Magnetic Properties and Magnetoresistance of Evaporated Fe-Si Films

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Fe/Si films were prepared by evaporating Fe and Si alternately at various substrate temperatures T_s between 100 and 623K. The nominal thicknesses of Fe and Si were fixed at 3.4 Å and 6 Å, respectively. The films prepared at T_s higher than 200 ~ 300K showed a superparamagnetic behavior, and also an isotropic magnetoresistance (MR) of 0.02%. On the other hand, the films at lower T_s showed an anisotropic MR similar to that observed in most ferromagnets. These results suggest that the film structure is changed from a granular like one at low T_s to a partially a continuous one at high T_s . These structure changes may come from an interdiffusion and/or a reaction between Fe and Si in the films.

Key words: Fe/Si, evaporated film, magnetoresistance, granular structure, substrate temperature

1. Introduction

In Fe/Si and Fe/FeSi multilayer films, the antiferromagnetic (AF) interlayer coupling across Si or an iron silicide spacer¹⁾⁻⁸⁾ has been observed. A problem of much consideration is to whether the spacer layer mediating the AF coupling is a crystalline (c-) FeSi^{1), 5)-7)} or an amorphous (a-) Si^{3), 4)}. In addition, even if the c-FeSi spacer may be responsible for the AF coupling, there remains a problem whether c-FeSi is a metal^{6), 7)} or a semiconductor^{1), 5)}. Those who assume the semiconductor spacer insisted that the AF coupling was induced by the thermal excitation of charge carriers in the spacer. This conclusion is based on the facts that the film becomes ferromagnetic with decreasing temperature, and the AF coupling changes into the ferromagnetic (F) one. On the other hand, those who assume the metallic spacer insisted that the AF coupling in Fe/FeSi multilayer films is of the same origin as that in Fe/Cr one. They claim that the change from the AF coupling to the F coupling observed in references 1)-4) may be caused by the presence of ferromagnetic bridges (pinholes) in the spacer layers.

In the case of Fe/Si multilayers with AF coupling, the magnetoresistance (MR) ratio is about 0.15% at 300Oe and at 298K⁴⁾, and it is not clear whether such a small MR ratio is attributed to the Si spacer or the intermixed FeSi interface structure. It is also considered that such an interface structure strongly depends on the substrate temperature during the preparation of specimens.

For the investigation of ferromagnet/semiconductor multilayers, it is required to reveal the origin of the AF coupling. But it is also expected to find the phenomena which reflect the electronic structure of a semiconductor spacer, and which differ from those in ferromagnet/metal multilayers. If we can prepare a granular like structure in Fe-Si system, we may obtain a clue to clear those questions. However, there is scarcely

such a report on the granular Fe-Si except one prepared by sputtering technique⁸⁾ because Fe and Si are apt to intermix or react.

Recently we attempted to realize the granular like structure in Fe-Si system by means of an alternate evaporation of Fe and Si⁹⁾, utilizing the fact that the evaporated film has a discontinuous island structure when the nominal thickness is about 10 Å. In the specimen prepared by the alternate evaporation, the volume ratio of Fe and Si was always fixed to 1:3 changing both nominal Fe and Si thicknesses from 20 Å to 1 Å. Then we have investigated how the total magnetic properties and the magnetoresistance change. In consequence, we found that the magnetic properties of our films change from ferromagnetic to superparamagnetic to paramagnetic, and the transport properties change from metallic to semiconductor with decreasing the thickness of Fe and Si. In addition, the MR changes from anisotropic MR to isotropic MR to small MR. Because the specimens with the middle nominal thickness show superparamagnetic and isotropic MR, we thought the granular like structure was partially realized. In this paper, in order to change the structure of the specimen, the substrate temperature during the preparation was varied, so we investigated how the magnetic properties and the magnetoresistance change.

2. Experiment

The FeSi films were prepared by evaporating Fe and Si alternately. Some pieces of Fe wire and a bit of Si powder were heated by each tungsten conical basket coated with Al₂O₃, and the two baskets were covered with a shutter alternately. The nominal thickness of Fe and Si was fixed at 3.4 Å and 6 Å, respectively. Each layer thickness was monitored by a quartz-crystal oscillator. Total thickness of film was about 2100 Å. We used silica substrate, and the substrate temperature (T_s) during the preparation of films was set at 100K, 318K, 473K, and 623K. The evaporation pressure was 10^{-6} – 10^{-7} Torr. The crystalline structure of the film was examined by X-ray diffraction and the magnetic properties were measured by a vibrating sample magnetometer. The resistivity measurement was carried out by the four terminal method. In this paper, for MR measurement, the electric current was perpendicular to the magnetic field in the plane of the film.

3. Results and discussion

As shown in Fig. 1, no peaks of Fe, Si, and FeSi were observed except a halo of the silica substrate around $2\theta = 26^\circ$ in X-ray diffraction pattern, which is due to the very thin nominal thickness or an amorphous structure of Si and FeSi.

Fig. 2 shows the magnetization curve for $T_s=623\text{K}$, $T_s=318\text{K}$, and $T_s=100\text{K}$. The specimens of $T_s=623\text{K}$ and $T_s=318\text{K}$ show a superparamagnetic behavior at R.T. The magnetization of these films increases at 77K. This result suggests that the specimens of $T_s=623\text{K}$, and

Fig. 1 X-ray diffraction pattern for Fe-Si films. (a) $T_s=623\text{K}$, (b) $T_s=318\text{K}$, (c) $T_s=100\text{K}$

$T_s=318\text{K}$ have not a continuous layered structure but a granular like structure. In Fig. 3, T_s dependence of the magnetization at 16kOe is shown. If all magnetization of Fe in these films is alive, the total magnetization of a film is 617 emu/cc. The dashed line in Fig. 3 denotes this value. Since the

Fig. 2 Magnetization curves at R.T. and 77K for Fe-Si films. (a) $T_s=623\text{K}$, (b) $T_s=318\text{K}$, (c) $T_s=100\text{K}$

magnetization increases with decreasing T_s , the interfacial diffusion and/or reaction decreases with

decreasing T_s , and the film of lower T_s may have a continuous layered like structure. When T_s is higher than about 300K, the magnetization is smaller

Fig. 3 Magnetization (16kOe) at R.T. and 77K as a function of the substrate temperature T_s .

Fig. 4 Resistivity at R.T. and 77K as a function of the substrate temperature T_s .

than a half of 617 emu/cc, thus a lot of Fe may react on Si in the interface.

For all samples, the temperature dependence of resistivity is semiconductor-like (the resistivity increases with decreasing measurement temperature). In Fig. 4 is shown T_s dependence of the resistivity at R.T. and 77K. The magnitude of resistivity decreases as T_s decreases. This also supports that Fe in the film has a fairly continuous layered structure for the lower T_s . On the contrary, for the higher T_s the resistivity is large because Fe may have a granular structure or a partially continuous structure.

Fig. 5 shows MR for $T_s=623\text{K}$, $T_s=318\text{K}$, and $T_s=100\text{K}$. At R.T., the resistivity decreases as the magnetic field increases for all T_s . This is an isotropic negative MR which is generally observed in granular or multilayers systems as Co-Cu. On the other hand, at 77K, all specimens show a positive MR (the resistivity increases with increasing the magnetic field). The magnitude of positive MR increases with higher T_s . We think that the positive MR is caused by the Lorentz force in the nonmagnetic Si or FeSi, but further investigation is required.

In addition, at around the zero magnetic field, there is a rapid MR change, which is an anisotropic magnetoresistance (AMR) generally observed in a ferromagnet. The component of AMR is the largest for $T_s=100\text{K}$, while the specimen of $T_s=623\text{K}$ does not show almost the component of AMR. This suggests that

the continuous layered ferromagnetic structure of Fe holds only for $T_s=100\text{K}$, and the diffusion or reaction causes the granular like structure with small AMR component for $T_s=318\text{K}$ and $T_s=623\text{K}$.

The relationship between T_s and the isotropic MR ratio in which the component of AMR is excluded is

Fig. 5 Magnetoresistance at R.T. and 77K for Fe-Si films. (a) $T_s=623\text{K}$, (b) $T_s=318\text{K}$, (c) $T_s=100\text{K}$

shown in Fig. 6 (a). We define the isotropic MR ratio as $(\rho_{10\text{kOe}} - \rho_{0\text{kOe}}) / \rho_{0\text{kOe}} \times 100\%$. The negative isotropic MR ratio is about 0.02% for all T_s . At R.T., the negative MR ratio increases with decreasing T_s , because the resistivity decreases with decreasing T_s . Fig. 6 (b) shows T_s dependence of the AMR ratio. In this paper, AMR ratio is defined as $\Delta \rho_{\text{AMR}} / \rho_{0\text{kOe}} \times 100\%$, $\Delta \rho_{\text{AMR}}$ is magnitude of the sharp peak around zero field in Fig. 5. Fig. 6 (b) suggests again that Fe becomes a continuous layered like structure for lower T_s . From Figs. 3 and 6, it is considered that a structure change from the continuous layered like one to granular like one may occur at about 200–300K. As we discussed above, it is plausible that the Fe/Si (3.4 Å/6 Å) films which were prepared by the alternate evaporation (at T_s higher than R.T.) have a partially granular like structure, since much interfacial diffusion and/or reaction between Fe and Si may occur for higher T_s .

4. Conclusion

The Fe/Si films were prepared by evaporating Fe and

Fig. 6 Magnetoresistance ratio as a function of the substrate temperature T_s . (a) Isotropic MR (b) AMR

Si alternately. The nominal thicknesses of Fe and Si were fixed at 3.4 Å and 6 Å, respectively. When the substrate temperature T_s during preparation is higher than R.T., the specimen shows superparamagnetism. On the contrary, the specimen prepared at $T_s=100\text{K}$ shows ferromagnetism. The transport property is semiconductive for all T_s . For lower T_s , the component of AMR becomes larger indicating a continuous layered ferromagnet, while for higher T_s the isotropic MR becomes main part suggesting a granular structure. The isotropic MR ratio is about 0.02% for all T_s .

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