Spin Freezing and Random Anisotropy in Tb-Fe-Si Amorphous Alloys

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(Received; May 6, 1998, Accepted; August 26, 1998)

The spin freezing and random magnetic anisotropy in Tb-Fe-Si amorphous alloys have been investigated. Zero-field and field-cooled magnetizations of $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($0 \leq x \leq 10$) amorphous alloys exhibit a magnetic cooling effect. The spin freezing temperature decreases with increasing x . The compensation temperature is lower than the spin freezing temperature. The present amorphous alloys exhibit a negative magnetization because the frozen spins can not be turned over below the compensation temperature.

Key words: Tb-Fe-Si amorphous alloy, random magnetic anisotropy, magnetic cooling effect, spin freezing temperature

1. Introduction

R-Fe ($R \equiv$ Rare-earth metal) amorphous alloys show a relatively large random magnetic anisotropy originated from the single-ion magnetic anisotropy of R atoms in low temperatures¹⁾. On the other hand, amorphous Tb-Fe thin film is a typical alloy as magneto-optical (MO) disks²⁾. During the recording on the MO disk, the film surface is pulse-heated by scanning of a laser beam. Therefore, high thermal stability of amorphous films is highly desired. An addition of Si to Tb-Fe binary amorphous alloys improves the thermal stability due to the strong covalent-type bonding³⁻⁶⁾.

In the present work, magnetic cooling effects of these alloys have been measured. The spin freezing and random anisotropy in an amorphous metal-metal type binary $\text{Tb}_{0.22}\text{Fe}_{0.78}$ alloy have been compared with these in amorphous metal-metalloid type ternary $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 2, 4, 6$ and 10) alloys.

2. Experimental

Amorphous $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 0, 2, 4, 6$ and 10) alloys were made by DC sputtering on a water-cooled Cu substrate of 1mm thickness. To make the amorphous alloys of about 0.3mm thickness, the sputtering was carried out for about 3 days. The amorphous state of the specimens was confirmed by X-ray diffraction with Mo $K\alpha$ radiation. The Cu substrate was removed by mechanical grinding.

The crystallization processes of the amorphous alloys were examined by a differential scanning calorimeter (DSC) at heating rate of about 0.4K/s. The crystallization temperature T_x was defined as the exothermic

peak of the DSC curve. The Curie temperature T_c was also defined as the λ -type endothermic peak¹⁾. Temperature and field dependences of DC magnetizations were measured with a SQUID magnetometer below 400K. Temperature dependence of DC magnetizations from room temperature to 420K for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 0, 2$ and 4) amorphous alloys was measured with a VSM.

3. Results and Discussion

The crystallization temperature T_x and the Curie temperature T_c of amorphous $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 0, 2, 4, 6$ and 10) alloys are given in Fig. 1. The temperature T_x increases rapidly from $x=0$ to 2 and increases gradually up to $x=10$. The addition of Si about 2% decreases the rate of the crystallization process, because of the strong covalent-type bonding in metal-metalloid type amorphous $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{98}\text{Si}_2$ alloy in comparison with the metallic bonding in the metal-metal type amorphous $\text{Tb}_{22}\text{Fe}_{78}$ alloy. This suggests that addition of Si improves the thermal stability for pulse-heated recording magneto-optical amorphous media³⁻⁶⁾. On the other hand, T_c decreases

Fig. 1 Concentration dependence of the crystallization temperature T_x and the Curie temperature T_c for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 0, 2, 4, 6$ and 10) amorphous alloys.

Fig. 2 Temperature dependence and concentration dependence of Zero-field cooled ZFC and field-cooled FC magnetizations for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 0$ and 2) amorphous alloys in a field of 50 Oe.

linearly with the addition of Si.

Although magnetization curves are sensitive to the strength of applied magnetic field and the demagnetization factor, the compensation temperature T_{comp} was confirmed in all of the present specimens⁷⁾ and it decreases with increasing Si concentration.

It should be noted that the spin freezing occurs in every present amorphous alloy. Zero-field cooled ZFC and field-cooled FC magnetizations of $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x=0$ and 2) amorphous alloys are shown in Fig. 2. The cooling effect of magnetization as a function of magnetic field for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{90}\text{Si}_{10}$ amorphous alloy is given in Fig. 3. From Figs. 2 and 3, these alloys exhibit a marked hysteresis between ZFC and FC magnetizations below T_c .

It is known that randomly oriented magnetic anisotropy causes frustration and the random anisotropy magnets exhibit a spin-glass like irreversibility⁸⁾. Although the spin-glass behavior due to the exchange frustration of Fe magnetic moments appears in many Fe-rich Fe-M ($M \equiv$ non-magnetic rare-earth elements such as Y and Zr), the spin freezing temperature of these Fe-M amorphous alloys lies below about 100K ⁹⁾. Therefore, the magnetic irreversibility observed above 400K for $x=0$ and $x=2$ should originate from the random magnetic anisotropy with a large single ion anisotropy energy on the Tb site. As seen in Fig. 2, the magnetic irreversibility appears just

Fig. 3 Temperature dependence and field dependence of ZFC and FC magnetizations for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{90}\text{Si}_{10}$ amorphous alloy.

below T_c , therefore, the spin freezing temperature is relatively close to the Curie temperature. The spin freezing temperature T_f is determined as the starting point of the hysteresis between ZFC and FC magnetizations.

In the present amorphous alloys, the relation among T_{comp} , T_f , T_c and T_x is confirmed as

$$T_{\text{comp}} < T_f < T_c < T_x. \quad (1)$$

It should be noted that T_{comp} is always lower than T_f . The present amorphous alloys exhibit a negative value of the field cooled DC magnetization because a coherent reversal of subnetwork magnetizations is prevented below T_{comp} as shown in Figs. 2 and 3.

The concentration dependence of the spin freezing temperature T_f for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ amorphous alloys in a field of 50 Oe is shown in Fig. 4. A broken line indicates the Curie temperatures T_c in Fig. 1. The temperature of T_f decreases linearly with increasing x . The slope of the decreasing of T_f is slightly steeper than that of T_c . The difference between T_c and T_f is relatively small, about $5 \sim 10\text{K}$. The each spin is frozen at a bottom in the potential well for random magnetic anisotropy energy. These spin are stable until the thermal energy of about $k_B T_f$ ($\approx k_B T_c$). This suggests that a potential energy of the saddle point for coherent

Fig. 4 Concentration dependence of the spin freezing temperature T_f for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 0, 2, 4, 6$ and 10) amorphous alloys in a field of 50 Oe. A broken line is the Curie temperature T_c in Fig 1.

reversal of subnetwork magnetizations is considerably high.

Figure 5 shows the magnetic field H dependence of the spin freezing temperature T_f for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{90}\text{Si}_{10}$ amorphous alloy. The Curie temperature T_c is also shown in the same figure. The temperature of T_f decreases with increasing of H ⁸⁾. Two curves of T_f and T_c divide the T - H area into three phases. Consequently, $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{90}\text{Si}_{10}$ amorphous alloy exhibits a paramagnetic, ferromagnetic and spin frozen phase as the temperature is lowered. A curvature of T_f - H line is similar to that of Dy-Ni amorphous alloys with a large random anisotropy¹⁰⁾. The difference in the T_f - H curve is discussed in terms of the ratio of the average local anisotropy (D) to the exchange strength (J). The Dy-Ni amorphous alloys have a relatively large value of D/J . Therefore, the ratio of D/J of the present amorphous alloys should be relatively large even Si is added, although it is difficult to evaluate the value of D/J due to a large irreversibility and a high temperature of T_f .

4. Summary

$(\text{Tb}_{0.22}\text{Fe}_{0.78})_{100-x}\text{Si}_x$ ($x = 0, 2, 4, 6$ and 10) amorphous alloys have been prepared by DC sputtering in order to investigate the spin freezing and random anisotropy. The main results are summarized as follows:

- (a) Zero-field-cooled and field-cooled magnetizations of these amorphous alloys exhibit a clear magnetic cooling effect.
- (b) The compensation temperature T_{comp} is always lower

Fig. 5 Magnetic field dependence of the spin freezing temperature T_f and the Curie temperature T_c for $(\text{Tb}_{0.22}\text{Fe}_{0.78})_{90}\text{Si}_{10}$ amorphous alloy.

than the spin freezing temperature T_f .

- (c) These amorphous alloys exhibit a negative magnetization below the compensation temperature.
- (d) The ratio of the average local anisotropy to the exchange strength should be relatively large.

Acknowledgments The authors are grateful to Professor M. Dezuca of Hakodate National College of Technology, for introduction of a DSC apparatus. Thanks are given to Professor Y. Iijima and Professor Y. Otani for valuable discussions.

References

- 1) Y. Mimura and N. Imamura: *Appl. Phys. Lett.*, **28**, 746 (1976).
- 2) Y. Suzuki, S. Takayama, F. Kirino and N. Ohta: *IEEE Trans. Magn.*, **MAG-23**, 2275 (1987).
- 3) K. Yamada, K. Fukamichi and Y. Iijima: Proc. Magneto-Optical Recording Int. Symp., Yamagata Japan, 1997, to be published in *J. Magn. Soc. Jpn.*, **22** (Suppl. No.S1), (1998).
- 4) K. Yamada, M. Oumi, Y. Iijima and K. Fukamichi: Abstracts of The 120th Spring Meeting of the Japan Institute of Metals, Tokyo, Japan, 88 (1997).
- 5) K. Yamada, Y. Iijima and K. Fukamichi: *Defect and Diffusion Forum*, **143-147**, 765 (1997).
- 6) K. Yamada, Y. Iijima and K. Fukamichi: *J. Mater. Res.* **8**, 2231 (1993).
- 7) T. Satoh, K. Fukamichi and Y. Satoh: *Sci. Rep. RITU*, **A-32**, 190 (1985).
- 8) T. Saito: *J. Magn. Soc. Jpn.*, **19**, 692 (1995).
- 9) K. Fukamichi, T. Goto, H. Komatsu and H. Wakabayashi: Proc. 4th Int. Conf. on Phys. Magn. Mater. (Poland), ed. by W. Gorkowski, H. K. Lachowicz and H. Szymczak, World Scientific Pub. Co Singapore, 354 (1989).
- 10) B. Dieny, B. Barbara, G. Fillion, M. Maeder and B. Michelutti: *J. de Phys.*, **48**, 1741 (1987).