4. *Magnetic characteristics of* $(Fe_{0.8}Co_{0.2})_{0.2}Ag_{0.8}$ *thin films annealed under various conditions.*

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 [7], the intrinsic H_k and saturated magnetization (M_s) values cannot be estimated. In this work, therefore, the H_k 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 figure 5. The M_s is estimated from the magnetization value at $H = 1$ T. In addition, the coercive field (H_c^*) associated with the MR characteristics is evaluated from the value of the magnetic field corresponding to the maximum in the MR (see figure 5). The H_c^* is in direct proportion to the H_c, which is obtained from the magnetic hysteresis loop.

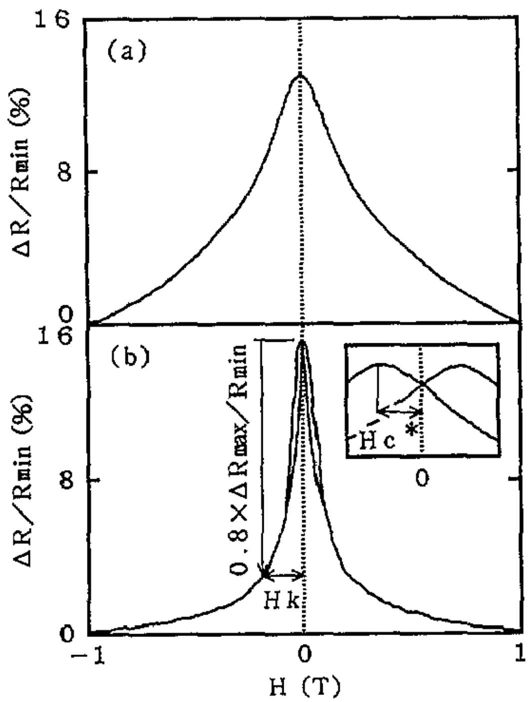

Figure 5. MR curves of (a) as-deposited and (b) annealed $(Fe_{0.4}Co_{0.6})_{0.2}Ag_{0.8}$ thin films.

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 increases 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 figures 2 and 4. This occurs because the dilution effect arising from the internal diffusion of the Ag atoms disappears. 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, suggesting that an increase in the grain size leads to little change in the band structure in this system. 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 single-domain structure [11]. 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 non-magnetic 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 non-magnetic atoms in the ferromagnetic grains negatively contributes to the realization of the large MR ratio, the interfacial diffusion between the magnetic and non-magnetic grains produces an opposite effect.

As shown in figure 5, annealing at an appropriate time and temperature is an effective way to reduce the H_k while increasing the MR ratio. Here the change in the H_k possesses a strong correlation with the decrease in the resistivity (ρ) arising from the improvement in the crystallinity (see figures 2 and 3). This result supports our idea that the RKKY interaction between the ferromagnetic grains dominates the H_k 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 alignment of the magnetization vectors 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 ferromagnetic grains that are not included in the RKKY interactions.

Similar structural, MR, and magnetic changes induced by annealing were also observed in the $(Fe_{1-x}Co_x)_{1-y}Ag_y$ and $(Ni_{1-x}Fe_x)_{1-y}Ag_y$ thin films with various x and y values. Here all the samples were prepared by using the vacuum deposition method on the substrates kept at room temperature. Therefore, the annealing effects described above are expected to be generally in effect for the deposited grain-type films. In contrast, the little annealing effect was observed in the grain-type alloy thin films that were sputtered at room temperature. The kinetic energy of sputtered atoms is high and the bombardment by either neutral or ionized inert gas ions results in heating of the substrate surface. Consequently, the migration effect of atoms in the sputtered films is higher than that in the deposited ones. This involves a similar contribution to that seen for annealing, resulting in the reduction in the annealing effects in the sputtered grain-type alloy thin films.

In conclusion, the internal diffusion of the non-magnetic atoms in the ferromagnetic grains, which induces the decrease in the M_s, negatively contributes to the realization of the large MR ratio. The interfacial diffusion between the magnetic and non-magnetic grains, which is related to the spin-dependent scattering for the conduction electrons at the grain boundary, produces an opposite contribution. The H_k associated with the MR characteristics is strongly correlated with ρ, suggesting that the RKKY interaction between the ferromagnetic grains dominates the H_k in the grain-type alloy thin films. From the application point of view, it is extremely important that annealing for an appropriate time and at an appropriate temperature reduces the H_k while increasing the MR ratio.

Acknowledgments

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