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Tunneling magnetoresistance and electron spin resonance study on Co–Al–O granular films

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Abstract

In this work, a series of Co–Al–O granular films were prepared using an $Ar–O_2$ reactive rf sputtering system. The optimal magnetoresistance (MR) ratio of 5.1% at room temperature was found in $Co_{44}Al_{23}O_{33}$ sample. After annealing at 300°C, MR ratio can be raised to 6.1%. We also measured the spectra of electron spin resonance for this series of samples and found an interesting correlation between the magnitude of MR ratio and the width of absorption field (ΔH). Our results show that the maximum tunneling magnetoresistance occurs when ΔH is the largest, indicating the important role of magnetic microstructure in the mechanism of granular tunneling. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Tunneling magnetoresistance; Granular film; Electron spin resonance

1. Introduction

The finding of giant magnetoresistance in the sputtered Co–Al–O granular films [1] opened a wide scope for studying the spin-dependent tunneling phenomena, which are closely related to the further applications of spin-electronics. Based on many reports [2–5], the magnitude of such "tunneling-type magnetoresistance (TMR)" depends strongly on the microstructures of samples. The microstructures of the samples are affected by many parameters, such as the composition ratio [2], the oxygen concentration [3], the substrate temperatures [4] and the interlayer roughness [5]. Furthermore, the anomalous temperature and bias-voltage dependence of MR value was observed, which was ascribed to a spin-dependent higher-order tunneling [6]. In the process of a simultaneous tunneling of electrons, an electronic charge is transferred from the charged large grain, via the two small ones, to the neutral large one. In the higher-order tunneling model, the enhancement of MR in the low temperature originates from the product of the probability of each tunneling event. Since the granular materials show a broad distribution of grain-size, this model provides a more precise picture to describe the detailed tunneling process. Yakushiji et al. [2] has pointed out that the geometry standard deviation of the LNDF (log-normal distribution function), estimated from the magnetic analysis, increases with Co-concentration. This suggests that some Co-grains couple ferromagnetically to form a large magnetic grain. Therefore, it is very possible that the mechanism of granular TMR involves not only the geometrical microstructure but also the magnetic microstructures within the films. Unfortunately, the information about the magnetic microstructures for Co–Al–O films is quiet lacking. One of the effective techniques to study the magnetic microstructure is the electron spin resonance (ESR). We thus conduct an ESR study for a series of Co–Al–O granular films and correlate the ESR results with the TMR results to investigate the influence of magnetic anisotropy on the optimization of TMR.

2. Experiments

Co–Al–O granular films were prepared using an $Ar–O_2$ reactive rf sputtering system with a base pressure

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of 10^{-8} Torr. The metal targets were two-inch disks of pure Co and Al. The ratio of Ar/O_2 was 20/1 at the sputtering pressure of 5×10^{-2} Torr. In the present work, we have changed the power of guns to vary the ratio of Co/Al, and the prepared six films are of the composition of $Co_{32}Al_{32}O_{36}$, $Co_{39}Al_{30}O_{31}$, $Co_{41}Al_{29}O_{30}$, $Co_{44}Al_{23}O_{33}$, $Co_{47}Al_{25}O_{28}$ and $Co_{50}Al_{23}O_{27}$. After the depositing, the as-made films were annealed at 300°C. Standard four-probe method was used to measure the resistivity with the applied field sweeping from 0 to 1.5 T. ESR measurements at room temperature were performed using a Bruker EMX spectrometer operating at 9.48 GHz.

3. Results and discussion

The MR ratio is defined as the difference between the resistance at 1.5 T and that at zero field, and divided by the resistance at zero field. Fig. 1 shows the typical plot of MR ratio vs. field. It indicates a sharp decrease of resistance at field <3 kOe, then the decrease rate becomes slower at high field. But it did not saturate at field of 1.5 T. Fig. 2 shows the MR ratio vs. Co-composition for as-deposited and annealed films. It indicates that the MR value has a maximum value of 5% and 6% for as-deposited (open circles) and annealed (closed circles) $Co_{44}Al_{23}O_{33}$ samples, respectively.

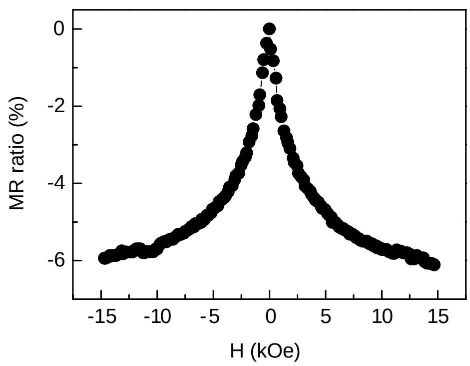

Fig. 1. MR ratio vs. applied field H.

Fig. 3 is a plot of $M\Delta H_\parallel$ vs. Co-concentration, where M is the magnetization and $\Delta H_\parallel$ is the absorption linewidth with the field parallel to the film surface. In principle, the linewidth is the convolution of the distribution of resonance fields with some intrinsic narrow linewidth, and it is proportional to $\delta H_i^2/M$ where H_i is the internal field. Therefore, the quantity stands for the amplitude of the internal field variation, and is more fundamentally related to the broadening mechanism. Fig. 3 shows a sharp arise of $M\Delta H_\parallel$ at 44 at% indicating a strong enhancement of field variation.

Furthermore, we follow the simple equations to calculate the anisotropy constants [7]:

$$(\omega/\gamma)^2 = H(H + 4\pi M - H_A),$$

and

$$(\omega/\gamma) = H_\perp - (4\pi M - H_{A1}),$$

where $H_{A1} = 2K_1/M$, $H_{A2} = 4K_2/M$, and $H_A = H_{A1} + H_{A2}$. $H_\perp$ is the resonance fields with field perpendicular to the film surface; γ is the gyromagnetic ratio, M is the saturation magnetization; H_A, H_{A1}, and H_{A2} are the anisotropy fields defined above, and K_1 and K_2 are the first and second order anisotropy constants. Fig. 4 is the plot for the anisotropy vs. the Co-concentration. K_1 and K_2 are all negative, suggesting that the magnetization prefer to lie on the plane of film surface. It can be seen from Fig. 4 that the first order anisotropy has a dramatic increase at 44 at%. Based on Figs. 2–4, the MR ratio is strongly correlated to the magnetic anisotropy of the Co-grains embedded in the Al–O matrix. Namely, at this particular Co-concentration, either the shape anisotropy of Co-grains or the coupling of inter-grain has a dramatic change. According to our tunneling electron microscopy (TEM) data, the size of

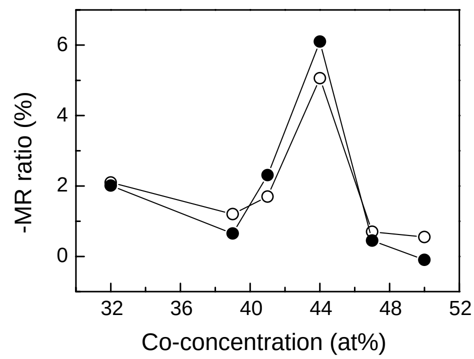

Fig. 2. MR ratio vs. Co-concentration.

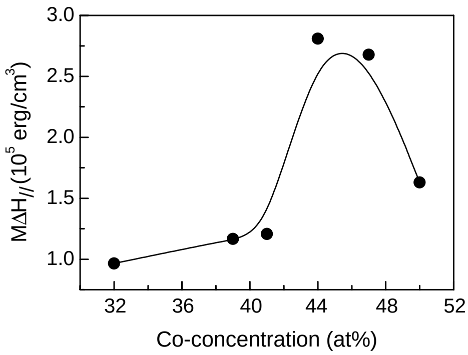

Fig. 3. $M\Delta H_\parallel$ vs. Co-concentration.

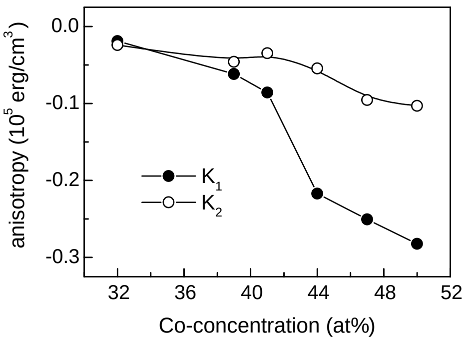

Fig. 4. Anisotropy constants vs. Co-concentration.

Co-grain does not show dramatic change at 44%, therefore, the former cause is not likely associated with the observed results. We therefore proposed that the magnetic inter-grain coupling may assist the tunneling process of the electrons when Co-grains size enlarges with increasing Co-concentration up to some optimal value.

In summary, we have studied the TMR and ESR properties of Co–Al–O granular films. The maximum MR ratio occurs at the sample with 44 at% of Co-concentration. For this composition, $M\Delta H_{\parallel}$ also shows a maximum value, which indicates the mechanism of granular TMR is involved with the microstructures of magnetization within the film. A dramatic increase of the magnetic anisotropy in this composition further suggests that the magnetic interaction between Co-grains is also important to determine the value of MR ratio.

Acknowledgements

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