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LETTER TO THE EDITOR

Giant magnetoresistance effect in grain-type alloy thin films

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Abstract. FeCo–Ag thin films with a grain structure, in which the FeCo alloy and the Ag metal are mutually insoluble, were prepared by a vacuum deposition method. The maximum magnetoresistance (MR) ratio observed in these films was 13.0% at room temperature in the magnetic field region between −1 and 1 T. Here the features of the MR curve corresponded well with those of the magnetization curve. This indicates the appearance of a giant MR effect in such grain-type alloy thin films. Though the magnetic anisotropy field (Hk) associated with the MR characteristics that were obtained is extremely large (>1 T), proper annealing was found to be effective for decreasing the Hk value without causing a drop in the MR ratio.

Monolayer-by-monolayer growth of single-crystal thin films has been successfully accomplished as a result of the recent progress in epitaxial techniques, as typified by molecular beam epitaxy. However, it is still difficult to suppress the diffusion between successive layers in the superlattice structure [1, 2]. In particular, superlattices with clean interfaces cannot be prepared using a sputtering method. This occurs because ion bombardment during synthesis provides knock-on mixing between layers. Recently, a giant magnetoresistance (GMR) effect has been observed in such sputtered metallic superlattices. Since the interdiffusion phenomenon strongly influences the physical properties of the superlattices that are grown, the origin of the GMR is believed to be an interfacial spin-dependent scattering [3–6].

In this letter, we propose that superlattices with diffused interlayers are equivalent to an aggregation of grains with various sizes. Here it is assumed that such a 'grain-type thin film' is composed of magnetic and non-magnetic metals; the Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction is expected to be present between magnetic grains through the mediation of the non-magnetic grains. The sign of the RKKY interaction alternates with the change in the distance between the magnetic spins [7]. In grain-type 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 [8–12], 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 the GMR effect is present. This idea prompted us to study the MR characteristics

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of grain-type thin films. Here it should be noted that GMR effect has been observed very recently in heterogeneous Co–Cu alloy films at low temperatures [13, 14].

In the present study, FeCo alloy and Ag metal, which are mutually insoluble, were selected as the components to prepare the grain-type thin films. Reported here are the results from analysis of the structures and the MR characteristics.

$Fe_{100-x}Co_x$ alloy (99.9%: $x = 0$, 20, 40 and 60) and Ag metal (99.9%) were deposited at the same time onto a glass substrate under vacuum at 2×10^{-5} Torr using a conventional deposition apparatus with an ultimate pressure of 2×10^{-6} Torr. The substrate was kept at ambient temperature during growth. The film thickness was monitored by use of a quartz oscillating thickness sensor. Deposition was continued until a total thickness of 3800 Å was reached. The deposition rates of both sources were adjusted to yield a film composition of $(Fe_{100-x}Co_x)_1Ag_4$. The structure of each sample that was prepared was characterized by using 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) installed into a scanning electron microscope was used for the elemental analysis. The MR was measured at room temperature, about 293 K, in a four-terminal geometry with an in-plane direct current (J) of 12 mA. A magnetic field (H) up to 1 T was applied perpendicular ($H \perp J$) and parallel ($H \parallel J$) to the current or perpendicular to the film plane ($H \top J$). The magnetization and hysteresis loop were measured using a vibrating sample magnetometer (Toei VSM-3S).

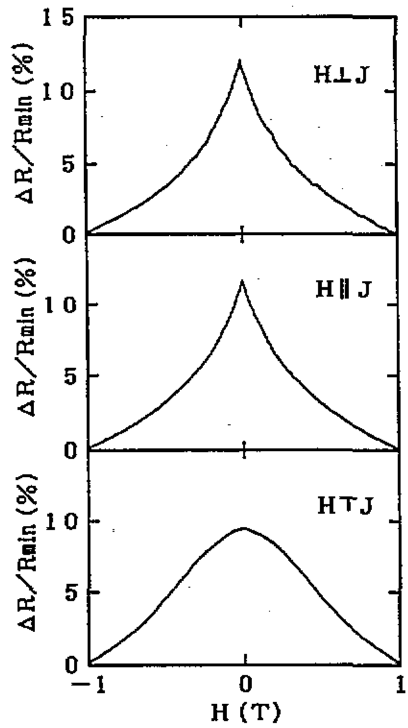

Figure 1. Magnetoresistance curves of $(Fe_{80}\,Co_{20})_1Ag_4$ thin films at room temperature with various arrangements between the external magnetic field (H) and the sense current (J).

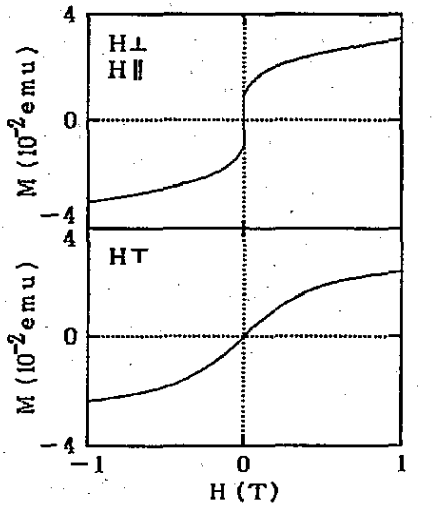

Figure 2. Magnetization curves of $(Fe_{80}Co_{20})_1Ag_4$ thin films at room temperature with various directions of the external magnetic field.

The MR and magnetization curves at room temperature measured in the magnetic field region between −1 and 1 T are shown in figures 1 and 2. The features of the MR curve correspond well with those of the magnetization curve. The MR data are displayed as

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

where a minimum in resistance, R_{min}, was observed at $H = \pm 1$ T in the applied magnetic field range. The grain-type alloy thin film that was prepared shows an MR ratio larger than 10%. In the present study, the maximum MR ratio obtained was 13.0% for the $(Fe_{40}Co_{60})_1Ag_4$ film at room temperature. Here it is worth noting that saturation of the MR was not observed in the applied magnetic field as shown in figure 1. This shows that the intrinsic MR ratios are larger than the evaluated values. Our studies on other grain-type thin films, for example NiFe–Ag, suggest that the MR characteristics observed in FeCo–Ag are not unique to that system. These indicate the appearance of a GMR effect in such grain-type alloy thin films, thus demonstrating that the GMR is not an inherent feature of superlattice systems. It is found that the MR characteristics observed are independent on the arrangements between H and J. That is, under the condition where the magnetic field was applied perpendicular to the current ($H \perp J$), the MR ratio was 12.2% in contrast to 11.8% for a field applied parallel to the current ($H \parallel J$). This implies that the GMR observed involves an anisotropic MR effect in the ordinary ferromagnetic materials. On the other hand, for the arrangement in which the field is applied perpendicular to the film plane ($H \top J$), the MR ratio decreased to 10.2% and the shape of the MR curve was different from those of the others. This occurs because of the demagnetizing field. Similar behaviour is also observed in the magnetization curve (see figure 2). From the above results, the GMR effect in the grain-type alloy thin film is concluded to have an isotropic nature after the correction of the demagnetizing field effect.

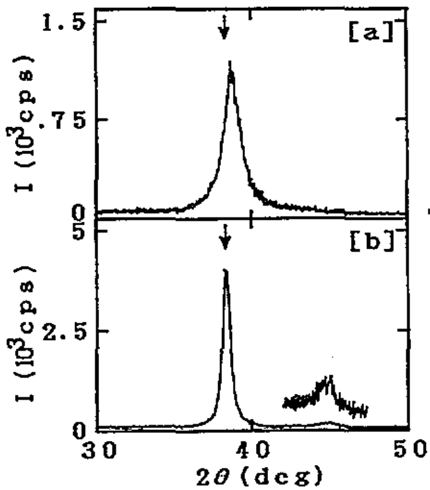

Figure 3. XRD patterns of as-deposited (*a*) and annealed (*b*) $(Fe_{80}Co_{20})_1Ag_4$ thin films. The arrows indicate the known position of the Ag(111) diffraction peak.

The magnetic anisotropy fields (Hk) associated with the MR characteristics that were obtained are extremely large, with values the same as those in superlattices with interlayer antiferromagnetic coupling. Since saturation of the MR was not observed in the present study, the Hk value cannot be estimated, but it must be larger than 1 T. Several reasons for the large Hk can be considered as follows. Though the FeCo alloy and the Ag metal are mutually insoluble, the grain structure is expected to be incomplete in the films deposited at room temperature. This will occur because the migration effect of atoms is low. In fact, the diffraction arising from the FeCo grains is not present in the XRD pattern as shown in figure 3(*a*). Instead, the Ag(111) diffraction is shifted to a higher angle compared with the position known for the bulk material. This indicates that the Fe and/or Co atoms substitute for some of the Ag sites. In such films, the grains possess various FeCo:Ag composition ratios and various magnetic characters. That is, the FeCo-rich phases are ferromagnetic and the grains containing many Ag atoms are non-magnetic or paramagnetic. Here the RKKY interaction exists between the ferromagnetic grains through the mediation of the non-magnetic and/or paramagnetic grains. It is extremely important that the 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. The dilution effect arising from the diffusion of Ag atoms and/or the size effect known for fine particles [15] 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 Hk is necessary for complete alignment of the magnetization vectors of all the ferromagnetic grains.

As described above, the inter-grain diffusion is considered to be one of the causes of the large Hk. Since the mutual insolubility of the FeCo alloy and the Ag metal is effective for elimination of interdiffusion between successive grains, annealing of the grain-type films should improve the Hk. Figure 3(*b*) shows the XRD pattern of

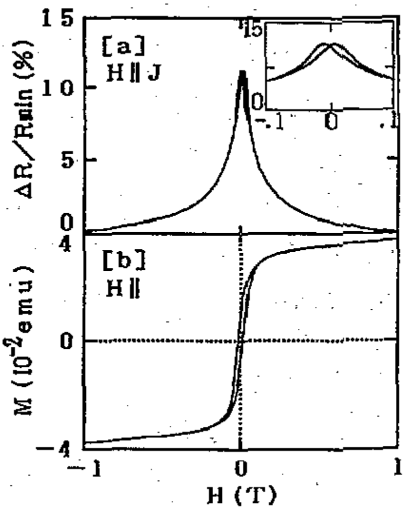

Figure 4. Magnetoresistance (*a*) and magnetization (*b*) curves of annealed $(Fe_{80}Co_{20})_1Ag_4$ thin films at room temperature.

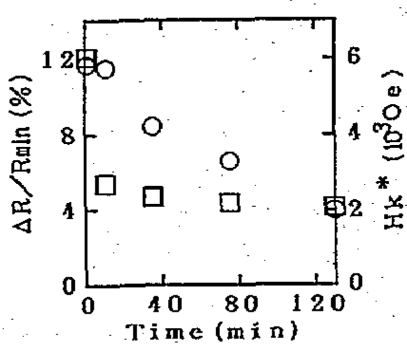

Figure 5. MR ratios and magnetic anisotropy fields at room temperature of $(Fe_{80}Co_{20})_1Ag_4$ thin films annealed at 773 K for various times. Hk^* is defined as the value of the magnetic field corresponding to the MR ratio at a value 80% below each MR ratio.

the FeCo–Ag film annealed at 773 K for 10 min under vacuum at 1×10^{-5} Torr. The Ag(111) peak is shifted to a lower angle compared with the as-deposited film. This is accompanied by the appearance of a new peak due to the FeCo grains. This indicates that the annealing makes the grain structure distinct. The MR and magnetization curves at room temperature for the annealed sample are shown in figure 4. In comparison with figure 1, the Hk is found to be decreased by annealing. Though proper annealing is effective for decreasing the Hk value without changing the MR ratio, the increasing annealing time and/or temperature cause a decrease in the MR ratio (see figure 5). This implies that appropriate diffusion between the magnetic and non-magnetic grains is necessary for the appearance of the GMR effect. Here we note that a decrease in the diffusion induces a change in the grain size. As shown in figure 3, the crystallinity of the thin film becomes higher after annealing. Therefore, the origin of the significant hysteresis observed in the MR and magnetization curves of the annealed samples is likely to be due to magnetocrystalline anisotropy associated with the change of the grain size [16]. In addition, annealing for the $(Fe_{80}Co_{20})_1Ag_4$ film causes the resistivity to drop from 53.5 to 19.5 $\mu\Omega$ cm and there is a 20% increase in the magnetization. These support the idea that there is improvement in the crystallinity and a decrease in the dilution effect, respectively.

In summary, the GMR effect was observed in grain-type alloy thin films consisting of FeCo-rich precipitate particles in a Ag-rich matrix. This indicates that the GMR effect is not an inherent feature of superlattice systems. Annealing at an appropriate temperature for an appropriate time was found to be an effective way to decrease the Hk value. The interdiffusion between the magnetic and non-magnetic grains is important for the appearance of the GMR effect.

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