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Itinerant-electron metamagnetism and the onset of ferromagnetism in Laves phase $Lu(Co_{1-x}Ga_x)_2$ compounds

H Saito, T Yokoyama and K Fukamichi†

Department of Materials Science, Graduate School of Engineering, Tohoku University, Sendai 980-77, Japan

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Abstract. Detailed magnetization measurements have been carried out for Laves phase $Lu(Co_{1-x}Ga_x)_2$ compounds which have a C15-type structure up to $x = 0.220$. A clear increase in the lattice constant occurs at room temperature in the vicinity of $x = 0.095$, corresponding to the critical concentration of the onset of ferromagnetism. A representative first-order metamagnetic transition was observed in the compounds with $x = 0.090$ and 0.093 in relatively low fields below 9 T.

The critical temperature T_0 from the first- to second-order metamagnetic transition in both compounds and the susceptibility maximum temperature T_{max} for several paramagnetic compounds were obtained. It should be noted that T_{max} practically coincides with T_0 for $x = 0.093$, very close to the critical concentration of the onset of ferromagnetism. This result is well explained by the recent theory associated with spin fluctuations. The temperature T_{max} shifts to lower temperatures with increasing x and disappears at $x = 0.095$ and above, correlated with the onset of ferromagnetism also. The spontaneous magnetization per Co atom exhibits a maximum value of 0.67 μ_B at $x = 0.110$, whereas the Curie temperature does not show a maximum value at the same composition.

1. Introduction

Itinerant-electron metamagnetism was first discussed by Wohlfarth and Rhodes (1962). They predicted that exchange-enhanced Pauli paramagnets which exhibit a maximum in the temperature dependence of susceptibility would show an itinerant-electron metamagnetic transition. Laves phase compounds such as YCo_2 and $LuCo_2$ belong to exchange-enhanced Pauli paramagnets, accompanied by a broad maximum in the temperature dependence of susceptibility (Lemaire 1966, Bloch *et al* 1971). The critical fields of the metamagnetic transition in Laves phase compounds with a C15-type structure such as YCo_2 (Cyrot and Lavagna 1979, Schwarz and Mohn 1984, Yamada and Shimizu 1985) and $LuCo_2$ (Yamada *et al* 1987) have been estimated theoretically. Recently, the metamagnetic transition in YCo_2 and $LuCo_2$ has been confirmed directly and their critical fields are about 70 and 75 T, respectively (Goto *et al* 1989, 1990). These values are lower than the theoretical values because of a large magnetovolume effect (Goto *et al* 1989).

By partial substitution of Co with Al, remarkable reductions of the critical field and onsets of ferromagnetism have been confirmed in $Y(Co_{1-x}Al_x)_2$ (Aleksandryan *et al* 1985, Sakakibara *et al* 1986, 1987, 1990a, b) and $Lu(Co_{1-x}Al_x)_2$ compounds (Endo *et al* 1987a, 1988, Gabelko *et al* 1987, Iijima *et al* 1990, Sakakibara *et al* 1987). The magnetic properties

† E-mail address: fukamich@material.tohoku.ac.jp

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in the ferromagnetic regions of $Lu(Co_{1-x}Al_x)_2$ compounds are different from those of $Y(Co_{1-x}Al_x)_2$ compounds regarded as weak ferromagnets (Yoshimura and Nakamura 1985). That is, the maximum values of the spontaneous magnetization are 0.14 μ_B/Co for $Y(Co_{1-x}Al_x)_2$ (Yoshimura and Nakamura 1985) and 0.66 μ_B/Co for $Lu(Co_{1-x}Al_x)_2$ (Iijima *et al* 1990). Furthermore, it has been reported that the magnetic state of $Lu(Co_{1-x}Al_x)_2$ compounds with $x = 0.125$ and 0.150 is classified as an intermediate regime between an itinerant weak and localized ferromagnetic state (Yoshimura *et al* 1988).

In both compounds mentioned above, no sharp metamagnetic transition has been observed in relatively low magnetic fields because of the onset of a weak ferromagnetism in $Y(Co_{1-x}Al_x)_2$ (Aleksandryan *et al* 1985, Sakakibara *et al* 1986, 1987, 1990a, b) and the coexistence of non-magnetic and magnetic states of Co atoms in $Lu(Co_{1-x}Al_x)_2$ (Endo *et al* 1987b, Shinogi *et al* 1987). On the other hand, it has been demonstrated that $Lu(Co_{1-x}Ga_x)_2$ compounds show a sharp itinerant-electron metamagnetic transition even in low magnetic fields (Murata *et al* 1993a–c) in contrast to $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Al_x)_2$. Therefore, $Lu(Co_{1-x}Ga_x)_2$ compounds are very useful to study the itinerant-electron metamagnetism.

It has been pointed out that the critical field H_{cr} of the metamagnetic transition and the temperature of susceptibility maximum T_{max} show a linear relationship in $Y(Co_{1-x}Al_x)_2$ compounds, suggesting that T_{max} of strongly exchange-enhanced compounds is closely correlated with the itinerant-electron metamagnetism (Sakakibara *et al* 1990a, b). More recently, itinerant-electron metamagnetism at finite temperatures has been discussed in the framework of spin fluctuations (Yamada 1993). According to this theory, the relationship between T_{max} and the critical temperature T_0 from the first- to second-order metamagnetic transition is given as a function of ac/b^2, where a, b and c are the Landau–Ginzburg coefficients of itinerant-electron metamagnets. However, the data on T_{max} and T_0 for Laves phase compounds, particularly in the concentration region very close to the onset of ferromagnetism, are insufficient. Furthermore, there is no report on the onset of ferromagnetism in $Lu(Co_{1-x}Ga_x)_2$ compounds, although ferromagnetic properties of in $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Al_x)_2$ are well known.

The purpose of the present study is to investigate the itinerant-electron metamagnetism and the onset of ferromagnetism in $Lu(Co_{1-x}Ga_x)_2$ compounds.

2. Experimental details

Alloying was performed by arc-melting in an argon gas atmosphere. One charge weight of about 15 g was melted several times. The weighed Lu content was kept slightly higher than the stoichiometric composition, namely, $Lu_{34}(Co_{1-x}Ga_x)_{66}$, to avoid the precipitation of any ferromagnetic phases due to the decrease in Lu composition through vaporization. These compounds were annealed at 1223 K for a week in an evacuated quartz tube for homogenization and subsequently quenched into water. The surface of the bulk specimens was eliminated in order to exclude the influence of oxidation. In magnetization measurements, bulk specimens were used because the powders are easily oxidized.

Since the magnetic properties are significantly sensitive to the concentration, a short step change in the Ga concentration x was carried out. The room-temperature lattice constant was determined by x-ray powder diffraction using Mo Kα radiation. Magnetization measurements were carried out up to 9 T with a SQUID magnetometer (Quantum Design) and an extraction-type magnetometer PPMS (Quantum Design). The composition of the samples was analysed by an electron probe microanalyzer (EPMA), and no significant deviation from $Lu(Co_{1-x}Ga_x)_2$ was confirmed.

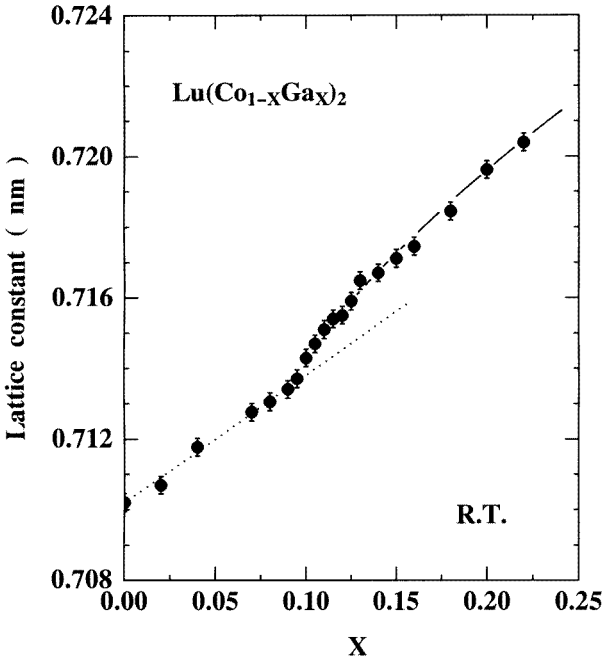

Figure 1. Concentration dependence of the room-temperature lattice constant for $Lu(Co_{1-x}Ga_x)_2$ compounds.

3. Results and discussion

X-ray diffraction analysis showed that all the samples with $x \leqslant 0.220$ have a C15-type Laves phase structure. A hexagonal C14-type structure phase, however, coexists with the C15-type structure phase above $x = 0.220$. Figure 1 shows the concentration dependence of the room-temperature lattice constant for $Lu(Co_{1-x}Ga_x)_2$. The lattice constant increases with increasing x, because the atomic size of Ga is larger than that of Co. Further, it deviates upward from the linear dotted lines above about $x = 0.095$, consistent with the previous results (Murata *et al* 1993a). It is important to point out that $Lu(Co_{1-x}Ga_x)_2$ shows the onset of ferromagnetism at $x = 0.095$ and above as given in the next figure. According to the self-consistent renormalization (SCR) theory (Moriya 1979, 1985), the mean-square amplitude of the local spin-density of fluctuations $\langle S_L^2 \rangle$ in itinerant ferromagnetic and nearly ferromagnetic alloys and compounds increases with increasing temperature in the paramagnetic ranges, leading to an extra lattice expansion in addition to the phonon term. The present result suggests that the value of the additional expansion at room temperature above about $x = 0.095$ is different from that below this composition, although all the samples are paramagnetic at room temperature as mentioned in connection with the next figure. In other words, the amplitude of $\langle S_L^2 \rangle$ at room temperature of the ferromagnets in the ground state would be different from that of the paramagnets in the ground state.

Shown in figure 2 are the magnetization curves at 4.2 K for $Lu(Co_{1-x}Ga_x)_2$ in the concentration range from $x = 0.090$ to 0.100. The curves for these compounds are quite sensitive to the Ga concentration x. The correction due to the demagnetizing field was not made since the approximated demagnetizing field was about two orders of magnitude

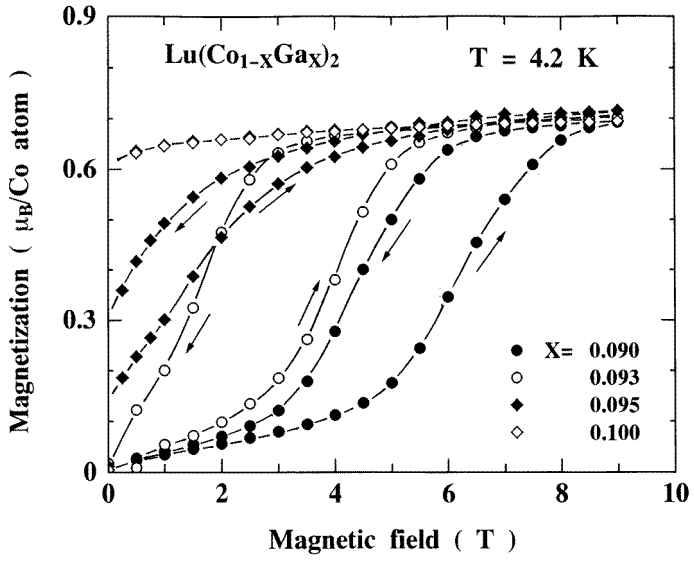

Figure 2. Magnetization curves at 4.2 K for $Lu(Co_{1-x}Ga_x)_2$ compounds in the concentration range from $x = 0.090$ to 0.100.

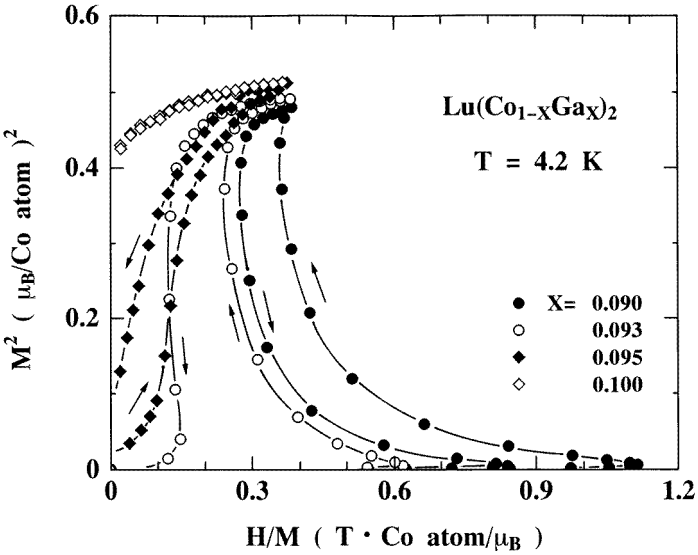

Figure 3. Arrott plots at 4.2 K for $Lu(Co_{1-x}Ga_x)_2$ compounds in the concentration range from $x = 0.090$ to 0.100.

smaller than the applied field. The critical transition field H_{cr} decreases with increasing x, and the compounds with $x \geqslant 0.095$ eventually become ferromagnets. A representative first-order metamagnetic transition in the magnetization curve is observed in the compounds with $x = 0.090$ and 0.093, while the curve of $x = 0.095$ indicates a spontaneous magnetization,

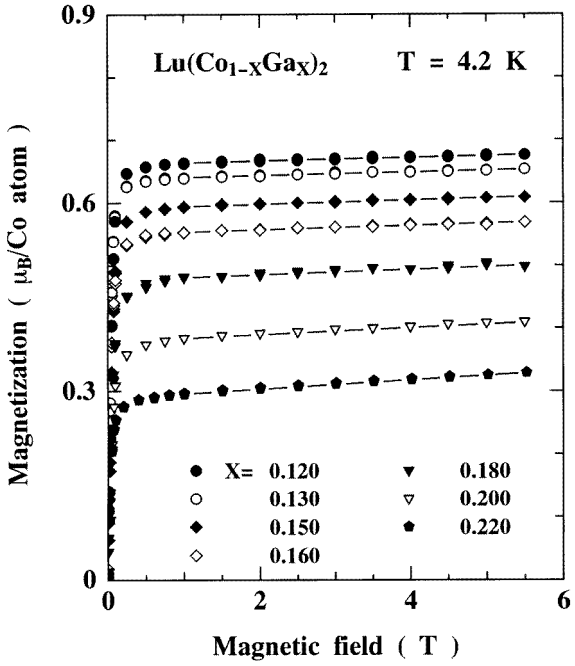

Figure 4. Magnetization curves at 4.2 K for Lu($Co_{1-x}Ga_x$)$_2$ compounds in the concentration range from $x = 0.120$ to 0.220.

confirmed clearly by the Arrott plots in figure 3. That is to say, in the curve of $x = 0.095$, the linear extrapolations to $H/M = 0$ in low fields give a positive M^2, indicating the onset of ferromagnetism. The magnetization curves of $x = 0.090$ and 0.093 exhibit a sharp metamagnetic transition from a paramagnetic to ferromagnetic state. It is worth noting that $x = 0.093$ is very close to the critical concentration of the onset of ferromagnetism. The magnetization curves at 4.2 K for Lu($Co_{1-x}Ga_x$)$_2$ in the concentration range from $x = 0.120$ to 0.220 are given in figure 4. All of the samples are ferromagnetic, and the spontaneous magnetization M_S decreases with increasing x. It has been reported that the sample with $x = 0.12$ is not ferromagnetic but paramagnetic and exhibits a metamagnetic transition (Murata *et al* 1993c) in contrast with the present results. This difference would come from the different annealing conditions. Namely, the present specimens were annealed at 1223 K for a week, but the previous one at 1073 K for a week (Murata *et al* 1993c). It was confirmed by EPMA that the present sample with $x = 0.090$ annealed under the former conditions has a homogeneous Ga concentration. On the other hand, a concentration gradient of Ga was confirmed in the sample annealed under the latter conditions. That is, the Ga concentration becomes lower toward the core in the crystal grains. These results indicate that the metamagnetic transition and other magnetic properties are dramatically affected by annealing conditions. The detailed relationship between the metamagnetic transition and the inhomogeneous concentration in Lu($Co_{1-x}Al_x$)$_2$ and Lu($Co_{1-x}Ga_x$)$_2$ will be reported elsewhere (Yokoyama *et al* 1997).

As seen from figures 2 and 3, the magnetization curves show a drastic change in the concentration range from $x = 0.090$ to 0.100. Therefore, it is necessary to carry out detailed measurements for each compound. In order to reveal vividly the remarkable concentration

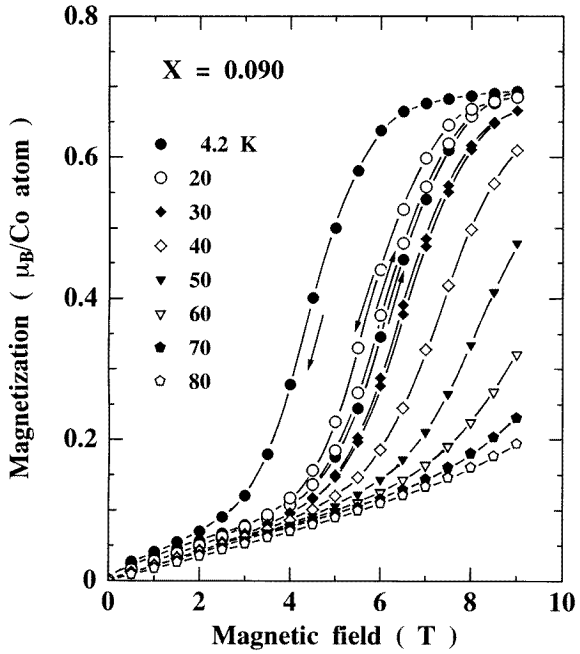

Figure 5. Magnetization curves as a function of temperature for the compound with $x = 0.090$.

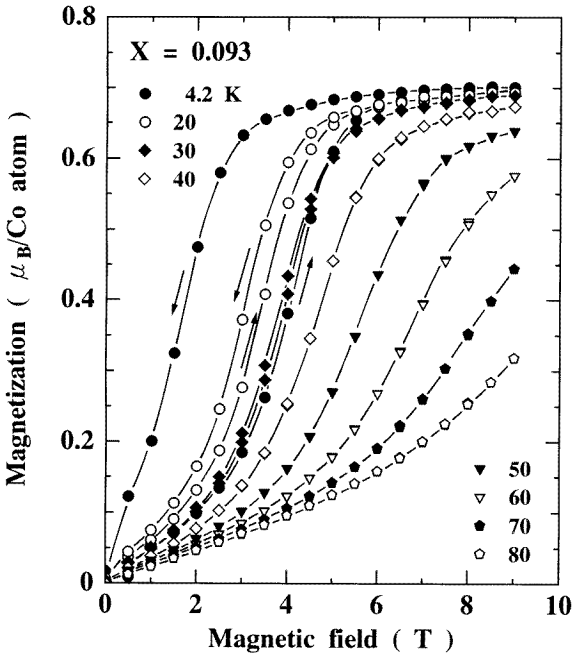

Figure 6. Magnetization curves as a function of temperature for the compound with $x = 0.093$.

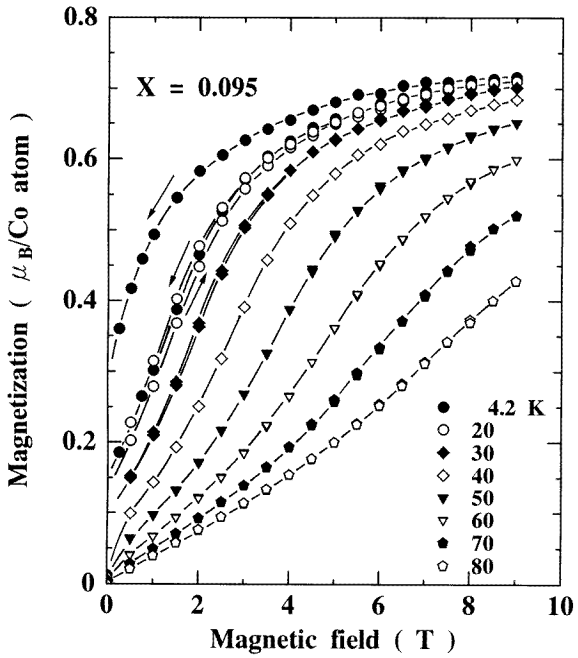

Figure 7. Magnetization curves as a function of temperature for the compound with $x = 0.095$.

dependence, the magnetization curves as a function of temperature for the compounds with a short step change in the concentration, $x = 0.090$, 0.093 and 0.095, are demonstrated in figures 5, 6 and 7, respectively. The temperature was increased in 10 K steps from 4.2 K for each magnetization measurement. The hysteresis in the curves becomes narrower with increasing temperature and disappears in the range between 30 K and 40 K. The disappearing temperature T_0 corresponds to the critical temperature from the first- to second-order transitions (Iijima *et al* 1990, Goto *et al* 1994). In order to obtain exactly T_0 for these compounds, we again carried out the magnetization measurements from 30 K to 40 K in 2 K increments. The values of T_0 for $x = 0.090$, 0.093 and 0.095 were determined to be 38, 38 and 36 K, respectively. It has been observed that the hysteresis of the metamagnetic transition in $Lu(Co_{0.91}Ga_{0.09})_2$ disappears at around 80 K (Murata *et al* 1993b), twice as high as that of the present specimen with the same composition practically. It is considered that T_0 increases with decreasing x, because the value of T_0 for $LuCo_2$ was estimated to be about 120 K (Yamada 1993). Therefore, the main reason for the difference mentioned above also could be attributed to the difference in the annealing conditions. The observed values of T_0 are almost the same because of the narrow concentration range. It is necessary to estimate T_0 for the compounds with a lower Ga concentration for detailed discussion. It has been reported that $H_{cr}(T)$ for YCo_2 (Goto *et al* 1994) and $Lu(Co_{0.91}Ga_{0.09})_2$ (Murata *et al* 1993b) increases in proportion to T^2. Such a relationship is also confirmed in the present compounds. Therefore, the values of H_{cr} at 0 K for $x = 0.090$, 0.093 and 0.095 determined from the $H_{cr}(T)$ versus T^2 are estimated to be 5.4, 2.9 and 1.0 T, respectively.

The magnetization curves as a function of temperature for the compound with $x = 0.100$ are given in figure 8. The Curie temperature T_C is estimated to be about 40 K from the inflection point in the thermomagnetization curve in a field of 10 mT. A metamagnetic

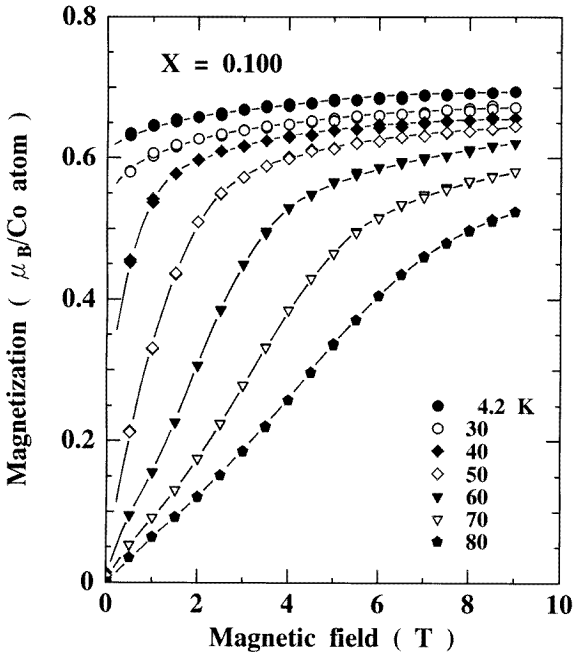

Figure 8. Magnetization curves as a function of temperature for the compound with $x = 0.100$.

transition behaviour still remains above T_C as seen from the figure, although no clear hysteresis is observed. To make the metamagnetic transition clear, the Arrott plots as a function of temperature for the compound with $x = 0.100$ are displayed in figure 9. In the paramagnetic region, these plots suggest that a metamagnetic transition occurs. For example, in the curve measured at 50 K, the broken and solid lines extrapolated to $H/M = 0$ from low fields and high fields give a negative and positive M^2, respectively. Similar phenomena are also observed in the curves at 60 and 70 K. Even above 70 K, such a metamagnetic transition would be expected by applying much higher magnetic fields. Consequently, the ferromagnetic compound with $x = 0.100$ exhibits the second-order metamagnetic transition in the paramagnetic region on applying external magnetic fields.

Itinerant-electron metamagnetism at finite temperatures has been discussed by taking into account spin fluctuations (Yamada 1993, Yamada and Terao 1994). According to this theory, the temperature dependence of the inverse slope of the Arrott plots in low magnetic fields $B(T)$ in the paramagnetic temperature range is given by

$$B(T) = b + \tfrac{14}{3}c\xi(T)^2 \tag{1}$$

where $\xi(T)^2$ is the mean-square amplitude of spin fluctuations and increases monotonically with increasing temperature. The coefficients b and c are respectively the fourth- and sixth-order terms in the Landau–Ginzburg coefficients in a uniform magnetization density. The coefficient b depends on the mode–mode coupling among spin fluctuations (Moriya 1985). It should be noted that the slopes of the Arrott plots in low magnetic fields for $x = 0.100$ are negative above 60 K. Therefore, the coefficient b should be negative because both c and $\xi(T)^2$ are positive, being a sure indication of the negative mode–mode coupling among spin fluctuations. It has been pointed out that an anomalously large negative value of pressure effect on the Curie temperature should be observed in ferromagnets with negative

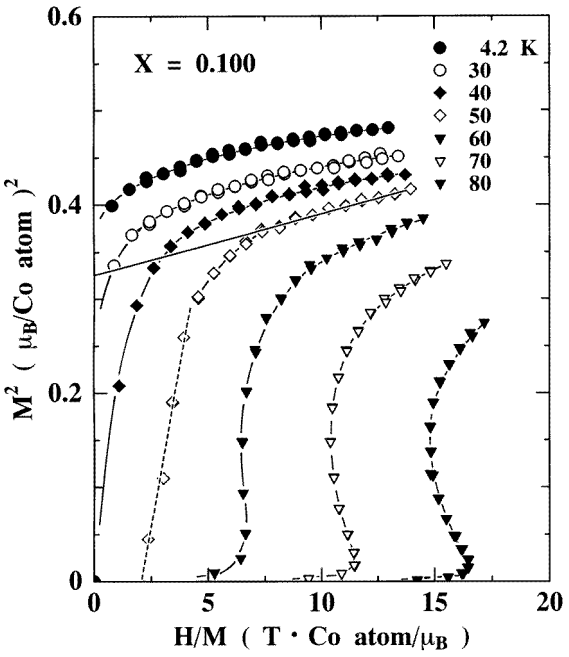

Figure 9. Arrott plots as a function of temperature for the compound with $x = 0.100$.

mode–mode coupling among spin fluctuations near the critical concentration of the onset of ferromagnetism (Yamada and Terao 1994). The experimental results of Lu($Co_{1-x}Ga_x$)$_2$ are in accord with this prediction. The systematic study of the pressure on the Curie temperature will be reported elsewhere (Saito *et al* 1997).

Figure 10 shows the temperature dependence of magnetic susceptibility for the paramagnetic compounds at 3 T, together with that of the specimen with $x = 0.090$ at 10 mT. Inverse susceptibility $\chi^{-1}(T)$ for strongly exchange-enhanced paramagnets is given by the following expression associated with spin fluctuations (Yamada 1993),

$$\chi^{-1}(T) = a + \tfrac{5}{3}b\xi(T)^2 + \tfrac{35}{9}c\xi(T)^4. \tag{2}$$

When $a > 0$, $b < 0$, $c > 0$ and $5/28 < ac/b^2 < 9/20$, with increasing $\xi(T)^2$, $\chi^{-1}(T)$ decreases at small $\xi(T)^2$ and increases at large $\xi(T)^2$. This means that the susceptibility $\chi(T)$ shows a maximum in the temperature. Strongly exchange-enhanced paramagnets YCo_2 (Lemaire 1966) and $LuCo_2$ (Bloch *et al* 1971) exhibit a broad maximum χ_{max} in the temperature dependence of susceptibility. Clearly, the temperature, T_{max}, which exhibits χ_{max} as indicated by the arrow, shifts to lower temperatures with increasing x. A similar phenomenon has been observed in Y($Co_{1-x}Al_x$)$_2$, (Sakakibara *et al* 1990a, b) and Lu($Co_{1-x}Al_x$)$_2$ (Iijima *et al* 1990, Sakakibara *et al* 1990b). Moreover, it has been reported that the data on H_{cr} and T_{max} as a function of x in Y($Co_{1-x}Al_x$)$_2$ indicate a linear relationship (Sakakibara *et al* 1990a), which is explained by considering the spin fluctuations (Yamada 1993a). In the present study, H_{cr} for the compounds with $x \leqslant 0.080$ could not be observed because of insufficient strength of magnetic fields. However, H_{cr} for Lu($Co_{1-x}Ga_x$)$_2$ compounds decreases with increasing Ga content (Murata *et al* 1993c, 1994), suggesting a linear relationship between H_{cr} and T_{max}. The temperature dependence

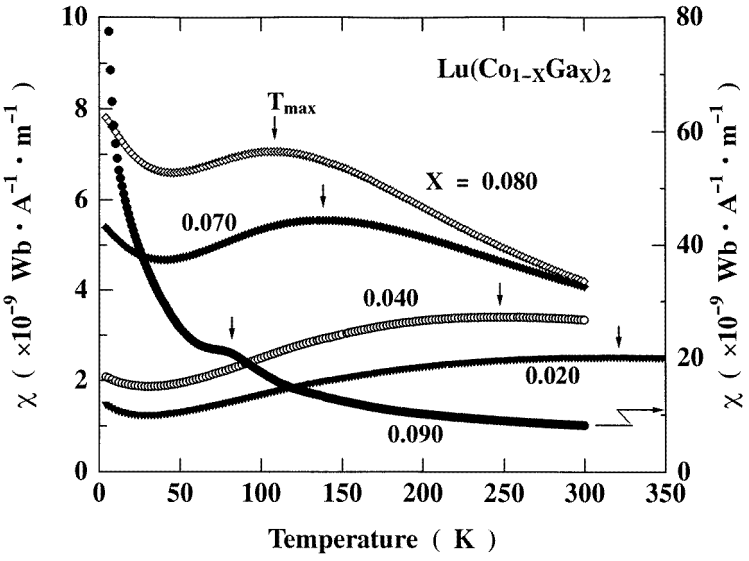

Figure 10. Temperature dependence of magnetic susceptibility χ for $Lu(Co_{1-x}Ga_x)_2$ compounds in the concentration range from $x = 0.020$ to 0.090.

of magnetic susceptibility for the compounds with $x = 0.093$, 0.095 and 0.100 is shown in figure 11. Since the compounds with $x = 0.095$ and 0.100 are ferromagnets, only paramagnetic regions are shown. All of the curves were measured in a low magnetic field of 10 mT enough to induce no ferromagnetic component. In the curve of the compounds with $x = 0.093$, a small peak corresponding to χ_{max} is observed around 30 K. The obtained results on T_{max}, T_0 and H_{cr} for $Lu(Co_{1-x}Ga_x)_2$ are listed in table 1. It should be noted that T_{max} almost coincides with T_0 for $x = 0.093$, very close to the critical composition of the onset of ferromagnetism. The value of T_0/T_{max} is given by the following expression (Yamada 1993),

$$\left(\frac{T_0}{T_{max}}\right)^2 = 1 - \sqrt{\frac{70}{19}}\sqrt{\frac{ac}{b^2} - \frac{5}{28}} \tag{3}$$

where a, b and c are the Landau–Ginzburg coefficients. This expression discloses that the value of T_0 of itinerant-electron metamagnets with the critical concentration of the onset of ferromagnetism is almost equal to that of T_{max} (Yamada 1993). The present experimental results for the compound with $x = 0.093$ are in consistent with the theory. The value of ac/b^2 for $x = 0.090$ is estimated to be about 0.34. It has been reported that T_0 for $(Y_{0.59}Lu_{0.41})(Co_{0.915}Al_{0.085})_2$ compound almost coincides with T_{max}, and the value of ac/b^2 is estimated to be 0.18 (Yamada *et al* 1993). However, the reported value of ac/b^2 seems to be rather small, because H_{cr} for $(Y_{0.59}Lu_{0.41})(Co_{0.915}Al_{0.085})_2$ is 9.5 T (Yamada *et al* 1993), higher than that of the present compounds with $x = 0.090$ and 0.093. In the paramagnetic temperature ranges, no χ_{max} is observed in the compounds with $x = 0.095$ and above in connection with the onset of ferromagnetism. The values of T_0 and H_{cr} for the compounds with lower concentration of x should be obtained by measuring in much higher magnetic fields.

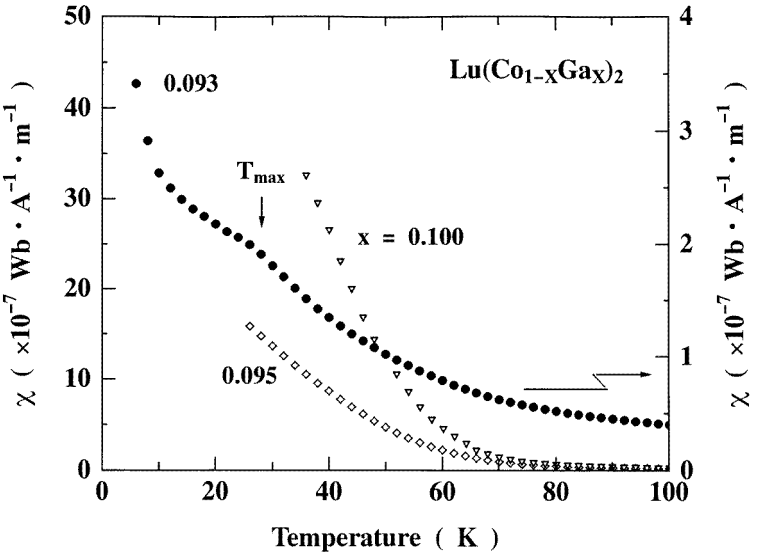

Figure 11. Temperature dependence of magnetic susceptibility χ for $Lu(Co_{1-x}Ga_x)_2$ compounds with $x = 0.093$, 0.095 and 0.100.

Table 1. The susceptibility maximum temperature T_{max}, the critical temperature T_0 from the first- to second-order metamagnetic transition and the critical transition field H_{cr} and from the paramagnetic to ferromagnetic state at 0 K for $Lu(Co_{1-x}Ga_x)_2$ compounds.

x	T_{max} (K)	T_0 (K)	H_{cr} (T)
0.020	320	—	—
0.040	246	—	—
0.070	138	—	—
0.080	108	—	—
0.090	~ 80	38	5.2
0.093	~ 30	38	2.9
0.095	—	36	1.0

The concentration dependences of the spontaneous magnetization M_S per Co atom at 4.2 K and the Curie temperature T_C for $Lu(Co_{1-x}Ga_x)_2$ are summarized in figure 12. All of the values of M_S and T_C were determined by the Arrott plots except T_C of the compounds with $x = 0.110$ and below, where these values were obtained from the inflection point in the thermomagnetization curves in a field of 10 mT, because no straight lines in the Arrott plots were obtained. With increasing x, the spontaneous magnetization increases significantly in a limited narrow composition range. It should be pointed out that such a marked increase has not been observed in $Lu(Co_{1-x}Al_x)_2$ (Iijima *et al* 1990, Endo *et al* 1987a, Gabelko *et al* 1987), because non-magnetic and magnetic states of Co atoms coexist in the composition range $0.06 \leqslant x \leqslant 0.12$ (Endo *et al* 1987b, Shinogi *et al* 1987). It is considered that the difference mentioned above comes also from the annealing conditions of the specimens as discussed in connection with figure 4.

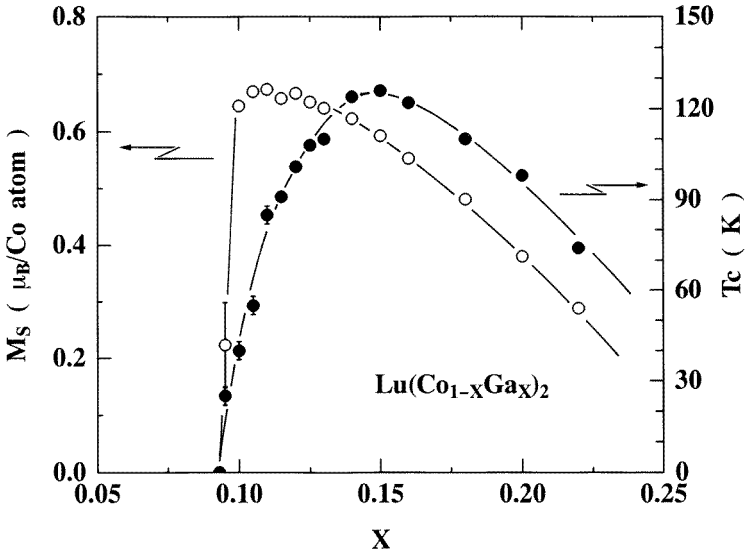

Figure 12. Concentration dependences of the spontaneous magnetization per Co atom M_S and the Curie temperature T_C for $Lu(Co_{1-x}Ga_x)_2$ compounds.

In YCo_2 and $LuCo_2$ compounds, the position of the Fermi level E_F lies just above a sharp and large peak of the density of states (DOS) calculated by a self-consistent tight-binding approximation method (Yamada *et al* 1984, Yamada and Shimizu 1985). By partial replacement of Co with Al, the peak is reduced and E_F shifts to the lower-energy side because of hybridization between the 3d electrons of Co and 3p electrons of Al (Aoki and Yamada 1989, 1992). The ferromagnetic state would be stabilized at a certain concentration where E_F goes across the peak of the DOS. For the $Lu(Co_{1-x}Ga_x)_2$ compounds, it is also suggested that E_F shifts to the lower-energy side and is located at the energy position of the peak of the DOS with increasing x, leading to the onset of ferromagnetism. The maximum spontaneous magnetization per Co atom is about 0.67 μ_B at $x = 0.110$, almost the same magnitude as that of the $Lu(Co_{0.89}Al_{0.11})_2$ compound (Iijima *et al* 1990). On the other hand, the maximum value of T_C 126 K at $x = 0.150$ is lower than that at $x = 0.156$ by about 20 K for the $Lu(Co_{1-x}Al_x)_2$ composition (Gabelko *et al* 1987). The noticeable phenomenon is that the composition with the highest T_C does not coincide with the composition which exhibits the largest M_S. A similar behaviour has been observed in $Lu(Co_{1-x}Al_x)_2$ compounds (Gabelko *et al* 1987) and conventional Fe–Ni Invar alloys (Crangle and Hallam 1963). The different concentration dependence between T_C and M_S for Fe–Ni alloys has been elucidated by the effective exchange coupling between local moment (Kakehashi 1990).

4. Summary

The room-temperature lattice constant and the magnetic properties of C15-type Laves phase $Lu(Co_{1-x}Ga_x)_2$ compounds have been investigated in order to discuss the relationship between T_0 and T_{max}, associated with the metamagnetic transition, where T_0 is the critical temperature from the first- to second-order transitions, and T_{max} is the susceptibility

maximum temperature. The critical concentration of the onset of ferromagnetism has been determined, and the characteristic concentration dependences of the spontaneous magnetization and the Curie temperature have been demonstrated. The main results are summarized as follows.

(a) A C15-type structure is maintained up to $x = 0.220$ and a clear increase in the lattice constant occurs in the vicinity of $x = 0.095$, corresponding to the onset composition of ferromagnetism.

(b) The ferromagnetic compound with $x = 0.100$ shows a second-order metamagnetic transition in paramagnetic regions on applying external magnetic fields.

(c) The slopes of the Arrott plots in low fields for the ferromagnetic compound with $x = 0.100$ are negative at paramagnetic temperatures, indicating a negative mode–mode coupling among spin fluctuations.

(d) The temperature T_{max} almost coincides with T_0 in the compound $x = 0.093$, very close to the critical concentration of the onset of ferromagnetism. This behaviour is well explained by the recent theory associated with spin fluctuations.

(e) The temperature T_{max} shifts to lower temperatures with increasing x and disappears at $x = 0.095$ and above, associated with the onset of ferromagnetism.

(f) The Ga concentration with a maximum spontaneous magnetization per Co atom does not coincide with the concentration exhibiting a maximum Curie temperature.

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