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Magnetic properties of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys

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Abstract. Several kinds of $La(Fe_xAl_{1-x})_{13}$ ($0.80 \leq x \leq 0.95$) amorphous alloys have been prepared by high-rate DC sputtering to investigate the magnetic properties. The magnetic phase diagram has been established by DC and AC magnetic field measurements. Above $x = 0.85$, re-entrant spin-glass behaviour has been observed in the concentration range where antiferromagnetic order occurs in the crystalline state. The high-field susceptibility in this concentration range is extremely large and the magnetization curves are not easily saturated. The magnetic moment estimated from the magnetization curves measured up to 380 kOe in pulsed magnetic fields shows a maximum around $x \sim 0.85$, corresponding to 80 at.% Fe. The Curie temperature in the amorphous state decreases with increasing x and it is higher than the Néel and Curie temperatures in the crystalline state. The spin-wave stiffness constant in the amorphous state is extremely small, suggesting a magnetic instability.

1. Introduction

No stable crystalline Fe–La intermetallic compounds exist because the heat of alloying between Fe and La is positive. Recently, however, the cubic $NaZn_{13}$-type compounds $La(Fe_xAl_{1-x})_{13}$ have been prepared successfully (van der Kraan *et al* 1983) by substitution of Al for a part of the transition metal in RT_{13} (R = rare-earth element, T = transition metal). The $La(Fe_xAl_{1-x})_{13}$ compounds can be stabilized in the concentration range $0.46 \leq x \leq 0.92$, and the lattice constant of these intermetallic compounds decreases linearly with increasing Fe content (Palstra *et al* 1984). It has been reported that the crystalline compound with $x = 0.91$ shows an antiferromagnetic long-range order at 4.2 K, which consists of ferromagnetic clusters coupled antiferromagnetically (Helmholdt *et al* 1986). A pressure-induced transition from the ferromagnetic to antiferromagnetic state has been observed at a pressure of $P \leq 0.1$ GPa in the $La(Fe_{0.86}Al_{0.14})_{13}$ crystalline compound (Abd-Elmegnid *et al* 1987). From the high-pressure experiments of Mössbauer effect on the magnetic properties of $La(Fe_{0.88}Al_{0.12})_{13}$, it has been pointed out that the Curie temperature and the average magnetic hyperfine field decrease abruptly at a critical pressure corresponding to the average Fe–Fe nearest-neighbour distance $d_c = 2.53$ Å (Ludorf *et al* 1989). As described above, it is clear that the magnetic properties of Fe are drastically affected by environment, such as atomic distance and coordination number. Furthermore, the cubic crystal structure of $La(Fe_xAl_{1-x})_{13}$ is composed of many icosahedral clusters (Palstra *et al* 1984),

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which is often correlated with the structures of amorphous and quasi-crystalline alloys (Kofalt *et al* 1986). A density difference between amorphous and crystalline alloys is observed (Konczos and Sas 1986), RFe_2 alloy systems show a distinct large difference (Fukamichi *et al* 1987) owing to a dense Laves-phase crystalline structure. These facts mean that the density is closely interrelated with the structure of both states.

The magnetic properties of $La_{1-y}Fe_y$ ($0.50 \leqslant y \leqslant 0.925$) amorphous alloys have been investigated (Wakabayashi *et al* 1990). The alloys with $y < 0.90$ show a re-entrant spin-glass behaviour at low temperatures. In the case of the alloys with $y \geqslant 0.90$ however, a direct transition from the paramagnetic to the spin-glass state was observed. Moreover, the extrapolation to $y = 1.0$ in the phase diagram suggests that amorphous Fe is a spin glass with a freezing temperature of about 110 K.

In the present study, the magnetic properties of several kinds of $La(Fe_xAl_{1-x})_{13}$ ($0.80 \leqslant x \leqslant 0.95$) amorphous alloys have been investigated and compared with the corresponding crystalline compounds and $La_{1-y}Fe_y$ amorphous alloys. The magnetic phase diagram of the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system has been established by AC and DC magnetic measurements. The spin-wave stiffness constants determined from the thermomagnetization curves have been compared with those of other Fe-based amorphous alloys. The density of the amorphous alloys and crystalline compounds has been measured at room temperature.

2. Experimental details

$La(Fe_xAl_{1-x})_{13}$ amorphous alloys about 0.3 mm thick were prepared by high-rate DC sputtering on a Cu substrate by using alloy targets made by arc melting in an argon atmosphere. The size of the alloy targets is about 50 mm in diameter. The argon gas pressure during sputtering was 40 mTorr, and the target voltage and the anode current were 1.0 kV and 6.0 A, respectively. The samples were confirmed to be in an amorphous state by x-ray diffraction. The Cu substrate was dissolved away in a solvent of CrO_3 (500 g) + H_2SO_4 (27 cm^3) + H_2O (1000 cm^3) around 350 K.

The magnetization measurements were carried out by an induction method up to 60 kOe using a superconducting magnet. The high-field magnetization measurements up to 380 kOe were made by an induction method with a wire-wound pulse magnet. The AC susceptibility measurements were carried out by a mutual induction method at 80 Hz and 1 Oe.

The room-temperature density of amorphous alloys and crystalline compounds was measured by the Archimedean method using toluene as the working fluid.

3. Results and discussion

Figure 1 shows the magnetization curves of four kinds of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys at 4.2 K. The magnetization for $x \leqslant 0.85$ is easily saturated in an external magnetic field less than 5 kOe, varying linearly in the high-field ranges in the same manner as that of conventional ferromagnetic alloys. On the other hand, in the concentration range $0.90 \leqslant x \leqslant 0.95$, the curves are not easily saturated even in a field of 60 kOe. Such a phenomenon becomes significant with increasing Fe content. This peculiar phenomenon has also been observed in many Fe-based amorphous alloys such as Fe–Zr (Hiroyoshi and Fukamichi 1982), Fe–Hf (Hiroyoshi *et al* 1985) and $La_{1-y}Fe_y$ (Wakabayashi *et al*

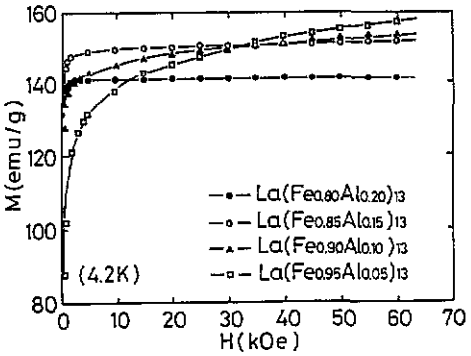


Figure 1. Magnetization curves of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys at 4.2 K in a steady field up to 60 kOe.

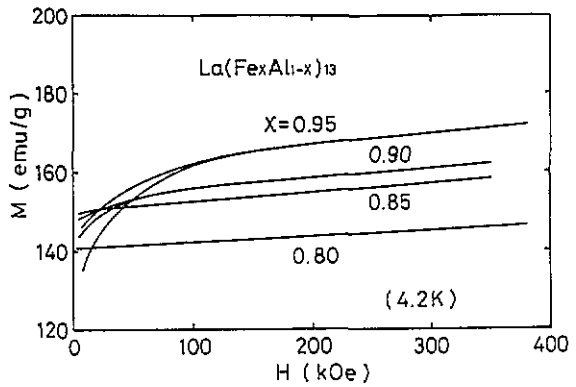
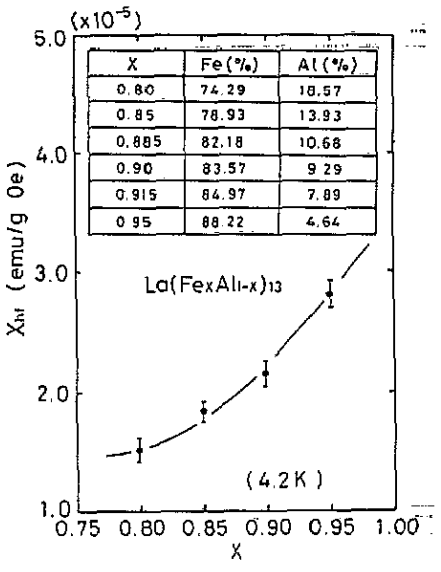


Figure 2. Magnetization curves of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys at 4.2 K in a pulsed field up to 380 kOe.

1990). It has been suggested that the frustrated antiferromagnetic interactions caused by the structural disorder in the amorphous state appear in the Fe-rich concentration range, resulting in the spin-glass state (Fukamichi *et al* 1989). Since the saturation is incomplete for $x \geq 0.90$ even at 60 kOe as described above, we have extended the magnetization measurements up to 380 kOe using the pulse magnet. As seen from figure 2, the magnetization curves for $x \geq 0.90$ seem to vary linearly with the applied field above 200 kOe. The saturation magnetization M_s and the high-field susceptibility χ_{hf} are obtained from the law of approach to saturation given by

$$M = M_s(1 - a/H - b/H^2) + \chi_{hf}H \tag{1}$$

where a and b are constants. The terms a/H and b/H^2 are concerned with the local and magnetocrystalline anisotropies, respectively. Since magnetocrystalline anisotropy in amorphous alloys is considered to be absent, M_s and χ_{hf} were determined by neglecting the b term in equation (1). The concentration dependence of χ_{hf} at 4.2 K is shown in figure 3. The value of χ_{hf} of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys increases with increasing Fe content. The concentration dependence of χ_{hf} is very similar to that of various crystalline and amorphous Invar-type alloys (Hiroyoshi *et al* 1978, 1983, Fukamichi *et*



X	Fe(%)	Al(%)
0.80	74.29	18.57
0.85	78.93	13.93
0.885	82.18	10.68
0.90	83.57	9.29
0.915	84.97	7.89
0.95	88.22	4.64

Figure 3. Concentration dependence of the high-field susceptibility χ_{hf} for $La(Fe_xAl_{1-x})_{13}$ amorphous alloys at 4.2 K. The table shows the alloy compositions of Fe and Al.

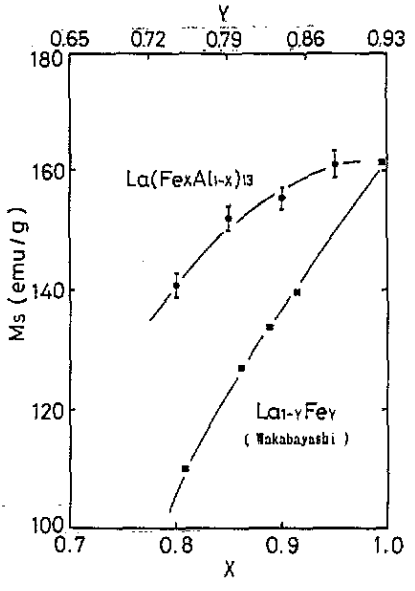


Figure 4. Concentration dependence of the saturation magnetization of the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system at 4.2 K, together with that of the $La_{1-y}Fe_y$ amorphous alloy system (Wakabayashi 1988).

al 1979). It is clear that the magnetization of the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system above $x = 0.90$ is sensitive to the external magnetic field. The concentration dependence of saturation magnetization M_s obtained from equation (1) for the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system is shown in figure 4. The curve for $La_{1-y}Fe_y$ amorphous alloys (Wakabayashi 1988) is also given in the same figure, for comparison. The value of M_s for $La(Fe_xAl_{1-x})_{13}$ amorphous alloys increases with increasing Fe content, showing a tendency to reach that of $La_{7.5}Fe_{92.5}$ amorphous alloy. The concentration dependence of magnetic moment μ of the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system is presented in figure 5. For comparison, the data on $La(Fe_xAl_{1-x})_{13}$ crystalline compounds (Palstra *et al* 1985) and $La_{1-y}Fe_y$ amorphous alloys (Wakabayashi *et al* 1990) are also presented in the same figure. In both $La(Fe_xAl_{1-x})_{13}$ and $La_{1-y}Fe_y$ amorphous alloy systems, the magnetic moment exhibits a similar concentration dependence, taking a broad maximum. The values of the former alloys are slightly lower than those of the latter ones. The magnetic moment of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys deduced from the pulsed field measurements shows a tendency to decrease gradually towards that of $La_{7.5}Fe_{92.5}$ amorphous alloy, which corresponds to the concentration of $La(Fe_xAl_{1-x})_{13}$ when $x = 1$. These results are consistent with the results obtained from the Mössbauer effect measurements (Wakabayashi *et al* 1989). The concentration dependence of the magnetic moment is slightly reduced by substitution of Al for a part of Fe in the present alloys. On the other hand, the magnetic moment of $La(Fe_xAl_{1-x})_{13}$ crystalline compounds increases linearly with increasing Fe content. In the figure, the values for the crystalline compounds in the antiferromagnetic region were determined in fields beyond the spin-flip transition (Palstra *et al* 1985) and are shown by the broken line. It should be noted that the magnetic moment of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys is

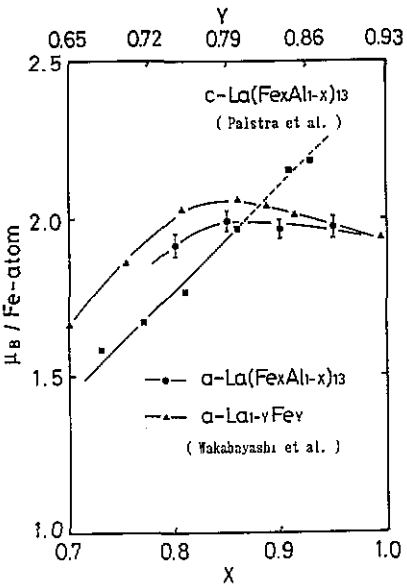


Figure 5. Concentration dependence of the magnetic moment μ_B per Fe atom of the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system, together with that of $La(Fe_xAl_{1-x})_{13}$ crystalline compounds (Palstra *et al* 1985) and $La_{1-y}Fe_y$ amorphous alloys (Wakabayashi *et al* 1990).

smaller than that of crystalline counterparts above $x = 0.85$. It is well known that the magnetic moment of Fe is significantly affected by local environment, such as the atomic distance of Fe–Fe pairs and the coordination number (Kouvel and Wilson 1961, Jaccarino and Walker 1965). Therefore, the difference in the magnetic properties mentioned above would originate from the difference in the above-mentioned environments between the amorphous and crystalline states. Recently, the correlation between the magnetic properties and the structure of Fe-rich amorphous $La_{1-y}Fe_y$ alloys has been investigated, using large-angle and small-angle x-ray scattering (Matsuura *et al* 1989). According to their results, the interatomic distance between Fe–Fe atoms is slightly shortened and the mean Fe–Fe coordination number is increased with increasing Fe content. Under such circumstances, the frustrated antiferromagnetic exchange interactions are considered to develop with increasing Fe content, resulting in re-entrant spin-glass behaviour at low temperatures. It is known that the ferromagnetic interaction is stable when the Fe–Fe coordination number in the nearest-neighbour shell of the pair distribution function is smaller than 6 and antiferromagnetic interactions prevail above it (Kakehashi 1990). In the case of $La(Fe_xAl_{1-x})_{13}$ crystalline compounds, the Fe atom sites are occupied by two different site atoms, Fe^{I} and Fe^{II}. The Fe^{I} atoms are surrounded by an icosahedron of 12 Fe^{II} atoms and the Fe^{II} atoms by nine nearest Fe^{II} atoms and one Fe^{I} atom. Furthermore, the Fe^{I}–Fe^{II} distance is shorter by about 2% than that of the Fe^{II}–Fe^{II} and decreases linearly with x from 2.510 Å for $x = 0.46$ to 2.431 Å for $x = 0.92$ (Palstra *et al* 1985). The atomic distance of Fe^{I}–Fe^{II} in $La(Fe_xAl_{1-x})_{13}$ crystalline compounds (Palstra *et al* 1985) is much smaller than that of $La_{1-y}Fe_y$ amorphous alloys (Matsuura *et al* 1989).

It is considered that the density is closely correlated with the structure of alloys as mentioned in the introduction. Therefore, the measurement of density is very important in the present alloy system. Figure 6 shows the concentration dependence of the room-temperature density of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys, together with that of the

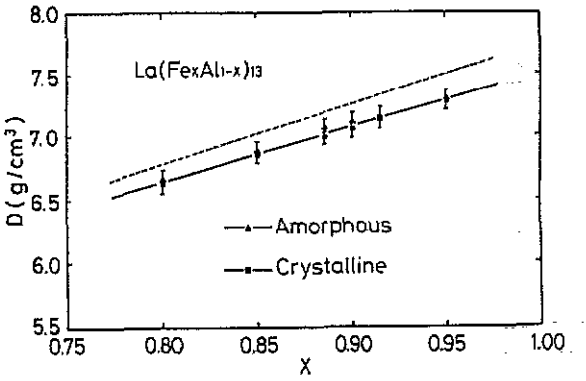


Figure 6. Concentration dependence of the room-temperature density of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys, together with that of $La(Fe_xAl_{1-x})_{13}$ crystalline compounds. The broken line shows Vegard's law.

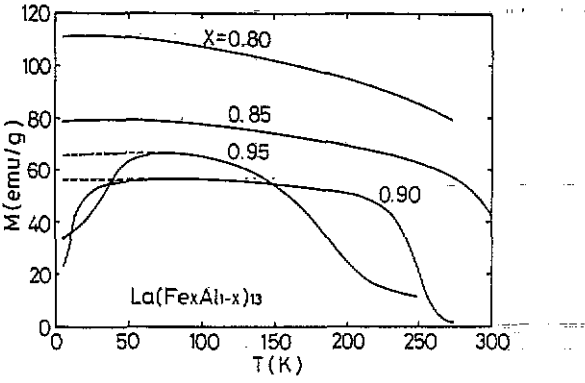


Figure 7. Temperature dependence of the zero-field-cooled and field-cooled magnetization in a 100 Oe field for $La(Fe_xAl_{1-x})_{13}$ amorphous alloys.

crystalline compounds. The broken line shows Vegard's law (Vegard 1921). The densities in the amorphous and crystalline states show a gradual increase with increasing x. Both of them are lower than the values obtained from Vegard's law. The difference from Vegard's law increases with increasing Fe content. Generally, the density difference between the amorphous and crystalline states is less than 2% and the crystalline state is denser (Konczos and Sas 1986). However, as seen from figure 6, both of them take almost the same values. Since the magnetic properties are different from each other in the amorphous and crystalline states, it is expected that the coordination number in these two states is not the same. Detailed structural analyses are now in progress.

As seen from figures 1 and 3, the magnetization curves are not easily saturated in high magnetic fields for the alloys in the concentration range $0.90 \leqslant x \leqslant 0.95$, and the high-field susceptibility χ_{hf} increases remarkably with increasing Fe content. These results suggest that the present alloys exhibit a spin-glass-like behaviour at low temperatures in this concentration range. Figure 7 shows the thermomagnetization curves of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys. The full curves represent the heating curves measured

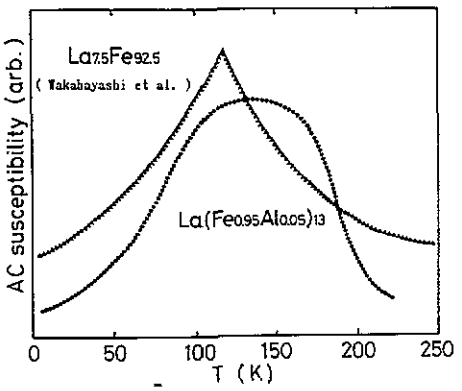


Figure 8. Temperature dependence of the AC susceptibility obtained in 1 Oe field at 80 Hz for $La(Fe_{0.95}Al_{0.05})_{13}$ amorphous alloy, together with that measured under the same conditions for $La_{7.5}Fe_{92.5}$ amorphous alloy (Wakabayashi *et al* 1990).

after cooling the samples from room temperature to 4.2 K in zero field. The broken curves show the cooling curves measured in a field of 100 Oe. In the case of the samples with $0.80 \leqslant x \leqslant 0.85$, the thermomagnetization curves are reversible between cooling and heating processes, being similar to those of normal ferromagnets. However, they indicate a characteristic hysteresis between the zero-field-cooled and field-cooled states for the alloys in the concentration range $0.90 \leqslant x \leqslant 0.95$. The hysteresis of the field-cooling effect at low temperatures becomes more remarkable with increasing Fe content. It is well known that a large random magnetic anisotropy exists in amorphous alloys when they contain rare-earth metals with a non-S state (Harris and Zobin 1977). However, the values of orbital and spin quantum numbers of La are 0 in $La(Fe_xAl_{1-x})_{13}$ amorphous alloys and the large random magnetic anisotropy is absent. The AC susceptibility of these amorphous alloys has been investigated in order to elucidate the spin-glass behaviour. Figure 8 shows the temperature dependence of AC susceptibility of $La(Fe_{0.95}Al_{0.05})_{13}$ amorphous alloy measured in an AC magnetic field of 1.0 Oe with a frequency of 80 Hz, together with that of $La_{7.5}Fe_{92.5}$ amorphous alloy measured under the same conditions (Wakabayashi *et al* 1990). As shown in the figure, the latter exhibits a characteristic cusp in the spin glasses. Namely, it undergoes a transition from a paramagnetic (PM) to a spin-glass (SG) state with decreasing temperature. On the other hand, the temperature dependence of AC susceptibility of the former is different from that of the latter, showing a broad maximum. It has been pointed out that such a broad maximum of Fe–Zr amorphous alloys is divided into two peaks in superposed DC fields, which correspond to the Curie and the spin freezing temperatures (Saito *et al* 1986). Therefore, it is considered that the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system exhibits a re-entrant-type spin-glass (RSG) behaviour.

The magnetic phase diagram of the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system obtained from the DC magnetization and AC susceptibility measurements is presented in figure 9, together with that of $La(Fe_xAl_{1-x})_{13}$ crystalline alloys (Palstra *et al* 1985). In the crystalline alloy system, the magnetic phase diagram is composed of three different magnetic orders: (i) in the low Fe concentration range, $0.46 \leqslant x < 0.62$, the temperature dependence curve of AC susceptibility shows a cusp, indicating a mictomagnetic-like state; (ii) with increasing Fe content, a ferromagnetic state appears in the concentration range $0.62 < x \leqslant 0.86$; (iii) in the higher Fe concentration range, $0.86 < x \leqslant 0.92$, the antiferromagnetic state is stable and the metamagnetic transition is induced by external fields of a few tesla (Palstra *et al* 1984). In the amorphous state, it should be noted

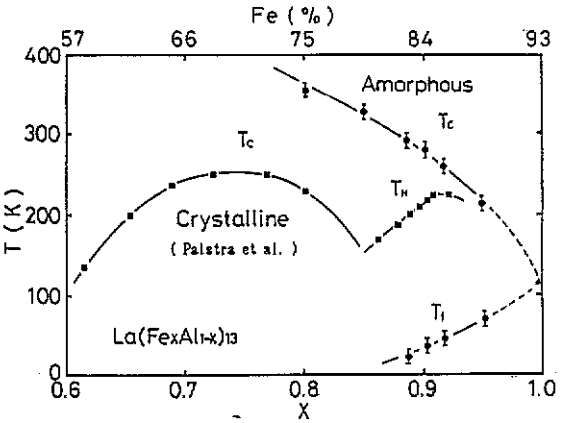


Figure 9. Magnetic phase diagram of the $La(Fe_xAl_{1-x})_{13}$ amorphous alloy system, compared with that in the crystalline state (Palstra *et al* 1985). The spin freezing temperature of $La_{7.5}Fe_{92.5}$ amorphous alloy is indicated by the full triangle on the right (Wakabayashi *et al* 1990).

that the re-entrant spin-glass behaviour appears in the concentration range of the antiferromagnetic state in the crystalline compounds as seen from figure 9. It is considered that the antiferromagnetic order in the crystalline compounds is stabilized with increasing Fe coordination number and with decreasing atomic distance in the Fe-rich concentration range (Abd-Elmegnid *et al* 1987). On the other hand, antiferromagnetic long-range order is absent in the amorphous alloys because of the structural disorder (Kaneyoshi 1983). Therefore, the origin of spin-glass-like behaviour at sufficiently low temperatures in the amorphous alloys would be due to the frustrated antiferromagnetic interactions caused by the structural disorder in the amorphous state. The Curie temperature T_C was determined from the point of steepest decrease of the thermomagnetization curves. The spin freezing temperature T_f was defined as the branch of these curves. In the case of Fe–Ce and Fe–Lu amorphous alloys, these two temperatures thus determined correspond to the points of steepest increase and decrease of AC susceptibility curves measured in 1 Oe at 80 Hz (Komatsu *et al* 1991). Strictly speaking, T_f and T_C may be slightly higher and lower, respectively, because they depend on the magnitude of the applied magnetic field (Saito *et al* 1988, Komatsu *et al* 1991). As seen from figure 9, T_C decreases and T_f increases with increasing Fe content, and they show a tendency to link up with the spin freezing temperature of $La_{7.5}Fe_{92.5}$ amorphous alloy (Wakabayashi *et al* 1987). It should be noted that the Curie temperature of the amorphous alloys is higher than the Curie and Néel temperatures of the crystalline compounds, making a striking contrast with many other Fe-based alloys (Fukamichi *et al* 1989).

The temperature dependence of the magnetization at low temperatures has been examined in order to get the spin-wave stiffness constant in the amorphous state. The present amorphous alloys show a remarkable magnetovolume effect, resulting in the Invar characteristics (Chiang *et al* 1991). The variation of magnetization with temperature for Invar alloys is given by the following expression (Wasserman 1990):

$$M = M(0,0)(1 - BT^{3/2} - CT^2 + \ldots). \tag{2}$$

Here $M(0, 0)$ is the magnetization at 0 K and zero magnetic field and B is related to the spin-wave stiffness constant given by the following expression:

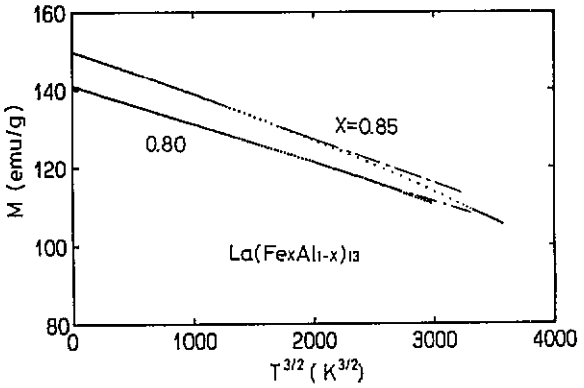


Figure 10. Temperature dependence of magnetization (M versus $T^{3/2}$ plots) measured in 1 T for $La(Fe_xAl_{1-x})_{13}$ ($x = 0.80$ and 0.85) amorphous alloys. The chain lines are a guide for the eye.

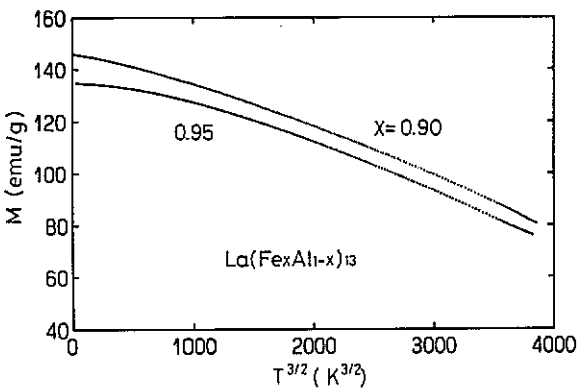


Figure 11. Temperature dependence of magnetization (M versus $T^{3/2}$ plots) measured in 1 T for $La(Fe_xAl_{1-x})_{13}$ ($x = 0.90$ and 0.95) amorphous alloys.

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

The coefficient C is the Stoner excitation term (Yamada 1983). The magnetization of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys obeys the $T^{3/2}$ temperature dependence at low temperatures only in the concentration range $0.80 \leqslant x \leqslant 0.85$, as shown in figure 10. The straight chain lines are a guide for the eye. On the other hand, such a linear dependence does not hold above $x = 0.85$ due to spin freezing, as seen from figure 11. The spin-wave stiffness constant D determined from equation (3) for $La(Fe_xAl_{1-x})_{13}$ amorphous alloys ($0.80 \leqslant x \leqslant 0.85$) is plotted in figure 12, and compared with the data on Fe–B and Fe–P amorphous alloys (Kazama *et al* 1978). It should be noted that the value of D for $La(Fe_xAl_{1-x})_{13}$ amorphous alloys is extremely small, suggesting an instability of ferromagnetism.

As is well known, Invar alloys exhibit various peculiarities in magnetic properties such as a large high-field susceptibility, small spin-wave stiffness constant, large pressure effect on the Curie temperature, large compressibility and so on (Fukamichi 1983). As

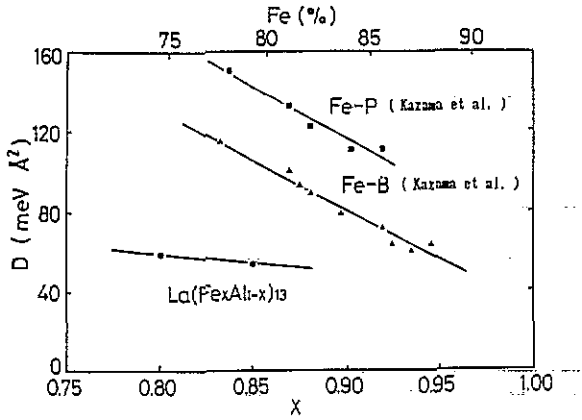


Figure 12. Concentration dependence of the spin-wave stiffness constant D of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys at 4.2 K, together with that of Fe–B and Fe–P amorphous alloys (Kazama *et al* 1978).

shown in figures 3 and 9, the concentration dependences of the high-field susceptibility and the Curie temperature are analogous to those of Fe–B and Fe–P amorphous alloys and Fe–Ni crystalline Invar alloys (Fukamichi *et al* 1979). Moreover, the spin-wave stiffness constant of the present alloys is about 50–60 meV Å^2, which is much smaller than that of Fe–B and Fe–P amorphous alloys (Kazama *et al* 1978), as shown in figure 12. As mentioned before, the present amorphous alloys exhibit the Invar characteristics. Furthermore, they also show anomalous elastic properties. These experimental results will be discussed in a following paper.

4. Summary

Magnetic properties of several kinds of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys prepared by high-rate DC sputtering have been investigated. The difference in the magnetic properties of $La(Fe_xAl_{1-x})_{13}$ alloys between the amorphous and crystalline states is discussed and compared with those of $La_{1-y}Fe_y$ amorphous alloys. The concentration dependence of the room-temperature density in the amorphous and crystalline states has been measured. The main results are summarized as follows:

The concentration dependence of the magnetic moment per Fe atom of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys is very similar to that of $La_{1-y}Fe_y$ amorphous alloys. The values of amorphous alloys are higher than those of crystalline counterparts below $x = 0.85$.

$La(Fe_xAl_{1-x})_{13}$ amorphous alloys show re-entrant spin-glass behaviour in the concentration range $0.85 < x \leq 0.95$, where the antiferromagnetic order takes place in the crystalline state.

The spin-glass state is responsible for a significantly large high-field susceptibility at 4.2 K.

The Curie temperature of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys is higher than that of the crystalline compounds.

The spin-wave stiffness constant of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys is much smaller than that of Fe–B and Fe–P Invar-type amorphous alloys.

The magnitude and the concentration dependence of the room-temperature density of $La(Fe_xAl_{1-x})_{13}$ amorphous alloys are very similar to those of the crystalline compounds.

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