CRYSTALLINE FERRIMAGNET-SPINGLASS-AMORPHOUS FERRIMAGNET PHASE TRANSITION IN Tb/Fe MULTILAYERS

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ABSTRACT—The crystallographic structure and the magnetic properties of Tb/Fe sputtered multilayers have been studied as a function of the thickness of Fe-layer (d_{Fe}). The crystallographic structure of Fe layer changes from bcc to amorphous at the critical thickness of Fe layer ($d_{Fe}^c \approx 17\text{\AA}$) with decreasing d_{Fe} . In corresponding to the change in the crystallographic phase, the magnetic properties of the saturation magnetization M_s , the Kerr rotation angle θ_K , the Curie temperature T_c and the perpendicular coercivity $H_c(1)$ change abruptly at the critical thickness d_{Fe}^c , where the Curie temperature drops to below room temperature and the magnetization curve shows the spinglass like or superparamagnetism like nature at room temperature. Below d_{Fe}^c , with decreasing d_{Fe} the Curie temperature T_c increases owing to the exchange interaction between Fe and Tb around the layer boundaries and then the saturation magnetization increases again approaching the value of the alloys with the same composition. The perpendicular coercivity also increases steeply with decreasing d_{Fe} .

KEYWORDS: Amorphous Fe, Tb/Fe Multilayer, Spinglass, Superparamagnetism, Sputtered Multilayer, Perpendicular Coercivity, Curie Temperature, Magneto-optical Kerr Rotation

INTRODUCTION

Compositionally modulated rare-earth/transition-metal films have been extensively studied in recent years for reasons of scientific interest and of application to magneto-optical recording [1-3]. Sato [2] reported that Tb/Fe multilayers have large perpendicular magnetic anisotropy for small layer thickness and suggested that the perpendicular anisotropy comes from the anisotropic distribution of Tb-Fe pairs aligned perpendicular to the film plane.

Yoden et. al. [4] studied the crystallographic structure of Dy/Fe multilayers and indicated that Fe layer changes from crystalline to amorphous at about $20\text{\AA}$ with decreasing Fe layer thickness (d_{Fe}) and there the magnetization at room temperature falls off to nearly zero. We have also shown that the crystallographic structure of Fe layer in evaporated Dy/Fe [5,6] and in sputtered DyFe/Fe multilayers [5] changes from bcc to amorphous via fcc at about $20\text{\AA}$, and the saturation magnetization falls off drastically at this point followed by the increase in the magnetization with decreasing d_{Fe} . Sellmyer and Shan [7,8] examined intensively the periodicity dependence of magnetic properties of Dy/Co, Dy/Fe and Tb/Fe multilayers, and indicated that a "kink" in the curve of magnetization versus d_{Fe} appears in Dy/Fe and Tb/Fe multilayers.

In this paper, we will discuss the magnetic phase transition arising from the crystallographic change in Tb/Fe sputtered multilayers.

EXPERIMENTS

Tb/Fe multilayers were prepared by RF magnetron sputtering at 6 mTorr Ar gas pressure from Tb and Fe targets onto glass substrates on a rotatable substrate table. Two type multilayers were prepared; Type I is the sequence of substrate/Tb/Fe/.../Fe/Tb/SiO₂ and Type II is the sequence of substrate/Fe/Tb/.../Tb/Fe/SiO₂. Here, SiO₂ layer of $500\text{\AA}$ thickness was overcoated for the protection. The thickness ratio of Fe to Tb layer (d_{Fe}/d_{Tb}) was held constant 1/1, 1/2, 1/3 and 1/4, and the bilayer thickness, $\Lambda = d_{Fe} + d_{Tb}$, was changed from 6 to $160\text{\AA}$ with the total thickness of about $1800\text{\AA}$. In this paper, we will discuss the results only for the films of $d_{Fe}/d_{Tb} = 1/2$.

The layered structure was confirmed in the range between 20 and $100\text{\AA}$ of periodic length (Λ) by the small angle X-ray diffraction (SA-XRD). The periodic length (Λ) was estimated from the peak position of SA-XRD for $\Lambda \geq 20\text{\AA}$, and calculated for $\Lambda < 20\text{\AA}$ from the sputtering rate which was corrected from SA-XRD and the total film thickness. The film composition was determined by Rutherford backscattering spectroscopy (RBS). The magnetic properties

Fig.1. X-ray diffraction of Tb/Fe multilayers of Type II with various thickness of Fe layer (d_{Fe}).

were measured with a vibrating sample magnetometer and a Kerr loop tracer at a wavelength of 780 nm. The crystallographic structure was studied by X-ray scattering method.

RESULTS AND DISCUSSION

X-ray diffraction spectra are shown in Fig.1 for several modulation periodicities (Λ). For $\Lambda = 170 \text{ Å}$ ($d_{Fe} = 56.6 \text{ Å}$, $d_{Tb} = 113.2 \text{ Å}$), we can recognize the clear peaks of (100), (002) and (112) planes of hcp Tb and (110) plane of bcc Fe. These peaks become smaller in intensity and broaden with decreasing Λ and only the broad peak due to glass substrate remains around 30° for $\Lambda \leq 50 \text{ Å}$ ($d_{Fe} \leq 17 \text{ Å}$, $d_{Tb} \leq 34 \text{ Å}$). These data imply that the crystallographic structure changes from crystalline to amorphous at around 17 Å of d_{Fe} with decreasing layer thickness similar to the evaporated Dy/Fe and the sputtered DyFe/Fe multilayers [5,6].

The magnetic hysteresis characteristics vary with the Fe layer thickness (d_{Fe}) as shown in Fig.2. Although the easy axis of magnetization lies in film plane for $d_{Fe} \geq 40 \text{ Å}$, the small loops appear for the perpendicular magnetization curves. The coercivity $H_c(1)$ of the perpendicular loop increases with decreasing d_{Fe} and the perpendicular loops become parallelogrammic for $d_{Fe} \leq 24 \text{ Å}$ as shown in Figs.2(a) and (b), because of the increase in the perpendicular anisotropy originated from the interfacial anisotropy. At the critical layer thickness, $d_{Fe}^c \approx 17 \text{ Å}$, of transition

Fig.2. Magnetic hysteresis loops for Type I films with various thickness of Fe layer (d_{Fe}).

in crystalline phase, the magnetization curves change abruptly and exhibit the spinglass like or the superparamagnetism like nature as shown in Fig.2(c); namely, both the perpendicular and in-plane magnetization curves, which coincide with each other in the experimental error, have not hysteresis characteristic and do not saturate even at 15 kOe. Below d_{Fe}^c , the rectangular hysteresis loops appear again for perpendicular applied field, and the perpendicular anisotropy energy and coercivity increase with decreasing d_{Fe}^c as shown in Figs.2(d) and (e).

The similar behaviour with change in d_{Fe} is also observed in polar Kerr loops as shown in Fig.3. In particular, the saturation Kerr rotation angle becomes nearly zero at the critical thickness d_{Fe}^c (Fig.3c). Below d_{Fe}^c , the polarity of Kerr rotation changes to Tb-rich characteristics from Fe-rich ones and rectangular loops with high coercivity appear as shown in Figs.3(d) and (e).

Fig.4 shows the saturation or spontaneous magnetization M_s as a function of d_{Fe} . Here, the values of M_s for non-saturated films around 17 Å of d_{Fe} are obtained from Arrott plot method and the polarity of M_s corresponds to that of negative polar Kerr rotation; namely + or - sign indicates the Fe- or Tb-moment dominant nature. The value of M_s decreases gradually with reduction of d_{Fe} and becomes zero around 17 Å . Below 17 Å , M_s is negative, namely Tb-moment is dominant. The absolute value of M_s increases again with decreasing d_{Fe} and approaches the value of alloyed amorphous film with the

Fig.3. Polar Kerr hysteresis loops measured from the film surface for Type I films of different d_{Fe} .

Fig.4. Saturation magnetization (M_s) as a function of d_{Fe} .

same Tb-composition, 33at.% [9].

The saturation Kerr rotation angle θ_K , which was measured through glass substrate at a wavelength of 780 nm, is plotted in Fig.5. The absolute value of θ_K is nearly constant 0.35° in the crystalline region of $d_{Fe} \geq 20\text{Å}$ for the films of Type II, while the value decreases with increasing d_{Fe} above 20Å for the films of Type I. Because the Kerr rotation at 780 nm wavelength comes mainly from Fe layer, with increasing d_{Fe} or increasing d_{Tb} , the intensity of light reflected from Fe layer decays for Type I films and thereby the apparent Kerr rotation angle decreases. The absolute value of θ_K falls off abruptly to zero at the critical thickness d_{Fe}^c , and it increases again gradually with decreasing d_{Fe} .

As shown in Fig.6, the perpendicular coercivity $H_c(l)$ increases slightly up to about 1 kOe with decreasing d_{Fe} to 20Å from 60Å in the crystalline region. At the critical thickness d_{Fe}^c , $H_c(l)$ drops to nearly zero and then increases drastically up to about 8 kOe with decreasing d_{Fe} in the amorphous region below d_{Fe}^c .

Fig.5. Saturation Kerr rotation angle θ_K measured through glass substrate at a wavelength of 780 nm.

Fig.6. Perpendicular coercivity $H_c(l)$ versus d_{Fe} .

We have seen in the above that the magnetic properties change abruptly at the critical Fe layer thickness, d_{Fe}^c , around 17Å in coincidence with the variation of crystalline structure in Fe layer. Plausibly, the spinglass like or the superparamagnetism like nature at $d_{Fe}^c \approx 17\text{Å}$ is closely connected with the amorphous structure of Fe layer, because the Curie temperature of amorphous Fe is below room temperature as reported in Refs. [4,10].

We, therefore, estimated the Curie temperature T_c by measuring the temperature dependence of the Kerr rotation angle θ_K . Temperature dependences of $|\theta_K|$ are plotted in Fig.7 for various d_{Fe} . For the film of $d_{Fe} = 20.4\text{Å}$, the value of $|\theta_K|$

Fig.7. Temperature dependence of θ_k at 780 nm for the films of Type I.

increases irreversibly with rising temperature above 200°C. This irreversible increase is caused by the crystallization of Fe around the layer boundaries, because the XRD peak of (110) bcc Fe increased after heated up to 300°C. In this case, the Curie temperature T_c was estimated by the extrapolation as shown by the broken line in Fig.7.

The Curie temperature T_c , thereby, is obtained as a function of d_{Fe} as shown in Fig.8. We can see that the Curie temperature T_c falls off abruptly at around 17 Å with decreasing d_{Fe} . The abrupt drop in T_c is probably caused by the phase transition from crystalline to amorphous at this critical layer thickness. This might be supported by the irreversible increase in $|\theta_k|$ above 200°C for the films of $d_{Fe} = 20.4$ Å as shown in Fig.7. Below the critical Fe layer thickness d_{Fe}^c , the Curie temperature rises again owing to the exchange interaction between Fe and Tb around the layer boundaries. Then, Fe moment and especially Tb moment increase, and the net saturation magnetization M_s increases with the polarity of Tb-dominant nature. The increase in T_c implies that the Tb atoms become active magnetically with decreasing d_{Fe} similarly to the T_c change against Tb composition in amorphous Tb-Fe alloys, in which T_c rises with increasing Tb composition [9].

SUMMARY

The crystallographic structure of Fe layer changed from bcc to amorphous at the critical thickness of Fe layer ($d_{Fe}^c \approx 17$ Å) with decreasing d_{Fe} . At the critical thickness d_{Fe}^c , the Curie temperature dropped to below room temperature and then the magnetization curve showed the spin-

Fig.8. Curie temperature T_c estimated from the temperature dependence of θ_k .

glass like or superparamagnetism like nature at room temperature. Below d_{Fe}^c , with decreasing d_{Fe} , the Curie temperature T_c increased owing to the exchange interaction between Fe and Tb around the layer boundaries, then the saturation magnetization increased again and the perpendicular coercivity increased drastically.

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REFERENCES

- [1] T.Morishita, Y.Togami and K.Tsushima, J.Phys.Soc.Japan 54,37(1985).
- [2] N.Sato, J.Appl.Phys.59,2514(1986).
- [3] S.Honda, M.Ohkoshi and T.Kusuda, IEEE Trans.Magn. MAG-22,1221(1986).
- [4] K.Yoden, N.Hosoito, K.Kawaguchi, K.Mibu and T.Shinjo, Jpn.J.Appl.Phys. 27,1680(1988).
- [5] S.Honda, S.Nishimura, M.Ohtsu and T.Kusuda, Res.Meeting of Magnetics, MAG-88-198,9(1988).
- [6] S.Honda, S.Nishimura and T.Kusuda, IEEE Trans.Magn. 25,4027(1989).
- [7] D.J.Sellmyer and Z.S.Shan, MRS Int'l.Mtg.Adv.Mats. 10,269(1989).
- [8] Z.S.Shan and D.J.Sellmyer, J.Appl.Phys. 67,5713(1990).
- [9] Y.Mimura and N.Imamura, Appl.Phys.Lett.28,746(1976).
- [10] N.Heiman, K.Lee, R.I.Potter and S.Kirkpatrick, J.Appl.Phys.47,2634(1976).