Magnetic phase diagrams of itinerant-electron metamagnetic $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) Laves-phase compounds

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(Received 20 April 2001; published 28 August 2001)

Detailed magnetization measurements of $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) Laves-phase compounds with a high quality of homogeneity in composition have been carried out. A first-order ferromagnetic transition at the Curie temperature T_{C1} has been confirmed in $Lu(Co_{1-x}Al_x)_2$ with $x=0.094$ and 0.095. For these compounds, in addition, a metamagnetic transition from the paramagnetic to the ferromagnetic state above T_{C1} is induced by applying external magnetic field. The critical temperature T_0 from the first- to the second-order metamagnetic transition was estimated to be 42 K for $Lu(Co_{0.905}Al_{0.095})_2$. The concentration dependence of T_0 of $Lu(Co_{1-x}Al_x)_2$ is not so remarkable. In the higher concentration of x, the ferromagnetic transition is of the second-order-type. The magnetic transitions in $Lu(Co_{1-x}Gal_x)_2$ are quite similar to those in $Lu(Co_{1-x}Al_x)_2$. In contrast with previous experimental results, the established magnetic phase diagrams in the vicinity of the onset of ferromagnetism for $Lu(Co_{1-x}M_x)_2$ are in good agreement with the theoretical phase diagram.

DOI: 10.1103/PhysRevB.64.134401 PACS number(s): 75.30.Kz, 75.50.Cc, 75.60.Ej

I. INTRODUCTION

RCo_2 (R=Y and Lu) Laves-phase compounds are enhanced Pauli paramagnets, which exhibit a broad maximum in the temperature dependence of magnetic susceptibility[1,2] due to the characteristic sharp peak in the density of states (DOS) just below the Fermi energy E_F.[3,4] In connection with such a shape of the DOS in these compounds, the metamagnetic transition (MT) from the paramagnetic to the ferromagnetic state has been expected to be induced by applying high magnetic fields.[5,6] The critical transition field B_C of the MT has been confirmed to be about 69 T for YCo_2,[7] and 74 T for $LuCo_2$.[8] By replacing Co with Al, a significant reduction of B_C and the onset of weak ferromagnetism have been observed in $Y(Co_{1-x}Al_x)_2$,[9–11] consistent with the band calculation.[12] For $LuCo_2$, B_C is also decreased and a ferromagnetic state is caused by the replacement of Co with Al,[11,13,14] or Ga.[15,16] A characteristic behavior of $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) systems is a sharp metamagnetic transition without a remarkable change in magnitude of the field-induced moment even in the low critical field region,[14–16] in contrast to that of $Y(Co_{1-x}Al_x)_2$.[9–11] This implies that the shape of the DOS for $Lu(Co_{1-x}M_x)_2$ is different from that for $Y(Co_{1-x}Al_x)_2$. Although no information on the band structure of $Lu(Co_{1-x}M_x)_2$ is available, our recent studies have revealed that a metamagnetic transition is caused by applying external magnetic field under high pressures for $Lu(Co_{0.900}M_{0.100})_2$ [M=Ga (Ref. 17) and Al (Refs. 18 and 19)], implying a characteristic sharp peak in the DOS just below the Fermi level E_F.

The itinerant-electron metamagnetic transition at finite temperatures has been discussed in terms of the Ginzburg-Landau-type free energy expansion by considering the renormalization effect of spin fluctuations on the expansion coefficients.[20] For the onset of the itinerant-electron metamagnetic transition, the fourth-order coefficient of the Landau expansion should be negative because a double minimum structure in the magnetic-free energy as a function of magnetization is necessary in the paramagnetic and the ferromagnetic states.[20] Under such a condition, it is expected that the compounds in the vicinity of the onset of ferromagnetism give rise to a first-order ferromagnetic transition at the Curie temperature.[21]

$Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) below $x=0.090$ are paramagnetic in the ground state and exhibit a metamagnetic transition from the paramagnetic to the ferromagnetic state by applying external magnetic field. On the other hand, the compounds with a higher concentration of x are ferromagnetic in the ground state.[22–25] It should be emphasized that the magnetic properties of these compounds are very sensitive to homogeneity in composition.[14] Consequently, detailed behavior in the vicinity of the onset of ferromagnetism has not been discussed yet. Recently, we have revealed that the sign of the fourth-order coefficient of the Landau expansion for $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) with $x=0.100$ is negative.[19] This result implies that the magnetic free energy has a double minimum structure in the paramagnetic and the ferromagnetic states. In connection with the characteristic of the magnetic free energy, it is strongly expected to exhibit a first-order ferromagnetic transition at the Curie temperature in the concentration near the onset of ferromagnetism.

In the present study, the magnetic properties have been investigated for well homogenized $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) with the composition in the vicinity of the onset of

ferromagnetism. Further, the first-order magnetic transition behavior at the Curie temperature has been examined. From these results, the magnetic phase diagrams have been established. These results are discussed in terms of the Moriya[26] and Yamada[20] theories based on the Ginzburg-Landau-type free energy expansion by taking spin fluctuations into consideration.

II. EXPERIMENT

Alloying was carried out by arc-melting in an argon gas atmosphere. The composition of Lu was kept slightly higher than the stoichiometric composition as $Lu_{34}(Co_{1-x}M_x)_{66}$ (M = Al and Ga) in order to avoid any other ferromagnetic precipitates such as $LuCo_3$ because of the loss of Lu during arc-melting. The appropriate heat-treatment conditions were already investigated in detail.[14,19] Therefore, the specimens were annealed at 1273 K for 240 h in evacuated quartz tubes for homogenization, followed by quenching into water. The surface oxidation of the bulk specimens was mechanically eliminated. The crystal structure was identified by x-ray powder diffraction using Cu $K\alpha$ radiation as a C15-type Laves phase without any other phases. Magnetizations were measured by an extraction-type magnetometer PPMS (Quantum Design) in magnetic fields up to 9 T. The temperature dependence of magnetization was measured with a SQUID magnetometer. The compositions of the specimens are given by the nominal value of x throughout the present paper.

III. RESULTS AND DISCUSSION

Moriya[26] and Yamada[20] have proposed theories for the itinerant-electron metamagnetic transition at finite temperatures by taking spin fluctuations into account because magnetic properties of itinerant-electron systems at finite temperatures are strongly influenced by spin fluctuations. In order to discuss the present experimental results, we give an outline of the theories mentioned above. In the ground state, the magnetic free energy $F(M)$ of itinerant-electron systems is expressed by

$$F(M)=\tfrac{1}{2}aM^2+\tfrac{1}{4}bM^4+\tfrac{1}{6}cM^6, \tag{1}$$

where M is the uniform magnetization. The coefficients a, b, and c are the Landau expansion coefficients. The value of a corresponds to the inverse susceptibility at $T=0$ K. The coefficients a, b, and c at finite temperatures are renormalized by thermal spin fluctuations and the magnetic free energy $F(M)$ is given by[20]

$$F(M)=\tfrac{1}{2}A(T)M^2+\tfrac{1}{4}B(T)M^4+\tfrac{1}{6}C(T)M^6. \tag{2}$$

The coefficients $A(T)$, $B(T)$, and $C(T)$ are functions of a, b, and c, and the mean-square amplitude of spin fluctuations, $\xi_P(T)^2$, which is proportional to T^2 at low temperatures.[20] These coefficients are given as

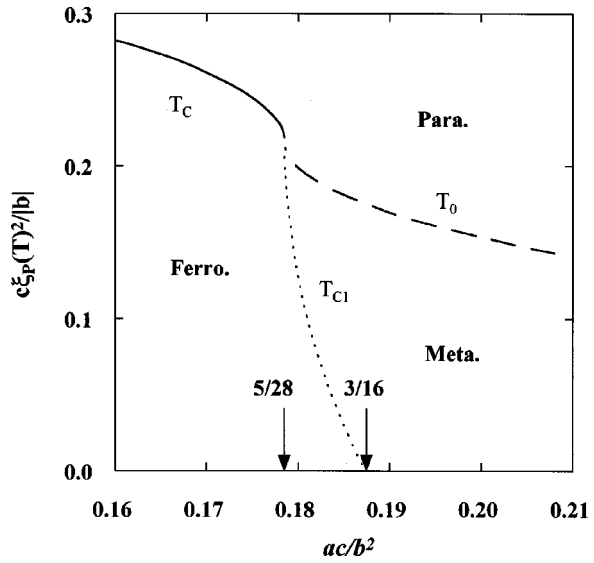

FIG. 1. Theoretical magnetic phase diagram under the conditions of $a>0$, $b<0$, and $c>0$ for the Landau expansion coefficients by considering spin fluctuations (Ref. 26).

$$A(T)=a+\tfrac{5}{3}b\xi_P(T)^2+\tfrac{35}{9}c\xi_P(T)^4,$$

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

$$C(T)=c.$$

The conditions of $a>0$, $b<0$, and $c>0$ with $3/16<ac/b^2<9/20$ are essential for the metamagnetic transition. On the other hand, the ground state becomes ferromagnetic under the condition of $0\leqq ac/b^2\leqq 3/16$. The theoretical magnetic phase diagram under the condition of $a>0$, $b<0$, and $c>0$ in the vicinity of the onset of ferromagnetism is shown in Fig. 1.[26] The ordinate corresponds to the temperature axis because $c\xi_P(T)^2/|b|$ also varies in proportion to T^2 at low temperatures. The abscissa is a measure of the concentration x, because ac/b^2 depends on x.[20] The phase transition between the paramagnetic and the ferromagnetic states occurs on the T_C line and the first-order phase transition between the paramagnetic and the ferromagnetic states takes place on the T_{C1} line. The first-order metamagnetic transition disappears on the T_0 line. Therefore, the first-order metamagnetic transition takes place between the T_0 and T_{C1} lines. In the figure, the ferromagnetic transition changes from the second order in the region $0\leqq ac/b^2\leqq 5/28$ to the first-order in the narrow region $5/28<ac/b^2<3/16$. Moreover, the metamagnetic transition occurs just above the Curie temperature T_{C1} in the narrow region, and T_{C1} steeply increases with decreasing ac/b^2 in the region $5/28<ac/b^2<3/16$.

Figure 2 shows the magnetization curves at 4.2 K for $Lu(Co_{1-x}Al_x)_2$ in the very narrow concentration range from $x=0.085$ to 0.100. The magnetization curves are very sensitive to the concentration of x.[14,19] The curves of $Lu(Co_{1-x}Al_x)_2$ with $x=0.085$ and 0.090 exhibit a clear metamagnetic transition from the paramagnetic to the ferro-

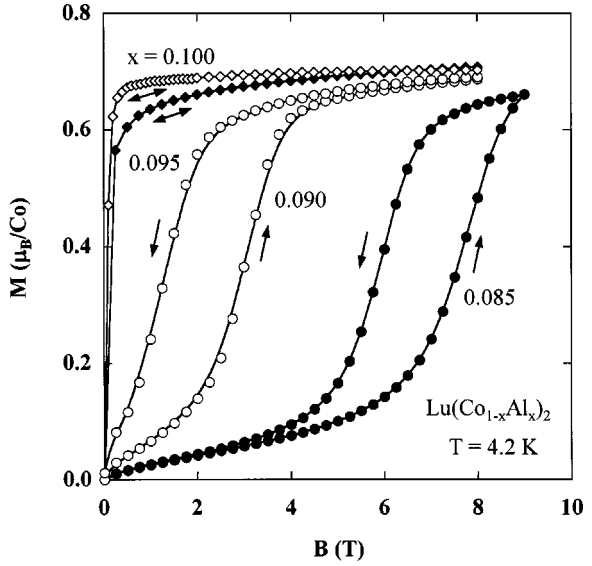

FIG. 2. Magnetization curves at 4.2 K for $Lu(Co_{1-x}Al_x)_2$ in the concentration range from $x=0.085$ to 0.100.

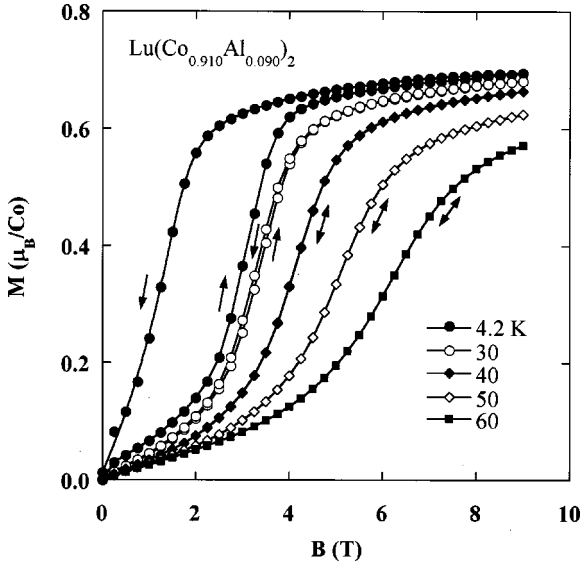

FIG. 4. Magnetization curves as a function of temperature for $Lu(Co_{0.910}Al_{0.090})_2$.

magnetic state, accompanied by an apparent hysteresis. It should be noted that the critical transition field B_C of the metamagnetic transition decreases with increasing x, and hence $Lu(Co_{1-x}Al_x)_2$ with $x=0.095$ and 0.100 become ferromagnetic in the ground state with almost the same magnitude of magnetization. From the figure, $x=0.095$ is very close to the critical concentration for the onset of ferromagnetism. The magnetization curves as a function of temperature are shown in Figs. 3 and 4 for the paramagnetic $Lu(Co_{1-x}Al_x)_2$ with $x=0.085$ and 0.090, respectively. The hysteresis in the curves becomes narrower with increasing temperature and eventually disappears in the temperature range between 30 and 40 K. By magnetization measurements from 30 to 40 K, the critical temperature T_0 from the first- to the second-order transition[20,26] for $Lu(Co_{1-x}Al_x)_2$ with $x=0.085$ and 0.090 was respectively determined to be 36 and 38 K, practically independent of x. It has been reported that the hysteresis of the metamagnetic transition in $Lu(Co_{0.94}Al_{0.06})_2$ disappears at around 60 K,[24] much higher than that of the present specimen with $x=0.085$. This difference would come from the different annealing condition responsible for homogeneity in composition, because the magnetic properties are very sensitive to homogeneity.[14,19]

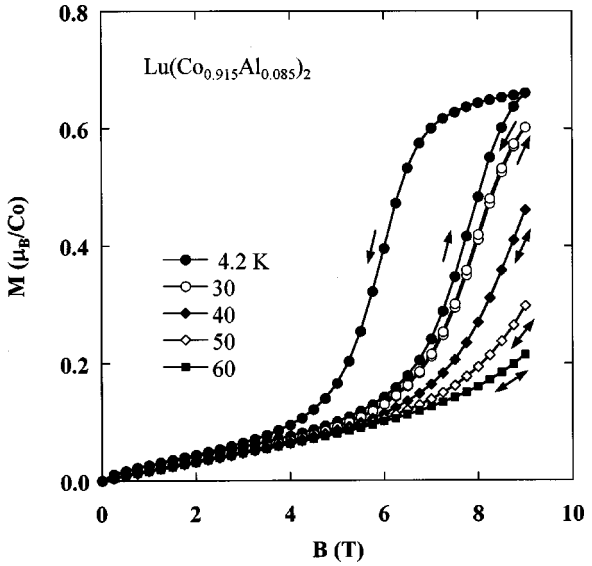

FIG. 3. Magnetization curves as a function of temperature for $Lu(Co_{0.915}Al_{0.085})_2$.

$Lu(Co_{1-x}Al_x)_2$ with $x=0.095$ and 0.100 are ferromagnetic in the ground state as shown in Fig. 2. In order to confirm the type of the ferromagnetic transition at the Curie temperature, the temperature dependence of magnetization was measured. Figure 5(a) shows the thermomagnetization curves in a magnetic field of 0.5 T for the ferromagnetic $Lu(Co_{1-x}Al_x)_2$ with $x=0.095$, 0.098, and 0.100. The compounds with $x=0.098$ and 0.100 are ferromagnetic and the Curie temperature T_C is determined to be 55 and 64 K, respectively, from the minimum point of $\partial M/\partial T$ in the thermomagnetization curve[17] in a magnetic field of 10 mT. The thermomagnetization curves exhibit no hysteresis, indicating that the second-order ferromagnetic transition. On the other hand, the curve of $Lu(Co_{0.905}Al_{0.095})_2$ exhibits a hysteresis around the Curie temperature $T_{C1}=28$ K, and the magnetization in the vicinity of the Curie temperature decreases more rapidly than that with $x=0.098$, reflecting the first-order ferromagnetic transition. Therefore, it is concluded that the first-order ferromagnetic transition occurs in $Lu(Co_{0.905}Al_{0.095})_2$. The first-order ferromagnetic transition is also observed in the thermomagnetization curve for $Lu(Co_{1-x}Ga_x)_2$ with $x=0.098$ as shown in Fig. 5(b). On the other hand, no hysteresis is confirmed in the curve for $x=0.100$. The latter behavior will be discussed in connection with Fig. 9.

The magnetization curves for $Lu(Co_{0.905}Al_{0.095})_2$ at several temperatures between 5 K and 80 K are shown in Fig. 6. The magnetization curve at 5 K exhibits a characteristic ferromagnetic behavior and a metamagnetic transition takes

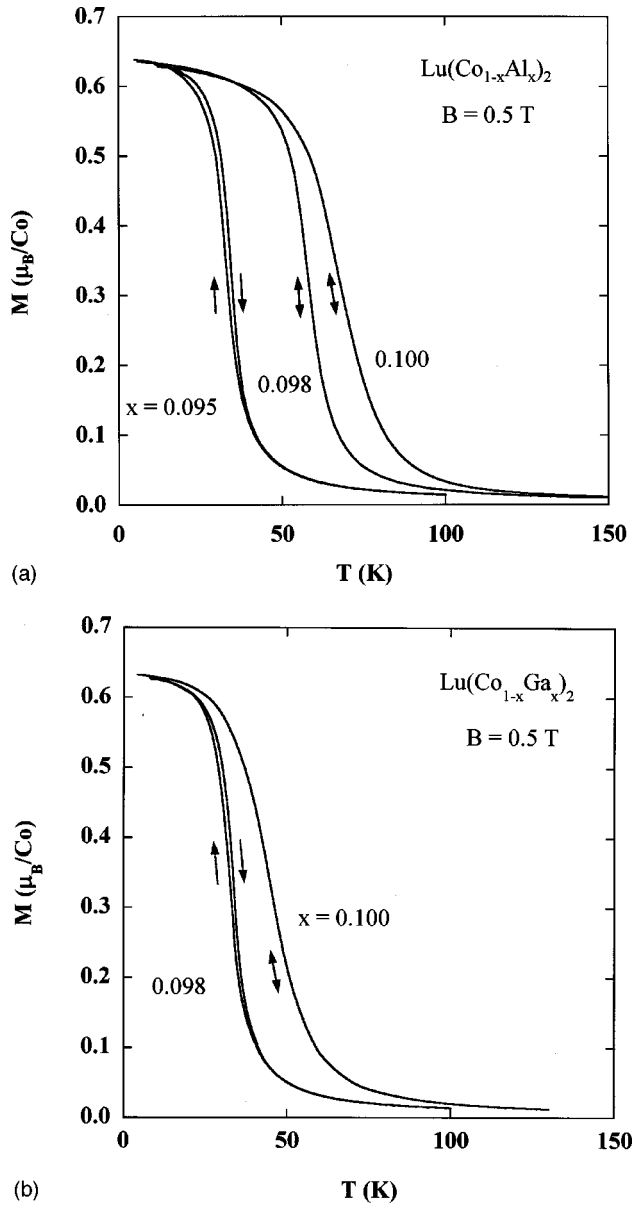

FIG. 5. (a) Heating and cooling thermomagnetization curves of $Lu(Co_{1-x}Al_x)_2$ with $x=0.095$, 0.098, and 0.100 in a magnetic field of 0.5 T. (b) Heating and cooling thermomagnetization curves of $Lu(Co_{1-x}Ga_x)_2$ with $x=0.098$ and 0.100 in a magnetic field of 0.5 T.

place just above $T_{C1}=28$ K, accompanied by hystereses as seen from the curves at 30 K. The magnetization curves of $Lu(Co_{0.902}Ga_{0.098})_2$ are similar to those of $Lu(Co_{0.905}Al_{0.095})_2$. In order to elucidate such behaviors, the magnetization curves were measured every 2 K just above T_{C1} in magnetic fields up to 3 T for $Lu(Co_{0.905}Al_{0.095})_2$. The first-order metamagnetic transition becomes broad with increasing temperature, resulting in the increase of B_C and finally disappears in the temperature range between 40 and 42 K as presented in Fig. 7. Such magnetic curves just above T_{C1} are also observed in the paramagnetic $Lu(Co_{1-x}Al_x)_2$ with $x=0.085$ and 0.090 as given in Figs. 3 and 4. The value of T_0 for $Lu(Co_{0.905}Al_{0.095})_2$ is estimated to be 42 K, slightly

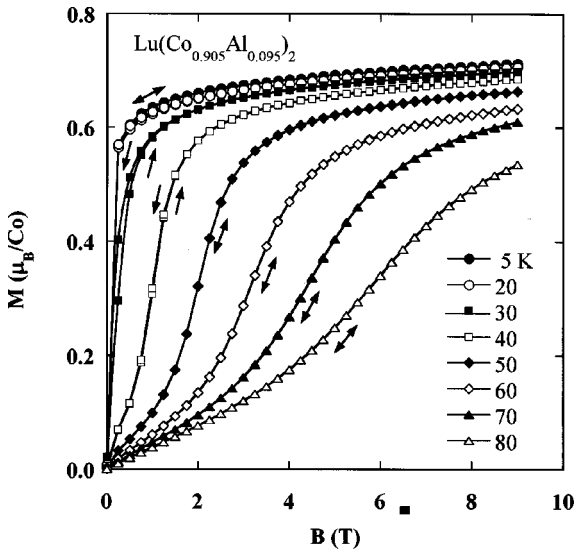

FIG. 6. Magnetization curves as a function of temperature for $Lu(Co_{0.905}Al_{0.095})_2$.

higher than that for the compounds with 0.085 and 0.090. This first-order ferromagnetic transition is similar to that observed in $Co(S_{0.90}Se_{0.10})_2$ pyrite[27] and $La(Fe_{0.88}Si_{0.12})_2$ with a $NaZn_{13}$-type structure,[28–30] which have a ferromagnetic ground state and exhibit an itinerant-electron metamagnetic transition above T_{C1} by applying external magnetic fields.

It has been theoretically discussed that the itinerant-electron metamagnetic transition is closely concerned with a double minimum structure in the paramagnetic ($M=0$) and the ferromagnetic ($M\neq 0$) states in the magnetic-free energy as a function of magnetization $F(M)$. When the minimum in the ferromagnetic state is lower than that in the paramagnetic state at 0 K, the system becomes ferromagnetic in the ground state. However, the thermal activation makes the ferromagnetic energy minimum higher than the paramagnetic one,

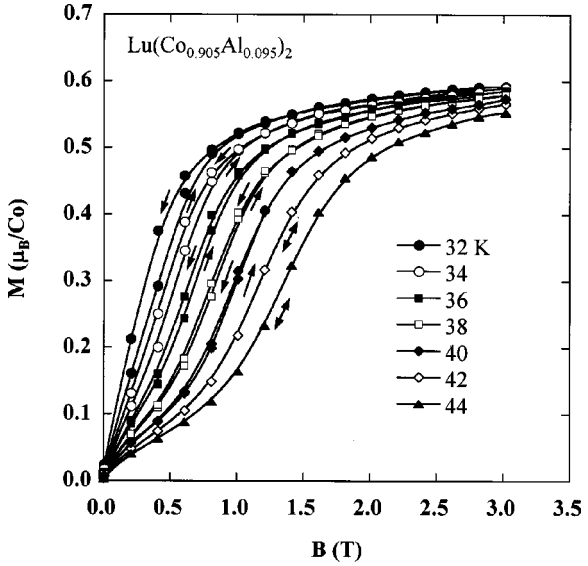

FIG. 7. Magnetization curves for $Lu(Co_{0.905}Al_{0.095})_2$ just above the Curie temperature T_{C1} in magnetic fields up to 3 T.

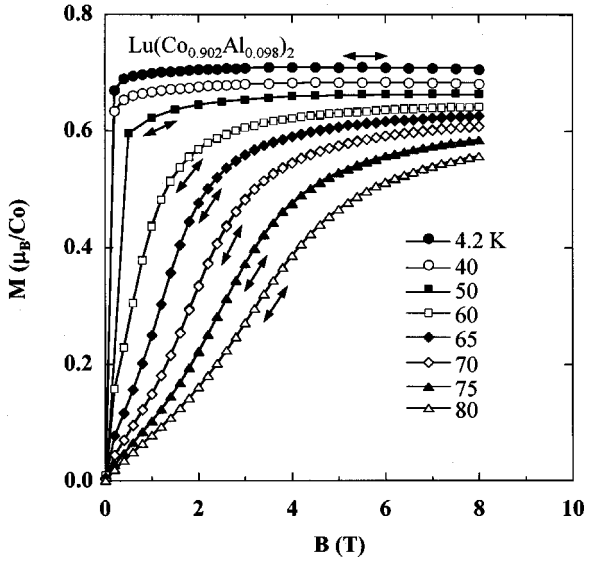

FIG. 8. Magnetization curves as a function of temperature for $Lu(Co_{0.902}Al_{0.098})_2$.

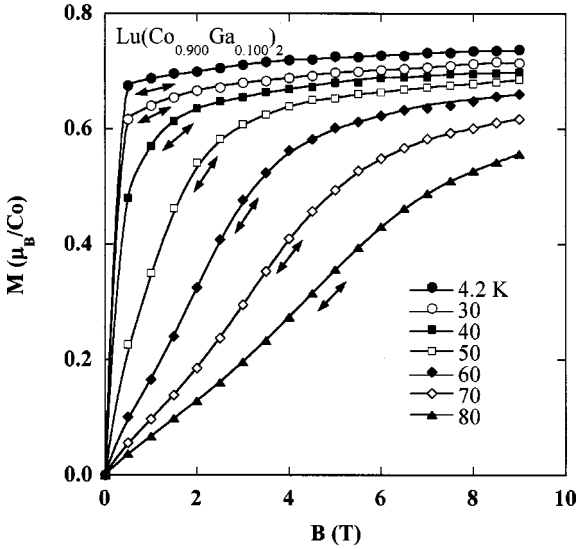

FIG. 9. Magnetization curves as a function of temperature for $Lu(Co_{0.900}Ga_{0.100})_2$.

resulting in the paramagnetic state. On applying magnetic field, the ferromagnetic energy minimum again becomes lower than the paramagnetic one and the first-order metamagnetic transition from the paramagnetic to the ferromagnetic state is induced. Therefore, the metamagnetic transition in the paramagnetic temperature region just above T_{C1} for $Lu(Co_{0.905}Al_{0.095})_2$ is attributed to the double minimum structure of the magnetic-free energy as a function of magnetization $F(M)$.

Figure 8 shows the magnetization curves as a function of temperature for $Lu(Co_{0.902}Al_{0.098})_2$ having a second-order ferromagnetic transition. In the paramagnetic temperature range, above $T_C = 55$ K, as shown in the figure, there are no hysteresis characterized as the metamagnetic transition, showing an *S*-shape with a slight curvature. This means that $Lu(Co_{0.902}Al_{0.098})_2$ exhibits a second-order-type metamagnetic transition in the paramagnetic temperature region by applying external magnetic field. Figure 9 shows the magnetization curves as a function of temperature for $Lu(Co_{0.900}Ga_{0.100})_2$ with $T_{C1} = 37$ K. It is expected that this compound also exhibits the metamagnetic transition in the paramagnetic region on applying external magnetic field. However, the magnetization curves above T_{C1} exhibit a slight *S*-shape-like curvature without any hysteresis. This is because T_{C1} is very close to T_0. Note that the thermomagnetization curve around T_{C1} given in Fig. 5(b) also exhibits no hysteresis due to the same reason. On the other hand, recently, the volume change in $Lu(Co_{1-x}Ga_x)_2$ at various temperatures has been confirmed by x-ray diffraction. In a temperature range from 20 to 40 K, a paramagnetic cubic phase and a ferromagnetic tetragonal phase coexist in $Lu(Co_{0.900}Al_{0.100})_2$, indicating the first-order ferromagnetic transition.[31]

The itinerant-electron metamagnetism has been discussed in terms of Yamada and Moriya theories based on the Ginzburg-Landau-type free energy expansion.[20,26] For the onset of the metamagnetic transition, the fourth-order term of the Landau expansion $B(T)$ should be negative, giving a negative slope of the Arrott plot. In order to discuss the condition for the metamagnetic transition, the Arrott plots for $Lu(Co_{1-x}Al_x)_2$ with $x = 0.095$, 0.098, and 0.100 just above T_C are presented in Fig. 10. A negative slope is confirmed in the plots for $x = 0.095$, indicating that the fourth-order term of the Landau expansion $B(T)$ is negative. The plots for $x = 0.098$ and 0.100 do not show a negative slope but an inflection which is also explained by the negative fourth-order term of the Landau expansion coefficient $B(T)$. Our previous study of the pressure effects for $Lu(Co_{0.900}Al_{0.100})_2$ revealed that the metamagnetic transition from the paramagnetic state induced by pressure to the ferromagnetic state is caused by applying external magnetic field.[19] Moreover, the Arrott-plots near the onset of ferromagnetism above T_{C1} in-

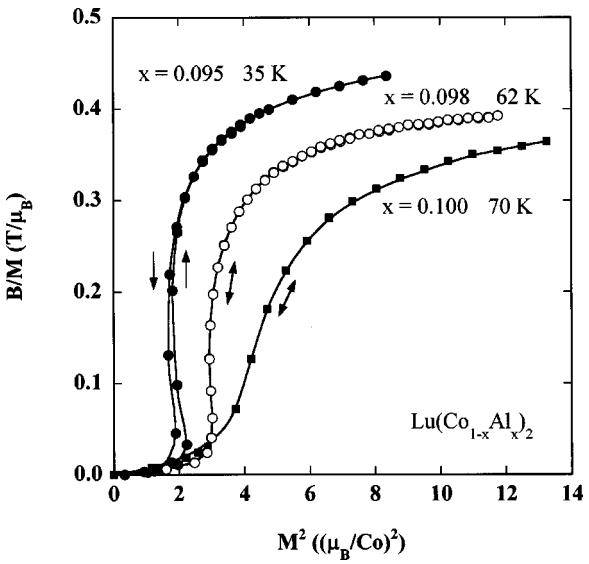

FIG. 10. Arrott plots for $Lu(Co_{1-x}Al_x)_2$. At 35 K for $x = 0.095$, at 62 K for $x = 0.098$, and at 70 K for $x = 0.100$.

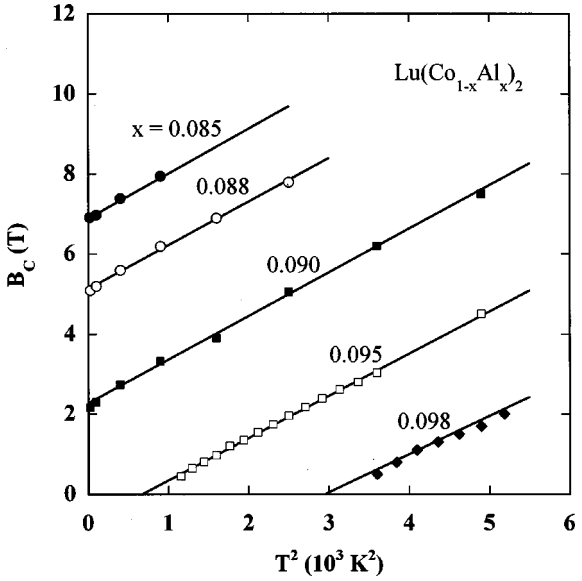

FIG. 11. The critical transition field B_C against the square of temperature T^2 for $Lu(Co_{1-x}Al_x)_2$ in the concentration range from $x=0.085$ to 0.098.

dicate a negative sloop for $Lu(Co_{0.900}Ga_{0.100})_2$.[17] Therefore, it is considered that $F(M)$ of $Lu(Co_{1-x}M_x)_2$ ($M=$Al and Ga) systems have a double minimum up to the composition in the vicinity of the onset of ferromagnetism.

For the compounds in the vicinity of the onset of ferromagnetism, it is difficult to determine the Curie temperature from the thermomagnetization curves, because the minimum point of $\partial M/\partial T$ sensitively depends on the magnitude of applying magnetic fields. In the present study, therefore, the following method has been adopted. It has been pointed out that B_C for itinerant-electron metamagnetic systems such as paramagnetic $Lu(Co_{1-x}Ga_x)_2$ Laves-phase[15,16] and $Co(S_{1-x}Se_x)_2$ pyrite[27] compounds is proportional to the

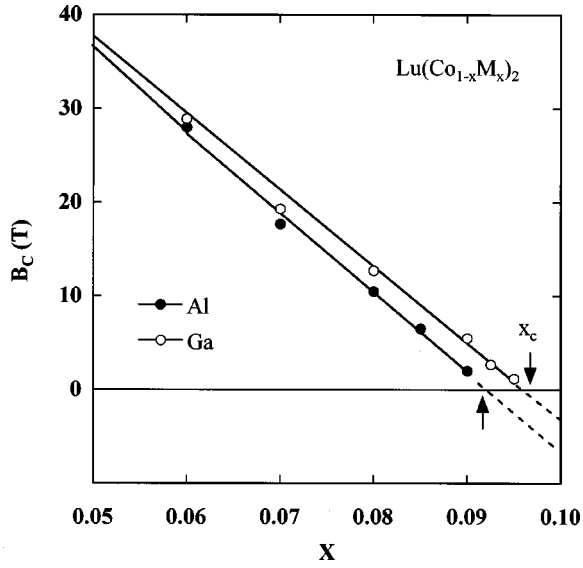

FIG. 12. Concentration dependence of the critical transition field B_C at 0 K for $Lu(Co_{1-x}M_x)_2$ ($M=$Al and Ga).

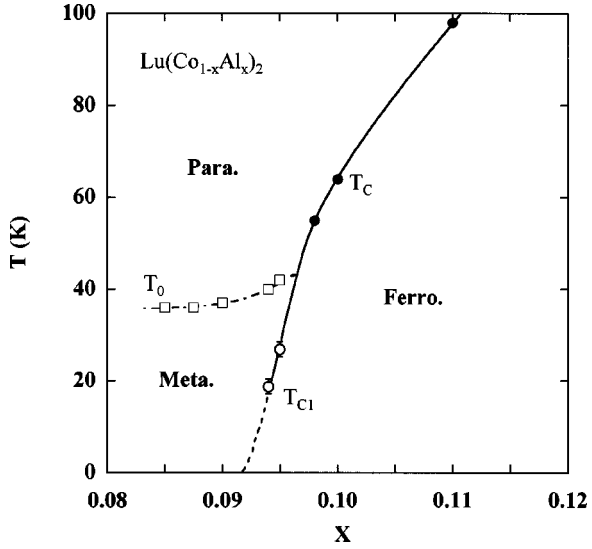

FIG. 13. Magnetic phase diagram of $Lu(Co_{1-x}Al_x)_2$ system in the vicinity of the critical concentration.

square of temperature T^2 at low temperatures. Furthermore, the observed T^2 dependence of B_C was elucidated from the mean-square amplitude of spin fluctuations, $\xi_P(T)^2$, which increases as T^2 at low temperatures.[20] In order to determine T_{C1}, B_C as a function of T^2 for $Lu(Co_{0.905}Al_{0.095})_2$ is shown in Fig. 11, together with the data of $Lu(Co_{1-x}Al_x)_2$ with $x=0.085$, 0.088, 0.090, and 0.098 for comparison. The value of B_C was defined as the average of the lower and higher critical transition fields determined at the peaks of the differential susceptibility in increasing and decreasing fields, respectively. In the figure, B_C is proportional to T^2 and the ferromagnetic compound with $x=0.095$ and the paramagnetic compounds with $x=0.085$, 0.088, and 0.090 have the same slope at low temperatures. As discussed above, the observed T^2 dependence of B_C originates from the thermal spin fluctuations $\xi_P(T)^2$ proportional to T^2 at low temperatures. The slope hardly depends on x, implying that the thermal spin fluctuation in the ferromagnetic compound with $x=0.095$ above T_{C1} is equal to that in the paramagnetic compounds. The value of B_C for the ferromagnet with $x=0.095$ should become zero at T_{C1}. From this relation, T_{C1} is estimated to be 26 K, practically in agreement with T_{C1} obtained by the thermomagnetization curves given in Fig. 5(a). In order to determine the critical concentration x_C of the onset of ferromagnetism, the concentration dependence of B_C at 0 K is shown in Fig. 12, together with the data of $Lu(Co_{1-x}Ga_x)_2$. Here B_C at 0 K is estimated by a linear extrapolation to $T=0$ in Fig. 11. As shown in the figure, B_C linearly decreases with increasing x. The critical concentration x_C at which B_C becomes zero corresponds to the onset of ferromagnetism. From this relation, the critical concentration x_C for $Lu(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ is estimated to be 0.092 and 0.096, respectively, indicating that $x=0.095$ for $Lu(Co_{1-x}Al_x)_2$ and 0.100 for $Lu(Co_{1-x}Ga_x)_2$ are very close to the critical concentration for the onset of ferromagnetism.

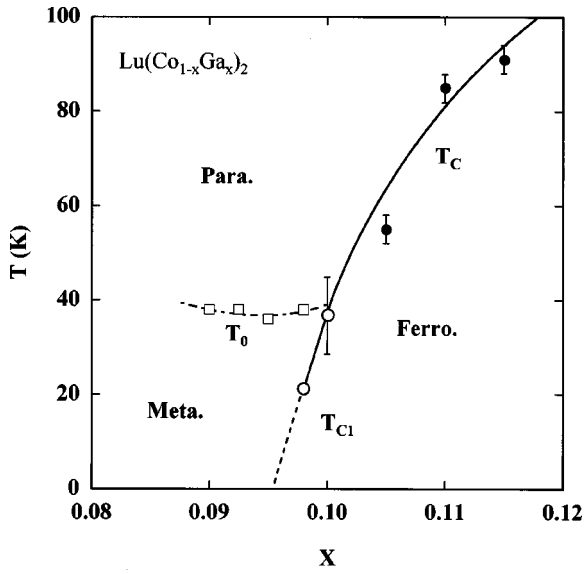

FIG. 14. Magnetic phase diagram of $Lu(Co_{1-x}Ga_x)_2$ system in the vicinity of the critical concentration.

Using the observed values of the Curie temperatures, T_C and T_{C1}, and the critical temperature T_0, the magnetic phase diagrams in the x-T plane in the vicinity of the critical concentration between metamagnetism and ferromagnetism for $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) systems were obtained as shown in Figs. 13 and 14, respectively. The value of T_{C1} steeply increases near the onset of ferromagnetism with increasing x, whereas T_0 is not so sensitive to the concentration of x. The experimental phase diagrams in the vicinity of the onset of ferromagnetism for (M=Al and Ga) systems reproduce the theoretical phase diagram as seen from Fig. 1. Note that the increase of x corresponds to the decrease of ac/b^2 shown Fig. 1. That is to say, Figs. 13 and 14 are equivalent to the view of the reverse side of Fig. 1. The ordinate in the theoretical phase diagram corresponds to the magnitude of the mean-square amplitude of spin fluctuations $\xi_P(T)^2$ at the Curie temperature as shown in Fig. 1. Therefore, the rapid increase in the Curie temperature of $Lu(Co_{1-x}M_x)_2$ with increasing x indicates the increase of $\xi_P(T)^2$ at the Curie temperature. Moreover, the onset of ferromagnetism in the ground state and the metamagnetic transition above T_{C1} by applying magnetic fields is also explained by the theoretical phase diagram based on the conditions of $a>0$, $b<0$ and $c>0$. What has to be noted is that the first-order ferromagnetic transition occurs in very narrow concentration ranges. Accordingly, the striking difference between the magnetic properties of $Lu(Co_{0.905}Al_{x0.095})_2$ in Fig. 6 and $Lu(Co_{0.902}Al_{x0.098})_2$ in Fig. 8 is explained on the basis of the proposed magnetic phase diagram shown in Fig. 1. Furthermore, in accordance with the results given in Fig. 9, the value of T_{C1} for $Lu(Co_{0.900}Ga_{0.100})$ almost meets the T_0 line as seen from Fig. 14.

Recently, it has been pointed out that the region of the first-order ferromagnetic transition at T_{C1} becomes wider by considering the magnetovolume coupling energy.[32] The present compound systems exhibit pronounced magnetovolume effects such as large pressure effects on the magnetization and the Curie temperature,[17–19,33] and a remarkable thermal expansion anomaly.[9,31] By considering the magnetovolume coupling energy, the Landau expansion coefficients should be replaced by $\tilde{a}=a+2\kappa C_{m\nu}P$, $\tilde{b}=b-2\kappa C_{m\nu}^2$, and $\tilde{c}=c$.[34] The first-order ferromagnetic transition occurs under the following condition:[32]

$$\frac{5}{28}-\eta<\frac{\tilde{a}\tilde{c}}{\tilde{b}^2}<\frac{3}{16} \quad \text{with} \quad \eta=\frac{2}{7|\tilde{b}|}\kappa C_{m\nu}^2. \tag{4}$$

Accordingly, the magnetovolume coupling energy widens the region of the first-order ferromagnetic transition at T_{C1}. The type of ferromagnetic transition changes to the second-order under the condition of $0\leqq\tilde{a}\tilde{c}/\tilde{b}^2\leqq 5/28-\eta$. The values of $\tilde{a}$, $\tilde{b}$, $\tilde{c}$, and $\kappa C_{m\nu}$ for $Lu(Co_{0.900}M_{0.100})_2$ have been determined by using magnetic behaviors under pressure.[17,19] The values of η and 5/28-η are given in Table I, together with these determined values. 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 $5/28-\eta$. This means that the ferromagnetic transition is of the second order. The experimental phase diagram given in Fig. 13 is consistent with the present value. Furthermore, 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 condition between the first- and the second-order ferromagnetic transition. The obtained value is in good agreement with the experimental phase diagram given in Fig. 14. Therefore, it is concluded that the present phase diagrams in the vicinity of the onset of ferromagnetism for $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) are well explained by the theoretical phase diagram.

The present results are in contrast with previous experimental data. That is, the phase diagram obtained from the present results for $Lu(Co_{1-x}Al_x)_2$ is very different from that proposed by Iijima *et al.*[24] Furthermore, the magnetic transition from a weakly ferromagnetic to strongly ferromagnetic state[23,35] was not ascertained in the present study. It should be concluded that these differences are closely associated with homogeneity in composition because the magnetic

TABLE I. The values of η and 5/28-2η, together with the Landau expansion coefficients $\tilde{a}$, $\tilde{b}$, $\tilde{c}$, and the magnetoelastic coupling constant $\kappa C_{m\nu}$ for $Lu(Co_{0.900}M_{0.100})_2$ Laves-phase compounds (M=Al and Ga).

	$\tilde{a}$ (10^2 cm^3/emu)	$\tilde{b}$ (10^{-2} cm^3/erg)	$\tilde{c}$ (10^{-6} cm^6/erg^2)	$\kappa C_{m\nu}$ ($10^{-3}\mu_B^{-2}$)	η	5/28-η
$Lu(Co_{0.900}Al_{0.100})_2$	6.9	−7.8	1.2	6.6	0.011	0.17
$Lu(Co_{0.900}Ga_{0.100})_2$	7.6	−8.3	1.4	7.4	0.014	0.16

properties of $Lu(Co_{1-x}M_x)_2$ (M = Al and Ga) exhibit a precipitous concentration dependence.[19]

IV. CONCLUSION

By using samples with a high quality of compositional homogeneity, the magnetization curves at various temperatures and thermomagnetization curves of $Lu(Co_{1-x}M_x)_2$ (M = Al and Ga) Laves-phase compounds have been measured. The type of the transition at the Curie temperature in the vicinity of the critical concentration of the onset of ferromagnetism has been investigated. From the observed Curie temperatures, T_C and T_{C1}, and the critical temperature T_0 between the first- and the second-order transition, the magnetic phase diagrams for these systems have been established. The main results are summarized as follows.

(a) The ferromagnetic transition changes from the first to the second order with increasing x and the first-order ferromagnetic transition occurs in a restricted narrow concentration region.

(b) The critical transition field B_C of the metamagnetic transition at 0 K linearly decreases with increasing x, indicating that the critical concentration x_C is estimated to be 0.094 for $Lu(Co_{1-x}Al_x)_2$ and 0.096 for $Lu(Co_{1-x}Ga_x)_2$.

(c) In the ground state, ferromagnetic $Lu(Co_{0.905}Al_{0.095})_2$ and $Lu(Co_{0.902}Al_{0.098})_2$ exhibit a hysteresis around T_{C1}, indicating a first-order ferromagnetic transition.

(d) Just above T_{C1}, a metamagnetic transition from the paramagnetic to the ferromagnetic state takes place in $Lu(Co_{0.905}Al_{0.095})_2$ and $Lu(Co_{0.902}Al_{0.098})_2$.

(e) The magnetic phase diagrams of $Lu(Co_{1-x}M_x)_2$ (M = Al and Ga) in the vicinity of the onset of ferromagnetism are well explained by the Moriya and Yamada theories in contrast with the previous experimental phase diagram.

ACKNOWLEDGMENT

One of the authors (H.S.) would like to thank the Japan Society for Promotion of Science for Young Scientists for support.

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