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The pseudogap and anisotropic thermal expansion in RMn_4Al_8 (R = La, Y, Lu and Sc)

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Abstract

The temperature dependence of the magnetic susceptibility shows a broad maximum at ∼550 and 630 K for $LuMn_4Al_8$ and $ScMn_4Al_8$, respectively, which can be interpreted as due to the presence of a pseudogap in the effective bands as in $LaMn_4Al_8$ and YMn_4Al_8. The anisotropic thermal expansion observed for RMn_4Al_8 (R = La, Y, Lu and Sc) and the sensitive volume dependence of the gap width throughout the RMn_4Al_8 system suggest dominant magnetic coupling in Mn spin chains along the *c* axis.

1. Introduction

The effect of geometry on the magnetism and the spin gap formation in magnets have been extensively investigated in recent decades. We have recently reported that $(La_{1-x}Y_x)Mn_4Al_8$ ($0 \leqslant x \leqslant 1$) is a very unique itinerant electron system, which shows a strongly concentration-sensitive broad maximum in the temperature dependence of the susceptibility [1]. To explain this behaviour, a pseudogap model, which assumes a partially filled gap in the spin excitation spectrum (see the inset of figure 2), was applied by referring to a well-known square band model proposed for FeSi [2]. In this case, the temperature of the maximum, T_{max}, roughly corresponds to the pseudogap width, Δ. This pseudogap model was originally applied to YMn_4Al_8 to explain the activation type temperature dependence of the nuclear spin–lattice relaxation rate observed in a high temperature range [3], which is a rare example for itinerant electron magnets. The strong concentration dependence of T_{max}, i.e. Δ, was interpreted as due to a (chemical) pressure or volume effect on the electronic structure. We also emphasized the characteristics of the Mn spin arrangement, which has a quasi-one-dimensional (1D) nature, and proposed that the origin of the gap formation is associated with the characteristic geometry. In the present study, to confirm whether the sensitive Δ is only due to a pressure or volume effect, we extend measurements to $LuMn_4Al_8$ and $ScMn_4Al_8$ with lattice volumes smaller than those of $(La_{1-x}Y_x)Mn_4Al_8$; the lattice parameters at room temperature are listed in table 1 [4–6]. If the 1D magnetic interaction is responsible for the gap formation, we expect anisotropic properties. Hence we measured temperature dependences of the lattice parameters

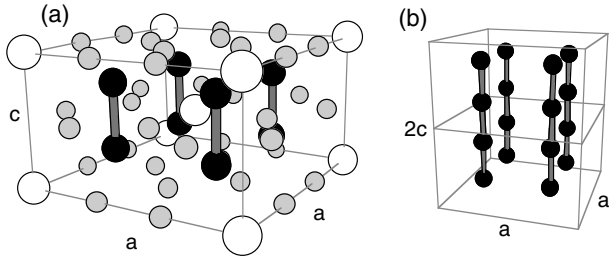

Figure 1. (a) The $CeMn_4Al_8$ type structure. White, black and grey marks represent Ce, Mn and Al atoms, respectively. (b) The Mn sublattice doubled along the *c* axis.

Table 1. Lattice parameters and pseudogap widths of RMn_4Al_8 (R = La, Y, Lu and Sc).

	a (Å)	*c* (Å)	d_{Mn-Mn} (Å)	Δ (K)
$LaMn_4Al_8$	9.031[a]	5.166[a]	2.583	250[d]
YMn_4Al_8	8.856[a]	5.103[a]	2.552	500[d]
$LuMn_4Al_8$	8.814[b]	5.083[b]	2.542	610
$ScMn_4Al_8$	8.7734[c]	5.0467[c]	2.5234	700

[a] From [4], [b] from [5], [c] from [6], [d] from [1].

for RMn_4Al_8 with R = La, Y, Lu and Sc using low temperature x-ray diffraction, which can give information on the anisotropy in polycrystalline samples.

The characteristics of RMn_4Al_8 are summarized as follows.

(1) The crystal structure is of tetragonal $CeMn_4Al_8$ type (space group $I4/mmm$; see figure 1(a)) derived from the $ThMn_{12}$ type, in which Mn atoms at the 8f site form linear chains along the *c* axis (see figure 1(b)); the intrachain Mn–Mn distance d_{Mn-Mn} (=2.5–2.6 Å) is much larger than the interchain distances 4.4–4.5 Å [4].
(2) $LaMn_4Al_8$ and YMn_4Al_8 show a relatively large electronic specific heat coefficient [7], indicating strong electron correlations; results for $LuMn_4Al_8$ and $ScMn_4Al_8$ were not found.
(3) In $(La_{1-x}Y_x)Mn_4Al_8$, the spin pseudogap can be controlled continuously and nearly uniformly over a wide range of Δ = 250–500 K by applying chemical pressure which induces anisotropic volume change [1].
(4) RMn_4Al_8 (R = La, Y, Lu and Sc) compounds basically do not show magnetic long range ordering; specific heat measurements indicate no phase transition for $LaMn_4Al_8$ and YMn_4Al_8 [7]. Recent experiments, however, suggested the development of magnetic correlations below ∼50 K [8, 9] and the appearance of static internal fields below ∼4 K [10] in $LaMn_4Al_8$.

2. Experimental procedures

Polycrystalline samples of RMn_4Al_8 were prepared by arc melting under an Ar atmosphere and annealed at 800 °C for one week in evacuated quartz tubes. The purities of starting materials, R (La, Y, Lu and Sc), Mn and Al, were 99.9, 99.98 and 99.99%, respectively. The susceptibility was measured by using a SQUID magnetometer (Quantum Design, MPMS-5) between 5 and 800 K at 10 kOe. Temperature dependences of lattice parameters between 10 and 300 K were

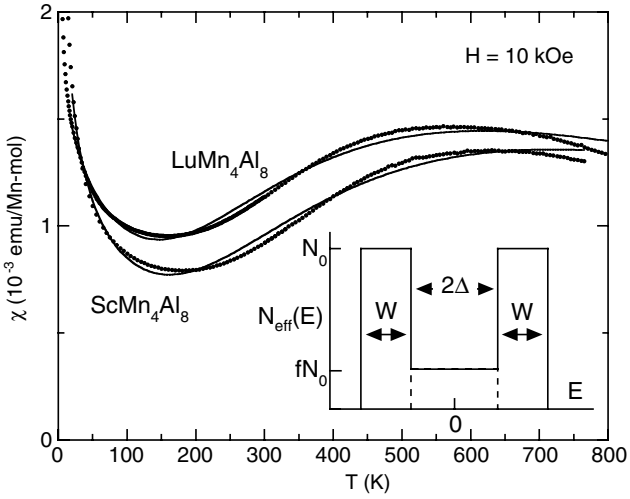

Figure 2. Temperature dependences of the susceptibility for $LuMn_4Al_8$ and $ScMn_4Al_8$. The inset shows the effective density of states assumed for the analysis. Solid curves represent the best fit with constraints $W = 2\Delta$ and $f = 25\%$.

measured using conventional x-ray powder diffraction with Cu Kα radiation. The Rietveld refinement was performed by using RIETAN2000 [11] to obtain lattice parameters.

3. Experimental results

3.1. Susceptibility

Figure 2 shows temperature dependences of the susceptibility measured for $LuMn_4Al_8$ and $ScMn_4Al_8$. Broad maxima were observed at $T_{max} \simeq 550$ and 630 K for $LuMn_4Al_8$ and $ScMn_4Al_8$, respectively, as in the cases of $LaMn_4Al_8$ and YMn_4Al_8 [3, 8, 12]. Upturns at low temperatures were seen, as in the cases of $(La_{1-x}Y_x)Mn_4Al_8$ [1]. Although these upturns may have arisen from an intrinsic origin partially [8, 9], in the cases of $LuMn_4Al_8$ and $ScMn_4Al_8$, it is likely that the upturns come from paramagnetic impurities and/or recovered spins near defects.

To analyse the results of the susceptibility, we apply the same pseudogap model as was used for $(La_{1-x}Y_x)Mn_4Al_8$ [1, 3, 8]. The model of the effective density of states (DOS), $N_{\rm eff}(E)$, is shown as the inset in figure 2, where W, Δ and f represent the bandwidth, the gap width and the relative fraction of the filled part in the gap, respectively. We assume thermal excitations in simple square bands separated by a partially filled gap, where the Fermi level, $E_{\rm F}$, is taken at the centre of the pseudogap. The experimental susceptibility was reproduced with two components as

$$\chi = -2\mu_{\rm B}^2 \int N_{\rm eff}(E) \frac{\partial f(E,T)}{\partial E}\,{\rm d}E + \frac{C}{T-\theta} \tag{1}$$

where $\mu_{\rm B}$ is Bohr magneton and $f(E,T)$ is Fermi distribution function. The second term, with C and θ fitting parameters, describes the low temperature upturn. The maximal density of states N_0 in $N_{\rm eff}(E)$ was also treated as a fitting parameter. In analysing the results for $(La_{1-x}Y_x)Mn_4Al_8$ [1], we assumed $W = 2\Delta$ and $f = 25\%$ since we obtained a common relations $W \simeq 2\Delta$ and $f \simeq 25\%$ for all the compounds by treating all the parameters as

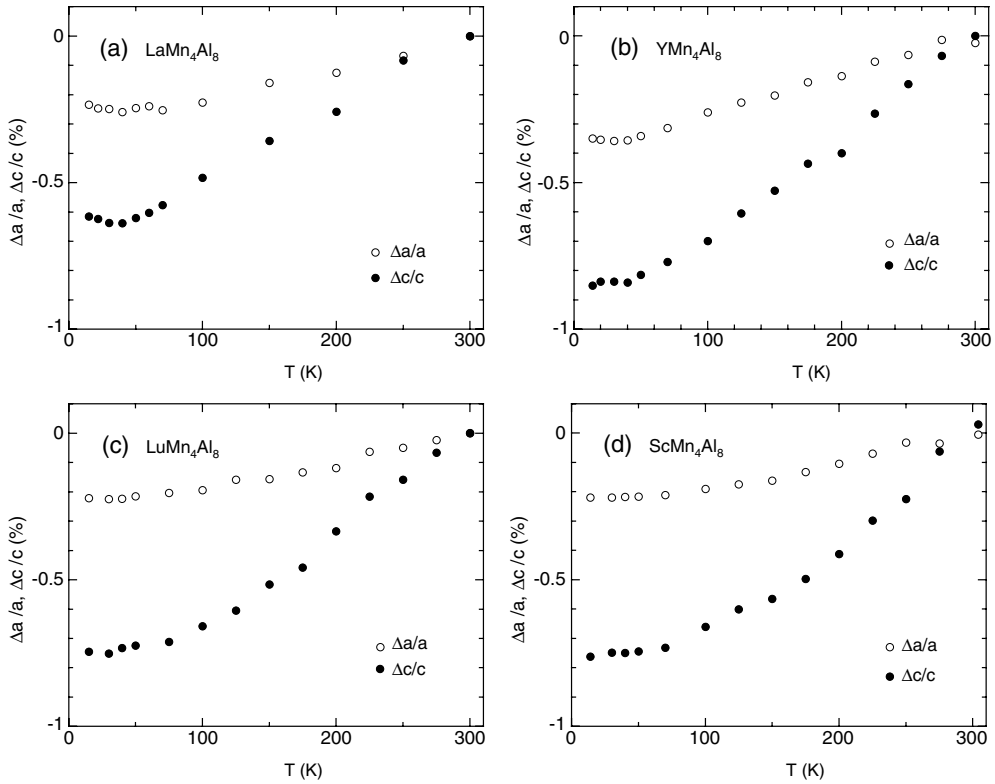

Figure 3. Relative temperature variations of lattice parameters for RMn_4Al_8 with R = La (a), Y (b), Lu (c) and Sc (d).

free variables. For $LuMn_4Al_8$ and $ScMn_4Al_8$, the relations $W \simeq 2\Delta$ and $f \simeq 25\%$ do not necessarily hold. The final refinement was, however, performed by imposing the constraints $W = 2\Delta$ and $f = 25\%$ to reduce the number of free parameters and to obtain approximate values of the characteristic energy for the RMn_4Al_8 compounds. Solid curves in figure 2 indicate best-fit results thus obtained. The values of Δ obtained are listed in table 1.

3.2. Thermal expansion

Figures 3(a), (b), (c) and (d) show thermal expansion curves measured for RMn_4Al_8 with R = La, Y, Lu and Sc, respectively. Relative temperature variations of lattice parameters, $\Delta a/a = [a(T) - a(300\ \mathrm{K})]/a(300\ \mathrm{K})$ and $\Delta c/c = [c(T) - c(300\ \mathrm{K})]/c(300\ \mathrm{K})$ are plotted against temperature. Anisotropic behaviours were commonly observed for all the compounds; the thermal expansion along the c axis is 2–4 times larger than that along the a axis. This fact is consistent with the interpretation that the magnetism in RMn_4Al_8 is dominated by the intrachain interaction. In the previous paper [8], we reported the averaged thermal expansion of polycrystalline $LaMn_4Al_8$ and YMn_4Al_8 measured by a strain gauge method. Averaged values of the present results agree quantitatively with the polycrystalline thermal expansion. For $LaMn_4Al_8$, both a and c increase with decreasing temperature below ∼40 K. This negative thermal expansion corresponds to the development of certain magnetic correlations at low temperatures [8].

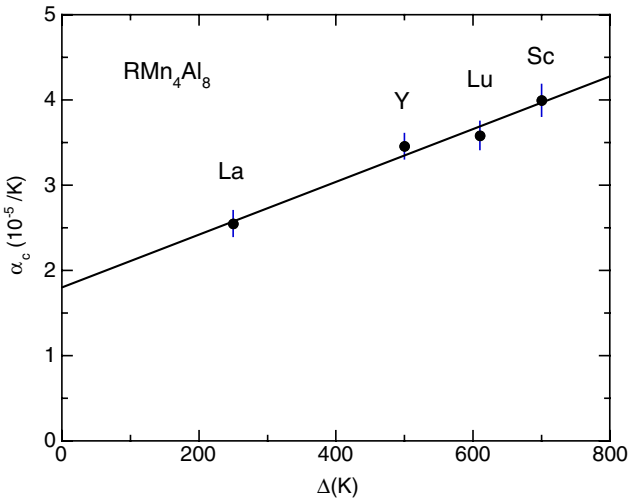

Figure 4. Thermal expansion coefficients along the c axis plotted against the pseudogap width Δ.

Focusing on temperature dependences of c, there is an apparent correlation between the temperature derivative and the characteristic energy. In figure 4, $\alpha_c = (1/c)(\mathrm{d}c/\mathrm{d}T)$, estimated roughly at around room temperature, is plotted as a function of Δ. The variation of α_c is mainly ascribed to a magnetic origin. One of the characteristics of itinerant electron magnets is soft or temperature-induced longitudinal spin fluctuations. The smaller α_c for $LaMn_4Al_8$ implies that the spin fluctuations are nearly saturated at room temperature, while the larger α_c means strong recovery of spins at around room temperature. The Δ dependence of α_c is thus a reasonable consequence for the soft spin gap system.

4. Discussion

It is surprising that the pseudogap width in RMn_4Al_8 can be controlled continuously over the wide range of $\Delta = 250$–700 K. In discussing the magnetovolume effect in RMn_4Al_8, the intrachain Mn–Mn distance $d_{\mathrm{Mn-Mn}}$ $(=c/2)$ is appropriate as the scale rather than the lattice volume. Figure 5 shows Δ plotted against $d_{\mathrm{Mn-Mn}}$ at room temperature. In the first approximation, it is reasonable to ascribe the variation of Δ entirely to the volume effect. In general, for classical 3d bands, we expect a volume effect $W = d^{-5}$, where d is the interatomic distance between 3d atoms [13]. A tentative fit of the data in figure 5 to $\Delta \propto d_{\mathrm{Mn-Mn}}^{-\Gamma}$ gives $\Gamma = -\ln\Delta/\ln d_{\mathrm{Mn-Mn}} = 48 \pm 3$. Noting the approximate relation $\Delta \propto W$ for RMn_4Al_8, the magnitude larger by one order than the classical expectation suggests that the electronic structure around E_{F} is not dominated by classical interactions, and that the gap is not an accidental consequence of the one-electron bare bands. This strong $d_{\mathrm{Mn-Mn}}$ dependence of Δ supports the idea that the 1D instability leads to the formation of the spin singlet-like state at low temperature. It is interesting to note that other 3d correlated electron systems, FeSi and $FeSb_2$, which show gap type temperature dependences of the susceptibility, have similar large values of Γ [14] and the anisotropic thermal expansion [15], respectively.

In conclusion, we observed anisotropic thermal expansion for RMn_4Al_8. This result supports the idea that the magnetic interaction in RMn_4Al_8 is strongly anisotropic. In the RMn_4Al_8 system, the pseudogap can be controlled continuously over a very wide range

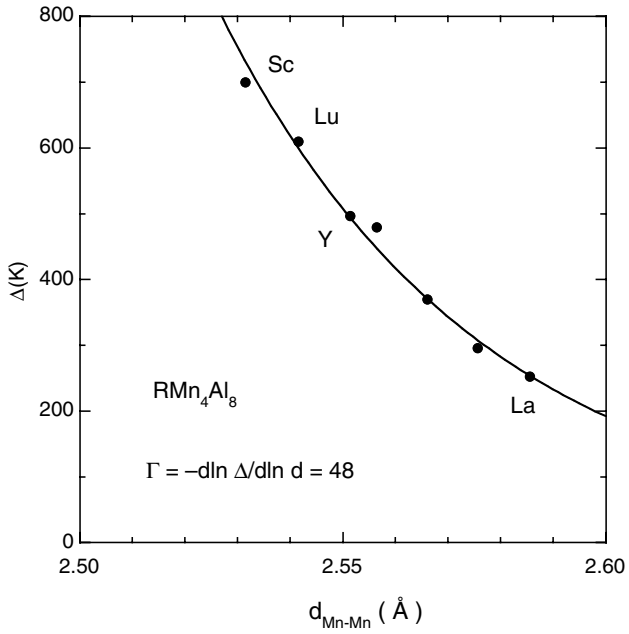

Figure 5. The width of the pseudogap estimated for RMn_4Al_8 (R = $(La_{1-x}Y_x)$, Lu and Sc) plotted against the intrachain Mn–Mn distance. Results for $(La_{1-x}Y_x)Mn_4Al_8$ were cited from [1].

of Δ = 250–700 K. The origin of the pseudogap may be associated with the quasi-1D arrangement of Mn spins.

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