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Random magnetic anisotropy and competition of interactions due to exchange fluctuations in amorphous Er–Cu alloys

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Abstract. Magnetic properties have been investigated for amorphous Er_xCu_{100-x} (x = 25, 30, 40, 50, 60 and 70) alloys prepared by high-rate DC sputtering. The paramagnetic Curie temperature θ_p of these alloys is positive and it means that ferromagnetic exchange interactions are predominant. However, a steep decrease of θ_p occurs below about $x = 50$, suggesting a development of some antiferromagnetic exchange interaction. These alloys exhibit spin glass-like behaviour, and the spin freezing temperature T_f decreases with decreasing Er content, but an abrupt change occurs around $x = 40 \sim 50$ in analogy with that of θ_p. The spin freezing is dominantly caused by the RMA in high Er concentration ranges above about $x = 50$. On the other hand, not only the RMA but the competition of ferromagnetic and antiferromagnetic interactions due to the exchange fluctuations contribute to the magnetic properties in low Er concentration ranges below about $x = 50$. Consequently, RMA becomes weaker, whereas the competition of the exchange interaction becomes stronger with decreasing Er content. The concentration dependence of magnetization per Er atom is also explained by considering both the random magnetic anisotropy and the competition of the exchange interaction.

1. Introduction

There has been considerable interest in the experimental and theoretical studies of the magnetic properties for amorphous non-S state rare earth (RE) alloys (Harris *et al* 1973, Coey 1978, O'Shea and Lee 1991). It has been well known that the magnetization curves of these amorphous alloys are not saturated easily (Boucher 1977a, b, Ferrer *et al* 1978) and they exhibit a pronounced large coercive force H_c at low temperatures (Rhyne *et al* 1974, Tissier *et al* 1980). In these alloys, the local electrostatic field acts on the localized 4f moment of RE and produces huge anisotropy randomly-oriented because of the random atomic structure in the amorphous state. Magnetic properties of these alloys have been explained in terms of such an anisotropy called random magnetic anisotropy (RMA) (Harris *et al* 1973). When the anisotropy is larger than the exchange interaction, the magnetic moments of RE are distributed randomly in a hemisphere having the applied field as the polar direction. It had been considered that this leads to the saturation moment of about half the free ion moment and such a magnetic order has been called 'asperomagnet' (Coey 1978). However, the absence of long-range magnetic order due to the RMA has been discussed theoretically (Aharony and Pytte 1980). In fact, ferromagnetic coupling is suppressed by the RMA, and spin glass-like behaviour has been observed in these amorphous alloys containing non-S state RE elements (Boucher 1977a, b, Coey and Molnar 1978, Molnar *et al* 1980). This magnetic structure has been named 'speromagnetic' because the origin of the spin

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freezing is the RMA rather than the competition of ferromagnetic and antiferromagnetic interactions (Sellmyer and Nafis 1985).

In spite of the absence of RMA the S-state Gd based alloys such as amorphous Gd-Al (Mizoguchi *et al* 1977a, b, McGuire *et al* 1978) and amorphous Gd-Cu (Heiman and Kazama 1978, McGuire *et al* 1978) alloys also exhibit spin glass behaviour in low concentration ranges of the magnetic element. The frustration in these alloys originates from the competition of the RKKY exchange interaction.

It has been pointed out that the fluctuations of the exchange interaction also exist in amorphous non-S state RE alloys (Boucher 1977a, b, Sellmyer and Nafis 1985). However, the systematic investigation of magnetic properties taking both the RMA and the exchange fluctuations into consideration for non-S state RE amorphous alloys has not yet been carried out. Therefore, the concentration dependence of the magnetic properties such as the magnetization, the paramagnetic Curie temperature θ_p and the spin freezing temperature T_f for the amorphous Er_xCu_{100-x} alloys have been investigated. The concentration dependence of magnetization for amorphous Er_xNi_{100-x} ($x = 33$, 50 and 80) and Gd_xAl_{100-x} ($x = 30$, 40, 50 and 60) alloys has also been investigated to clarify the effect of the RMA and the fluctuations of exchange interaction on the magnetization. In this paper, we will show that both the RMA and the exchange fluctuations play an important role in the magnetic properties of the amorphous Er_xCu_{100-x} alloys, depending on the Er concentration.

2. Experiment

Alloy targets with a diameter of about 50 mm were made by arc-melting 99.9 wt.% pure Er and Gd, 99.99% Cu and Al, and 99.9% Ni in an argon gas atmosphere purified with a Ti getter. Bulk amorphous Er_xCu_{100-x} ($x = 25$, 30, 40, 50, 60 and 70) and Er_xNi_{100-x} ($x = 33$, 50 and 80) alloys about 0.2 $\sim$ 0.3 mm thick were prepared by high-rate DC sputtering on a water-cooled Cu substrate. The argon gas pressure during sputtering was 40 mTorr and the target voltage was 1.0 kV. The Cu substrate was removed from the samples by mechanical polishing. Amorphous $Gd_{30}Al_{70}$ and $Gd_{40}Al_{60}$ alloys were made by high-rate DC sputtering and $Gd_{50}Al_{50}$ and $Gd_{60}Al_{40}$ alloys by rapidly melt-quenching using a single-roller apparatus. Their amorphous state was confirmed by x-ray diffraction using Cu-Kα radiation. The magnetization up to 55 kOe, the temperature dependence of the DC magnetic susceptibility and the magnetic cooling effect were measured with a SQUID magnetometer (Quantum Design). Very high-field measurements up to 360 kOe were made using a pulse magnet. The AC magnetic susceptibility measurement was carried out by a mutual induction method with 80 Hz in 1 Oe.

3. Results and discussion

The DC magnetic susceptibility of amorphous Er_xCu_{100-x} alloys exhibits a Curie–Weiss type temperature dependence in wide temperature ranges as shown in figure 1. The effective magnetic moment μ_{eff} is deduced from the Curie constant given by the following conventional expression;

$$\chi - \chi_0 = \frac{C}{(T - \theta_p)} \tag{1}$$

where χ_0 is the temperature independent susceptibility and θ_p the paramagnetic Curie temperature. The effective magnetic moment μ_{eff} of these amorphous alloys is very close to $9.6\mu_B$ of free Er^{3+} ion. Figure 2 shows the concentration dependence of θ_p for the amorphous Er_xCu_{100-x} alloys. All these alloys exhibit a positive value of θ_p, indicating that ferromagnetic Er–Er exchange interactions are predominant. It is worth noting that the value of θ_p decreases rapidly below about $x = 50$. This concentration dependence suggests that antiferromagnetic interactions increase rapidly below this concentration because the magnitude of θ_p is proportional to the sum of all exchange interactions. The magnetic susceptibility of these alloys deviates from the Curie–Weiss law at very low temperatures, indicating the existence of a magnetic ordering. Many amorphous alloys containing rare-earth ion often exhibit spin glass-like behaviour (Boucher 1977a, b, Coey and Molnar 1978). Therefore, the magnetic field cooling effect has been investigated. Figure 3 displays the temperature dependence of zero field-cooled (ZFC) and field-cooled (FC) magnetizations of the amorphous Er_xCu_{100-x} alloys in a field of 100 Oe. An apparent hysteresis between ZFC and FC magnetizations is observed. In addition, the temperature dependence of AC magnetic susceptibility shows a characteristic cusp of the spin glass as shown in figure 4. Other amorphous Er–Cu alloys exhibit similar behaviour. In this manner, the amorphous Er_xCu_{100-x} alloys exhibit spin glass-like behaviour. From the AC magnetic field measurements, the spin freezing temperature was determined and its concentration dependence is given in figure 5. The spin freezing temperature T_f increases with the increase of the Er content. However, the concentration dependence of T_f is not linear and exhibits an abrupt increase about $x = 50$, being analogous to that of θ_p. As shown in the inset of figure 5, such a concentration dependence has also been observed in amorphous Dy–Cu alloys (Molnar *et al* 1980), although the existence of the steep change and its origin have not been discussed. It is worth noting that the Cu content of the steep change of T_f for the amorphous Er–Cu alloys is close to that of the occurrence of the spin freezing for amorphous Gd–Cu alloys (McGuire *et al* 1978). The magnetic properties of amorphous Gd–Cu alloys have already been investigated (Mizoguchi *et al* 1977a, McGuire *et al* 1978, Heiman and Kazama 1978) and its magnetic phase diagram has been reported (McGuire *et al* 1978). For high Gd contents, they are ferromagnetic, and then spin glass state occurs with decreasing Gd content, indicating the existence of competition of positive and negative exchange interactions owing to the fluctuations of the interaction. The magnetic properties of the amorphous Er–Cu alloys with a large RMA would also be affected by the fluctuations of exchange interaction in a similar manner to the amorphous Gd–Cu alloys. That is, the spin glass-like behaviour in high Er concentration ranges above about $x = 50$ is considered to be caused by the RMA. Therefore, the magnetic structure in high concentration above about $x = 50$ is speromagnetic. On the other hand, not only the RMA but the competition of ferromagnetic and antiferromagnetic interactions due to the exchange fluctuations contribute to the spin glass-like behaviour in low Er contents below about $x = 50$, and hence the magnetic structure in low concentration would be not simple speromagnetic but more complex one.

The magnetic properties of amorphous non-S state RE alloys have been explained by the HPZ (Harris–Plischke–Zuckermann) model (Harris *et al* 1973). Taking into account the exchange fluctuations, which often induce the competition of ferromagnetic and antiferromagnetic interactions, the following Hamiltonian has been proposed (Sellmyer and Nafis 1985)

$$H = -\sum(J_0 + \Delta J_{ij})S_iS_j - D\sum(n_i \cdot S_i)^2 - g\mu_B H\sum J \qquad (2)$$

where J_0 is the average ferromagnetic exchange interaction and ΔJ_{ij} represents the exchange fluctuations, S the total angular momentum and n denotes the direction of the local

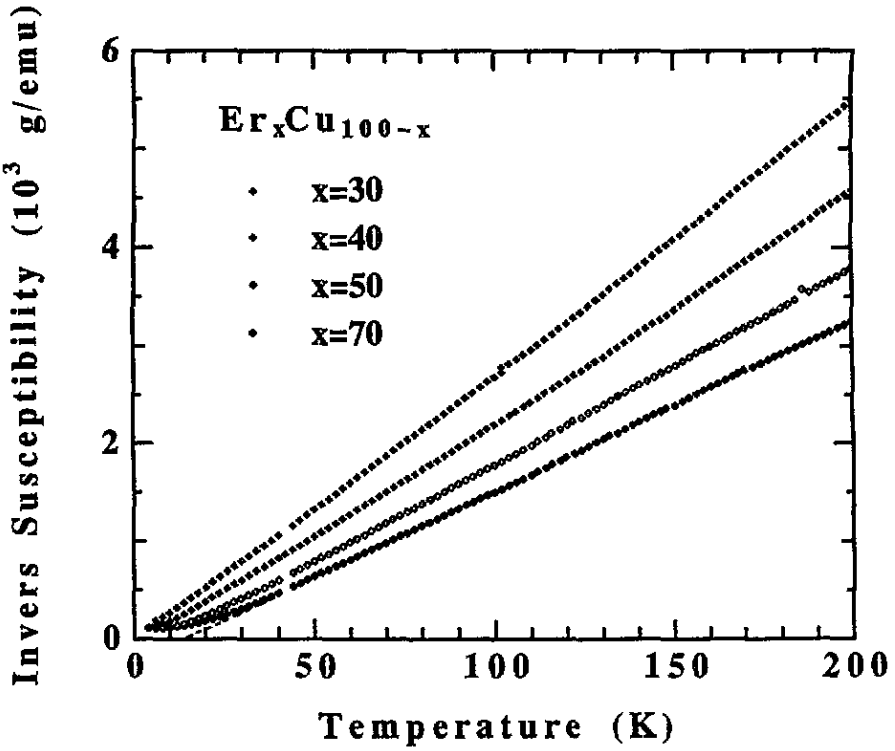

Figure 1. Temperature dependence of inverse DC magnetic susceptibility for amorphous Er_xCu_{100-x} (x = 30, 40, 50 and 70) alloys.

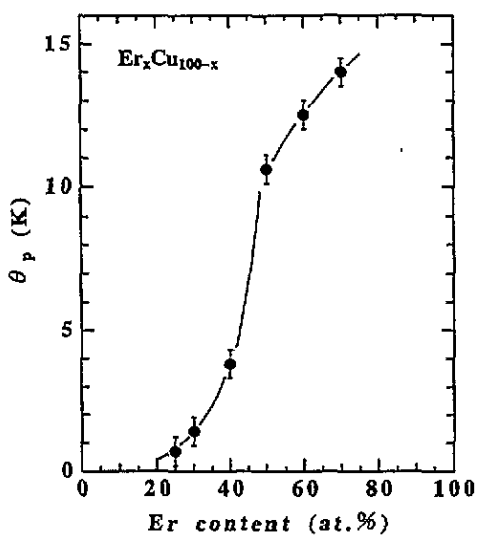

Figure 2. Concentration dependence of the paramagnetic Curie temperature θ_p for amorphous Er_xCu_{100-x} (x = 30, 40, 50, 60 and 70) alloys.

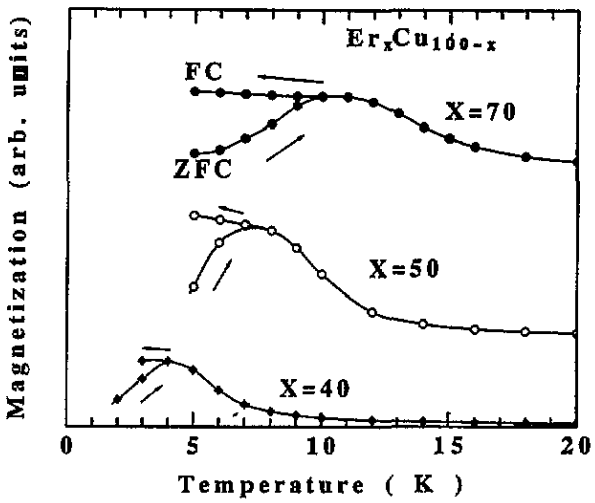

Figure 3. Temperature dependence of the ZFC and FC magnetizations for amorphous Er_xCu_{100-x} ($x = 40$, 50 and 70) alloys in a field of 100 Oe.

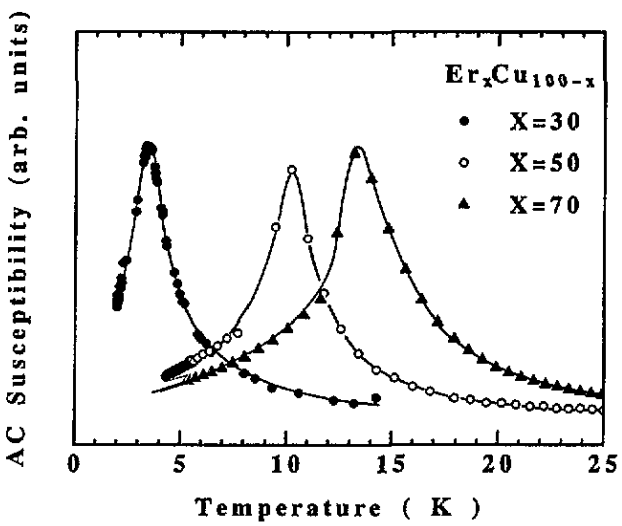

Figure 4. Temperature dependence of the AC magnetic susceptibility for amorphous Er_xCu_{100-x} ($x = 30$, 50 and 70) alloys in a field of 1 Oe with 80 Hz.

anisotropy D at site i and, as mentioned above, both the RMA and the competition exist in the amorphous Er–Cu alloys with low Er content. It is important to separate both effects in order to understand the magnetic properties of the amorphous Er–Cu alloys.

We discuss the fluctuation effect in amorphous S-state Gd-based alloys which has no orbital angular momentum in order to exclude the effect of the RMA. Because of the well-shielded nature of 4f electrons of RE, the exchange interaction of alloys and compounds containing RE element arises from the RKKY interaction (Ruderman and Kittel 1954, Kasuya 1956, Yoshida 1957). As is well known the function oscillates and gives both

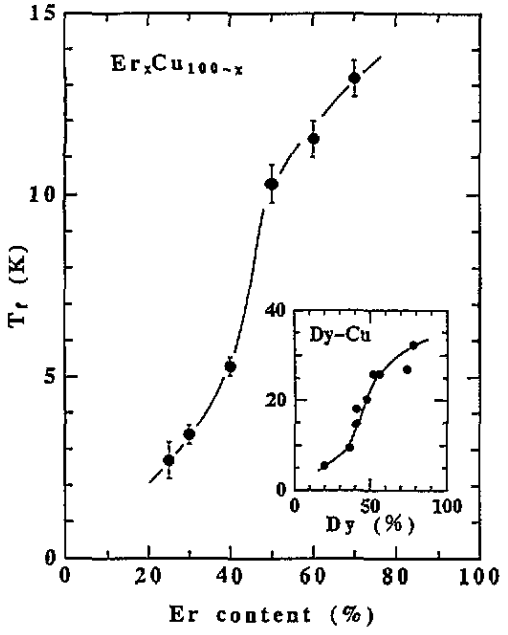

Figure 5. Magnetic phase diagram for amorphous Er_xCu_{100-x} (x = 25, 30, 40, 50, 60 and 70) alloys, together with that for amorphous Dy–Cu alloys in the inset (Molnar *et al* 1980).

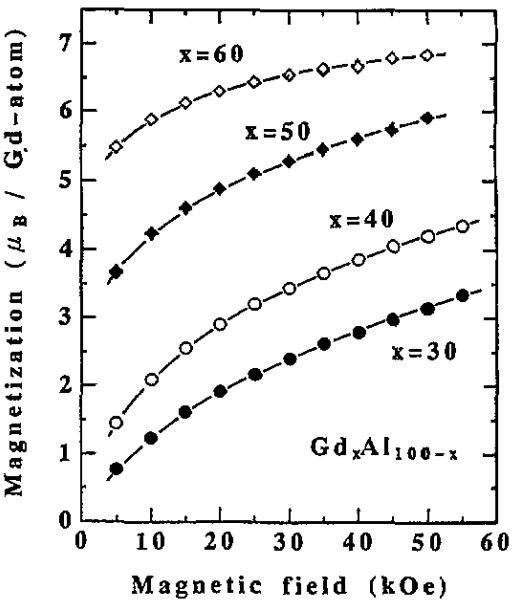

Figure 6. Magnetization per Gd atom up to 20 kOe at 4.2 K for amorphous Gd_xAl_{100-x} (x = 30, 40, 50 and 60) alloys.

ferromagnetic and antiferromagnetic interactions. It has been reported that the value of q increases in accordance with the decrease in the Gd concentration x, leading to the increase in the antiferromagnetic interaction. The effect of this exchange fluctuation is observed in the magnetization process. Figure 6 displays the magnetization curves up to 20 kOe for amorphous Gd_xAl_{100-x} (x = 30, 40, 50 and 60) alloys which have fluctuations of the RKKY interaction with no RMA. The magnetization per Gd atom decreases with decreasing Gd content because of increase in the competition of ferromagnetic and antiferromagnetic interactions. The intrinsic concentration dependence of the magnetic state for these four samples is so large that some difference related to the preparation technique was not observed.

Comparing the amorphous Gd_xNi_{100-x} alloys with the amorphous Gd_xCu_{100-x}, it is considered that the value of x for the amorphous Gd_xNi_{100-x} alloys lie on a lower q region, enhancing the ferromagnetic RKKY interaction (Hattori *et al* 1995). In fact, an amorphous $Gd_{26}Ni_{74}$ alloy is ferromagnetic with T_c = 38 K (Asomoza *et al* 1979), although an amorphous $Gd_{31}Cu_{69}$ alloy is a spin glass (McGuire *et al* 1978). In the amorphous RE–Ni system, therefore, the influence of fluctuations of the RKKY exchange interaction on magnetic properties is very small compared with that of the amorphous RE–Cu alloys. Therefore, the effect of the RMA on the magnetization is clarified by investigation of the amorphous non-S state RE–Ni alloys (Hattori *et al* 1995). Figure 7 displays the magnetization per Er atom of the amorphous Er_xNi_{100-x} alloys up to 55 kOe at 4.2 K. It should be noted that Ni is regarded as non-magnetic in the present amorphous Er_xNi_{100-x} ($x \geqslant 33$) alloys (Hattori *et al* 1995). The magnitude of magnetization per Er atom decreases with increasing Er content as seen from the figure. This result indicates that

the ferromagnetic couplings decrease with the increase in Er content. That is to say, the RMA, which suppresses the ferromagnetic coupling, becomes stronger with increasing Er content (Hattori *et al* 1995). Note that the concentration dependence of magnetization per RE atom is opposite to that of the amorphous Gd–Al alloys as seen in figure 5.

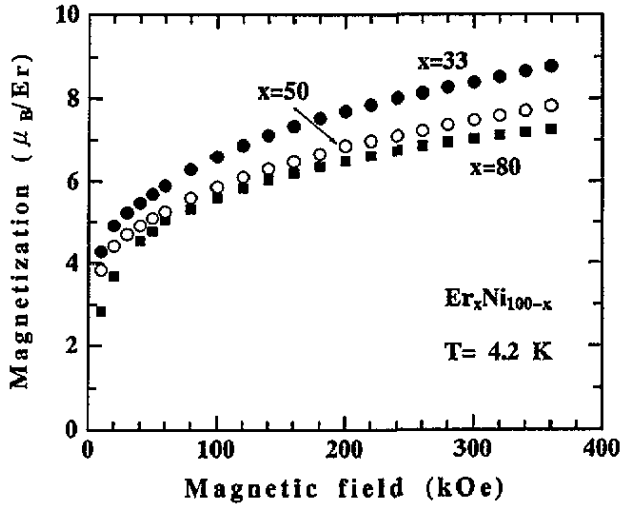

Figure 7. Magnetization per Er atom up to 360 kOe at 4.2 K for amorphous Er_xNi_{100-x} (x = 33, 50 and 80) alloys.

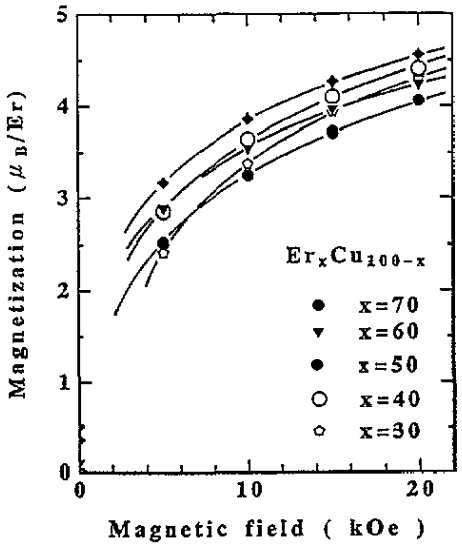

Figure 8. Magnetization per Er atom up to 20 kOe at 4.2 K for amorphous Er_xCu_{100-x} (x = 30, 40, 50, 60 and 70) alloys.

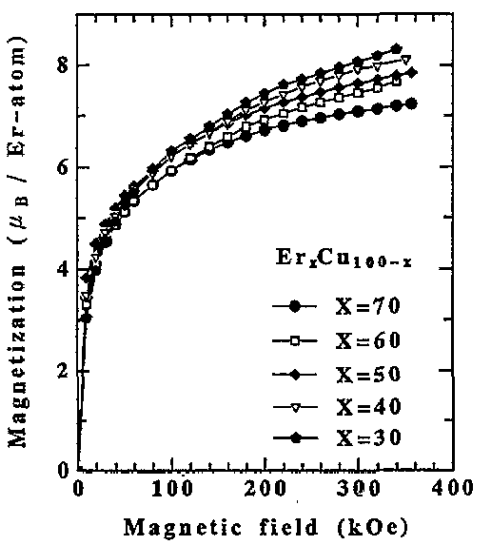

Figure 9. Magnetization curves up to 360 kOe for amorphous Er_xCu_{100-x} (x = 30, 40, 50, 60 and 70) alloys.

In the case of the amorphous Er–Cu alloys, not only the RMA but also the exchange fluctuations contribute to the concentration dependence of magnetization. Figure 8 shows the magnetization per Er atom of the amorphous Er_xCu_{100-x} alloys up to 20 kOe at 4.2 K. The magnetization increases with decreasing Er content from $x = 70$ to 50 due to a decrease in the RMA in analogy with that of the amorphous Er_xNi_{100-x} ($x = 33$, 50 and 80) alloys. On the contrary, in low concentration ranges the magnetization decreases with decreasing Er content from 50 to 30 in accordance with increase in the fluctuations of the RKKY exchange interaction in a similar manner to the amorphous Gd_xAl_{100-x} alloys. Therefore, the concentration dependence of magnetization up to 20 kOe is explained by the effect of both the fluctuations of the RKKY interaction and the RMA. It is important to bear in mind that the Er content which exhibits an abrupt change of magnetization and θ_p is same to each other.

The random magnetic anisotropy constant D is related to the high-field susceptibility, χ_{hf}, and given by the following expression (Asomoza *et al* 1979)

$$D = \frac{g^2\mu_B^2}{3\chi_{hf}}\left(1 + \frac{3k\theta_p\chi_{hf}}{g^2J^2\mu_B^2}\right) \tag{3}$$

where θ_p is the paramagnetic Curie temperature, k_B the Boltzmann constant, g the Landé g-factor and J the total angular momentum. In order to estimate the constant D, we have extended the magnetization measurements up to 360 kOe using a pulse magnet. Figure 9 shows the field dependence of magnetization per Er atom up to 360 kOe for the amorphous Er_xCu_{100-x} alloys at 4.2 K. The magnetization curves are not saturated but vary almost linearly with the applied magnetic field above 200 kOe. It should be emphasized that the magnitude of magnetization above 200 kOe increases with decreasing Er content, contrasting with the magnetization up to 20 kOe in figure 8. Figure 10 shows the concentration dependence of magnetization for the amorphous Er_xCu_{100-x} alloys at 20, 50, 100, 200 and 350 kOe. The concentration dependence of magnetization at 20 kOe exhibits a maximum at $x = 50$ owing to the balance of both the RMA and the exchange fluctuations as mentioned above. With increasing applied magnetic field, this concentration dependence becomes vague, and the magnetization above 200 kOe exhibits a monotonic decrease with increasing Er content. This result clearly indicates that the effect of exchange fluctuations is reduced and the RMA becomes dominant in very high magnetic fields. Therefore, we can exclude the effect of exchange fluctuations and estimate the magnitude of the RMA by means of the high-field magnetization measurements. The value of χ_{hf} was obtained by fitting the magnetization curves above 200 kOe at 4.2 K in figure 9. In figure 11 the concentration dependence of the random magnetic anisotropy constant D for the amorphous Er_xCu_{100-x} alloys is shown, together with that for the amorphous Er_xNi_{100-x} alloys. The magnitude of D for the amorphous Er_xCu_{100-x} alloys in the present study is comparable to the results reported previously for amorphous $Er_{25}Ni_{75}$ (Asomoza *et al* 1979) alloy and amorphous $(RE_{75}Au_{25})_{90}B_{10}$ (RE = Pr, Tb and Er) alloys (Sellmyer *et al* 1980). The value of D increases with the Er content, namely the RMA becomes larger in a similar manner to the amorphous Er_xNi_{100-x} ($x = 33$, 50 and 80) alloys. Strictly speaking, the increment of D is much more remarkable above 50% Er.

4. Summary

The magnetic properties of the amorphous Er_xCu_{100-x} ($x = 25$, 30, 40, 50, 60, and 70) alloys have been investigated. Both the random magnetic anisotropy and the fluctuations of the RKKY exchange interaction play an important role in the magnetic properties, depending on the Er concentration. The main results are summarized as follows.

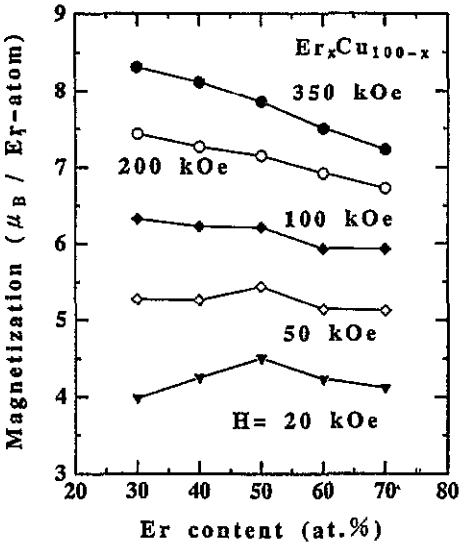

Figure 10. Concentration dependence of the magnetization at 20, 50, 150 and 350 kOe for amorphous Er_xCu_{100-x} (x = 30, 40, 50, 60 and 70) alloys.

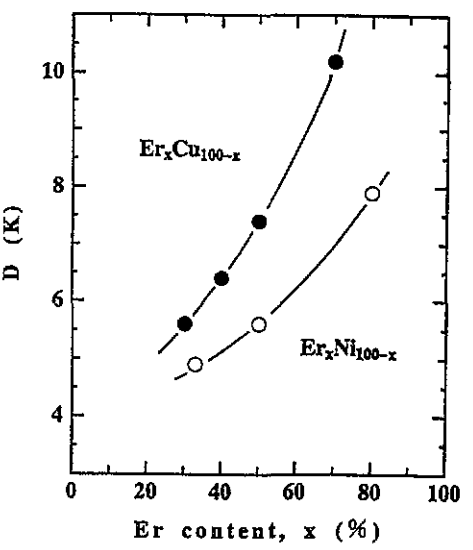

Figure 11. Concentration dependence of the random magnetic anisotropy constant D for amorphous Er_xCu_{100-x} (x = 30, 40, 50 and 70) alloys, together with that for amorphous Er_xNi_{100-x} (x = 33, 50 and 80) alloys.

(i) With decreasing Er content, the paramagnetic Curie temperature θ_p exhibits a steep decrease below about $x = 50$, indicating the steep increase in antiferromagnetic interactions.

(ii) Amorphous Er–Cu alloys show spin glass-like behaviour and the spin freezing temperature T_f increases with increasing Er content, and its increase is steep about $x = 50$, being analogous to the concentration dependence of θ_p.

(iii) In high Er concentration ranges above about $x = 50$, the random magnetic anisotropy dominantly causes the spin glass-like state. On the other hand, the spin glass state comes from both the fluctuation of the RKKY exchange interactions and the random magnetic anisotropy in low concentration ranges below about $x = 50$.

(iv) The magnetization per Er atom increases with decreasing Er content because of the decrease in the random magnetic anisotropy in high concentration range above about $x = 50$. However, it decreases with decreasing Er content because of the fluctuations of the RKKY exchange interaction in low Er contents below about $x = 50$.

(iv) In very high magnetic field above 200 kOe, the random magnetic anisotropy dominantly suppresses the ferromagnetic couplings, and the effect of the competition due to the exchange fluctuations on the magnetization is negligibly small.

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