Tunneling Magnetoresistance in FeCo-SiO₂ Granular Films

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Abstract---In heterogeneous Fe-, Co- and Fe₃₈Co₆₂-SiO₂ films prepared on glass substrates, the magnetic and transport properties have been examined. The resistivity increases drastically with decreasing volume fraction of the transition metal, and the transport property changes from the metallic to the tunneling conductivity at the percolation points. At these points, the magnetism also varies from the ferromagnetic to the superparamagnetic, which causes the tunneling giant magnetoresistance (MR). The magnetization and MR curves have been systematically analyzed well with the summation of two Langevin functions, corresponding to the two type superparamagnetic grains having the different sizes. The maximum MR ratios are about 3.6% for Fe-, 3.8% for Co- and 4.4% for Fe₃₈Co₆₂-SiO₂. These values are discussed with the degree of the spin polarization.

I. INTRODUCTION

A tunneling magnetoresistance (TMR) was observed early in granular metals embedded in insulators, such as Ni-SiO₂ [1,2] and Co-SiO₂ [3], and in the artificial layered structures of ferromagnetic metal-insulator-metal junctions [4]. Recently, a quite large TMR has been observed in Fe/Al₂O₃/Fe [5] and CoFe/Al₂O₃/Co or NiFe junctions [6] and in Co-Al-O granular magnetic films [7].

In this paper, we will examine the composition dependencies of the magnetic properties and the magnetoresistance (MR) ratio for Fe-, Co- and Fe₃₈Co₆₂-SiO₂ heterogeneous granular films. In particular, the relation between the MR ratio and the saturation magnetization will be discussed.

II. EXPERIMENTAL

Heterogeneous granular films were prepared on glass substrates by rf sputtering at 10 mTorr Ar gas pressure from a composite target consisting of Fe and Co plates placed asymmetrically on a SiO₂ disk. The asymmetry in target composition brought the gradient in the film composition. Thus, we can simultaneously obtain the samples having various metal compositions on a substrate with a single run. However, the atomic ratio of Fe to Co was controlled to be uniform over the whole substrate. We measured the atomic composition of Si in the films by Rutherford Backscattering Spectroscopy (RBS), and the atomic ratio of Fe to Co was measured by Particle Induced X-ray Emission (PIXE). Then, the volume fraction of metal, x , was calculated from the bulk densities of the

metal and SiO₂. Thus, we prepared the heterogeneous granular Fe-, Co- and Fe₃₈Co₆₂-SiO₂ films having various volume fraction x .

The crystalline structure was examined by X-ray diffraction method and the magnetic properties were measured with a vibrating sample magnetometer. The magnetoresistance was obtained using dc method with in-plane field, H , up to 15 kOe at room temperature.

III. RESULTS AND DISCUSSION

The resistivity increases drastically with decreasing volume fraction of the transition metal, x , and the transport property changes from the metallic to the tunneling conductivity at the critical points; around 45vol.% for Fe-, 50vol.% for Fe₃₈Co₆₂- and 55vol.% for Co-SiO₂. At around these percolation points, the magnetism also varies from the ferromagnetic to the superparamagnetic, which causes the TMR. Typical magnetization and MR curves in the tunneling region are shown in Fig.1 for Fe₃₈Co₆₂-SiO₂ films for various compositions x . These magnetization curves can be expressed systematically with the summation of two Langevin functions;

$$\begin{aligned} M(H, T) &= M_s^{(S)} \left[\coth(\mu_s H / kT) - (kT / \mu_s H) \right] \\ &+ M_s^{(L)} \left[\coth(\mu_L H / kT) - (kT / \mu_L H) \right] \quad (1) \\ &= M_s^{(S)} L_s(H) + M_s^{(L)} L_L(H) \end{aligned}$$

where $M_s^{(S)}$ and $M_s^{(L)}$ are the saturation magnetizations,

Fig.1. Magnetization and MR curves of $\text{Fe}_{38}\text{Co}_{62}\text{-SiO}_2$ films for various compositions, x .

Fig.2. MR ratio versus x for Fe-, $\text{Fe}_{38}\text{Co}_{62}$ - and Co-SiO₂ films. These data were obtained at room temperature by applying in-plane fields up to 15 kOe.

μ_S and μ_L are the magnetic moments of unit magnetic cell, T is the temperature, k is the Boltzmann constant, and S and L mean the smaller and larger grains, namely the S-component and the L-component, respectively. Furthermore, the MR curves are expressed by the sum of two squared Langevin functions;

$$\frac{\rho(H) - \rho(0)}{\rho(0)} = -A_S L_S^2(H) - A_L L_L^2(H), \quad (2)$$

where $\rho(H)$ and $\rho(0)$ are the resistivities at applied fields H

and 0, and A_S and A_L are the MR ratios at $H = \infty$ for the S- and L-components, respectively. The relation (2) indicates the tunneling occurs mainly between nearest-neighbor grains which are equal or nearly equal in size [2]. From the fitting parameters μ_S and μ_L , we can evaluate the size of the superparamagnetic granules. Assuming a spherical grain structure, the grain radii are estimated to be ranged from about 10 Å (for 12vol.%) to 15 Å (for 40vol.%) for the S-component, and from about 15 Å (for 12vol.%) to 25 Å (for 40vol.%) for the L-component. These values are consistent with the data observed in the transmission electron microscopy.

The MR ratio measured with in-plane fields up to 15 kOe is represented in Fig.2 as a function of x . The MR ratio increases with x and reaches the maxima at around the percolation points; 3.6% at 37vol.% for Fe-, 4.4% at 42vol.% for $\text{Fe}_{38}\text{Co}_{62}$ -, and 3.8% at 44vol.% for Co-SiO₂. At the same x (≤ 40 vol.%), the MR ratio becomes larger with the order of Co-, Fe- and $\text{Fe}_{38}\text{Co}_{62}$ -SiO₂, namely the order of the saturation magnetization of the transition metals; 1422 emu/cm³ for Co, 1714 emu/cm³ for Fe, and 1782 emu/cm³ for $\text{Fe}_{38}\text{Co}_{62}$.

A theoretical relation [8] indicates that the MR ratio is approximately proportional to the square of the spin polarization, p^2 , of the magnetic metal. Paraskevopoulos *et al.* [9] indicated experimentally that the spin polarization, p , is proportional to the saturation magnetization, M_s , for the 3d-transition metals. Therefore, it is expected that the MR ratio normalized by M_s^2 is independent of the kind of the transition metal. Figure 3 shows the values of MR ratio/ M_s^2 as a function of x . We can see that the values for the Fe- and $\text{Fe}_{38}\text{Co}_{62}$ -systems agree well with each other, while the value of the Co system is larger than the others. The reason for the discrepancy is not made clear.

Now, we plotted in Fig.4 the values of A_L/M_s^2 and A_S/M_s^2 , which were calculated using eq.(2) and which correspond the MR ratio at $H = \infty$ arising from the larger and smaller superparamagnetic granules, respectively. The values of A_L/M_s^2 increase with x , and those for three systems agree well with each other. On the other hand, the values of A_S/M_s^2 show the weak dependence on x and they become larger with the order of Fe-, $\text{Fe}_{38}\text{Co}_{62}$ -, and Co-systems. In other words, the S-component of Fe granules exhibits the small MR ratio compared with the Co granules. This implies that the spin polarization of Fe is strongly affected by the surroundings. It is suggested that the discrepancy in Fig.3 is caused by the decrease in the value of p of Fe atoms due to the surrounding effects.

Fig.3. MR ratio normalized by M_s^2 as a function of x for Fe-, $\text{Fe}_{38}\text{Co}_{62}$ - and Co-SiO₂ films, where the data of MR ratio were obtained from Fig.2. The values of M_s used were 1422 emu/cm³ for Co-, 1714 emu/cm³ for Fe-, and 1782 emu/cm³ for $\text{Fe}_{38}\text{Co}_{62}$ -SiO₂ films.

Fig.4. Composition dependence of the calculated values of A_L/M_s^2 and A_S/M_s^2 for Fe-, $\text{Fe}_{38}\text{Co}_{62}$ - and Co-SiO₂ films.

IV. CONCLUSION

In heterogeneous Fe-, Co- and $\text{Fe}_{38}\text{Co}_{62}$ -SiO₂ films prepared on glass substrates, the resistivity increased drastically with decreasing x , and the transport property changed from the metallic to the tunneling conductivity at the percolation compositions. The magnetism also varied at these points from the ferromagnetic to the superparamagnetic, which caused the TMR. The MR ratio increased with x , and reached the maxima; about 3.6% for Fe-, 3.8% for Co- and 4.4% for $\text{Fe}_{38}\text{Co}_{62}$ -SiO₂ at around the percolation points. These values were discussed using the degree of the spin polarization. The magnetization and MR curves were systematically analyzed well with the summation of two Langevin functions. The MR ratio for the L-component for three systems increased with x and the values of normalized by M_s^2 agreed well with each other.

On the other hand, the MR ratio of the S-component depended little on x , and increased with the order of Fe-, $\text{Fe}_{38}\text{Co}_{62}$ -, and Co-SiO₂.

ACKNOWLEDGEMENTS

The authors are grateful to F. Nishiyama for RBS and PIXE measurements. This research was partly supported by a Grant-in-Aid for Science Research for the Ministry of Education, Culture and Science of Japan.

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