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Itinerant-electron metamagnetism of $(Y, Lu)(Co, Al)_2$ at finite temperature

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Abstract. The metamagnetic transition and paramagnetic susceptibility for the pseudo-binary compound $(Y, Lu)(Co, Al)_2$ with the cubic Laves-phase structure are measured. The observed results are analysed on the basis of phenomenological spin-fluctuation theory for itinerant-electron metamagnetism. A satisfactory agreement between experiment and theory is obtained qualitatively.

Two characteristic phenomena in itinerant-electron metamagnetism, i.e., a field-induced first-order phase transition from the paramagnetic to the ferromagnetic state at lower temperature and a maximum in the temperature dependence of susceptibility at higher temperature, are the consequence of the special feature of the density of states near the Fermi level. Recently, these phenomena have been studied intensively from both experimental and theoretical points of view [1]. It has been found that the paramagnetic compounds YCo_2 and $LuCo_2$ with the cubic Laves-phase structure show a clear metamagnetic transition (MT) at an extremely high magnetic field of about 700 kOe at low temperature [2]. These compounds show also a maximum in the temperature dependence of the susceptibility around room temperature [1].

On the other hand, it has been observed for the pseudo-binary compounds $Y(Co, Al)_2$ and $Lu(Co, Al)_2$ that the critical field of the MT decreases with increasing concentration of Al and a ferromagnetic state is stabilized at a certain concentration of Al [3–6]. As the lattice constant is increased by the substitution of Al, then it can be said that the ferromagnetic state is stabilized by lattice expansion in these systems. However, Gabelko *et al* [7] have explicitly shown that the lattice expansion induced by the substitution of Al does not play an important role in the onset of the ferromagnetic moment or in the decrease of the critical field of the MT. For the pseudo-binary $(Y, Lu)(Co, Al)_2$ compounds in which an appropriate amount of Y atoms is replaced by Lu atoms to keep the volume constant when the Co atoms are substituted by Al atoms, they have observed similar results to those for $Y(Co, Al)_2$ and $Lu(Co, Al)_2$ without the lattice expansion. As pointed out by Aoki and Yamada [8], the hybridization between p states of Al and d states of Co in $Y(Co, Al)_2$ is so strong that the characteristic peak of the density-of-states curve, which is responsible for the MT, becomes broad and small. The onset of the ferromagnetic moment and the decrease of the critical field of the MT may be attributed to this broad peak together with the shift of the Fermi level.

While many experimental and theoretical works have been carried out to study the MT for YCo_2, $LuCo_2$ and related pseudo-binary systems at low temperature, there are only

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a few concerned with the MT at finite temperature. It has been observed for Y(Co, Al)$_2$ [5], Lu(Co, Al)$_2$ [6] and (Y, Lu)(Co, Al)$_2$ [7] that the critical field of the MT increases with increasing temperature as T^2 at low T and the MT disappears at a certain temperature. Recently, Yamada [9] has proposed a model for the itinerant-electron metamagnetism at finite temperature. By using the phenomenological spin-fluctuation theory, he has obtained a relation between three characteristic temperatures T_0, T_1 and $T_{\max}$, at which the field-induced MT disappears, the temperature-induced first-order transition of the spontaneous moment occurs and the susceptibility reaches a maximum in its temperature dependence, respectively.

In this work, our observed data of T_0 and $T_{\max}$ for $(Y_{1-t}Lu_t)(Co_{1-x}Al_x)_2$ are analysed by the spin-fluctuation model for the MT [9]. Two polycrystalline compounds with the paramagnetic ground state and the same lattice parameter 7.219 ± 0.003 Å, $(Y_{0.59}Lu_{0.41})(Co_{0.915}Al_{0.085})_2$ (referred to as sample A) and $(Y_{0.636}Lu_{0.364})(Co_{0.925}Al_{0.075})_2$ (referred to as sample B), have been chosen for measurements. The magnetization curves observed for these samples have already been reported in [7]. The critical field H_c of the MT observed at 4.2 K is comparatively low (about 95 kOe) for the former sample, while it is substantially higher (about 180 kOe) for the latter one with lower Al concentration, as shown in figure 1. A small difference in the Al concentration gives a large difference in the observed values of H_c. The sample preparation and magnetization measurements in a magnetic field up to 270 kOe have been described in [7]. A vibrating-sample magnetometer was used for the measurements of magnetic susceptibility.

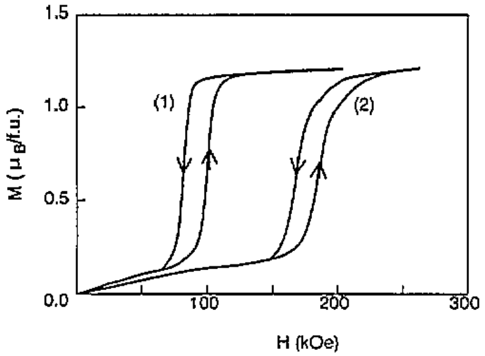

Figure 1. Field dependences of the magnetization at 4.2 K for the samples A (curve (1)) and B (curve (2)) observed in [7].

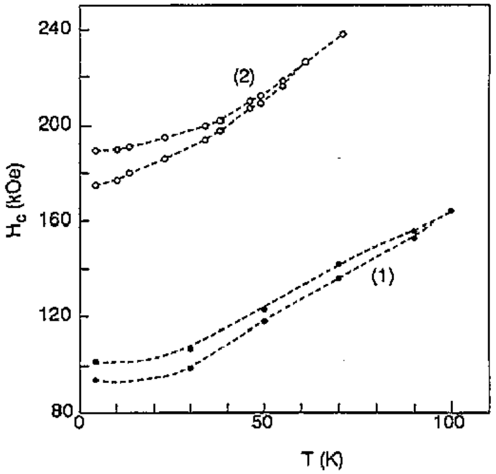

Figure 2. Temperature dependences of the critical field H_c of the MT for the samples A (filled circles) and B (open circles).

In figure 2, the temperature variation of the critical field H_c observed for the A and B samples is shown for increasing and decreasing external field. The value of H_c is evaluated by the field at which the derivative curve dM/dH passes through a maximum. Two characteristic tendencies are clearly seen in this figure: H_c increases and the MT is '*smeared*' (the hysteresis in the magnetization curve becomes narrower) with increasing temperature. These observed results are consistent with the spin fluctuation model for the MT [9]. The values of T_0 for the samples A and B, at which the hysteresis in the magnetization curve disappears, are evaluated as 95 and 60 K, respectively. Figure 3 shows the temperature dependence of the magnetic susceptibility observed for the samples A and B. In accordance with [7], they are paramagnetic down to 4.2 K as the Al concentration is less than 0.1. The values of $T_{\max}$ are about 100 and 125 K for samples A and B, respectively. In table 1, the observed values of H_c at 4.2 K and T_0, and of T_0 and $T_{\max}$ for the two compounds together with those for pure YCo$_2$ [5] are shown.

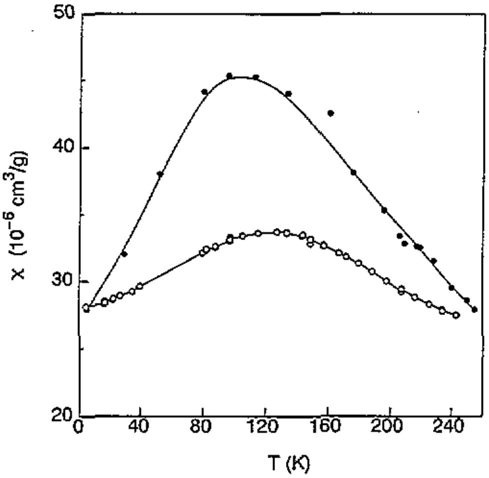

Figure 3. Temperature dependences of the paramagnetic susceptibility for the samples A (filled circles) and B (open circles).

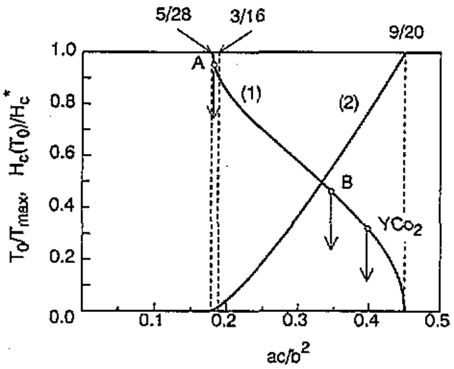

Figure 4. Estimated values of $T_0/T_{\max}$ (curve (1)) and $H_c(T_0)/H_c^*$ (curve (2)) as functions of ac/b^2. Arrows indicate the values of ac/b^2 estimated from the observed values of $T_0/T_{\max}$ for the samples A and B and for YCo$_2$ [5].

Table 1. The observed values of H_c at 4.2 K, $H_c(T_0)$, T_0 and $T_{\max}$ and estimated values of ac/b^2 and H_c^* for $(\mathrm{Y}_{1-t}\mathrm{Lu}_t)(\mathrm{Co}_{1-x}\mathrm{Al}_x)_2$ together with those for pure YCo$_2$ [5].

t	x	H_c (kOe)	$H_c(T_0)$ (kOe)	T_0 (K)	$T_{\max}$ (K)	ac/b^2	H_c^* (kOe)
0.41	0.085	95	160	95	100	0.18	—
0.365	0.075	180	225	60	125	0.34	430
0	0	700[a]	800	80	250	0.40[b]	1030

[a] Observed at 10 K [5].

[b] Yamada (1992) [13].

The equation of state for the magnetization M and the magnetic field H is written by the spin-fluctuation model for the MT [9] as

$$H = A(T)M + B(T)M^3 + C(T)M^5 \tag{1}$$

where

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

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

$$C(T) = c. \tag{4}$$

Here, a, b and c are coefficients of $|m(r)|^2$, $|m(r)|^4$ and $|m(r)|^6$ in the free energy density expanded with respect to the square of the magnetization density $m(r)$. $\xi(T)$ denotes the mean square root of the fluctuating magnetization in the paramagnetic state. It is noted that the M dependence of $\xi(T)$ under the magnetic field is neglected here. Thus the quenching of the spin fluctuations by the magnetic field is not taken into account in the present theory.

Neglecting the T dependences of the Landau coefficients a, b and c, the dependences on T of $A(T)$, $B(T)$ and $C(T)$ can be seen through their dependences on $\xi(T)$, as $\xi(T)$ is a monotonically increasing function of T [9]. When $a > 0$, $b < 0$ and $c > 0$, at which the MT may occur, $A(T)$ decreases with increasing $\xi(T)$ at small $\xi(T)$ and increases at large $\xi(T)$. Thus, $A(T)$ has a minimum at a certain value of $\xi(T)$. This means that the susceptibility $\chi(T)$ $(=A(T)^{-1})$ shows a maximum in the T dependence. As shown in [9], the temperature $T_{\max}$, at which $\chi(T)$ reaches a maximum, is given by

$$\xi(T_{\max})^2 = \tfrac{3}{14}|b|/c \tag{5}$$

The field-induced MT from the paramagnetic to the ferromagnetic state is shown from equation (1) to occur when the following conditions are satisfied [10]

$$A(T) > 0 \qquad B(T) < 0 \qquad C(T) > 0 \qquad \tfrac{3}{16} < A(T)C(T)/B(T)^2 < \tfrac{9}{20}. \tag{6}$$

It can be easily shown by equations (2)–(4) that the value of $A(T)C(T)/B(T)^2$ increases monotonically with increasing $\xi(T)$ and diverges at $T_{\max}$ as $B(T_{\max}) = 0$. When $a > 0$, $b < 0$, $c > 0$ and $\frac{3}{16} < ac/b^2 < \frac{9}{20}$, the value of $A(T)C(T)/B(T)^2$ becomes larger than $\frac{9}{20}$ at $T > T_0$, where T_0 is given by [9]

$$\xi(T_0)^2 = \tfrac{3}{14}(|b|/c)\left(1 - \sqrt{(\tfrac{70}{19})}\,\sqrt{ac/b^2 - \tfrac{5}{28}}\right). \tag{7}$$

By the microscopic spin-fluctuation theory, Lonzarich and Taillefer [11] and Moriya [12] have shown that $\xi(T)$ is proportional to T at low T. Then, from equations (5) and (7) the ratio between T_0 and $T_{\max}$ is obtained as [13]

$$(T_0/T_{\max})^2 = 1 - \sqrt{\tfrac{70}{19}}\sqrt{ac/b^2 - \tfrac{5}{28}} \tag{8}$$

where T_0 and $T_{\max}$ are assumed to be low enough. From equation (8) with the observed values of T_0 and $T_{\max}$, the value of ac/b^2 for YCo_2 was estimated as 0.4 [13].

The critical field $H_c(T_0)$ at $T = T_0$ is obtained from the condition that the equation $\partial H/\partial M = 0$ has an equal root, i.e., the hysteresis in the magnetization curve disappears. We get

$$H_c(T_0)/H_c^* = \left[\tfrac{70}{19}\left(ac/b^2 - \tfrac{5}{28}\right)\right]^{5/4} \tag{9}$$

where

$$H_c^* = \tfrac{6}{25}\sqrt{\tfrac{3}{10}|b|/c}\; b^2/c \tag{10}$$

which is the value of $H_c(T_0)$ at $ac/b^2 = \frac{9}{20}$. In terms of $\chi(0)(= 1/a)$ and $\xi(T_{\max})$ given by equation (5), H_c^* is rewritten as

$$H_c^* = \tfrac{6}{25}\sqrt{\tfrac{7}{5}}\,(b^2/ac)\,\xi(T_{\max})/\chi(0). \tag{11}$$

In figure 4, the values of $T_0/T_{\max}$ and $H_c(T_0)/H_c^*$ given by equations (8) and (9) are shown as a function of ac/b^2. From these curves and the observed values of $T_0/T_{\max}$, the values of ac/b^2 can be evaluated as shown in table 1. The following results are obtained by comparison between the present theory and experiment.

(i) In the $(Y_{1-t}Lu_t)(Co_{1-x}Al_x)_2$ system, the critical value $\frac{3}{16}$ ($\simeq 0.188$) for ac/b^2, corresponding to the ferromagnetic boundary of an itinerant-electron paramagnet, is attained at $x = 0.09$ [7]. Therefore, nearly the same value of ac/b^2 for the sample A ($x = 0.085$) is expected. The value of ac/b^2 estimated from the observed $T_0/T_{\max}$ is a little smaller than $\frac{3}{16}$ as shown by an arrow in figure 4. This means that the ferromagnetic state becomes stable at 0 K, which is incompatible with the observed result. However, $\chi(T)$ shows a very broad maximum as shown in figure 3. Therefore, a few percent error is involved in the estimation of $T_{\max}$. Moreover, the value of T_0 has also a few percent error in itself. As the value of ac/b^2 estimated above is very close to the critical value, so changes of only a few percent in $T_{\max}$ and T_0 are able to give a value of ac/b^2 larger than $\frac{3}{16}$. In any case, the theoretical prediction that the values of T_0 and $T_{\max}$ for sample A are very close to each other has been confirmed by the present observed results.

(ii) As long as $T_0/T_{\max}$ decreases monotonically with decreasing Al concentration, the value of ac/b^2 increases in the same way, being about 0.34 for sample B and 0.40 for pure YCo_2 [13]. The Landau coefficient b is negative for YCo_2 and should be positive for the ferromagnetic compounds at $x > 0.9$ with the second-order magnetic phase transition at the Curie temperature. Then, the absolute value of b will decrease with increasing x. Assuming that the Landau coefficient c does not depend so much on x, $\xi(T_{\max})$ given by equation (5) decreases with increasing x. As $\xi(T_{\max})$ is a monotonically increasing function of $T_{\max}$, then the value of $T_{\max}$ decreases with increasing x, which is consistent with the observed result.

(iii) As shown in figure 4, the reduced critical field $H_c(T_0)/H_c^*$ increases with decreasing Al concentration. The values of H_c^* for sample B and YCo_2 are 430 and 1030 kOe, respectively, which are estimated from the value of ac/b^2 obtained above and the observed value of $H_c(T_0)$. As $\xi(T_{\max})$ is proportional to $T_{\max}$ at low $T_{\max}$, then H_c^* given by equation (11) is proportional to $T_{\max}$ and also to $\chi(0)^{-1}$. The observed value of $T_{\max}$ for sample B is about half of that for pure YCo_2. Moreover, the value of $\chi(0)$ for the former is about three times larger than that for the latter. Then, the value of H_c^* for sample B becomes much smaller than that for pure YCo_2, which is also consistent qualitatively with the result estimated above.

In this way, the spin fluctuation model developed here has been able to explain qualitatively the main features in the high-temperature behaviours observed in the (Y,Lu)(Co,Al)$_2$ system, revealing a correlation between the low-field and high-field properties. In the present theory, the temperature dependences of the Landau coefficients a, b and c are neglected. They originate from the Fermi distribution functions involved in the respective expression of a, b and c [14]. However, their dependences on temperature will be weak, compared with the contributions from the spin fluctuations, because the effective degenerate temperature in the Fermi distribution function is rather high. Moreover, in the present model the suppression of spin fluctuations by the external magnetic field is not taken into account. Such a quenching of spin fluctuations may affect the critical field of the MT at finite temperature. This problem is left for future work.

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