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Itinerant electron metamagnetism and spin fluctuations in nearly ferromagnetic metals $Y(Co_{1-x}Al_x)_2$

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Abstract. High-field magnetisation- and temperature-dependent susceptibility of Laves phase inter-metallic compounds $Y(Co_{1-x}Al_x)_2$ are investigated in the strongly exchange-enhanced paramagnetic region ($0 \leq x \leq 0.11$) in pulsed magnetic fields up to 100 T. In the whole concentration range, a sharp metamagnetic transition is observed in the low-temperature magnetisation process, while the low-field susceptibility exhibits a maximum at finite temperatures. The transition field B_c and the temperature of the susceptibility maximum T_{max} obtained as a function of x indicate a linear relationship, i.e. B_c/T_{max} = constant. The result clearly suggests that the susceptibility maximum of the nearly ferromagnetic metals is strongly correlated with itinerant electron metamagnetism.

1. Introduction

There is a class of materials called nearly ferromagnetic metals. This group of metals, consisting of transition metals and compounds such as Pd, Ni_3Ga, $TiBe_2$ and YCo_2, are characterised by strongly exchange-enhanced Pauli paramagnetism and large electronic specific heats and exhibit a number of unusual properties.

One of the interesting properties in the ground state ($T = 0$) of the nearly ferromagnetic metals is itinerant electron metamagnetism (IEM). This is a phenomenon that a paramagnetic metal undergoes a first-order phase transition to a ferromagnetic state at a high magnetic field with a discontinuous jump in the magnetisation. Although the first idea was presented a long time ago (Wohlfarth and Rhodes 1962), the experimental realisation has been difficult until now because the critical field of the transition is estimated to be very high, in the range of 10^2 T, for materials such as Pd (Jarlborg and Freeman 1981), YCo_2 (Cyrot *et al* 1979, Schwarz and Mohn 1984, Yamada *et al* 1987), $LuCo_2$ and $ScCo_2$ (Yamada *et al* 1987).

Recently, the pseudo-binary compounds $Y(Co_{1-x}Al_x)_2$ were found to exhibit increasing exchange-enhanced paramagnetism with increasing x and to become ferromagnetic at $x_c = 0.12$ (Yoshimura and Nakamura 1985, Yoshimura *et al* 1988). Subsequently, we examined the low-temperature magnetisation process of $Y(Co_{1-x}Al_x)_2$ up to 42 T and found a clear metamagnetic transition in the region $0.06 \leq x < 0.12$ (Sakakibara *et al*

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1986, 1987). Aleksandryan *et al* (1985) also observed similar results. We concluded from the concentration dependence of the transition that the observed metamagnetic transition is IEM inherent in YCo_2 and roughly estimated the critical field B_c of YCo_2 to be about 100 T. Similar but less sharp metamagnetic transitions were found in $Sc(Co_{1-x}Al_x)_2$ (Ishiyama and Endo 1987) and $Lu(Co_{1-x}Al_x)_2$ (Sakakibara *et al* 1987, Gabelko *et al* 1987, Endo *et al* 1988). In this paper, we have extended the measurement in $Y(Co_{1-x}Al_x)_2$ up to about 100 T using a single-turn coil system in order to explore the transition for lower x. We have succeeded in observing IEM in the whole paramagnetic concentration range $0 \leq x < 0.12$ and in particular directly confirmed that IEM does occur in YCo_2 at $B_c = 69$ T (Goto *et al* 1990), a somewhat lower field than the previously estimated values.

Another important property of the nearly ferromagnetic metals can be seen in the temperature variation of the magnetic susceptibility $\chi(T)$. As is well known, most of the nearly ferromagnetic metals exhibit a maximum of $\chi(T)$ at finite temperatures (Hoare and Matthews 1952, Acker *et al* 1981, Lemaire 1966, Givord and Lemaire 1971, Collings *et al* 1969). These facts motivated us to examine $\chi(T)$ in $Y(Co_{1-x}Al_x)_2$. We have observed that $\chi(T)$ exhibits a well defined maximum and the temperature of the susceptibility maximum T_{max} shows a systematical variation with x.

It is widely recognised that the finite-temperature properties of an itinerant electron magnet cannot be explained simply by the one-electron density of states but are strongly renormalised by interactions between particles (spin fluctuations). Several thermodynamical properties of weakly ferromagnetic metals can be explained successfully by the spin fluctuation theory (Moriya 1985). As for the nearly ferromagnetic metals, however, the origin of the singular behaviour of $\chi(T)$ is not fully understood, and there still seems to be controversy in the theoretical explanations (Barnea 1975, Misawa 1978, 1988, Yamada *et al* 1984, Lonzarich and Taillefer 1985). On the other hand, it seems to be generally accepted that IEM has its origin in the special band structures near the Fermi level and can be accounted for quantitatively by band calculations (Jarlborg and Freeman 1981, Yamada *et al* 1987). In this sense, the relation between the high-temperature property $\chi(T)$ and the low-temperature magnetisation processes $M(B)$ of the nearly ferromagnetic metals seems to be obscure.

In this paper, we present the first systematic experimental results on both IEM and the susceptibility maximum in nearly ferromagnetic metals. The present system, $Y(Co_{1-x}Al_x)_2$, is unique in the sense that it shows both clear susceptibility maxima and IEM in wide but accessible ranges by varying the Al concentration x. We demonstrate that there is a simple correlation between the $\chi(T)$ behaviour and $M(B)$, i.e. B_c/T_{max} is almost constant in this system. The result implies that both phenomena should be discussed on the same grounds.

2. Experimental details

The specimens were prepared by arc melting the metals, followed by annealing at 950 °C for 2 weeks. X-ray powder diffraction confirmed that they were essentially of single phase. No significant impurity reflection was observed in the diffraction patterns, from which we roughly estimated the amount of impurity phases possibly contained in our specimens to be less than 3%.

Magnetisation measurements below 42 T were done with a slow-pulse magnet (rise time 4 ms) in the temperature range 4.2–300 K. The magnetic susceptibility $\chi(T)$ was

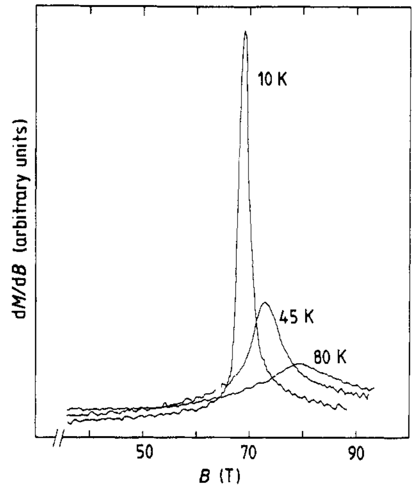

Figure 1. Differential susceptibility versus field for YCo_2 at $T = 10$ K, 45 K and 80 K. The sharp peak seen at 10 K indicates the occurrence of IEM at 69 T.

obtained as the slope (dM/dB) of the magnetisation curve at 8 T. We used this definition because even a small amount of ferromagnetic impurity phase significantly alters the initial slope of the magnetisation.

Very high magnetic fields up to 100 T (fast-pulse fields) were generated by a single-turn coil system (Nakao *et al* 1985). The magnetisation of the sample was directly measured by an induction method at temperatures down to 10 K. Details of the experimental set-up have already been published (Takeyama *et al* 1988). The inhomogeneity of the field produced by a single-turn coil of 16 mm diameter was about 2% within the dimensions of the pick-up coils (2 mm × 2 mm × 4 mm). In order to avoid Joule heating during a field rise time of 3 μs, samples were ground to fine powder of particle size less than 50 μm and cast into a rod shape of 1.5 mm diameter and 5 mm length with epoxy (Stycast 1266). The estimated temperature rise due to the heating was not more than a few kelvins at the lowest temperature. In order to subtract background noise from the signal, each measurement was done with two shots of the pulse field generation, with and without the sample in the pick-up coil. However, the field-generating coil was completely damaged and had to be replaced at each shot. This procedure resulted in poor reproducibility of the background, leading to an error of the order of 10% in the magnetisation values. On the other hand, B_c (position of the sharp peak in dM/dB) could be measured within an accuracy of ±2%.

3. Results

Figure 1 shows the field dependence of the differential susceptibility of YCo_2 taken in the increasing field scan at three temperatures $T = 10$ K, 45 K and 80 K. The sharp peak seen in dM/dB for $T = 10$ K is clearly of a first-order magnetic transition, indicating the existence of IEM in this compound (Goto *et al* 1990). No appreciable change was found in B_c at temperatures between 10 K and 20 K. Therefore, the result of 10 K represents the ground-state behaviour of the system. At higher temperatures, B_c shows a small positive shift, resulting in rapid broadening of the peak. The peak can still be seen but is very weak and almost vanishing at 80 K.

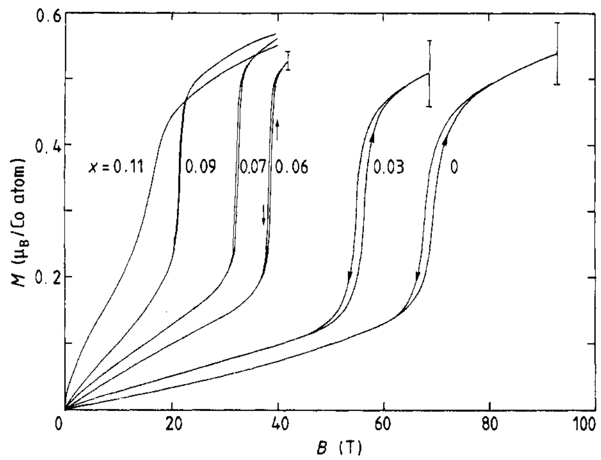

Figure 2. Low-temperature magnetisation of $Y(Co_{1-x}Al_x)_2$. The results for $x \geqslant 0.06$ are taken in the slow-pulse fields at 4.2 K (Sakakibara *et al* 1987) while those for $x = 0$ and 0.03 are measured in the fast-pulse fields at 10 K. Vertical bars indicate the estimated error in the magnetisation values.

Low-temperature magnetisation measurements were also made for the other samples and the representative magnetisation curves are displayed in figure 2, together with the previous results obtained in the slow-pulse fields up to 42 T. A clear metamagnetic transition can be seen in the whole range $0 \leqslant x \leqslant 0.11$. Since the transition shows a small hysteresis for $x \leqslant 0.09$, we define the low-temperature critical field B_{c0} by the average of the values in the increasing and decreasing field scans. It is noticed that the rate of the field scan at B_{c0} is typically 10^4 T s^{-1} and 3×10^7 T s^{-1} for the slow-pulse and the fast-pulse field measurements, respectively. Clearly, the higher rate of the field scan resulted in a larger hysteresis for $x = 0$ and 0.03. Nevertheless, it is remarkable that the hysteresis is at most 2 T for these samples, implying that the transition occurs in a time scale of 10^{-7} s. The apparent broadening of the transition for $x = 0$ and 0.03 is considered to be due to a degraded field homogeneity involved in the fast-pulse field measurements.

The critical field B_{c0} of $Y(Co_{1-x}Al_x)_2$ is summarised in figure 3. The open squares represent the present high-field results, which are fairly consistent with the previous low-field results given by full circles (Sakakibara *et al* 1986, 1987) and open triangles (Aleksandryan *et al* 1985). It is found that the slope of the B_{c0} versus x curve shows a slight decrease below $x = 0.06$. This is the reason why the previous experiments below 42 T (Sakakibara *et al* 1986) overestimate the critical field of YCo_2. The broken line in figure 3 is an extrapolation of the linear part of the B_{c0} versus x plot between $x = 0.05$ and 0.07. It is noted that the broken line crosses the horizontal axis at around $x_c = 0.12$, above which the spontaneous moment is known to appear (Yoshimura and Nakamura 1985). However, the observed B_{c0} does not follow this line and metamagnetism seems to coexist with ferromagnetism in the narrow region above x_c. The reason for this behaviour is not clear at present. Probably, it may be related to the fact that the ferromagnetic transition at x_c is second order in this system whereas metamagnetism in itself is first order.

The magnetisation jump ΔM of the transition is also plotted in figure 3, with the same symbols. Here, ΔM is estimated by integrating the peak in dM/dB. The magnitude

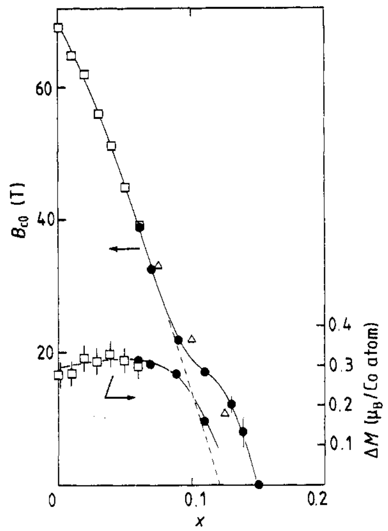

Figure 3. Critical field and magnetisation jump of the metamagnetic transition in $Y(Co_{1-x}Al_x)_2$ as a function of Al concentration x: open squares, data from the present high-field measurement performed at 10 K; full circles, results at 4.2 K obtained by Sakakibara *et al* (1986, 1987); open triangles, results at 4.2 K obtained by Aleksandryan *et al* (1985); broken line, extrapolation of the linear part of the B_c versus x plot.

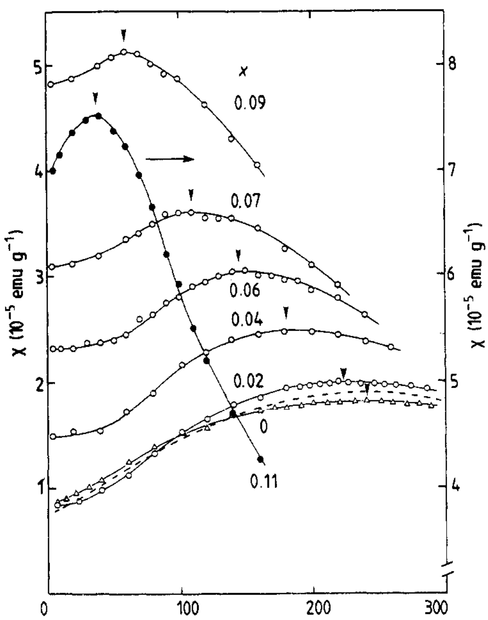

Figure 4. Temperature dependence of the magnetic susceptibility of $Y(Co_{1-x}Al_x)_2$: open circles, open triangles, present work in which $\chi(T)$ is defined as a differential susceptibility at 8 T; broken curve, $\chi(T)$ for YCo_2 obtained by Lemaire (1966); arrowheads, temperature T_{max} of the susceptibility maximum.

of ΔM is found to be essentially constant below $x = 0.1$ having the value 0.3 μ_B/Co atom, in accord with the band calculation of YCo_2 (Yamada *et al* 1987).

It is well known that $\chi(T)$ for YCo_2 increases with increasing temperature and becomes a maximum at around 240 K (Lemaire 1966), followed by a Curie–Weiss law at higher temperatures. Our $\chi(T)$ results for $Y(Co_{1-x}Al_x)_2$ are given in figure 4 by open circles and open triangles, together with the previous data for YCo_2 indicated by the broken curve. All of our samples show this typical behaviour of nearly ferromagnetic metals. Clearly, $\chi(0)$ is essentially an increasing function of x, implying that the exchange enhancement is growing with increasing x. The fact that $\chi(T)$ for $x = 0$ exceeds that for $x = 0.02$ at low temperatures may be an extrinsic effect. We noticed that the low-temperature susceptibility is sensitive to a ferromagnetic impurity phase and therefore somewhat sample dependent.

It should be emphasised that the temperature T_{max} of the susceptibility maximum shows a systematical shift against the Al content x. We show T_{max} versus x plot in figure 5, where T_{max} continuously decreases as the system approaches the critical concentration x_c indicated by the arrow. However, T_{max} does not disappear at x_c but tends to have a finite value similar to the B_{c0} behaviour in figure 3. This fact might suggest that IEM in this system does not have a strong relevance to the appearance of weak ferromagnetism at x_c. In order to discuss this point in detail, however, great care should be taken in the sample homogeneity. We therefore do not discuss the region near x_c in this paper.

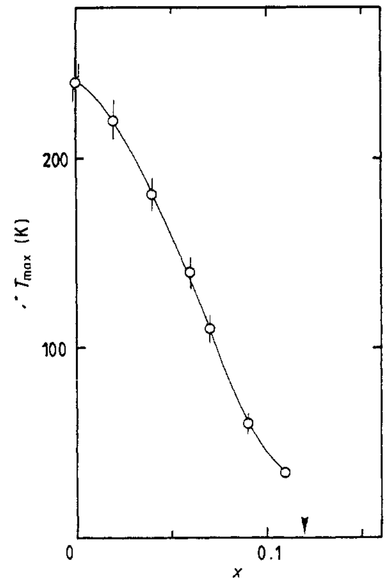

Figure 5. Temperature of the susceptibility maximum versus Al concentration for $Y(Co_{1-x}Al_x)_2$: arrowhead, ferromagnetic critical concentration x_c.

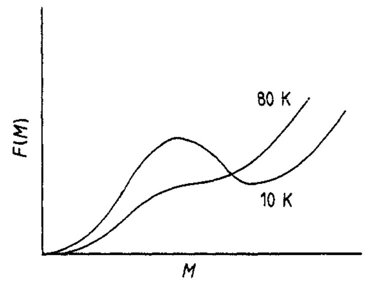

Figure 6. Sketch of the magnetic part of the free energy of YCo_2 at 10 and 80 K.

4. Discussion

In the band model of IEM, the magnetic free energy of the system is expanded in the form

$$F(M) = \tfrac{1}{2}aM^2 + \tfrac{1}{4}bM^4 + \ldots \qquad (1)$$

where the coefficients a, b, . . . are generally functions of temperature. Under the field B, the equilibrium state is realised by making the energy $G = F(M) - MB$ a minimum. For the nearly ferromagnetic metals, a is always positive since the system is paramagnetic at $B = 0$. If b is also positive, G has only a single minimum under B and therefore M increases monotonically with increasing B. In the case $b < 0$, however, G may take a double minimum at some field B_c with the presence of positive higher-order terms in (1). Then a sudden jump in M occurs at B_c, leading to IEM. A more detailed discussion has been given by Shimizu (1982). The coefficients at $T = 0$ in (1) can be in principle calculated from the density of states $\rho(\varepsilon)$ with a mean-field approximation for the electron–electron interactions. The origin of a negative b is considered to be a special band structure near ε_F, such as $\rho''(\varepsilon_F) > 0$. This condition is realised when ε_F lies just nearby a sharp peak in $\rho(\varepsilon)$, as is often the case with nearly ferromagnetic metals. A more quantitative assessment of IEM at $T = 0$ is given by the detailed calculation of $\rho(\varepsilon)$ with field (Yamada *et al* 1987), which shows reasonably good agreement with our experimental results for YCo_2 (Goto *et al* 1990).

The phenomenological explanation of IEM given above is considered to work at any temperature. Up to the present, however, there is no established theory that can deal with IEM quantitatively at finite temperatures, because of difficulties in evaluating the coefficients in (1) as a function of temperature. Here we consider what is implied by the results in figure 1. First, as the transition at 10 K is remarkably sharp, G should take a clear double-minimum structure at low temperatures. When the temperature is increased, we know from figure 4 that the coefficient a in (1) first decreases, since $\chi(T) \propto a^{-1}$. This leads to a slower increase in $F(M)$ in the small-M region. From the fact that B_c increases with increasing T, however, the finite-temperature $F(M)$ should exceed the low-temperature value in the larger-M region. This situation is illustrated schematically in figure 6. From these considerations, it follows that the absolute value of the coefficient b (<0) is a rapidly decreasing function of T. This explains the rapid broadening of the transition seen in figure 1 as well. The double-minimum structure of G at high temperatures becomes so weak that the transition may be easily smeared by thermal fluctuations.

The rapid decrease in the magnitude of b with increasing temperature is consistent with the precise measurement of the temperature-dependent non-linear magnetisation of YCo_2 (Bloch *et al* 1975, Schinkel 1978). It is also consistent with the previous models for the magnetic transitions in RCo_2 with R ≡ Gd, Tb, Dy, Ho and Er (Bloch *et al* 1975, Cyrot *et al* 1979), in which the magnetic ordering is either first order (R ≡ Dy, Ho and Er) or second order (R ≡ Gd and Tb) depending on the ordering temperature. However, we notice that the broadening of the metamagnetic transition is serious even at 80 K, contrary to these models where a sharp metamagnetic transition of the Co 3d band due to an exchange field from the rare-earth site is assumed to occur up to 200 K.

Microscopic evaluation of the temperature dependence of the free-energy coefficients in (1) is a difficult problem in itinerant electron magnetism. For example, the origin of the temperature variation of the coefficient a, which leads to the susceptibility maximum as shown in figure 4, is still an unsolved question in nearly ferromagnetic metals. At an early stage of the work, this problem was dealt with within the Stoner model in which the coefficients are derived from $\rho(\varepsilon)$ and the temperature dependence comes from the Fermi distribution function $f(\varepsilon)$ (Edwards and Wohlfarth 1968, Bloch *et al* 1975). By assuming special structures in $\rho(\varepsilon)$, this model could qualitatively reproduce the behaviour of $\chi(T)$. An advantage of this model is that it can treat both the ground-state magnetisation process and the high-temperature susceptibility within the same framework.

On the basis of the realistic band structure, however, it is now generally recognised that the Stoner model does not give a quantitative explanation of the thermodynamical properties of the itinerant electron systems. This can also be shown by the simple consideration as follows. The magnetic anomaly of YCo_2 occurs at about 70 T in the ground state. Considering that the Zeeman energy in this system is enhanced by a Stoner factor S of about 10 (Yoshimura *et al* 1988), the energy separation between zero-field ε_F and the sharp peak of $\rho(\varepsilon)$ responsible for IEM can be roughly estimated to be of order 10^3 K, in accordance with the band calculation (Yamada *et al* 1984). An attempt to exlain $\chi(T)$ by the smearing of $f(\varepsilon)$ then contradicts the fact that the $\chi(T)$ anomaly occurs at a much lower temperature, about 200 K. The rapid change in the b-term in $F(M)$ is also difficult to explain by this model, since the thermal smearing of $f(\varepsilon)$ should be small at 80 K compared with the relevant energy scale of order 10^3 K.

At present, the most successful theory that can quantitatively explain the finite-temperature properties of itinerant electron system is the spin fluctuation theory. According to the theory, the coefficient a is renormalised by the thermal spin fluctuations as (Moriya 1985, Lonzarich and Taillefer 1985)

$$a = a_0 + \tfrac{5}{3} b_0 \langle m^2 \rangle \tag{2}$$

where a_0 and b_0 are essentially the $T = 0$ values of the coefficients of M^2 and M^4 terms,

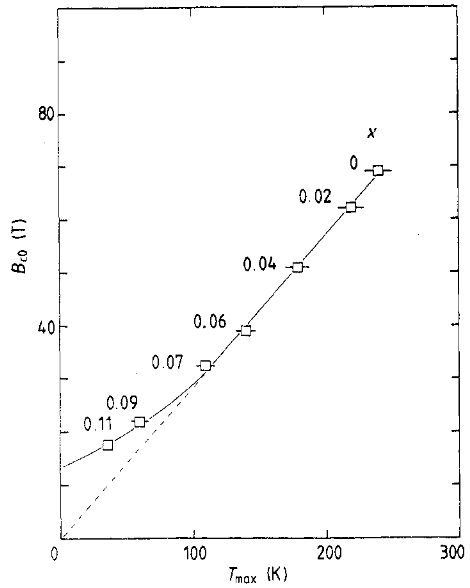

Figure 7. Critical field of IEM versus temperature of the susceptibility maximum for $Y(Co_{1-x}Al_x)_2$.

respectively, in the free energy. In principle they can be determined from the known band structures. The thermal spin fluctuation $\langle m^2\rangle$ is a strongly increasing function of temperature and thus leads to the strong temperature dependence of $\chi(T)$. This model explains the Curie–Weiss behaviour of weakly ferromagnetic metals above T_C with the usual positive b_0. If equation (2) is simply applied to the nearly ferromagnetic metals where b_0 is considered to be negative, then the initial increase in $\chi(T)$ may be explained. In order to explain the susceptibility maximum and the Curie–Weiss behaviour at higher temperatures, however, inclusion of the higher-order terms is necessary (Lonzarich and Taillefer 1985). Such an attempt was actually made in the discussion of temperature-induced ferromagnetism (Moriya 1986), where the renormalised free energy was given by retaining up to the sixth-order term in M. Although quantitative estimation was not given, the theory seems to reproduce the susceptibility maximum and the rapid temperature variation of the coefficient b (<0). Detailed analysis of experiments along this line is desired.

It should be noticed that there is a different approach to the problems from the Fermi liquid theory (Misawa 1978, 1988, Barnea 1975). According to the theory, the $T^2 \ln(T/T^*)$ term is generally introduced in the expression for $\chi(T)$, where T^* is a characteristic temperature, and the susceptibility maximum is predicted without assuming special band structures. The model also leads to the non-linear magnetisation at $T = 0$ through the term $H^2 \ln H$ (Misawa 1978). However, the possibility of IEM has not been discussed so far in the Fermi liquid theory except as a broad maximum in the field variation of the susceptibility. As discussed in the following, we believe that ground-state metamagnetism is relevant to the occurrence of a susceptibility maximum in nearly ferromagnetic metals.

We find that all the samples that show the susceptibility maximum are metamagnetic in the ground state. For those samples, the critical fields B_{c0} are plotted against the temperatures T_{max} of the susceptibility maxima in figure 7. Except for the region close to x_c, the result reveals a quite simple relation between B_{c0} and T_{max}, i.e. $B_{c0}/T_{max} \simeq$

constant. This fact implies that the susceptibility maximum is strongly correlated to IEM and possibly these two phenomena have the same origin. The obtained value of $B_{c0}/T_{max} \simeq 0.29\,T\,K^{-1}$ may not be a universal constant but may depend on the system. In fact, we performed preliminary experiments on Pd with a T_{max} of 90 K (Hoare and Matthews 1952) but could not observe any metamagnetic transition up to 120 T in accordance with the band calculation which predicts B_{c0} to be over 200 T (Jarlborg and Freeman 1981). The above correlation between the susceptibility maximum and metamagnetism is consistent with the experimental observations in other nearly ferromagnetic metals $TiBe_2$ and Ni_3Ga, where the former exhibits a susceptibility maximum in both the temperature and the field variation (Acker *et al* 1981) while the latter does not (Schinkel *et al* 1973). Similar indications of the correlation can be seen in some pseudo-binary compounds (Endo *et al* 1988, Sakakibara *et al* 1988).

Since the band calculations are considered to give a sound explanation of IEM in the ground state, the correlation of B_{c0} and T_{max} shown in figure 7 may naturally imply that the susceptibility maximum is also of band-structure origin. That is, the phenomenon may be explained within the spin fluctuation theory by properly taking into account the band parameters which lead to IEM (Takagi and Yasuoka 1985). It is noticed that in figure 2 the transitions for $x \leq 0.09$ occur at nearly the same moment value of about 0.15 μ_B Co atom with almost constant ΔM, although the initial susceptibility increases strongly with increasing x. This fact suggests that the magnetisation curve in this concentration range is roughly scaled by an effective field $B^* = SB$. Thus the dominant implicit parameter in figure 7 may be the exchange enhancement factor S, the structure of the density of state being essentially unchanged. Then, the systematic shift of T_{max} implies that the thermal effect is also strongly enhanced by S. We should notice, however, that an alternative interpretation of figure 7 might be possible. In the recent theory of spin fluctuations, it is argued that the role of the zero-point spin fluctuations is important and the free energy is strongly renormalised by interactions even in the ground state (Takahashi 1986). Under such a circumstance, an explanation of IEM might be different, such that the strong external field suppresses the fluctuations and stabilises the ferromagnetic state.

In conclusion, we have found the close relation between metamagnetism and the susceptibility maximum in $Y(Co_{1-x}Al_x)_2$ in the strongly exchange-enhanced paramagnetic region. The results are consistent with the model that both metamagnetism and the susceptibility maximum in nearly ferromagnetic metals are of special band-structure origin. In order to clarify this point further, more experimental and theoretical studies are desired.

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