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Characteristic spin fluctuations in $Y(Mn_{1-x}Al_x)_2$

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Abstract. The thermal expansion and magnetic susceptibility have been measured for $Y(Mn_{1-x}Al_x)_2$ pseudobinary Laves phase intermetallic compounds. A large volume change at T_N and a remarkable enhancement of the thermal expansion coefficient above T_N were observed in YMn_2. The substitution of Al for Mn makes these anomalies less clear. The temperature dependence of the susceptibility, being almost temperature independent for YMn_2, becomes of the Curie–Weiss type as the Al content increases. Such behaviour is explained as the characteristic features of spin fluctuations. The amplitude of local spin fluctuations at the Mn site has been estimated on the basis of the phenomenological theory of magnetovolume effects. It has been revealed that the magnetic ground state of Al-substituted compounds is a spin glass.

1. Introduction

Recent developments in the theory of spin fluctuations give us a unified picture of the magnetism of transition metals and alloys ranging from weakly itinerant electron ferro- or antiferromagnetism to local moment systems (Moriya 1979). One of the most important quantities characterising the magnetism of a particular metal is the amplitude of the local spin fluctuations, S_L, and its temperature dependence. For the local moment limit, S_L remains constant below and above the Curie or the Néel temperature. On the other hand, for a weakly itinerant electron ferromagnet, it varies with temperature as was shown by the self-consistent renormalisation (SCR) theory of spin fluctuations (Moriya and Kawabata 1973a, b). The effects of spin fluctuations are detected by several experimental techniques, such as neutron scattering and NMR. The estimation of S_L, however, is difficult because there is no experimental method to measure S_L directly except inelastic neutron scattering measurements over a wide energy and momentum transfer space, which is hardly attained in the present neutron scattering facilities.

Magnetovolume effects give us fruitful information on the estimation of S_L because longitudinal spin fluctuations give rise to a volume expansion proportional to S_L^2. The negative thermal expansion coefficient observed in weakly itinerant electron ferromagnets and the large spontaneous volume magnetostriction of Invar-type alloys are explained by a reduction of S_L with increasing temperature up to T_C (Shiga and Nakamura 1969, Moriya and Usami 1980). In addition to the reduction of S_L^2 below T_C, the SCR theory predicts a recovery of S_L^2 above T_C, leading to an additional volume expansion or an enhancement of thermal expansion coefficient (TEC) above T_C. Actually, several authors claim the enhancement of TEC above T_C for some weakly itinerant electron ferromagnets, such as

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MnSi (Matsunaga *et al* 1982), $ZrZn_2$ (Ogawa 1983) and Ni_3Al (Suzuki and Masuda 1985). Brommer and Franse (1984) have shown, however, that their analyses depend critically on the estimate of TEC due to anharmonic lattice vibrations and hence the enhancement of TEC is not conclusive. Therefore, it is very desirable to find and investigate a material that shows more distinct spin fluctuations and remarkable magnetovolume effects.

Recently, we have found that the Laves phase intermetallic compound YMn_2 is an antiferromagnet with a Mn moment of $2.7\,\mu_B$ and $T_N \sim 100$ K (Nakamura *et al* 1983). This compound shows a first-order phase transition at T_N with a large temperature hysteresis of 20 K accompanied by a huge volume change of about 5%. Above T_N, the magnetic susceptibility increases with increasing temperature, indicating itinerant electron magnetism. Therefore, the large volume change can be attributed to the volume shrinkage due to a remarkable reduction of Mn moments at T_N. We also expect the recovery of S_L above T_N and hence an enhancement of TEC.

We found that the magnetic character of YMn_2 is highly sensitive to the substitution of a third element (Yoshimura *et al* 1986). For instance the substitution of a small amount of Al for Mn notably reduces the thermal expansion anomaly. We have interpreted this change as the result of a transition from an itinerant electron system to a local moment system (Shiga *et al* 1986).

In this paper we will show a large enhancement of TEC above the Néel temperature in YMn_2 and depression of the enhancement by the substitution of Mn by Al, which leads the system to the local moment limit. By analysing thermal expansion data, we estimated the temperature dependence of the amplitude of local spin fluctuations.

2. Experiment

The samples were prepared by argon arc-melting followed by annealing at 800 °C for seven days. We used 99.9% pure Y, 99.9% pure Mn and 99.99% pure Al as raw materials. No foreign phase other than C15 was detected by x-ray analysis. The magnetic susceptibility was measured by a magnetic torsion balance. The thermal expansion was measured by both a differential transformer-type dilatometer and x-ray diffraction. The disagreement between the results obtained by the two measurements was observed for the Al-free compounds, and this may be due to microcracks introduced by a large volume change at the transition temperature; we therefore employed the temperature dependence of the lattice parameter as the thermal expansion curve for $0 \leqslant x \leqslant 0.1$. Since the x-ray result agreed with the dilatometric curve for $x = 0.1$, dilatometric measurements were used to give the thermal expansion curves of the compounds with $x \geqslant 0.1$. The lattice parameters of YMn_2 at high temperatures were measured partly by neutron diffraction.

3. Results

The concentration dependence of the lattice parameter at room temperature is shown in figure 1. The lattice expands far more rapidly than Vegard's law as x increases from zero. The lattice parameters at 4.2 K, which were measured directly by x-ray diffraction for $x \leqslant 0.05$ and estimated from thermal expansion curves for $x \geqslant 0.1$, are also plotted in figure 1. In this case, the deviation from Vegard's law has been reduced.

The temperature dependence of the magnetic susceptibility (χ–T) curves of

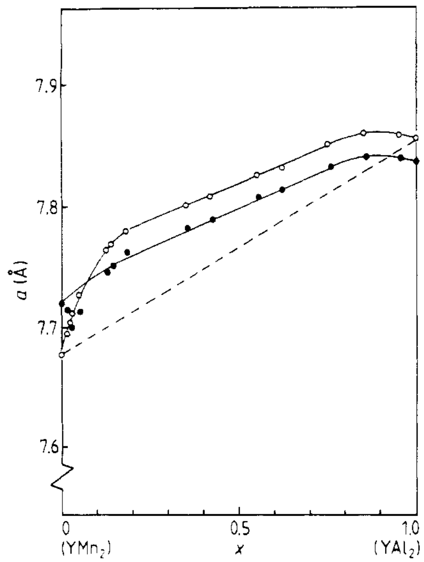

Figure 1. Concentration dependence of the lattice parameter of $Y(Mn_{1-x}Al_x)_2$ at room temperature (open circles) and at 4.2 K (full circles). The broken line indicates Vegard's law at room temperature. The full curves are to guide the eye.

$Y(Mn_{1-x}Al_x)_2$ with $0 \leqslant x \leqslant 0.05$ is shown in figure 2. For YMn_2, a discontinuity is observed around 100 K, where a distinct volume change occurs with a temperature

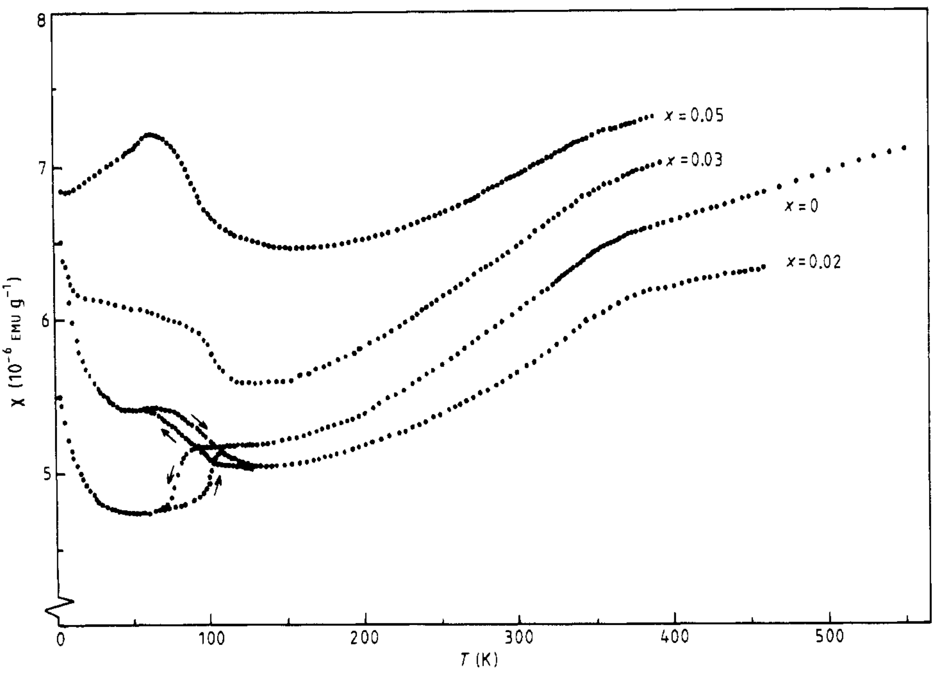

Figure 2. Temperature dependence of the susceptibility of $Y(Mn_{1-x}Al_x)_2$ for $0 \leqslant x \leqslant 0.05$. Arrows indicate heating and cooling processes. $H = 8.28$ kOe.

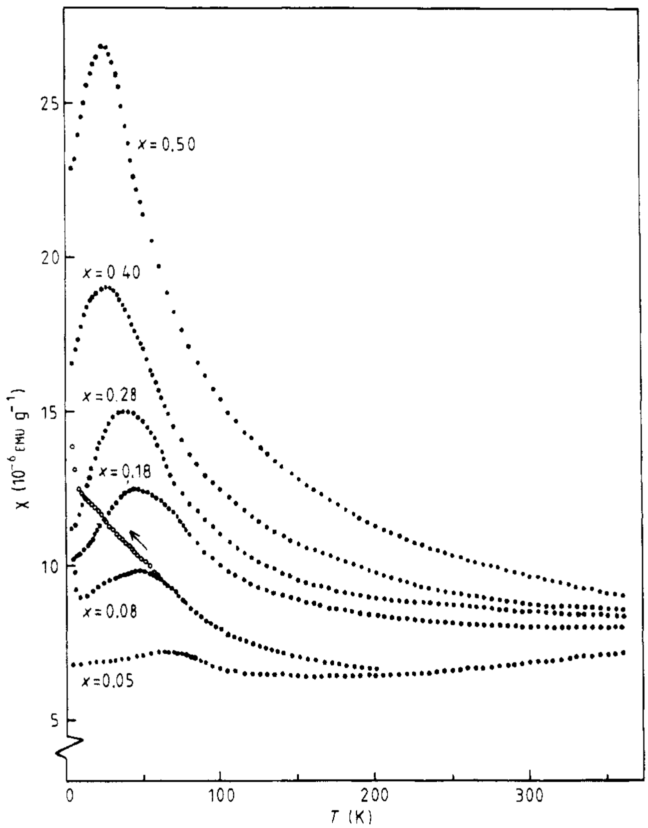

Figure 3. Temperature dependence of the susceptibility of $Y(Mn_{1-x}Al_x)_2$ for $0.05 \leqslant x \leqslant 0.50$. Open circles for $x = 0.08$ indicate a field-cooling process. $H = 8.28$ kOe.

hysteresis of about 20 K. This temperature is the Néel temperature, T_N, where the first-order phase transition takes place. The susceptibility increases with increasing temperature above T_N and shows a trend of saturation at high temperatures. The substitution of a small amount of Al for Mn gives rise to distinct changes in the χ–T curves, particularly around T_N. The temperature hysteresis disappears for $x = 0.03$ and a maximum is observed at 60 K for $x = 0.05$. The maximum becomes more distinct with increasing x, and the overall feature looks like an antiferromagnet with localised moments as seen in figure 3.

Figure 4 shows the temperature dependence of the lattice parameter of YMn_2 up to 900 K. A large volume change is observed at T_N, which may be ascribed to the spontaneous volume magnetostriction. Just above T_N, the temeprature derivative of the curves, that is the thermal expansion coefficient TEC, is very large, being as large as $50 \times 10^{-6} K^{-1}$ at 200 K. At high temperatures, TEC decreases with increasing temperature, in contrast to the usual temperature dependence in metals, and it approaches a normal value.

Thermal expansion curves of $Y(Mn_{1-x}Al_x)_2$ measured by either a dilatometer or x-ray diffraction are shown in figure 5. A substitution of Mn by Al gives rise to dramatic changes in the thermal expansion curves as follows. (i) The volume change at T_N decreases and almost disappears at $x = 0.1$, beyond which the thermal expansion curve does not

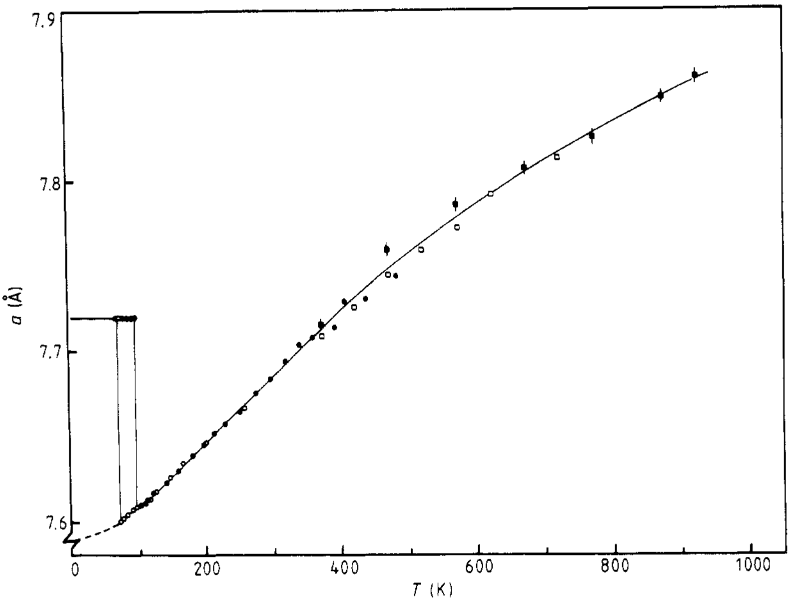

Figure 4. Temperature dependence of the lattice parameter of YMn_2. The broken curve indicates a hypothetical lattice parameter for paramagnetic YMn_2 estimated from $Y_{0.98}Sc_{0.02}Mn_2$ (see text). ● ($T\uparrow$), ○ ($T\downarrow$), x-ray (4, 4,0) Fe K_α; □, x-ray (5, 5, 3) Cu K_α; ▮ neutron (3, 3, 1) $\lambda = 1.418$ Å.

change so much with Al concentration. (ii) For $x = 0.02$, a discontinuity is still observed at T_N, indicating the first-order phase transition, while the variation of the lattice parameter becomes continuous at T_N for $x \geqslant 0.03$, indicating a second-order transition. (iii) TEC above T_N decreases with increasing x and reaches a normal value of about $16 \times 10^{-6}\mathrm{K}^{-1}$ for $x = 0.1$.

In figure 6 the TEC at room temperature is plotted as a function of x. It should be noted that the TEC has normal values of $15 \times 10^{-6}\ \mathrm{K}^{-1}$ for $x > 0.1$ and increases sharply for $x < 0.1$ with decreasing x, indicating an enhancement of TEC in YMn_2.

4. Discussion

4.1. *Effect of spin fluctuations*

The characteristic features of thermomagnetic and thermal expansion curves of $Y(Mn_{1-x}Al_x)_2$ are summarised as follows.

(i) The lattice parameter at room temperature increases more rapidly than Vegard's law, while that at 4.2 K varies nearly linearly with x except for $0.02 \leqslant x \leqslant 0.05$.

(ii) The χ–T curve becomes of the Curie–Weiss type.

(iii) The volume change at T_N, that is the spontaneous volume magnetostriction, decreases with increasing x and disappears for $x > 0.1$.

(iv) The TEC at room temperature decreases sharply with increasing x and reaches a normal value for $x > 0.1$.

We will show that such characteristic behaviour is consistently explained by assuming

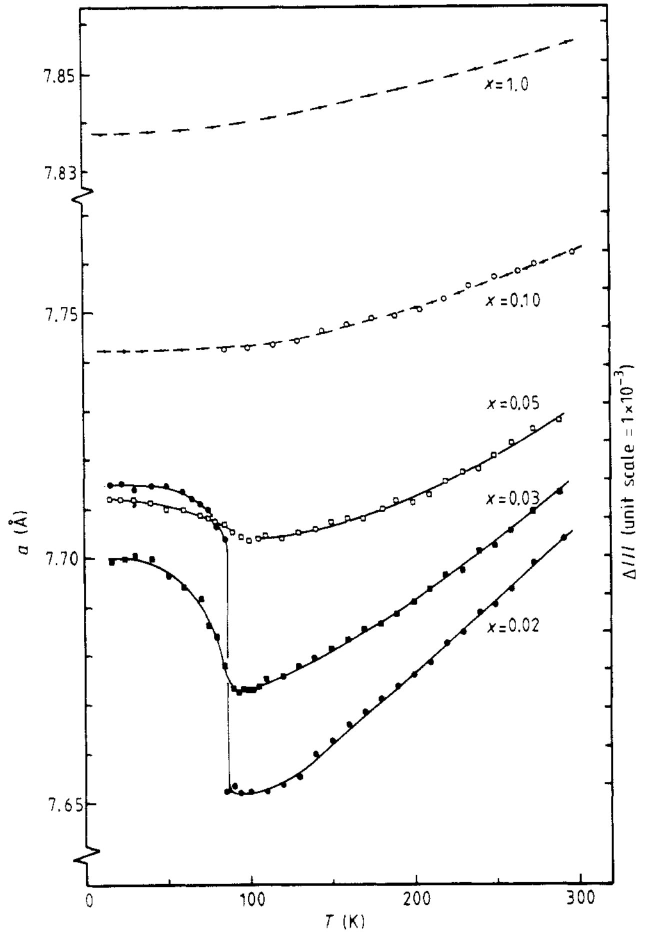

Figure 5. Thermal expansion curves of $Y(Mn_{1-x}Al_x)_2$ for $x \geqslant 0.02$ measured by either x-ray diffraction (heating process) (circles and squares) or a dilatometer (broken curves). Full curves are to guide the eye.

characteristic spin fluctuations in this system and by applying a phenomenological theory of magnetovolume effects (Shiga 1981). In this theory the magnetic volume change ω_m is decomposed into two contributions, $\omega_m = \omega_b + \omega_i$, where ω_b is a band term and ω_i an interaction term. The former term is proportional to the square amplitude of the local spin fluctuations or, in other words, the square of the local moment m_l: $\omega_b = k_b m_l^2$. The interaction term is proportional to the pair correlation function between local moments, namely $\omega_i = k_i \langle m_i m_j \rangle$. However, noting the fact that no thermal expansion anomaly is detected at the spin-glass freezing temperature for $x > 0.1$, where the χ–T curves show a sharp peak, we may neglect the latter contribution as compared with the former. Hereafter, we take into consideration only the band term and hence use a relation $\omega_m = k_b(1-x)m_{Mn}^2$ for the magnetic volume change in this system, where m_{Mn} is the Mn moment and the factor $(1-x)$ is the fractional number of Mn atoms.

Now we discuss how the Mn moment in YMn_2 collapses above T_N. The magnetic volume expansion accompanied by the onset of the local moment of $1\,\mu_B$ is roughly

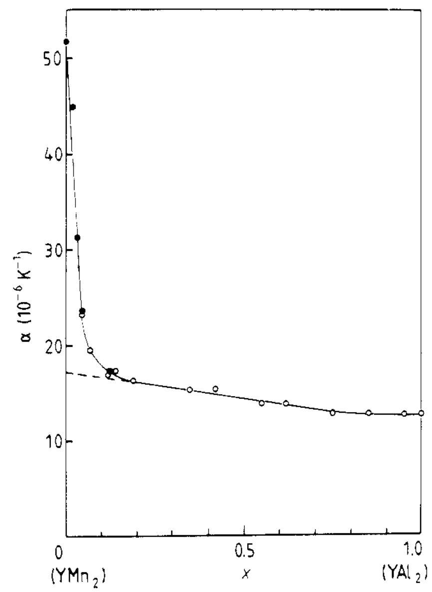

Figure 6. Concentration dependence of the thermal expansion coefficient of $Y(Mn_{1-x}Al_x)_2$ at room temperature. Full circles are determined by x-ray measurements and open circles by dilatometric measurements. The broken line is an extrapolated line from $x > 0.2$.

estimated as 1% for 3d transition metals (Janak and Williams 1976). Therefore, a volume change of about 7% is expected for YMn_2 if the Mn moment of 2.7 μ_B collapses completely above T_N. The observed value of 5% suggests that the Mn moment does not collapse to zero above T_N but remains finite due to spin fluctuations.

According to the SCR theory, the amplitude of spin fluctuations grows with increasing temperature in a weak ferromagnet or antiferromagnet above its Curie or Néel temperature, which means that the magnitude of the local moment m_l increases with increasing temperature. On the other hand, in the local moment limit, fluctuations are saturated at all temperatures and hence m_l becomes constant. Taking these theoretical predictions into consideration, we propose a characteristic feature of spin fluctuations in the $Y(Mn_{1-x}Al_x)_2$ system as shown schematically in figure 7. For YMn_2, m_{Mn}^2 is suddenly reduced at T_N and recovers remarkably above T_N with a trend of saturation at higher temperatures. As Al is substituted for Mn the type of spin fluctuation gradually changes and finally becomes that of the local moment limit around $x = 0.1$. Furthermore, we assume that the Mn moment at 0 K remains nearly constant with varying Al concentration. On the basis of this model, the characteristic features of this system are consistently explained as follows:

(1) *Concentration dependence of the lattice parameter.* We may write the lattice parameter of $Y(Mn_{1-x}Al_x)_2$ as

$$\begin{aligned} a(x) &= a_A(1-x) + a_B x + \omega_m(T) \\ &= a_A(1-x) + a_B x + \tfrac{1}{3}k(1-x)m_{Mn}^2 \end{aligned} \qquad (1)$$

where a_A is the lattice parameter of a hypothetical non-magnetic YMn_2, a_B that of YAl_2 and k the magnetovolume coupling constant (Shiga 1981). As shown by the chain

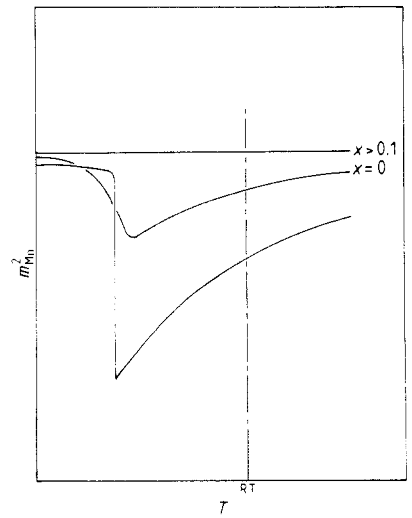

Figure 7. Schematic features of temperature dependence of the magnitude of the Mn moment in the system $Y(Mn_{1-x}Al_x)_2$.

line in figure 7, m^2_{Mn} at room temperature increases sharply with increasing x for $x<0.1$, giving rise to a rapid increase in the lattice parameter. For $x>0.1$, m_{Mn} takes a maximum value of about $3\mu_B$ and the lattice parameter varies linearly. On the other hand, if the Mn moment at 0 K has a constant value for all concentrations, the lattice parameter should obey Vegard's law. In fact, the deviation from Vegard's law is less distinct at 4.2 K. However, in the dilute Al region, $0.02\leqslant x\leqslant 0.05$, we have observed a downward deviation. Noting the fact that the hyperfine field at ^{55}Mn nuclei has the same value in this concentration range as in YMn_2 (Yoshimura *et al* 1986), we ascribe the deviation from Vegard's law not to the decrease of Mn moment but to some unknown factors which violate the relation (1).

(2) *Thermal expansion.* The volume thermal expansion of magnetic materials may be given as

$$\omega(T)=\omega_{\text{lattice}}+\omega_m$$

$$=\omega_{\text{lattice}}+k(1-x)m^2_{Mn}. \tag{2}$$

Since the Mn moment is constant for $x>0.1$, the thermal expansion is ascribed only to anharmonic lattice vibrations and appears normal. For $x<0.1$, the temperature dependence of m^2_{Mn} contributes appreciably to $\omega(T)$. The enhancement of TEC just above T_N for a small x is a result of the recovery of the spin fluctuation amplitude, and the trend of saturation of m^2_{Mn} at high temperatures gives rise to a decrease of TEC as seen in figure 4. The decrease of the volume change at T_N with increasing x is easily understood as a result of the decrease of the change in m^2_{Mn} at T_N.

(3) *χ–T curves.* The trend that the χ–T curve approaches the Curie–Weiss type with increasing x is in accordance with the present model, assuming a transition from itinerant antiferromagnetism to the local moment limit.

(4) NMR. The resonance frequency of zero-field NMR of ^{55}Mn at 1.4 K remains constant around 110 MHz with increasing x (Yoshimura *et al* 1986), indicating that the Mn moment is constant at low temperatures. The intensity of the signal decreases slightly with increasing x. The rate of decrease, however, is much slower than that of the systems $Y(Mn_{1-x}Co_x)_2$ and $Y(Mn_{1-x}Fe_x)_2$, where the Mn moment collapses on substitution of Co or Fe. Furthermore, no paramagnetic NMR signal in an applied field was detected in the system Y(Mn–Al)$_2$, in contrast to the system Y(Mn–Co)$_2$ or Y(Mn–Fe)$_2$ where the signal has clearly been observed around the frequency for the Knight shift of ^{55}Mn, $^{55}K=0$, indicating the existence of non-magnetic Mn atoms. These results provide microscopic evidence that the Mn moment remains constant at 0 K for substitution of Mn by Al.

4.2. Estimation of m_{Mn}^2

We estimate the amplitude of local spin fluctuations at the Mn site, m_{Mn}, by analysing the thermal expansion curve and the concentration dependence of the lattice parameter on the basis of the phenomenological theory of magnetovolume effects (Shiga 1981).

The lattice parameter for an Al fraction x at a given temperature T can be given by

$$a(x, T)=a^0(x, 0)+\Delta a(x, T)+\tfrac{1}{3}k(1-x)m_{\mathrm{Mn}}^2 \qquad (3)$$

where $a^0(x, 0)$ represents the lattice parameter at 0 K in the hypothetical non-magnetic state (that is, $m_{\mathrm{Mn}}^2=0$), $\Delta a(x, T)$ is the thermal expansion due to lattice vibrations and k is the magnetovolume coupling constant. Each term was estimated as follows. For $a^0(x, 0)$, we write Vegard's law as

$$a^0(x, 0)=a^0(0. 0)(1-x)+a^0(1, 0)x. \qquad (4)$$

In order to estimate $a^0(0, 0)$, we used the thermal expansion curve of $Y_{0.98}Sc_{0.02}Mn_2$, which is in a non-magnetic state even at 4.2 K (Wada *et al* 1987). The temperature dependence of the lattice parameter above 100 K is almost the same for YMn_2 and $Y_{0.98}Sc_{0.02}Mn_2$. Then we extrapolated the thermal expansion curve of YMn_2 above T_N to 0 K using that of $Y_{0.98}Sc_{0.02}Mn_2$ as shown by a broken curve in figure 4. This procedure enables us to estimate the spontaneous volume magnetostriction at 0 K, $\omega_s(0)$, of YMn_2 and the coupling constant k as

$$\omega_s(0)=3(a(0, 0)-a^0(0, 0))/a^0((0, 0)=k(2.7\,\mu_B)^2.$$

We have $\omega_s(0)=5.1\times10^{-2}$ and $k=7\times10^{-3}/\mu_B^2$. $a^0(1, 0)$ was estimated from the lattice parameter at room temperature and the thermal expansion curve of YAl_2. For an estimation of $\Delta a(x, T)$, we assume that the Mn moment has its full T-independent value over the whole temperature range in the Al-rich region as discussed above and, therefore, the thermal expansion is due only to lattice vibrations for $x>0.2$. For $x<0.2$, we assume that the unenhanced TEC at room temperature lies on the extrapolated line from $x>0.2$ as shown by the broken line in figure 6. Using these values and the Debye model, we estimated $\Delta a(x, T)$ for $x<0.2$. These procedures are shown schematically in figure 8. The estimated Mn moments are plotted against temperature in figure 9 for $0\leqslant x\leqslant 0.30$. The Mn moment at low temperatures has an x-independent value of $2.7\pm0.3\,\mu_B$, which is consistent with the NMR results. It should be noted that the Mn moment in YMn_2 is reduced to about 1 μ_B just above T_N and recovers to 2 μ_B at room temperature. This value of 2 μ_B at room temperature agrees with a preliminary result of neutron scattering on YMn_2 (Deportes *et al* 1987).

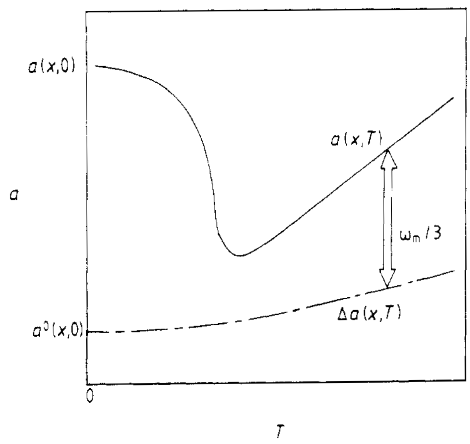

Figure 8. Illustrative figure showing the procedure for estimating the magnetic volume expansion ω_m. $a(x, T)$ means the lattice parameter of $Y(Mn_{1-x}Al_x)_2$ at the temperature T. $a^0(x, 0)$ is the lattice parameter of $Y(Mn_{1-x}Al_x)_2$ in a hypothetical paramagnetic state. $\Delta a^0(x, T)$ (chain curve) indicates the thermal expansion curve due to anharmonic lattice vibrations.

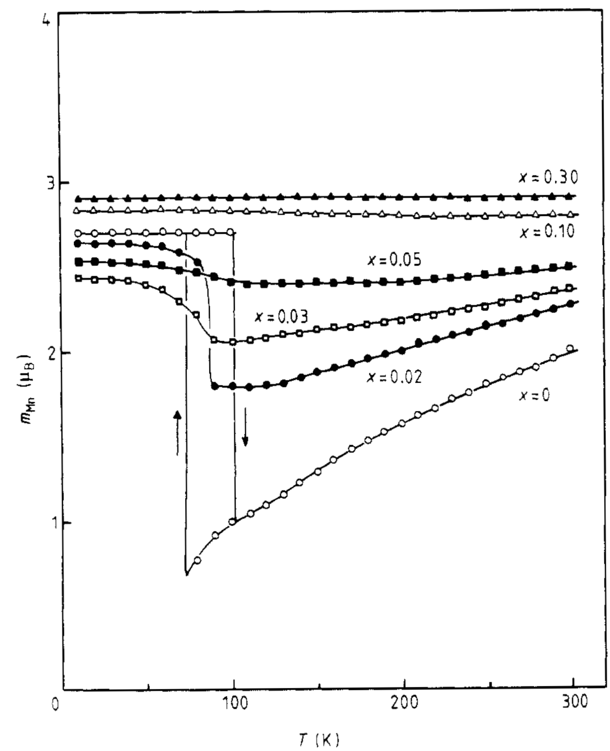

Figure 9. Temperature dependence of the Mn local moment, m_{Mn}, in the system $Y(Mn_{1-x}Al_x)_2$ estimated from the analyses of thermal expansion curves.

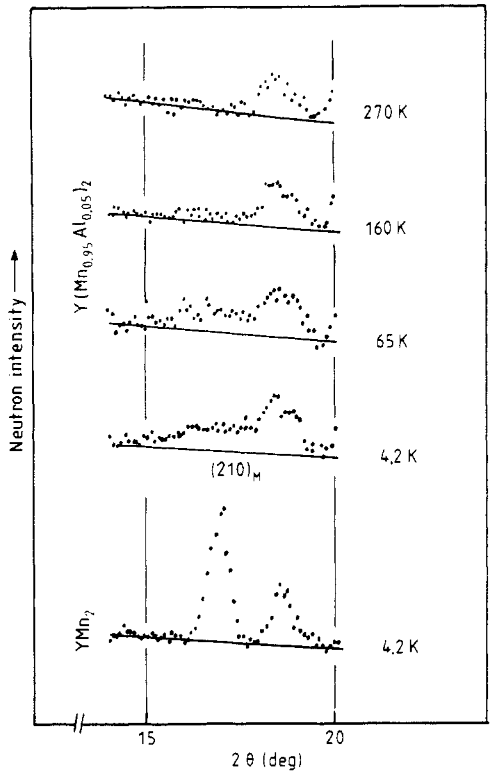

Figure 10. Neutron diffraction patterns of YMn_2 and $Y(Mn_{0.95}Al_{0.05})_2$ around the (210) magnetic Bragg peak.

4.3. Magnetic structure at low temperatures

So far, we have shown that the Mn moment is stabilised by the substitution of Al for Mn but have not mentioned the ground-state spin structure. The sharp peak in the χ–T curve implies an antiferromagnetic spin ordering for $x \geqslant 0.05$. We have carried out neutron diffraction experiments for $Y(Mn_{0.95}Al_{0.05})_2$ at several temperatures. The spectra around the (210) magnetic peak are shown in figure 10. First, it should be noted that the intensity of the magnetic peak is notably reduced and becomes diffuse on the addition of Al, implying the collapse of a long-range antiferromagnetic order. Second, the broad magnetic 'peak' does not disappear at the temperature of susceptibility maximum, T_M, but persists far above T_M as seen in figure 11, which shows the temperature dependence of the intensity of the magnetic 'peak' as well as that of the susceptibility. Furthermore, we have found that the maximum in the χ–T curve disappears with field cooling as shown in figure 3. From these facts, it is likely that the 'peak' in the χ–T curves is due to spin-glass freezing. On the other hand, the intensity of the diffuse magnetic peak seems to correlate with the thermal expansion anomaly and hence the spontaneous volume magnetostriction as seen in figure 11. Although a microscopic picture of the magnetic structure is not clear for the Al-substituted system, these observations suggest that the YMn_2-type antiferromagnetism, whose Néel temperature is nearly the same as that of pure YMn_2, still coexists in this

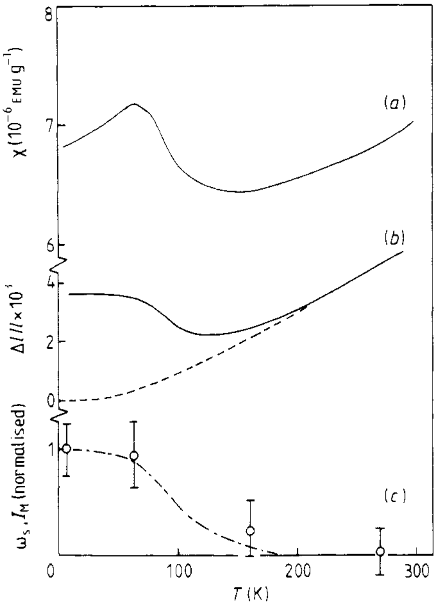

Figure 11. Temperature dependence of (*a*) the susceptibility (*b*) the thermal expansion and (*c*) the intensity of the magnetic Bragg peak (open circles) of $Y(Mn_{0.95}Al_{0.05})_2$. The chain curve in (*c*) indicates the spontaneous volume magnetostriction, ω_s.

sample as fine clusters. Recently, Motoya (1986) has made neutron diffraction and susceptibility measurements on the same system with different values of x. He also observed a collapse of the magnetic Bragg peaks and irreversibility in the thermomagnetic curves.

4.4. *Effect of Al substitution*

It is an interesting problem why the substitution of Al stabilises the Mn moment. The expansion of the lattice may explain this, because it makes the band width narrower, leading to a higher density of states. On the other hand, the substitution of Fe, Co or Ni for Mn, or Sc for Y, which causes a contraction of the lattice, may make the Mn atom non-magnetic.

Another possible reason would be that substituted Al atoms cut 3d–3d bonds between Mn atoms because Al has no 3d state near the Fermi level, which causes the local 3d band width to become narrower. We cannot conclude which mechanism is dominant in the present case.

5. Concluding remarks

(i) It has been revealed that the Mn moment in the antiferromagnetic YMn_2 collapses at T_N, which gives rise to a large volume contraction, and it recovers rapidly with increasing temperature, resulting in a distinct enhancement of thermal expansion coefficient.

(ii) When Al is substituted for Mn, the system approaches the local moment limit and thermal expansion anomalies disappear.

(iii) The amplitude of the local spin fluctuations was estimated as a function of temperature by analysing the thermal expansion curve.

(iv) The ground state of Al-substituted compounds has been found to be a spin-glass state.

Finally, we emphasise that the variations of amplitude of spin fluctuations with temperature and concentration are remarkable in the present system. Therefore, it is highly desirable to carry out other experiments such as neutron scattering to detect spin fluctuations more directly.

Acknowledgments

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