Pressure Effect on Itinerant-Electron Ferromagnetic Properties of Lu(Co$_{0.90}$Al$_{0.10}$)$_2$ Laves Phase Intermetallic Compound

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

*Institute for Solid State Physics, The University of Tokyo, Roppongi Minato-ku, Tokyo 106-0032, Japan

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Itinerant-electron magnetic properties of Lu(Co$_{0.90}$Al$_{0.10}$)$_2$ Laves phase intermetallic compound under high pressures up to 1.0 GPa have been investigated. The compound has a critical concentration of the onset of ferromagnetism. The spontaneous magnetization decreases with increasing pressure and disappears above 0.6 GPa. In the paramagnetic state induced by applying pressure, a metamagnetic transition from a paramagnetic to ferromagnetic state occurs in magnetic fields. An anomalously large negative pressure dependence of the Curie temperature has been observed, which is explained by the Landau expansion of free energy taking spin fluctuations into consideration.

Key words: itinerant-electron, pressure effect, Curie temperature, spontaneous magnetization, spin fluctuations

1. Introduction

RM$_2$ (R and M indicate rare earth and 3d transition metals, respectively) Laves phase intermetallic compounds show variety of magnetic properties. Some of them exhibit large magnetovolume effects such as a large spontaneous volume magnetostriiction and a large effect of pressure on the Curie temperature. RCo$_2$ compounds are strongly exchange-enhanced Pauli paramagnets when R is non-magnetic element such as Y and Lu. These compounds exhibit a metamagnetic transition from a paramagnetic to ferromagnetic state by applying high magnetic fields$^{1, 2}$. The origin of the metamagnetic transition is associated with a special band structure which exhibits a sharp peak of the density of state (DOS) just below the Fermi level$^3$.

By a partial replacement of Co by Al, a critical transition field $H_c$ is reduced to a relatively low magnetic field$^4$. Eventually, ferromagnetism is established above $x = 0.105$. Thus, pseudo-binary Lu(Co$_{1-x}$Al$_x$)$_2$ Laves phase compounds are suitable to study itinerant-electron metamagnetism and ferromagnetism.

Magnetic properties such as the spontaneous magnetization $M_s$ and the Curie temperature $T_C$ are expected to be sensitive to pressure because the applying pressure results in an increase of the band width.

Recently, itinerant-electron metamagnetism and ferromagnetism have been discussed in terms of spin fluctuations$^6$. According to this theory, a large negative pressure dependence of the Curie temperature $T_C$ is expected near the critical concentration between the ferromagnetic and metamagnetic state$^7$.

In the present study, the pressure effects on the Curie temperature $T_C$ and the spontaneous magnetization $M_s$ of Lu(Co$_{0.90}$Al$_{0.10}$)$_2$ Laves phase intermetallic compound have been investigated. The experimental results of the pressure dependence of $T_C$ and $M_s$ are discussed by the Landau expansion of free energy taking spin fluctuations into account$^7$.

2. Experimental

The specimen was prepared by arc-melting in an argon gas atmosphere. The Lu content was kept slightly higher than the stoichiometric composition to avoid a precipitation of ferromagnetic phase due to a loss of Lu during arc-melting. The compound was homogenized at 1273 K for a week in an evacuated quartz tube followed by quenching into water. The crystal structure was identified by x-ray as a C15-type Laves phase without any other phases. The specimen was compressed in a Teflon cell filled with a Fluorinate in high pressure Cu-15 Ti clamp. The effect of hydrostatic pressure on the magnetization was measured in fields up to 9 T by an extraction-type magnetometer. Hydrostatic pressures at low temperatures were calibrated by a superconducting transition temperature of Pb.

3. Results and discussion

The temperature dependence of magnetization in a magnetic field in 0.5 T at various pressures is given in Fig. 1. The composition of the specimen corresponds to the critical concentration of the onset of ferromagnetism. The spontaneous magnetization $M_s$ and the Curie temperature $T_C$ show large negative pressure dependences. In particular, an anomalously large negative pressure dependence of $T_C$ has been observed. The inset shows the pressure dependence of $T_C$ obtained from $M^2$-T plots. With increasing pressure, $T_C$ decreases linearly and disappears at about 0.6 GPa, showing the disappearance of ferromagnetism. In connection with the remarkable pressure effect on $T_C$, the present system is also expected to show a significant influence on the magnetization process by applying pressure. Shown in Fig. 2 is the magnetization curves at 4.2 K as a function of pressure. It should be noted that the magnetization curves are also
Fig. 1 Temperature dependence of magnetization at various pressures in a magnetic field in 0.5 T. The inset shows the pressure dependence of the Curie temperature $T_c$.

The magnetization curve is very sensitive to the pressure. At atmospheric pressure, the ferromagnetic state is stable. With increasing pressure, $M_r$ obtained by the Arrott-plots decreases and disappears above 0.6 GPa. In the paramagnetic state induced by applying pressure, the metamagnetic transition occurs in high magnetic fields and its critical transition field $H_{cr}$ increases with increasing pressure, suggesting that the paramagnetic state becomes much stabler. This phenomenon is explained by considering a decrease of the DOS at the Fermi level caused by applying pressure.

The magnetic free energy $F$ of the itinerant-electron systems is expressed by the following expression,

$$ F = -\frac{1}{2} a M^2 + \frac{1}{4} b M^4 + \frac{1}{6} c M^6, \quad (1) $$

where $a$, $b$, and $c$ are the Landau coefficients. For the metamagnetic transition, the condition of $a>0$, $b<0$ and $c>0$ is required because $F$ takes a double minimum in the magnetic field under this condition. By examining the dependence on the magnetization of the free energy for electrons at finite temperature in the band model, the coefficients can be in principle determined from the DOS near the Fermi level. It is clear that the sign of $b$ is negative because the metamagnetic transition occurs by applying magnetic field in the paramagnetic state induced by pressure. The negative $b$ is correlated with a special band structure near the Fermi level. That is, this condition is realized when the Fermi level lies just near a sharp peak of the DOS. Therefore, the present result implies that there is a sharp peak of the DOS just below the Fermi level. At finite temperatures, on the other hand, the free energy coefficients are renormalised by thermal spin fluctuations\(^7\). According to Yamada and Terao, the pressure dependence of $T_c$ for ferromagnets with a negative sign of $b$ can be expressed by the following equation\(^7\),

$$ \frac{\partial \xi_p(T_c^2)}{\partial P} = -\frac{3\kappa C_{mv}}{\sqrt{35b}} \left[ \frac{5}{28} \frac{ac}{b^2} \right]^{1/2}, \quad (2) $$

where $\xi_p(T_c^2)$, $\kappa$ and $C_{mv}$ are the mean-square amplitude of spin fluctuations at $T_c$, the compressibility and the magneto-volume coupling constant, respectively. The value of $\partial \xi_p(T_c^2)/\partial P$ is proportional to $\partial T_c^2/\partial P$, because $\xi_p(T_c^2)$ is proportional to $T_c^2$. This equation indicates that a significantly large negative value of $\partial T_c^2/\partial P$ is observed near the critical concentration of the onset of ferromagnetism because the condition of $ac/b^2 = 5/28$ is very near the paramagnetic state. Therefore, the present result can be explained by the theory associated with the spin fluctuations under the condition of $a>0$, $b<0$ and $c>0$.

As seen from Figs. 1 and 2, it is considered that the reduction of $T_c$ is different from that of $M_r$, because $M_r$ is reduced more significantly at high pressures. To make this point clear, $T_c/M_r(0)$ and $M_r/M_r(0)$ against pressure is plotted in Fig. 3. The values of $T_c(0)$ and $M_r(0)$ are $T_c$ and $M_r$ at atmospheric pressure, respectively. With increasing pressure, $M_r$ begins to decrease drastically above a critical pressure while $T_c$ shows a monotonic decrease. The pressure coefficients of $\partial \ln T_c/\partial P$ and $\partial \ln M_r/\partial P$ are estimated to be 1.63 GPa\(^{-1}\) and 0.26 GPa\(^{-1}\), respectively. Such a significant difference between the
magnitude of $\partial \ln M_0 / \partial P$ and $\partial \ln T_c / \partial P$ should be noted. In the case of weak ferromagnets with the coefficients condition of $a<0$, $b>0$ and $c=0$, the ratio of $\partial \ln T_c / \partial P$ to $\partial \ln M_0 / \partial P$, that is, $\partial \ln T_c / \partial \ln M_0$ is unity in the Stoner-Wohlfarth theory and $3/2$ in the theory associated with spin fluctuations. As mention above, Lu(Co$_{0.90}$Al$_{0.10}$)$_2$ compound satisfies the condition of $a>0$, $b<0$ and $c>0$. The difference between the magnitude of $\partial \ln T_c / \partial P$ and $\partial \ln M_0 / \partial P$ would be responsible for an extremely large value of $\partial T_c / \partial P$ under the condition of $a>0$, $b<0$ and $c>0$.

4. Conclusion

Effects of pressure on the Curie temperature $T_c$ and the spontaneous magnetization $M_s$ of itinerant-electron ferromagnetic Lu(Co$_{0.90}$Al$_{0.10}$)$_2$ Laves phase intermetallic compound have been investigated. The main results are summarized as follows:

(a) Both $M_s$ and $T_c$ decrease with increasing pressure and disappear at about 0.6 GPa.

(b) In the paramagnetic state induced by applying pressure, a metamagnetic transition from a paramagnetic to ferromagnetic state occurred in high magnetic fields. This means that the sign of the coefficient $b$ is negative, suggesting that there is a sharp peak of DOS just below the Fermi level.

(c) Due to the negative sign of $b$, a large negative pressure dependence of $T_c$ is observed and its pressure coefficient is much larger than that of $M_s$.

(d) These phenomena are explained by the Landau expansion taking spin fluctuations into account under the condition of $a>0$, $b<0$ and $c>0$.

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References