Magnetoresistance characteristics of grain-type alloy thin films of various compositions

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Abstract. Grain-type alloy thin films consisting of fine ferromagnetic grains, which are composed of Fe, Co, Ni, or their alloys, in non-magnetic Ag, Bi, or Mg matrices were prepared by a vacuum deposition method. Structural, magnetic, and magnetoresistance characteristics were studied on such films as a function of the composition and concentration. A giant magnetoresistance (GMR) effect was observed over a broad range of magnetic concentrations in the Ag-based systems, except the Ni–Ag one, at room temperature. The appearance of the GMR effect is probably correlated with the Curie temperature of the magnetic component and the atomic radius ratio of the magnetic and non-magnetic components.

1. Introduction

When two kinds of metals that are mutually insoluble are evaporated or sputtered at the same time, each metal is expected individually to form fine grains in the prepared films. Here it is assumed that such a 'grain-type alloy thin film' is composed of magnetic and non-magnetic metals. The Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction is present between magnetic grains through the mediation of the polarization wave in the conduction band. The sign of the RKKY interaction alternates with the change in distance between the magnetic spins [1]. In the grain-type alloy thin films, therefore, a random magnetic state may be produced by a proper combination of ferromagnetic and antiferromagnetic RKKY interactions. From our studies on rare-earth superlattices [2–6], in which a long-range RKKY interaction exists between the magnetic layers, it is expected that the random orientation that exists in the grain-type film changes to a ferromagnetic arrangement as the external magnetic field increases. This magnetic feature is similar to that of a magnetic–non-magnetic superlattice with interlayer antiferromagnetic coupling, such as Fe/Cr, in which a giant magnetoresistance (GMR) effect is present.

From the structural and magnetic points of view described above, we have studied the MR characteristics of grain-type alloy thin films [7, 8]. Similar studies have been carried out by some groups based on a different strategy [9–17]. In the present study, the grain-type alloy thin films were prepared by a vacuum deposition method for various insolubility systems. We report here their MR characteristics as a function of the composition and concentration.

2. Experiment

Fe, Co, Ni, and their alloys were selected as the magnetic component in the grain-type alloy thin films. In order to study the magnetic component dependence, Ag was an obvious choice because all magnetic metals and alloys are totally or almost totally immiscible in Ag under equilibrium conditions [18]. For the non-magnetic component dependence study, Mg and Bi were used as the counterpart of Fe. The magnetic and non-magnetic components (99.9% purity) were deposited at the same time onto a glass substrate under vacuum at 1×10^{-6} Torr using a conventional deposition apparatus with an ultimate pressure of 5×10^{-7} Torr. The substrates were at room temperature prior to the deposition. No attempt was made to regulate the temperature nor was the substrate temperature measured during the deposition. The film thickness was monitored by use of a quartz oscillating thickness sensor. Deposition was continued until a total thickness of 100 nm was reached. The structure of each sample was characterized by x-ray diffraction (XRD) with a RIGAKU RAD-2C x-ray diffractometer using Cu Kα radiation. A wavelength dispersive x-ray spectrometer (JEOL JXA-840), in which the diameter of the electron beam used was 1 μm, installed into a scanning electron microscope was used for the elemental analyses. The MR was measured at room temperature, about 293 K, in a four-terminal geometry with an in-plane direct current of between 0.01 and 1 mA. A magnetic field (H) of up to 1.5 T was applied parallel to the current. Magnetization (M) and hysteresis loops were measured using a vibrating sample magnetometer (TOEI VSM-3S).

3. Magnetic concentration dependence

MR curves observed in the prepared grain-type alloy thin films can be classified into four cases, as shown in figure 1. When the concentration of the magnetic component was too small, no magnetic field dependence was observed in the electronic transport properties (case (a)). Here, only the Bi-based films, in which the magnetic concentration is similarly small, possess a MR curve as shown in case (a'). In contrast, an anisotropic MR (AMR) effect, which is known in the ordinary ferromagnetic materials, was present in the films with large magnetic concentrations (case (d)). The GMR effect was observed in the samples with appropriate magnetic concentrations (cases (b) and (c)). In case (c), however, the observed GMR effect involved the AMR effect. This suggests that the ferromagnetic grains link up with each other in a part of the film since the magnetic concentration was somewhat large. The appropriate magnetic concentration for the appearance of the GMR effect is dependent on the composition of the grain-tyupe alloy thin films, as shown in figure 2. Here note that the MR related to the GMR effect changes negatively with increasing magnetic field. In contrast, since the magnetic field is applied parallel to the current, the AMR effect possesses a positive MR change. In addition, as described below, the MR change for the GMR effect is inactive in comparison with that for the AMR effect, indicating that the magnetic anisotropy field (H_k) associated with the GMR characteristics is extremely large. These are the important differences between the GMR and AMR effects.

Figure 3 shows the MR curves for the Co–Ag films at various Co concentrations. Since saturation of the MR was not observed in the present study, the H_k-value cannot be estimated, but it must be larger than 1.5 T. One of the reasons for the large H_k is considered to be the existence of the 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
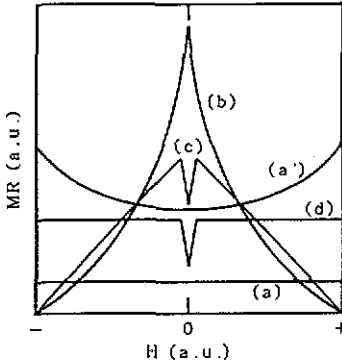
Figure 1. Typical MR curves for grain-type alloy thin films. The concentration of magnetic component is: (a), too small; (a′), too small in Bi-based films; (b), appropriate; (c), somewhat large; and (d), too large for the appearance of the GMR effect.
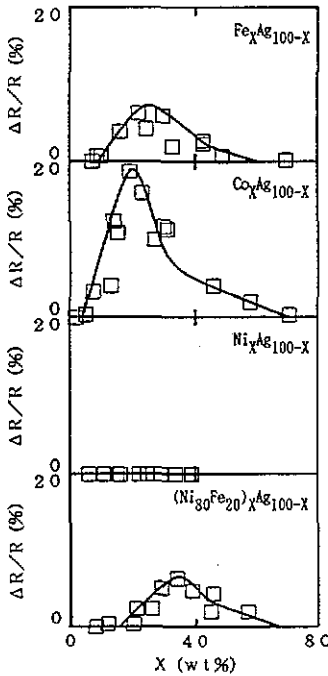

Figure 2. MR ratios at room temperature for (Fe, Co, Ni, $Ni_{80}Fe_{20})_X Ag_{100-X}$ thin films at various magnetic concentrations.

$\cos(2\kappa_F D$ nm$)/D$ nm^3, in which κ_F is the Fermi wavevector and D nm the distance between the magnetic spins. The dilution effect arising from the diffusion of Ag atoms and/or the size effect known for fine particles [19] induce a dispersion of magnetic spins in the ferromagnetic grains. This further weakens the RKKY interaction between the magnetic grains. In addition, the free ferromagnetic grains that are not in the RKKY interaction's network possess a superparamagnetic nature. Therefore, a large magnetic field is necessary for complete alignment of the magnetization vectors for all the ferromagnetic grains. This

magnetic feature reflects on the large H_k-value in the grain-type alloy thin films. As shown in figure 3, the MR change becomes steep as the Co concentration increases. The XRD measurements indicated that these films possess similar structural characteristics, for example crystallinity and diffusion state. Therefore, the difference in shape between three MR curves is due to that in the Co concentration. This supports our idea that the RKKY interaction between the ferromagnetic grains dominates the H_k value. Namely, as the magnetic concentration in the grain-type alloy thin films increases, the distance between the ferromagnetic grains becomes short, resulting in a stronger RKKY interaction.
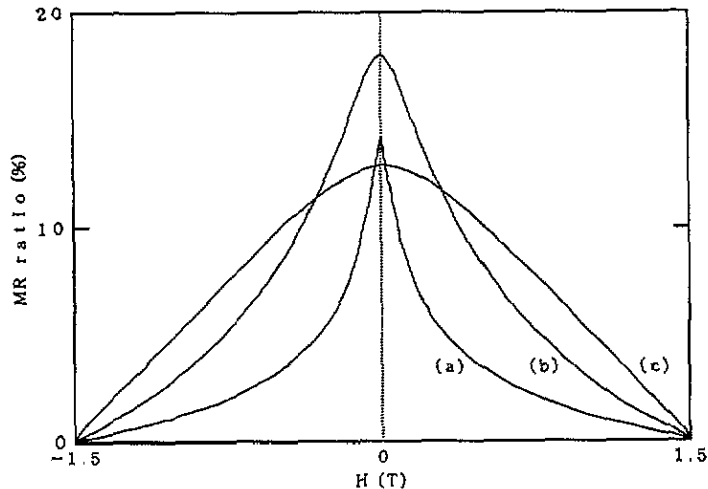

Figure 3. MR curves for $Co_X Ag_{100-X}$ thin films. (a), $X = 30$ wt%; (b), $X = 20$ wt%; (c) $X = 13$ wt%.

4. Magnetic component dependence

In order to ignore the AMR effect and estimate only the GMR effect, the MR ratio is displayed as follows in the present study:

$$\Delta R/R = [R_{max} - R(1.5)]/R(1.5)$$

where $R(1.5)$ is the MR at $H = 1.5$ T. Since R_{max} is equal to $R(1.5)$, the MR ratio in cases (a), (a$'$) and (d) of figure 1 is estimated to be zero. In other cases, R_{max} is observed at the coercive field where the magnetization is zero (see figure 4 in [7]). The MR data at room temperature obtained for all the Ag-based samples that were prepared are shown in figure 2. Starting from the dilute limit, the MR ratio rises as the magnetic concentration (X) increases. This behaviour is attributed to the increasing concentration of magnetic scattering centres. The best result in each system was obtained in the $Fe_{22}Ag_{78}$, $Co_{19}Ag_{81}$ and $(Ni_{80}Fe_{20})_{34}Ag_{66}$ films with a maximum MR ratio as high as 6.6, 18.7 and 6.5%, respectively. The MR ratios in the grain-type alloy thin films with appropriate magnetic concentrations were significantly larger than the AMR ratios in the ordinary magnetic films. This indicates that the GMR effect

is observed over a broad range of magnetic concentrations in all the systems except for Ni–Ag at room temperature. After reaching the peak, the MR ratio starts to decrease. As described above, this is due to the increasing probability of coalescence of the ferromagnetic grains.

From the experimental results described above, the question arises as to why the materials containing Ni metal were the only ones not to have the GMR effect at room temperature. The $M-H$ hysteresis loop measurement indicated that this appears reasonable because the Ni–Ag films are not ferromagnetic at room temperature, though the Ni concentration is large enough. Here the dilution effect due to the diffusion of the non-magnetic Ag atoms in the Ni grains can be considered as one of the reasons for this. In fact, the diffraction arising from the Ni grains was not present in the XRD patterns of as-deposited films. Instead, the Ag(111) diffraction was shifted to a higher angle compared with the position known for the bulk material. This indicates that the Ni atoms substitute for some of the Ag sites. This is the same, however, for another three systems (Fe–Ag, Co–Ag and permalloy–Ag) and cannot account for the absence of the GMR effect in the Ni–Ag films. Such a structural characteristic of the prepared grain-type alloy thin films occurs because the migration effect of atoms in the films deposited at room temperature is small. In addition, it is known that the Ni and Ag metals are mutually only slightly soluble [18]. Figure 4 shows the relation between the maximum MR ratio obtained in each system and the Curie temperature of each magnetic component in the bulk state. Here the MR data of $Fe_Y Co_{100-Y}$–Ag ($Y = 40$ and 80 wt%) films obtained in a previous study [7] are also described in this figure. It is found that the maximum MR ratio and the Curie temperature correlate with each other. Therefore, we note the difference in the Curie temperature between the magnetic components in the grain-type alloy thin films as one of the reasons why the maximum MR ratio is dependent on the magnetic component.
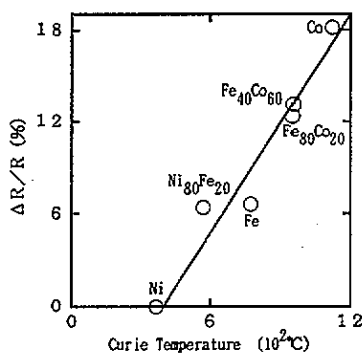

Figure 4. Maximum MR ratios for Ag-based grain-type alloy thin films of vrious magnetic components versus the Curie temperature of each magnetic component in the bulk state.

5. Non-magnetic component dependence

In this section, the composition in the samples is expressed in at.% to clarify the discussion. Figure 5 shows the MR curves for $Fe_{53}Bi_{47}$ and $Fe_{68}Mg_{32}$ thin films with the maximum MR ratio in each system. In the Fe–Bi film, the AMR effect is always superposed on
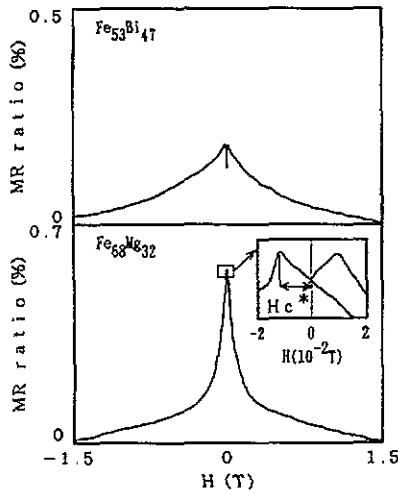
Figure 5. MR curves for $Fe_{53}Bi_{47}$ and $Fe_{68}Mg_{32}$ thin films.

the GMR one. Since this phenomenon was observed even in the samples with small Fe concentrations, the presence of the AMR effect cannot be explained by networking of the ferromagnetic grains, as discussed for figure 1(c). Therefore, we note the difference in resistivity (ρ) between the Fe and Bi metals. In their bulk states, ρ_{Fe} (= 9.71 $\mu\Omega$ cm) is smaller than ρ_{Bi} (=116 $\mu\Omega$ cm). If this relation is maintained between both metals with a grain structure, conduction electrons pass through the Fe grains, resulting in the observation of the AMR effect. On the other hand, a significant hysteresis is observed in the MR curve of the $Fe_{68}Mg_{32}$ film. As shown in figure 5, the coercive field (H_c^*) associated with the MR characteristics, which shows the amplitude of the hysteresis, is evaluated from the value of the magnetic field corresponding to the maximum in the MR. H_c^* is directly proportional to H_c, which is obtained from the $M-H$ hysteresis loop. As the Fe concentration increases, the H_c^*-value increases. Therefore, the increases in H_c^* (= hysteresis amplitude) is due to the increase in the Fe grain size, and indicates that the ferromagnetic grains possess a single-domain structure [20].

The MR data at room temperature obtained for the Fe–Ag, Fe–Bi and Fe–Mg thin films at various Fe concentrations are shown in figure 6. It is found that the maximum MR ratios in the Fe–Bi and Fe–Mg systems are smaller than that in the Fe–Ag system, though the magnetic component is the same for all three systems. This indicates that the MR characteristics of the grain-type alloy thin films possess a strong non-magnetic component dependence. We consider the difference in the atom size between the component metals as one of the reasons for this. In the Fe–Ag system, the ratio in their atomic radii (r_A/r_{Fe}) is 1.16. In contrast, r_{Bi}/r_{Fe} and r_{Mg}/r_{Fe} are 1.25 and 1.29, respectively. As described above, the phase separation is incomplete in the films deposited at room temperature because the migration effect of atoms is small. When internal diffusion of the non-magnetic atoms occurs in the Fe grains, the Curie temperature is expected to decrease. Such a dilution effect arising from the diffusion phenomenon becomes stronger as the atomic radius ratio between the components in the grain-type alloy film increases. This is reasonable because the structural and magnetic disorder are enhanced by the diffusion of the larger atoms. Here it is extremely important that a decrease in the Curie temperature induces a decrease in the magnetization. Accordingly, in comparison with the Fe–Ag sysem, the spin-dependent scattering for conduction electrons is decreased in the Fe–Bi and Fe–Mg systems, resulting
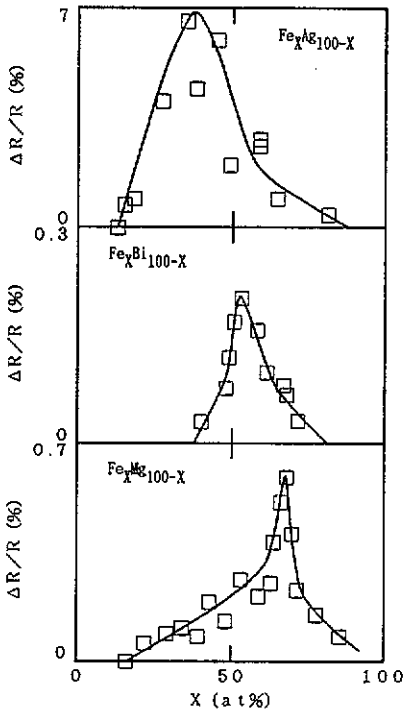
Figure 6. MR ratios at room temperature for $Fe_X(Ag, Bi, Mg)_{100-X}$ thin films at various magnetic concentrations.

in a small MR ratio. As shown in figure 6, the Fe concentration corresponding to the maximum MR ratio in each system becomes larger with increasing atomic radius ratio. These experimental results support the above discussion. Specifically, in the Fe–Mg system, the large Fe concentration is necessary for cancelling the dilution effect due to the Mg atoms.

6. Summary

The GMR effect was observed over a broad range of magnetic concentrations in the Ag-based grain-type alloy thin films containing Fe, Co, their alloys or permalloy, which were prepared by a vacuum deposition method. The best result was obtained in the $Co_{19}Ag_{81}$ film with a maximum MR ratio of as high as 18.7% at room temperature in the magnetic field region between -1.5 and 1.5 T. The H_k value associated with the GMR characteristics is dominated by the RKKY interaction between the ferromagnetic grains. A high Curie temperature in the magnetic component and a small atomic radius ratio between the magnetic and non-magnetic components are likely to be important for the appearance of the GMR effect.

References

[1] Ruderman M and Kittel C 1954 *Phys. Rev.* **96** 99
Kasuya T 1956 *Prog. Theor. Phys.* **16** 45
Yosida K 1957 *Phys. Rev.* **106** 893

[2]   Maeda A, Satake T, Fujimori T, Tajima H, Kobayashi M and Kuroda H 1989 *J. Appl. Phys.* **65** 3845

[3]   Maeda A, Satake T, Fujimori T and Kuroda H 1990 *J. Phys.: Condens. Matter* **2** 245

[4]   Maeda A, Satake T, Fujimori T and Kuroda H 1991 *J. Phys.: Condens. Matter* **3** 1967

[5]   Maeda A, Satake T and Kuroda H 1991 *J. Phys.: Condens. Matter* **3** 5241

[6]   Maeda A, Satake T and Kuroda H 1991 *Phys. Lett.* **157A** 178

[7]   Maeda A, Kume M, Oikawa S, Shimizu Y and Doi M 1993 *J. Phys.: Condens. Matter* **5** L189

[8]   Maeda A, Kume M, Oikawa S, Shimizu Y and Doi M 1993 *J. Phys.: Condens. Matter* **5** 4641

[9]   Berkowitz A E, Mitchell J R, Carey M J, Young A P, Zhang S, Spada F E, Parker F T, Hutten A and Thomas G 1992 *Phys. Rev. Lett.* **68** 3745

[10] Xiao J Q, Jiang J S and Chien C L 1992 *Phys. Rev. Lett.* **68** 3749

[11] Barnard J A, Waknis A, Tan M, Haftek E, Parker M R and Watson M L 1992 *J. Magn. Magn. Mater.* **114** L230

[12] Jiang J S, Xiao J Q and Chien C L 1992 *Appl. Phys. Lett.* **61** 2362

[13] Xiao J Q, Jiang J S and Chien C L 1992 *Phys. Rev.* B **46** 9266

[14] Carey M J, Young A P, Starr A, Rao D and Berkowitz A E 1992 *Appl. Phys. Lett.* **61** 2935

[15] Tsoukatos A, Wan H, Hadjipanayis G C and Li Z G 1992 *Appl. Phys. Lett.* **61** 3059

[16] Zhang S 1992 *Appl. Phys. Lett.* **61** 1855

[17] Xiong P, Xiao G, Wang J Q, Xiao J Q, Jiang J S and Chien C L 1992 *Phys. Rev. Lett.* **69** 3220

[18] Hansen M 1958 *Constitution of Binary Alloys* (New York: McGraw-Hill)

[19] Frei E H, Shtrikman S and Treves D 1958 *Phys. Rev.* **109** 1522

[20] Luborsky F E and Paine T O 1960 *J. Appl. Phys.* **31** 68S