Differential magnetic susceptibility of amorphous Fe-Y alloys

1993 J. Phys.: Condens. Matter 5 3003

(http://iopscience.iop.org/0953-8984/5/18/022)

J. Phys.: Condens. Matter 5 (1993) 3003–3010. Printed in the UK

Differential magnetic susceptibility of amorphous Fe–Y alloys

A Fujita, H Komatsu†, K Fukamichi and T Goto‡
Department of Materials Science, Faculty of Engineering, Tohoku University, Sendai 980, Japan

Received 5 January 1993

Abstract. The spin-glass behaviour of amorphous Fe_xY_{100-x} alloys ($70 \leqslant x \leqslant 90$) has been investigated by means of differential magnetic susceptibility dM/dH. In low magnetic fields, the dM/dH versus T curves show the peaks, indicating that the spin-glass state occurs through two steps. Two freezing temperatures are defined from the two peaks in the curve, i.e. the higher temperature is T_g and the lower temperature is T_f. From the results of the magnetization measurement as a function of temperature, i.e. the M versus T curve, it is indicated that the magnetization starts to decrease at T_g and a thermal irreversibility appears below T_f with decreasing temperature. With increase in the applied magnetic field, a third peak appears at around 150 K in the dM/dH versus T curve for amorphous $Fe_{84}Y_{16}$ alloy. The position of this peak corresponds to the upper inflection point in the M versus T curve, being defined as the Curie temperature T_C. Therefore, re-entrant behaviour appears in this system on application of a magnetic field. The concentration dependences of T_C and T_f for a field of 500 Oe are very similar to those obtained in zero field for other amorphous Fe–RE alloys (RE: rare-earth metal) such as Fe–Lu and Fe–Ce systems.

1. Introduction

The results obtained by investigation of the amorphous Fe_2Y alloy (Pickart *et al* 1974, Rhyne *et al* 1974) show that this alloy has a magnetic property with no long-range order and with hysteresis phenomena. Subsequently, magnetic susceptibility, Mössbauer effect and neutron scattering studies were carried out for amorphous Fe_2Y alloy (Forester *et al* 1979a,b) and these results indicate that amorphous Fe_2Y shows a spin-glass behaviour. The magnetic properties of amorphous Fe_xY_{100-x} with x = 32–88 at.% were investigated by magnetization and Mössbauer effect measurements (Chappart *et al* 1978, 1981, Coey *et al* 1981), and spin-glass behaviour with no ferromagnetic phase was observed even in much higher-reconcentration range than in conventional dilute spin-glass alloys. However, it has been revealed that the spin-glass behaviour in the amorphous Fe–Y system is unique, after the reports of the spin-glass behaviour in amorphous Fe–Zr alloys (Hiroyoshi and Fukamichi 1981, Saito *et al* 1986) and the systematic investigations on amorphous Fe–RE alloys (RE ≡ Y, La, Ce and Lu) (Fukamichi *et al* 1988, 1989a,b, Wakabayashi *et al* 1990, Goto *et al* 1991). That is, except for Y, these alloys exhibit a magnetic phase transition from the paramagnetic to the ferromagnetic phase before the spin-glass phase appears. On the other hand, amorphous Fe–Y alloys have no ferromagnetic phase over the whole range of compositions (Coey *et al* 1981) and the magnetic phase changes from the paramagnetic to the spin-glass state without passing through the ferromagnetic phase with decreasing

---

† Present address: Mitsui Petrochemical Industries Ltd., Nagaura, Sodegaura, Chiba 299-02, Japan.
‡ Institute for Solid State Physics, The University of Tokyo Roppongi, Minato-ku, Tokyo 106, Japan.

0953-8984/93/183003+08$07.50

temperature. All the other systems mentioned above show a direct transition from the paramagnetic to the spin-glass state at almost the same temperature above 90 at.% Fe, so it is considered that this spin glass behaviour is concerned with the intrinsic magnetism of amorphous pure Fe (Fukamichi *et al* 1989a,b). The spin-glass behaviour in amorphous pure Fe has been supported theoretically (Kakehashi 1991).

The differential susceptibility is sensitive to the magnetic phase transition because it is the second differential of the free energy with respect to the magnetic field. Therefore, it is well known that a transition from the paramagnetic to the ferromagnetic state results in a divergent-type peak at the Curie temperature in the curve of the temperature dependence of the differential susceptibility. The effectiveness of the measurement of differential susceptibility for spin-glass systems was considered in both calculation and experiments (Nieuwenhuys *et al* 1978). The calculation was based on the theory of the model proposed by Sherrington and Kirkpatrick (1975) and Kirkpatrick and Sherrington (1978). According to this model, the spin-glass state is determined by not only the magnetization M but also the spin-glass order parameter Q; $m = 0$ and $q \neq 0$. The free energy is obtained from the model of Sherrington and Kirkpatrick (1975) by using M and Q. Therefore, the differential magnetic susceptibility reflects the change in M and Q, and the calculation demonstrates that the differential magnetic susceptibility has a maximum at the spin-glass transition temperature. Accordingly, it is interesting to investigate in detail especially the spin-glass behaviour of amorphous Fe–Y alloys.

2. Experimental details

The amorphous samples used in the present investigation were prepared by DC high-rate sputtering at an argon gas pressure of 40 mTorr and a target voltage of 1.0 kV. They were accumulated onto a water-cooled copper substrate until their thickness became about 0.1–0.2 mm. The sputtered samples were confirmed as amorphous by x-ray diffraction. The Cu substrate was dissolved in a heating solvent of CrO_3 (500 g) + H_2SO_4 (27 cm^3) + H_2O (1000 cm^3) at 350 K. The magnetization was measured from 4.2 K to room temperature in various magnetic fields up to 1 kOe by means of a SQUID magnetometer. The differential magnetic susceptibility was calculated numerically from the adjoining two points of magnetization curves measured at each temperature.

3. Results and discussion

The temperature dependence of the differential magnetic susceptibility $\mathrm{d}M/\mathrm{d}H$ of amorphous $Fe_{84}Y_{16}$ alloy obtained at various DC fields is shown in figure 1. The $\mathrm{d}M/\mathrm{d}H$ curve measured in a field H of 10 Oe shows only one maximum and it becomes clearer with increasing magnetic field, eventually splitting into two peaks. Two peaks are also observed in the $\mathrm{d}M/\mathrm{d}H$ versus T curve at low temperatures for alloys with different Fe–Y compositions. This means that the spin-glass transition proceeds through two steps in the external magnetic field for the present amorphous Fe–Y system. Two transition temperatures T_f and T_g are defined as the boundary points of each step in the process of the spin-glass transition from the positions of each peak; the higher temperature is T_g and the lower temperature T_f. Both peaks shift to lower temperatures on increase in the applied field H, and the peak corresponding to T_f becomes sharp, although the peak corresponding to T_g becomes uncertain. As seen from the figure, the peak for T_g disappears and a third broad peak appears at around 150 K in a field of 700 Oe. According to the results from experiments on the conventional spin-glass system $PdFe_{0.0035}$ + 6.5 at.% Mn, and from the model calculation for this system (Nieuwenhuys *et al* 1978), the appearance of a new peak in the dM/dH versus T curve due to the contribution of ferromagnetic property is observed with increasing external magnetic field, although the dM/dH curve for this system shows a single peak at zero field and this peak is regarded as corresponding to the Curie temperature. The temperature of the third peak in the dM/dH versus T curve for amorphous $Fe_{84}Y_{16}$ alloy should also be the Curie temperature T_C. Further discussion of this peak will be given when we consider figure 3.

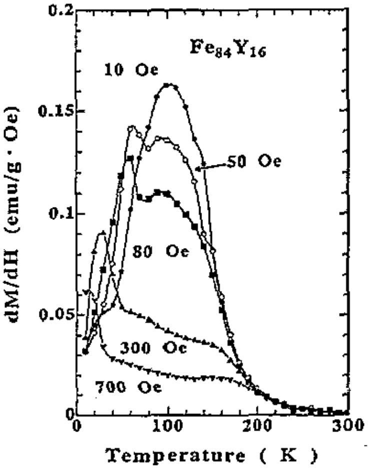

Figure 1. Temperature dependence of the differential magnetic susceptibility dM/dH measured at 10, 50, 80, 300 and 700 Oe for amorphous $Fe_{84}Y_{16}$ alloy.

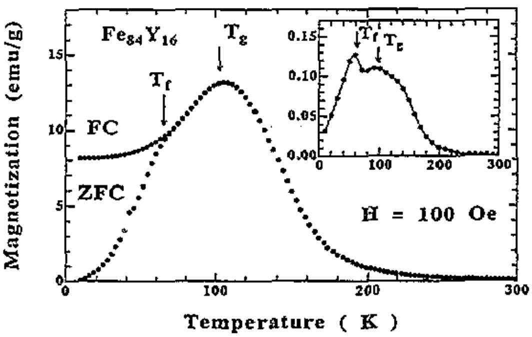

Figure 2. Temperature dependence of the magnetization for amorphous $Fe_{84}Y_{16}$ alloy at 100 Oe, measured after zero-field cooling (ZFC) and after field cooling (FC) at 100 Oe. The inset shows the dM/dH curve as a function of temperature for the same alloy at 100 Oe. The arrows indicate T_g and T_f.

Figure 2 shows the temperature dependence of the magnetization M measured at 100 Oe for amorphous $Fe_{84}Y_{16}$ in the zero-field-cooled and field-cooled states. The inset shows the dM/dH versus T curve at 100 Oe for the same alloy. The abscissa and ordinate axes show the temperature and dM/dH, respectively. As shown in the figure, the temperature T_g corresponds to the maximum and T_f to the shoulder where the zero-field-cooling line starts to deviate from the field-cooling line. The magnetization decreases in the region below T_g, and a marked thermal hysteresis of the magnetization appears in the region below T_f. Theoretical investigations of the spin-glass transition have been made in particular by using a mean-field model and the transitions described by de Almeida and Thouless (AT) (1978) and by Gabey and Toulouse (GT) (1981) are generally referred to in order to analyse the experimental results. The AT transition was originally derived from an Ising spin model using the replica symmetry trick (Kirkpatrick and Sherrington 1975) and it is defined according to AT as the change from the state in which the replica symmetry is stable to the state in which it is broken. According to the Heisenberg spin model with the hypothesis that average interaction is ferromagnetic, it is predicted by GT that only the transverse component of spin can initially freeze, i.e. the GT transition occurs, and subsequently the AT transition occurs with decreasing temperature. In this model, the replica symmetry is already broken after the occurrence of the GT transition; so the AT transition temperature for the vector spin with the general dimensions is considered as the crossover point from the state with weak irreversibility to the state with strong irreversibility (Elderfield and Sherrington 1984). From this point of view, T_g should be the GT transition temperature T_{GT}, and the decrease in magnetization below T_g is due to the freezing of the transverse component of spins. On the other hand, T_f should be at the AT transition temperature T_{AT} because an apparent thermal irreversibility is observed as a result of the broken replica symmetry, namely the freezing of the longitudinal component of spins below T_f.

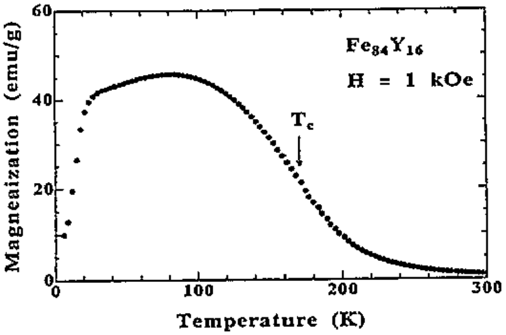

Figure 3. Temperature dependence of the magnetization for amorphous $Fe_{84}Y_{16}$ alloy at 1 kOe. The arrow indicates the Curie temperature T_C.

Figure 3 shows the temperature dependence of the magnetization, the M versus T curve, for amorphous $Fe_{84}Y_{16}$ alloy at 1 kOe. The temperature of the third peak which appears in the differential susceptibility as shown in figure 1 corresponds to the upper inflection point of the M versus T curve. Therefore, the temperature of the third peak is confirmed as the Curie temperature T_C. As mentioned above, the magnetic properties of amorphous Fe–Y system are unique, compared with those of other amorphous Fe-rich Fe–RE alloys (RE rare-earth metals) (Fukamichi *et al* 1989a,b). Amorphous Fe_xRE_{100-x} alloys exhibit re-entrant spin-glass behaviour in the Fe-rich region (except for $x > 90$ at.%) but in amorphous Fe–Y alloys there is a direct change from the paramagnetic to the spin-glass state. On close observation, the cusp of the AC susceptibility for amorphous $Fe_{80}Y_{20}$ alloy is relatively broad compared with that of amorphous $Fe_{90}Y_{10}$ alloy, at which Fe concentration other amorphous Fe–RE alloys have a narrow temperature range of the ferromagnetic phase or show almost a direct spin-glass transition. This implies that amorphous $Fe_{80}Y_{20}$ alloy lies at a composition very near to the occurrence of the ferromagnetic transition before freezing. Therefore, long-range ferromagnetic interactions are induced, or ferromagnetic-like alignments of spins are realized by applying an external magnetic field.

The field dependences of T_f, T_g and T_C for amorphous $Fe_{84}Y_{16}$ alloy are shown in figure 4(*a*). Both T_f and T_g strongly depend on the field strength and shift towards lower temperatures as the field increases. A re-entrant behaviour can be seen at around 400 Oe. Figures 4(*b*) and 4(*c*) show the field dependences of T_f, T_g and T_C for amorphous $Fe_{80}Y_{20}$ and $Fe_{90}Y_{10}$ alloys. The temperature T_C for amorphous $Fe_{80}Y_{20}$ alloy appears at 200 Oe and this strength of field is lower than that for amorphous $Fe_{84}Y_{16}$ alloy. On the other hand, the temperature T_C for amorphous $Fe_{90}Y_{10}$ alloy can be observed above 1.5 kOe, and this strength of field is rather higher than that of amorphous $Fe_{84}Y_{16}$ alloy and of amorphous $Fe_{80}Y_{20}$ alloy. T_f for amorphous $Fe_{90}Y_{10}$ alloy is observed even at 1.5 kOe and this disappears at 500 Oe for amorphous $Fe_{84}Y_{16}$ alloy and at 300 Oe for amorphous

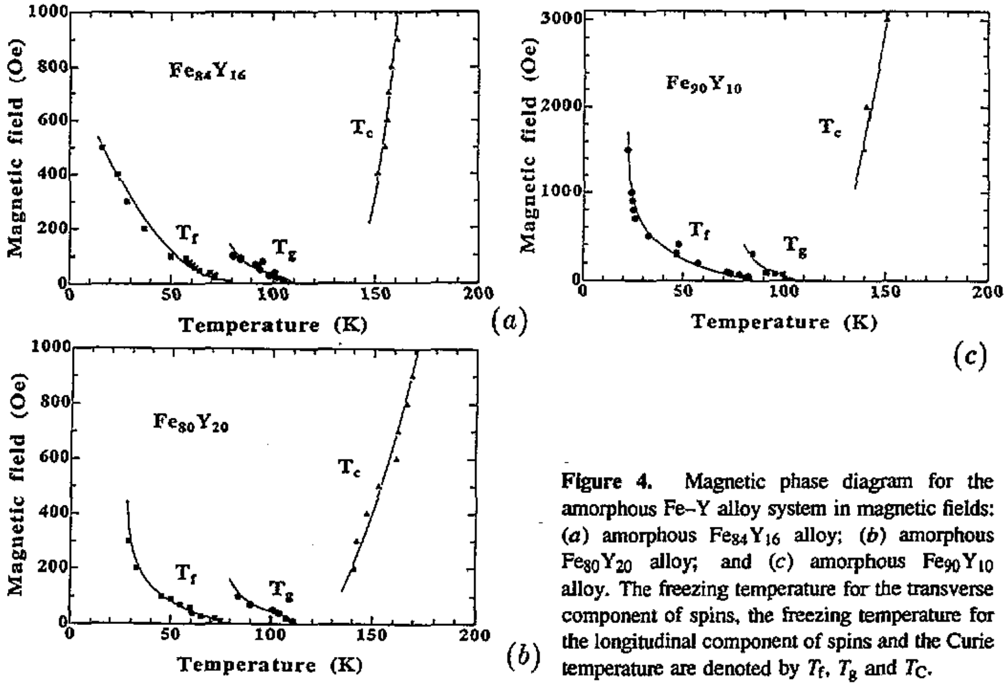

Figure 4. Magnetic phase diagram for the amorphous Fe–Y alloy system in magnetic fields: (*a*) amorphous $Fe_{84}Y_{16}$ alloy; (*b*) amorphous $Fe_{80}Y_{20}$ alloy; and (*c*) amorphous $Fe_{90}Y_{10}$ alloy. The freezing temperature for the transverse component of spins, the freezing temperature for the longitudinal component of spins and the Curie temperature are denoted by T_f, T_g and T_C.

$Fe_{80}Y_{20}$. It is therefore considered that ferromagnetic interaction becomes unstable while the antiferromagnetic interaction is developed, and frustration of both interactions becomes stronger with increasing Fe concentration.

The field dependences of T_{GT} and T_{AT} are theoretically simulated in the H versus T plane and these two transition temperatures are predicted to obey certain power laws (AT and GT). From the above discussion, T_g and T_f should vary as T_{GT} and T_{AT}. However, T_g and T_f do not obey the power laws predicted for T_{GT} and T_{AT}. This disagreement could arise for the following two reasons. Experimentally, it is known that the field dependence of the spin-glass transition temperatures varies as a function of the time scale of measurements (Salamon and Herman 1978), because the spin-glass state shows a marked relaxation phenomenon with different times depending on the system. Furthermore, the GT and AT transitions are derived from the Ising or the Heisenberg model; so, strictly speaking, these models are not suitable for the itinerant system in which spins fluctuate not only in the transverse direction but also in the longitudinal direction. However, the occurrence of the freezing of the transverse component of spins in Fe-rich amorphous alloy was also confirmed by Mössbauer effect measurements in magnetic fields for amorphous Fe–Zr alloys (Ghafari *et al* 1988, 1989, Ryan *et al* 1987), for amorphous Fe–La alloys (Wakabayashi *et al* 1989) and for amorphous Fe–Lu alloys (Goto *et al* 1991). For amorphous Fe–Lu alloys (Goto *et al* 1991), the temperature variations in coercive field and remanent magnetization have also been measured within the concentration region in which re-entrant behaviour appears and the results indicate that the re-entrant spin-glass phase of the amorphous Fe–Lu system is divided into two types of phase with weak irreversibility and strong irreversibility. From the above discussion, the spin-glass state of the amorphous Fe–Y system is also divided into two phases with a freezing of the transverse component of spins and with a strong irreversibility. Further investigations, e.g. AC susceptibility measurements with a DC bias field or Mössbauer effect measurements, are needed to study in detail these two freezing

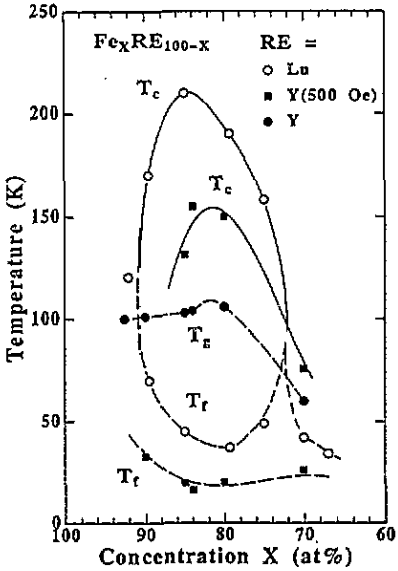

Figure 5. Concentration dependences of the Curie temperature T_C and the freezing temperature T_f for the longitudinal component of spins at 500 Oe for amorphous Fe–Y alloys. The spin-freezing temperature T_g in zero field for amorphous Fe–Y alloys and the Curie temperature T_C and spin freezing temperature T_f in zero field for amorphous Fe–Lu alloys in zero field (Fukamichi *et al* 1989) are also shown, for comparison.

steps in the spin glass.

The temperatures T_f and T_C in the same field clearly exhibit different concentration dependences. Figure 5 shows the concentration dependences of T_f and T_C defined from the dM/dH curve measured at 500 Oe. The temperatures T_f and T_C for amorphous Fe–Lu alloys (Goto *et al* 1991) and T_g for amorphous Fe–Y alloys (Fukamichi *et al* 1989a,b) determined from the AC susceptibility are also shown in the same figure, for comparison. As shown in the figure, T_C and T_f at 500 Oe for amorphous Fe–Y alloys have the same tendency as those in zero field for amorphous Fe–Lu alloys. For amorphous Fe–Y alloys, T_C at 500 Oe increases with decreasing Fe concentration and starts to decrease beyond 80 at.% Fe, and T_f at 500 Oe continues to decrease up to the same concentration, while the maximum of T_C and the minimum of T_f in zero field for amorphous Fe–Lu alloys also occur at around 85 at.% Fe.

Recently, the magnetism of amorphous transition metals and alloys has been theoretically discussed on the basis of a finite-temperature theory, and an itinerant-type spin-glass state has been proposed (Kakehashi 1991). According to this theory, the spin glass of Fe-rich amorphous Fe-RE alloys is a cluster spin glass. In the CPA approximation, the ferromagnetic interaction of the intracluster and the antiferromagnetic interaction reflected from the effective medium are frustrated, and it is predicted that the ferromagnetic phase which appears during the re-entrant behaviour originates from these ferromagnetic clusters and their growth. In the case of the amorphous Fe–Y system, it is considered that frustration of the ferromagnetic and antiferromagnetic interactions is stronger than for other amorphous Fe–RE systems, resulting in the lack of a ferromagnetic phase; the external field assists the ferromagnetic interactions and growth of clusters, and then the Curie temperature appears when a magnetic field is applied. However, this theory for an itinerant-type spin glass neglects both the influence of external fields and the transverse fluctuation for simplicity; so information on the freezing of the transverse component of spins and the field dependence of its freezing temperature cannot be obtained.

In an Fe-rich amorphous system, the Fe moment and the interaction between its nearest-neighbour Fe moments are mainly determined by the d-electron number and the local environment effect (LEE) related to the fluctuation in the atomic configuration of the first-nearest-neighbour shell (Kakehashi 1991). The magnitude of this structural fluctuation affects the stability of ferromagnetism; a large fluctuation makes the ferromagnetic phase unstable and the frustration becomes stronger, that is to say T_C becomes lower and T_f higher with increasing structural fluctuation. The lack of a ferromagnetic phase in amorphous Fe–Y alloys suggests that the fluctuation in the atomic configuration is larger than other amorphous Fe–RE alloys. In the binary alloy system, the LEE is also characterized by the coordination number (CN) and the short-range order (SRO) parameter of each element; so the size effect of the second element should be introduced in the LEE (Kakehashi *et al* 1992). The theory of electronic-structure calculations in amorphous transition-metal alloys thus predicts the concentration dependence of the electronic configuration for amorphous Fe–Zr alloys (Kakehashi *et al* 1992). The magnetic phase diagram for amorphous Fe–Zr alloys can be simulated numerically (Yu *et al* 1992), accounting for the LEE predicted by the above theory (Kakehashi *et al* 1992); the calculated result is qualitatively consistent with the experimental result (Hiroyoshi and Fukamichi 1981). Similarity between the magnetic phase diagram in zero field for amorphous Fe–RE alloys and that in an external field for amorphous Fe–Y alloys suggests that the concentration dependences of T_C and T_f in the external field for amorphous Fe–Y alloys also reflect the CN and the SRO parameter through the LEE. Furthermore, the CN and the SRO parameter are affected by not only the size of element but also the chemical affinity of the second element for Fe. A strong affinity allows the second element to be located on the first-nearest neighbour of the Fe site; so the CN of Fe around the Fe site decreases and the SRO parameter changes. Therefore, fluctuations in distance and in coordination between nearest Fe atoms should become larger than for pure amorphous Fe in binary amorphous alloys in which the elements have a strong affinity. In the crystalline Fe–Y system, many compounds exist in the Fe-rich region. In this system, the most stable distance between Fe atoms and the CN of Fe around change depending on the Fe concentration. When this fact is taken into account, it is believed that a large fluctuation in structure due to the fluctuation in concentration and the affinity of Y for Fe causes the different magnetic properties of amorphous Fe–Y alloys from those of other amorphous Fe–RE alloys. As a result of the present investigation, it is revealed that the spin-glass behaviour in the amorphous Fe–Y system is essentially the same as in other amorphous Fe-RE alloys.

4. Conclusion

The differential magnetic susceptibility measurements for amorphous Fe–Y alloys have been presented in order to investigate the spin-glass behaviour. In the external magnetic field, three types of transition temperature appear although, in zero field, only one freezing temperature is observed and comparison between the magnetic phase diagram in external fields for amorphous Fe–Y alloys with those in zero field for other amorphous Fe–RE (RE ≡ rare-earth metals) is also presented. The results are summarized as follows.

(1) The temperature dependence of the differential magnetic susceptibility dM/dH for amorphous Fe–Y alloys exhibits two peaks in low-temperature ranges on application of a weak magnetic field, and two freezing temperature T_g and T_f are defined from the positions of these peaks.

(2) The magnetization starts to decrease below T_g and thermal irreversibility appears below T_f; both T_g and T_f shift towards lower-temperature ranges with increasing magnetic field.

(3) When a strong magnetic field is applied, a third peak, the temperature of the inflection point of the magnetization corresponds, appears in the $\mathrm{d}M/\mathrm{d}H$ curve indicating that this temperature corresponding to the third peak is the Curie temperature T_C.

(4) A re-entrant spin-glass behaviour is observed in the field versus temperature plane. Both T_f and T_g shift towards lower temperatures while T_C shifts towards a higher temperature with increasing magnetic field.

(5) The concentration dependences of T_C and T_f in external magnetic fields are analogous to those of amorphous Fe–RE alloys in zero field. This implies that the spin-glass behaviour in the amorphous Fe–Y system is essentially the same as those of other amorphous Fe–RE alloys.

Acknowledgment

This work was supported by a Grant-in-Aid for Scientific Research (04402045) from the Ministry of Education, Science and Culture in Japan.

References

Chappart J, Arrese-Boggiano R and Coey J M D 1978 *J. Magn. Magn. Mater.* **7** 175–7

Chappart J, Coey J M D, Liénard A and Rebouillat J P 1981 *J. Phys. F: Met. Phys.* **11** 2727–44

Coey J M D, Givord D, Liénard A and Rebouillat J P 1981 *J. Phys. F: Met. Phys.* **11** 2707–25

de Almeida J R L and Thouless D J 1978 *J. Phys. A: Math. Gen.* **11** 983–90

Elderfield D and Sherrington D 1984 *J. Phys. C: Solid State Phys.* **17** 5595–602

Forester D W, Koon N C, Schelleng J H and Rhyne J J 1979a *Solid State Commun.* **30** 177–80

—— 1979b *J. Appl. Phys.* **50** 7336–41

Fukamichi K, Komatsu H, Goto T and Wakabayashi H 1988 *Physica* B **149** 276–80

Fukamichi K, Goto T, Komatsu H and Wakabayashi H 1989a *Proc. 4th Int. Conf. on the Physics of Magnetic Materials (Poland, 1989)* ed W Gorkouski, H K Lachowicks and H Szymczak (Singapore: World Scientific) p 354–81

Fukamichi K, Komatsu H, Goto T, Wakabayashi H and Matsuura M 1989b *Proc. MRS Int. Meet. on Advanced Materials* vol 11, p 285–99

Gabey M and Toulouse G 1981 *Phys. Rev. Lett.* **47** 201–4

Ghafari M, Keune W, Brand A, Day R K and Dunlop J B 1988 *Mater. Sci. Eng.* **99** 65–8

Ghafari M, Chmielek N, Keune W and Foley C P 1989 *Physica* B **161** 222–4

Goto T, Kuroda K, Fukamichi K, Komatsu H, Morimoto S and Ito A 1991 *J. Phys. Soc. Japan* **60** 4286–99

Hiroyoshi H and Fukamichi K 1981 *Phys. Lett.* **85A** 242–4

Kakehashi Y 1991 *Phys. Rev.* B **42** 10820–31

Kakehashi Y, Tanaka H and Yu M 1992 *Phys. Rev.* B at press

Kirkpatrick S and Sherrington D 1978 *Phys. Rev.* B **17** 4384–403

Nieuwenhuys G J, Stocker H, Verbeek B H and Mydosh J A 1978 *Solid State Commun.* **27** 197–9

Pickart S J, Rhyne J J and Alperin H A 1974 *Phys. Rev. Lett.* **33** 424–7

Rhyne J J, Schelleng J H and Koon N C 1974 *Phys. Rev.* B **10** 4672–9

Ryan D H, Coey J M D, Batalla E, Altounian Z and Ström-Olsen J O 1987 *Phys. Rev.* B **35** 8630–8

Saito N, Hiroyoshi H, Fukamichi K and Nakagawa K 1986 *J. Phys. F: Met. Phys.* **16** 911–9

Salamon M B and Herman R M 1978 *Phys. Rev. Lett.* **41** 1506–9

Sherrington D and Kirkpatrick S 1975 *Phys. Rev. Lett.* **29** 1792–6

Wakabayashi H, Goto T, Fukamichi K, Komatsu H, Morimoto S and Ito A 1989 *J. Phys. Soc. Japan* **58** 3383–91

Wakabayashi H, Goto T, Fukamichi K and Komatsu H 1990 *J. Phys.: Condens. Matter* **2** 417–29

Yu M, Kakehashi Y and Tanaka H 1992 submitted