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Magnetisation and magnetovolume effect in amorphous $R_{0.2}Fe_{0.8}$ ($R \equiv Y, Pr, Nd, Sm, Gd, Tb, Dy$ and Er) alloys

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Abstract. Magnetisation, thermal expansion and forced volume magnetostriction of amorphous $R_{0.2}Fe_{0.8}$ ($R \equiv Y, Pr, Nd, Sm, Gd, Tb, Dy$ and Er) alloys have been measured to make clear the contribution of R elements to both the Fe moment m_{Fe} and the magnetovolume effect. The value of m_{Fe} in these alloys ranges from $1.6\mu_B$ to $2.3\mu_B$, depending on the R elements. All $R_{0.2}Fe_{0.8}$ alloys show an Invar property. The forced volume magnetostriction at about Curie temperature shows the same R dependence of the mean magnetic moment \bar{m} . The local environmental effect on the Fe moment in the amorphous state is discussed in comparison with the corresponding crystalline R–Fe compounds.

1. Introduction

The local environment and molecular field strongly affect the magnetic moment of 3d transition metals (TMs). The instability of 3d magnetic moments due to these effects is found prominently in rare-earth (R)–TM compounds. In the RCO_2 compounds the molecular field from an R sublattice yields a magnetic polarisation on Co and gives rise to a spontaneous volume magnetostriction (Beille *et al* 1976). In RMn_2 the Mn magnetic moment appears when the lattice constant of the compound is more than critical value (Shiga 1988). In the R_2Fe_{17} and R_6Fe_{23} compounds the magnetovolume effect is attributed to either the latent antiferromagnetic coupling between Fe atoms in a dumb-bell site (Givord and Lemaire 1974) or the weak ferromagnetism (Inoue and Shimizu 1985).

The magnetic properties of amorphous R–Fe alloys have been studied using thin films (Heiman *et al* 1975, Taylor *et al* 1978) and rapidly quenched alloys (Buschow and van der Kraan 1981). Although it was suggested that the Fe moments in the amorphous R–Fe alloys are influenced by the local environment and the molecular field, their dependence on the R element and on the R concentration is still open to question. Furthermore, only a little experimental work has so far been carried out on the magnetovolume effect of amorphous R–Fe alloys (Buschow *et al* 1977).

Ishio *et al* (1986, 1988) investigated the magnetic properties and the magnetovolume effect of amorphous $Y_{1-x}Fe_x$ ($0.4 \leq x \leq 0.8$) alloys. $Y_{0.2}Fe_{0.8}$ exhibits the largest magnetovolume effect of the amorphous Y–Fe alloys as well as a wide distribution of hyperfine field. In this paper the magnetisation and magnetovolume effect of amorphous $R_{0.2}Fe_{0.8}$ ($R \equiv Y, Pr, Nd, Sm, Gd, Dy, Tb$ and Er) alloys are described. The contribution of R elements to both the Fe moment and the magnetovolume effect is discussed.

2. Experimental details

Amorphous $R_{1-x}Fe_x$ samples ($R \equiv Pr, Nd, Sm, Gd, Tb, Dy, Er$ and Y ; $x = 0.4–0.8$) were prepared by melt spinning in an Ar atmosphere. Molten alloys were ejected under

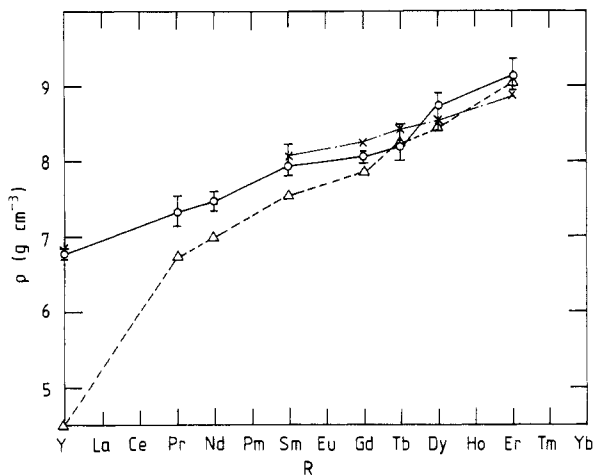


Figure 1. Density ρ for amorphous $R_{0.2}Fe_{0.8}$ alloys at room temperature (\circ). The ρ -values for crystalline R_6Fe_{23} compounds (\times) (Herbst and Croat 1984) and for R elements (Δ) (Spedding *et al* 1956) are also shown.

an Ar pressure of 0.5 kgf cm^{-2} through a quartz nozzle (0.15–0.2 mm in diameter) onto the edge of a Cu disc, rotating with a surface velocity of 42 m s^{-1} . The amorphous samples prepared were about 1 mm wide, $10 \mu\text{m}$ thick and less than 10 cm in length. The amorphous structure was confirmed by x-ray Debye–Scherrer analysis. The density of amorphous alloys was measured by comparing the specimen weight in air and toluene at room temperature according to the Archimedean principle.

Magnetisation was measured using a magnetic balance and a vibrating magnetometer in magnetic fields of up to 16 kOe, on heating samples up to 700 K after cooling to 4.2 K in a magnetic field of 15 kOe. The Curie temperature T_C was determined by Arrott plots.

Thermal expansion was measured with a differential transformer from 77 to about 500 K. Creep took place during thermal expansion measurements above about 500 K. The magnetostriction λ was measured by a three-terminal capacitance method (Ishio and Sato 1988) in magnetic fields H up to 20 kOe. Magnetostriction measurements were made on heating samples, after cooling to 4.2 K without a magnetic field. The forced volume magnetostriction $\partial\omega/\partial H$ was defined by the gradient of a volume expansion $(\Delta l/l)_{\parallel} + 2(\Delta l/l)_{\perp}$ between 10 and 20 kOe with respect to H .

3. Experimental results

3.1. Density

The density ρ of amorphous $R_{0.2}Fe_{0.8}$ alloys at room temperature is shown in figure 1. As the R atomic number rises, the value of ρ increases from about 7.3 g cm^{-3} for $R \equiv \text{Pr}$ to about 9.1 g cm^{-3} for $R \equiv \text{Er}$. The values of ρ (Herbst and Croat 1984) for R_6Fe_{23} compounds which have very similar chemical compositions ($x = 0.793$) to $R_{0.2}Fe_{0.8}$ are also shown in the figure. It should be noted that the ρ -values for both states agree within experimental error and show the same trend with respect to R.

3.2. Magnetic moment

The temperature dependence of magnetisation σ for amorphous $R_{0.2}Fe_{0.8}$ alloys is shown in figure 2. While σ for $R \equiv \text{Pr}$, Nd and Y decreases monotonically with heating, σ for

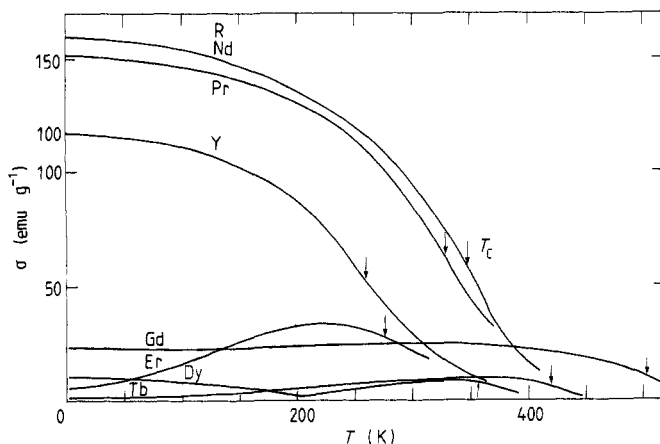


Figure 2. Temperature dependence of the magnetisation for amorphous $R_{0.2}Fe_{0.8}$ alloys.

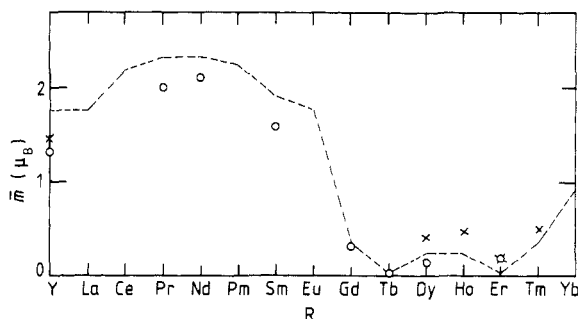


Figure 3. Mean magnetic moment \bar{m} per atom for amorphous $R_{0.2}Fe_{0.8}$ alloys (O). The \bar{m} -values for R_6Fe_{23} compounds (x) (Herbst and Croat 1984) are also shown. See the text for the meaning of the broken curve.

R = Tb, Dy and Er exhibits ferrimagnetic behaviour. The mean magnetic moment \bar{m} per atom at 4.2 K derived from σ -values is plotted against R in figure 3. The Fe moment is considered to be parallel to the magnetic moments of light rare earths (LRS) and antiparallel to those of heavy rare earths (HRS). The \bar{m} -values calculated by assuming a magnetic moment of $2.2\mu_B$ for Fe and the magnetic moment gJ of R^{3+} ions for R elements (also by assuming collinear alignment of magnetic moments) are indicated by the broken line in the figure. The \bar{m} -values for LRS are somewhat smaller than the calculated values, whereas the \bar{m} -values for HRS are roughly on the line. The \bar{m} -values for R_6Fe_{23} (Herbst and Croat 1984) are slightly larger than \bar{m} for amorphous alloys.

The value of the Fe moment evaluated by subtracting R moments gJ from \bar{m} is plotted in figure 4. The Fe moment m_{Fe} ranges from $1.6\mu_B$ for Y to $2.3\mu_B$ for Er. $m_{Fe} = 1.8\mu_B$ for $Nd_{0.2}Fe_{0.8}$ is smaller than the value of $2.0\mu_B$ deduced from Mössbauer spectroscopy (Nagayama *et al* 1988), whereas the value for Y-Fe agrees well with the Mössbauer data (Ishio *et al* 1988). The inconsistency of m_{Fe} in $Nd_{0.2}Fe_{0.8}$ may be attributed to a reduction in the Nd moment from its free-ion value because of crystal-field interactions, which has been frequently observed in Sm-TM and Nd-TM compounds (Buschow *et al* 1973, Herbst *et al* 1982). With due regard to this situation, one may say that, although m_{Fe} is recovered to some extent by replacing Y by other R elements, the dependence of m_{Fe} on R (R = Pr-Er) is not outstanding.

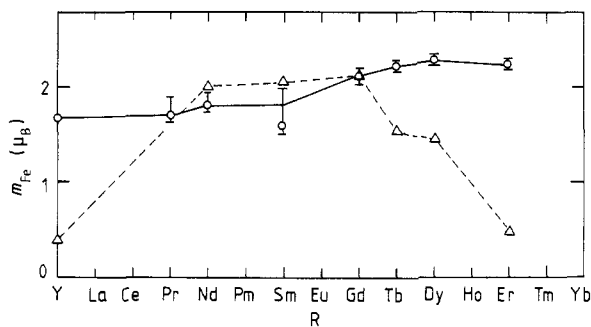


Figure 4. Fe magnetic moment m_{Fe} for amorphous $R_{0.2}Fe_{0.8}$ alloys (○). The m_{Fe} -values for $R_{0.6}Fe_{0.4}$ alloys (△) (Buschow and van der Kraan 1981, Nagayama *et al* 1988, Ishio *et al* 1988) are also shown.

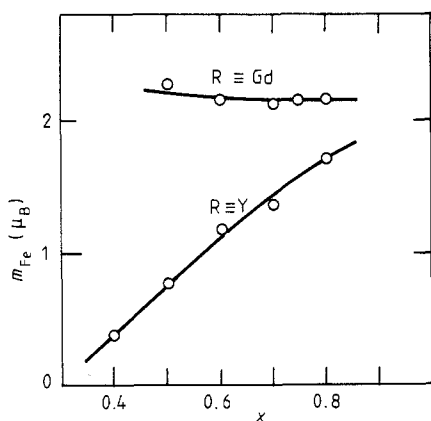


Figure 5. Fe magnetic moment m_{Fe} for amorphous $Y_{1-x}Fe_x$ and $Gd_{1-x}Fe_x$ alloys.

The composition dependence of m_{Fe} for Y-Fe and Gd-Fe is shown in figure 5. In contrast with a remarkable decrease in m_{Fe} for Y-Fe with decreasing x , the m_{Fe} of Gd-Fe does not show an appreciable change in the composition range measured.

3.3. Curie temperature

The Curie temperature T_C of $R_{0.2}Fe_{0.8}$ alloys is shown in figure 6. The R dependence of T_C is quite similar to those for R-Fe compounds; namely the highest T_C is found for $R \equiv Gd$ and the lowest for $R \equiv Y$. Such a general trend has been explained by the molecular-field effect caused by R-Fe exchange interactions, whose contribution to T_C is proportional to the de Gennes factor (Heiman *et al* 1975, Belorizky *et al* 1987) and reaches a maximum at Gd.

3.4. Forced volume magnetostriction

The temperature dependence of the forced volume magnetostriction $\partial\omega/\partial H$ of amorphous $R_{0.2}Fe_{0.8}$ alloys is shown in figure 7. The $\partial\omega/\partial H$ -values for $R \equiv Pr$, Nd and Er increase monotonically up to T_C and then decrease with further increasing temperature. The R dependences of $\partial\omega/\partial H$ taken at $T = 77$ K and at about T_C are shown in figure 8. Here the values of $\partial\omega/\partial H$ for $R \equiv Tb$ and Dy at 77 K were evaluated by extrapolation from high temperatures as indicated by the broken lines in figure 7, because a large

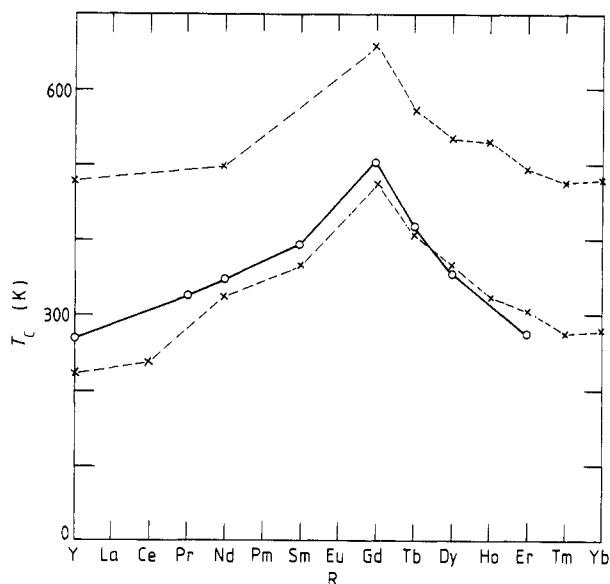


Figure 6. Curie temperatures for amorphous $R_{0.2}Fe_{0.8}$ alloys (\circ), together with those for crystalline R_6Fe_{23} (\bullet) and R_2Fe_{17} (\times) compounds (Buschow 1977).

coercive force of more than 10 kOe appeared below 200–230 K owing to magnetic compensation, causing an outward decrease in $\partial\omega/\partial H$ below this temperature (Ishio 1988). At about T_C , $\partial\omega/\partial H$ -values for LRS are much larger than those of HRS. Note that the R dependence of $\partial\omega/\partial H$ at T_C is quite similar to that of the mean magnetic moment \bar{m} (see figure 3). The value of $\partial\omega/\partial H$ at 77 K has a maximum of $67 \times 10^{-10} \text{ Oe}^{-1}$ for $R \equiv Y$.

The $\partial\omega/\partial H$ -values for amorphous $R_{1-x}Fe_x$ ($R \equiv \text{Pr, Nd, Gd, Dy and Y}$) alloys decreased rapidly with increasing x , and $\partial\omega/\partial H$ for $R \equiv \text{Gd and Dy}$ even became negative at $x = 0.4\text{--}0.7$. The details have been published elsewhere (Ishio 1988).

3.5. Thermal expansion

The thermal expansion curves of $R_{0.2}Fe_{0.8}$ alloys are shown in figure 9. All $R_{0.8}Fe_{0.2}$ alloys show an Invar property below T_C , suggesting that the R dependence of the spontaneous volume magnetostriction is not notable. The thermal expansion of $R_{1-x}Fe_x$ ($R \equiv \text{Nd and Gd}$; $0.4 \leq x \leq 0.8$) is shown in figures 10 and 11. The Invar property for Gd-Fe and Nd-Fe disappears with decreasing x .

4. Discussion

4.1. R dependence of both Invar effect and Fe magnetic moment in $R_{0.2}Fe_{0.8}$ alloy

The electronic band structure of the crystalline Y_6Fe_{23} compound was calculated by Inoue and Shimizu (1985), who suggested a weak ferromagnetic character and Invar effect. The Invar property as well as the large forced volume magnetostriction observed in the amorphous $Y_{0.2}Fe_{0.8}$ alloy seem to be explained by this model by assuming similarity of the atomic short-range order (and hence of the resultant electronic structure) in the amorphous and the crystalline states. The R elements in $R_{0.2}Fe_{0.8}$ alloys may

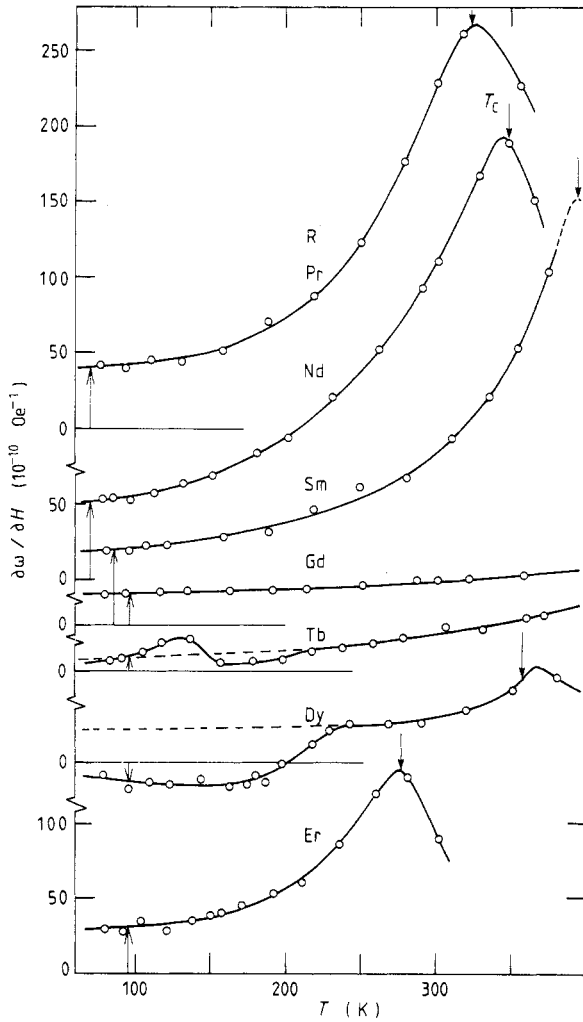


Figure 7. Temperature dependence of the forced volume magnetostriction of amorphous $R_{0.2}Fe_{0.8}$ alloys.

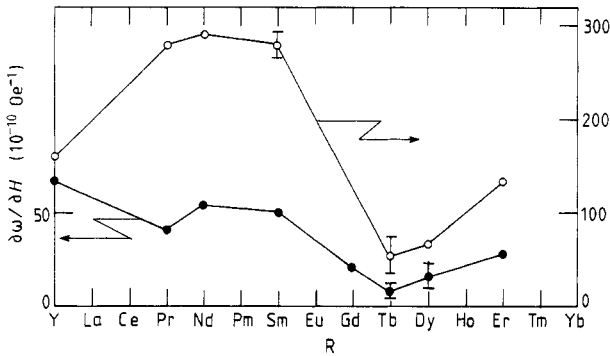


Figure 8. R dependence of the forced volume magnetostriction $\partial\omega/\partial H$ at 77 K (●) and at T_c (○).

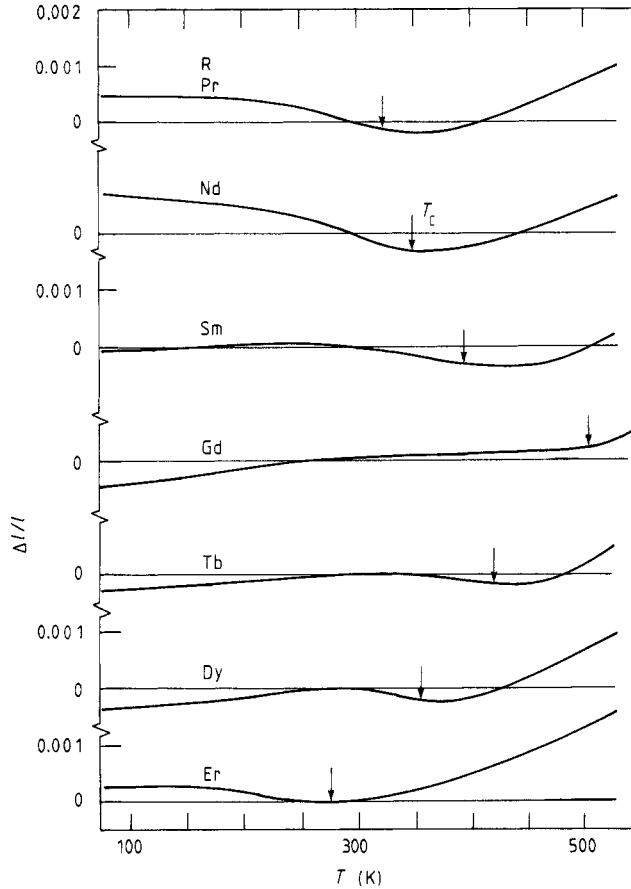


Figure 9. Thermal expansion curves for amorphous $R_{0.2}Fe_{0.8}$ alloys.

add the molecular field acting on Fe without a significant change in the band structure from that of $Y_{0.2}Fe_{0.8}$. One can therefore expect an enhancement of both the Fe moment and the magnetovolume effect, particularly for $R \equiv Gd$ and Tb which have a strong molecular field. Contrary to this expectation, however, the spontaneous volume magnetostriction is almost the same for all $R_{0.2}Fe_{0.8}$ and the enhancement of Fe moment for $R \equiv Gd$ and Tb is not clear. This is in contrast with crystalline RCo_2 , in which Gd and Tb sublattices induce a magnetic moment of about $1\mu_B$ on Co and yield a spontaneous volume magnetostriction (Beille *et al* 1976).

According to Kemeny *et al* (1983), Fe is non-magnetic when the Fe molar volume V_{Fe} is less than 6.6 cm^3 . The Fe moment increases rapidly at around $V_{Fe} = 6.7\text{--}7.1$ and saturates to about $2.2\mu_B$ at $V_{Fe} > 7.2$. A strong magnetovolume effect occurs reasonably in the critical region. The value of V_{Fe} in the amorphous $R_{0.2}Fe_{0.8}$ alloys can be estimated from the density ρ by assuming that the R molar volume V_R is the same as V_R in the pure metal and that the density ρ of the amorphous state is given by $(0.2M_R + 0.8M_{Fe}) / (0.2V_R + 0.8V_{Fe})$, where M_R and M_{Fe} are the atomic weights for R and Fe. The value of V_{Fe} thus obtained decreases from about 7.24 for $R \equiv Pr$ to 6.1 for $R \equiv Er$. Despite the variation in V_{Fe} through the critical region, m_{Fe} for $R_{0.2}Fe_{0.8}$ does not change appreciably. This suggests that the effect of R elements through the molar volume (or lattice constant) is seemingly negligible in this series, at least from the macroscopic point of view.

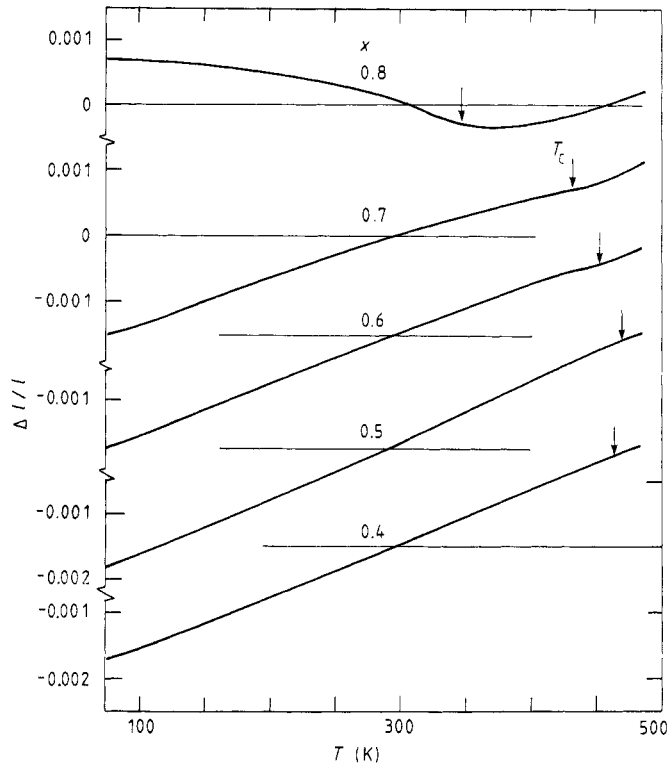


Figure 10. Thermal expansion curves for amorphous $\text{Nd}_{1-x}\text{Fe}_x$ alloys.

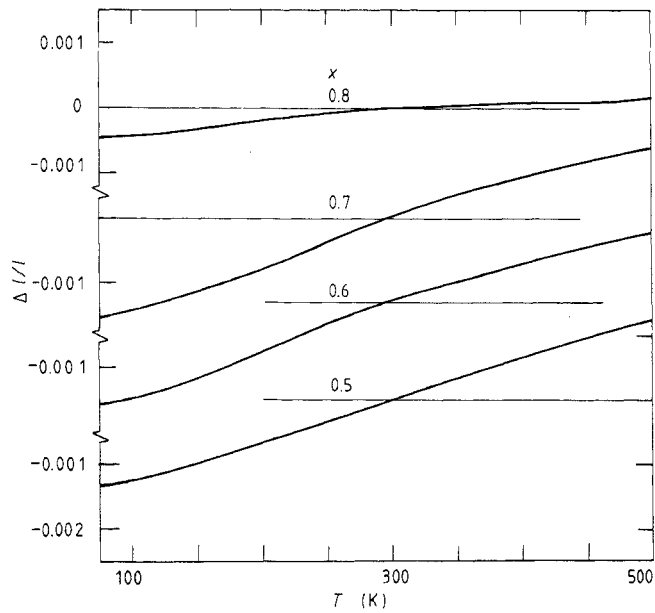


Figure 11. Thermal expansion curves for amorphous $\text{Gd}_{1-x}\text{Fe}_x$ alloys.

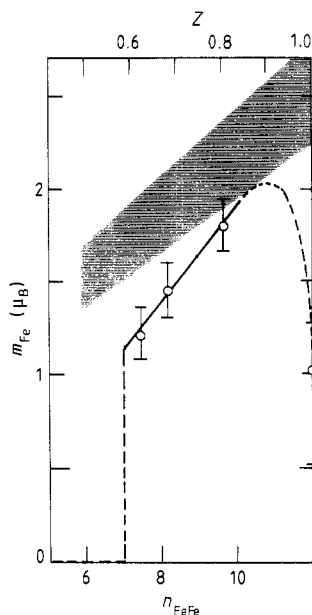


Figure 12. Fe magnetic moment as a function of the number n_{FeFe} of Fe nearest neighbours in the amorphous Y-Fe alloys (Ishio *et al* 1988). The Fe moment in crystalline R-Fe compounds is shown in the shaded area (see text).

From the above experimental results and considerations, it is concluded that the contribution of R elements to both the Fe moment and the spontaneous volume magnetostriction through the molecular field or the molar volume is very small in amorphous $\text{R}_{0.2}\text{Fe}_{0.8}$ alloys. The magnetic instability of Fe was found in Y, Zr-Fe alloys containing more than 80% Fe (Ishio *et al* 1988). The Fe moment begins to reduce its magnitude when the number n_{FeFe} of Fe nearest neighbours is 10 or more and becomes about $1\mu_{\text{B}}$ for $n_{\text{FeFe}} = 12$ (figure 12). It is likely that such an instability in the Fe moment also exists in $\text{R}_{0.2}\text{Fe}_{0.8}$ (R \equiv Pr-Er) amorphous alloys, yielding an Invar effect. In other words, R elements as well as Y and Zr stabilise the amorphous structure, but they contribute little to the instability of the Fe moment in this composition range. Fe moments less than $1\mu_{\text{B}}$ tend to couple antiferromagnetically with other moments (Kakehashi 1988a). These Fe components give rise to a decrease in T_{C} for the amorphous $\text{R}_{0.2}\text{Fe}_{0.8}$ alloys in comparison with that for R_6Fe_{23} and are responsible for spin-glass behaviour for alloys containing more than 90% Fe (Saito *et al* 1986, Kakehashi 1989).

4.2. Composition dependence of m_{Fe} in $\text{R}_{1-x}\text{Fe}_x$ alloys

The Fe moment of Y-Fe increases with increasing x , while that of Gd-Fe does not show an appreciable change with respect to x (see figure 5). To confirm this contrast between Y and Gd, m_{Fe} for $\text{R}_{0.6}\text{Fe}_{0.4}$ alloys measured by Mössbauer spectroscopy[†] (Buschow and van der Kraan 1981, Nagayama *et al* 1988) is plotted in figure 4. The m_{Fe} -values for Er, Dy and Tb show a similar trend to m_{Fe} for Y when studied as a function of the Fe concentration x . The local environmental effect was proposed by Chappert *et al* (1981) to explain the composition dependence of m_{Fe} in amorphous $\text{Y}_{1-x}\text{Fe}_x$ thin films. Ishio *et al* (1988) obtained the dependence of m_{Fe} on the number n_{FeFe} of Fe neighbours, in rapidly quenched amorphous Y-Fe alloys by assuming a coordination of 12 (see figure

[†] Amorphous $\text{R}_{1-x}\text{Fe}_x$ alloys except for R \equiv Gd and Y show a huge coercive force at $x < 0.7$ because of random magnetic anisotropy, so that magnetic fields far greater than 20 kOe are necessary to saturate magnetisation. Therefore, Mössbauer data for $x = 0.4$ are compared here with the results for $x = 0.8$.

12). The local environmental effect in crystalline compounds was studied by Gubbens *et al* (1978) and Pszczola *et al* (1985). The dependence of m_{Fe} on $Z = n_{\text{FeFe}}/(n_{\text{FeFe}} + n_{\text{FeR}})$ for R-Fe compounds (R \equiv Y, Ce, Nd, Gd, Tb, Dy, Er and Th) is shown as a shaded area in figure 12. This $m_{\text{Fe}}-Z$ relation is analogous to the $m_{\text{Fe}}-n_{\text{FeFe}}$ relation for the amorphous Fe-Y alloys. The decrease in m_{Fe} in compounds is understood as the gradual decrease in the density of states near the Fermi energy with decreasing x (Inoue and Shimizu 1985, Malozemoff *et al* 1984). Such a modification of the band structure seems to explain the decrease in m_{Fe} for R \equiv Dy and Er as well as for R \equiv Y, although one must take account of the distribution of the atomic short-range order (and hence of the magnetic moment) in the amorphous state.

In the crystalline case, R \equiv Gd and Y show almost the same local environmental effect. It is unlikely that the $m_{\text{Fe}}-Z$ (or $m_{\text{Fe}}-n_{\text{FeFe}}$) relation for the amorphous state differs entirely from that of the crystalline state. The difference between the $m_{\text{Fe}}-x$ relations for Gd and Y might be associated with the structural change in the amorphous state (Matsuura *et al* 1987, Buschow and van der Kraan 1981). A detailed structural study is necessary before further discussion of this point.

4.3. Forced volume magnetostriction

In alloys containing more than two types of magnetic element, the magnetic order parameter is the total magnetisation M ($M = M_{\text{d}} + M_{\text{R}}$ where M_{d} is the magnetisation of the 3d TM and M_{R} is the magnetisation of the R metal) (Shimizu 1982). Therefore, the forced volume magnetostriction is given as a function of the total magnetisation phenomenologically:

$$\partial\omega/\partial H = \kappa C_M M \chi_M \quad (1)$$

where κ is the compressibility. C_M and χ_M are the magnetovolume coupling constant and the susceptibility for total magnetisation, respectively. The resemblance of the R dependence of $\partial\omega/\partial H$ at T_C to that of \bar{m} (in figure 3) indicates that the forced volume magnetostriction near T_C is seemingly determined by the total magnetisation.

On the contrary, the Fe and R moments align collinearly at low temperatures, being parallel (or antiparallel) to magnetic fields. The Fe moment has a larger high-field susceptibility than the moments of R elements because of the good localisation of 4f electrons. Therefore χ_M in equation (1) may be approximated by χ_{Fe} and hence

$$\partial\omega/\partial H = \kappa C_M M (\pm\chi_{\text{Fe}}). \quad (2)$$

The sign in parentheses corresponds to whether the Fe moment is parallel or antiparallel to applied magnetic fields. Although a temperature of 77 K is not necessarily low enough to suppress a spin-wave contribution to susceptibility, the value of $\partial\omega/\partial H$ at 77 K reflects the high-field susceptibility of Fe moments more than around T_C . The smaller $\partial\omega/\partial H$ for R \equiv Pr-Er ($(8-54) \times 10^{-10} \text{ Oe}^{-1}$) than that for Y ($67 \times 10^{-10} \text{ Oe}^{-1}$) may indicate a reduction in χ_{Fe} due to the replacement of Y by other R elements, which is consistent with somewhat larger m_{Fe} for $\text{R}_{0.2}\text{Fe}_{0.8}$ (R \equiv Pr-Er) than for $\text{Y}_{0.2}\text{Fe}_{0.8}$.

5. Summary and conclusion

The Fe magnetic moment in amorphous $\text{R}_{0.2}\text{Fe}_{0.8}$ is about $1.6\mu_{\text{B}}$ for R \equiv Y and recovers to some extent by replacing Y with other R elements. However, the dependence of m_{Fe}

on R is not outstanding. All $R_{0.2}Fe_{0.8}$ alloys show the Invar property, which is attributable to the nature of Fe atoms surrounded by more than about ten Fe nearest neighbours in the amorphous state.

The decrease in the Fe moment with decreasing x in $R_{1-x}Fe_x$ alloys (R = Er, Tb and Y) is explained by the local environmental effect, which is almost the same as that in the crystalline case.

The forced volume magnetostriction of $R_{0.2}Fe_{0.8}$ alloys at T_C was determined from the total magnetisation, showing the same R dependence as the mean magnetic moment \bar{m} . The forced volume magnetostriction at 77 K for R \equiv Pr-Er is smaller than that for R \equiv Y, which seems consistent with the recovery of m_{Fe} obtained by replacing Y by other R elements.

Mössbauer measurements for $R_{0.2}Fe_{0.8}$ alloys are now in progress; this is expected to give further information about the local environmental effects on Fe moment and a clue to the relation between the magnetovolume effect and the instability of the Fe moment.

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