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Tunnel-type Magnetoresistance in Metal-nonmetal Granular Films Prepared by Tandem Deposition Method

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The relationship between the tunnel-type magnetoresistance(TMR) and the structure of metal-nonmetal granular system (Fe-Co)-Al-O films has been examined. Films were prepared by a tandem deposition method with rotating a substrate holder. The average diameter of metallic granules is found to decrease with an increase in the rotating speed of substrate holder without any change of film composition. And the MR ratio increases with a decrease of the diameter of metallic granules. The influence of the granule diameter on the MR ratio is discussed by so-called coulomb blockade effect. Furthermore, we studied the effect of a substrate temperature($T_{sub.}$) on the MR and the structure of films. The MR ratio and electrical resistivity of the films increase with an increase of $T_{sub.}$. The magnetization measurements suggests that these increases are related with the decrease of the amount of magnetic impurities in the Al-oxide intergranule. The phase separation between the metallic granule and the oxide intergranule may take place in the film prepared on the higher $T_{sub.}$.

Key words : magnetoresistance, nano-granular structure, spin-dependent tunneling, thin film, tandem deposition method

1. Introduction

The metal-nonmetal granular films such as Co-Al-O system show a giant magnetoresistance associated with a spin-dependent tunneling (TMR)[1)-3)]. The films were composed of metallic granules and oxide insulating network-like intergranule and have large electrical resistivities over $10^4\ \mu\Omega$cm. The TMR observed in those films is attributed to the tunneling conductance through a thin oxide layer between granules, and the conductance depends on the magnetization directions of the magnetic granules[3)].

The TMR of nano-granular film is known to be closely related with the film's structure such as the diameter of a metallic granule and the thickness of the oxide intergranule[4)5)]. We have already announced the relationship between the structure and TMR of nano-granular films prepared by the tandem deposition method in detail[6)]. The tandem deposition is the method that a substrate holder is rotated as a substrate passes above of a metal target and oxide target alternately. The schematic illustration for tandem deposition is shown in Fig.1. This method is very useful to make nano-granular film, the structure of the films is controlled by changing the rotation speed of a substrate holder, and the size of the metallic granules in the film is changed independently of the film composition[6)].

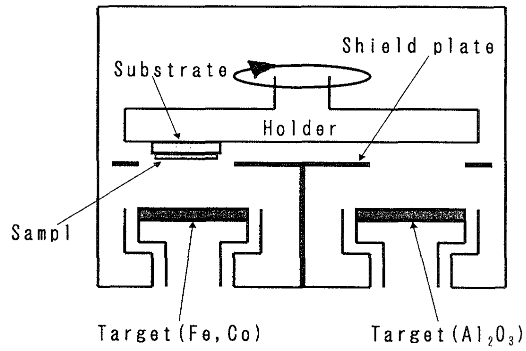

Fig.1 Schematic illustration of the equipment for tandem deposition.

In this paper, we will report the relation between the structure and TMR of (Fe,Co)-Al-O nano-granular film prepared by a tandem deposition method, and also we will describe the effects of the substrate temperature($T_{sub.}$) during deposition on the structure and TMR of the films.

2. Experimental Procedure

The alloy films were prepared by a conventional RF sputtering method in argon atmosphere with a metal(Fe or Co : 75mmϕ) and oxide(Al_2O_3 : 75mmϕ) target on a glass(Corning #7059) substrate. The rotation speed of substrate holder was changed in the range of 1.86 - 11.54 rpm. The composition of the film was controlled by changing the input RF power into the target. The chemical composition was analyzed by energy dispersion spectroscopy (EDS). The electrical resistivity (ρ) and the magnetoresistance (MR) were measured by a DC four-terminal method. The magnetization was measured by using vibrating sample magnetometer (VSM). The micro structure of the films was examined by X-ray diffraction(XRD) using CuKα and transmission electron microscopy (TEM). All the measurements were carried out at room temperature.

3. Results and Discussion

3.1 Structure and TMR

Figure 2 shows a cross-sectional view of TEM

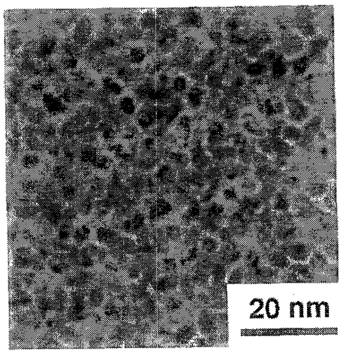

Fig.2 Cross sectional TEM observation for a Co-Al-O film prepared at rotation speed of 4.48 rpm.

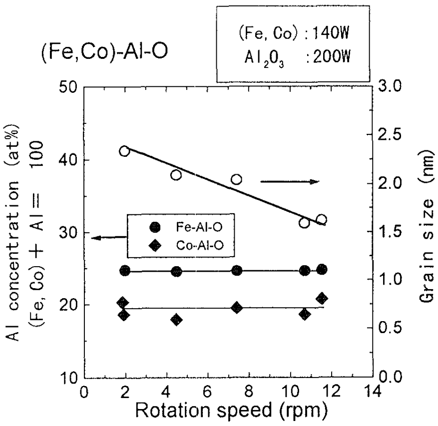

Fig.3 The average grain size of Fe in Fe-Al-O films and the Al concentration in films as a function of the rotation speed.

observation of a Co-Al-O film. As the deposition condition of this film, input RF power into Co and Al_2O_3 target are 140W and 200W respectively, the rotation speed of the substrate holder is 4.48rpm, and the substrate is cooled by water. This film shows MR ratio of about 4% at 10kOe. As seen in the Fig.2, the structure of the film consists of the Co based metallic granules with a diameter of about 2 - 4 nm and Al_2O_3 based intergranules with a width of about 0.5 - 1 nm. This structure is also observed in a TEM photograph[6]. These TEM observations indicate that the film prepared by tandem deposition has granular structure, not layered structure.

Figure 3 shows the relation between the rotation speed of the substrate holder and the average grain size of Fe in Fe-Al-O film and Al concentration in films prepared on water-cooled substrate. The average grain size is calculated from half-value width of bcc-Fe(110) line. The Al concentration is independent upon the rotation speed of substrate holder, while the grain size of Fe is decreased with increasing the rotation speed. Thus, the structure of film can be controlled by changing rotation speed independent of film composition. The grain size of Co in Co-Al-O film can not estimate from X-ray diffraction patterns, because the line at $2\theta=44^{\circ}$ is unsymmetric, the patterns show that the film is composed of hcp and fcc

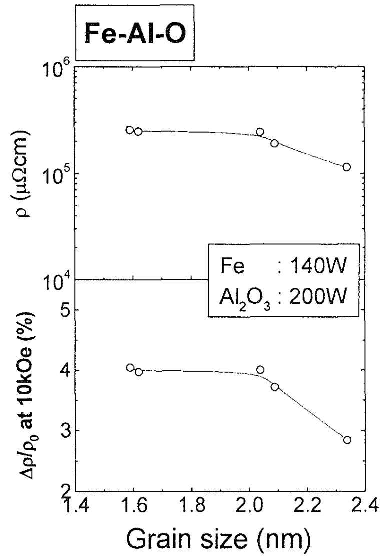

Fig.4 Dependence of the grain size of Fe on ρ and $\Delta\rho/\rho_0$ at 10 kOe for Fe-Al-O films.

phases.

Figure 4 shows the relation between the average grain size of Fe and ρ and MR ratio ($\Delta\rho/\rho_0$) of Fe-Al-O films prepared on water-cooled substrate. ρ and $\Delta\rho/\rho_0$ are constant upto 2.0nm and decrease with increasing the grain size of Fe. The results may be explained by so-called coulomb blockade effect[7]. In the case of Co-Al-O, $\Delta\rho/\rho_0$ increase with increase of rotation speed like case of Fe-Al-O expect for the result at 1.92rpm[6]. As shown in Fig.3, grain size estimated from XRD is propotional to the rotation speed in Fe-Al-O, however, the change of film structure with rotation speed is not observed in Co-Al-O films. We have been examining the relationship between the rotation speed and structure of Co-Al-O film by TEM observation.

3.2 Effects of substrate temperature

Figure 5 shows the effect of $T_{sub.}$ on ρ and $\Delta\rho/\rho_0$ of Fe-Al-O films. Both ρ and $\Delta\rho/\rho_0$ increase with increasing $T_{sub.}$.

To reveal the reason why the MR ratio increases with $T_{sub.}$, we examined the structure of film by XRD. The XRD patterns of Fe-Al-O films with various $T_{sub.}$ are shown in Fig.6. Each films is observed to be similar in pattern, and this means that the granular structure of each film is almost unchanged. We can not obtain information of the reason of the increase from the XRD patterns.

On the other hand, the magnetization of Fe-Al-O films increase with increase of $T_{sub.}$(Fig.7), which suggests that the apparent phase separation of Fe and Al-oxide may take place in the film prepared on the higher $T_{sub.}$, namely, the amount of magnetic impurities (Fe atom) in the Al-oxide intergranule decrease. If a considerable amount of magnetic impurities are contained in Al-oxide tunneling barriers, spin relaxation may occur, resulting in the breakdown of spin conservation. Therefor, it is reasonable results that $\Delta\rho/\rho_0$ increase with $T_{sub.}$.

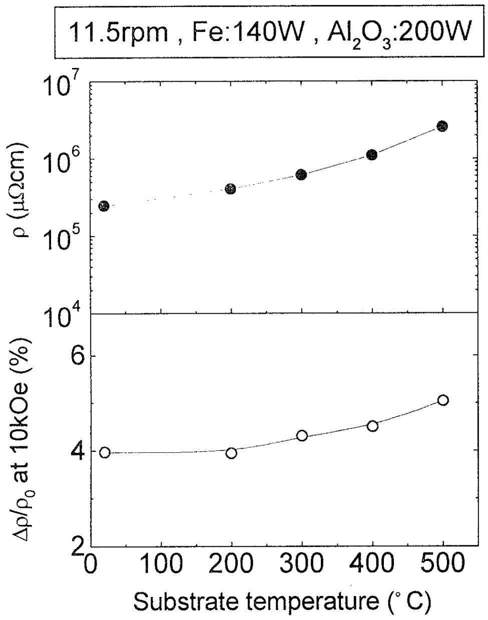

Fig.5 Dependence of the $T_{sub.}$ on ρ and $\Delta\rho/\rho_0$ at 10 kOe Fe-Al-O films.

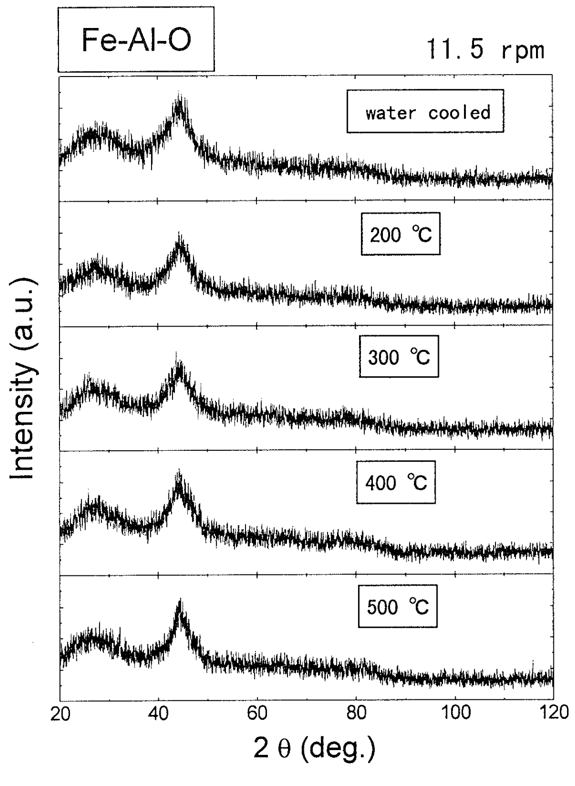

Fig.6 X-ray diffraction patterns of Fe-Al-O films prepared at various Tsub..

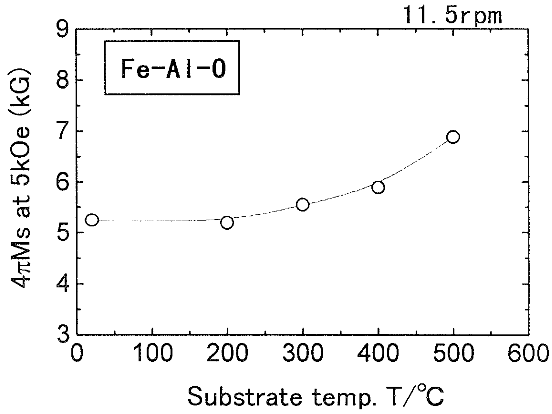

Fig.7 Dependence of the $T_{susb.}$ on the magnetization at 5kOe for Fe-Al-O films.

4. Conclusion

The relation between the structure and TMR of (Fe,Co)-Al-O nano-granular film and also effects of the substrate temperature during deposition on the structure and TMR of the films have been investigated.

1)The average size of metal granule in the films decreases with increasing rotation speed of a substrate holder. The ρ and $\Delta\rho/\rho_0$ increase with decrease of average diameter of metal granule of the film. It may be explained by so-called coulomb blockade effect.

2)The ρ and $\Delta\rho/\rho_0$ increase with increase of $T_{sub.}$. The result of magnetization measurements suggests that these increases are related with decrease of the magnetic impurities in Al-oxide intergranule.

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References

1)H.Fujimori, S.Mitani, and S.Ohnuma : *Mater. Sci. Eng.*, **B31**, 219 (1995)
2) H.Fujimori, S.Mitani, and S.Ohnuma : *J. Magn. Magm. Mat.*, **156**, 311 (1996)
3) S.Mitani, H.Fujimori, and S.Ohnuma : *J. Magn. Magn. Mat.*, **165**, 141 (1997)
4)N.Kobayashi, S.Ohnuma, T.Masumoto, S.Mitani, and H.Fujimori : *J. Magn. Soc. Japan*, **21**, 461 (1997)
5)M.Ohnuma, K.Hono, E.Abe, H.Onodera, S.Mitani and H.Fujimori : *J. Appl. Phys.*, **82**, 5646 (1997)
6) N.Kobayashi, S.Ohnuma, T.Masumoto, and H.Fujimori : *J. Magn. Soc. Japan*, **22**, 581 (1998)
7)S.Maekawa, and J.Inoue : *J. Magn. Magn. Mat.*, **156**, 315 (1996)