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Pressure effect on the magnetic properties of amorphous $La(Fe_xAl_{1-x})_{13}$ alloys composed of icosahedral clusters

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Abstract. The pressure effects on the magnetic properties have been investigated for amorphous $La(Fe_xAl_{1-x})_{13}$ ($0.80 \leqslant x \leqslant 0.95$) alloys prepared by high-rate DC sputtering. The pressure derivatives of magnetization $\partial M/\partial P$ and the Curie temperature $\partial T_C/\partial P$ are significantly large. The concentration dependences of $\partial M/\partial P$ and $\partial T_C/\partial P$ are similar to those of various magnetovolume and magnetoelastic properties such as the spontaneous volume magnetostriction ω_S, forced volume magnetostriction $\partial\omega/\partial H$ and compressibility κ.

The pressure derivative of the Curie temperature $\partial T_C/\partial P$ is also large, and the linear relation of $\partial \ln T_C/\partial P$ versus T_C plot is accounted for by taking inhomogeneity into consideration. The linear plot of $T_C/\partial P$ versus $T_C^{-1/3}$ is explained in terms of spin fluctuations. In connection with the peculiar amorphous structure composed of icosahedral clusters, both $\partial T_C/\partial P$ and $\partial \ln T_C/\partial P$ are different from those of amorphous Fe-based binary alloys. The value of $\partial T_C/\partial P$ determined from the pressure dependence of the spin-wave stiffness constant D is reasonable. The spin freezing temperature T_f is increased with increasing pressure, consistent with the theory concerned with itinerant electron spin-glass systems.

1. Introduction

Many data on Invar anomalies in various amorphous Fe-based alloys are available (Fukamichi 1983, Fukamichi *et al* 1989). A large compressibility of Fe–B amorphous alloy has been confirmed by x-ray diffraction under pressure (Tomizuka *et al* 1984). A marked pressure effect on the electrical resistivity of amorphous Zr–Fe alloys has been correlated to their large compressibility (Shirakawa *et al* 1983). The pressure effect on the Curie temperature of Zr–Fe, Hf–Fe, Sc–Fe, Nd–Fe–B and La–Fe amorphous alloys is prominent (Fukamichi *et al* 1983, Fukamichi *et al* 1985, Fukamichi *et al* 1986, Goto *et al* 1988). Experimental and theoretical results of the pressure effect on the Curie temperature of weak ferromagnets in both crystalline and amorphous alloys have recently been discussed in detail (Wagner and Wohlfarth 1981). Furthermore, a general relation between the pressure effect on the Curie temperature and the magnetization has been derived (Inoue and Shimizu 1982). More recently, the pressure effect on the magnetic properties of amorphous Fe has been investigated on the basis of a finite-temperature theory of local-environment effects (Kakehashi 1993). The pressure effect on the magnetization is expected to give valuable

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information. However, the study for amorphous alloys is not so active as that for crystalline alloys (Nakamura *et al* 1971).

Crystalline $La(Fe_xAl_{1-x})_{13}$ compounds with a cubic $NaZn_{13}$ type of structure can be stabilized in the concentration range $0.46 \leqslant x \leqslant 0.92$ (Palstra *et al* 1984). These compounds have icosahedral clusters composed of Fe and Al atoms. The nearest-neighbour configuration in the clusters is similar to that in γ-Fe (Palstra *et al* 1985). It is interesting to note that the icosahedral clusters are often correlated to the structures of amorphous and quasicrystalline alloys (Kofalt *et al* 1986). The existence of the icosahedral clusters in amorphous $La(Fe_xAl_{1-x})_{13}$ ($0.80 \leqslant x \leqslant 0.95$) alloys has been confirmed by x-ray diffraction (Matsubara *et al* 1992) and the Mössbauer effect (Chiang *et al* 1994). The interatomic atomic distances of the Fe–Fe nearest neighbour in both crystalline and amorphous states (Ludorf *et al* 1989, Matsubara *et al* 1992, Chiang *et al* 1994) are very close to the critical distance of the competition between ferromagnetic and antiferromagnetic interactions (Wassermann 1990).

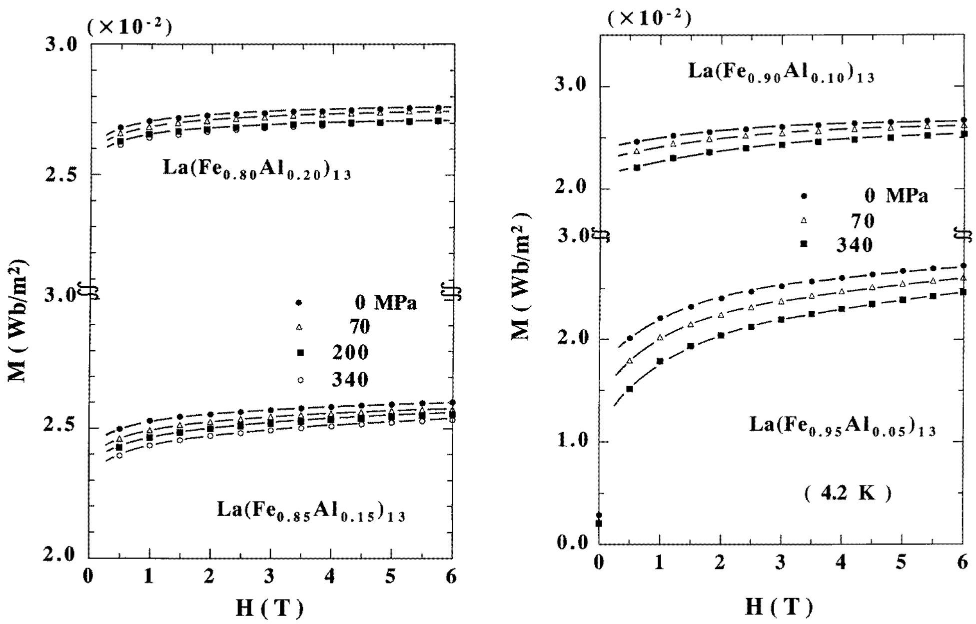
Under such circumstances, the magnetic state becomes unstable, and various anomalous magnetic and elastic properties emerge. The $La(Fe_{1-x}Al_x)_{13}$ alloys in the crystalline and amorphous states exhibit a pronounced thermal expansion anomaly (Palstra *et al* 1985, Chiang *et al* 1992). The pressure effect on magnetic properties of a crystalline $La(Fe_{0.88}Al_{0.12})_{13}$ has been investigated. Instability of Fe magnetic moment is attributed to γ–Fe-like environment and the critical distance of the Fe–Fe nearest-neighbour distance (Ludorf *et al* 1989). Large anomalies in the thermal expansion and elastic properties of the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys have been related to their large magnetovolume effect (Chiang *et al* 1992). The compressibility κ obtained from the data on Brillouin scattering of the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys is large (Yoshihara *et al* 1994). Furthermore, the concentration dependence of the giant forced volume magnetostriction has been demonstrated (Fukamichi *et al* 1995). Large pressure effects on the Curie and Néel temperatures of crystalline $La(Fe_{0.88-x}Co_xAl_{0.12})_{13}$ alloys have been discussed in the context of the instability of the ferromagnetic state (Medvedeva *et al* 1992).

Judging from the above, marked pressure effects on the magnetic properties are expected. In the present paper, therefore, the pressure effects on the magnetization, the Curie temperature, the spin-wave stiffness constant and the spin freezing temperature of the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys prepared by high-rate DC sputtering have been investigated. These data are compared with those for amorphous Fe-based binary alloys and discussed in connection with large magnetovolume effects and magnetic instability.

2. Experimental details

The alloy targets for sputtering were made by arc melting in an argon gas atmosphere. Several kinds of amorphous $La(Fe_xAl_{1-x})_{13}$($0.80 \leqslant x \leqslant 0.95$) samples about 0.3 mm thick were prepared by high-rate DC sputtering on a water-cooled Cu substrate. The Cu substrate was dissolved in a dilute chromic acid kept at about 350 K. Their amorphous state was confirmed by x-ray diffraction. More detailed procedures have been described elsewhere (Chiang *et al* 1991).

Conventional precipitation hardening Be–Cu alloys contain several per cent ferromagnetic Co precipitates and impede the precise magnetic measurements. Therefore, clamp pressure cells made of a precipitation hardening Ti–Cu alloy were used for the magnetic measurements. The magnetization curves up to 6 T at 4.2 K and the thermomagnetization curves at 1 T were obtained by using an extraction-type magnetometer with a superconducting magnet. The pressure effect on the magnetization was obtained at 4.2 K and the pressure effect on the Curie temperature was determined from the thermomagnetization curves measured at 1 T. The pressure effect on the magnetic cooling effect was measured with the same magnetometer from 4.2 to 300 K under different hydrostatic pressures. Fluorinate No 70 (C_6F_{14}) was used as a pressure transmitting fluid and the applied pressures were calibrated by the shift of the superconducting transition temperature of pure Pb.



(*a*) (*b*)

Figure 1. (*a*) Magnetization curves up to 6 T at 4.2 K under different pressures for the amorphous $La(Fe_{0.80}Al_{0.20})_{13}$ and $La(Fe_{0.85}Al_{0.15})_{13}$ alloys. (*b*) Magnetization curves up to 6 T at 4.2 K under different pressures for the amorphous $La(Fe_{0.90}Al_{0.10})_{13}$ and $La(Fe_{0.95}Al_{0.05})_{13}$ alloys.

3. Results and discussion

Figure 1 shows the magnetization curves up to 6 T under different pressures for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys which exhibit a large thermal expansion anomaly (Chiang *et al* 1991). The magnetization decreases with increasing applying pressure, and the magnitude of decrease in the magnetization depends on the magnetic state. Re-entrant spin-glass behaviour in the amorphous alloys has been observed in the concentration range $0.85 < x \leqslant 0.95$ (Chiang *et al* 1991), although their crystalline counterparts are antiferromagnetic (Palstra *et al* 1985). Ferromagnetic states are observed below $x = 0.85$ in both the amorphous state (Chiang *et al* 1991) and the crystalline state (Palstra *et al* 1985). In more detail, therefore, the decrease in the magnetization of the ferromagnetic amorphous alloys with $x = 0.80$ and 0.85 is not remarkable, as seen from figure 1(*a*), whereas the decrease of the re-entrant spin-glass amorphous alloys with $x = 0.90$ and 0.95 is significant, as shown in figure 1(*b*).

The pressure dependence of magnetization measured at 4.2 K and 6 T is plotted in figure 2. The decrease in the magnetization becomes remarkable with increasing x. The

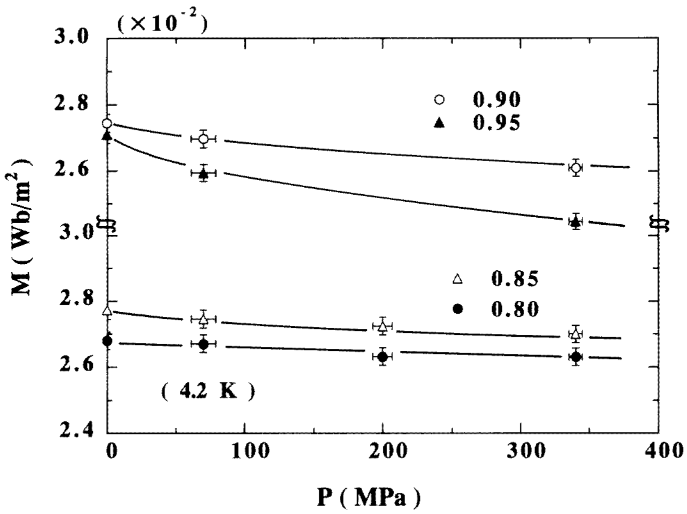


Figure 2. The pressure dependence of the magnetization measured at 4.2 K and 6 T for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys.

concentration dependence of the pressure derivative $\partial M/\partial P$ defined as the initial slope is given in figure 3. The magnitude of $\partial M/\partial P$ increases markedly in the re-entrant spin-glass concentration regime. We should notice that the spontaneous volume magnetostriction ω_s and the forced volume magnetostriction $\partial\omega/\partial H$ also drastically increase with increasing x (Fukamichi *et al* 1995). From Maxwell's relation for the free energy, the pressure effect on the magnetization is equivalent to $\partial\omega/\partial H$, namely $\partial\omega/\partial H = -\rho\partial M/\partial P$, where ρ is the mass density and H is the applied magnetic field. Therefore, the concentration dependence of the forced volume magnetostriction $\partial\omega/\partial H$ (Fukamichi *et al* 1995) is plotted in the same figure for comparison. As is expected, both $-\partial M/\partial P$ and $\partial\omega/\partial H$ show a very similar trend.

The magnetic properties of Fe are prominently governed by the environment such as the coordination number and the Fe–Fe interatomic distance. Various theoretical and experimental results show that the dramatic change in the magnetism from the antiferromagnetic to the ferromagnetic state occurs in the vicinity of 0.25 nm for the Fe–Fe interatomic distance in γ-Fe (Wassermann 1990). In fact, the average magnetic hyperfine field of crystalline Fe–Ni alloys shows a marked decrease below 0.25 nm (Abd-Elmeguid *et al* 1988). The pressure effect on the Mössbauer effect in a crystalline $La(Fe_{0.88}Al_{0.12})_{13}$ compound has been investigated and it has been pointed out that an abrupt change in the average magnetic hyperfine field occurs at the critical pressure of about 4.5 GPa, corresponding to the Fe–Fe interatomic distance of about 0.253 nm (Ludorf *et al* 1989). In the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys, the icosahedral clusters are composed of Fe and Al atoms and the coordination number of Fe depends on x, but the Fe–Fe nearest-neighbour interatomic distance is scarcely changed by x, being about 0.255 nm (Chiang *et al* 1994). With increase in the number of nearest-neighbour Fe, therefore, the ferromagnetism becomes unstable and hence the re-entrant spin-glass state emerges, resulting in the remarkable pressure effect mentioned above.

In figure 4, the temperature dependence of magnetization $M(T)$ measured at 1 T for the ferromagnetic amorphous alloys with $x = 0.80$ and 0.85 exhibits a linear relation between

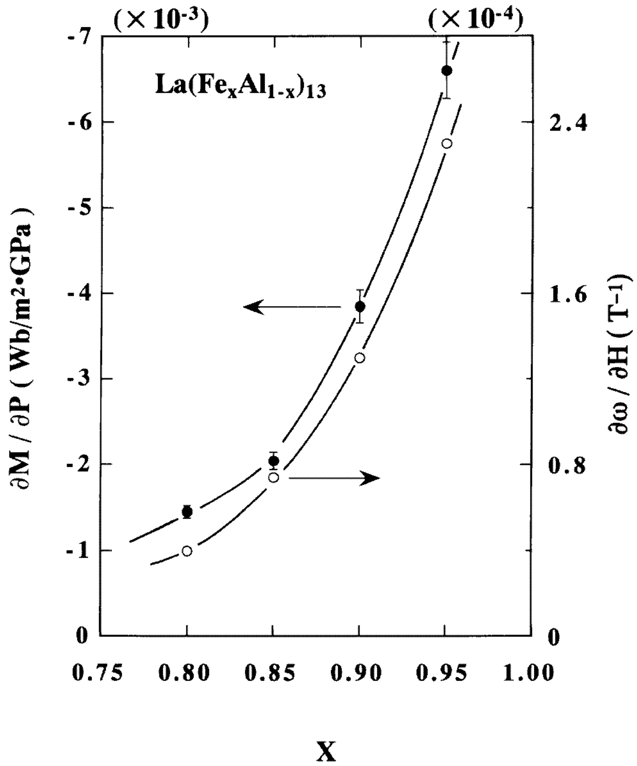


Figure 3. The concentration dependence of the pressure derivative of magnetization, $\partial M/\partial P$, for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys, together with that of the forced volume magnetostriction (Fukamichi *et al* 1995).

M and $T^{3/2}$ over a wide temperature range, which is dominated by the spin-wave excitation given by the following Bloch formula:

$$M(T) = M_0(1 - BT^{3/2} \ldots) \tag{1}$$

with

$$B = 2.612 g\mu_B/M(0)(k_B/4\pi D)^{3/2}$$

where D is the spin-wave stiffness constant and k_B the Boltzmann constant. The obtained D values under 0 Pa are 51.5 for $x = 0.80$ and 48.5 meV Å^2 for $x = 0.85$. These values are much smaller than those of amorphous Fe–metalloid alloys (Kazama *et al* 1978), which exhibit similar Invar effects (Fukamichi 1983), suggesting that the ferromagnetic state of the present amorphous alloys is more unstable. The spin-wave stiffness constant of the amorphous alloy with $x = 0.80$ has been determined by Brillouin scattering (Yoshihara *et al* 1994), but the value is much larger than the present result. In a similar manner as above, the values obtained by neutron inelastic scattering for Invar-type alloys are almost twice as large as those derived from low-temperature magnetization. The reasons for such a discrepancy are not fully understood yet. An additional low-lying magnetic excitation with a quadratic dispersion has been proposed as a possible origin (Ishikawa *et al* 1979).

The pressure derivative of the spin-wave stiffness constant $\partial D/\partial P$ is obtained from the thermomagnetization curve as a function of pressure. These results will be used in

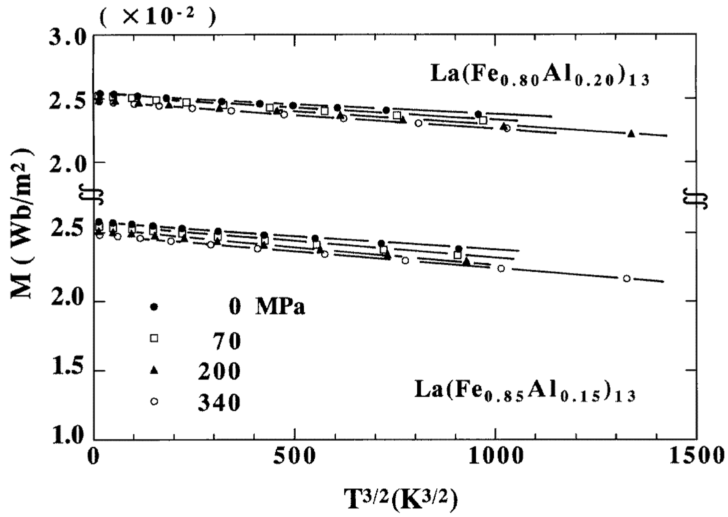


Figure 4. The temperature dependence of magnetization in the form M–$T^{3/2}$ under different pressures for the ferromagnetic amorphous $La(Fe_xAl_{1-x})_{13}$ alloys with $x = 0.80$ and 0.85.

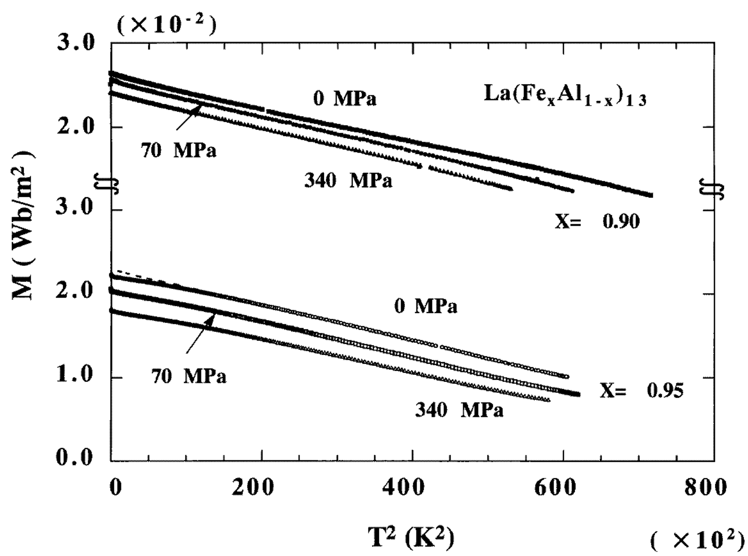


Figure 5. The temperature dependence of magnetization in the form M versus T^2 under different pressures for the re-entrant spin-glass amorphous $La(Fe_xAl_{1-x})_{13}$ alloys with $x = 0.90$ and 0.95.

the discussion in connection with figure 8. As shown in figure 5, on the other hand, the thermomagnetization of the re-entrant spin-glass amorphous alloys with $x = 0.90$ and 0.95 does not obey the $T^{3/2}$ law but obeys the T^2 law over a wide temperature range except for low-temperature ranges due to the Stoner-type excitation in weak ferromagnets (Nakai *et al* 1983). Note that the deviation from the straight line in the low-temperature ranges is due to the spin-glass state. With increasing pressure, the derivative becomes more significant in a similar manner as for the alloy with $x = 0.95$. Comparing the two figures under such pressures, it is considered that the spin-glass state is not created by reducing the Curie temperature for the alloys with $x = 0.80$ and 0.85 because no deviation is confirmed at low temperatures in figure 4 in contrast to the data in figure 5. The Curie temperature and the magnetization differ in the pressure coefficients, that is,

$$\partial \ln T_C/\partial P > \partial \ln M_S/\partial P. \tag{2}$$

This relation has been reported for crystalline $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys (Nakamura *et al* 1971), non-stoichiometric Ni_3Al compounds (Buis *et al* 1976) and crystalline Ni–Cr alloys (Tange *et al* 1981). Such behaviour would be explained by considering a large compressibility κ (Tange *et al* 1981) and additionally a positive Landau coefficient obtained from the Arrott plot (Edwards and Wohlfarth 1968). On the other hand, some weak ferromagnetic compounds (Franse 1979) and amorphous $Zr_{10}(Fe_xNi_{1-x})_{90}$ (Tange *et al* 1988a, b) exhibit an opposite relation. The band calculation for amorphous Fe mentioned before (Kakehashi 1993) also gives an opposite tendency, depending on input parameters. The present results fall within the latter case as shown in figure 6. Since the bulk modulus $B = \kappa^{-1}$ is obtained from the second volume derivative of the total energy, the notations $B\partial M/\partial P$ and $B\partial T_C/\partial P$ are more meaningful. The experimental value of κ has been obtained from the data on the Brillouin scattering (Fukamichi *et al* 1995). The concentration dependences of these two pressure derivatives calculated by using the experimental data are very similar to each other. With increasing x, these two pressure effects become remarkable in accordance with the ferromagnetic instability. Taking into account both thermal spin fluctuations and local magnetic moment fluctuations with respect to the structural disorder, the theoretical investigations also have revealed that the markedly large values of these two pressure effects are associated with the ferromagnetic instability (Kakehashi 1993).

From the M versus $T^{3/2}$ plots in figure 4, the spin-wave stiffness constant D is obtained for the ferromagnetic alloys with $x = 0.80$ and 0.85. For weak itinerant ferromagnets, the relation between and $\partial \ln M_S/\partial P$ has been obtained by expanding the free energy in a power series of magnetization (Wagner and Wohlfarth 1981). On the other hand, the following general relation for the pressure effects mentioned above has been given without using such an expansion of the free energy (Inoue and Shimizu 1982).

$$\partial \ln T_C/\partial P = 5\kappa/3 + (C_{eff}/\chi_{hf}T_C)\,\partial \ln M_S/\partial P \tag{3}$$

where C_{eff} is the effective Curie constant and χ_{hf} is the high-field susceptibility defined as $\partial M/\partial H$. In the weak ferromagnetic limit, that is, $(C_{eff}/\chi_{hf}T_C) = 1$, this equation is reduced to the expression obtained by expansion. The study of $(C_{eff}/\chi_{hf}T_C)$ for crystalline Fe–Ni and amorphous $Fe_{90-x}Ni_xZr_{10}$ alloys has been carried out and it has been pointed out that the $C_{eff}/\chi_{hf}T_C$ versus T_C plots for both alloy systems give unity around $T_C = 290$ K (Tange *et al* 1992). It is important to note that the Curie temperature of the present amorphous alloys is close to this temperature (Chiang *et al* 1991).

In itinerant electron ferromagnets for the weak limit, the pressure dependence of the spin-wave stiffness constant D is given by the following relation (Gustafson and Phillips 1969):

$$\partial \ln(D/a_\ell^2)/\partial P = \partial \ln T_C/\partial P \tag{4}$$

where a_ℓ is the lattice constant and would be converted to $\sqrt{2}$ times the Fe–Fe nearest-neighbour distance in the amorphous state in analogy with an fcc structure. The validity of this relation has been confirmed in crystalline Fe–Ni Invar alloys (Gustafson and Phillips 1969). The pressure derivative $\partial D/\partial P$ obtained from figure 4 is given in figure 7. The effect for the alloy with $x = 0.85$ is more remarkable compared with that of the alloy with $x = 0.80$ because of the increase in ferromagnetic instability. Figure 8 shows the concentration

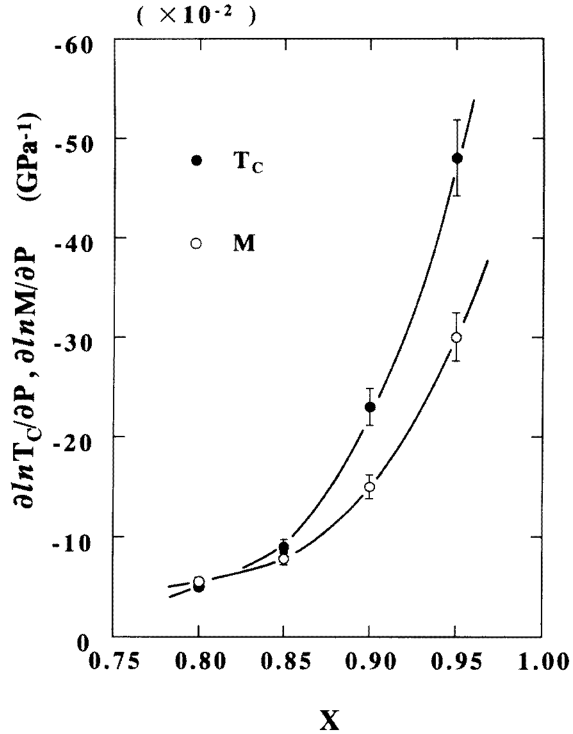


Figure 6. The concentration dependence of the pressure coefficients of the Curie temperature, $\partial \ln T_C/\partial P$, and the magnetization, $\partial \ln M/\partial P$, for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys.

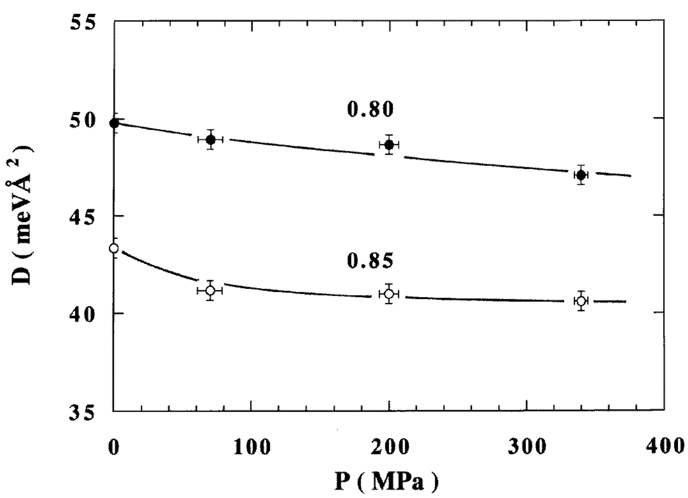


Figure 7. The pressure dependence of the spin-wave stiffness constant D for the ferromagnetic amorphous $La(Fe_xAl_{1-x})_{13}$ alloys with $x = 0.80$ and 0.85.

dependence of the pressure derivative of the Curie temperature $\partial T_C/\partial P$ determined from the thermomagnetization curves and equation (4) for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys,

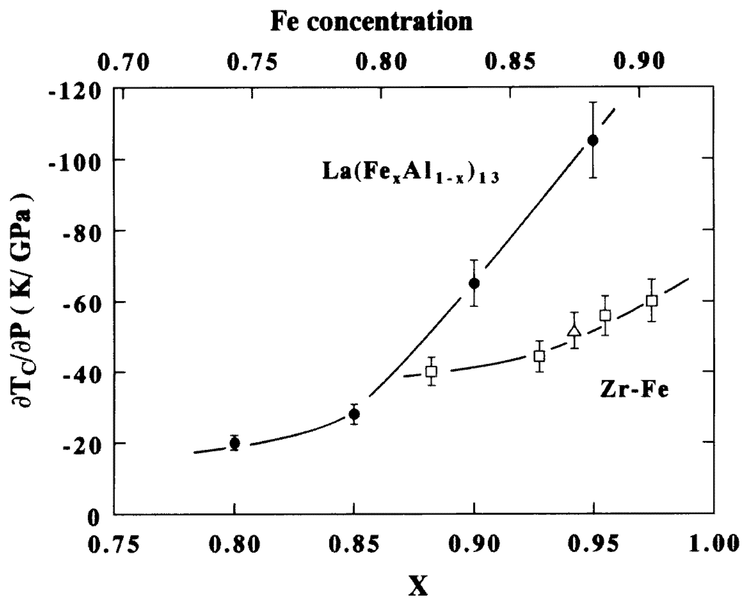
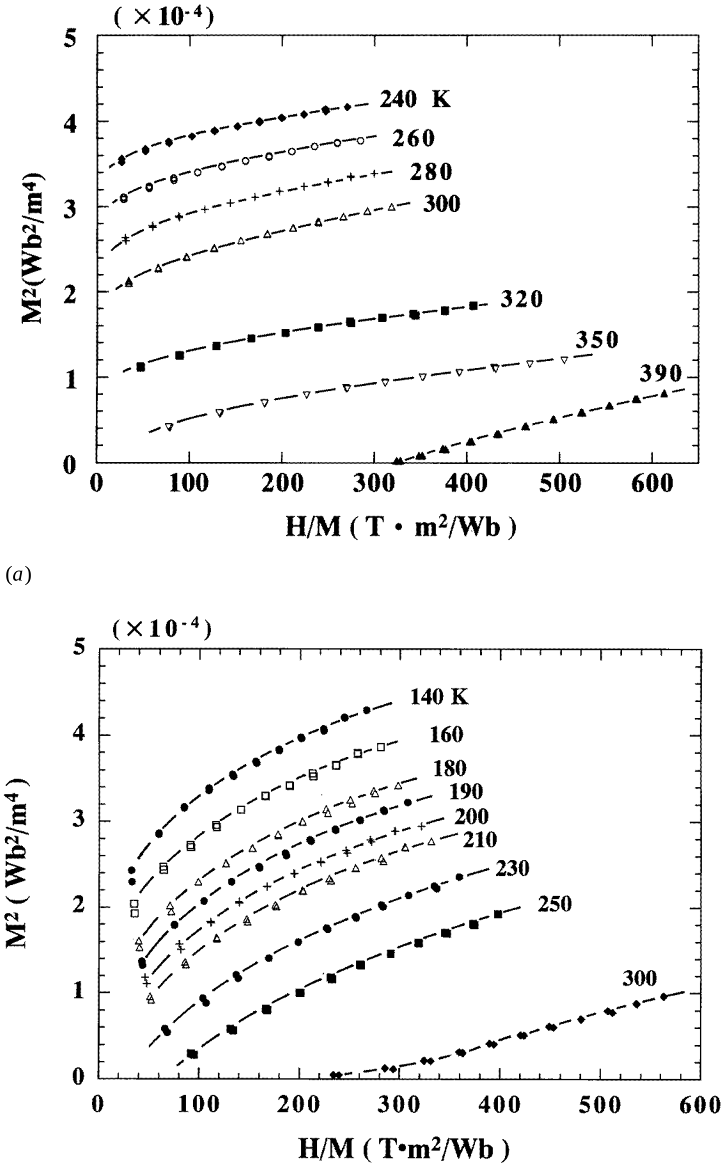


Figure 8. The concentration dependence of the pressure derivative of the Curie temperature $\partial T_C/\partial P$ for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys (–● –), together with that for amorphous Zr–Fe (–□–) and $La_{0.125}Fe_{0.875}$ (–△–) alloys (Fukamichi *et al* 1985, Goto *et al* 1988).

together with that of amorphous Zr–Fe and $La_{0.125}Fe_{0.875}$ alloys (Fukamichi *et al* 1985, Goto *et al* 1988) for comparison. It should be noted that the data for amorphous Hf–Fe and Sc–Fe alloys (Fukamichi *et al* 1985) are very similar to those for amorphous Zr–Fe alloys. The magnitude of $\partial T_C/\partial P$ for the former ternary alloys composed of icosahedral clusters is much larger than that for the latter binary alloys at the same Fe concentration. The number of the nearest-neighbour Fe atoms pairs in the amorphous $La(Fe_{0.95}Al_{0.05})_{13}$ alloy is about 10.3 and that in amorphous $La_{0.12}Fe_{0.88}$ alloy which has almost the same Fe concentration is about 9.7, although there is no distinct difference in the Fe–Fe interatomic distance between these two amorphous alloys. These data imply that the electronic structure of the present amorphous ternary alloys is closer to that of γ-Fe, compared with that of amorphous Fe-based binary alloys. The magnitude of $\partial T_C/\partial P$ significantly increases with increasing x, similar to that of the pressure derivative of magnetization $\partial M/\partial H$, high-field susceptibility χ_{hf} (Chiang *et al* 1991) and forced volume magnetostriction $\partial\omega/\partial H$ (Fukamichi *et al* 1995), which are closely correlated with the large spontaneous volume magnetostriction ω_S. The magnitude of ω_S is dominantly governed by the magnetoelastic coupling constant κC. Here κ is the compressibility and C is the coupling constant defined in terms of the Landau theory of phase transition (Shimizu 1980). The values of ω_S for the present amorphous alloys are extremely large (Chiang *et al* 1992), and hence the magnetoelastic coupling constant κC is also large (Fukamichi *et al* 1995). The marked pressure effects on the magnetization and the Curie temperature are also closely correlated with the large compressibility κ. The values of κ for the present alloys have been investigated at room temperature (Yoshihara *et al* 1994) and the values at 0 K estimated from the temperature dependence of Young's modulus are large (Fukamichi *et al* 1995), consistent with the present pressure effects. The magnitude of $\partial T_C/\partial P$ is proportional to κC and the high-field susceptibility χ_{hf} and is given by the following equation (Wagner and Wohlfarth 1981):

$$-(\partial T_C/\partial P) = 2\kappa C \chi_{hf} T_C. \tag{5}$$

The values of κC and χ_{hf} for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys are eminently large (Fukamichi *et al* 1995). Since κC is associated with the spontaneous volume magnetostriction ω_S, the ΔE effect (Chiang *et al* 1992) and the forced volume magnetostriction $\partial\omega/\partial H$, these values show a similar concentration dependence.



(*a*)

(*b*)

Figure 9. (*a*) Arrott plots of the amorphous $La(Fe_{0.80}Al_{0.20})_{13}$ ferromagnetic alloy. (*b*) Arrott plots of the amorphous $La(Fe_{0.95}Al_{0.05})_{13}$ re-entrant spin-glass alloy.

The Arrott plots of homogeneous alloys exhibit straight lines with the slopes independent of the temperatures, and the deviations give an indication that the materials are not magnetically homogeneous (Brommer and Franse 1990). Figure 9(*a*) and (*b*) shows the Arrott plots of the amorphous $La(Fe_{0.80}Al_{0.20})_{13}$ ferromagnetic and $La(Fe_{0.95}Al_{0.05})_{13}$ re-entrant spin-glass alloys, respectively. Both plots exhibit a strong curvature in connection with an inhomogeneous magnetic state. The Arrott plots of inhomogeneous systems have been discussed using a variant of Landau–Ginzburg theory taking into account cooperative spin fluctuations (Herzer *et al* 1980) and also using a superparamagnetic model (Acker and Huguenin 1979). It should be noted that the curvature of the re-entrant spin glass is more considerable than that of the ferromagnetic alloy. It has been pointed out that $\partial \ln T_C/\partial P$ versus T_C plot becomes linear for magnetically inhomogeneous systems. Substituting $\chi_1 - \chi_2 T_C$ for χ_{hf} in equation (5), the following empirical expression yields, giving a spin-glass-like peak for $T_C = 0$ (Wagner and Wohlfarth 1981),

$$\partial \ln T_C/\partial P = (1/T_C)\partial T_C/\partial P = -a + bT_C \qquad (6)$$

with

$$a = 2\kappa C\chi_1 \qquad b = 2\kappa C\chi_2.$$

Shown in figure 10 is the plot in the form of equation (6) for the present amorphous alloys, together with that of amorphous Zr–Fe and $La_{0.125}Fe_{0.875}$ alloys (Fukamichi *et al* 1985, Goto *et al* 1988), for comparison. A linear relationship is observed although the data show some scatter. It should be noted that the line of amorphous $La(Fe_xAl_{1-x})_{13}$ alloys is different from that of the amorphous binary alloys. That is, the pressure effect of the former is much stronger than that of the latter in the alloys which have the same Curie temperature. Such a distinct difference could be attributed to the difference in the amorphous structure as pointed out in the discussion of figure 8.

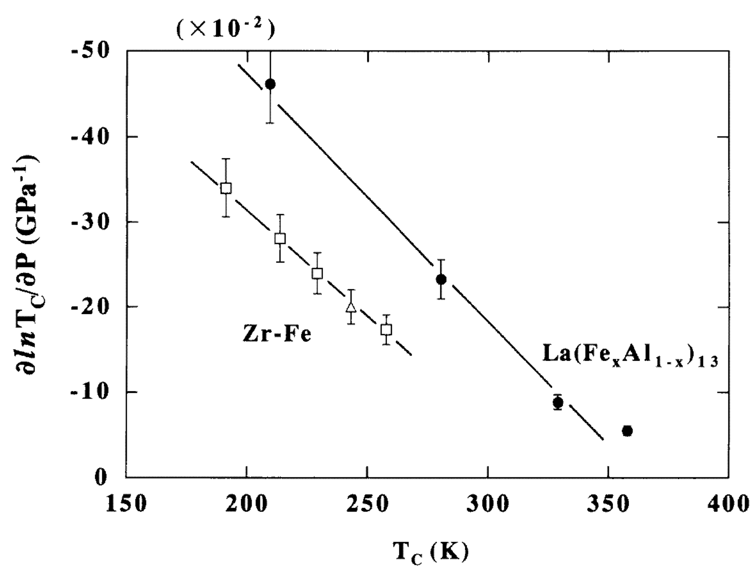


Figure 10. A pressure coefficient of the Curie temperature, $\partial \ln T_C/\partial P$, versus T_C plots for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys (–● –), together with that for amorphous Zr–Fe (–□–) and $La_{0.125}Fe_{0.875}$ (–△–) alloys (Fukamichi *et al* 1985, Goto *et al* 1988).

Even in inhomogeneous systems, spin fluctuations play an important role in magnetic properties (Fujita *et al* 1994a, b). Furthermore, it is worth noting that the giant forced volume magnetostriction $\partial\omega/\partial H$ of amorphous Fe-based alloys is explained by the finite-temperature theory of the local environment effect taking the spin fluctuation into consideration (Kakehashi 1993). The thermal expansion anomaly in a wide temperature range for amorphous Fe-based alloys is attributed to spin fluctuations (Fujita *et al* 1994a, b). The pressure effect due to spin fluctuations is given by the following expression (Wohlfarth 1980):

$$-\partial T_C/\partial P = 2\kappa C N(\varepsilon_F)\mu_B^2 T_F^{4/3} T_C^{-1/3} \tag{7}$$

where $N(\varepsilon_F)$ is the density of states at the Fermi level, μ_B the Bohr magneton and T_F the effective degeneracy temperature. As seen from figure 11, the $\partial T_C/\partial P$ versus $T_C^{-1/3}$ plot gives a relatively good linear relationship for the present amorphous $La(Fe_xAl_{1-x})_{13}$ alloys. The plots for amorphous Zr–Fe and $La_{0.125}Fe_{0.875}$ alloys (Fukamichi *et al* 1985, Goto *et al* 1988) are given in the same figure, for comparison. The slope for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys is about twice that for the amorphous binary alloys. Such a difference would be connected with the difference in the electronic structure due to the structural difference as mentioned in connection with figures 8 and 10. For conclusive discussion, further detailed information is necessary because the expression (7) is composed of several physical parameters.

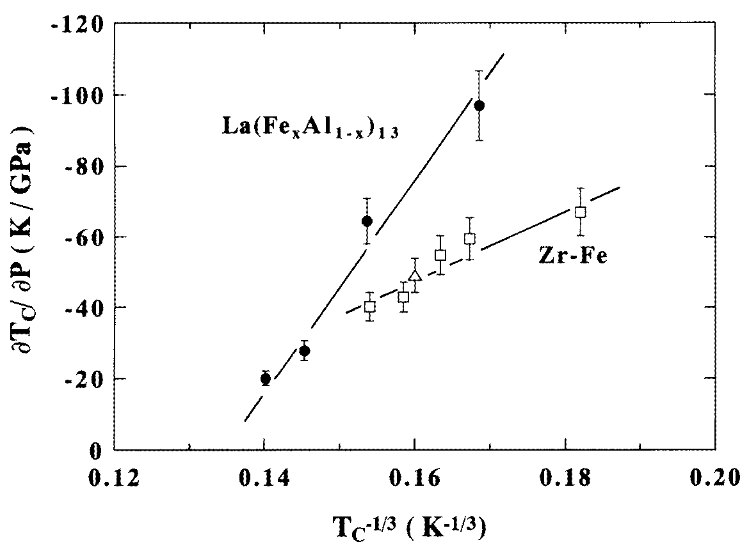


Figure 11. A pressure derivative of the Curie temperature, $\partial T_C/\partial P$, versus $T_C^{-1/3}$ plot for the amorphous $La(Fe_xAl_{1-x})_{13}$ alloys (–●–), together with that for amorphous Zr–Fe (–□–) and $La_{0.125}Fe_{0.875}$ (–△–) alloys (Fukamichi *et al* 1985, Goto *et al* 1988).

The marked pressure effect on the spin-glass state has been reported for amorphous La–Fe (Goto *et al* 1988) and Ce–Fe alloys (Fukamichi *et al* 1989). In the present study, the pressure effect is also investigated at 10^{-2} T using the same clamp pressure cell. The thermomagnetization curves of the amorphous $La(Fe_{0.90}Al_{0.10})_{13}$ alloy in figure 12 exhibit typical characteristics of the re-entrant spin glass, indicating that the Curie temperature is reduced and the spin freezing temperature is increased on application of hydrostatic pressure. Note that theoretical analysis for itinerant electron spin glasses gives a consistent explanation

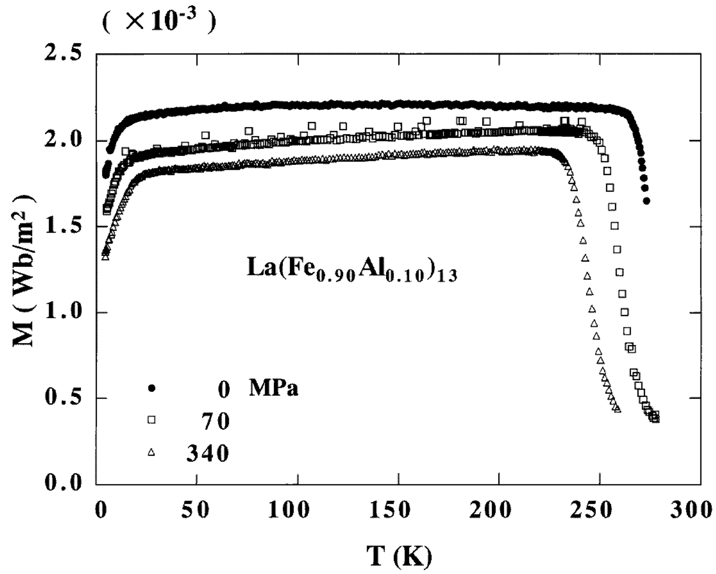


Figure 12. Thermomagetization curves at 10^{-2} T under hydrostatic pressures for the amorphous $La(Fe_{0.90}Al_{0.10})_{13}$ alloy.

by taking the spin fluctuations into consideration (Kakehashi 1993). Since the spin freezing temperature T_f is very sensitive to the magnetic field (Fukamichi *et al* 1995), it should be borne in mind that the present result is different from that reported before because of the different experimental conditions (Chiang *et al* 1991).

The pressure effects on the Curie temperature and the spin freezing temperatures have been obtained numerically by taking the derivative $\partial T_C/\partial V$ with respect to the volume V for amorphous Fe-based alloys (Kakehashi 1993). The calculated T_C decreases with increasing bandwidth, while T_f increases until the ferromagnetism disappears at a critical pressure. These calculated results are consistent with the experimental results for amorphous La–Fe alloys (Goto *et al* 1988). On the other hand, the decreases in both the temperatures with pressure have been reported for an amorphous $Y_{19}Fe_{81}$ alloy prepared by ion sputtering onto a nitrogen-cooled Al substrate (Andreenko *et al* 1993). The present results are in accord with the former, namely, a decrease in T_C and an increase in T_f occur. These shifts for the amorphous alloy with $x = 0.95$ are more remarkable than those of the amorphous alloy with $x = 0.90$, associated with the magnetic instability.

4. Conclusions

The pressure effects on the magnetization, the Curie temperature, the spin-wave stiffness constant and the spin freezing temperature have been investigated for amorphous $La(Fe_xAl_{1-x})_{13}$ alloys composed of icosahedral clusters. The following conclusions were derived from the results and discussion.

(i) The pressure effect on the Curie temperature T_C is very considerable and becomes more significant with increasing x. The magnitude of $\partial T_C/\partial P$ is much larger than that of amorphous Fe-based binary alloys, associated with the difference in the amorphous structure.

(ii) The pressure coefficient of the Curie temperature $\partial \ln T_C/\partial P$ is very large. The difference in the magnitude between the amorphous $La(Fe_xAl_{1-x})_{13}$ and amorphous Fe-based binary alloys could also be attributed to the difference in the amorphous structure.

(iii) The linear relationship between $\partial \ln T_C/\partial P$ and T_C is explained in terms of magnetic inhomogeneity. On the other hand, the spin fluctuations bring about the linear $\partial T_C/\partial P$ versus $T_C^{-1/3}$ plot.

(iv) The decrease in the magnetization on application of pressure is significantly large and its concentration dependence is similar to that of the forced volume magnetostriction $\partial\omega/\partial H$.

(v) The spin-wave stiffness constant determined from the $T^{3/2}$ dependence of magnetization is decreased on application of hydrostatic pressure, similar to the pressure effect on the Curie temperature.

(vi) The spin freezing temperature T_f is increased and the Curie temperature T_C is decreased on application of hydrostatic pressure, consistent with the theoretical calculations.

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