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Itinerant-electron metamagnetism and magneto-volume effects in $Lu(Co_{1-x}Al_x)_2$ Laves phase compounds

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Abstract

The itinerant-electron metamagnetic transition (MT) and the effects of hydrostatic pressure on the critical transition field B_C of the MT, on the spontaneous magnetization M_S and on the Curie temperature T_C have been investigated for well homogenized $Lu(Co_{1-x}Al_x)_2$ Laves phase compounds.

The critical field B_C decreases with increasing x, maintaining a linear relationship with the inverse susceptibility at the temperature where the susceptibility exhibits a maximum value, $\chi^{-1}(T_{max})$. On applying pressure, the magnetization M of the ferromagnetic compound with $x = 0.100$ is drastically decreased at a critical pressure, resulting in a paramagnetic state. In addition, the metamagnetic transition from the paramagnetic to the ferromagnetic state is induced by applying an external magnetic field.

The effect of pressure on the Curie temperature T_C is extremely large and negative in the vicinity of the critical concentration for the onset of ferromagnetism. The pressure coefficient of the Curie temperature, $\partial \ln T_C/\partial P$, is much larger than that of the spontaneous magnetization, $\partial \ln M_S/\partial P$, below $x = 0.150$. These results can be explained by the theory for itinerant ferromagnets having a negative coefficient b of the fourth-order term in the Landau expansion. The Landau expansion coefficients estimated from the experimental results are in accord with the theories. From these estimated values, it is concluded that the magneto-volume effect decreases the critical transition field B_C. It has been confirmed that the results for $Lu(Co_{1-x}Al_x)_2$ are very much analogous to those for $Lu(Co_{1-x}Ga_x)_2$.

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1. Introduction

YCo_2 and $LuCo_2$ Laves phase compounds are strongly exchange-enhanced Pauli paramagnets [1, 2] and exhibit a metamagnetic transition (MT) from the paramagnetic to the ferromagnetic state under high magnetic fields. The MT is associated with a sharp peak in the density of states (DOS) just below the Fermi energy E_F [3, 4]. The critical transition field B_C of the MT has been demonstrated to be about 69 T for YCo_2 [5] and 74 T for $LuCo_2$ [6]. These values are smaller by about 20 T than the values calculated by using the electronic structure of d electrons in the magnetic field [7]. These compounds both show a broad maximum χ_{max} in the temperature dependence of the magnetic susceptibility $\chi(T)$ at finite temperatures. On replacing Co with a non-magnetic element such as Al and Ga, both B_C and the susceptibility-maximum temperature, T_{max}, are significantly decreased [8–12]. A linear relation between B_C and T_{max} for $Y(Co_{1-x}Al_x)_2$ compounds has been pointed out [8]. These results imply that the susceptibility maximum χ_{max} is relevant to the shape of the DOS curve.

In quasi-binary systems such as $Y(Co_{1-x}Al_x)_2$ [8, 13, 14], $Lu(Co_{1-x}Al_x)_2$ [10, 15, 16] and $Lu(Co_{1-x}Ga_x)_2$ [17], the ferromagnetic state becomes stable above a critical concentration x_C. Below x_C, these compounds exhibit a MT under an applied magnetic field, accompanied by a large positive volume magnetostriction [18, 19]. This volume expansion makes the bandwidth narrower and the DOS at E_F higher, giving rise to a stable ferromagnetic state. Therefore, B_C for the MT is also expected to be influenced by the volume magnetostriction.

Itinerant-electron metamagnetism has been considered taking into account the effect of spin fluctuations on the free energy given by the Landau expansion [20–23]. Magnetic properties of $Lu(Co_{1-x}Ga_x)_2$ [24, 25] and $Co(S_{1-x}Se_x)_2$ [26] have been considered successfully using the theory of itinerant-electron metamagnetism at finite temperatures and under high pressures. According to the theory, a double-minimum structure in the paramagnetic and the ferromagnetic states in the free energy is closely related to the itinerant-electron metamagnetism when one considers spin fluctuations [21]. An anomalously large negative value of the pressure dependence of T_C is expected due to Invar effects in such materials [21]. In fact, it has been reported that $Lu(Co_{1-x}Ga_x)_2$ [25] and $Co(S_{1-x}Se_x)_2$ [26] each exhibit a large negative pressure dependence of T_C in the vicinity of the critical concentration for the onset of ferromagnetism.

For $Lu(Co_{1-x}Al_x)_2$ with $x = 0.06$ and 0.08, on the other hand, an extremely broad hysteresis in the metamagnetic transition curves has been observed [16]. In addition, the coexistence of paramagnetic and ferromagnetic Co atoms in the compound with $x = 0.08$ has been indicated by NMR experiment [27]. However, detailed considerations of magnetic properties of $Lu(Co_{1-x}Al_x)_2$ in the vicinity of the critical concentration for the onset of ferromagnetism have not been made yet. Recently, it has been pointed out by the present authors that the existing broad metamagnetic transitions mentioned above should be attributed to inhomogeneity of the Al concentration [28]. Accordingly, detailed investigations are of great interest for $Lu(Co_{1-x}Al_x)_2$ compounds with a high quality of homogeneity in composition.

In the present study, we have investigated the itinerant-electron metamagnetic transition and the effects of pressure on the spontaneous magnetization M_S and the Curie temperature T_C for well homogenized $Lu(Co_{1-x}Al_x)_2$. The experimental results have been analysed in terms of the Landau expansion, taking spin fluctuations into consideration. Moreover, the influence of the magneto-volume effect on B_C has been considered quantitatively. For comparison, several kinds of magnetic data for $Lu(Co_{1-x}Ga_x)_2$ have been presented, because the magnetic properties of $Lu(Co_{1-x}Ga_x)_2$ are very similar to those of $Lu(Co_{1-x}Al_x)_2$.

2. Experimental procedure

The specimens were prepared by arc melting in an argon gas atmosphere. In order to avoid any other ferromagnetic precipitates such as $LuCo_3$ arising through the loss of Lu during alloying, the nominal Lu composition was kept slightly higher than the stoichiometric composition, as $Lu_{34}(Co_{1-x}Al_x)_{66}$. The appropriate condition for homogenization was confirmed to be annealing at 1273 K for a week, similarly to in our previous study [28]. Therefore, the specimens were homogenized under the same conditions in evacuated quartz tubes; this was followed by quenching into water. The oxidized surface of the annealed specimen was removed mechanically. The crystal structure was identified by x-ray diffraction as a C15-type Laves phase without any other phases.

Magnetization measurements at ambient pressure were carried out up to 9 T with an extraction-type magnetometer. High magnetic fields up to 45 T and ultrahigh magnetic fields up to 90 T were produced by a wire-wound pulse magnet and a single-turn coil, respectively. Magnetizations at ambient pressure were measured by an induction method with a set of compensated pick-up coils. On the other hand, the magnetization measurements under high pressures were carried out with an extraction-type magnetometer equipped with a non-magnetic pressure clamp made of a Cu–2.3 at.% Ti alloy in magnetic fields up to 9 T. The susceptibility of the clamp is negligibly small: 5×10^{-8} emu g^{-1} at 1.8 K. The specimen was compressed in a Teflon cell filled with Fluorinert in a clamp cylinder up to 1.0 GPa. The applied hydrostatic pressures at low temperatures were calibrated by measuring the shift of the superconducting transition temperature of Pb [26]. Throughout the present paper, the compositions of the specimens are indicated by giving the nominal value of x.

3. Results and discussion

Magnetic properties of $Lu(Co_{1-x}M_x)_2$ (M = Al and Ga) Laves phase compounds are very sensitive to the annealing conditions. Together with the data (A) reported by Endo *et al* [16], the magnetization curve of the $Lu(Co_{0.920}Al_{0.080})_2$ compound annealed at 1273 K for 168 h (B) is shown in figure 1. The former exhibits a spontaneous magnetization with a broad hysteresis in the metamagnetic transition curves, whereas the latter exhibits a clear MT in relatively low fields. The insets in figure 1 depict the composition analyses of the specimens annealed at 1173 K for 100 h (A) and 1273 K for 100 h (B), respectively. Our annealing was carried out at 1173 K for 100 h, like that by Endo *et al* [16]. The oscillating white lines in the figure show concentration fluctuations of Al in the specimens. Specimen (A) exhibits strong oscillations due to inhomogeneity—that is, the grain boundaries have higher Al content—accompanied by a concentration gradient. On the other hand, specimen (B) annealed at 1273 K for 168 h exhibits very weak oscillations with noise levels, being homogeneous in composition. In order to confirm the annealing effect more clearly, the present magnetization curves were compared with available data for specimens with similar compositions. The magnetization curves of $Lu(Co_{1-x}Al_x)_2$ with $x = 0.070$ and 0.090 are presented in figure 2. The existing data for the specimens annealed at 1123 K for 180 h are given by the dotted curves [29, 30]. It should be noted that the temperature at which the specimens were annealed is the same as that in the investigations by Endo *et al* [16], although the annealing time is about double. On the other hand, the present results for the specimens annealed at 1273 K for 168 h are given by the solid curves. The metamagnetic transition given by the solid lines is much sharper, as compared with the dotted lines relating to imperfect homogenization. It is important to stress that the magnetization below the metamagnetic transition field for $Lu(Co_{0.930}Al_{0.070})_2$ exhibits an excellent linear increase without any responses from ferromagnetic impurities. From the

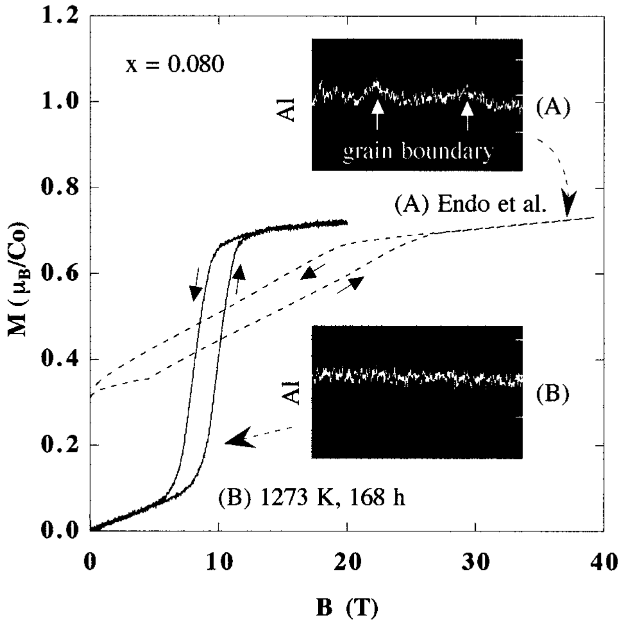

Figure 1. The magnetization curve for $Lu(Co_{0.920}Al_{0.080})_2$ annealed at 1273 K for 168 h, together with the data reported by Endo *et al* [16]. The insets show the composition profiles in the specimens annealed at 1123 K for 100 h and 1273 K for 168 h. The abscissa shows the Al concentration and the ordinate displays the cross section of the specimen.

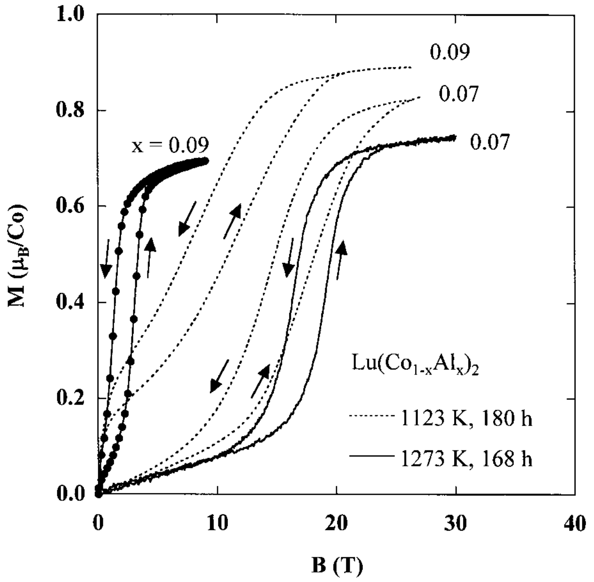

Figure 2. Magnetization curves for $Lu(Co_{1-x}Al_x)_2$ with $x = 0.070$ and 0.090, together with the results reported by Gabelko *et al* [30, 31].

present results, the appropriate condition for homogenization is concluded to be annealing at 1273 K for 168 h. Detailed experimental procedures and results have been reported elsewhere

[28]. The present clear metamagnetic transition strictly excludes the possibility of a transition from a weakly ferromagnetic to a strongly ferromagnetic state in $Lu(Co_{1-x}Al_x)_2$ [29, 30]. Gabelko *et al* have also discussed the relation between the magnetic phase and heterogeneity in $(Y_{1-t}Lu_t)(Co_{1-x}Al_x)_2$ annealed at 1123 K and 180 h, and arrived at the same conclusion [31].

In order to determine the critical field of the MT, B_C, for $Lu(Co_{1-x}Al_x)_2$, magnetization measurements were carried out. Figure 3 shows the magnetization curves obtained at 4.2 K for $Lu(Co_{1-x}Al_x)_2$. The data obtained at 10 K for the compounds with $x = 0.020$ and 0.040 are also given, by dotted curves, in the same figure, together with the data for $LuCo_2$ [6]. The magnetizations of both compound systems were calibrated using the data measured up to 5.5 T with a SQUID magnetometer. A clear MT with a large hysteresis is observed, and the critical transition field B_C of the MT decreases with increasing x, maintaining almost the same magnetization jump in magnitude. Here, B_C was defined as the average of the lower and higher critical fields determined at the peaks of the differential susceptibility in increasing and decreasing fields. For itinerant-electron metamagnetic systems such as paramagnetic $Lu(Co_{1-x}Ga_x)_2$ [32] and $Co(S_{1-x}Se_x)_2$ [26], B_C is in proportion to the square of the temperature T^2 at low temperatures. Therefore, B_C for the present $Lu(Co_{1-x}Al_x)_2$ sample is also directly proportional to T^2 and can be written in the low-temperature region as

$$B_C(x, T) = B_C(x, 0) + \alpha T^2 \quad (1)$$

where $B_C(x, 0)$ is the average critical field at $T = 0$ K and α is a constant. Because α remains almost constant as concentration is varied, B_C for $Lu(Co_{1-x}Al_x)_2$ with $x = 0.020$ and 0.040 is respectively estimated to be about 70 T and 51 T at 4.2 K, about 0.2 T lower than the values at 10 K. The magnetization curves of $Lu(Co_{1-x}Ga_x)_2$ are quite similar in shape to those of $Lu(Co_{1-x}Al_x)_2$ as given in figure 4. That is to say, these compound systems also exhibit a clear MT and B_C decreases with increasing x; this is accompanied by almost the same magnetization jump in magnitude.

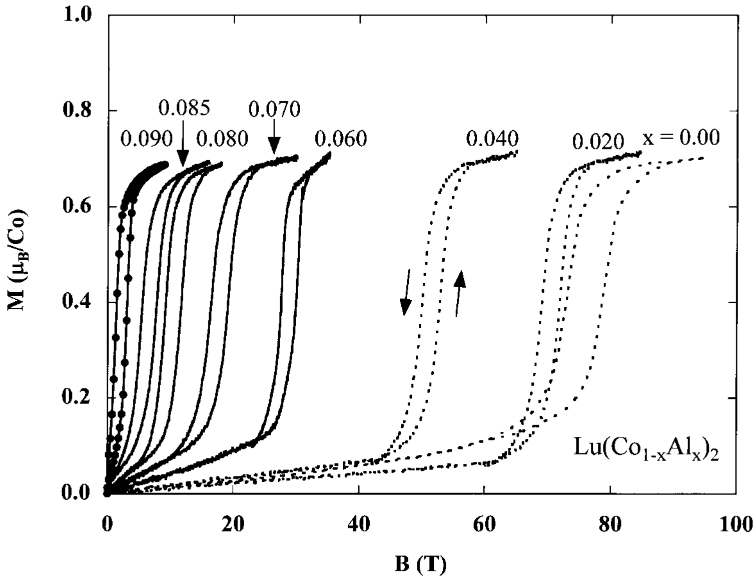

Figure 3. Magnetization curves for $Lu(Co_{1-x}Al_x)_2$ in the concentration range from $x = 0.020$ to 0.090, together with the curve for $LuCo_2$ for comparison [6]. The data obtained at 4.2 K are given by the solid curves, and those obtained at 10 K by the dotted curves.

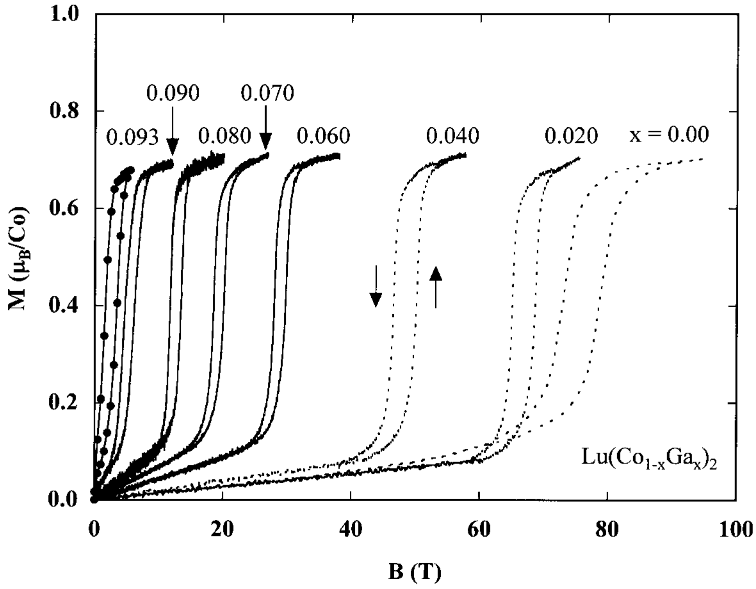

Figure 4. Magnetization curves for $Lu(Co_{1-x}Ga_x)_2$ in the concentration range from $x = 0.020$ to 0.093, together with the curve for $LuCo_2$ for comparison [6]. The data obtained at 4.2 K are given by the solid curves, and those obtained at 10 K by the dotted curves.

In connection with the MT in the Laves phase pseudo-binary systems, the temperature dependence of the susceptibility, $\chi(T)$, exhibits a broad maximum [8–12]. As shown in figure 5, $Lu(Co_{1-x}Al_x)_2$ below $x = 0.080$ shows a clear maximum χ_{max} at the temperature

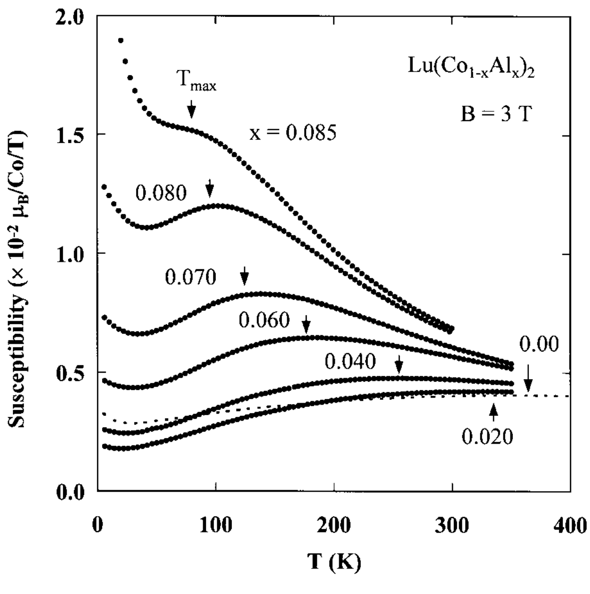

Figure 5. The temperature dependence of the susceptibility, $\chi(T)$, for $Lu(Co_{1-x}Al_x)_2$ in a magnetic field of 3 T. The result for $LuCo_2$ is given by the dotted line.

defined as T_{max} in the $\chi(T)$ curve as indicated by the arrows, and T_{max} decreases with increasing x. The dashed curve shows $\chi(T)$ for $LuCo_2$ annealed under the present conditions: 1273 K for 168 h. The present value of T_{max} for $LuCo_2$ is 360 K, slightly higher than the reported value [1]. Figure 6 shows the data for $Lu(Co_{1-x}Ga_x)_2$ [24], together with the additional new results. These curves are similar to those for $Lu(Co_{1-x}Al_x)_2$. Note that the scales of $\chi(T)$ are different for the compound with $x = 0.090$ and those with $x \leqslant 0.080$ as indicated in the figure. A linear relation between B_C and T_{max} in $Y(Co_{1-x}Al_x)_2$ has been pointed out, implying that the MT and χ_{max} originate from the same cause [8]. However, each compound system shows a different B_C–T_{max} relation, indicating that T_{max} is not necessarily connected with just B_C but may also be governed by some additional factors related to thermal spin fluctuations [33]. Figure 7 shows the relation between B_C and $\chi(T_{max})^{-1}$ for several kinds of Co-based quasi-binary compound. It is noteworthy that the B_C–$\chi(T_{max})^{-1}$ plots for Co-based Laves phase compounds follow a universal straight solid line with a slope of about 0.4 μ_B/Co. This result means that the mean square amplitude of the spin fluctuations at T_{max}, $\xi_p(T_{max})$, for these compounds is almost the same, regardless of the kind of compound system [33]. From the universal straight line of B_C–$\chi(T_{max})^{-1}$, the value of T_{max} for $Lu(Co_{0.915}Al_{0.085})_2$ is estimated to be about 85 K, although no clear maximum is observed in the susceptibility curve as indicated by the arrows in figure 5.

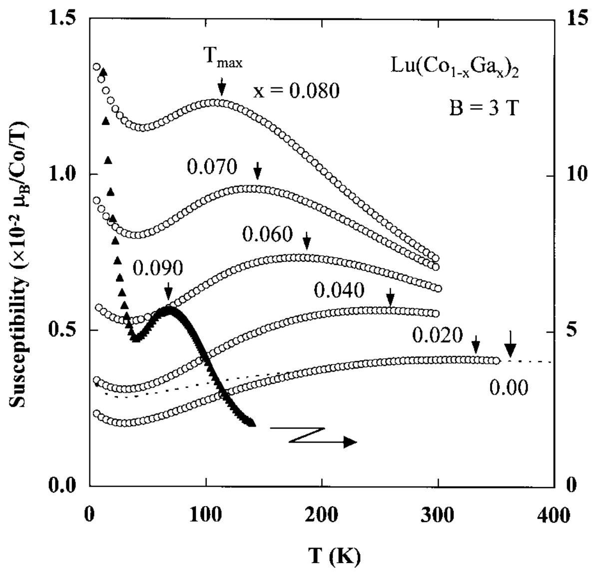

Figure 6. The temperature dependence of the susceptibility, $\chi(T)$, for $Lu(Co_{1-x}Ga_x)_2$ in a magnetic field of 3 T. Note that the scale of $\chi(T)$ for the compound with $x = 0.090$ is different from that for the compounds with $x \leqslant 0.080$. The result for $LuCo_2$ is given by the dotted line.

Shown in figure 8 is the concentration dependence of the spontaneous magnetization M_S at 4.2 K obtained from the Arrott plots for $Lu(Co_{1-x}Al_x)_2$, together with the data reported before [30] and the data for $Lu(Co_{1-x}Ga_x)_2$ [24] for comparison. The concentration dependence of M_S at 4.2 K for $Lu(Co_{1-x}Al_x)_2$ is quite similar to that of $Lu(Co_{1-x}Ga_x)_2$. The critical composition of the onset of ferromagnetism is higher than that reported by Gabelko *et al* [30]. This difference originates from the different annealing conditions responsible for the homogeneity (see figure 2). Figure 9 shows the concentration dependence of the Curie temperature T_C for $Lu(Co_{1-x}Al_x)_2$, together with the existing data for $Lu(Co_{1-x}Al_x)_2$ [30] and $Lu(Co_{1-x}Ga_x)_2$

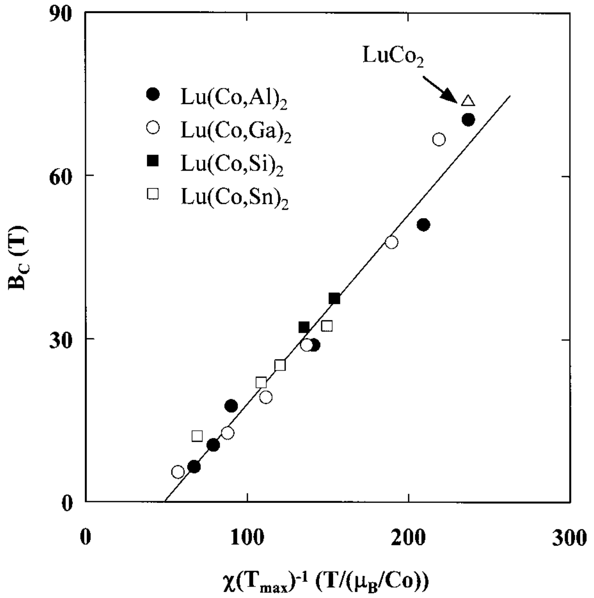

Figure 7. The critical field of the metamagnetic transition, B_C, obtained at 4.2 K ($B < 40$ T) and 10 K ($B \geqslant 40$ T) versus the inverse susceptibility at the susceptibility-maximum temperature, $\chi(T_{max})^{-1}$, for $Lu(Co_{1-x}Al_x)_2$, together with those for $Lu(Co_{1-x}Ga_x)_2$ [33], $Lu(Co_{1-x}Si_x)_2$ [33], $Lu(Co_{1-x}Sn_x)_2$ [33] and $LuCo_2$ [6].

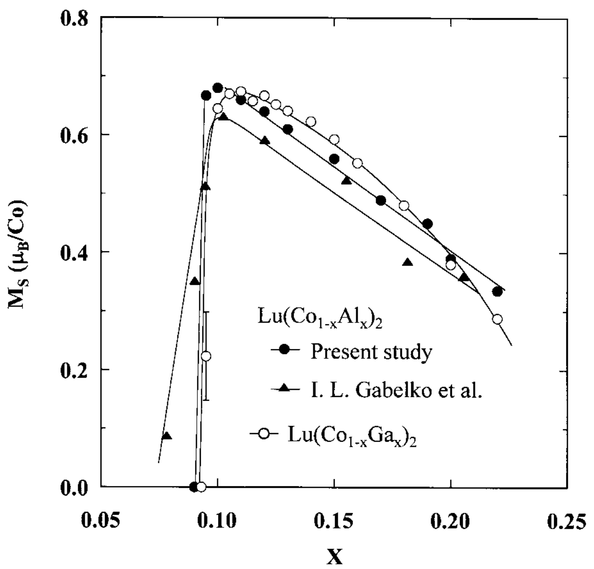

Figure 8. The concentration dependence of the spontaneous magnetization M_S at 4.2 K for $Lu(Co_{1-x}Al_x)_2$, together with the results reported by Gabelko *et al* [30] and also those for $Lu(Co_{1-x}Ga_x)_2$ [24].

[24] for comparison. The present results are analogous to those for $Lu(Co_{1-x}Ga_x)_2$, rather than the previous data for $Lu(Co_{1-x}Al_x)_2$ [30]. Strictly speaking, the values for $Lu(Co_{1-x}Al_x)_2$ are

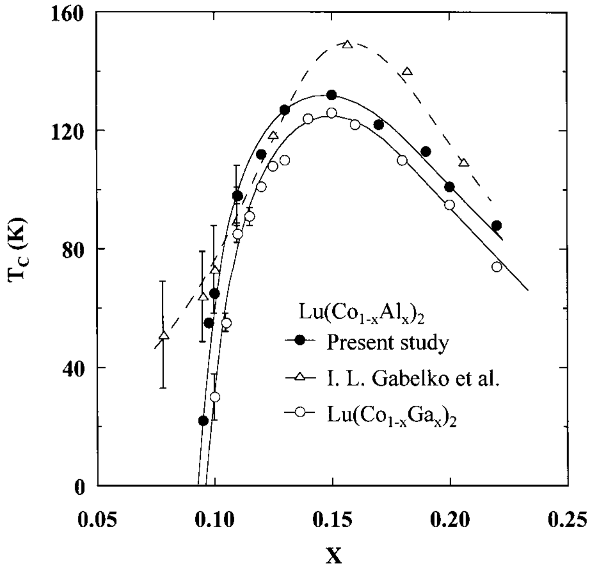

Figure 9. The concentration dependence of the Curie temperature T_C for $Lu(Co_{1-x}Al_x)_2$, together with the results reported by Gabelko *et al* [30] and those for $Lu(Co_{1-x}Ga_x)_2$ [24].

slightly higher than those for $Lu(Co_{1-x}Ga_x)_2$. Furthermore, it is interesting to note that the concentration with the highest T_C in figure 9 does not coincide with the concentration where M_S exhibits the largest value in figure 8. Thus the increase in T_C and the decrease in M_S with increasing x are observed around $x = 0.100$–0.150. As is well known, these behaviours are characteristic of conventional Invar alloys such as Fe–Ni, Fe–Pt and Fe–Pd [34], as well as amorphous Invar alloys [35, 36].

The concentration dependence of the high-field susceptibility χ_{hf} at 4.2 K for the compound $Lu(Co_{1-x}Al_x)_2$ is shown in figure 10, together with that of $Lu(Co_{1-x}Ga_x)_2$. The value of χ_{hf} was obtained from the law of approach to saturation. The two compound systems are quite similar to each other in concentration dependence—that is, χ_{hf} decreases rapidly up to about $x = 0.120$, and then increases with increasing x. The concentration dependence of χ_{hf} in the vicinity of the onset of ferromagnetism is analogous with that of Fe–Ni Invar alloys [34]. Such a large χ_{hf} is related to a pronounced forced volume magnetostriction, as well as significant pressure dependences of the spontaneous magnetization M_S and the Curie temperature T_C. From the present results, marked negative pressure dependences of M_S and T_C are expected for $Lu(Co_{1-x}Al_x)_2$ in the vicinity of the onset of ferromagnetism in analogy with the case for $Lu(Co_{1-x}Ga_x)_2$ [25]. On the other hand, the increase of χ_{hf} in the high-concentration range is attributed to the magnetic weakness; i.e., both the spontaneous magnetization M_S and the Curie temperature T_C significantly decrease with increasing x as seen from figures 8 and 9, respectively.

Figure 11 shows the thermomagnetization curves as a function of pressure for the representative samples of $Lu(Co_{1-x}Al_x)_2$ with $x = 0.100$ and 0.200 in a magnetic field of 0.5 T. The curves for both compounds clearly indicate negative pressure dependence. In particular, the curves for $x = 0.100$ are significantly affected by pressure in comparison with those for $x = 0.200$, indicating that the ferromagnetic state in the vicinity of the onset of ferromagnetism is very unstable. Shown in figure 12 is the effect of pressure on the Curie temperature T_C determined from the minimum point of $\partial M/\partial T$ in the thermomagnetization curve for

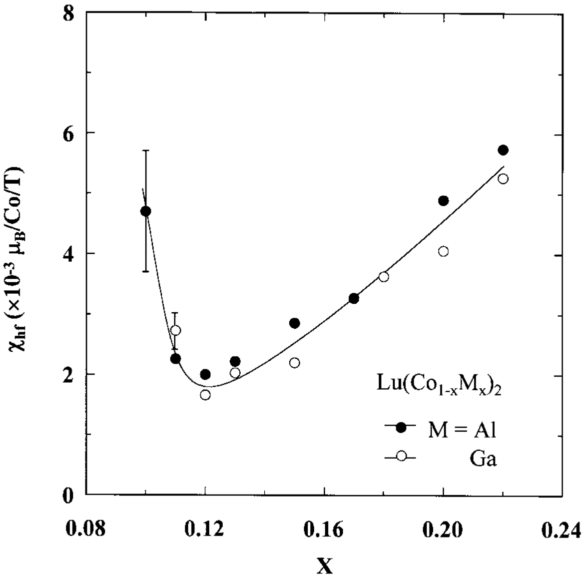

Figure 10. The concentration dependence of the high-field susceptibility χ_{hf} obtained at 4.2 K for $Lu(Co_{1-x}Al_x)_2$, together with that for $Lu(Co_{1-x}Ga_x)_2$.

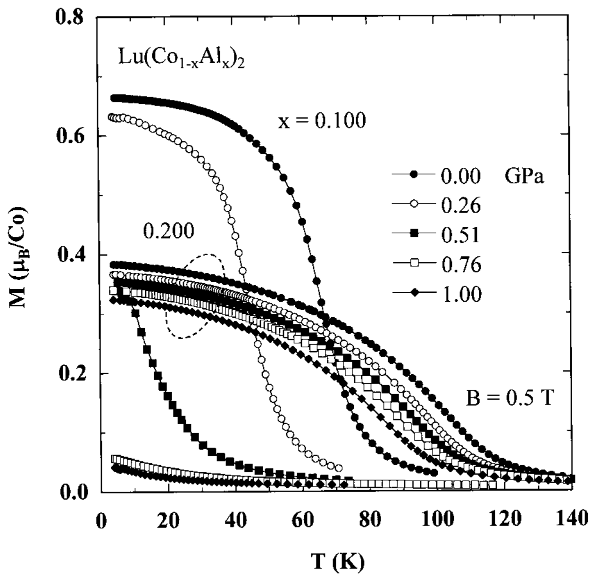

Figure 11. Thermomagnetization curves at various pressures for $Lu(Co_{1-x}Al_x)_2$ with $x = 0.100$ and 0.200 in a magnetic field of 0.5 T.

$Lu(Co_{1-x}Al_x)_2$. This method was reliable for the $Lu(Co_{1-x}Ga_x)_2$ data [25]. As shown in the figure, the high-concentration compounds exhibit a linear decrease of T_C, but the line for $x = 0.100$ is slightly curved due to the instability of the ferromagnetism. Figure 13 shows the concentration dependence of the pressure derivative of the Curie temperature, $\partial T_C/\partial P$, for $Lu(Co_{1-x}Al_x)_2$, together with that for $Lu(Co_{1-x}Ga_x)_2$ [25] for comparison. The magnitude of

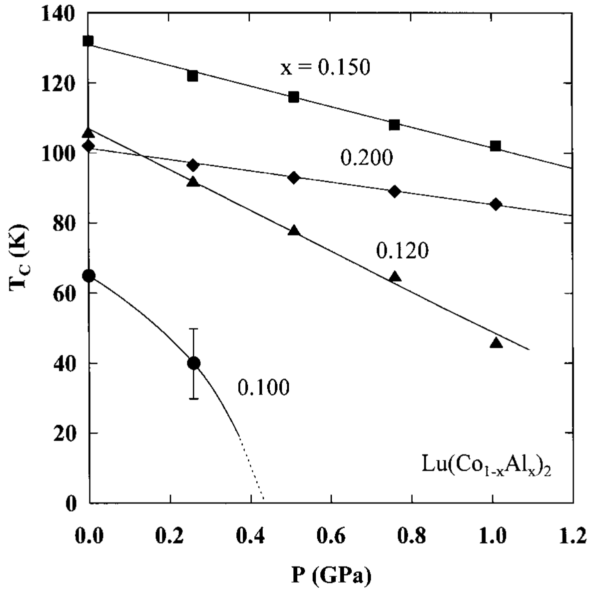

Figure 12. The effect of pressure on the Curie temperature T_C for $Lu(Co_{1-x}Al_x)_2$.

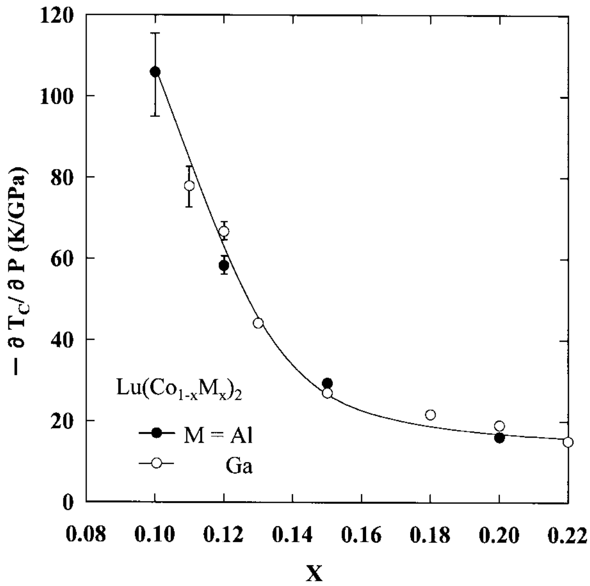

Figure 13. The concentration dependence of the pressure derivative of the Curie temperature, $\partial T_C/\partial P$, for $Lu(Co_{1-x}Al_x)_2$, together with that for $Lu(Co_{1-x}Ga_x)_2$ [25].

$\partial T_C/\partial P$, for both compound systems, is very sensitive to the concentration and a significantly large negative value is observed in the concentration toward the onset of ferromagnetism. The concentration dependence of $\partial T_C/\partial P$, for both compound systems, is comparable to that for conventional and amorphous Invar-type alloys [34–36]. Note that, in connection with figures 8 and 9, we have pointed out the similarity of the concentration dependences of the spontaneous magnetization and the Curie temperature for $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$, conventional and amorphous Invar alloys.

To explain the pressure dependence of T_C, the effect of spin fluctuations should be taken into account because the magnetic properties are influenced by spin fluctuations. Taking the magneto-volume coupling energy into consideration, the magnetic free energy F of itinerant-electron systems is written as

$$F = \frac{1}{2}\tilde{a}M^2 + \frac{1}{4}\tilde{b}M^4 + \frac{1}{6}\tilde{c}M^6 \tag{2}$$

with

$$\tilde{a} = a + 2\kappa C_{mv}P \qquad \tilde{b} = b - 2\kappa C_{mv}^2 \qquad \tilde{c} = c$$

where M is the uniform magnetization and the coefficients $\tilde{a}$, $\tilde{b}$ and $\tilde{c}$ are functions of a, b, c and the magneto-elastic coupling constant, κC_{mv}. The coefficients a, b and c are the Landau expansion coefficients. The conditions $\tilde{a} > 0$, $\tilde{b} < 0$ and $\tilde{c} > 0$ with $3/16 < \tilde{a}\tilde{c}/\tilde{b}^2 < 9/20$ are necessary for the MT. A negative $\tilde{b}$ is related to a positive curvature of the DOS at E_F [37] as well as negative mode–mode couplings among spin fluctuations [38]. The coefficients in equation (2) are renormalized by thermal spin fluctuations at finite temperatures. In the spin-fluctuation theory, the pressure dependence of T_C for ferromagnets under the conditions $\tilde{a} > 0$, $\tilde{b} < 0$ and $\tilde{c} > 0$ can be considered taking the pressure effect on the mean square amplitude of the spin fluctuations at T_C, $\xi_P(T_C)^2$, into account. $\partial\xi_P(T_C)^2/\partial P$ is given by the following expression [39]:

$$\frac{\partial\xi_P(T_C)^2}{\partial P} = -\frac{3\kappa C_{mv}}{\sqrt{35}|\tilde{b}|}\left(\frac{5}{28} - \frac{\tilde{a}\tilde{c}}{\tilde{b}^2} - \frac{2\kappa C_{mv}^2}{7|\tilde{b}|} + \frac{4\kappa^2 C_{mv}^4}{35\tilde{b}^2}\right)^{-1/2}. \tag{3}$$

The value of $\partial\xi_P(T_C)^2/\partial P$ is proportional to $\partial T_C^2/\partial P$, because $\partial\xi_P(T_C)^2$ varies in proportion to T_C^2 at low temperatures [20]. Equation (3) means that a significantly large negative value of $\partial T_C/\partial P$ is observed in the vicinity of the critical concentration for the onset of ferromagnetism because the condition $\tilde{a}\tilde{c}/\tilde{b}^2 = 5/28$ is close to $\tilde{a}\tilde{c}/\tilde{b}^2 = 3/16$, the condition for the critical concentration [21]. Recently, the values of $\tilde{a}$, $\tilde{b}$ and $\tilde{c}$ have been estimated by the fixed-spin-moment method for Fe_3Pt Invar alloy having a large negative value of $\partial T_C/\partial P$, satisfying the conditions $\tilde{a} > 0$, $\tilde{b} < 0$ and $\tilde{c} > 0$ with $\tilde{a}\tilde{c}/\tilde{b}^2 \fallingdotseq 5/28$ [21, 40]. Consequently, the observed large negative value of $\partial T_C/\partial P$ can also be explained by the conditions mentioned above. Further, the results for $Lu(Co_{1-x}Al_x)_2$ mean that the mean square amplitude of the spin fluctuations is very sensitive to the pressure in the vicinity of the critical concentration for the onset of ferromagnetism because $\partial\xi_P(T_C)^2/\partial P$ is proportional to $\partial T_C^2/\partial P$ at low temperatures in analogy with the results for $Lu(Co_{1-x}Ga_x)_2$ [25].

In order to facilitate discussion of the effect of pressure on the spontaneous magnetization M_S, the magnetization curves for $Lu(Co_{0.900}Al_{0.100})_2$ at 4.2 K as a function of pressure are presented in figure 14. By applying pressure, M_S at 4.2 K is easily dissipated and a paramagnetic state induced. In this paramagnetic state, the metamagnetic transition (MT) is caused by applying an external magnetic field. The observed MT in the magnetic curves is of first order with a clear hysteresis. Therefore, it is evident that the coefficient $\tilde{b}$ is negative, implying a positive curvature of the DOS at E_F due to the sharp peak of the DOS just below E_F. Figure 15 shows the effect of pressure on M_S obtained from the Arrott plots at 4.2 K for $Lu(Co_{1-x}Al_x)_2$. The value of M_S for the compound with $x = 0.100$ begins to decrease drastically above a certain pressure, whereas M_S for the compounds with $x \geqslant 0.120$ shows a linear decrease with increasing pressure up to 1.0 GPa. The concentration dependence of the pressure coefficient of M_S, $\partial \ln M_S/\partial P$, for $Lu(Co_{1-x}Al_x)_2$ is shown in figure 16, together with that for $Lu(Co_{1-x}Ga_x)_2$ [25] for comparison. The value of $\partial \ln M_S/\partial P$, for both compound

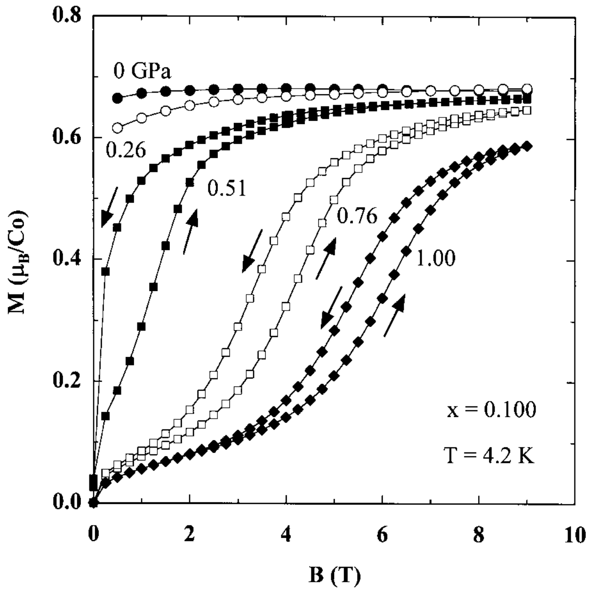

Figure 14. Magnetization curves at 4.2 K as a function of pressure for $x = 0.100$.

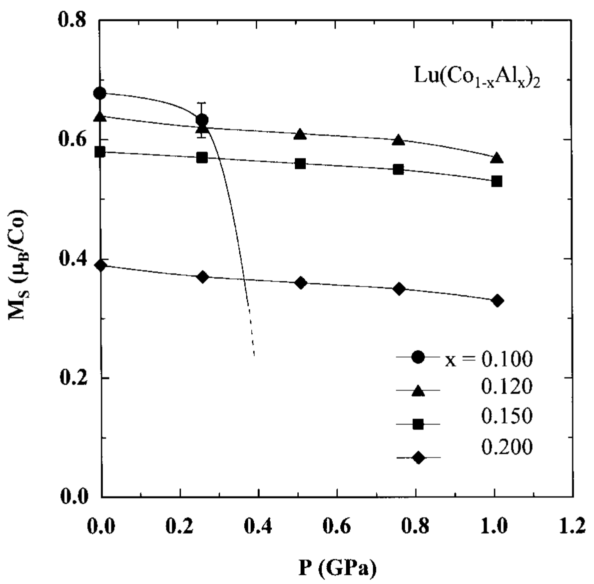

Figure 15. The effect of pressure on the spontaneous magnetization M_S for $Lu(Co_{1-x}Al_x)_2$.

systems, decreases up to about $x = 0.120$ and then slightly increases with increasing x. Its magnitude is proportional to κC_{mv} and χ_{hf} and is given by the following equation [41]:

$$-\frac{\partial \ln M_S}{\partial P} = 2\kappa C_{mv}\chi_{hf}. \tag{4}$$

The value of κC_{mv} is estimated to be about (4–7) $\times$ 10^{-3} μ_B^{-2}. Note that the concentration dependence of $\partial \ln M_S/\partial P$ is analogous to that of χ_{hf} at 4.2 K as given in figure 10.

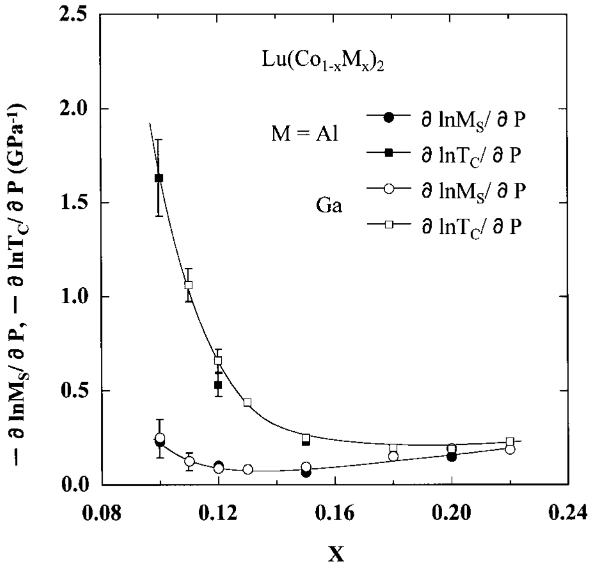

Figure 16. Concentration dependences of the pressure coefficients, $\partial \ln T_C/\partial P$ and $\partial \ln M_S/\partial P$, for $Lu(Co_{1-x}Al_x)_2$, together with those for $Lu(Co_{1-x}Ga_x)_2$ [25].

Figure 16 also shows the concentration dependence of the pressure coefficients of $\partial \ln T_C/\partial P$ for $Lu(Co_{1-x}Al_x)_2$, together with that for $Lu(Co_{1-x}Ga_x)_2$ [25]. Both of the coefficients, $\partial \ln M_S/\partial P$ and $\partial \ln T_C/\partial P$, for the former system are essentially the same as those of the latter system. It is worth noting that the magnitude of $\partial \ln T_C/\partial P$ is much larger than that of $\partial \ln M_S/\partial P$ below around $x = 0.150$, showing a rapid decrease of T_C in comparison with the decrease of M_S. Therefore, the sign of $\tilde{b}$ for the compounds with $x \leqslant 0.150$ is negative, implying that the positive curvature of the DOS at E_F remains up to around $x = 0.150$. Accordingly, the present results suggest that there is a sharp peak of the DOS just below E_F up to around $x = 0.150$. In figure 17, the concentration dependence of $\partial \ln T_C/\partial \ln M_S$ for $Lu(Co_{1-x}Al_x)_2$ is plotted against the concentration x, together with that for $Lu(Co_{1-x}Ga_x)_2$ [25]. The value of $\partial \ln T_C/\partial \ln M_S$, for both of the compound systems, increases with decreasing x and becomes about 7.2 for $Lu(Co_{0.900}Al_{0.100})_2$. According to the theory of weakly ferromagnetic itinerant-electron systems with the conditions $a < 0$, $b > 0$ and $c = 0$, a value of 3/2 for $\partial \ln T_C/\partial \ln M_S$ is obtained by taking spin fluctuations into consideration [42]. On the other hand, the present results are much larger than this value in the vicinity of the onset of ferromagnetism. Recently, our previous study of $Lu(Co_{1-x}Ga_x)_2$ [25] revealed that $\partial \ln T_C/\partial \ln M_S$ is determined from the Landau expansion coefficients $\tilde{a}$, $\tilde{b}$ and $\tilde{c}$ and that the magnitude of $\partial \ln T_C/\partial \ln M_S$ becomes large near the onset of ferromagnetism under the conditions $\tilde{a} > 0$, $\tilde{b} < 0$ and $\tilde{c} > 0$. As mentioned above, the sign of $\tilde{b}$ for $Lu(Co_{1-x}Al_x)_2$ is negative up to around $x = 0.150$, corresponding to the Al concentration where a large magnitude of $\partial \ln T_C/\partial \ln M_S$ is observed. Consequently, the present results can also be explained under the same conditions for the Landau expansion coefficients.

Large magneto-volume effects have been observed for $Lu(Co_{1-x}M_x)_2$ (M = Al and Ga) compounds as discussed in connection with figures 11–17. The magneto-volume effects influence not only the pressure dependences of T_C and M_S, but also the critical field B_C of the MT. In order to facilitate discussion of the influence of the magneto-volume effects on B_C, the pressure dependence of B_C was estimated from figure 14. The value of B_C was defined as

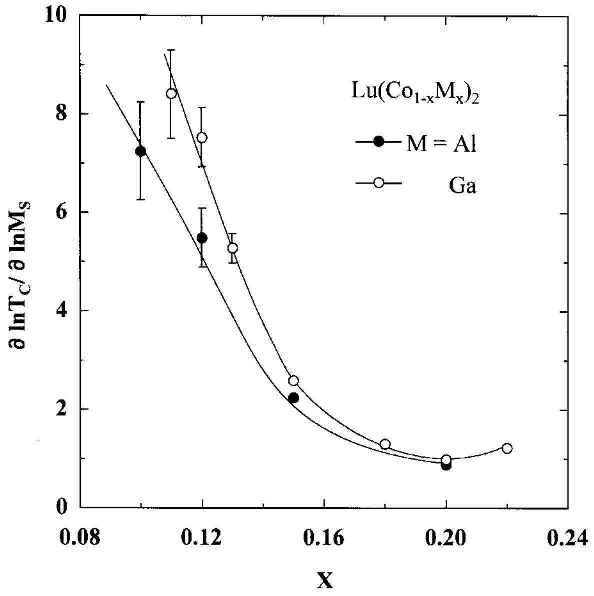

Figure 17. The concentration dependence of $\partial \ln T_C/\partial \ln M_S$ for $Lu(Co_{1-x}Al_x)_2$, together with that for $Lu(Co_{1-x}Ga_x)_2$ [25].

the average of the lower and higher critical fields determined at the peaks of the differential susceptibility in increasing and decreasing fields. Figure 18 shows the pressure dependence of B_C at 4.2 K for $Lu(Co_{0.900}Al_{0.100})_2$, together with that for $Lu(Co_{0.900}Ga_{0.100})_2$ [25] for comparison. The value of B_C increases linearly with the pressure as seen from the figure, and the value of $\partial B_C/\partial P$ is estimated to be 9.2 T GPa^{-1} for $Lu(Co_{0.900}Al_{0.100})_2$, slightly smaller than the value of 12 T GPa^{-1} for $Lu(Co_{0.900}Ga_{0.100})_2$. The critical pressure P_1 defined as the pressure where the transition field becomes zero is estimated to be 0.43 GPa by a linear extrapolation to $B_C = 0$. The effect of pressure on the width of the hysteresis ΔB_C defined as the difference between the lower and higher critical fields for $Lu(Co_{0.900}Al_{0.100})_2$ is given in figure 19, together with that for $Lu(Co_{0.900}Ga_{0.100})_2$ [25]. The critical pressure P_2 at which the first-order MT disappears is estimated to be 2.1 for the former and 1.3 GPa for the latter by a linear extrapolation. From figures 18 and 19, the values of P_1 and P_2 for $Lu(Co_{0.900}Al_{0.100})_2$ are both slightly larger than the values of $P_1 = 0.13$ and $P_2 = 1.3$ for $Lu(Co_{0.900}Ga_{0.100})_2$ [25]. These results imply that the ferromagnetic state of the former is more stable than that of the latter, in accordance with the larger M_S in figure 8 and the higher T_C in figure 9.

The magneto-elastic coupling constant, κC_{mv}, and the Landau expansion coefficients for $Lu(Co_{0.900}Al_{0.100})_2$ are estimated from the present experimental results in order to facilitate discussion of the influence of the magneto-volume effects on B_C. The effect of thermal spin fluctuations on the free energy is negligibly small because the present magnetization measurement temperature $T = 4.2$ K is low enough to allow neglect of the thermal spin fluctuations. Taking the magneto-volume coupling energy into consideration [22], the magnetic equation of state is written as

$$B(\omega, H) = \tilde{a}(P)M + \tilde{b}M^3 + \tilde{c}M^5. \tag{5}$$

The Landau expansion coefficients a and b are modified by the magneto-volume coupling as given in equation (2). It should be stressed that only the coefficient a in equation (2) is affected by the pressure dependence of the volume. The value of B_C becomes zero when the value of

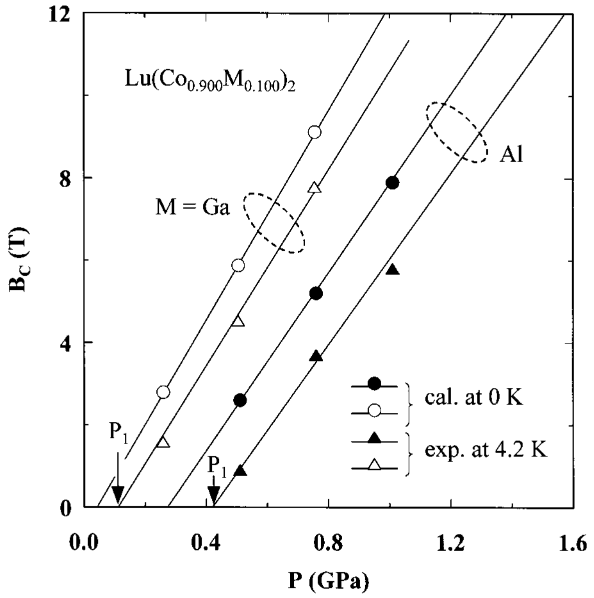

Figure 18. Pressure dependences of the critical transition field B_C at 4.2 K and the calculated B_C at 0 K for $Lu(Co_{0.900}Al_{0.100})_2$, together with those for $Lu(Co_{0.900}Ga_{0.100})_2$ [25].

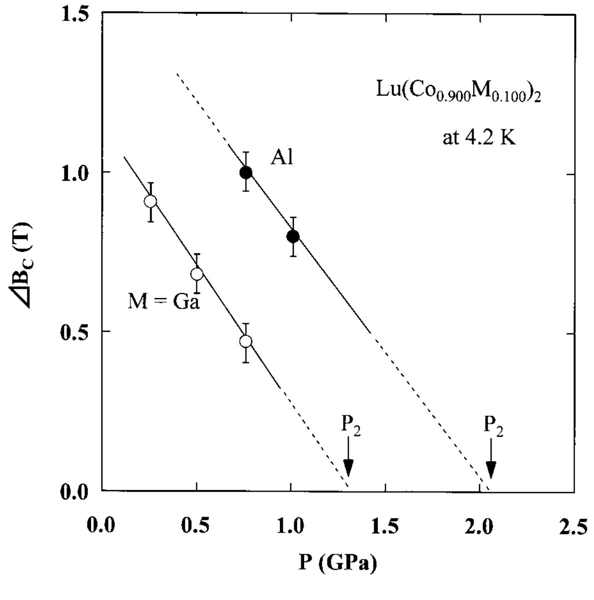

Figure 19. The effect of pressure on the width of the hysteresis of the critical field ΔB_C at 4.2 K for $Lu(Co_{0.900}Al_{0.100})_2$, together with that for $Lu(Co_{0.900}Ga_{0.100})_2$ [25].

$\tilde{a}\tilde{c}/\tilde{b}^2 = 3/16$, and the first-order MT disappears when $\tilde{a}\tilde{c}/\tilde{b}^2 = 9/20$ [20, 43]. Therefore, the observed values of P_1 and P_2 are connected by the following equations, respectively:

$$\frac{\tilde{a}\tilde{c}}{\tilde{b}^2} = \frac{(a + 2\kappa C_{mv} P_1)c}{(b - 2\kappa C_{mv}^2)^2} = \frac{3}{16} \tag{6a}$$

and

$$\frac{\tilde{a}\tilde{c}}{\tilde{b}^2} = \frac{(a + 2\kappa C_{mv} P_2)c}{(b - 2\kappa C_{mv}^2)^2} = \frac{9}{20}. \qquad (6b)$$

From equation (5), the pressure effect on M_S^2 at $T = 0$ is given by the following equation:

$$M_S^2 = \frac{|\tilde{b}|}{2\tilde{c}} \left\{ 1 + \sqrt{1 - \frac{4\tilde{a}\tilde{c}}{\tilde{b}^2}} \right\}. \qquad (7)$$

The measured value of M_S^2 for the compound with $x = 0.10$ is 0.46 $(\mu_B/\text{Co})^2$ (=6.0 × 10^4 emu^2 cm^{-6}) at $T = 4.2$ K under ambient pressure. The values of $\tilde{b}/a$ (=β), $\tilde{c}/a$ (=γ) and $\kappa C_{mv}/a$ (=δ) can be determined from equations (6*a*), (6*b*) and (7). Using the experimental result for B_C under pressure, the coefficient a is obtainable because two minima of F given by the following equation are equal to each other at B_C:

$$F = \frac{1}{2}(a + 2\delta a P)M^2 + \frac{1}{4}\beta a M^4 + \frac{1}{6}\gamma a M^6 - M B_C. \qquad (8)$$

Then, the values of $\tilde{b}$, $\tilde{c}$ and κC_{mv} are calculated by using the estimated value of a and the available value of the compressibility $\kappa = 8.5 \times 10^{-13}$ dyn cm^{-2} for $LuCo_2$ [44]. The estimated values of the Landau expansion coefficients $\tilde{a}$, $\tilde{b}$, $\tilde{c}$ and κC_{mv} for $x = 0.100$ are listed in table 1, together with those for $LuCo_2$ [18, 23], $Lu(Co_{0.900}Ga_{0.100})_2$ [25], $Co(S_{0.9}Se_{0.1})_2$ [26] and Fe_3Pt [40, 45] for comparison. The values of κC_{mv} for $LuCo_2$ and Fe_3Pt were estimated from the forced volume magnetostriction, while that for $Lu(Co_{0.900}Ga_{0.100})_2$ was estimated in the same way as in the present study. For all of these compounds, F has two local minima in the paramagnetic and the ferromagnetic states and the energy in the ferromagnetic state is lower than that in the paramagnetic state, indicating that the ferromagnetic state is stable in the ground state. From these results, a first-order ferromagnetic transition at T_C is expected. In fact, there is experimental evidence for such transitions in $Lu(Co_{0.900}Ga_{0.100})_2$ [25], $Co(S_{0.9}Se_{0.1})_2$ [26] and Fe_3Pt [46].

Table 1. Estimated values of the Landau expansion coefficients $\tilde{a}$, $\tilde{b}$, $\tilde{c}$ and the magneto-elastic coupling κC_{mv}, for $Lu(Co_{0.900}Al_{0.100})_2$, together with those for the compounds $LuCo_2$ [18, 23], $Lu(Co_{0.900}Ga_{0.100})_2$ [25], $Co(Se_{0.9}Se_{0.1})_2$ [26] and Fe_3Pt [40, 45].

	$\tilde{a}$ (10^2 cm^3 emu^{-1})	$\tilde{b}$ (10^{-2} cm^3 erg^{-1})	$\tilde{c}$ (10^{-6} cm^3 erg^{-2})	κC_{mv} (10^{-3} μ_B^{-2})
$Lu(Co_{0.900}Al_{0.100})_2$	6.9	−7.8	1.2	6.6
$Lu(Co_{0.900}Al_{0.100})_2$	7.6	−8.3	1.4	7.4
$LuCo_2$	269	−242	65	15.2
$Co(S_{0.9}Se_{0.1})_2$	2.2	−5.4	2.3	
Fe_3Pt	4.1	−5.9	1.3	2.4

According to the Landau expansion, as mentioned above, the first-order ferromagnetic transition occurs under the following condition at $P = 0$ [39]:

$$\frac{5}{28} - \eta < \frac{\tilde{a}\tilde{c}}{\tilde{b}^2} < \frac{3}{16} \qquad (9)$$

with

$$\eta = \frac{2}{7|\tilde{b}|}\kappa C_{mv}^2.$$

Therefore, the region of the first-order ferromagnetic transition at T_{C1} becomes wider if one takes the magneto-volume coupling energy into consideration. In the case of $0 \leqslant \tilde{a}\tilde{c}/\tilde{b}^2 \leqslant 5/28 - \eta$, the type of ferromagnetic transition changes to a second-order one. The value of $\tilde{a}\tilde{c}/\tilde{b}^2$ for $Lu(Co_{0.900}Ga_{0.100})_2$ is estimated to be 0.16, very close to $5/28 - \eta$ under the conditions that are operative between the first- and the second-order ferromagnetic transitions. On the other hand, the value of $\tilde{a}\tilde{c}/\tilde{b}^2$ for $Lu(Co_{0.900}Al_{0.100})_2$ is estimated to be 0.14, slightly smaller than $5/28 - \eta$, indicating a second-order ferromagnetic transition, in good agreement with the experimental results [47]. Consequently, it is concluded that the present estimated Landau expansion coefficients, $\tilde{a}$, $\tilde{b}$ and $\tilde{c}$, are reasonable. The estimated value of κC_{mv} for $Lu(Co_{0.900}Al_{0.100})_2$ in the paramagnetic state is very close to the value 6.9×10^{-3} $(\mu_B/\text{Co})^{-2}$ for paramagnetic $Lu(Co_{0.920}Ga_{0.080})_2$ [25]. The value of κC_{mv} is decreased by the MT from the paramagnetic to the ferromagnetic state due to the change of the spin-fluctuation spectra [18]. Therefore, $Lu(Co_{0.900}Al_{0.100})_2$ is expected to have a smaller value of κC_{mv} in the ferromagnetic state.

Finally, the influence of the magneto-volume effects on the critical transition field B_C is considered quantitatively in terms of the Landau expansion involving $\tilde{b}$ and b for $Lu(Co_{0.900}Al_{o.100})_2$. Here, $\tilde{b}$ and b are respectively the fourth-order-term coefficients in the Landau expansion with and without the magneto-volume effect in equation (2). The value of B_C without a magneto-volume effect should be obtained by comparison of the free energies in the ferromagnetic and paramagnetic states in the magnetic field. In figure 18, the experimental value is smaller by about 2 T than the calculated value at the same pressure, indicating that B_C is decreased by the magneto-volume coupling energy. Moreover, the calculated values of P_1 and P_2 are 0.28 and 1.7 GPa, respectively, smaller than the experimental results given in figures 18 and 19. In other words, it is clear that the ferromagnetic state becomes more stable on taking the magneto-volume effect into account, reducing the value of B_C. $Lu(Co_{1-x}Ga_x)_2$ [25] exhibits a striking resemblance to $Lu(Co_{1-x}Al_x)_2$ as seen from figures 18 and 19. Therefore, the magneto-volume effect evidently affects the itinerant-electron metamagnetism.

4. Conclusions

The itinerant-electron metamagnetic transition and the effect of pressure on the spontaneous magnetization M_S at 4.2 K, on the Curie temperature T_C and on the critical transition field B_C of the metamagnetic transition have been investigated for well homogenized $Lu(Co_{1-x}Al_x)$ Laves phase compounds. The results have been analysed in terms of the Landau expansion, taking spin fluctuations and magneto-volume effects into consideration. The relation between the critical transition field and the magneto-volume effect has been considered quantitatively. The magnetic properties of $Lu(Co_{1-x}Al_x)_2$ have been confirmed to be in analogy with those of $Lu(Co_{1-x}Ga_x)_2$. The main results are summarized as follows:

(a) Magnetic properties are very sensitive to the annealing conditions, in connection with the compositional homogeneity.

(b) The critical transition field for the metamagnetic transition, B_C, and the susceptibility-maximum temperature, T_{max}, both decrease with increasing x. The relation between B_C and the inverse of the susceptibility maximum, $\chi(T_{max})^{-1}$, follows a universal straight line with a slope of about 0.4 μ_B/Co.

(c) The pressure derivative of the Curie temperature, $\partial T_C/\partial P$, exhibits an extremely large negative value in the vicinity of the critical concentration of the onset of ferromagnetism, which is explained under the conditions $\tilde{a} > 0$, $\tilde{b} < 0$ and $\tilde{c} > 0$ with $\tilde{a}\tilde{c}/\tilde{b}^2 = 5/28$.

This result means that the mean square amplitude of the spin fluctuations is very sensitive to the pressure.

(d) The concentration dependence of the high-field susceptibility, χ_{hf}, and the pressure coefficient of the spontaneous magnetization, $\partial \ln M_S/\partial P$, exhibit similar tendencies at 4.2 K, associated with large magneto-volume effects. These values show marked increases in the vicinity of the critical concentration of the onset of ferromagnetism.

(e) The magnetization M of $Lu(Co_{0.900}Al_{0.100})_2$ exhibits a drastic decrease at relatively low pressures resulting in a paramagnetic state. In the pressure-induced paramagnetic state, a first-order metamagnetic transition is caused by applying an external magnetic field.

(f) The pressure coefficient of the Curie temperature, $\partial \ln T_C/\partial P$, is much larger than the pressure coefficient of the spontaneous magnetization, $\partial \ln M_S/\partial P$, for the compounds with $x \leqslant 0.150$ due to the negative sign of the coefficient $\tilde{b}$ of the fourth-order term in the Landau expansion. A significantly large value of $\partial \ln T_C/\partial \ln M_S$ in the vicinity of the onset of ferromagnetism can be explained under the conditions $\tilde{a} > 0$, $\tilde{b} < 0$ and $\tilde{c} > 0$.

(g) The experimental value of the critical transition field B_C is smaller than the calculated value without a magneto-volume effect at the same pressure, revealing that the magneto-volume effect reduces B_C for itinerant-electron metamagnetism.

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