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Itinerant-electron metamagnetism of the Laves-phase compounds $Lu(Co_{1-x}Ga_x)_2$ under high pressures with high magnetic fields

H. Saito, T. Yokoyama, and K. Fukamichi
Department of Materials Science, Graduate School of Engineering, Tohoku University, Aoba-yama 02, Sendai 980-8579, Japan

K. Kamishima and T. Goto
Institute for Solid State Physics, The University of Tokyo, Roppongi, Minato-ku, Tokyo 106-0032, Japan
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Magnetic properties of the Laves phase compounds $Lu(Co_{1-x}Ga_x)_2$ ($0.10 \leqq x \leqq 0.22$) have been investigated under the duplicate conditions, i.e., high magnetic fields with high pressures. The pressure effect on the Curie temperature T_C is extremely large in the vicinity of $x=0.10$, which is close to the critical concentration for the onset of ferromagnetism. The spontaneous magnetization M_S around $x=0.10$ is drastically decreased and a paramagnetic state is induced by applying pressure. The paramagnetic state is changed into a ferromagnetic state by applying magnetic field, accompanied by a metamagnetic transition. The Landau coefficients up to the sixth-order terms of the magnetization for $x=0.10$ were estimated from the experimental data. The pressure dependence of the critical transition field H_C for $x=0.10$ is consistent with the results calculated from the obtained coefficients. For the compounds with $x \leqq 0.15$, the pressure coefficient of the spontaneous magnetization, $\partial \ln M_S/\partial P$, is much larger than the pressure coefficient of the Curie temperature, $\partial \ln T_C/\partial P$, implying that there is a peak of the density of state just below the Fermi energy. The variation of $\partial \ln T_C/\partial \ln M_S$ as a function of x near the critical concentration of the onset of ferromagnetism can be explained by the theory for itinerant ferromagnets having a negative coefficient b of the fourth-order term in the Landau expansion. [S0163-1829(99)02413-3]

I. INTRODUCTION

Laves phase compounds RCo_2 (R=Y and Lu) are enhanced Pauli paramagnets which exhibit a broad maximum in the temperature dependence of susceptibility[1,2] due to their 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 occur by applying high magnetic fields.[5,6] The critical field H_C of MT has been demonstrated to be about 69 for YCo_2,[7] and 74 T for $LuCo_2$.[8] By replacing Co with Al, a significant reduction in H_C and the onset of weak ferromagnetism have been observed in $Y(Co_{1-x}Al_x)_2$,[9–11] consistent with a band calculation.[12] For $LuCo_2$, H_C also decreases and a ferromagnetic state becomes stable by the replacement of Co with Al (Refs. 11, 13, and 14) or Ga.[15,16] A characteristic behavior of $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) systems is a sharp transition in relatively low magnetic fields without a remarkable change in the magnitude of magnetization,[14–16] in contrast to $Y(Co_{1-x}Al_x)_2$.[9–11] Moreover, the spontaneous magnetization M_S and the Curie temperature T_C of $Lu(Co_{1-x}M_x)_2$ are several times as large as those of $Y(Co_{1-x}Al_x)_2$.[16–18] These results suggest that the shape of the DOS for $Lu(Co_{1-x}M_x)_2$ differs from that of $Y(Co_{1-x}Al_x)_2$. However, the effects of replacing Co with M on their band structures have not been discussed yet.

Magnetic properties of itinerant-electron systems strongly depend on the band structure. Therefore the values of M_S, T_C, and H_C for this system are expected to be sensitive to the pressure because the application of pressure results in an increase of the bandwidth, reducing the DOS at E_F. Pressure dependences of these values have been discussed by taking into account the effect of spin fluctuations on the magnetic free energy given by the Landau expansion, revealing that T_C and H_C are significantly influenced by pressure.[19–22]

In the present study, we have measured the magnetic properties of $Lu(Co_{1-x}Ga_x)_2$ compounds under the duplicate conditions, i.e., high pressures with high magnetic fields. The experimental results of the pressure dependences of M_S, T_C, and H_C are analyzed in terms of the Landau expansion by considering spin fluctuations.[19–22] Moreover, the effect of replacing Co with Ga on their band structures will be discussed.

II. EXPERIMENTAL

The alloying of Lu, Co, and Ga was carried out by arc melting in an argon gas atmosphere. The Lu composition was adjusted slightly higher than the stoichiometric composition as $Lu_{34}(Co_{1-x}Ga_x)_{66}$ to avoid any other ferromagnetic precipitates such as $LuCo_3$. The ingots were melted several times, followed by annealing at 1223 K for a week in an evacuated quartz tube for homogenization, and subsequently quenched into water. The oxidized surface of the annealed specimen was mechanically removed. X-ray powder-diffraction analyses identified the specimens as a single $C15$-type Laves phase.

Magnetization measurements at ambient pressure were carried out up to 9 T with an extraction-type magnetometer. Magnetization measurements under high pressures were carried out with an extraction-type magnetometer equipped with

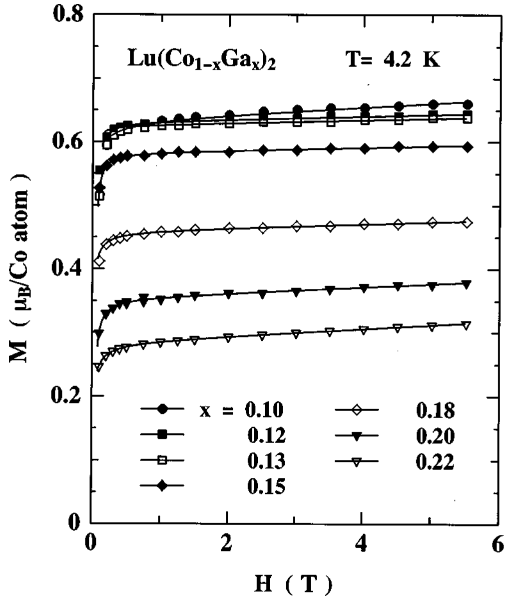

FIG. 1. Magnetization curves at 4.2 K for $Lu(Co_{1-x}Ga_x)_2$ compounds in the concentration range from $x=0.10$ to 0.22 at ambient pressure.

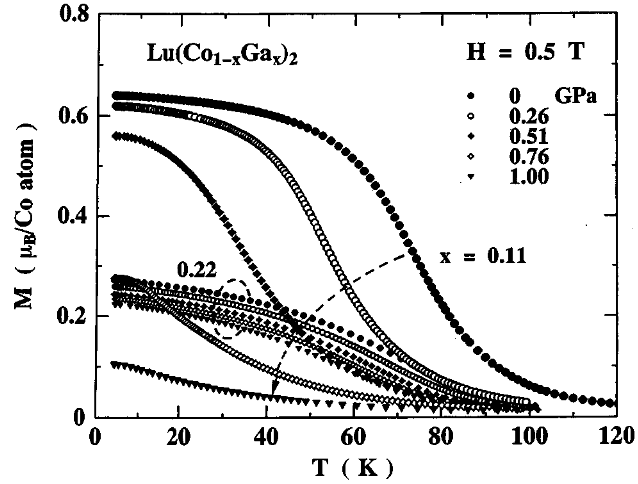

FIG. 2. Temperature dependence of magnetization at various pressures for $Lu(Co_{1-x}Ga_x)_2$ compounds with $x=0.11$ and 0.22 in a magnetic field of 0.5 T.

a nonmagnetic pressure clamp made of a Cu-3 wt. % Ti alloy in magnetic fields up to 9 T.[23] The susceptibility of the clamp is extremely small, that is, 5×10^{-8} emu/g at 1.8 K.[23] Hydrostatic pressures were applied up to 1.00 GPa in a Teflon cell filled with a Fluorinert in the clamp cylinder, and calibrated by measuring the shift of the superconducting transition temperature of Pb. Forced-volume magnetostriction was measured up to 5 T by a capacitance method.

III. RESULTS AND DISCUSSION

The magnetic properties of the Laves phase $Lu(Co_{1-x}M_x)_2$ (M=Al and Ga) systems are very sensitive to annealing condition. Our previous studies revealed that insufficient homogenization of the composition results in a concentration gradient of Al or Ga in the crystal grains.[14,16] On the other hand, the spontaneous magnetization M_S increases significantly in a limited narrow concentration range.[14,16] Therefore it should be emphasized that insufficient homogenization leads to a strange magnetization curve around the critical concentration of the onset of ferromagnetism associated with the coexistence MT due to inhomogeneity as $Lu(Co_{1-x}Al_x)_2$ compounds.[13] In order to confirm homogeneity in the $Lu(Co_{1-x}Ga_x)_2$, the magnetization curves at 4.2 K at ambient pressure were measured in the concentration range from $x=0.10$ to 0.22. All of the samples exhibit a characteristic ferromagnetic magnetization curve as shown in Fig. 1 even near the critical concentration without any strange magnetization curves. Therefore, it is considered that the samples are excellent in homogeneity.

Shown in Fig. 2 is the temperature dependence of magnetization M at various pressures for $x=0.11$ and 0.22 in a magnetic field of 0.5 T. The magnetizations of both compounds clearly show a negative pressure dependence. However, the magnetization of $x=0.11$ strongly depends on the pressure compared to that of $x=0.22$, indicating that the ferromagnetic state is unstable due to the concentration near the onset of ferromagnetism.

For the compounds near the onset of ferromagnetism, it is difficult to determine the Curie temperature T_C from the M-T curves. In the present study, the following method has been adopted. Figure 3 shows the magnetization curves for $x=0.10$ just above T_C at ambient pressure. Each curve shows a inflection point, suggesting the occurrence of the MT. It has been reported that the critical transition field of the MT H_C for itinerant-electron metamagnetic systems such as $Lu(Co_{1-x}Ga_x)_2$ (Refs. 15 and 16) and $Co(S_{1-x}Se_x)_2$ (Ref. 23) increases in proportion to the square of temperature T^2. Using this relation, the temperature of $H_C=0$, namely, T_C, is obtainable. The temperature dependence of H_C versus T^2 plot for $x=0.10$ is displayed in Fig. 4. The value of H_C was determined as the peak of differential susceptibility. The

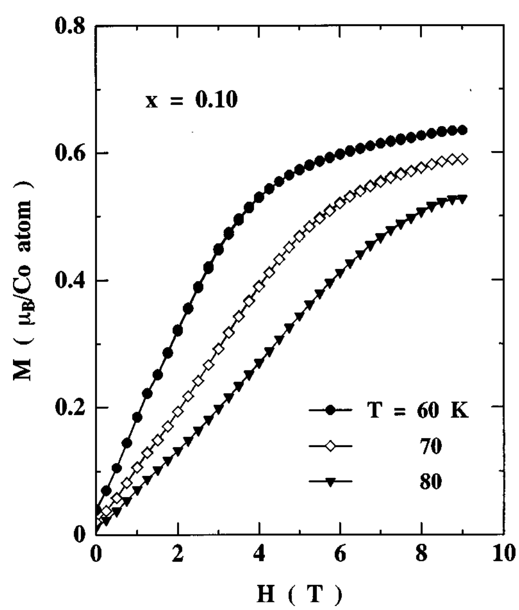

FIG. 3. Magnetization curves for $Lu(Co_{0.90}Ga_{0.10})_2$ at ambient pressure just above the Curie temperature T_C.

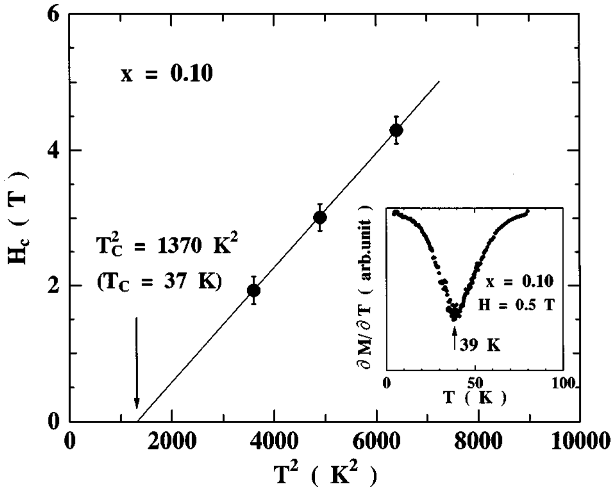

FIG. 4. Temperature dependence of the critical fields H_C for $Lu(Co_{0.90}Ga_{0.10})_2$ in the form of H_C versus T^2. The inset shows the temperature dependence of the temperature derivative of magnetization, $\partial M/\partial T$, for $x=0.10$ at ambient pressure in a magnetic field of 0.5 T.

plot shows a good linear relationship and T_C^2 given by a linear extrapolation to $H_C=0$ is estimated to be 1370 K^2 ($T_C=37$ K). The inset shows the temperature dependence of the temperature derivative of magnetization, $\partial M/\partial T$, for $x=0.10$ at ambient pressure in field of 0.5 T. As indicated by the arrow, the curve takes a minimum at $T=39$ K, practically in accord with T_C obtained by the H_C-T^2 relation. Shown in Fig. 5 is the pressure dependence of T_C determined from the minimum point for $Lu(Co_{1-x}Ga_x)_2$. In all of the compounds, T_C decreases significantly with increasing pressure. For $x=0.10$ and 0.11, the ferromagnetic state disappears and the paramagnetic state is developed by applying pressure. Figure 6 shows the concentration dependence of the pressure derivative of the Curie temperature, $\partial T_C/\partial P$, for $Lu(Co_{1-x}Ga_x)_2$. The magnitude of $\partial T_C/\partial P$ first de-

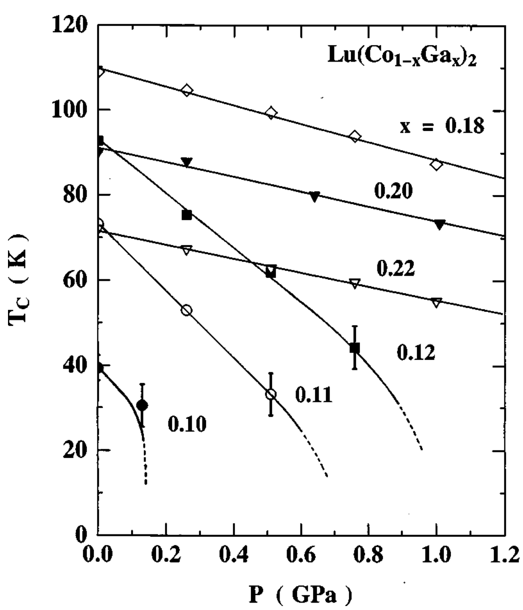

FIG. 5. Pressure dependence the Curie temperature T_C for $Lu(Co_{1-x}Ga_x)_2$ compounds.

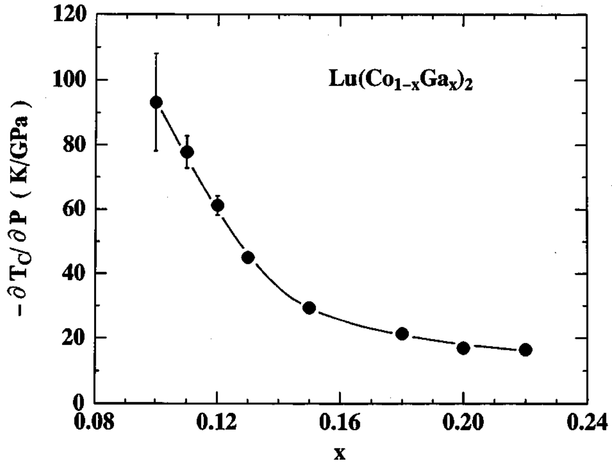

FIG. 6. Concentration dependence of the pressure derivative of the Curie temperature, $\partial T_C/\partial P$, for $Lu(Co_{1-x}Ga_x)_2$ compounds.

creases rapidly and becomes sluggish with increasing Ga concentration. It should be noted that the magnitude of $\partial T_C/\partial P$ is extremely large in the vicinity of $x=0.10$, close to the critical concentration of the onset of ferromagnetism.

To explain the present results mentioned above, the effect of spin fluctuations should be taken into account because magnetic properties of itinerant-electron systems at finite temperatures are strongly influenced by spin fluctuations. The magnetic free energy F of itinerant-electron systems is expressed by,

$$F=\frac{1}{2}aM^2+\frac{1}{4}bM^4+\frac{1}{6}cM^6, \tag{1}$$

where the coefficients a, b, and c are determined from the shape of the density of states (DOS) around the Fermi energy E_F.[24,25] The conditions of $a>0$, $b<0$, and $c>0$ with $\frac{3}{16}<ac/b^2<\frac{9}{20}$ are necessary for the metamagnetic transition MT.[26] A negative b is related to a positive curvature of the DOS at E_F (Ref. 24) as well as negative mode-mode couplings among spin fluctuations.[27] At finite temperatures, the coefficients in Eq. (1) are renormalized by thermal spin fluctuations.[19] According to the spin-fluctuation theory, the pressure dependence of T_C for ferromagnets under the conditions of $a>0$, $b<0$, and $c>0$ can be discussed by considering the pressure effect of the mean-square amplitude of spin fluctuations at T_C, $\xi_p(T_C)^2$, which is given by the following expression:[20]

$$\xi(T_C)^2=\frac{3|b|}{14c}\left(1+2\sqrt{\frac{7}{5}}\sqrt{\frac{5}{28}-\frac{(a+2\kappa C_{\mathrm{mv}}P)c}{b^2}}\right), \tag{2}$$

where κ and C_{mv} are the compressibility and the magnetovolume coupling constant, respectively. Hence $\partial\xi_p(T_C)^2/\partial P$ at $P=0$ is expressed as[20]

$$\frac{\partial\xi_p(T_C)^2}{\partial P}=-\frac{3\kappa C_{\mathrm{mv}}}{\sqrt{7}|b|}\left(\frac{5}{28}-\frac{ac}{b^2}\right)^{-1/2}. \tag{3}$$

The value of $\partial\xi_p(T_C)^2/\partial P$ is proportional to $\partial T_C^2/\partial P$, since $\xi_p(T_C)^2$ is proportional to T_C^2 at low temperatures.[19] Equa-

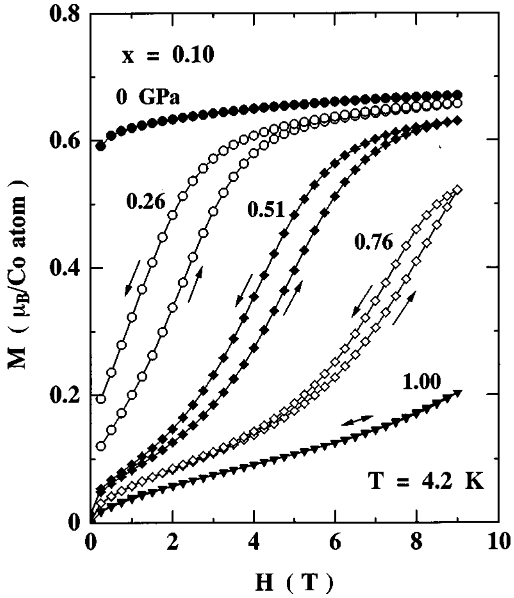

FIG. 7. Pressure and magnetic field dependences of the magnetization for $Lu(Co_{0.90}Ga_{0.10})_2$ at 4.2 K.

tion (3) indicates that a significantly large value of $\partial T_C/\partial P$ with a negative sign is observed near the critical concentration of the onset of ferromagnetism because the condition for $ac/b^2=\frac{5}{28}$ is very close to the paramagnetic state.[19] The band calculations for Invar-type alloys such as Fe_3Ni and Fe_3Pt which exhibit a large $\partial T_C/\partial P$ lead to the conditions of $a>0$, $b<0$, and $c>0$ with $ac/b^2\approx\frac{5}{28}$.[28] Consequently, the present result can be explained by the same conditions of the Landau coefficients.

In order to prove the negative sign of b for the present compounds, we have measured the magnetization process under pressures because the negative b is necessary for the MT. Shown in Figs. 7 and 8 are the pressure and field dependences of the magnetization for $x=0.10$ and 0.11 at 4.2 K, respectively. With increasing pressure, the ferromagnetic state disappears and the paramagnetic state appears. It is noteworthy that the MT from the paramagnetic state induced by pressure to the ferromagnetic state occurs by applying external magnetic field in the both compounds. The MT observed in the both compounds is first order because a clear hysteresis occurs in the magnetization curves. A first-order MT is also observed for $x=0.12$ at 4.2 K at 1.00 GPa as shown in the inset in Fig. 8. Therefore, it is clear that the coefficient b for $x\leqq 0.12$ is negative, implying a positive curvature of the DOS at E_F due to the sharp peak of the DOS just below E_F.[29] The observed MT is relatively broad. It has been pointed out that the coexistence of the magnetic and nonmagnetic Co atoms around H_C results in a broad MT in $Co(S_{1-x}Se_x)_2$ (Ref. 30) and $Lu(Co_{1-x}Al_x)_2$ (Ref. 31) systems. Therefore the relatively broad transition in the present compounds could be explained by the same reason.

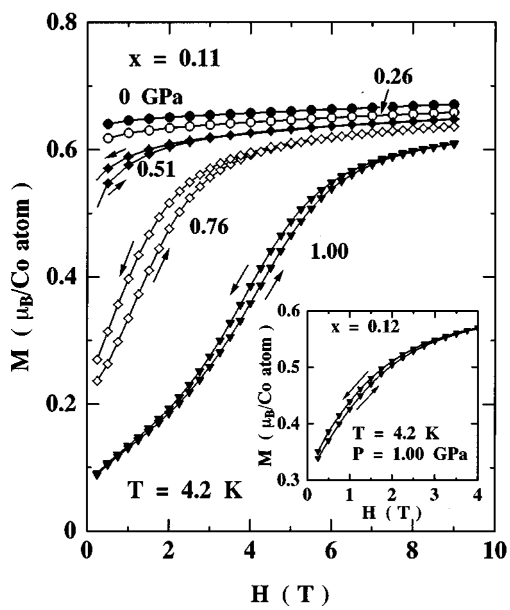

FIG. 8. Pressure and magnetic field dependences of the magnetization for $Lu(Co_{0.89}Ga_{0.11})_2$ at 4.2 K. The inset shows the magnetization curve for $Lu(Co_{0.88}Ga_{0.12})_2$ at 4.2 K at 1.00 GPa.

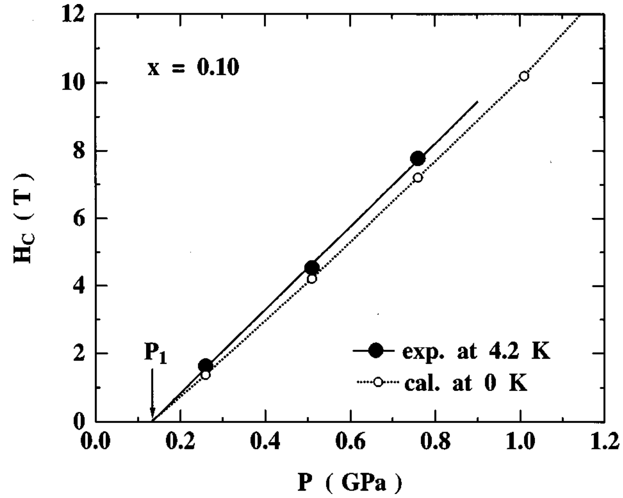

FIG. 9. Pressure dependence of the experimental critical transition field H_C at 4.2 K of $Lu(Co_{0.90}Ga_{0.10})_2$ (●), together with the calculated H_C at 0 K (○).

The pressure dependence of H_C at 4.2 K for $x=0.10$ is given by the solid circle in Fig. 9. The field of H_C was defined as the average of the lower and higher critical fields determined respectively at the peaks of the differential susceptibility in increasing and decreasing fields. The value of H_C increases linearly with the pressure as seen from the figure. The critical pressure P_1 at which the transition field becomes zero is estimated to be 0.13 GPa by a linear extrapolation to $H_C=0$, and the value of $\partial H_C/\partial P$ is obtained as 12 T/GPa for $x=0.10$. The values of P_1 for $x=0.11$ and 0.12 are also obtained as 0.71 and about 1 GPa, respectively. The pressure dependence of the width of hysteresis ΔH_C for $x=0.10$ is shown in Fig. 10. The value of ΔH_C determined as the average of the lower and higher magnetization at H_C decreases linearly with pressure. The critical pressure P_2 at which the first-order MT disappears is estimated to be 1.3 GPa by the linear extrapolation, being smaller by about one order of magnitude that of the estimated value for $LuCo_2$.[22] This difference would come from the difference between the Landau coefficients of both compounds since the value of P_2 strongly depends on these coefficients.[21,22]

Using the present experimental results, the Landau coef-

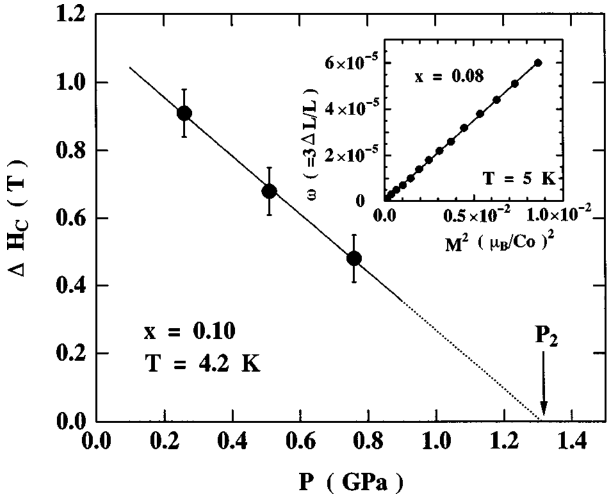

FIG. 10. Pressure dependence of the width of hysteresis of the critical field ΔH_C for $Lu(Co_{0.90}Ga_{0.10})_2$. The forced-volume magnetostriction ω versus M^2 plot for $Lu(Co_{0.92}Ga_{0.08})_2$ in the paramagnetic state is given in the inset.

ficients for $x=0.10$ can be estimated. Effects of spin fluctuations on the free energy are neglected because the present measurement temperature ($T=4.2$ K) of the magnetization curves is low enough to ignore the thermal spin fluctuations. When the values of ac/b^2 are $\frac{3}{16}$ and $\frac{9}{20}$, H_C becomes zero and the first-order MT disappears.[19,26] Because the term of $2\kappa C_{\mathrm{mv}}P$ is added to the coefficient a under pressures,[20] the observed values of P_1 and P_2 are given by the following equations, respectively:

$$\frac{(a+2\kappa C_{\mathrm{mv}}P_1)c}{b^2}=\frac{3}{16} \tag{4a}$$

and

$$\frac{(a+2\kappa C_{\mathrm{mv}}P_2)c}{b^2}=\frac{9}{20}. \tag{4b}$$

From Eq. (1), the pressure effect of M_S^2 at $T=0$ is associated with the following equation:

$$M_S^2=\frac{|b|}{2c}\left(1+\sqrt{1-\frac{4(a+2\kappa C_{\mathrm{mv}}P)c}{b^2}}\right). \tag{5}$$

The measured M_S^2 at $T=4.2$ K and $P=0$ GPa for the compound with $x=0.10$ is $0.36\mu_B^2(=4.67\times10^4\ \mathrm{emu^2/cm^6})$. From Eqs. (4a), (4b), and (5), the values of a, b, and c can be determined if the value of κC_{mv} is available. For $Lu(Co_{1-x}Ga_x)_2$ system, the MT from the paramagnetic to the ferromagnetic state reduces κC_{mv} due to the change of the spin fluctuation spectrum.[32] Furthermore, $P_1 \ll P_2$ as given in Figs. 9 and 10. Therefore the value of κC_{mv} for $x=0.10$ should be obtained in the paramagnetic state. In the present paper, we assume that the value is equal to that of the paramagnetic compound $Lu(Co_{0.92}Ga_{0.08})_2$ which is near the concentration of $x=0.10$. In order to determine κC_{mv}, the magnetization curve and the forced-volume magnetostriction ω for $Lu(Co_{0.92}Ga_{0.08})_2$ with H_C of 12.7 T at $T=5$ K in the paramagnetic state were measured. The results are plotted against M^2 in the inset in Fig. 10. The value of ω, defined as three times of the fractional change in length $\Delta L/L$, was measured at parallel to the applied magnetic field because the volume magnetostriction of the similar Laves phase compounds $Y(Co_{1-x}Al_x)_2$ is almost isotropic.[33,34] A linear relationship between ω and M^2 gives the value of κC_{mv} as $6.9\times10^{-3}\mu_B^{-2}(=5.0\times10^{-8}\ \mathrm{cm^3/erg})$. The estimated values of the Landau coefficients a, b, and c for $x=0.10$ are listed in Table I, together with those of $LuCo_2$,[22] for comparison. The coefficients of $Lu(Co_{0.90}Ga_{0.10})_2$ are much smaller than those of $LuCo_2$, associated with a very large H_C of 74 T.[8] The paramagnetic susceptibility χ at 0 K $(=a^{-1})$ for $Lu(Co_{1-x}Ga_x)_2$ increases with increasing x,[16] accompanied by an increase of the DOS at E_F. The higher the DOS at E_F, the smaller the Landau coefficients,[25,35] consistent qualitatively with the present results. Moreover, ac/b^2 for $x=0.10$ is estimated to be 0.16, very close to $\frac{3}{16}$ under the condition of the onset of ferromagnetism.[19,26] To elucidate the validity of the estimated coefficients, H_C for $x=0.10$ is calculated using these coefficients. The value of H_C is calculated as the magnetic field at which two minima of F, given by the following equation, are equal each other:

TABLE I. Estimated values of the Landau coefficients a, b, and c for $Lu(Co_{0.90}Ga_{0.10})_2$, together with those for $LuCo_2$ (Ref. 22).

	a (10^2 cm^3/emu)	b (10^{-2} cm^3/erg)	c (10^{-6} cm^6/erg^2)
$Lu(Co_{0.90}Ga_{0.10})_2$	7.1	−7.7	1.3
$LuCo_2$ (Ref. 22)	269	−242	65

$$F=\frac{1}{2}(a+2\kappa C_{\mathrm{mv}}P)M^2+\frac{1}{4}bM^4+\frac{1}{6}cM^6-MH. \tag{6}$$

The resultant values are plotted against pressure in Fig. 9 by the open circle. As seen from the figure, the calculated values of H_C versus pressure are in agreement with the experimental values, although the former values are slightly smaller than the latter values. Consequently, the experimental coefficients for $x=0.10$ are reasonable.

It has been reported that an itinerant-electron ferromagnet $Co(S_{0.90}Se_{0.10})_2$ shows the MT under pressure.[23] The value of $\partial H_C/\partial P$ is 4.25 T/GPa,[23] being about one-third of that of $Lu(Co_{0.90}Ga_{0.10})_2$. This difference would mainly come from the different value of κC_{mv} because a large κC_{mv} leads to a very sensitive pressure dependence of H_C. The spontaneous volume magnetostriction for CoS_2 is extremely small,[36] compared with that of ferromagnetic $Lu(Co_{0.90}Al_{0.10})_2$,[37] suggesting that κC_{mv} for $Co(S_{0.90}Se_{0.10})_2$ is much smaller than that of $Lu(Co_{0.90}Ga_{0.10})_2$.

The pressure dependence of M_S obtained from the Arrott plots for $Lu(Co_{1-x}Ga_x)_2$ at $T=4.2$ K is given in Fig. 11. The value of M_S for the compounds with $x\leqq0.12$ begins to decrease drastically at relatively low pressures, which is closely correlated with the negative b. For the compounds with $x\geqq0.15$, M_S shows a linear decrease with pressure up to 1.00 GPa. The data of the compound with $x=0.13$ slightly deviate from the straight line at high pressures, suggesting a marked decrease in M_S under much higher pressures. Figure 12 shows the concentration dependence of the pressure coefficient, $\partial \ln M_S/\partial P$, for $Lu(Co_{1-x}Ga_x)_2$, together with that of

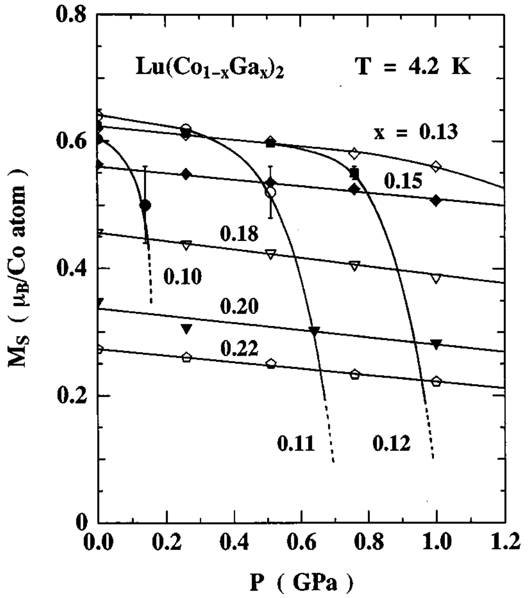

FIG. 11. Pressure dependence of the spontaneous magnetization M_S for $\mathrm{Lu(Co_{1-x}Ga_x)_2}$ compounds.

T_C. The value of $\partial \ln M_S/\partial P$ for $x=0.10$ is not shown because it is difficult to determine $\partial M_S/\partial P$ due to the vicinity of the critical concentration of the onset of ferromagnetism. It should be noted that the magnitude of $\partial \ln T_C/\partial P$ is larger than $\partial \ln M_S/\partial P$ up to around $x=0.15$, showing a rapid decrease in T_C compared with that of M_S and the negative b for the compounds with $x \leqq 0.15$. In other words, the positive curvature of the DOS at E_F remains up to around $x=0.15$. For the compounds with $x \geqq 0.18$, there is no significant difference between both the magnitudes, in contrast to the results for $x \leqq 0.12$. The present results suggest that there is a sharp peak of the DOS just below E_F up to around $x=0.15$, and the peak would become broader with increasing x. The band calculations reveal that the sharp peak of the DOS just below E_F for $\mathrm{YCo_2}$ is broadened by a partial replacement of Co with Al,[12] consistent with the present results.

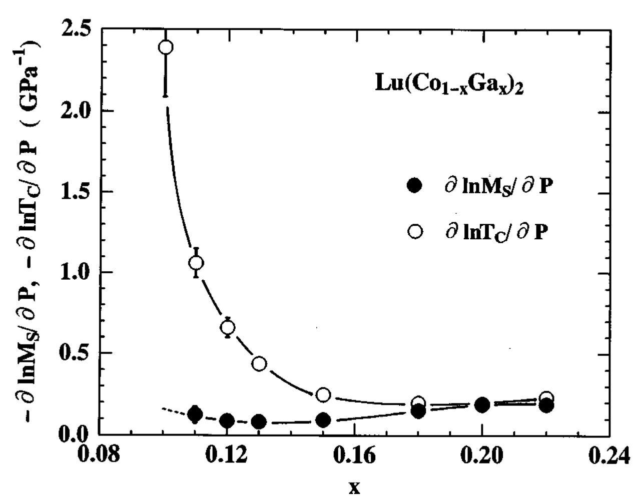

FIG. 12. Concentration dependence of the pressure coefficient of the spontaneous magnetization, $\partial \ln M_S/\partial P$ (●), together with the pressure coefficient of the Curie temperature, $\partial \ln T_C/\partial P$ (○), for $\mathrm{Lu(Co_{1-x}Ga_x)_2}$.

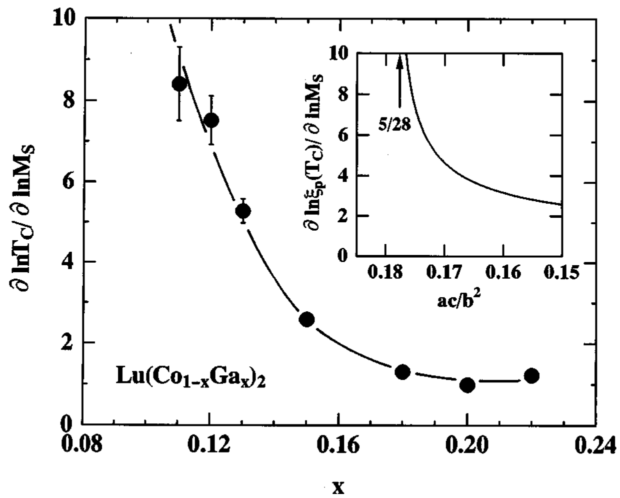

FIG. 13. Concentration dependence of $\partial \ln T_C/\partial \ln M_S$ for $\mathrm{Lu(Co_{1-x}Ga_x)_2}$. The inset shows $\partial \ln \xi_p(T_C)/\partial \ln M_S$ versus ac/b^2.

Concentration dependence of the ratio of $\partial \ln T_C/\partial P$ to $\partial \ln M_S/\partial P$, i.e., $\partial \ln T_C/\partial \ln M_S$ for $\mathrm{Lu(Co_{1-x}Ga_x)_2}$, is plotted in Fig. 13. The value of $\partial \ln T_C/\partial \ln M_S$ is much larger than unity in the vicinity of $x=0.10$, and approaches unity with increasing x. In the case of a weak ferromagnet with $a<0$, $b>0$, and $c=0$, $\partial \ln T_C/\partial \ln M_S$ is unity for the Stoner-Wohlfarth theory[38] and $\frac{3}{2}$ for the spin fluctuation theory.[39] Therefore a significant difference between the both magnitudes around the onset of ferromagnetism would come from the conditions of $a>0$, $b<0$, and $c>0$. Using Eqs. (2) and (5), $\partial \ln \xi_p(T_C)/\partial \ln M_S$, which is equal to $\partial \ln T_C/\partial \ln M_S$, is given by

$$\frac{\partial \ln \xi_p(T_C)}{\partial \ln M_S} = \frac{\sqrt{\frac{7}{5}}\left(1+2\sqrt{\frac{1}{4}-\frac{ac}{b^2}}\right)\sqrt{\frac{1}{4}-\frac{ac}{b^2}}}{\left(1+2\sqrt{\frac{7}{5}}\sqrt{\frac{5}{28}-\frac{ac}{b^2}}\right)\sqrt{\frac{5}{28}-\frac{ac}{b^2}}}. \tag{7}$$

Remarkable is that $\partial \ln \xi_p(T_C)/\partial \ln M_S$ is only determined from the coefficients, i.e., the band structure around E_F. The value of $\partial \ln \xi_p(T_C)/\partial \ln M_S$ versus ac/b^2 is shown in the inset. With increasing ac/b^2, the ferromagnetic state becomes unstable, resulting in a divergent large value near $ac/b^2 = \frac{5}{28}$ which is close to the onset of ferromagnetism. Consequently, the concentration dependence of $\partial \ln T_C/\partial \ln M_S$ for $\mathrm{Lu(Co_{1-x}Ga_x)_2}$ near the critical concentration of the onset of ferromagnetism can be explained by the conditions of $a>0$, $b<0$, and $c>0$.

IV. SUMMARY

The magnetic properties of the Laves phase $\mathrm{Lu(Co_{1-x}Ga_x)_2}$ system have been investigated in high pressures with high magnetic fields. Effect of pressure on the Curie temperature T_C, the spontaneous magnetization M_S, and the critical transition field H_C were measured. These results were analyzed under the conditions of the coefficients $a>0$, $b<0$, and $c>0$ for the Landau coefficients. The effect of replacing Co with Ga on the shape of the density of states around the Fermi energy E_F has been also discussed. The main results are summarized as follows:

(a) For the compounds with $x \leqq 0.12$, the spontaneous magnetization M_S shows a drastic decrease at relatively low pressures. In the paramagnetic state induced by pressure, a first-order metamagnetic transition is caused by applying magnetic fields.

(b) The observed value of H_C versus pressure P for $Lu(Co_{0.90}Ga_{0.10})_2$ is consistent with the results calculated from the Landau coefficients.

(c) The pressure derivative of the Curie temperature, $\partial T_C/\partial P$, is significantly large at around the critical concentration of the onset of ferromagnetism.

(d) 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 $x \leqq 0.15$ due to a negative sign of the coefficient b of the fourth-order term in the Landau expansion, implying that there is the peak of the DOS just below E_F up to around $x = 0.15$.

(e) Concentration dependence of the ratio of the pressure coefficients, $\partial \ln T_C/\partial \ln M_S$, near the onset of ferromagnetism can be explained by taking into account the negative sign of the coefficient b.

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