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# Spin fluctuations in amorphous $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$ alloys consisting of icosahedral clusters

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Received 14 June 1994, in final form 30 September 1994

**Abstract.** Thermal properties of spin fluctuations have been investigated for amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  ( $0.90 \leq x \leq 0.975$ ) alloys. The temperature  $T$  dependence of magnetization  $M$  for the present alloys exhibits an  $M^2-T^{4/3}$  relation below the Curie temperature  $T_C$  in the  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  amorphous alloy and its relation becomes poor with increasing Ni concentration. The inverse magnetic susceptibility shows a peculiar temperature dependence above  $T_C$  owing to the saturation of the thermal growth of spin fluctuations. The temperature dependence of the inverse magnetic susceptibility is influenced by the external magnetic field. The distribution of spin fluctuations is localized in both wavenumber and frequency space in the low-Ni-concentration range, and it becomes broader with increasing Ni concentration. The generalized Rhodes–Wohlfarth plots for the present alloy system indicate that the magnetic susceptibility is dominated by the thermal variation in spin fluctuations.

## 1. Introduction

The magnetic properties of crystalline 3d transition metals and alloys have been explained in terms of energy band models. The Stoner (1938) model can be applied to the ground state, but the excited motion of 3d electrons can be treated only as a single-particle excitation and the interactions between the excited electrons and holes are neglected in this model (Murata and Doniach 1972, Moriya and Kawabata 1973, 1974). The thermal fluctuations of local spin density are excited at finite temperatures, and the magnetic properties of these metals and alloys are affected by the spin fluctuations. In particular, the spin fluctuations in weak ferromagnets dominate the thermal variation in magnetic properties (Moriya and Kawabata 1973, 1974). This theory has treated crystalline alloys and compounds, and the spin fluctuations in non-crystalline systems remain to be investigated. Recent theoretical treatment for the electronic state of amorphous alloys shows that the magnetic properties of 3d-metal-based amorphous alloys are also affected by the thermal spin fluctuations, i.e. many features of the magnetic properties of 3d-transition-metal-based amorphous alloys are explained by the spin fluctuations (Kakehashi 1989, 1991).

It has been pointed out that the ferromagnetic properties of Ni collapse as a result of structural disorder (Kakehashi 1991, Tanaka and Takayama 1992), while many of the Ni-based amorphous alloys exhibit weak ferromagnetic properties in the Ni-rich region (Liénard and Rebouillat 1978, Buschow 1984, Frémy *et al.* 1984). The appearance of ferromagnetism in amorphous Ni-based alloys is caused by the smaller concentration fluctuation, compared with that of amorphous Fe-based alloys (Kakehashi 1991, Tanaka and Takayama 1992).

It is noteworthy that amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys have a well defined local unit, i.e. icosahedral clusters composed of Ni and Al (Matsubara *et al* 1994). The structural analyses for the present amorphous alloys have revealed that the clusters randomly coordinate the La atom and every La atom has no La atom in the nearest-neighbour site (Matsubara *et al* 1994). The detail structures of the amorphous  $\text{La}(\text{TM}_x\text{Al}_{1-x})_{13}$  alloys ( $\text{TM} \equiv \text{Fe}, \text{Ni}$  or  $\text{Co}$ ) have been discussed elsewhere (Matsubara *et al* 1992, Chiang *et al* 1994, Matsubara *et al* 1994). The formation of such well defined icosahedral clusters reduces the concentration fluctuation in contrast with other binary amorphous alloys, and hence the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloy system is expected to exhibit weakly itinerant electron ferromagnetism. Moreover, the magnetic properties can easily be controlled by the Al concentration. Accordingly, this system is useful for investigating the spin fluctuations in amorphous alloys.

## 2. Experimental details

The target alloys for sputtering were made by arc melting. Amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  ( $0.90 \leq x \leq 0.975$ ) samples were prepared by high-rate DC sputtering. The sputtering on a water-cooled Cu substrate was carried out for 2.5 d and the substrate was mechanically ground off to obtain the bulk amorphous samples about 0.1 mm thick for the magnetization measurement. Magnetization at 4.2 K was measured up to 55 kOe in a steady field with a SQUID magnetometer, and the thermomagnetization curve at 100 Oe was obtained in the temperature range from 4.2 K to room temperature. Magnetization measurements up to 400 kOe in pulsed fields were also carried out at 4.2 K to evaluate the saturation magnetization.

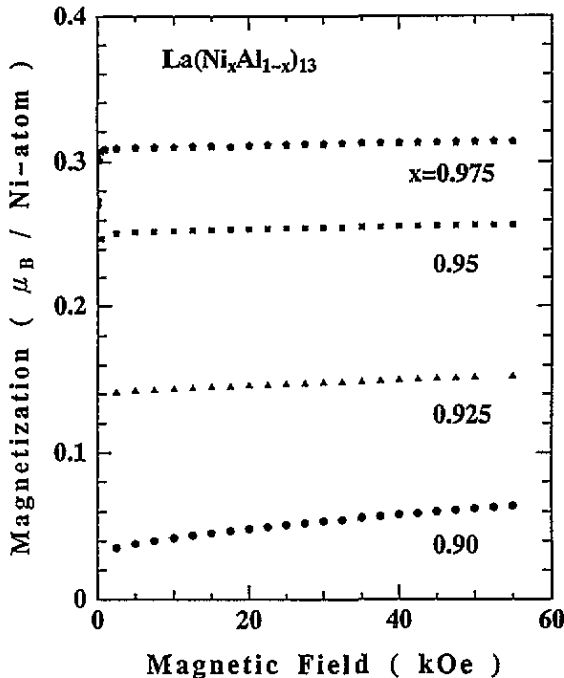


Figure 1. Magnetization curves at 4.2 K for amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys.

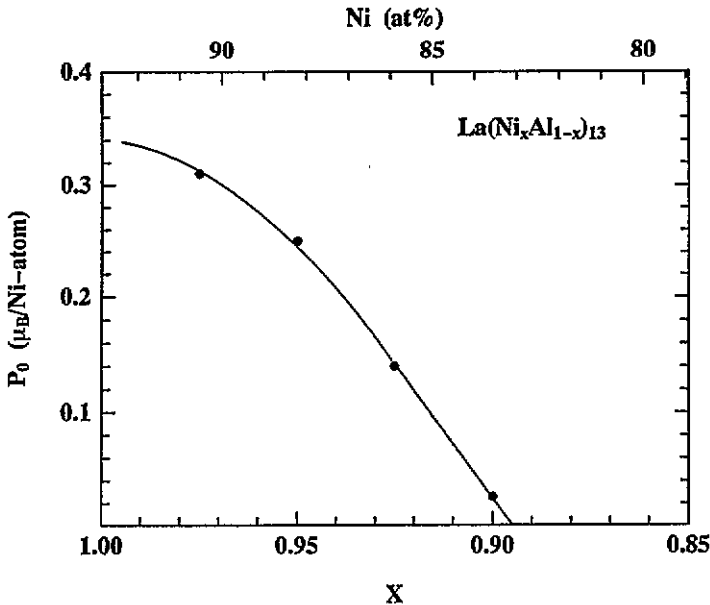


Figure 2. Concentration dependence of the spontaneous magnetic moment for amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys.

### 3. Results and discussion

Figure 1 shows the magnetization curves at 4.2 K for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys. In the highest-Ni-concentration alloy, the magnetization is almost saturated above 5 kOe, while the high-field susceptibility becomes larger as the Ni concentration decreases. In particular, the high-field susceptibility is very large for the amorphous  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  alloy because the Ni concentration is very close to the critical region for the appearance of ferromagnetism. The concentration dependence of the spontaneous magnetic moment  $P_0$ , per Ni atom for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys is plotted in figure 2. In the vicinity of the critical concentration of the disappearance of ferromagnetism, the saturation magnetic moment linearly decreases with the decrease in the Ni concentration. In the higher-Ni-concentration range, however, the decrease in  $P_0$  with increasing concentration becomes sluggish. The value of  $P_0$  extrapolated to  $x = 1.0$  is about  $(0.35\text{--}0.4)\mu_B$ , being fairly consistent with the magnetic moment of the amorphous La-Ni binary alloy for a Ni concentration of around 93% (Buschow 1984).

The self-consistent renormalization (SCR) theory (Moriya and Kawabata 1973, 1974) predicts that the temperature dependence of magnetization for weakly itinerant electron ferromagnets follows the  $M^2\text{--}T^{4/3}$  relation below the Curie temperature  $T_C$ . The thermal variation in spontaneous magnetization  $M(0, T)$  of the amorphous  $\text{La}(\text{Ni}_{0.925}\text{Al}_{0.075})_{13}$  alloy is plotted in the form of  $M^2(0, T)/M^2(0, 0)\text{--}T^\beta/T_C^\beta$  ( $\beta = \frac{4}{3}$  and 2) in figure 3. The value of  $M(0, T)$  is derived from the intercept of the Arrott plots. Apparently, a linear relation is observed in a wide temperature range in the plots with  $\beta = \frac{4}{3}$ , compared with those with  $\beta = 2$ . That is to say, the former plots exhibit a linear relation in a wide temperature range except for low temperatures, whereas the latter do only in a limited low-temperature range. Such a  $T^{4/3}$  dependence is also predicted by considering both longitudinal and transverse spin fluctuation effects (Lonzarich and Taillefer 1985). According to this model,

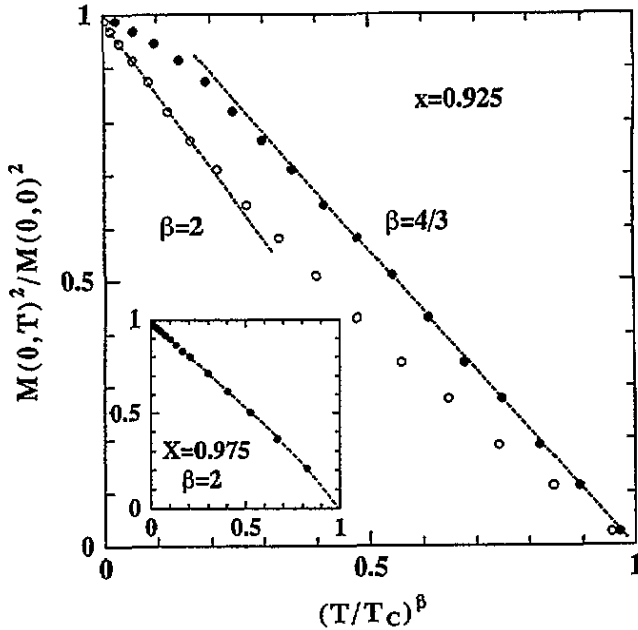


Figure 3. Temperature dependence of the spontaneous magnetization for the amorphous  $\text{La}(\text{Ni}_{0.925}\text{Al}_{0.075})_{13}$  alloy in the form of an  $M^2(0, T) - T^\beta$  ( $\beta = \frac{4}{3}$  and 2). The inset shows the  $M^2(0, T) - T^2$  plot for the amorphous  $\text{La}(\text{Ni}_{0.975}\text{Al}_{0.025})_{13}$  alloy.

the  $T^{4/3}$  dependence of  $M^2(0, T)$  is restricted to the temperatures close to  $T_C$ , and the  $T^2$  dependence will be dominant in almost the whole temperature range below  $T_C$  (Lonzarich and Taillefer 1985). Hereafter, this theory is referred to as the LT model. The results for the amorphous  $\text{La}(\text{Ni}_{0.925}\text{Al}_{0.075})_{13}$  alloy are consistent with the SCR theory rather than with the LT model. On the other hand, the thermal variation in  $M^2(0, T)$  is quadratic for the amorphous  $\text{La}(\text{Ni}_{0.975}\text{Al}_{0.025})_{13}$  alloy, as seen in the inset of figure 3. The coefficient of  $T^2$  is a measure of the contribution from the thermal spin fluctuations in addition to that from Stoner-type single-particle excitations. The single-particle excitation makes a small contribution, compared with the spin fluctuations, to the quadratic thermal variation in  $M^2(0, T)$ , because it results from the thermal smearing of the Fermi surface (Stoner 1938, Moriya 1985). When the thermal demagnetization due to the single-particle excitation is expressed in the form  $M^2(0, T)/M^2(0, 0) = S(T_C^2 - T^2)$ , the coefficient  $S$  is equal to  $T_C^{-2}/2$  (Edwards and Wohlfarth 1968). On the other hand, the coefficient of  $T^2$  is enhanced by spin fluctuations via a paramagnon-like mode of fluctuation (Makoshi and Moriya 1975, Lonzarich and Taillefer 1985, Moriya 1985). The estimated value of  $S = T_C^{-2}/2$  for the amorphous  $\text{La}(\text{Ni}_{0.925}\text{Al}_{0.075})_{13}$  alloy is  $0.64 \times 10^{-4}$ , while the observed value of the slope of the  $M(0, T)^2 - T^2$  plot is  $1.86 \times 10^{-4}$ , which is about three times the calculated value. A similar comparison has been carried out for the amorphous  $\text{La}(\text{Ni}_{0.975}\text{Al}_{0.025})_{13}$  alloy and the ratio of the two values is about 1.5. Therefore, an enhancement effect of  $S$  due to the spin fluctuations exists, but it becomes smaller with increasing Ni concentration. The Curie temperature  $T_C$  of the amorphous  $\text{La}(\text{Ni}_{0.975}\text{Al}_{0.025})_{13}$  alloy is 220 K, which is about 130 K higher than  $T_C$  for the amorphous  $\text{La}(\text{Ni}_{0.925}\text{Al}_{0.075})_{13}$  alloy. In the former alloy, the ferromagnetic properties are further stabilized, and hence the exciting energies of both longitudinal and transverse spin fluctuations turn out to be closer. Consequently,

the quadratic thermal variation in  $M^2(0, T)$  in a wide temperature range for the amorphous  $\text{La}(\text{Ni}_{0.975}\text{Al}_{0.025})_{13}$  alloy is explained qualitatively by the LT model.

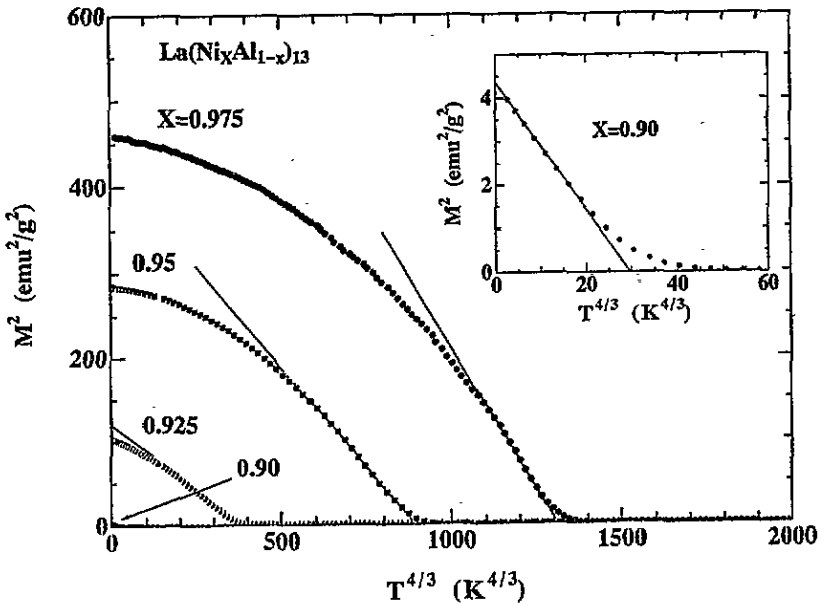


Figure 4. Temperature dependence of the magnetization for amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys in the form of an  $M^2-T^{4/3}$  plot. The inset shows the result for the amorphous  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  alloy.

The validity of the  $M^2-T^{4/3}$  relation can be checked for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys with various Ni concentrations in figure 4. We compare  $M(0, T)$  and the in-field magnetization  $M(H, T)$  ( $H = 100$  Oe) and confirm that  $M(H, T)$  is also characterized by the  $M^2-T^{4/3}$  relation below the Curie temperature. The data plotted in figure 4 are the in-field magnetization  $M$  for all samples. The inset shows the result for the amorphous  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  alloy at temperatures below  $T_C$ . The square  $M^2$  of the magnetization for these alloys varies linearly with  $T^{4/3}$  in a relatively wide temperature range below  $T_C$ . Although the spin fluctuations are influenced by the external magnetic field (Takeuchi and Masuda 1979, Ikeda *et al* 1991), the  $M^2-T^{4/3}$  relation observed in the present alloys is intrinsic, because the measurement was carried out in a magnetic field of 100 Oe, whose energy is too small to make a drastic change in the spin fluctuations. Only the slight deviation from the straight line around  $T_C$  may be due to the strength of the external magnetic field. The  $M^2-T^{4/3}$  relation is observed in weakly ferromagnetic compounds such as  $\text{Ni}_3\text{Al}$  (Sasakura *et al* 1984) and  $\text{Y}(\text{Co}_x\text{Al}_{1-x})_2$  (Yoshimura and Nakamura 1985), and it is considered that the longitudinal spin fluctuations dominate their magnetic properties at finite temperatures. This relation is invalid in the localized moment system because the transverse spin fluctuations are dominant (Moriya and Kawabata 1973, Moriya and Takahashi 1978). Generally speaking, there exists an interpolation region between these two limits and spin fluctuations gradually change as the system changes from the weakly itinerant electron ferromagnet limit to the localized magnetic moment limit (Moriya and Takahashi 1978). In fact, the  $M^2-T^{4/3}$  curve is fairly flat even at low temperatures

for the amorphous  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  alloy, indicating that the  $M^2-T^{4/3}$  relation holds in wide temperature ranges, while the relation becomes poor at lower temperatures with increasing  $T_C$  and holds only near  $T_C$ . The Curie temperature  $T_C$  linearly decreases with increasing Ni concentration in the vicinity of the critical concentration of the disappearance of ferromagnetism as seen in figure 5. These temperatures are deduced from the linear relation between  $M^2$  and  $T^{4/3}-T_C^{4/3}$  (Moriya and Kawabata 1973). Note that the difference between the Curie temperatures obtained by this method and from the Arrott plots is less than  $\pm 5$  K. Correspondingly, spin-wave excitation is observed in the high-Ni-concentration region. In the amorphous  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  alloy, the linear part of the  $M-T^{3/2}$  plot is not observed, while the  $T^{3/2}$  variation is observed at low temperatures in amorphous  $\text{La}(\text{Ni}_{0.925}\text{Al}_{0.075})_{13}$ ,  $\text{La}(\text{Ni}_{0.95}\text{Al}_{0.05})_{13}$  and  $\text{La}(\text{Ni}_{0.975}\text{Al}_{0.025})_{13}$  alloys. The linear range is about  $(0.15-0.17) T_C$  for the former two alloys and about  $0.3T_C$  for the latter alloy. These results are consistent with the concentration dependences of the  $M^2(0, T)-T^2$  and  $M^2-T^{4/3}$  plots as mentioned before. All these characteristics are well explained by the shift in the nature of spin fluctuations from the weakly itinerant electron ferromagnet limit to the spin localized magnetic moment limit.

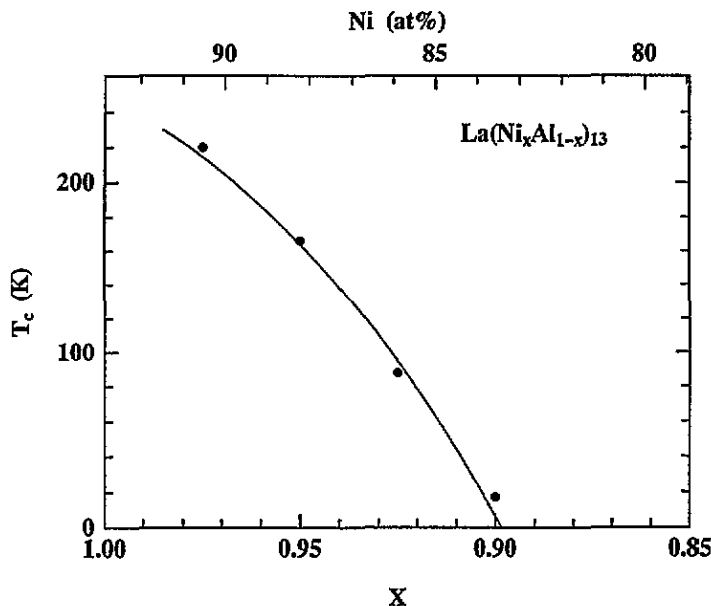


Figure 5. Concentration dependence of the Curie temperature for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys.

To elucidate the variation in characteristics of spin fluctuations for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloy system, the Arrott plots at low temperatures are analysed using the proposed theory (Takahashi 1986). According to this theory, the sum of the thermal and the zero-point spin fluctuations are considered to be almost independent of the temperature. At low temperatures where the thermal spin fluctuations are negligibly small, the magnetization process is expressed by the following equation:

$$-2C \left( \frac{T_C}{T_0} \right)^{4/3} k_B T_A P_s + \frac{4}{15} \frac{k_B T_A^2 P_s^3}{T_0} = 2\mu_B H \quad (1)$$

where the constant  $c$  is 0.3353, and  $k_B$  and  $P_s$  are the Boltzmann constant and the magnetic moment per atom, respectively. The temperatures  $T_A$  and  $T_0$  are correlated with the spectral width of the spin fluctuations in wavenumber and frequency space, respectively, through the following relation:

$$\text{Im}[\chi(q, \omega)] = \chi(q) \frac{\omega \Gamma_q}{\omega^2 + \Gamma_q^2} \quad (2)$$

with

$$\chi(q) = \frac{\kappa^2 \chi(0)}{\kappa^2 + q^2} \quad \kappa^2 \chi(0) = \frac{N_0}{2} \frac{q_B^2}{k_B T_A}$$

and

$$\Gamma_q = 2\pi \frac{k_B T_0}{q_B^3} q(\kappa^2 + q^2)$$

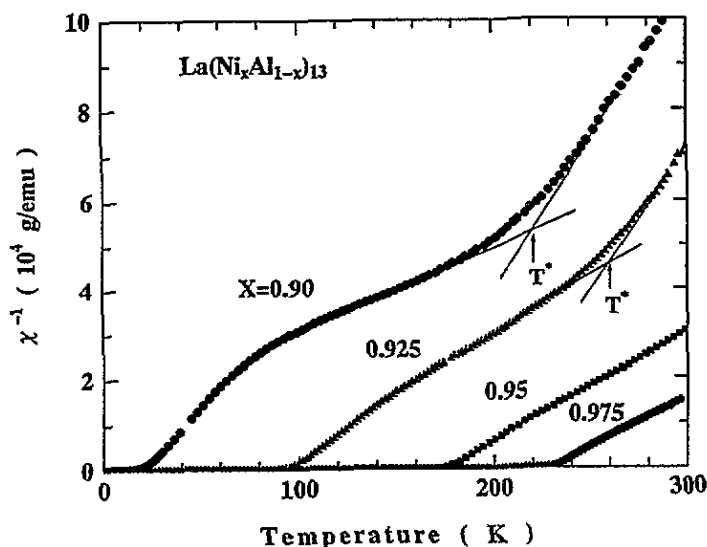
where  $\text{Im}[\chi(q, \omega)]$ ,  $\chi(q)$ ,  $\kappa$  and  $q_B$  are the spectrum of spin fluctuations, the dynamical susceptibility, the magnetic correlation length and the upper limit of the wavenumber, respectively (Takahashi 1986). The spin fluctuations of weakly itinerant electron ferromagnets and of localized magnetic moments are characterized by the spectrum width in the wavenumber and frequency space. In weakly itinerant ferromagnets, the local spin density fluctuates with long wavelength and low frequency, resulting in a narrow width of the spectrum. On the other hand, the spectral width of the localized magnetic moment system is broad, because the local spin density is fixed on each site in real space (Moriya and Kawabata 1973, Takahashi 1986). The temperatures  $T_A$  and  $T_0$  are much higher than  $T_C$  in the limit of weakly itinerant electron ferromagnets, while  $T_C/T_A$  and  $T_C/T_0$  become close to unity as the system shifts to the localized magnetic moment limit (Takahashi 1986). Using equation (1), the values of  $T_A$  and  $T_0$  can be estimated from the slope and the intercept of the Arrott plots, respectively, and the results obtained are listed in table 1. Strictly speaking, these parameters should be determined from microscopic measurements such as inelastic neutron scattering and NMR measurements. Both  $T_A$  and  $T_0$  for crystalline Ni are estimated (Takahashi 1986) from the results of neutron scattering experiments (Böni and Shirane 1985). As shown in table 1, nevertheless, the present estimated values are reasonable, being in agreement with the values for crystalline Ni. The following two points should be noted. First, the magnetization process in the present alloys depends on the spectral width of zero-point spin fluctuations at temperatures where the thermal spin fluctuations are negligibly small (Takahashi 1986). Second, if the correlation between these temperatures and the 3d electron bands is considered (Moriya 1985), the structure of 3d electron bands in the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys may be comparable with that in crystalline Ni. The concentration dependences of  $T_A$  and  $T_0$  indicate that the spectral width of spin fluctuations becomes broader with increasing Ni concentration. Thus, the concentration dependence of  $T_A$  indicates that the characteristics of spin fluctuations disappear from the weakly itinerant electron ferromagnetic limit and belong to the interpolation region as the Ni concentration increases.

The inverse magnetic susceptibility  $\chi^{-1}$  exhibits a unique temperature dependence as shown in figure 6; that is,  $\chi^{-1}$  becomes convex upwards just above  $T_C$  and subsequently undergoes an upturn. It has been pointed out that the  $\chi^{-1}$ - $T$  curve is convex upwards when the longitudinal stiffness is small, and the average of local spin-density fluctuations



**Table 1.** The parameters  $T_A$  and  $T_0$  correlated with the spectral width of spin fluctuations in wavenumber and frequency space, respectively, together with the ratio  $T_C/T_0$  of the Curie temperature to  $T_0$  for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys. The parameters for crystalline Ni deduced from the neutron diffraction measurement (Böni and Shirane 1985, Takahashi 1986) are also shown, for comparison.

|                | $T_A$<br>( $10^4$ K) | $T_0$<br>( $10^3$ K) | $T_C/T_0$ |
|----------------|----------------------|----------------------|-----------|
| $x = 0.90$     | 6.00                 | 3.35                 | 0.004     |
| $x = 0.925$    | 3.59                 | 2.06                 | 0.042     |
| $x = 0.95$     | 2.47                 | 1.54                 | 0.11      |
| $x = 0.975$    | 2.28                 | 1.82                 | 0.12      |
| Crystalline Ni | 1.26                 | 2.66                 | 0.42      |



**Figure 6.** Temperature dependence of the inverse magnetic susceptibility above  $T_C$  for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys.

$S_L^2$  exhibits a marked increase with increasing temperature above  $T_C$  (Moriya and Kawabata 1973, 1974, Moriya 1985). Therefore, the longitudinal stiffness of the local spin density in the present system is small and  $S_L^2$  should rapidly increase with increasing temperature just above  $T_C$ . Moreover, the drastic upturn in the  $\chi^{-1}-T$  curves means that a different temperature dependence of  $S_L^2$  occurs. Such a change may be explained by the saturation of  $S_L^2$  predicted from the SCR theory (Moriya and Takahashi 1978, Takahashi *et al* 1983). Because of the charge neutrality, the magnitude of  $S_L^2$  has an upper limit and it is impossible to exceed this limit even though  $S_L^2$  can increase with increasing temperature just above  $T_C$ . It is known that the saturation of  $S_L^2$  is observed in  $\text{Co}(\text{Se}_x\text{S}_{1-x})_2$  pyrite compounds (Adachi *et al* 1969, Inoue and Yasuoka 1979). These compounds exhibit a break in the slope of the  $\chi^{-1}-T$  curve at the saturation temperature  $T^*$  (Adachi *et al* 1969), because only the transverse fluctuations contribute to the magnetic susceptibility after saturation of  $S_L^2$ . The saturation temperature  $T^*$  indicated by the arrow for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys is estimated from the intersection of the straight lines obtained from the  $\chi^{-1}-T$  curve before

and after the change in the slope. The temperature  $T^*$  slightly increases with increasing Ni concentration. Since the spin fluctuations are influenced by the magnetic field, the field dependence of the  $\chi^{-1}-T$  curve has been investigated. The results for fields of 100 and 200 Oe are given in figure 7. As seen from the figure, the  $\chi^{-1}-T$  curve clearly changes with the magnetic field. That is, in a higher magnetic field, the curvature of  $\chi^{-1}-T$  between  $T_C$  and  $T^*$  is less significant and the upturn in the slope is reduced. The external magnetic field tends to suppress the thermal growth of spin fluctuations (Takeuchi and Masuda 1979), resulting in a change in the  $\chi^{-1}-T$  curve from convex upwards to linear. Needless to say,  $T^*$  is influenced by the magnetic field, stemming from the suppression of the spin fluctuations. Therefore, the change in  $\chi^{-1}$  induced by the magnetic field is one piece of evidence for the existence of correlation between the unique property of  $\chi^{-1}$  and thermal spin fluctuations in the present amorphous alloys.

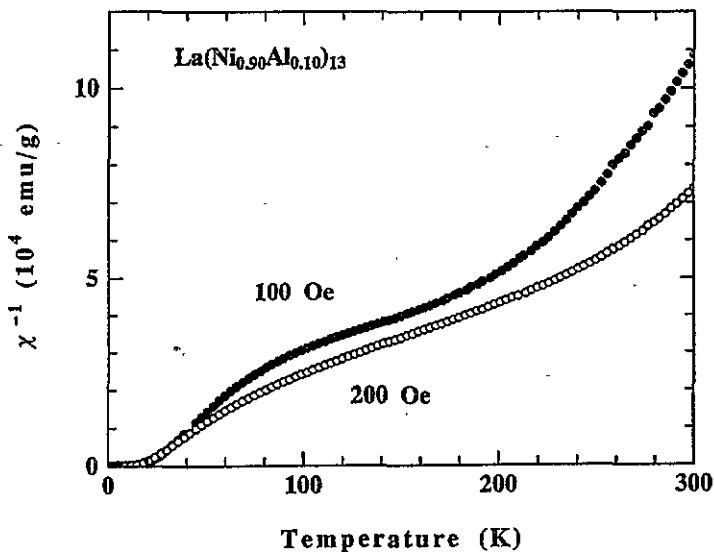


Figure 7. Temperature dependence of the inverse magnetic susceptibility for the amorphous  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  alloy measured at magnetic fields of 100 and 200 Oe.

Figure 8 shows the generalized Rhodes–Wohlfarth (GRW) plots for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys, together with the theoretical line and with the plots of various weakly itinerant electron ferromagnets such as crystalline Pd–Ni alloys,  $\text{ZrZn}_2$  and  $\text{Ni}_3\text{Al}$  (Takahashi 1986). These plots proposed on the basis of the spin fluctuations (Takahashi 1986) are a modification of the Rhodes–Wohlfarth (1963) plot. The abscissa and ordinate are the ratio  $P_{\text{eff}}/P_s$  of the effective moment  $P_{\text{eff}}$  to the saturation moment  $P_s$  and the ratio  $T_C/T_0$ , respectively. The saturation magnetic moment of the amorphous  $\text{La}(\text{Ni}_{0.90}\text{Al}_{0.10})_{13}$  and  $\text{La}(\text{Ni}_{0.925}\text{Al}_{0.075})_{13}$  alloys is deduced from the magnetization curves measured in a pulsed field up to 400 kOe. The effective moment  $P_{\text{eff}}$  for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys is obtained from the straight-line part of the  $\chi^{-1}-T$  curve in a field of 100 Oe just below  $T^*$ . Although the characteristics of the  $\chi^{-1}-T$  curve depend on the external magnetic field, the difference between the slopes of the straight line is small as seen in figure 7. The ratio  $T_C/T_0$  is one parameter affected by the increased ratio of  $S_L^2$  to the temperature above  $T_C$  (Takahashi 1986) and the thermal variation in  $\chi^{-1}$  is directly connected with the increase

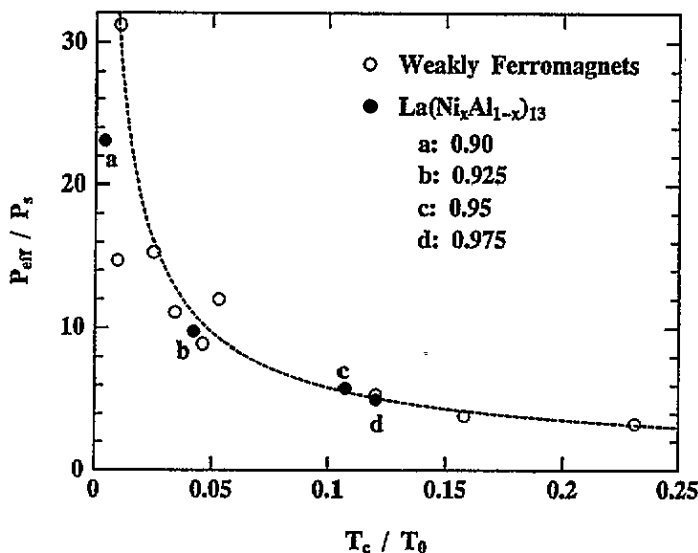


Figure 8. GRW plots for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys, together with the theoretical line and the plots for various weakly ferromagnets in the crystalline state (Takahashi 1986).

in  $S_L^2$  (Moriya and Kawabata 1973). The ratio  $P_{\text{eff}}/P_s$  strongly depends on  $T_c/T_0$  and it has been confirmed that the experimental values for various weakly itinerant electron ferromagnets are situated along the line in the  $P_{\text{eff}}/P_s$ - $T_c/T_0$  plane. The GRW plots for the amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys show a similar tendency to those of the crystalline alloys and compounds mentioned above. Consequently, the GRW plots for the present amorphous alloys indicate that  $P_{\text{eff}}/P_s$  is fairly well correlated with  $T_c/T_0$ ; that is, the variation in  $\chi^{-1}$  above  $T_c$  depends on the thermal variation in  $S_L^2$ . In the low-Ni-concentration region,  $T_c/T_0$  is small and  $\chi^{-1}$ - $T$  is dominated by  $S_L^2$ . Additionally, the directional fluctuations of the local moment contribute to the variation in  $\chi^{-1}$  with increasing Ni concentration, resulting in a decrease in  $P_{\text{eff}}/P_s$ .

Since well defined icosahedral clusters exist, the concentration fluctuations are reduced in the present amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys; discussion on the spin fluctuations leads to a successful interpretation. However, we must not forget that there is some influence from the concentration fluctuations in other amorphous alloys containing no icosahedral clusters (Wagner and Wohlfarth 1982). In this case, both spin fluctuations and concentration fluctuations should be taken into consideration, especially around the critical concentration (Wohlfarth 1983).

#### 4. Conclusion

Spin fluctuations in the weakly itinerant electron ferromagnetic amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys composed of icosahedral clusters have been investigated. The influence of the thermal variation in spin fluctuations has been observed in the thermal variation in magnetization below and above the Curie temperature  $T_c$  and in the magnetization process at 4.2 K. The concentration dependence of spin fluctuations has also been investigated. The main results are summarized as follows.

(a) The temperature dependence of the magnetization exhibits an  $M^2-T^{4/3}$  dependence below the Curie temperature  $T_C$ . This relation is well established in lower-Ni-concentration regions.

(b) The  $M^2-T^{4/3}$  relation becomes poor, while the  $M^2-T^2$  relation holds in a wide temperature range with increasing Ni concentration. Correspondingly, at low temperatures, an  $M-T^{3/2}$  relation due to the spin-wave excitation is well defined in the high-Ni-concentration region.

(c) The inverse magnetic susceptibility  $\chi^{-1}$  curve is convex upwards just above  $T_C$  and exhibits a marked upturn in the slope at high temperatures owing to the saturation of the thermal growth of spin fluctuations.

(d) The magnetization process at 4.2 K indicates that the distribution of spin fluctuations is localized in these spaces in low-Ni-concentration regions, while this distribution becomes broader with increasing Ni concentration.

(e) In the low-Ni-concentration region, the GRW plots indicate that the local spin density is thermally variable and the local spin-density fluctuations dominate the characteristics of  $\chi^{-1}$ . With increasing Ni concentration, the directional fluctuations of the local moment additionally contribute to the thermal variation in  $\chi^{-1}$ .

(f) The magnetic properties of amorphous  $\text{La}(\text{Ni}_x\text{Al}_{1-x})_{13}$  alloys belong to the weakly itinerant electron ferromagnet limit in the low-Ni-concentration regions and shift to the interpolation region with increasing Ni concentration.

## Acknowledgments

We would like to thank Dr T H Chiang, Messrs N Ohashi, M Yoshida and H Yokoyama for their experimental assistance. We also wish to thank Professor E Matsubara of the Faculty of Engineering, Kyoto University, and Professor Y Waseda of the Institute for Advanced Material Processing, Tohoku University, for their useful discussions on structural analyses. The present study has been supported by Research Fellowships of the Japan Society for Promotion of Science for Young Scientists.

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