Transactions of the Materials Research Society of Japan 24 [1] 89-92 (1999)

Multilayer Effect on Granular-type Giant Magnetoresistance of Co-Cu Film

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Abstract: We investigated the multilayer effect on the granular-type giant magnetoresistance (GMR) of Co-Cu films. The granular structure (cluster size and position) was controlled by annealing and by formation of a multilayered structure. Alloy films and multilayers were fabricated by rotating substrates at high speed and at low speed, respectively. Granular-type GMR property depended on Co composition and annealing temperature. The maximum MR ratio was observed in an as-sputtered alloy film containing 19.7 at% cobalt. The granular-type MR ratio decreased as the periodic thickness of the multilayer increased. This can be explained in terms of a composition shift in local areas and an increase in ineffective area. On the other hand, alloy films containing little (12.7 at%) cobalt showed that the change in MR ratio with annealing was dependent on the type of GMR, which depended on Cu layer thickness. The increase in MR ratio in granular-type areas is attributed to the increase in size of the Co clusters to the nanometer scale after annealing. The decreased MR ratio in multilayer-type areas can be explained by a decrease in interlayer magnetic exchange coupling as the multilayered structure was destroyed by annealing.

Keywords: giant magnetoresistance, granular type, ferromagnetic cluster, multilayered structure

1. INTRODUCTION

The giant magnetoresistance (GMR) phenomenon is based on the spin-dependent scattering of conduction electrons at the interface between nonferromagnetic and ferromagnetic matters [1]. MR ratios of over ten percent at room temperature have been reported. GMR materials are classified into two types: multilayer and granular systems. The multilayer type consists of ferromagnetic layers separated by a nonferromagnetic layer, while the granular type has a structure in which ferromagnetic clusters are dispersed in a nonferromagnetic matrix [2], [3]. The GMR effect occurs when the magnetization direction of the ferromagnetic matters is arranged by applying an external magnetic field. A granular type GMR material, as shown in Fig. 1, consists of ferromagnetic clusters (Co) dispersed in a nonferromagnetic (Cu) matrix. The magnetization vectors of each Co cluster are random, without any magnetic field. The electrical resistance decreases with arranging the magnetization vectors to parallel by the applied field. The generally reported fabrication method for granular GMR materials is for the ferromagnetic clusters to be precipitated by annealing a supersaturation solid solution. Granular GMR materials have been fabricated by also other methods, such as by ionized cluster beam [4] and by annealing of a NiFe/Ag multilayer [5]. The ion-

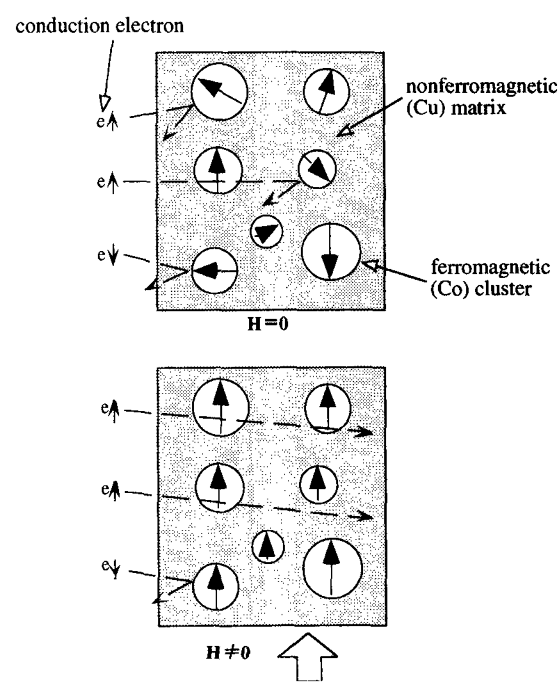

Fig. 1 GMR phenomenon of the granular type.

ized cluster beam method was able to control cluster size in as-deposited films, and the GMR property at low fields was improved in annealed NiFe/Ag multilayers.

In this paper, we investigate granular-type GMR in Co-Cu films. In particular, the granular structure (cluster size and position) was controlled by annealing and by constructing a multilayered structure.

2. EXPERIMENTAL

Co-Cu films were fabricated by dual-source rf magnetron sputtering from separate Co and Cu targets. Co-Cu alloy films and Co/Cu multilayers were fabricated by rotating glass substrates at high speed and at low speed, respectively. The Co composition, analyzed by electron probe microanalysis, was in the range of 11.1 to 26.4 at%. The vacuum annealing conditions were as follows. The vacuum pressure was less than 1×10^{-5} Torr. The temperature ranged from 100 to 600℃ and the time was 10 min. The heating rate was 10℃/min, and the cooling method was air cooling. MR loops were measured by the dc two-point method at room temperature, and a magnetic field (up to 13 kOe) was applied parallel to the measuring current. The MR ratio was defined by the equation: $\Delta\rho=(\rho_0-\rho_S)/\rho_S \times 100$, where ρ_0 and ρ_S are the electrical resistance without a field and with the maximum field. Magnetic property was measured by a vibrating sample magnetometer, with the magnetic field applied up to 15 kOe at room temperature. Film structure was observed by the X-ray diffraction method. (111) and (200) diffraction patterns of the fcc copper phase were measured in all samples.

3. RESULTS AND DISCUSSION

3.1 Co-Cu Alloy Film

The GMR property of the Co-Cu alloy film depended on Co composition and annealing temperature. As-sputtered film exhibited a maximum MR ratio of 2.5% at a 19.7 at% cobalt composition, and the MR ratio decreased with increasing annealing temperature. Alloy films containing little cobalt exhibited an MR ratio that increased with annealing, but the MR ratio did not change with annealing in films containing a lot of cobalt.

Figure 2 shows changes in the MR and magnetization curves before and after annealing at 500℃ for a sample with 19.7 at% Co. The MR ratio decreased from 2.1 to 1.0% with annealing, and the saturated magnetic field also decreased. The decrease in the MR ratio is attributed to the increased size of Co clusters. This is consistent with

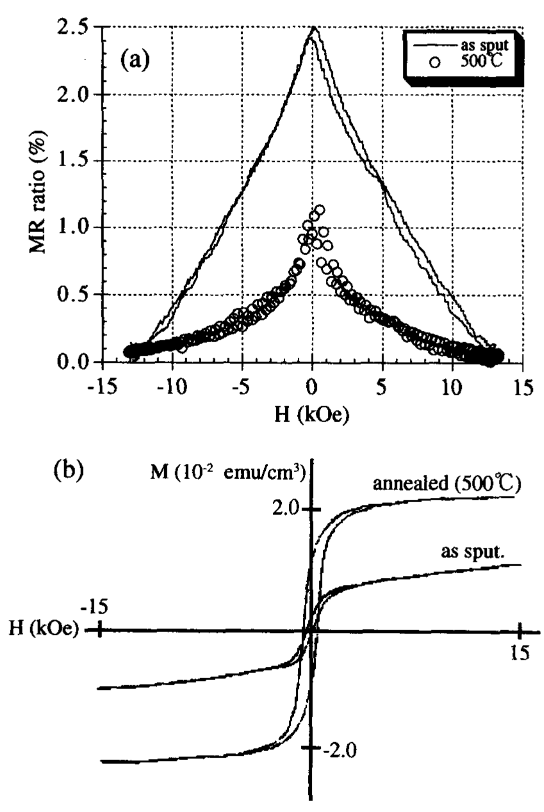

Fig. 2 MR (a) and MH (b) curves of Co-Cu alloy film. (Co composition 19.7 at%)

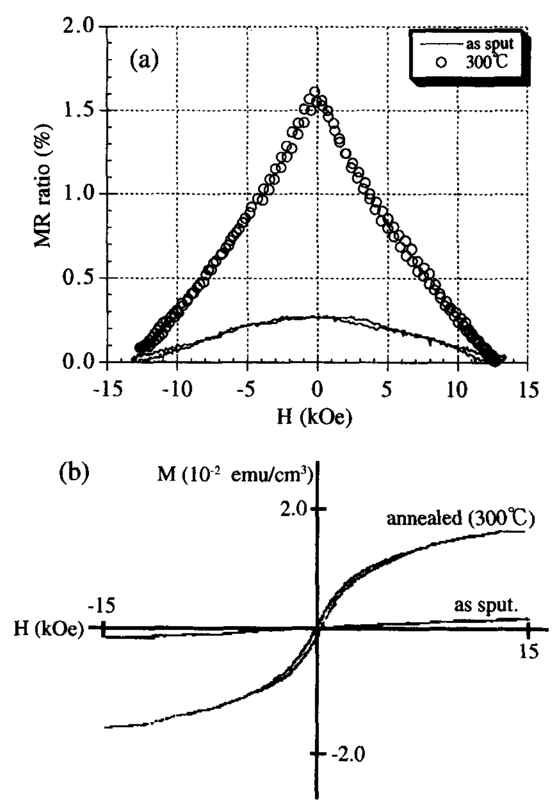

Fig. 3 MR (a) and MH (b) curves of Co-Cu alloy film. (Co composition 12.7 at%)

the magnetization curve shown in Fig. 2(b). Granular material with larger-size ferromagnetic clusters has several disadvantageous effects on GMR property. The interface between the nonferromagnetic and ferromagnetic materials decreases, reducing spin-dependent interface scattering. Furthermore, magnetic percolation will occur in alloy films in which a number of clusters are combined by annealing. With granular-type GMR, the magnetization of the various clusters must be random, and this is not observed in a ferromagnetic single material.

In contrast, GMR property improved with annealing in a Co-Cu alloy film containing 12.7 at% cobalt. As shown in Fig. 3(a), the MR ratio increased from 0.3 to 1.6% with annealing at 300℃. The improved GMR property can be explained in terms of the Co clusters increasing to an appropriate size. Figure 3(b) shows the changes in magnetization curves before and after annealing. Magnetization also increased with annealing, as did the MR ratio. The Co cluster size of the as-sputtered film is on the atomic order, where very small ferromagnetic clusters will display a thermally unstable magnetization, i.e., superparamagnetism. Thus, the magnetization and the MR ratio are very low at room temperature within the measuring field range. The Co clusters increase in size to the nanometer level by precipitating atomic order clusters with annealing, and magnetization and the granular-type GMR effect will then appear.

3.2 Co/Cu Multilayer

Co/Cu multilayers consisting of 19.7 at% cobalt and 12.7 at% cobalt were fabricated by rotating substrates at low speed. The effect of the multilayer on GMR property was then investigated.

Small angle x-ray reflectivity generally oscillates due to multiple reflection and interference in a multilayered structure. Figure 4(a) shows the relationship between the first peak intensity of small angle x-ray reflectivity and Cu layer thickness in an as-sputtered Co/Cu multilayer containing 19.7 at% cobalt. The peak intensity decreases with decreasing the Cu layer thickness. It is found from Fig. 4(a) that a multilayered structure is formed at a Cu thickness of more than about 0.6 nm, and that a granular structure is formed at less than 0.6 nm. A relatively high-qual-

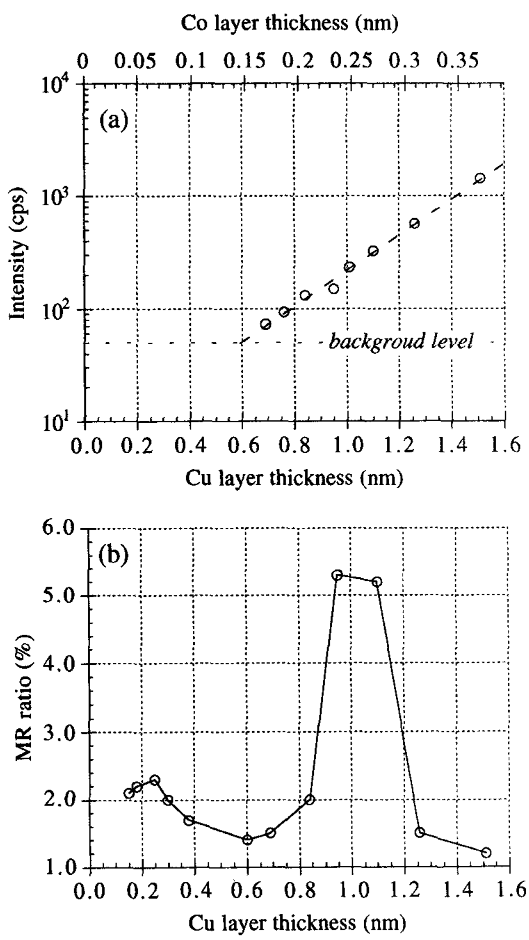

Fig. 4 Intensity of small angle x-ray reflectivity (a) and MR ratio (b) in Co/Cu multilayer film. (19.7 at% Co)

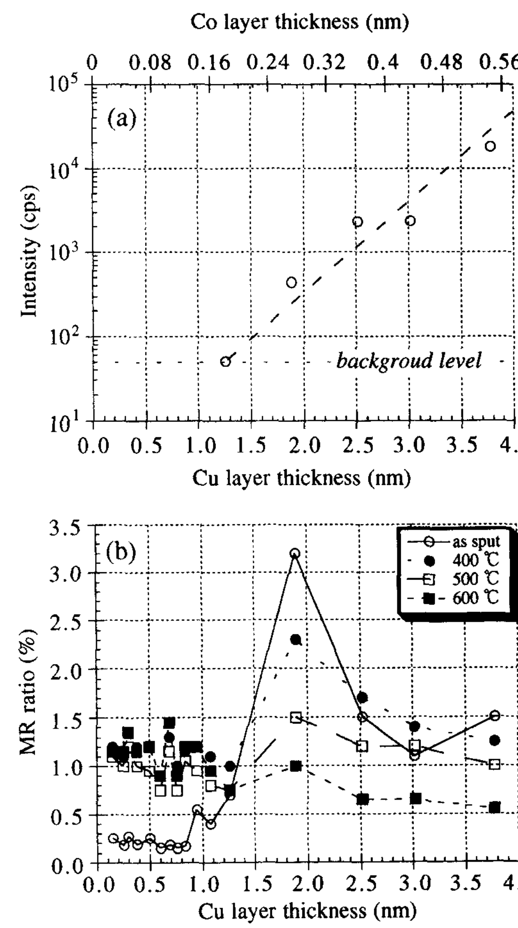

Fig. 5 Intensity of small angle x-ray reflectivity (a) and MR ratio (b) in Co/Cu multilayer film. (12.7 at% Co)

ity multilayered structure appears to have been formed, since the Co layer thickness is thinner than one atomic layer at this periodic length. The MR ratio of the Co/Cu multilayer, as shown in Fig. 4(b), oscillates in the Cu layer thickness range of 0.15 to 1.5 nm. It has been already reported that the MR ratio of multilayer-type GMR material oscillates with a nonferromagnetic layer thickness. This phenomenon is interpreted in terms of interlayer magnetic exchange coupling between ferromagnetic layers separated by a nonferromagnetic layer. The typical periodic thickness of the Cu layer is approximately 1 nm for the Co/Cu multilayer [6], [7]. Thus, the MR ratio peak in a 0.9 nm-thick Cu layer is attributed to the multilayer-type GMR effect. On the other hand, the MR ratio decreases as the Cu layer thickness increases in the range from 0.15 to 0.6 nm. This is attributed to the granular-type GMR effect. A relatively large MR ratio appears at a thinner Cu layer of less than one atomic layer thickness, because a homogeneous granular structure is formed. The MR ratio decreases with increasing the Cu layer thickness. The decrease in MR ratio can be explained by the formation of an inhomogeneous granular structure when the Cu layer thickness increases. The Co composition shifts in local areas and the ineffective area increases. As a result, the granular type GMR effect decreases, and thereafter shifts to the multilayer type with a Cu layer approximately 0.6 nm thick.

Figure 5(a) shows the relationship between the first peak intensity of small angle x-ray reflectivity and Cu layer thickness for an as-sputtered Co/Cu multilayer containing 12.7 at% cobalt. It is found from Fig. 5(a) that a multilayered structure is formed at Cu thickness of more than 1.2 nm, and that a granular structure is formed at less than about 1.2 nm. The as-sputtered Co/Cu multilayer, as shown in Fig. 5(b), exhibits a single MR ratio peak in an approximately 1.8 nm-thick Cu layer. The MR ratio is relatively small with Cu layer thicknesses of less than 0.8 nm, similar to Fig. 3(a), since a granular structure is formed with a low Co composition. The GMR effect shifts from the granular type to the multilayer type at a Cu layer about 1.2 nm thick. An MR ratio peak attributed to interlayer exchange coupling is observed at with a Cu layer about 1.8 nm thick. The first MR peak of the multilayer type was not observed with a Cu layer 1 nm thick, because a continuous Co layered structure was not formed at this periodic thickness. Moreover, it is found from Fig. 5(b) that the change in MR ratio with annealing is dependent on the type of GMR. The MR ratio increased in granular structure areas and decreased in multilayered structure areas, with increasing annealing temperature. The increasing MR ratio in granular areas, as mentioned above, can be explained in terms of the formation of nanometer-scale Co clusters by annealing. On the other hand, the decreasing MR ratio in multilayer areas is attributed to a decrease in interlayer exchange coupling as the multilayered structure is destroyed by annealing.

4. CONCLUSION

We investigated the multilayer effect on granular-type GMR in Co-Cu films. Granular structure (Co cluster size and position) was controlled by annealing and by constructing a multilayered structure. The granular-type GMR property depended on Co composition and annealing temperature. Co clusters appropriate for granular GMR were formed in an alloy film containing 19.7 at% cobalt, where the maximum MR ratio was observed. In a multilayer fabricated by rotating the substrate at low speed, the granular-type MR ratio decreased with increasing periodic thickness. This can be explained in terms of a composition shift in local areas and an increased ineffective area. On the other hand, alloy films containing little (12.7 at%) cobalt exhibited an MR ratio that increased with annealing. Moreover, this change in MR ratio with annealing was dependent on the type of GMR, which depended on Cu layer thickness. The increasing MR ratio in granular type areas is attributed to the fact that Co clusters were of the nanometer scale after annealing. The decrease in MR ratio in multilayer type areas can be explained by the fact that interlayer magnetic exchange coupling becomes small because the multilayered structure was destroyed with annealing.

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(Received December 11, 1998; accepted February 28, 1999)