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1998 J. Phys.: Condens. Matter 10 3625

(http://iopscience.iop.org/0953-8984/10/16/014)

J. Phys.: Condens. Matter **10** (1998) 3625–3634. Printed in the UK PII: S0953-8984(98)90655-5

Magnetovolume effects in metamagnetic itinerant-electron systems $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$

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Received 13 January 1998, in final form 13 February 1998

Abstract. High-field magnetization and magnetostriction measurements have been performed for typical metamagnetic itinerant-electron systems $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ with $0 \leqslant x \leqslant 0.12$ at 4.2 K in pulsed magnetic fields up to 40 T. The substitution of Al or Ga for Co significantly decreases the critical field of the metamagnetic transition. The magnetovolume coupling constant n_{dd} in the paramagnetic state is nearly constant up to about $x = 0.05$ and then decreases with increasing x. Moreover, the value of n_{dd} is considerably reduced by the metamagnetic transition. The variation of n_{dd} produced by the change of x or the metamagnetic transition is considered to originate from the change of the spin fluctuation spectrum.

1. Introduction

The magnetic properties of the cubic Laves phase compounds RCo_2 (R:Y or rare-earth element) have intensively been studied in recent years with interest in the instability of the d-electron magnetism and in the giant magnetostriction [1, 2]. The RCo_2 compounds with nonmagnetic R = Y and Lu are typical of exchange-enhanced Pauli paramagnets and exhibit itinerant electron metamagnetism, that is a first-order field-induced transition from the Pauli paramagnetic to the ferromagnetic state in the itinerant electron system. The critical field of the transition has been determined to be 69 T for YCo_2 and 74 T for $LuCo_2$ by means of direct magnetization measurements in ultrahigh magnetic fields up to 100 T [3, 4].

In the substituted systems $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$, the susceptibility is enhanced and the critical field decreases with increasing x [5, 6]. Above a critical concentration $x_c \cong 0.12$, both systems become ferromagnetic. Note that the spontaneous magnetization of $Y(Co_{1-x}Al_x)_2$ in the ferromagnetic concentration region $0.12 < x \leqslant 0.19$ is very small compared with the magnetization in the ferromagnetic state produced by the metamagnetic transition in the paramagnetic concentration region $x \leqslant 0.12$ [5]. In the concentration region $0.12<x<0.15$, a metamagnetic transition from the weakly ferromagnetic to a stronger ferromagnetic state is observed. The origin of the weakly ferromagnetic state was discussed by Dubenko *et al* [7]. In the case of the $Lu(Co_{1-x}Ga_x)_2$ system, however, the spontaneous magnetization in the ferromagnetic concentration region is consistent with the magnetization in the ferromagnetic state produced by the metamagnetic transition in the paramagnetic concentration region [6]. No metamagnetic transition occurs in the ferromagnetic region.

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0953-8984/98/163625+10$19.50

In the itinerant d-electron system, the appearance of magnetic moment is accompanied by a large volume magnetostriction. The volume magnetostriction $\omega = \delta V/V$ is usually related to the d-electron magnetic moment M_d:

$$\omega = n_{dd} M_d^2 \tag{1}$$

where n_{dd} is the magnetovolume coupling constant. The value of n_{dd} can be determined directly by measuring both the magnetization and the magnetostriction of the system. An alternative indirect method for determination of n_{dd} is the measurement of thermal expansion [8]. Thermal expansion measurements on RCo_2 with magnetic R, in which the d-electron moment (Co moment) is induced by an exchange field from the R sublattice, give the information about n_{dd}.

According to magnetostriction measurements of paramagnetic YCo_2 and $LuCo_2$ in high magnetic fields up to 32 T, the estimated values of n_{dd} are $n_{dd} = 1.2 \times 10^{-2}$ and 1.6×10^{-2} $(\mu_B/\mathrm{Co})^{-2}$, respectively [9]. On the other hand, magnetostriction measurements of ferromagnetic $Y(Co_{1-x}Al_x)_2$ in high magnetic fields up to 12 T give smaller values of n_{dd} ranging from 6×10^{-3} to 9×10^{-3} $(\mu_B/\mathrm{Co})^{-2}$ [10, 11]. The values of n_{dd} deduced from thermal expansion measurements on RCo_2 compounds with magnetic R are scattered in a wide range [8]. This scattering comes mostly from uncertainty in the estimation of the Co moment in the compounds. Recently, a reliable value of $n_{dd} = 8.1 \times 10^{-3}$ $(\mu_B/\mathrm{Co})^{-2}$ has been given by thermal expansion measurements of $GdCo_2$ by means of x-ray diffraction [2]. The Co moment in $GdCo_2$ can be uniquely determined by measuring the magnetization since the Gd^{3+} ion has no orbital moment. It should be noted that the value of n_{dd} in ferrimagnetic RCo_2 with magnetic R is remarkably smaller than that in paramagnetic YCo_2 and $LuCo_2$. This suggests that n_{dd} may depend on the magnetic state of the d-electron system because the system of ferrimagnetic RCo_2 feels the exchange field from the R sublattice which is higher than the critical fields of the metamagnetic transition in YCo_2 and $LuCo_2$.

In the present study, we have performed magnetization and forced magnetostriction measurements on the $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ systems with $0 \leqslant x \leqslant 0.12$ in pulsed high magnetic fields up to 40 T to reveal the change of the magnetovolume coupling constant produced by the substitution of Al or Ga and by the metamagnetic transition.

2. Experiment

Polycrystalline bulk samples of $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ with $0 \leqslant x \leqslant 0.12$ were prepared by arc melting of the constituent elements of 99.9% purity in an Ar atmosphere, followed by annealing at 850 °C for a week. No extraneous phases except for the C15 Laves phase were detected by x-ray diffraction. For this study, we selected high-quality samples which exhibit sharp metamagnetic transitions and shaped them into cubes of about 2.5 mm. The shaped samples were used for both magnetization and magnetostriction measurements.

The high-field magnetization was measured at 4.2 K using an induction method in pulsed magnetic fields up to 40 T with a rise time of about 5 ms. The experimental error of the magnetization is estimated to be within ±2%. The longitudinal and transverse magnetostrictions, $\lambda_\parallel$ and $\lambda_\perp$, were also measured at 4.2 K in pulsed magnetic fields up to 40 T with a capacitance method [12]. The experimental error of $\lambda_\parallel$ and $\lambda_\perp$ is within ±5%. The volume magnetostriction ω was calculated using the relation

$$\omega = \lambda_\parallel + 2\lambda_\perp. \tag{2}$$

3. Experimental results

As a typical example, we show in figure 1 the magnetostriction curves of paramagnetic $Y(Co_{0.89}Al_{0.11})_2$ and ferromagnetic $Lu(Co_{0.88}Ga_{0.12})_2$ measured at 4.2 K in pulsed high magnetic fields up to 40 T. $Y(Co_{0.89}Al_{0.11})_2$ has a sharp metamagnetic transition to the ferromagnetic state. The linear magnetostriction depends on the direction of the applied field, that is $\lambda_\parallel \neq \lambda_\perp$, indicating anisotropic expansion. In the case of $Lu(Co_{0.88}Ga_{0.12})_2$, the linear magnetostrictions $\lambda_\parallel$ and $\lambda_\perp$ exhibit a jump from zero value in low field and have negative and positive values, respectively. The observed anisotropic magnetostriction is associated with the large negative magnetostriction constant λ_{100} of the Co subsystem [13]. However, the volume magnetostriction calculated from (2) is nearly zero around zero field, indicating the cancellation of the anisotropic linear magnetostrictions.

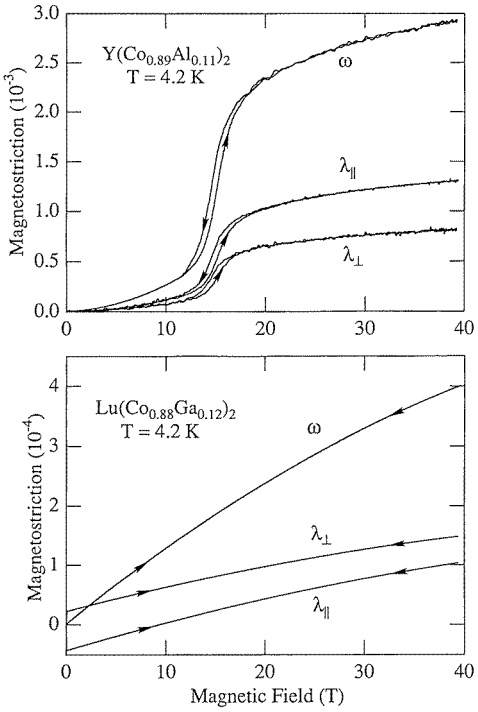

Figure 1. Longitudinal, transverse and volume magnetostriction curves of paramagnetic $Y(Co_{0.89}Al_{0.11})_2$ and ferromagnetic $Lu(Co_{0.88}Ga_{0.12})_2$ for $T = 4.2$ K in pulsed high magnetic fields up to 40 T. $Y(Co_{0.89}Al_{0.11})_2$ exhibits a metamagnetic transition at about 15 T.

The magnetization and volume magnetostriction curves of the $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ compounds are shown in figures 2 and 3. All the compounds except for $Lu(Co_{0.88}Ga_{0.12})_2$ are paramagnetic. The results of the magnetization measurements are in good agreement with those of powdered samples [5, 6]. The hysteresis of the magnetization curve associated with the metamagnetic transition is slightly different from that of the volume magnetostriction curve. This comes probably from different conditions in the magnetization and magnetostriction measurements: the sample was set in a Teflon tube in the former

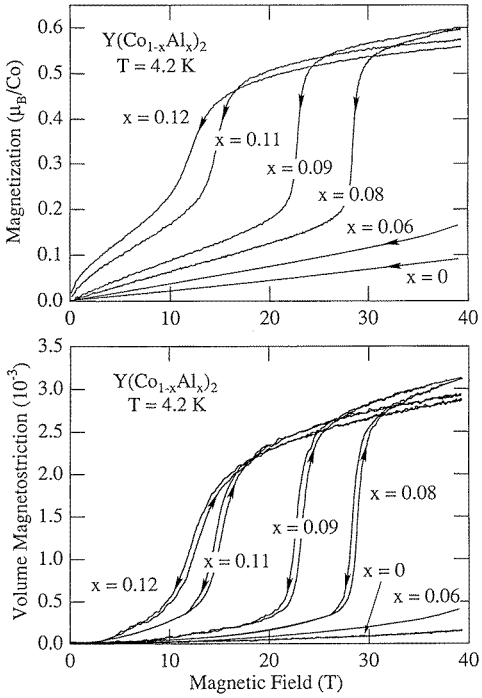

Figure 2. High-field magnetization and volume magnetostriction curves of $Y(Co_{1-x}Al_x)_2$ for $T = 4.2$ K in pulsed high magnetic fields up to 40 T. For simplicity, the magnetization curves measured in increasing fields are eliminated.

and was fixed with epoxy resin in a capacitance cell in the latter. Since the hysteresis appears only in the transition region from the paramagnetic to the ferromagnetic state, the magnetization and magnetostriction data in both states are not affected by the measurement conditions.

In order to estimate the magnetovolume coupling constant n_{dd} of all the samples, we plotted the volume magnetostriction as a function of the square of magnetization $M^2(B)$ in the paramagnetic state and as a function of $[M^2(B) - M^2(B_c^+)]$ in the ferromagnetic state, as shown in figures 4 and 5, where B_c^+ is the field value at which the metamagnetic transition terminates. (In the case of ferromagnetic $Lu(Co_{0.88}Ga_{0.12})_2$, B_c^+ means the saturation field.) The volume magnetostriction in both states increases linearly with $M^2(B)$, which is consistent with (1).

The estimated values of the magnetovolume coupling constant n_{dd} in the $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ systems are plotted as a function of x in figure 6. In the paramagnetic state of the $Y(Co_{1-x}Al_x)_2$ system, the value of n_{dd} is nearly constant up to $x = 0.06$ and then decreases abruptly with increasing x. In the concentration region $0.08 \leqslant x \leqslant 0.12$, the decrease of n_{dd} becomes small. It should be noted that the values of n_{dd} in these systems are remarkably reduced by the metamagnetic transition from the paramagnetic to the ferromagnetic state. This clearly indicates that the magnetovolume coupling constant depends on the magnetic state. In the ferromagnetic state, the coupling constant is nearly independent of the concentration x.

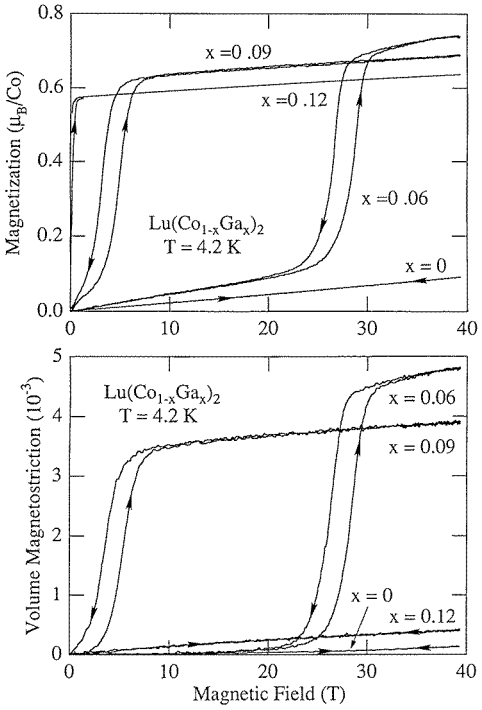

Figure 3. High-field magnetization and volume magnetostriction curves of $Lu(Co_{1-x}Ga_x)_2$ for $T = 4.2$ K in pulsed high magnetic fields up to 40 T.

4. Discussion

The metamagnetic compounds YCo_2 and $LuCo_2$ have the characteristics of exchange-enhanced Pauli paramagnets and their resistivity shows a temperature dependence $\rho(T) = \rho(0) + AT^2$ at low temperatures. The coefficient A is very large compared with isostructural compounds with small susceptibility, such as $ZrCo^2$ and $HfCo_2$. Baranov *et al* [14] found that YCo_2 and $LuCo_2$ satisfy the Kadowaki–Woods relation [15], $A/\gamma = 1.0 \times 10^{-5}$ $\mu\Omega$ cm (K mol $mJ^{-1})^2$, as well as heavy-fermion compounds, where γ is the electronic specific-heat coefficient. This relation for the d electron systems can be explained in terms of thermal spin fluctuation theory [16]. These results indicate that YCo_2 and $LuCo_2$ are typical of spin fluctuation systems. In these compounds, the d band is formed by the hybridization between the d-electron states of Co and the partner element Y or Lu. The Fermi level exists in a local valley between two peaks in the density of states curve of the d band. The spin fluctuations in the d band are considered to develop due to the high value and positive curvature of the density of states curve near the Fermi level [17]. The occurrence of the metamagnetic transition is also considered to originate from such a shape of the density of states curve near the Fermi level [18]. When Al (Ga) atoms are partially substituted for the Co atoms in YCo_2 ($LuCo_2$), the local peak being just below the Fermi level becomes broad and the peak position approaches to the Fermi level [19]. In the $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ systems, therefore, the susceptibilities and spin fluctuations are more enhanced and the critical field of the metamagnetic transition decreases with increasing x. Above the critical concentration $x_c \cong 0.12$, ferromagnetism appears [20].

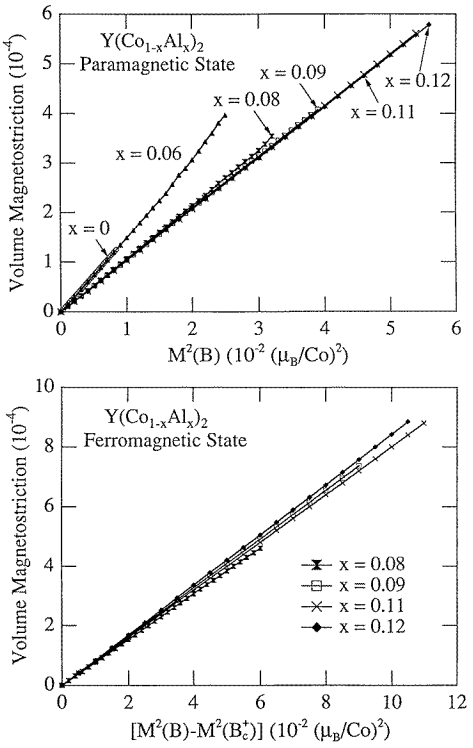

Figure 4. Volume magnetostriction of $Y(Co_{1-x}Al_x)_2$ as a function of $M^2(B)$ in the paramagnetic state and as a function of $[M^2(B) - M^2(B_c^+)]$ in the ferromagnetic state.

In the paramagnetic region $0 \leqslant x < x_c$, the susceptibility shows an anomalous temperature dependence: with increasing temperature, the susceptibility increases and then decreases through a maximum value. The susceptibility anomaly can be elucidated well with a theory of itinerant electron metamagnetism at finite temperatures based on the spin fluctuation model, as well as the characteristics of the metamagnetic transition [21]. In this theory, however, only the effects of thermal spin fluctuations are included and those of quantum spin fluctuations are ignored.

In the presence of uniform magnetization, the local spin fluctuation amplitude $\langle S^2 \rangle$ can be expressed in the following sum of contributions [22]:

$$\langle S^2 \rangle = \langle \delta S^2 \rangle_{zp} + \langle \delta S^2 \rangle_{th} + \sigma^2/4 \tag{3}$$

where $\langle \delta S^2 \rangle_{zp}$ and $\langle \delta S \rangle_{th}$ are respectively the quantum and thermal spin fluctuation amplitudes, $\sigma = (M_d/N_0\mu_B)$ is the magnetization per magnetic atom in units of μ_B and N_0 the number of magnetic atoms in the sample. $\langle \ldots \rangle$ represents the thermal average. In the ground state ($T = 0$ K), the thermal term vanishes and the quantum term reduces to the zero-point spin fluctuation amplitude. Since we measured the magnetostriction of $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ at 4.2 K, the thermal term is negligibly small. Takahashi and Sakai pointed out that even in the ground state the zero-point spin fluctuations play important roles in the magnetovolume effects in weakly ferromagnetic systems [22] and in the magnetization processes of itinerant metamagnetic systems [23]. They proposed theories of the magneto-volume effects and the metamagnetic transition based on the assumption that the local spin

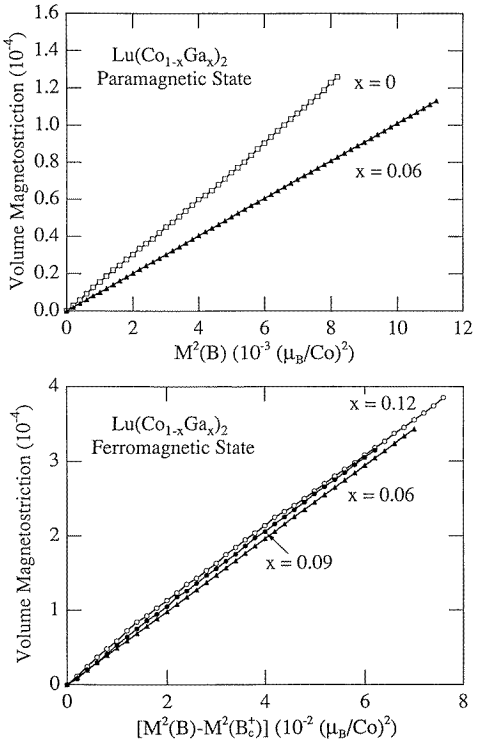

Figure 5. Volume magnetostriction of $Lu(Co_{1-x}Ga_x)_2$ as a function of $M^2(B)$ in the paramagnetic state and as a function of $[M^2(B) - M^2(B_c^+)]$ in the ferromagnetic state.

fluctuation amplitude is nearly constant, $\langle S^2 \rangle \cong$ constant. Experimental investigations seem to support the validity of this assumption [24, 25].

According to the theory of the magneto-volume effects [20], the free energy is expanded in terms of the wave vector and frequency-dependent spin fluctuation amplitude by

$$F = \frac{V}{2\kappa}\omega^2 + \frac{1}{2}\sum_{q,\nu}\chi^{-1}(q,\nu)S_{q,\nu}S_{-q,-\nu} \tag{4}$$

where the first term represents the elastic energy of the crystal with the volume V and the compressibility κ and the volumestriction $\omega = \delta V/V$. $\chi(q,\nu)$ is the dynamical spin susceptibility. Therefore, the generalized magnetovolume coupling constant is given by

$$\lambda(q,\nu) = -\frac{1}{2N_0}\frac{\partial}{\partial\omega}\chi^{-1}(q,\nu). \tag{5}$$

In the ground state, the volume magnetostriction can be described by

$$\omega = \omega_{zp} + \omega_{mag} = \frac{\kappa\bar{\lambda}N_0}{V}\langle\delta S^2\rangle_{zp} + \frac{\kappa\lambda_0 N_0}{V}\frac{\sigma^2}{4} = \frac{\kappa\lambda_0 N_0}{4V}(1-d)\sigma^2 + \text{constant} \tag{6}$$

where λ_0 is defined by $\lambda_0 \equiv \lambda(0,0)$. $\bar{\lambda}$ is the effective coupling constant for the zero-point spin fluctuation amplitude given by

$$\bar{\lambda} = \int_0^1 \mathrm{d}x^2\lambda(q_B x, 0) \tag{7}$$

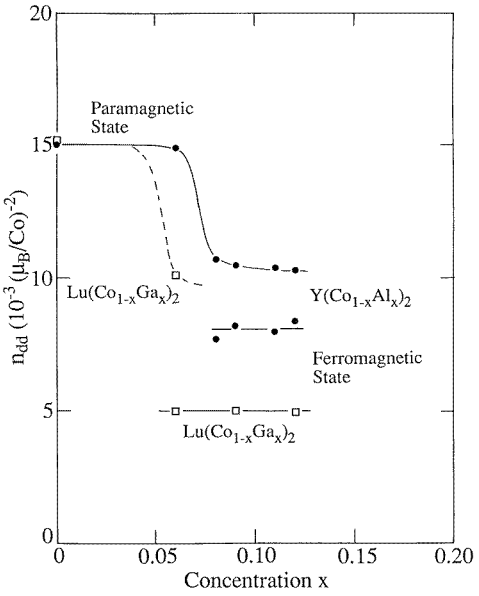

Figure 6. Estimated values of magnetovolume coupling constant n_{dd} of $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ as functions of x.

where q_B is the effective zone-boundary wave number, equal to $(6\pi N_0/V)^{1/3}$, and d is the ratio between $\bar{\lambda}$ and λ_0, $d = \lambda/\bar{\lambda}_0$. We obtain the last line of (6) using the relation $\langle \delta S^2 \rangle_{zp} + \sigma^2/4 =$ constant. The correction term d originates from the zero-point spin fluctuations. Note that the magnetoelastic coupling constants λ_0 and $\bar{\lambda}$ are usually different from each other. If there is no q-dependence of $\lambda(q, 0)$, both constants have the same value and the volume magnetostriction becomes zero.

As shown in figure 6, the magnetovolume coupling constant n_{dd} of $Y(Co_{1-x}Al_x)_2$ for 4.2 K in the paramagnetic state is nearly constant up to $x \cong 0.06$ and then decreases abruptly with increasing x in the concentration region $0.06 < x < 0.08$. In the region $0.08 \leqslant x \leqslant 0.12$, the decrease of n_{dd} becomes small. It should be noted that the metamagnetic transition from the paramagnetic to the ferromagnetic state remarkably reduces n_{dd}. In the ferromagnetic state, n_{dd} is nearly independent of x. These characteristics are observed also in the $Lu(Co_{1-x}Ga_x)_2$ system. According to (6), the variation of the coupling constant n_{dd} in $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ due to the change of x or the metamagnetic transition mainly comes from the term $\lambda_0(1-d)(= \lambda_0 - \bar{\lambda})$. This means that the dynamical spin fluctuation spectrum in the ground state is greatly modified by changing x or by the metamagnetic transition, which results in the change of n_{dd}.

In the itinerant electron system, spin fluctuations enhance the electronic specific heat coefficient γ. Therefore, the variation of n_{dd} will correlate with that of γ. The γ-values of $Lu(Co_{1-x}Ga_x)_2$ and $Y(Co_{1-x}Al_x)_2$ are plotted as functions of x in figure 7, together with those of $Lu(Co_{1-x}Al_x)_2$ for comparison [6, 26, 27]. With increasing x, the γ-value of $Y(Co_{1-x}Al_x)_2$ is nearly constant up to $x \cong 0.05$ and then increases abruptly in the region $0.06 < x < 0.08$. The γ-value continues to increase in the region $0.09 \leqslant x \leqslant 0.12$, but the increment becomes smaller. Goto *et al* suggested from the temperature dependence of the metamagnetic transition field in YCo_2 that the γ-value is reduced by the transition [1]. They estimated the reduction of γ to be $\Delta\gamma = 11$ mJ K^{-2} mol^{-1}. Using the γ-value in

the paramagnetic state (34 mJ K^{-2} mol^{-1}), we can evaluate the one in the ferromagnetic state to be 23 mJ K^{-2} mol^{-1} for YCo_2. This value is also plotted in figure 7. In the case of $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Al_x)_2$, the γ-value is also nearly constant up to $x = 0.04$ and then increases with x. The appearance of ferromagnetism largely suppresses it. In ferromagnetic $Lu(Co_{1-x}Al_x)_2$ with $0.1 \leqslant x \leqslant 0.15$, the γ-value is nearly independent of x. It should be noted that the $Lu(Co_{0.88}Ga_{0.12})_2$ sample used for specific heat measurement is paramagnetic and exhibits the metamagnetic transition to the ferromagnetic state in a magnetic field lower than 1 T. The γ-value is 36 mJ K^{-2} mol^{-1} in the paramagnetic state and is reduced to 21 mJ K^{-2} mol^{-1} in the ferromagnetic state. The latter value is consistent with the γ-value of ferromagnetic $Lu(Co_{1-x}Al_x)_2$ in zero field.

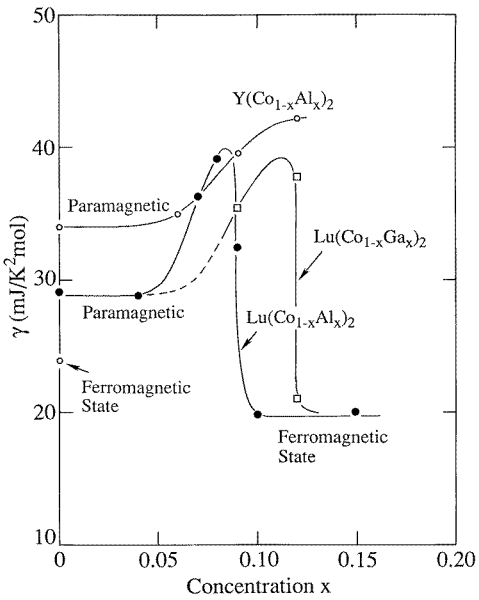

Figure 7. γ-values of $Y(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Al_x)_2$ as functions of x. These data are given in [6, 26, 27], respectively.

The experimental results on the magnetovolume coupling constant n_{dd} and the γ-value indicate that the large variation of n_{dd}, which is caused by the change of x or the metamagnetic transition, closely correlates with that of the γ-value in the $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ systems: with increasing x, the n_{dd}- and γ-values are nearly constant up to $x \cong 0.05$ and then rapidly change. The metamagnetic transition to the ferromagnetic state reduces both values. These facts suggest that the observed variation of n_{dd} originates mainly from the change of the spin fluctuation spectrum. Yoshimura *et al* [28] found that the spin fluctuation spectrum in the $Y(Co_{1-x}Al_x)_2$ system suddenly changes when going from the paramagnetic to the weakly ferromagnetic phase.

5. Conclusion

In this study, we have measured the high-field magnetization and magnetostriction of the itinerant systems $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ with $0 \leqslant x \leqslant 0.12$ at $T = 4.2$ K in pulsed magnetic fields up to 40 T. Both systems exhibit the metamagnetic transition

to the ferromagnetic state except for ferromagnetic $Lu(Co_{0.88}Ga_{0.12})_2$. With increasing concentration x, the critical field of the transition rapidly decreases. The observed volume magnetostriction is proportional to the square of magnetization in both the paramagnetic and ferromagnetic states. The magnetovolume coupling constant n_{dd} in the paramagnetic state is nearly constant up to about $x = 0.05$ and then decreases with increasing x. The coupling constant is considerably reduced by the metamagnetic transition. We have discussed the variation of n_{dd} due to the change of x or the metamagnetic transition on the basis of the Takahashi theory [22]. The variation is considered to be attributed to the change of the spin fluctuation spectrum.

Acknowledgments

The authors thank Y Takahashi and H Yamada for useful discussions, and H Mitamura and K Koui for supplying the samples of $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ used in this study. The stay of MIB at ISSP for one year was financially supported by the Ministry of Education, Science and Culture of Japan.

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