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Itinerant electron metamagnetic transition in exchange enhanced paramagnetic compounds $Lu(Co_{1-x}Sn_x)_2$

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Abstract. Magnetization measurements of C-15 type Laves phase compounds $Lu(Co_{1-x}Sn_x)_2$ have been carried out up to 100 T in order to investigate the itinerant electron metamagnetic transition. By partial replacement of Co by Sn, the lattice constant increases and the critical magnetic field decreases linearly with increasing x. The susceptibility increases until $x = 0.04$ and then decreases above $x = 0.06$. A clear itinerant electron metamagnetic transition has been observed in the whole concentration range. It has been demonstrated that this transition occurs even in a pure compound $LuCo_2$ and its critical magnetic-transition field is determined to be 74 T, not so different from the value predicted by the band calculation. On the other hand, the observed magnetization jump of $LuCo_2$ is about 0.5 μ_B/Co, which is much larger than the theoretical value.

1. Introduction

C-15 type Laves phase compounds such as YCo_2, $LuCo_2$ and $ScCo_2$ have been investigated extensively from both experimental and theoretical viewpoints. These systems exhibit a maximum in the temperature dependence of magnetic susceptibility (Lemaire 1966, Bloch *et al* 1971, Ishiyama *et al* 1984, Yoshimura *et al* 1987). The magnitudes of susceptibility and electronic specific heat coefficient of these exchange-enhanced Pauli paramagnets are about ten times (Gschneider Jr and Ikeda 1983) larger than those of unenhanced Pauli paramagnetic compounds (Burzo and Laforest 1972). These physical properties have been successfully explained by the spin fluctuation theory (Konno and Moriya 1987).

Itinerant electron metamagnetic transition, that is, the first-order field induced transition from a paramagnetic to a ferromagnetic state was first discussed on the basis of the band model (Wohlfarth and Rhodes 1962). The occurrence of the first-order magnetic phase transition in some exchange-enhanced Pauli paramagnets has been predicted by many workers (Cyrot and Lavagna 1979, Wohlfarth 1980, Jarlborg and Freeman 1981). The theoretical estimation of the critical magnetic fields for YCo_2, $LuCo_2$ and $ScCo_2$ has been made by using the tight-binding approximation for d-electrons (Shimizu 1982, Yamada and Shimizu 1985, Yamada *et al* 1987). However, the

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experimental realization is very difficult because the critical magnetic field is estimated to be very high, about 100 T.

It has been reported that the partial replacement of Co by Al is effective for the metamagnetic transition in lower magnetic fields. Systematic studies of the magnetic properties of the pseudo-binary compounds $Y(Co_{1-x}Al_x)_2$ revealed that the lattice constant increases and the metamagnetic transition occurs in relatively low magnetic fields (Aleksandryan *et al* 1985, Sakakibara *et al* 1986, Sakakibara *et al* 1987), resulting in the magnetic enhancement with increasing x (Yoshimura and Nakamura 1985). In the same way, the itinerant electron metamagnetic transition in the compounds $Lu(Co_{1-x}Al_x)_2$ has been investigated by several authors. The observed metamagnetic transition, however, is unusually broad because of the magnetic heterogeneity of Co atoms (Shinogi *et al* 1987, Sakakibara *et al* 1987) and the critical magnetic field of $LuCo_2$ estimated experimentally (Endo *et al* 1988) is very high compared with the theoretical value (Yamada *et al* 1987). On the other hand, recently, we have developed new pseudo-binary compounds $Lu(Co_{1-x}Sn_x)_2$ which show a clear metamagnetic transition. In the present study, the itinerant electron metamagnetic transition has been investigated systematically and compared with the theoretical considerations for YCo_2 and $LuCo_2$.

2. Experiment

The alloys were made by arc melting in an argon atmosphere. The Lu content was kept slightly higher than the stoichiometric composition because of some loss of Lu due to vaporization and oxidation during the alloying. Annealing of $Lu(Co_{1-x}Sn_x)_2$ for homogenization was carried out at 1073 K for 2 weeks in a vacuum quartz tube. The magnetic susceptibility was measured with a SQUID magnetometer. The samples were powdered for the magnetization measurements in pulsed high magnetic fields with a particle size of less than 50 μm and shaped into a rod with epoxy resin to eliminate any eddy current effect. High magnetic fields up to 42 T were generated with a multilayer pulse magnet. Ultra-high magnetic fields up to 105 T were generated by means of a fast capacitor discharge into a copper single-turn coil with a 100 kJ capacitor bank (Nakao *et al* 1985). The magnetization was measured by an induction method with well balanced pick up coils. Details of the experimental procedure have already been described (Sakakibara *et al* 1990a). The room temperature lattice constants were determined by x-ray diffraction.

3. Results and discussion

Temperature dependence of the magnetic susceptibility measured in 3 T for five compounds $Lu(Co_{1-x}Sn_x)_2$ is shown in figure 1. The susceptibility exhibits a broad maximum characterizing exchange-enhanced Pauli paramagnets (Lemaire 1966, Bloch *et al* 1971, Ishiyama *et al* 1984, Yoshimura *et al* 1987, Sakakibara *et al* 1990a, b). The temperature of this maximum T_m shifts to lower temperature ranges with increasing Sn concentration up to $x = 0.04$ and then goes up above $x = 0.06$. In compounds $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Al_x)_2$, the temperature of the maximum T_m has been confirmed to be closely related to the critical magnetic field B_c for itinerant electron metamagnetism and the value of B_c/T_m becomes constant (Sakakibara *et al* 1990b). In the present $Lu(Co_{1-x}Sn_x)_2$, the linear relationship between B_c and T_m is not maintained in the wide concentration

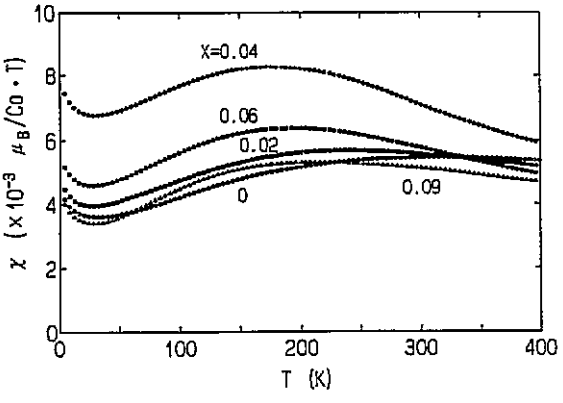

Figure 1. Temperature dependence of the magnetic susceptibility of the pseudo-binary compounds $Lu(Co_{1-x}Sn_x)_2$.

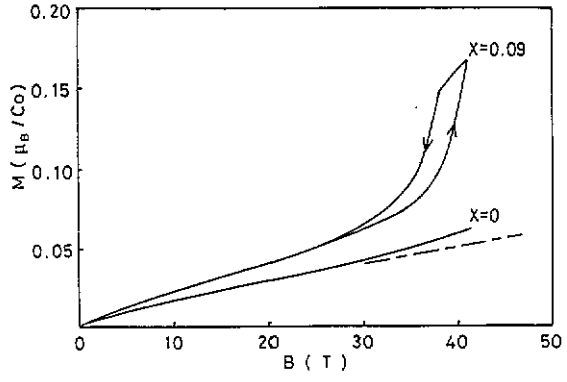

Figure 2. Magnetization curves of the pseudo-binary compounds $Lu(Co_{1-x}Sn_x)_2$ up to 42 T at 4.2 K in long pulse fields.

range $x = 0$–0.09. It is expected, however, that the itinerant electron metamagnetic transition takes place in a relatively low magnetic field because the value of T_m for $Lu(Co_{1-x}Sn_x)_2$ is lower than that of $LuCo_2$ as seen from figure 1.

The magnetization curves of $x = 0$ and $x = 0.09$ for $Lu(Co_{1-x}Sn_x)_2$ measured in magnetic fields up to 42 T are presented in figure 2. As expected, the itinerant metamagnetic transition occurs at the rather low field of 37 T for $x = 0.09$. It is noteworthy that the magnetization of $LuCo_2$ deviates slightly upward from the linear dotted line above 30 T, giving an indication of the metamagnetic transition. Therefore, higher magnetic fields are necessary to set off the metamagnetic transition in $LuCo_2$. On the other hand, the convex upward curvature observed below 10 T may originate from some ferromagnetic inclusions undetected by x-ray diffraction. The enhancement susceptibility in the low temperature ranges in figure 1 is attributed to the same cause.

Figure 3 shows the magnetization curves measured in ultra-high magnetic fields up to 94 T. A clear itinerant electron metamagnetic transition is observed, accompanied by a hysteresis of several Teslas due to the first-order magnetic phase transition in a similar manner to that in $Y(Co_{1-x}Al_x)_2$ (Sakakibara *et al* 1990a). The critical magnetic fields B_c of the transition are determined from the centres of the hysteresis curves. In the present Laves phase compounds, B_c shifts to a higher field with decreasing x and eventually $LuCo_2$ exhibits a sharp itinerant electron metamagnetic transition at 74 T, which is lower by 20 T than the recent calculated value of 94 T (Yamada *et al* 1987). The

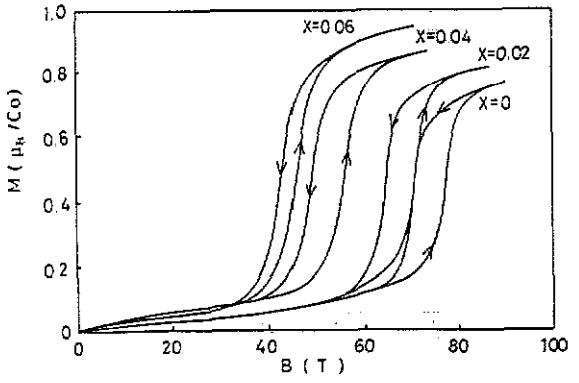

Figure 3. Magnetization curves of the pseudo-binary compounds $Lu(Co_{1-x}Sn_x)_2$ up to 92 T at 4.2 T in ultra-high magnetic fields.

Table 1. Theoretical and experimental values of the critical magnetic fields B_c and the magnetization jump ΔM for the Laves phase compounds YCo_2, $LuCo_2$ and $ScCo_2$.

		YCo_2	$LuCo_2$	$ScCo_2$
$B_c(T)$	Theoretical[a]	89	94	120
	Experimental	69[b]	74	105<
$\Delta M(\mu_B/Co)$	Theoretical[a]	0.25	0.28	0.30
	Experimental	0.27	0.49	—

[a] H Yamada *et al* 1987.
[b] T Goto *et al* 1989.

value of B_c for $LuCo_2$ has been estimated to be about 180 T from the Al concentration dependence of the critical magnetic field (Endo *et al* 1988). This over-estimation of B_c may be due to the unusually broad transition in the compounds $Lu(Co_{1-x}Al_x)_2$ (Shinogi *et al* 1987). By the nuclear magnetic resonance of ^{59}Co, this broad transition has been attributed to the coexistence of non-magnetic and magnetic Co atoms (Shinogi *et al* 1987). On the other hand, it should be noted that the clear itinerant electron metamagnetic transition in YCo_2 has been realized in a pulsed ultra-high magnetic field (Goto *et al* 1989) and its critical magnetic field of 69 T is not so different from the theoretical value of 89 T (Yamada *et al* 1987).

For the Laves phase compounds YCo_2 (Goto *et al* 1989) and $LuCo_2$, the critical magnetic fields B_c and the magnetization jump ΔM obtained from the band calculations (Yamada *et al* 1987) and those values from the experiments are given in table 1 for comparison. The experimental values of B_c are smaller than the theoretical ones. In the tight-binding approximation, the volume of $LuCo_2$ was assumed to be constant in the magnetic field (Yamada *et al* 1987). However, a large volume change associated with the metamagnetic transition has been observed in $Y(Co_{1-x}Al_x)_2$ (Wada *et al* 1988). Such a remarkable volume magnetostriction is also expected in $LuCo_2$. The theoretical critical magnetic fields for YCo_2 and $LuCo_2$ are considered to be reduced by taking into account this volume change. The magnetization jump ΔM is evaluated to be 0.25 and 0.28 μ_B/Co for YCo_2 and $LuCo_2$, respectively (Yamada *et al* 1987). The theoretical value is consistent with the observed value of 0.27 μ_B/Co for YCo_2 (Goto *et al* 1989, Sakakibara *et al* 1990a) but the observed value for $LuCo_2$ is estimated to be about 0.49 μ_B, which is

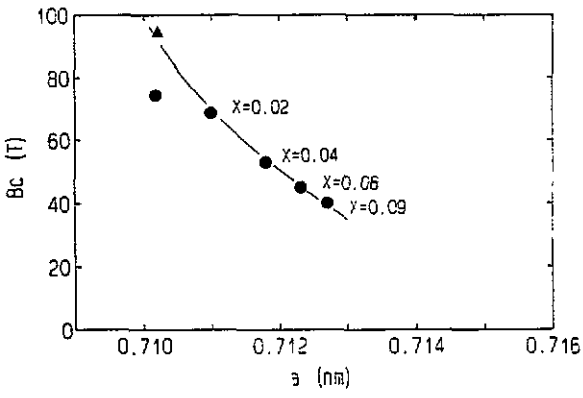

Figure 4. Relationship between the critical magnetic field and the room temperature lattice constant of the pseudo-binary compounds $Lu(Co_{1-x}Sn_x)_2$. ▲, theoretical results; ●, experimental results.

apparently higher than that of YCo_2, being inconsistent with the band calculation (Yamada *et al* 1987). It is interesting to note that the ferromagnetic phase of the compounds $Lu(Co_{1-x}Al_x)_2$ with high magnetization accompanies a remarkable spontaneous volume magnetostriction, showing the Invar effect (Gabelko *et al* 1987, Iijima *et al* 1990), even though the ferromagnetic compounds $Y(Co_{1-x}Al_x)_2$ exhibit no remarkable volume effect. The magnetization is not saturated easily, as seen from figure 3, after transition, and is accompanied by a large high-field susceptibility in a similar manner to that of YCo_2 (Goto *et al* 1989). On the other hand, the saturation magnetic moment of Co of YCo_2 amorphous alloy is estimated to be $1.0\,\mu_B$ (Fukamichi *et al* 1986). This suggests that the magnetic moment of $LuCo_2$ is also saturated to the same value in the much higher fields.

In the pseudo-binary compounds $Sc(Co_{1-x}Al_x)_2$, no itinerant electron metamagnetic transition has been observed up to 52 T (Ishiyama *et al* 1987). Therefore, the critical magnetic field of $ScCo_2$ is considered to be much higher than that of YCo_2 and $LuCo_2$ as predicted theoretically (Yamada *et al* 1987). In the present study, the magnetization measurement of $ScCo_2$ has been attempted up to 105 T. No indication of itinerant electron metamagnetic transition, however, is observed in the magnetization curve. Consequently, it has become apparent that the critical magnetic field of $ScCo_2$ is not too low, compared with the theoretical value, in contrast to the results of YCo_2 and $LuCo_2$.

It has been pointed out that the lattice expansion promotes a tendency to induce the metamagnetic transition (Yoshimura and Nakamura 1985). In the present compounds, therefore, the lattice constant has been measured. The room temperature lattice constant of the compounds $Lu(Co_{1-x}Sn_x)_2$ increases with increasing Sn content. Figure 4 shows the critical magnetic field B_c versus the lattice constant a for these compounds. The theoretical point (Yamada *et al* 1987) and four experimental points above $x = 0.02$ give a smooth curve, being slightly convex downward. The experimental value of $LuCo_2$ deviates from the line. This fact suggests that the critical magnetic field lower than the theoretical field for $LuCo_2$ is realized by the forced volume magnetostriction induced by the ultra-high magnetic field as mentioned in the discussion of table 1. Furthermore, it is expected that the magnetostriction of the pseudo-binary compounds is not so large because the critical magnetic field changes linearly with the lattice constant as shown in the figure.

It has been demonstrated, although theoretical explanations are not yet given, that there is a linear correlation between the critical magnetic field B_c and the temperature

of susceptibility maximum T_m in not only the pseudo-binary compounds $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Al_x)_2$ but also $U(Pt_{1-x}Pd_x)_3$ and $CeRu_2Si_2$ (Sakakibara *et al* 1990b, De Visser *et al* 1987, 1988, Mignot *et al* 1988). As mentioned in the discussion of figure 1, however, T_m of the present compounds $Lu(Co_{1-x}Sn_x)_2$ does not show a monotonic concentration dependence. Therefore, it is apparent that the critical magnetic field of the compounds $Lu(Co_{1-x}Sn_x)_2$ is closely correlated to the lattice constant rather than the temperature of susceptibility maximum.

In the case of $Y(Co_{1-x}Al_x)_2$, the susceptibility at 0 K (Sakakibara *et al* 1990a) and the electronic specific heat coefficient γ becomes larger with the Al content in the paramagnetic concentration range (Wada *et al* 1989). The maximum value of γ is concerned with the appearance of ferromagnetism (Wada *et al* 1989). However, the susceptibility of the present compounds $Lu(Co_{1-x}Sn_x)_2$ increases at the beginning and decreases above $x = 0.06$ as seen from figure 1 and the coefficient γ of these compounds also shows a similar tendency, accompanied by the maximum value in the concentration dependence curve without the onset of ferromagnetism (Murata *et al* 1990). The different properties in these two compound systems would be originated from the difference of the substitutional elements. Since Sn atoms have d-electrons, the Fermi level of $LuCo_2$ may be modified effectively, compared with that of YCo_2 by Al. Although the width of d-bands becomes narrower by addition of Sn, due to the lattice expansion, the Fermi level may be shifted to the high energy side, resulting in a decrease in the density of state at the Fermi level when the concentration of Sn exceeds some critical value. According to the theoretical consideration (Wohlfarth and Rhodes 1962), the critical magnetic field B_c has the following relation;

$$B_c \propto M_{onset}/\chi_0$$

where M_{onset} and χ_0 are the magnetization at the onset of metamagnetic transition and the susceptibility at 0 K, respectively. The reason why the critical magnetic field decreases as shown in figure 3 although the susceptibility becomes small above $x = 0.06$ as seen from figure 1 might be related to the decrease in M_{onset}. By addition of Sn, the d-bands would move downward relative to the Fermi level as predicted by CPA calculations (Velicky *et al* 1968), resulting in the increase in the second derivative of the density of state at the Fermi surface. This effect would bring about the decrease in the value of M_{onset}. In order to investigate the effect of the deferent substitutional element on the itinerant electron metamagnetic transition, studies on the magnetic properties and the low temperature specific heat of other pseudo-binary compounds such as $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ are in progress.

4. Summary

The itinerant electron metamagnetic transition in the pseudo-binary compounds $Lu(Co_{1-x}Sn_x)_2$ have been investigated in the ultra-high magnetic fields. The critical magnetic field of $LuCo_2$ has been compared with the theoretical value and the results for YCo_2. The critical magnetic field change in the present compounds $Lu(Co_{1-x}Sn_x)_2$ is closely correlated with the lattice constant. The main results are summarized as follows;

(1) The temperature of susceptibility maximum T_m of $Lu(Co_{1-x}Sn_x)_2$ first shifts to lower temperature ranges with increasing Sn concentration up to $x = 0.04$ and then increases above $x = 0.06$.

(2) The susceptibility χ of $Lu(Co_{1-x}Sn_x)_2$ increases with x up to $x = 0.04$ and decreases above $x = 0.06$.

(3) The critical magnetic field of $LuCo_2$ is 74 T which is slightly higher than that of YCo_2 and lower by about 20 T than the theoretical value calculated from the density of state.

(4) The magnetization jump of $LuCo_2$ is 0.49 μ_B/Co, which is apparently larger than the theoretical value.

(5) The linear relationship between the critical magnetic field and the temperature of susceptibility maximum is not observed in $Lu(Co_{1-x}Sn_x)_2$.

(6) The critical magnetic field becomes lower with increasing Sn content in accordance with the increase in the room temperature lattice constant.

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