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Itinerant electron metamagnetism and a large decrease in the electronic specific heat coefficient of Laves-phase compounds $Lu(Co_{1-x}Ga_x)_2$

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Received 14 September 1992, in final form 10 November 1992

Abstract. High-field magnetization and low-temperature specific heat of the nearly ferromagnetic compounds $Lu(Co_{1-x}Ga_x)_2$ have been investigated. A sharp metamagnetic transition has been observed in these compounds. The critical field H_c decreases with increasing x and becomes lower than 1 T at x = 0.12. However, no spontaneous magnetization is confirmed even above x = 0.15. With increasing x, the electronic specific heat coefficient γ increases and reaches 36 mJ K⁻² mol⁻¹ for x = 0.12 in 0 T, being 1.5 times as large as that of LuCo₂. Such a mass enhancement is discussed in terms of the spin fluctuation. The γ value of the ferromagnetic state for the specimen with x = 0.12 in a magnetic field of 14.6 T is reduced about 50% compared with that of the paramagnetic state in zero field. This means that the metamagnetic transition results in a drastic reduction of the spin fluctuation in the compound.

1. Introduction

Recently, the mass enhancement of itinerant electrons due to spin fluctuations in 3d and 4f compounds has been studied intensively. Co-based Laves-phase compounds such as YCo₂ and LuCo₂ have been regarded as strongly exchangeenhanced paramagnets (Lemaire 1966, Givord and Lemaire 1971). The electronic specific heat coefficient γ and the magnetic susceptibility χ of these compounds are several times as large as those of an unenhanced Pauli paramagnet YNi₂ (Bloch *et al* 1971, Burzo and Laforest 1972). These properties in exchange-enhanced paramagnets have been discussed in terms of the spin fluctuation (Konno and Moriya 1987).

LuCo₂ has been expected to exhibit an itinerant electron metamagnetic transition in a high magnetic field (Schinkel 1978, Yamada *et al* 1987). In fact, such a metamagnetic transition has been confirmed in 74 T (Goto *et al* 1989, 1990, Sakakibara *et al* 1990b). The magnetic moments have been estimated to be 0.15 and 0.65 $\mu_{\rm B}$ /Co in the paramagnetic and ferromagnetic states, respectively (Goto *et al* 1990). By a partial replacement of Co by other elements such as Al, Sn and so on, the critical field of the metamagnetic transition is reduced. Eventually, ferromagnetism has been established beyond x = 0.10 for Lu(Co_{1-x}Al_x)₂ (Shinogi *et al* 1987, Sakakibara *et al* 1987). The itinerant electron metamagnetic transition in Lu(Co_{1-x}Al_x)₂ has been investigated by several authors. The observed metamagnetic transition, however, is broad because of the magnetic heterogeneity of Co atoms (Shinogi *et al* 1987, Sakakibara *et al* 1987). On the other hand, recently, pseudo-binary compounds Lu(Co_{1-x}Ga_x)₂ and Lu(Co_{1-x}Sn_x)₂ have been developed (Murata *et al* 1991). A clear metamagnetic transition in a relatively low field and a remarkable enhancement of exchange interaction have been confirmed in these compounds.

The magnetic field effect on the spin fluctuation has been discussed by many authors (Brinkman and Engelsberg 1968, Béal-Monod *et al* 1968, Ueda 1976). It has been suggested that huge magnetic fields are required to reduce the electronic specific heat coefficient γ , related to the spin fluctuation (Brinkman and Engelsberg 1968). Remarkable magnetic field effects are expected in two systems of Lu(Co_{1-x}Ga_x)₂ and Lu(Co_{1-x}Sn_x)₂ compounds, because they have a high γ value and the former has a low metamagnetic transition field. Therefore, these two compounds are good candidates for investigations on the suppression of the spin fluctuation caused by high magnetic fields. In the present paper, we have measured low-temperature specific heats of Lu(Co_{1-x}Ga_x)₂ and Lu(Co_{1-x}Sn_x)₂ in magnetic fields up to 14.6 T and have discussed the metamagnetism and the suppression of the spin fluctuation.

2. Experimental details

The alloying was done by arc-melting in an argon gas atmosphere. The Lu content was kept slightly higher than stoichiometry because some loss occurs during alloying owing to vaporization and oxidization of Lu. $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Sn_x)_2$ compounds were annealed at 1073 K for a week in a vacuum quartz tube for homogenization. The specimens were powdered with a particle size of less than 50 μ m to eliminate the eddy current effect in pulsed high magnetic fields. High magnetic fields up to 42 T were generated with a pulse magnet. Ultra-high magnetic fields up to 105 T were generated by a fast capacitor discharge into a copper singleturn coil with a 100 kJ capacitor bank (Nakao et al 1985). The magnetization was measured by an induction method with well balanced pick-up coils. Details of the experimental procedure in high fields have already been described (Sakakibara et al 1990a). The low-temperature specific heat was measured by a conventional heat-pulse method using a mechanical heat switch. The button-shaped polycrystalline sample of about 2 g was attached to a copper addenda. The temperature was measured with a CGR resistance thermometer calibrated in magnetic fields of 0, 6, 10 and 14.6 T.

3. Results and discussion

Figure 1 shows the magnetization curves of $Lu(Co_{1-x}Ga_x)_2$ with x = 0.06, 0.09 and 0.12 obtained at 4.2 K in pulsed fields up to 40 T. A clear metamagnetic transition with a large hysteresis is observed in the compounds with x = 0.06 and 0.09, and the critical field H_c decreases with increasing x, being accompanied by a remarkable increase in the initial susceptibility χ_0 . The values of H_c obtained from the centre of the hysteresis are 36 T for x = 0.06 and 6 T for x = 0.09 at 4.2 K.

Figure 2 shows the concentration dependence of the critical field H_c of $Lu(Co_{1-x}Ga_x)_2$, together with that of $Lu(Co_{1-x}Sn_x)_2$ (Murata *et al* 1991) and $Lu(Co_{1-x}Al_x)_2$ (Sakakibara *et al* 1990b) for comparison. The value of H_c for LuCo₂

is 74 T at 8 K as reported previously (Goto et al 1990, Sakakibara et al 1990b, Murata et al 1991). The partial replacement of Co by Ga is effective in reducing H_c in this manner. The decrease of H_e has been explained on the basis of the increase of the density of states due to a lattice expansion, because the atomic size of Ga, Sn and Al is larger than that of Co. In general, the larger the lattice parameter, the narrower the 3d band width becomes, resulting in higher density of states. However, as shown in figure 2, the concentration dependence of H_c for $Lu(Co_{1-x}M_x)_2$ compounds depends on the kind of M. Namely, the value of H_c decreases remarkably with increasing x and ferromagnetism is developed around x = 0.09 for M = Al. On the other hand, the value for M = Sn initially decreases and then remains constant about H = 40 T above x = 0.06. The Lu(Co_{1-x}Ga_x)₂ compounds are considered to show an intermediate property, because the concentration dependence of H_c decreases to below 1 T without the appearance of the spontaneous magnetization. In the latter two cases, the substitutional elements Sn and Ga have d electrons, being different from Al. It has been pointed out that the hybridization effects between the 3d states of Co and 3p state of substitutional atoms are important in the high-content region (Aoki and Yamada 1990). Moreover, it has been experimentally confirmed that the d-electron concentration influences the magnetic properties of $M(Co_{1-x}Al_x)_2$ with M = Y and Lu, whose lattice parameter was kept constant (Gabelko et al 1991). Therefore, the hybridization and d-electron concentration effects are important to explain the magnetic properties of the present pseudo-binary compounds.



Figure 1. High-field magnetization curves of $Lu(Co_{j-x}Ga_x)_2$ measured at 4.2 K in pulsed fields up to 40 T



Figure 2. Concentration dependence of the critical field H_c of Lu(Co_{1-x}Ga_x)₂, together with that of Lu(Co_{1-x}Al_x)₂ (Sakakibara *et al* 1990b) and Lu(Co_{1-x}Sn_x)₂ (Murata *et al* 1991) at 4.2 K.

The Arrott plots for x = 0.12 at 4.2 K are shown in figure 3, because the magnetization process seems to be ferromagnetic. The broken and chain lines in this figure represent extrapolations of the linear parts of the plots below H/M = 100 Oe g/emu and above H/M = 2300 Oe g/emu, respectively. The extrapolation to H/M = 0 along the broken line gives a negative M^2 at 4.2 K in 0 T, showing no spontaneous magnetization. On the other hand, the M^2 value obtained along the chain line shows a positive value, and a hysteresis takes place

Table 1. Experimental values of the electronic specific heat coefficient γ_{exp} for YCo₂ (Muraoka *et al* 1977) and LuCo₂, together with the theoretical values γ_{th} calculated by tight-binding approximation (Yamada *et al* 1984, 1985).

	$\gamma_{exp} (mJ K^{-2} mol^{-1})$	$\gamma_{\rm th} \ ({\rm mJ} \ {\rm K}^{-2} \ {\rm mol}^{-1})$
LuCo ₂	24.6	12.7 ^a
YCo2	36.2 ^b	13.7°

* Yamada et al (1985). ^b Muraoka et al (1977). ^c Yamada et al (1984).

between these two lines. Apparently, a metamagnetic transition occurs in the compound with x = 0.12. We have tried to replace Co by Ga up to x = 0.15; however, no spontaneous magnetization has been observed in a similar manner to that of $Lu(Co_{1-x}Sn_x)_2$ (Murata *et al* 1991), in contrast with that of $Y(Co_{1-x}Al_x)_2$ (Yoshimura and Nakamura 1985) and $Lu(Co_{1-x}Al_x)_2$ (Endo *et al* 1988).



Figure 3. Arrott plots up to 5.5 T for $Lu(Co_{0.86}Ga_{0.12})_2$. The broken and chain lines are explained in the text.

The concentration dependence of electronic specific heat coefficient γ in 0 T is shown in figure 4. The γ value of LuCo₂ is 24.6 mJ K⁻² mol⁻¹, being comparable to previous data (Ikeda *et al* 1984). The γ value increases with x and reaches 37.9 mJ K⁻² mol⁻¹ at x = 0.12, being about 1.5 times as large as that of LuCo₂. Similar behaviour has been observed in other Co-based Laves-phase compounds such as Y(Co_{1-x}AJ_x)₂ (Wada *et al* 1990a) and Lu(Co_{1-x}AJ_x)₂ (Wada *et al* 1990b). The experimental values of the electronic specific heat coefficient γ_{exp} of YCo₂ (Muraoka *et al* 1979) and LuCo₂ are listed in table 1, together with the theoretical values γ_{th} calculated by the tight-binding approximation (Yamada *et al* 1984, 1985) for comparison. The values of γ_{th} for both compounds are much lower than γ_{exp} . This means that the enhancement of the electronic specific heat coefficient due to the spin fluctuations is considerably large. The electronic specific heat coefficient γ is given by

$$\gamma = \frac{1}{3}\pi^2 k_{\rm B}^2 N_{\rm A} N(E_{\rm F})(1+\alpha) \tag{1}$$

where $N(E_{\rm F})$ is the density of states at the Fermi level, $k_{\rm B}$ the Boltzmann constant, $N_{\rm A}$ Avogadro's number and α the mass enhancement factor due to spin fluctuation, electron-phonon interaction and so on. The factor α is important to explain the large γ value for the present compounds.

Low-temperature specific heat and magnetization measurements were also carried out on Lu(Co_{1-x}Sn_x)₂ prepared by the same method, for comparison. Figure 5 shows the concentration dependence of the electronic specific heat coefficient γ of Lu(Co_{1-x}Sn_x)₂, together with the initial susceptibility χ_0 . The γ value increases with increasing x, reaches a maximum around x = 0.04 and subsequently decreases with a further increase. Such a maximum is also observed in $Y(Co_{1,m}Al_m)_2$ and $Lu(Co_{1-x}Al_x)_2$ at the critical concentration for the appearance of ferromagnetism (Wada et al 1990b). However, the maximum is not due to the appearance of ferromagnetism in the present case, because no spontaneous magnetization has been confirmed (Murata et al 1991). The large value of γ for Lu(Co_{1-x}Sn_x)₂ is attributed to spin fluctuation, which is also reflected in the large value of the exchange enhancement factor. The initial susceptibility χ_0 and the electronic specific heat coefficient γ are related to the density of states of the 3d electrons at the Fermi level, $N(E_{\rm F})$. At low Sn concentrations, the behaviours of Lu(Co_{1-x}Sn_x)₂ and Lu(Co_{1-r}Ga_x)₂ are considered to be the same as that of Lu(Co_{1-r}Al_x)₂. Since the width of d bands becomes narrower by addition of Sn or Ga owing to lattice expansion, the density of states at the Fermi level becomes higher at the beginning and this effect causes the decrease of H_c . In the high Sn concentration regions, on the other hand, the concentration dependence of H_c for $Lu(Co_{1-x}Sn_x)_2$ is different from that for $Lu(Co_{1-x}Ga_x)_2$ as shown in figure 2. As is well known, the d bands of Sn atoms exist at a lower energy level than that of Co atoms. The local density of states on Co atoms would move downward relative to the Fermi level by substituting Sn atoms, as predicted by the coherent potential approximation (Velicky et al 1968), and result in the decrease of the density of states, because the position of the Fermi level lies just above the sharp peak of d bands in LuCo₂ (Yamada et al 1984). Such a consideration would also apply to the case of M = Ga. The d bands of Ga exist at a higher energy level than that of Sn, because Ga is a fourth periodic element, while Sn is a fifth one. Therefore, the decrease in the density of states is not remarkable compared with that in M = Sn. More noteworthy is that the H_c value decreases monotonically for Lu(Co_{1-x}Sn_x)₂ as shown in figure 2, although the γ and χ values become small above x = 0.06 as show in figure 6. A possible mechanism is considered as follows, although the reason for this behaviour is not clear at the present stage. The itinerant electron metamagnetism is characterized by a double-minimum structure in the free-energy curve. If the double-minimum structure is enhanced, such a decrease of H_c would be possible without increase of the γ and χ values. However, there is no conclusive evidence that the substitution of Sn enhances the double-minimum structure. Since this structure is considered to originate from a special density of states near the Fermi level with a positive sign of the second differential, detailed information on the band structure in $Lu(Co_{1-x}Sn_x)_2$ compounds is necessary for further discussion.

Figures 6 and 7 show the representative results on the heat capacity measurements at low temperatures for $Lu(Co_{1-x}Ga_x)_2$ with x = 0.09 and x = 0.12 in different magnetic fields. As is well known, the low-temperature specific heat is given by the following expression:

$$C/T = \gamma + \beta T^2 + \delta(T) \tag{2}$$

where the first, second and third terms are the electronic, lattice and magnetic contributions to the specific heat, respectively. The plots of C/T versus T^2 of





Figure 4. Concentration dependence of the electronic specific heat coefficient in zero field, γ , of Lu(Co_{1-x}Ga_x)₂.

Figure 5. Concentration dependence of the electronic specific heat coefficient in zero field, γ , and initial susceptibility, χ_0 , of Lu(Co_{1-x}Sn_x)₂.

 $Lu(Co_{1-x}Ga_x)_2$ show a straight line except below 4 K² and the slope of the lines in a magnetic field is nearly the same as that in zero field. An upturn in the C/Tversus T^2 curve is obtained at lower temperatures for the high Ga content samples. Such behaviour has often been observed in many other alloy systems around the critical concentration of the appearance of ferromagnetism (Konno and Moriya 1987, Wada et al 1990a), although no definitive explanation has been given. An analysis of the low-temperature specific heats has been carried out for $Y(Co_{1-r}Al_r)_2$ (Wada et al 1990a) in terms of the self-consistent renormalization (SCR) theory (Konno and Moriya 1987). However, the estimated γ value of bare density of states for $Y(Co_{0.88}Al_{0.12})_2$, 0.85 mJ K⁻² mol⁻¹, is unreasonably small. These results suggest that the observed upturn in the C/T versus T^2 curves of $Y(Co_{1-x}Al_x)_2$ is not well described by a simple SCR theory in the critical concentration range for the appearance of ferromagnetism. For nearly ferromagnets, a contribution of a form $\delta(T) = T^2 \ln T$ to the specific heat capacity is predicted by the paramagnon theory (Brinkman and Engelsberg 1968). The experimental results have been checked by considering this term, but the curves obtained are not well fitted. Recently, it has been pointed out that the paramagnon theory is applicable only in extremely low-temperature regions and not suitable for quantitative comparison with the experimental results (Shioda et al 1988). The conventional $C/T = \gamma + \beta T^2$ is considered to be a good expression above 5 K^2 for the present results from the discussion mentioned above. Therefore, the γ value is linearly extrapolated to $T^2 = 0$, neglecting the upturn part.

The field dependence of the electronic specific heat coefficient γ_H for $Lu(Co_{1-x}Ga_x)_2$ is shown in figure 8. The γ_H value in H = 14.6 T is significantly reduced compared with γ in H = 0 T. The value of γ for x = 0.12 is 37.8 mJ K⁻² mol⁻¹ and a remarkable reduction is caused above H = 6 T. In both ferromagnetic and paramagnetic states of YCo₂, band calculations have been made by the tight-binding approximation (Yamada *et al* 1984). Considering that the change in the density of states at the Fermi level before and after the metamagnetic transition



Figure 6. Low-temperature specific heat of Lu($Co_{0,91}Ga_{0,09}$)₂ plotted in the form of C/T versus T^2 as a function of magnetic field.



Figure 7. Low-temperature specific heat of $Lu(Co_{0.68}Ga_{0.12})_2$ plotted in the form of C/T versus T^2 as a function of magnetic field.

is relatively small, the large decrease of γ value is attributed to the suppression of the spin fluctuations due to the metamagnetic transition. The γ value for x = 0.09decreases with increasing magnetic field and no sharp reduction is observed in spite of the sharp transition as seen from figure 1. This is probably due to the difference in the form of specimens, that is, the specimens for the magnetization measurements are powders and those for the specific heat measurements are bulk samples. It has been reported that the metamagnetic transition in Lu(Co_{1-x}Al_x)₂ is accompanied by a large magneto-volume effect (lijima et al 1989). This affects the magnetization process of the metamagnetism, that is, it would become hard for the transition in the bulk samples for low-temperature specific heat measurements to take place and fine powder samples for magnetization measurements are effective in releasing the stresses due to the magneto-volume effect. It is noteworthy that the γ_H value after the metamagnetic transition is about 19 mJ K^{-2} mol⁻¹, being comparable to the data on the ferromagnet $Lu(Co_{0.90}Al_{0.10})_2$ (Wada et al 1990b). The concentration dependence of the γ value for $Y(Co_{1-x}AI_x)_2$ shows a broad maximum in the weakly ferromagnetic region, while the value for $Lu(Co_{1-x}Al_x)_2$ sharply decreases above x = 0.09 (Wada et al 1990b). The spontaneous Co moment in $Y(Co_{1-x}Al_x)_2$ is 0.15 $\mu_{\rm B}$ and not so large, that is, the molecular field of these compounds is small and the spin fluctuations play a dominant role even in the weakly ferromagnetic region. On the other hand, both spontaneous Co moments in $Lu(Co_{1-x}Al_x)_2$ and induced Co moments in Lu(Co_{1-x}Ga_x)₂ are about 0.7 $\mu_{\rm B}$, being considerably larger than the spontaneous moment in $Y(Co_{1-x}Al_x)_2$, and hence a large molecular field sufficiently suppresses the spin fluctuation. However, the γ value in the ferromagnetic state is still large, compared with that of many other ferromagnetic materials. This is consistent with the fact that the magnetization is not saturated easily after transition for $Lu(Co_{1-x}Ga_x)_2$, as shown in figure 1, being accompanied by a large high-field susceptibility. The Co moment in RCo₂ and $(R_x Y_{1-x})Co_2$ (R = Tb, Ho, Er, Nd, Gd, Tm, Pr) system is induced by the magnetic R element (Beille et al 1978, Steiner et al 1978, Goto et al 1989) and is saturated to 1.0 $\mu_{\rm B}$ /Co (Goto et al 1989).

As mentioned above, the large decrease in the value of γ is found in $Lu(Co_{1-x}Ga_x)_2$. It is, therefore, interesting to investigate the γ_H of $Lu(Co_{1-x}Sn_x)_2$ compounds, which have a larger γ and higher H_c than those of $Lu(Co_{1-x}Ga_x)_2$ compounds. Figure 9 shows the magnetic field dependence of the electronic specific heat coefficient for $Lu(Co_{0.96}Sn_{0.04})_2$, together with that of $LuCo_2$ (Ikeda *et al* 1984) for comparison. The reduction ratio in 10 T, $(\gamma - \gamma_{10})/\gamma$, of $Lu(Co_{0.96}Sn_{0.04})_2$ is 1.5% and smaller than that of $LuCo_2$, although its γ value is comparable with that of $Lu(Co_{1-x}Ga_x)_2$. These data are summarized in table 2. The reduction ratio between $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Sn_x)_2$ will correspond to the difference of the critical field, that is, the values of these samples are about 46 T and 0.2 T for $Lu(Co_{0.96}Sn_{0.04})_2$ and $Lu(Co_{0.88}Ga_{0.12})_2$, respectively. Therefore, the main suppression of the spin fluctuation is not correlated with the high γ value but with the occurrence of the metamagnetic transition.

Table 2. The electronic specific heat coefficients in zero field, γ , and in 10 T, γ_{10} , its reduction ratio in 10 T, $(\gamma - \gamma_{10})/\gamma$, and the critical field H_c for Lu(Co_{0.91}Ga_{0.09})₂, Lu(Co_{0.88}Ga_{0.12})₂ and Lu(Co_{0.96}Sn_{0.04})₂, together with those for LuCo₂ (lkeda *et al* 1984).

	γ ($H = 0$ T)	γ_H (H = 10 T)	$(\gamma - \gamma_{10})/\gamma$ (%)	H _c (T)
$Lu(Co_{0.91}Ga_{0.09})_2$	35.7	29.4	17.6	6
$Lu(Co_{0.88}Ga_{0.12})_2$	37.9	21.0	44.6	0.2
Lu(Co _{0.96} Sn _{0.04}) ₂	33.0	32.5	1.5	46
LuCo ₂	26.6 ^a	23.8ª	10.3 ^a	74

^a Ikeda et al (1984).





Figure 8. Magnetic field dependence of the electronic specific heat coefficient γ_H of Lu(Co_{1-x}Ga_x)₂ with x = 0.09 and x = 0.12. The full triangle indicates the data on Lu(Co_{0.90}Al_{0.10})₂ (Wada *et al* 1990b) for comparison.

Figure 9. Magnetic field dependence of the electronic specific heat coefficient γ_H of Lu(Co_{0.96}Sn_{0.04})₂, together with that of LuCo₂ (Ikeda *et al* 1984) for comparison.

4. Conclusions

To study the magnetic field effect on the spin fluctuation of itinerant electron metamagnets, the magnetization and low-temperature specific heat of the pseudobinary Laves-phase compounds $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Sn_x)_2$ have been investigated in high magnetic fields. The main results are summarized as follows:

(i) A sharp metamagnetic transition is observed and the critical field H_c decreases with increasing x.

(ii) The critical field H_c of Lu(Co_{1-x}Ga_x)₂ compounds decreases with increasing x and becomes below 1 T at x = 0.12, whereas that of Lu(Co_{1-x}Sn_x)₂ remains about 40 T. No ferromagnetic state is established in zero field for both compounds.

(iii) Since the mass enhancement due to the spin fluctuation becomes remarkable by substitution of Ga and Sn for Co, the electronic specific heat coefficient increases with x and reaches about 35 mJ K^{-2} mol⁻¹.

(iv) The γ value of Lu(Co_{1-x}Ga_x)₂ compounds in a magnetic field is drastically decreased above the critical field of the metamagnetic transition, reflecting a drastic reduction of the spin fluctuation.

(v) The γ value of Lu(Co_{0.88}Ga_{0.12})₂ after the transition is comparable to the reported value of the ferromagnet Lu(Co_{0.90}Al_{0.10})₂, suggesting that the large molecular field due to the induced Co moments sufficiently suppresses the spin fluctuation.

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