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1999 J. Phys.: Condens. Matter 11 8095

(http://iopscience.iop.org/0953-8984/11/41/312)

J. Phys.: Condens. Matter **11** (1999) 8095–8101. Printed in the UK PII: S0953-8984(99)04562-2

# Itinerant electron metamagnetism of $Y(Co_{1-x}Al_x)_2$ under high pressure and high magnetic fields

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Received 26 May 1999

**Abstract.** The magnetization curves of itinerant electron metamagnets $Y(Co_{1-x}Al_x)_2$ have been measured in high magnetic fields up to 42 T under high pressure to 1 GPa. The critical field of the metamagnetic transition $B_c$ was found to increase with pressure with a rate $dB_c/dP = 7.8$ T GPa$^{-1}$ for $x = 0.075$ and 7.4 T GPa$^{-1}$ for $x = 0.09$. The initial volume compressibility amounts to $8.7 \times 10^{-3}$ GPa$^{-1}$ at 77 K. The decrease of $B_c$ due to Al substitution for Co in $Y(Co_{1-x}Al_x)_2$ has been considered to originate from two separate mechanisms: the change of the electronic structure produced by Al substitution under the condition of constant volume and the change by the increase in the interatomic distance. The two contributions are estimated to be nearly equal.

## 1. Introduction

The intermetallic compounds $YCo_2$ and $LuCo_2$ are typical of exchange-enhanced Pauli paramagnets. External high magnetic fields induce a ferromagnetic state in these compounds. Such a first-order metamagnetic transition, which is called itinerant electron metamagnetism, was directly observed in $YCo_2$ and $LuCo_2$ at critical fields $B_c = 69$ T and 74 T by measuring the magnetization in ultra-high magnetic fields up to 100 T [1]. In $Y(Co_{1-x}Al_x)_2$, the critical field of the metamagnetic transition decreases when the concentration of aluminium increases [2]. The compounds with $0.12 < x < 0.20$ show weakly ferromagnetic behaviour [3].

The occurrence of the metamagnetic transition is considered to originate from a special shape of the density of states curve near the Fermi level [4]. 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 density of states near the Fermi level has a positive curvature [5], which enhances the spin fluctuations and the susceptibility. An applied magnetic field increases the density of states at the Fermi level $N(E_f)$ and the itinerant metamagnetic transition to a field-induced ferromagnetic state occurs when the Stoner condition for the appearance of ferromagnetism is satisfied.

The application of hydrostatic pressure to the itinerant metamagnetic compound is very useful for examining the mechanism of the metamagnetic transition. The change in the interatomic distance by external pressure leads to the increase in the bandwidth. As a result, the value of $N(E_f)$ can be finely reduced. Goto *et al* [6] showed that external pressure suppresses ferromagnetism in ferromagnetic $Lu(Co_{0.88}Ga_{0.12})_2$. A broad metamagnetic transition appears

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with increasing pressure and the critical field increases linearly with $dB_c/dP = 10$ T GPa$^{-1}$. Similar results were obtained recently for $Lu(Co_{0.90}Al_{0.10})_2$, the paramagnetic composition of which is very close to the ferromagnetic concentration range [7]. No study has been reported for Y systems. It is known, however, that the magnetic behaviour of $Y(Co_{1-x}Al_x)_2$ differs from that of the compounds with Lu. In the case of $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Al_x)_2$ the spontaneous magnetization in the ferromagnetic concentration range is consistent with the field-induced magnetization in the paramagnetic concentration range [8, 9]. In contrast, the compounds with Y show only a weak spontaneous moment considerably lower than that induced by magnetic fields [2, 3].

Recently, we developed a new instrument which can measure precisely the magnetization curve of the sample under high pressure up to 1 GPa in pulsed high magnetic fields up to 45 T. In this study, we have measured the magnetization curve of paramagnetic $Y(Co_{1-x}Al_x)_2$ with $x = 0.075$ and 0.09 at 4.2 K under high pressure to clarify the origin of the change in the metamagnetic transition due to Al substitution.

## 2. Experimental details

Polycrystalline samples of $Y(Co_{1-x}Al_x)_2$ have been prepared in a semi-levitation crucible by induction melting of the constituents under an argon atmosphere, followed by annealing at 900 °C for a week. Powder x-ray diffraction patterns were obtained using monochromatized Cu $K_\alpha$ radiation. The samples are confirmed to be single phase with the cubic Laves phase structure.

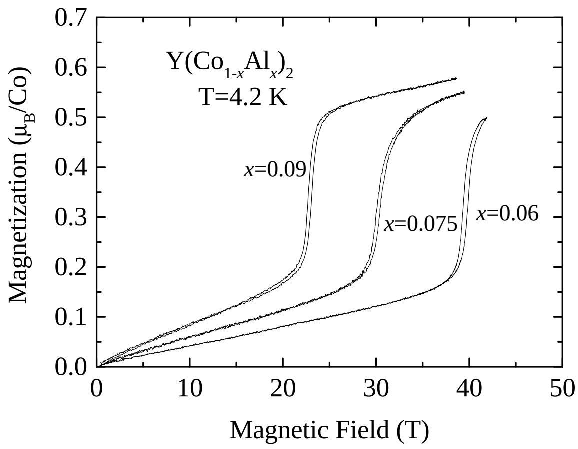


**Figure 1.** Magnetization curves of $Y(Co_{1-x}Al_x)_2$ samples with different $x$ for 4.2 K at ambient pressure.

The sample magnetization under high pressure to 0.86 GPa was measured in pulse magnetic fields up to 42 T with a duration time of about 20 ms. In order to reduce eddy current in the material of a high-pressure clamp cell, a metal–ceramic hybrid clamp cylinder was used. A commercial BeCu alloy was employed as the material of the outer cylinder and locking screws. The inner cylinder was made of high-purity zirconia together with the piston and piston caps. Because the BeCu clamp cylinder is relatively thin, applied magnetic field is reduced only by 4% in the high-pressure cell and the phase shift between inner and outer magnetic fields is as low as 0.2%. The sample was set inside a small coaxial-type pick-up coil for measuring the magnetization. This assembly was inserted in a Teflon cell filled with a liquid pressure medium, Fomblin, in the clamp cylinder. The lead wires from the pick-up coil passed through the BeCu electrical feedthrough. The wires were glued by the mixture of epoxy resin, Stycast 1266, and alumina powder. High pressure was applied at room temperature and clamped in by the locking screw. The produced pressure at low temperatures was calibrated by means of the resistivity measurement of manganine wire, for which the pressure dependence of the resistivity is known with high accuracy.

In order to check the absolute value of the sample magnetization measured, the magnetization curve under high pressure was also measured with an extraction-type magnetometer in magnetic fields up to 9 T produced by a superconducting magnet. A nonmagnetic clamp cell made of high-purity TiCu alloy was used to produce high pressure up to 1 GPa. The details of the measurement system were described elsewhere [10].

The initial compressibility of the sample was measured with strain gauges attached on the surface of the sample in a Teflon cell filled with a liquid pressure medium in a piston–cylinder high-pressure apparatus. These measurements were performed at 293 and 77 K under high pressure to 0.5 GPa. A high-purity aluminium was used as a reference material [11].

## 3. Experimental results

The high-field magnetization curves of $Y(Co_{1-x}Al_x)_2$ were reported by several groups [6, 12]. The sharp metamagnetic transition from the paramagnetic to the ferromagnetic state was observed for $x \leqslant 0.09$. For this study, we selected three samples with different Al concentrations: $x = 0.06$, 0.075 and 0.09. Figure 1 shows the magnetization curves of these samples up to 42 T measured at 4.2 K for ambient pressure. The samples exhibit relatively sharp metamagnetic transitions between 20 and 40 T. The initial susceptibility increases and the critical field of the transition $B_c$ decreases with increasing $x$, in good agreement with the previous studies [6, 12].

The concentration dependence of the room-temperature lattice constant is shown in figure 2. The present data are consistent with the data of Yoshimura and Nakamura [3] measured in a wide range of Al content. For $0.05 \leqslant x \leqslant 0.20$, the lattice parameter increases linearly with $x$: $a(x) = a(0) + 0.665x$ (Å).

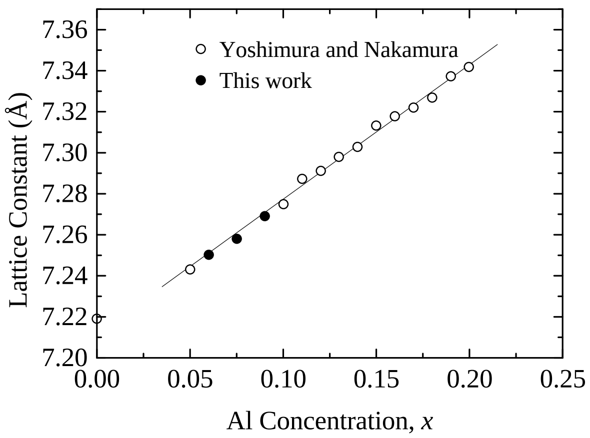


**Figure 2.** Concentration dependence of the lattice constant of $Y(Co_{1-x}Al_x)_2$. The data of Yoshimura and Nakamura [3] are also plotted for comparison.

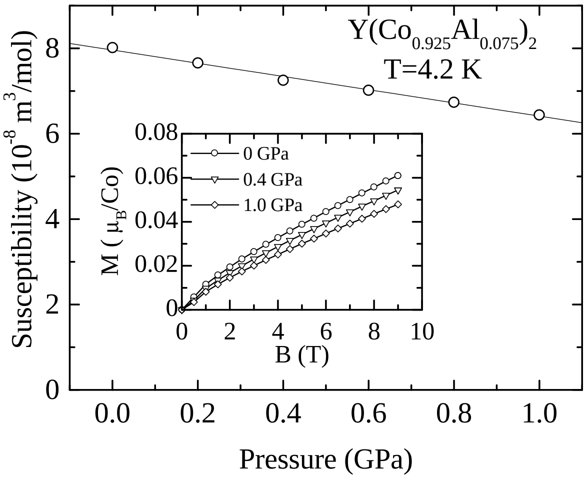


**Figure 3.** Pressure dependence of the susceptibility of $Y(Co_{1-x}Al_x)_2$ with $x = 0.075$ determined from the slope of the magnetization curves measured at 4.2 K in steady magnetic field up to 9 T. The inset shows the magnetization curves at several pressures.

The effect of applied pressure on the magnetization curve of $Y(Co_{0.925}Al_{0.075})_2$ was examined in detail in steady magnetic fields up to 9 T using the extraction-type magnetometer (figure 3). The paramagnetic susceptibility decreases with increasing pressure. For a Pauli paramagnet, the susceptibility is proportional to the density of states at the Fermi level: $\chi_p \propto N(E_f)$. Applied pressure reduces the value of $\chi_p$ and makes it more difficult to satisfy the Stoner condition for ferromagnetism: $IN(E_f) > 1$, where $I$ is the exchange parameter. Hence, high pressure is expected to increase the critical field of the metamagnetic transition.

The magnetization curves under pressure were measured for two samples with $x = 0.075$ and 0.09 in magnetic fields up to 42 T. Figure 4 shows the magnetization curves of $Y(Co_{0.925}Al_{0.075})_2$ for 4.2 K measured at different pressures. The metamagnetic transition was clearly observed for all pressures. The magnetic hysteresis indicates that the metamagnetic transition is of first-order and the type of the transition does not change with pressure. The average critical field of the transition $B_c$ increases linearly with pressure for both samples, as shown in figure 5. The values of $dB_c/dP$ are equal to 7.8 T $GPa^{-1}$ for $x = 0.075$ and 7.4 T $GPa^{-1}$ for $x = 0.09$. These values are slightly lower than 10 T $GPa^{-1}$ reported for $Lu(Co_{0.88}Ga_{0.12})_2$ [6].

In order to compare the change of $B_c$ due to the aluminium substitution with the change produced by external pressure, we need to know the volume compressibility $\kappa = d\ln V/dP$. The value $\kappa = 9.5\ 10^{-3}$/GPa was already reported for $YCo_2$ at room temperature [13]. We measured the compressibility of our samples $Y(Co_{1-x}Al_x)_2$ at room and liquid nitrogen temperatures. We obtained $\kappa = 9.3 \times 10^{-3}$ $GPa^{-1}$ at 293 K. When the temperature decreases, the compressibility slightly decreases to $8.7 \times 10^{-3}$ $GPa^{-1}$ at 77 K. Such a behaviour of the compressibility is typical of metals [14]. The obtained compressibility values are the same for the samples with different Al content within the experimental error.

## 4. Discussion

When Al (Ga) atoms are partially substituted for Co in $YCo_2$, this itinerant electron system is magnetically enhanced and approaches the onset of ferromagnetism. The increase of the

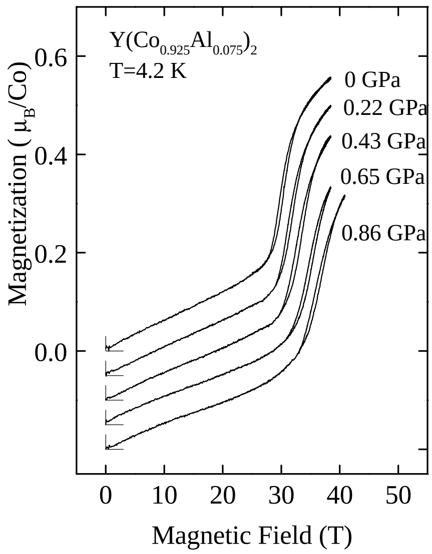


**Figure 4.** High-field magnetization curves of $Y(Co_{0.925}Al_{0.075})_2$ sample for 4.2 K at different pressures.

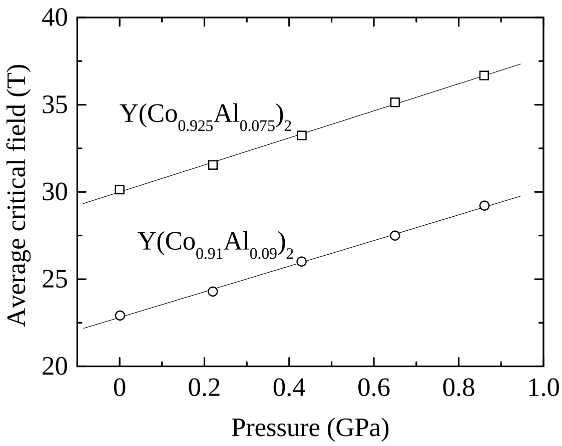


**Figure 5.** Pressure dependence of the average critical field of $Y(Co_{0.925}Al_{0.075})_2$ and $Y(Co_{0.91}Al_{0.09})_2$ at 4.2 K.

density of states at the Fermi level $N(E_f)$ with increasing Al (Ga) content is usually considered to be responsible for the enhancement.

Several interpretations were given to explain the increase of $N(E_f)$ in $Y(Co_{1-x}Al_x)_2$. Some authors considered the volume effect as the main contribution. When an aluminium atom with metallic radius $r_{Al} = 1.432$ Å is substituted for cobalt with $r_{Co} = 1.252$ Å [15], the lattice parameter increases. This leads to the narrowing of the d band and the increase of the density of states at the Fermi level [3, 16]. Armitage *et al* [17] found that the critical pressure to destroy the ferromagnetism of $Y(Co_{1-x}Al_x)_2$ with $0.12 \leqslant x \leqslant 0.20$ is $P_c \sim 0.9$ GPa. Since the lattice expansion of $Y(Co_{0.85}Al_{0.15})_2$ from $YCo_2$ is equivalent to a chemical pressure of −4 GPa, the appearance of ferromagnetism may be impossible in the $Y(Co_{1-x}Al_x)_2$ system with the same volume as $YCo_2$.

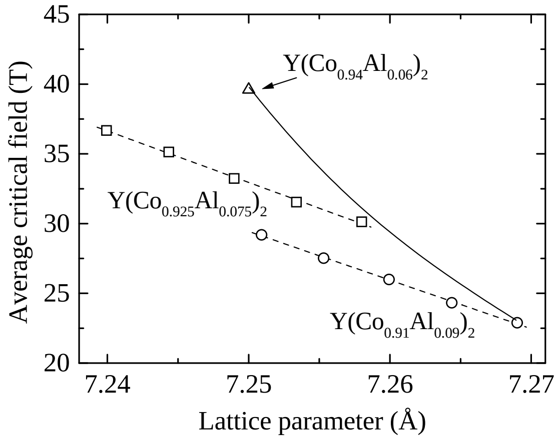


**Figure 6.** Average critical fields of $Y(Co_{1-x}Al_x)_2$ as functions of the lattice parameter. The circle, square and triangle indicate $x = 0.09$, $x = 0.075$ and $x = 0.06$, respectively. The dashed lines represent the data calculated from the values of $B_c$ determined under high pressure and the compressibility.

An alternative interpretation assumes that the increase of $N(E_f)$ comes mainly from the decrease of the 3d-electron concentration when Co ($3d^7$ configuration) is partially substituted by Al ($3d^0$ configuration) [12]. Since the Fermi level of $YCo_2$ lies in the region of $dN(E)/dE < 0$ [5, 18], the decrease of the d-electron concentration leads to the increase of $N(E_f)$ in the rigid-band model. In order to examine the effects due to the decrease in the electron concentration, a series of $(Y_{1-t}Lu_t)(Co_{1-x}Al_x)_2$ samples, which have nearly the same lattice parameter, were studied [19]. The critical field of the metamagnetic transition was found to decrease with increasing $x$ by 6.5–7 T per at.% of Al. This value is very close to those for $Y(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Al_x)_2$ where the lattice parameter changes with $x$. On the other hand, the value of $B_c$ for $Lu(Co_{1-x}Si_x)_2$ also decreases in spite of the decrease in the lattice parameter with increasing $x$ [20].

The above explanations are too simple because they do not take into account the change of the density of state curve near the Fermi level due to the p–d hybridization produced by the Al substitution. The change of $N(E_f)$ is more complicated in $Y(Co_{1-x}Al_x)_2$ than that expected from the rigid band model. Up to now there are no band calculations for $Y(Co_{1-x}Al_x)_2$ compounds substituted randomly. According to detailed band calculations of $YCo_2$ and ordered $Y_2Co_3Al$ [18], a sharp peak just below the Fermi level in the density of states of $YCo_2$ is considerably reduced and broadened in $Y_2Co_3Al$ because the hybridization between the Co 3d and Al 3p states is strong. The Fermi level in $Y_2Co_3Al$ is shifted to the lower energy side of the broad peak due to the decrease in the number of 3d electrons. Therefore, we expect that the Fermi level of $Y(Co_{1-x}Al_x)_2$ moves across the peak in the concentration region $0 < x < 0.25$. Since the peak value is relatively large, a ferromagnetic state is considered to appear at a certain concentration in this region [18].

The determination of both the pressure dependence of $B_c$ and the volume compressibility for $Y(Co_{1-x}Al_x)_2$ with different $x$ makes it possible to separate two contributions, which come from the changes of the electronic structure produced by the Al substitution (under the condition of constant volume) and by the volume increase. Figure 6 shows the average critical fields of the metamagnetic transition in $Y(Co_{1-x}Al_x)_2$ as functions of the lattice parameter. Two dashed lines represent the changes of the critical field produced by the application of high pressure in the samples with $x = 0.075$ and 0.09, whereas the solid line shows the change on Al substitution (at ambient pressure). It is evident that the change on Al substitution is about twice larger than that only produced by external pressure. This indicates that both contributions are nearly equal in the $Y(Co_{1-x}Al_x)_2$ system.

## 5. Conclusion

In order to clarify the origin of the change of the metamagnetic transition in $Y(Co_{1-x}Al_x)_2$, we have measured the high-field magnetization curve under high pressure and the volume compressibility of the $Y(Co_{1-x}Al_x)_2$ compounds with different $x$. The critical field of the metamagnetic transition increases linearly with pressure. The change of $B_c$ due to the Al substitution is found to be about twice that produced by external pressure. These experimental results indicate that both the increase in the bandwidth due to the expansion of the interatomic distance and the change of the electronic structure near the Fermi level due to the Al substitution contribute nearly equally to the change of $B_c$ in $Y(Co_{1-x}Al_x)_2$.

## Acknowledgments

The authors thank V S Gaviko and H Kageyama for x-ray diffraction measurements. The stay of NVM at ISSP was supported by the Ministry of Education, Science and Culture of Japan.

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