High Field Magnetization of Itinerant Electron Metamagnetic Laves Phase Lu(Co_<1-x>Ga_x)_2 Compounds(Magnetism)

High Field Magnetization of Itinerant Electron Metamagnetic Laves Phase $Lu(Co_{1-x}Ga_x)_2$ Compounds[*]

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Synopsis

Nearly ferromagnetic pseudobinary $Lu(Co_{1-x}Ga_x)_2$ compounds have been investigated.   The lattice constant increases with increasing Ga concentration, x, especially above x=0.09.   High field magnetization and magnetoresistance of $Lu(Co_{1-x}Ga_x)_2$ compounds have been measured in static and pulsed magnetic fields.   No significant difference is obtained between the static and pulsed fields. A clear itinerant electron metamagnetic transition is observed and its critical field increases with increasing temperature for x=0.09.   It is suggested from the discussion based on the Clausius-Clapeyron equation that the magnetic entropy of these compounds decreases above the critical field.   This is consistent with the recent result of the electronic specific heat coefficient in the magnetic fields. The magnetoresistance of the ferromagnetic state after the transition is reduced about 30% compared with that of the paramagnetic state.   This means that the metamagnetic transition results in a reduction of the spin fluctuation.

I.   Introduction

Ever   since   the   theoretical   prediction   of   the   itinerant   electron metamagnetism by Wohlfarth and Rhodes[1], many experimental and theoretical works have been made for strongly exchange enhanced Pauli paramagnets such as Pd[2,3], $TiBe_2$[4,5], YCo_2[6-10], $LuCo_2$[11]   and $ScCo_2$[11].   These   metal   and compounds show a large electronic specific heat coefficient, γ, and an exchange enhanced   paramagnetic   susceptibility[12].   Several   band   calculations   have   been carried out for the occurrence of the itinerant electron metamagnetic   transition in these materials.   However, the estimated critical field values of the transition, H_c, are too large, being 1000 kOe for YCo_2[10] and 1200 kOe for $LuCo_2$[11].

Recently, we have succeeded in direct observations of the itinerant electron metamagnetic transition in Laves phase YCo_2[12] and $LuCo_2$[13][14] compounds and their H_c values are 690 kOe and 740 kOe, respectively[15]. Since the metamagnetism is the first order transition from strongly enhanced paramagnetism to ferromagnetism, the appearance of unique properties resulting from the large change of the electronic state is expected. However, the direct comparison of these states at the metamagnetic transition has not been reported, because observed critical fields are in the range of 700 KOe and too high for us to study the magnetic properties in detail. By partial substitution of Al for Co, a remarkable reduction of H_c has been confirmed in Laves phase $Y(Co_{1-x}Al_x)_2$[16-18] and $Lu(Co_{1-x}Al_x)_2$[19,20] compounds. However, the magnetization jump due to the metamagnetic transition in $Y(Co_{1-x}Al_x)_2$ compounds smears out in the high Al concentration region[17,20]. Furthermore, the magnetization curves of $Lu(Co_{1-x}Al_x)_2$ compounds are unusually broad owing to a magnetic heterogeneity[22,23]. More recently, Laves phase $Lu(Co_{1-x}Ga_x)_2$ compounds with a C15 type have been found to show a clear metamagnetic transition at low magnetic fields[24]. Therefore, $Lu(Co_{1-x}Ga_x)_2$ compounds would be suited for the studies of the itinerant electron metamagnetic properties. In the present paper, the attention will be paid to the high field properties of $Lu(Co_{1-x}Ga_x)_2$.

II. Experimental

The alloying was made by arc-melting in an argon gas atmosphere, repeated several times. The Lu content was kept slightly higher than the stoichiometry to avoid the precipitation of the ferromagnetic $LuCo_3$ compound. $Lu(Co_{1-x}Ga_x)_2$ compounds were annealed at 1073K for a week in an evacuated quartz tube for homogenization. The room temperature lattice constants were determined by X-ray powder diffraction. Static high magnetic fields up to 170 kOe were generated with a water-cooled magnet of the High Field Laboratory for Superconducting Materials of Tohoku University (HFLSM) and the magnetization was measured using a vibrating sample magnetometer. High field magnetization up to 400 kOe was also measured at the Megagauss Laboratory of Institute for Solid State Physics, The University of Tokyo, using an induction method in the long pulsed fields from 4.2K to 200K. The powdered specimens were used to avoid eddy current effects under a transient pulsed fields. The magnetoresistance was measured by a conventional 4-probe method in the static fields up to 140 kOe using a superconducting magnet at 4.2K.
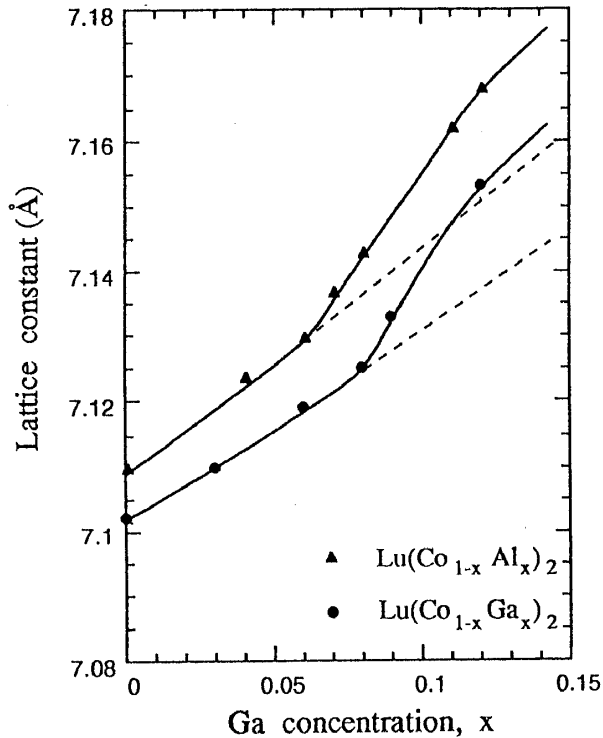
III. Results and Discussion

Fig. 1 Concentration dependence of the lattice constants of $Lu(Co_{1-x}Ga_x)_2$, together with that of $Lu(Co_{1-x}Al_x)_2$[25], for comparison.

In all samples, no other phase was detected by X-ray diffraction analysis. Figure 1 shows the concentration dependence of the lattice constant of $Lu(Co_{1-x}Ga_x)_2$ at room temperature, together with that of $Lu(Co_{1-x}Al_x)_2$[25] for comparison. These $LuCo_2$ and its pseudobinary compounds have a C15 type cubic structure. In this figure, the lattice constant of $Lu(Co_{1-x}Ga_x)_2$ at x=0 differs from that of $Lu(Co_{1-x}Al_x)_2$ at x=0. It should be noted, however, that the lattice constant at x=0 of the present data is in good agreement with the reported value in the ASTM card. The lattice constants of both systems increase with increasing x. This increase corresponds to Vegard's law; the atomic size of Ga and Al is larger than that of Co. In high x regions, the plots deviate upward from the linear dotted lines. A similar deviation has been observed in $Y(Co_{1-x}Al_x)_2$[16] compounds. In general, the lattice constant is expressed by the following expression[26];

$$a(T)=a^0+\Delta a(T)+1/3\kappa m^2_{Co},\qquad(1)$$

where a^0 represents the lattice parameter at T=0K in the hypothetical nonmagnetic state, $\Delta a(T)$ is the thermal expansion due to the lattice vibrations and κ the magnetovolume coupling constant and m^2_{Co} the magnetic moment of the Co atom. It should be noted that the m^2_{Co} is correlated to the amplitude of local spin fluctuations, that is, $(2\mu_B S_L)^2$ [26]. The magnetovolume effect expressed by the third term is the origin of this deviation. In the case of $Lu(Co_{1-x}Al_x)_2$ compounds, the spontaneous magnetization is revealed above x=0.08 at 4.2K[24] and this causes the lattice expansion. Although the present $Lu(Co_{1-x}Ga_x)_2$ compounds are paramagnetic in the whole concentration regions[24], a similar deviation could be observed as shown in Fig. 1. The magnetovolume

Magnetization (emu/g)

25
20
15
10
5
0

$Lu(Co_{0.91} Ga_{0.09})_2$

(a) static fields, 4.2K

0   50   100   150   200   250

Magnetic field (kOe)

Magnetization (emu/g)

25
20
15
10
5
0

$Lu(Co_{0.91} Ga_{0.09})_2$

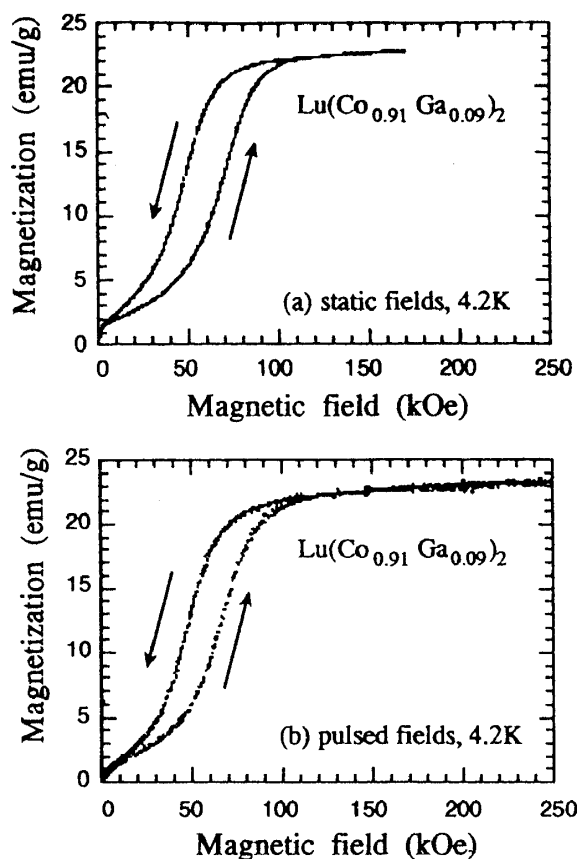(b) pulsed fields, 4.2K

0   50   100   150   200   250

Magnetic field (kOe)

Fig. 2 High field magnetization curves of $Lu(Co_{0.91}Ga_{0.09})_2$ measured at 4.2K: (a) and (b) represent the results obtained in static and pulsed fields, respectively.

change is ascribed not only to the spontaneous magnetization but also to the Co moment in the strongly paramagnetic state due to spin fluctuations. Since the critical fields of the $Lu(Co_{1-x}Ga_x)_2$ compounds with x=0.09 and 0.12 are low, also the paramagnetic susceptibility is strongly enhanced, the spin fluctuations would be remarkable. Such a remarkable spin fluctuation in these compounds would cause the deviation from Vegard's law. Detailed study on this respect is now in progress.

The critical field, H_c, determined by using a one-turn coil system[27] for $LuCo_2$ is 740 kOe at 8K. By partial substitution of Co by Ga, H_c is remarkably reduced and becomes below 100 kOe, being in accessible ranges for water-cooled and superconducting magnets. High field magnetization curves of $Lu(Co_{0.91}Ga_{0.09})_2$ measured at 4.2K by using a water-cooled magnet up to 170 kO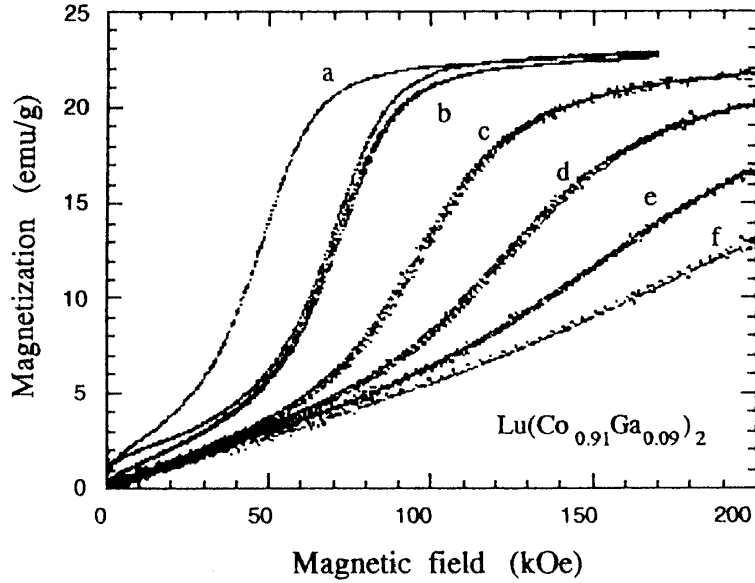e are shown in Fig. 2 (a). A clear magnetization jump around 60 KOe is observed at 4.2K, accompanied by a hysteresis. In order to confirm the scan speed dependence of the magnetization, high field magnetization curves of the same compound in the pulsed fields are shown in Fig. 2 (b). No significant difference is observed between Figs. 2 (a) and (b).

Figure 3 shows high field magnetization curves of $Lu(Co_{0.91}Ga_{0.09})_2$ compounds measured at various temperatures in a water-cooled magnet up to 170 kOe. Since the value of H_c increases with increasing temperature, high field results in the pulsed fields up to 400 kOe were also obtained, and the results up to 200 kOe at high temperatures are also shown in the same figure. The higher the temperature, the broader the transition and the smaller the magnetization jump. In the case of the magnetic transition, Maxwell's thermodynamical relation is represented as follows;

Fig. 3 High field magnetization curves of $Lu(Co_{0.91}Ga_{0.09})_2$ at various temperatures: (a) 4.2k and (b) 40K, measured in static fields, (c) 60K, (d) 80K, (e) 100K and (f) 120K measured in pulsed fields.
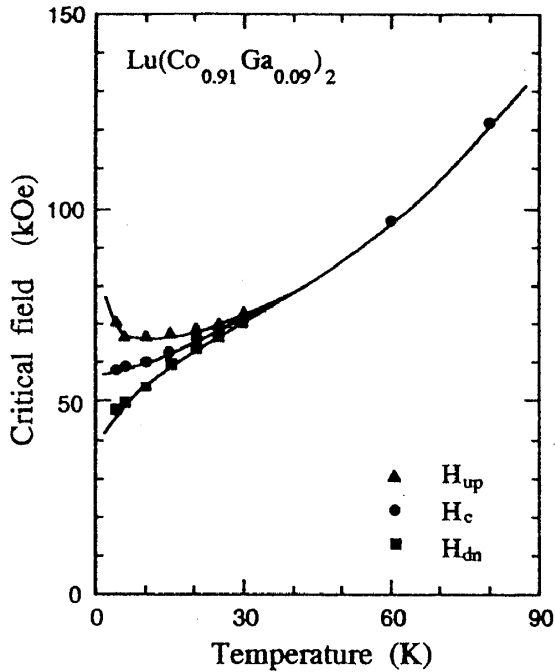

Fig. 4 Temperature dependence of the critical fields of $Lu(Co_{0.91}Ga_{0.09})_2$ in the process of increasing (H_{up}) and decreasing (H_{dn}) field scans and their average field (H_c).

$$\left(\frac{\partial H}{\partial T}\right)_M = -\left(\frac{\partial S}{\partial M}\right)_T \qquad (2),$$

where H, T, S and M are the magnetic field, the temperature, the entropy and the magnetization, respectively. Since ($\partial H/\partial T)>0$ for the present compound, the magnetic entropy is considered to decrease at the transition. This would be correlated to that the magnetic entropy of the nearly ferromagnetic state due to the spin fluctuation is suppressed by a large applied field.

Figure 4 shows temperature dependence of the critical fields of $Lu(Co_{0.91}Ga_{0.09})_2$ compound with increasing field scan ($H_{u\,p}$) and decreasing field scan ($H_{d\,n}$). The H_c represents the average value of $H_{u\,p}$ and $H_{d\,n}$. The critical fields $H_{u\,p}$ and $H_{d\,n}$ of the transition are defined by

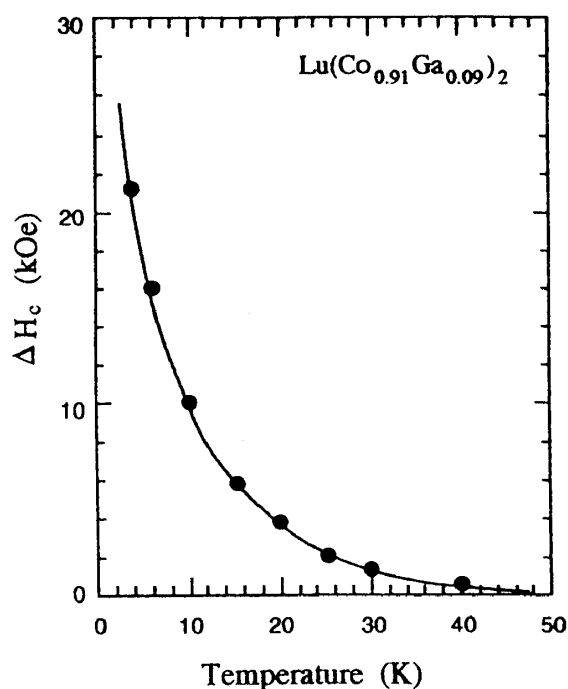the value at the peaks of the differential susceptibility in increasing and decreasing fields, respectively. The temperature dependence of the width of the hysteresis, $\Delta H_c = H_{u\,p} - H_{d\,n}$, is shown in Fig. 5. The value of ΔH_c rapidly decreases with increasing temperature. The logarithmic plot of ΔH_c linearly decreases with increasing temperature for the specimen with $x = 0.09$[28]. The observed temperature dependence of ΔH_c can be fitted by the following expression;

$$\Delta H_c = 31.1 \exp (-0.106T) \quad (3).$$

This suggests that some contribution from the thermal excitation process occurs. On the other hand, the temperature dependence of the critical field, H_c, linearly increases against T^2 and could be expressed by the following expression;

$$H_c = 56.9 + 0.0135 T^2 \quad (4).$$

Fig. 5 Temperature dependence of the width of the hysteresis, ΔH_c.

The metamagnetic transition in the present compounds is considered to be the first order and the magnetic transition behavior is considered to obey the following Clausius-Clapeyron equation[28,29];

$$\Delta S = -\Delta M \left(\frac{\partial H_c}{\partial T} \right)_M \quad (5),$$

where ΔS and ΔM are the entropy change and the magnetization jump due to the transition, respectively. In the low temperature limit, a leading term of the entropy of many metals is related to the electronic specific heat. On the other hand, the change in critical field, H_c, is proportional to T^2 as given by eq. (4). Then, Eq. (5) implies that ΔS is proportional to T at low temperatures, and results in a discontinuous change of electronic specific heat coefficient, $\Delta \gamma$;

$$\Delta \gamma = -2 \alpha \Delta M \quad (6),$$

where α is the coefficient of T^2 in Eq. (4), being 0.0135 (kOe/K^2) for $Lu(Co_{0.91}Ga_{0.09})_2$ compound. The magnetizations at the critical field are estimated to be 5.0 and 21.5 emu/g in the paramagnetic and ferromagnetic

phases, respectively, resulting in $\Delta M=16.5$emu/g at 4.2K. Therefore, the value of $\Delta \gamma$ is estimated to be -13.1 mJ/K^{2}mol from the observed magnetization data. Recently, low temperature specific heat of $Lu(Co_{1-x}Ga_x)_2$ compounds has been studied in the magnetic fields up to 146 kOe[24]. The γ value of the specimen with x=0.09 is about 35.5mJ/K^{2}mol in the paramagnetic state and a remarkable reduction is caused in the magnetic fields and results in 25mJ/K^{2}mol at 14.6 kOe, giving the $\Delta \gamma$ value of -10.5 mJ/K^{2}mol. Considering the fact that the transition of the bulk samples is broader than that of the powdered one, the $\Delta \gamma$ value is estimated from the magnetization in good agreement with that of the specific heat experiment.
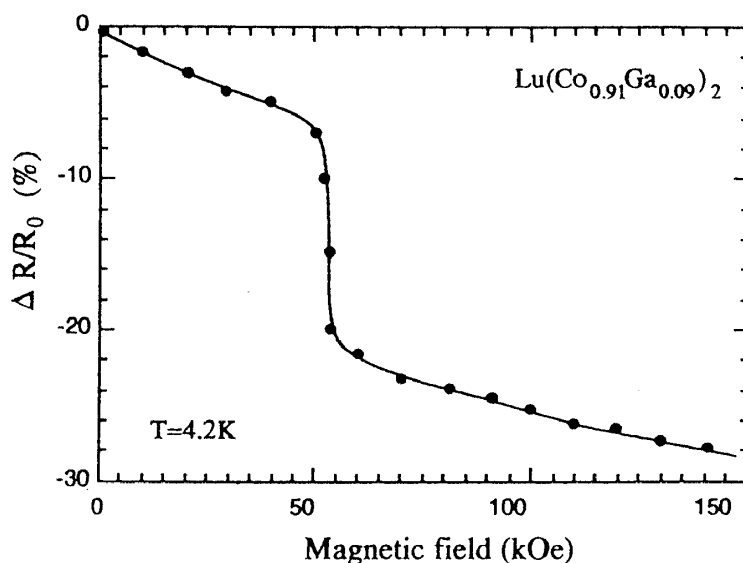

Fig. 6 Relative change in the magnetoresistance of $Lu(Co_{0.91}Ga_{0.09})_2$ at 4.2K as a function of external field.

Figure 6 shows the relative change in the magnetoresistance, $\Delta R/R_0$, of $Lu(Co_{0.91}Ga_{0.09})_2$ at 4.2K as a function of external field. The value of $\Delta R/R_0$ is remarkably reduced at about 50 kOe corresponding to H_c of the metamagnetic transition. This discontinuity of $\Delta R/R_0$ at H_c is due to the appearance of the magnetic ordering of the d-electrons, resulting in the significant change in s-d scattering, that is, the large decrease of $\Delta R/R_0$ would be attributed to the suppression of the spin fluctuations due to the metamagnetic transition. This effect also reflects the suppression of the electronic specific heat coefficient, as mentioned before[24]. From these results, it is concluded that the large molecular field due to the itinerant electron metamagnetic transition sufficiently suppresses the spin fluctuation. Through the studies of the itinerant metamagnetic $Lu(Co_{1-x}Ga_x)_2$ compounds, the direct comparison of the physical properties between the paramagnetic and ferromagnetic states have been performed in the present study and further studies are now in progress.

IV. Conclusions

High field magnetization, magnetoresistance and the lattice constant of metamagnetic Laves phase $Lu(Co_{1-x}Ga_x)_2$ compounds have been investigated and the main results are summarized as follows:

(1) The lattice constant increases with increasing x, particularly above x=0.09.

(2) No significant difference in magnetization curves of $Lu(Co_{0.91}Ga_{0.09})_2$ between the static and pulsed fields is observed. This means that the magnetization does not depend on the magnetic field scan rate.

(3) The critical field of the metamagnetic transition, H_c, increases with increasing temperature, T, in proportion to T^2.

(4) The width of hysteresis of the metamagnetic transition, ΔH_c, exponentially decreases with increasing T. This suggests some contribution from the thermal excitation process.

(5) The magnetoresistance, $\Delta R/R_0$, of $Lu(Co_{0.91}Ga_{0.09})_2$ drastically decreases above H_c. This behavior would come mainly from a significant reduction of the spin fluctuation.

Acknowledgments

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