Theory of itinerant-electron metamagnetism: II. The origin of the susceptibility maximum

1998 J. Phys.: Condens. Matter 10 5373

(http://iopscience.iop.org/0953-8984/10/24/014)

J. Phys.: Condens. Matter **10** (1998) 5373–5382. Printed in the UK PII: S0953-8984(98)89564-7

Theory of itinerant-electron metamagnetism: II. The origin of the susceptibility maximum

Yoshinori Takahashi and Tôru Sakai

Faculty of Science, Himeji Institute of Technology, 1475-2 Kanaji, Kamigori-cho, Ako-gun, 678-1297 Japan

Received 26 November 1997, in final form 17 March 1998

Abstract. We give a theoretical explanation for the phenomenon of the magnetic susceptibility showing a maximum at low temperature T_m for those compounds that show itinerant-electron metamagnetism. We also derive the linear relation between the metamagnetic transition field H_m and T_m observed experimentally for $Y(Co_{1-x}Al_x)_2$, paying particular attention to the effects of the zero-point quantum spin fluctuations and assuming the existence of almost energetically degenerate paramagnetic thermodynamic states.

1. Introduction

In spite of the long history of research in the field of itinerant-electron metamagnetism, there still remain important problems unresolved, a typical example of which is that of the occurrence of the susceptibility maximum observed at low temperature in the temperature dependence of the magnetic susceptibility (for $TiBe_2$ see Acker *et al* (1981, 1984), Gerhardt *et al* (1983); for YCo_2 see Yoshimura *et al* (1988a), Sakakibara *et al* (1990), for instance). The phenomenon has long been treated theoretically in terms of the Stoner–Wohlfarth theory based on the single-particle Hartree–Fock approximation (Wohlfarth and Rhodes 1962, Shimizu 1981, 1982). The idea, however, seems to contradict our current belief that the magnetic properties should be explained in terms of the collective low-lying spin-fluctuation excitations rather than single-particle excitations. In fact, a lot of observed magnetic properties of weak itinerant-electron ferromagnets and antiferromagnets are well described on the basis of the self-consistently renormalized (SCR) spin-fluctuation theory (Moriya 1985, Lonzarich and Taillefer 1985) at finite temperature. The ground state is not an exception. In our previous paper (Takahashi and Sakai 1995), we therefore proposed a new idea for describing the itinerant-electron metamagnetism in the ground state, paying particular attention to the role of the quantum zero-point spin fluctuations present even in the ground state. The purpose of this paper is to extend our previous study to finite temperature and to propose a mechanism giving rise to the susceptibility maximum as mentioned above.

The simplest idea based on the single-particle picture is that of assuming that the free energy $F(M)$ as a function of the uniform magnetization M can be expanded in the following form:

$$F(M) = F(0) + \frac{a}{2}M^2 + \frac{b}{4}M^4 + \frac{c}{6}M^6 + \cdots. \tag{1}$$

The coefficients are assumed to be obtained from the form of the density-of-states curve around the Fermi energy in the case where $M = 0$. If the fourth-order coefficient b

0953-8984/98/245373+10$19.50

becomes negative, the free energy will have a double-minimum structure, leading to a first-order metamagnetic transition in the presence of an external magnetic field H. The band-structure calculations show the existence of energetically almost degenerate non-magnetic and magnetic local minima (Schwarz and Mohn 1984, Hathaway and Cullen 1991), which are regarded as supporting the above picture. However, one has to assume adequate and sometimes artificial forms of the density-of-states curves in order to reproduce experimental results. Even in the SCR spin-fluctuation theory, such an expansion is assumed for the ground state and finite-temperature properties are discussed taking into account the effects of thermal spin fluctuations (Moriya 1986, Yamada 1993).

According to the above picture, it seems to be very difficult to interpret the magnetization process observed for the $Y(Co_{1-x}Al_x)_2$ system; i.e. the magnetization process for these compounds shows good linearity if plotted in the form of an Arrott plot (an M^2 versus H/M plot) after the metamagnetic transition. Because of the presence of the sixth-order term in (1), good linearity of the plot will not be expected from the above picture. On the other hand, our idea (Takahashi and Sakai 1995) was to assume the presence of two almost energetically degenerate exchange-enhanced thermodynamic states with different spin-fluctuation spectra. We then interpreted the metamagnetism as the transition from one exchange-enhanced paramagnetic state to another state, possibly magnetic, in the presence of an external magnetic field. The idea was motivated by the belief that the magnetic properties of the system have to be determined from the response of the spin-fluctuation excitation to the perturbation applied externally to the system. We have also emphasized the significant roles of the zero-point quantum spin fluctuations (Takahashi 1986, 1990, 1992, 1994, 1997) in dealing with the magnetization, which have been confirmed experimentally (Yoshimura *et al* 1988a, b, Shimizu *et al* 1990, Nakabayashi *et al* 1992).

Before proceeding to the next section, we again summarize the observed properties of itinerant-electron metamagnetism for later comparison between theory and experiment. Explaning these properties is the subject of our present study.

(i) The metamagnetism occurs around the critical region where the magnetism occurs or disappears when the external parameters, such as the volume of the system and the alloying concentration, are varied.

(ii) The transition is first order.

(iii) Accompanied by the metamagnetic transition, its magnetic susceptibility χ shows a maximum at around the temperature T_m with increasing temperature. The maximum structure of χ disappears in the presence of an external magnetic field higher than H_m, i.e. for $H > H_m$.

(iv) The temperature T_m is well correlated with the metamagnetic transition field H_m; for instance $H_m \propto T_m$ for $Y(Co_{1-x}Al_x)_2$ in the ground state (Sakakibara *et al* 1990).

(v) The temperature dependence of the metamagnetic transition field is given by

$$H_m = H_{m0} + \xi T^2 \tag{2}$$

at low temperature (Goto *et al* 1994).

The plan of the paper is as follows. On the basis of simple thermodynamic arguments, in the next section, we derive various behaviours occurring in relation to the metamagnetism as mentioned above. We make a brief comparison between the theory and the experiments on the $Y(Co_{1-x}Al_x)_2$ system in the following section. The final section is devoted to discussion.

2. Simple thermodynamic arguments

Following Takahashi and Sakai (1995), let us assume the presence of almost degenerate thermodynamic states whose free energies are denoted by $F_1(M,T)$ and $F_2(M,T)$, by taking into account their explicit temperature dependence. We assume that both of these states are paramagnetic, though one of the states, described by $F_2(M,T)$, is almost magnetic in the sense that it would become ferromagnetic classically if there were no spin-fluctuation effects. The state is therefore called pseudo-magnetic hereafter. It is important to recognize that the spin-fluctuation mechanism as well as the significant role of the quantum spin fluctuations underlie the above assumption, which is also supported by experiments. The spin-fluctuation spectrum, observed by means of NMR relaxation measurements (Yoshimura *et al* 1988b) on the itinerant metamagnetic system $Y(Co_{1-x}Al_x)_2$, for instance, shows sudden changes as the critical Al concentration for the appearance of ferromagnetism is crossed.

At low temperature, the above free energies are expanded in terms of the temperature T and the uniform magnetization M as follows:

$$\begin{aligned} F_1(M,T) &= F_1(0,0) + \frac{1}{2\chi_1}M^2 - \frac{1}{2}\gamma_1 T^2 + \cdots \\ F_2(M,T) &= F_2(0,0) + \frac{1}{2\chi_2}M^2 - \frac{1}{2}\gamma_2 T^2 + \cdots \end{aligned} \tag{3}$$

where the γs stand for the linear coefficients of the electronic specific heat, with enhancement due to the effect of spin fluctuations. In order for the metamagnetic transition to take place, the above two thermodynamic states have to be almost degenerate, or, to be precise, the free energy $F_1(0,0)$ has to be slightly lower than $F_2(0,0)$:

$$\Delta F(0,0) = F_2(0,0) - F_1(0,0) > 0.$$

The first-order metamagnetic transition takes place from one of the paramagnetic states to a pseudo-magnetic state in the presence of the external magnetic field, if both of the conditions

$$\gamma_2 > \gamma_1 \qquad \chi_2 > \chi_1 \tag{4}$$

are satisfied. It is therefore important to check the validity of the above conditions in actual cases.

In the case of the $Y(Co_{1-x}Al_x)_2$ system, the specific heat measurements (Wada *et al* 1989, 1990) suggest that the first condition $\gamma_2 > \gamma_1$ is satisfied. The coefficients γ for the ferromagnetic samples show larger values than those for the paramagnetic samples. It is reasonable to associate the γ-values of our pseudo-magnetic states with those of the ferromagnetic samples. The inequality of the γ-values is also consistent with our natural view that the entropy in the magnetic or pseudo-magnetic state grows faster than that in the non-magnetic state. The second condition is also satisfied in general. According to our earlier discussion (Takahashi and Sakai 1995), the value of χ_1 can be estimated as the observed value of the magnetic susceptibility at $T = 0$ K, because the system will be in the paramagnetic state in the ground state. On the other hand, the temperature dependence of the susceptibility at higher temperature mainly reflects the free energy of the pseudo-magnetic state. The value of χ_2 is estimated by smooth extrapolation of the susceptibility values at high temperature above the susceptibility maximum to the $T = 0$ limit. The presence of the susceptibility maximum therefore automatically ensures the inequality $\chi_2 > \chi_1$.

Let us now discuss the simple energetics by comparing the free energies of $\Phi_1(H,T)$ and $\Phi_2(H,T)$ in the presence of the external field H, defined by

$$\Phi_1(H,T) = F_1(M,T) - HM \qquad \Phi_2(H,T) = F_2(M,T) - HM. \tag{5}$$

We first introduce the metamagnetic transition temperature T_m and the transition field H_m, which characterize the energetic crossover between the paramagnetic and pseudo-magnetic states. After minimization of $\Phi_1(H,T)$ and $\Phi_2(H,T)$ with respect to M, the free-energy minima in the presence of the magnetic field H are, respectively, given by

$$\begin{aligned}\Phi_1(H,T) &= F_1(0,0) - \frac{1}{2}\chi_1 H^2 - \frac{1}{2}\gamma_1 T^2 + \cdots \\ \Phi_2(H,T) &= F_2(0,0) - \frac{1}{2}\chi_2 H^2 - \frac{1}{2}\gamma_2 T^2 + \cdots .\end{aligned} \tag{6}$$

From the condition that the free energies coincide with each other at the metamagnetic transition field in the ground state, H_m is defined by

$$H_m^2 = \frac{2\,\Delta F(0,0)}{\Delta\chi} \qquad (\Delta\chi = \chi_2 - \chi_1). \tag{7}$$

On the other hand, in the absence of the external field we define T_m on the basis of the same condition by

$$T_m^2 = \frac{2\,\Delta F(0,0)}{\Delta\gamma} \qquad (\Delta\gamma = \gamma_2 - \gamma_1). \tag{8}$$

From (7) and (8), the following relation between T_m and H_m is derived:

$$H_m = (\Delta\gamma/\Delta\chi)^{1/2} T_m. \tag{9}$$

The physical meaning of the parameters H_m and T_m introduced above is clarified by the following discussion. First, the condition $\Phi_1(H_m,0) = \Phi_2(H_m,0)$ is equivalent to the Maxwell rule for the first-order transition employed by Takahashi and Sakai (1995), i.e. at $H = H_m$ the tangents to the free energies $F_1(M,T)$ at $M = M_1$ and $F_2(M,T)$ at $M = M_2$ coincide with each other:

$$\begin{aligned}&\frac{\partial F_1(M_1,0)}{\partial M} = \frac{\partial F_2(M_2,0)}{\partial M} = H_m \\ &\frac{F_2(M_2,0) - F_1(M_1,0)}{M_2 - M_1} = \frac{\Delta F(0,0) + M_2^2/2\chi_2 - M_1^2/2\chi_1}{M_2 - M_1} = H_m.\end{aligned} \tag{10}$$

Therefore H_m is simply the metamagnetic transition field. It is also easy to see that at around the temperature T_m the susceptibility shows a maximum in its temperature dependence. Let us imagine an idealized situation in which, although the sample is very clean, because of some slight relaxation processes it always relaxes to its lowest energy state as soon as possible. Then the temperature dependence of the magnetic susceptibility of the sample $\chi(T)$ is given by

$$\chi(T) = \begin{cases} \chi_1(T) & \text{for } T < T_m \\ \chi_2(T) & \text{for } T \geqslant T_m \end{cases} \tag{11}$$

where $\chi_1(T)$ and $\chi_2(T)$ are the magnetic susceptibilities for the paramagnetic and the pseudo-magnetic states, respectively. Because the susceptibility of the pseudo-magnetic state is greater in magnitude, the magnetic susceptibility shows a steep rise at the temperature T_m. In actual experimental situations, the transition will occur gradually for various reasons, and the sudden increase of the susceptibility will be broadened to show a maximum at around T_m. This is why we associate T_m with the temperature of the susceptibility maximum. The observed linear relation between H_m and T_m is thus derived as the natural consequence of our equation (9).

The above simple explanation based on the energetic comparison is also consistent with the external field dependence of the magnetic susceptibility reported for $TiBe_2$ (Monod *et al*

1980). Though the magnetic susceptibility shows a maximum at low temperature in the presence of the weak field, it only decreases monotonically with temperature, showing no maximum structure for the field strength $H > H_m$. The reason for this is that, for such field strengths, the pseudo-magnetic state is always more stable than the paramagnetic one.

Before concluding this section, we briefly deal with the temperature dependence of the metamagnetic transition field H_m at low temperature. At finite temperature, by simply extending the above discussions, equation (7) is simply replaced by the following form:

$$H_m^2 = \frac{2\,\Delta F(0,T)}{\Delta\chi(T)} \simeq \frac{2\,\Delta F(0,0) - \Delta\gamma\,T^2 + \cdots}{\Delta\chi(0) - \Delta A\,T^2 + \cdots} \tag{12}$$

where we assume that the temperature dependence of the magnetic susceptibility difference $\Delta\chi$ is proportional to T^2 at low temperature. By expanding the above expression, equation (12), in terms of T^2, we obtain

$$\begin{aligned} H_m^2 &= \frac{1}{\Delta\chi(0)}\left(2\,\Delta F(0,0) - \Delta\gamma\,T^2 + \frac{2\,\Delta F(0,0)}{\Delta\chi(0)}\,\Delta A\,T^2\right) + \cdots \\ &= H_{m0}^2\left[1 + \left(\frac{H_{m0}^2\,\Delta A}{\Delta\gamma} - 1\right)\left(\frac{T}{T_m}\right)^2\right] + \cdots \end{aligned} \tag{13}$$

where H_{m0}^2 stands for the metamagnetic transition field in the ground state given by (7). The temperature dependence of H_m observed for YCo_2 can be understood if the condition

$$H_{m0}^2\,\Delta A > \Delta\gamma \tag{14}$$

is satisfied. The above result can also be derived with the use of the Clausius–Clapeyron relation. The condition (10) is expressed in the following form for finite temperature:

$$F_2(M_2,T) - F_1(M_1,T) = H_m\,\Delta M \qquad \Delta M = M_2 - M_1. \tag{15}$$

By differentiating both sides of the above equation with respect to T under constant magnetizations M_2 and M_1, we obtain the Clausius–Clapeyron relation

$$-\Delta S(T) = \frac{\mathrm{d}H_m}{\mathrm{d}T}\,\Delta M \tag{16}$$

where $\Delta S(T)$ is the entropy difference, given by

$$\begin{aligned} \Delta S(T) &= -\left(\frac{\partial F_2(M_2,T)}{\partial T} - \frac{\partial F_1(M_1,T)}{\partial T}\right) \\ &= \Delta\gamma\,T + \frac{M_2^2}{2\chi_2^2(T)}\frac{\mathrm{d}\chi_2(T)}{\mathrm{d}T} - \frac{M_1^2}{2\chi_1^2(T)}\frac{\mathrm{d}\chi_1(T)}{\mathrm{d}T} \\ &= \Delta\gamma\,T + \frac{H_m^2}{2}\frac{\mathrm{d}\,\Delta\chi(T)}{\mathrm{d}T} = -(H_m^2\,\Delta A - \Delta\gamma)T = \Delta\gamma_{\mathrm{eff}}\,T. \end{aligned} \tag{17}$$

In deriving the last line of (17) from the second, we use the following relation valid at the metamagnetic transition:

$$M_2/\chi_2 = M_1/\chi_1 = H_m.$$

With the use of (16) and (17) we finally obtain

$$\frac{\mathrm{d}H_m}{\mathrm{d}T} = \frac{T}{\Delta M}(H_m^2\,\Delta A - \Delta\gamma) = \frac{T}{\Delta\chi\,H_m}(H_m^2\,\Delta A - \Delta\gamma).$$

If we neglect the difference between H_{m0} and H_m at low temperature, the observed positive $\mathrm{d}H_m/\mathrm{d}T$ is also found to be consistent with the condition (14). The above result also indicates that we have to be careful when drawing any conclusions as regards the sign of

$\Delta\gamma$ from the field dependence of the electronic linear specific coefficient. The observed negative $\Delta\gamma_{\rm eff}$ (Goto *et al* 1994) is consistent with the positive deviation of H_m at finite temperature. This, however, does not necessarily contradict our assumption of a positive value of $\Delta\gamma$, since the observed T-linear specific heat coefficient consists of the sum of two contributions.

3. Comparison with experiments

The purpose of this section is to check the validity of our idea by comparing the theoretical consequences with actual observations and by estimating the parameters appearing in the theory, to see whether these have reasonable values. Before going into detail, let us first discuss the temperature dependence of the magnetic susceptibility in real experimental situations. It is in general quite difficult to imagine perfect-crystals samples. Even in very carefully prepared single crystals, there are usually various subtle inhomogeneities, such as defects and local strains. All of these will become sources of the distribution of the value of $\Delta F(0,0)$ throughout the whole of the samples. Only the value averaged over these distributions is meaningful and this is what is measured experimentally.

In the following, we assume that the values of $\Delta F(0,0)$ will distribute around a mean value $\Delta\bar{F}(0,0)$, i.e. $\Delta F(0,0) = \Delta\bar{F}(0,0) + u$. The statistical variable u stands for the deviation from the mean value. The free-energy difference $\Delta F(0,T)$ is then regarded as a statistical variable, i.e.

$$\begin{aligned}\Delta F(0,T) &= \Delta\bar{F} + u - \frac{1}{2}\Delta\gamma\, T^2 + \cdots \\ &= u - \frac{1}{2}\Delta\gamma\,(T^2 - T_m^2) + \cdots \\ &\simeq u - \Delta\gamma\, T_m(T - T_m)\end{aligned} \tag{18}$$

where, with the use of the average value of $\Delta\bar{F}$, we redefined T_m via

$$T_m^2 = 2\,\Delta\bar{F}/\Delta\gamma. \tag{19}$$

According to (11), we obtain the following condition determining whether the system in the statistical ensemble is either in the pseudo-magnetic or the paramagnetic state:

$$\begin{aligned} &u > \Delta\gamma\, T_m(T - T_m) \qquad \text{for paramagnetic states} \\ &u < \Delta\gamma\, T_m(T - T_m) \qquad \text{for pseudo-magnetic states.}\end{aligned} \tag{20}$$

If we introduce a distribution function $D(u)$ for the deviation u, the probabilities $p_1(T)$ and $p_2(T)$ for finding the system either in the paramagnetic state or in the pseudo-magnetic state at given temperature T are, respectively, given by

$$p_1(T) = \int_{\Delta\gamma\, T_m(T-T_m)}^{\infty} D(u)\ \mathrm{d}u \qquad p_2(T) = \int_{-\infty}^{\Delta\gamma\, T_m(T-T_m)} D(u)\ \mathrm{d}u. \tag{21}$$

The observed magnetic susceptibility, averaged over the ensemble of the samples, is given by

$$\chi(T) = p_1(T)\chi_1(T) + p_2(T)\chi_2(T) \tag{22}$$

where $\chi_1(T)$ and $\chi_2(T)$ represent the susceptibilities for the paramagnetic and the pseudo-magnetic states, respectively.

At low temperature, below T_m, $\chi(T)$ is mainly given by $\chi_1(T)$. As we increase the temperature it gradually changes until it is given by $\chi_2(T)$ at around T_m. In order to see

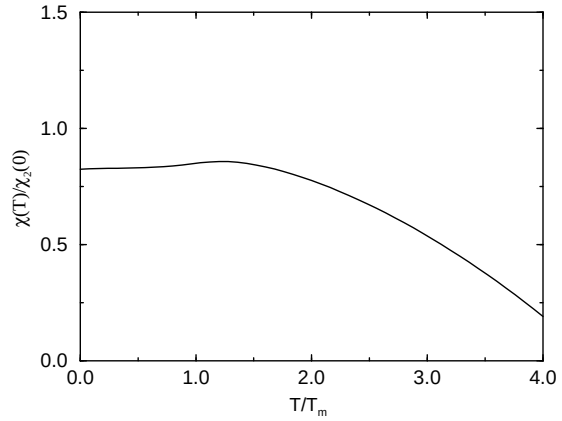

Figure 1. The temperature dependence of the magnetic susceptibility evaluated with the use of equation (24) for $\delta/\Delta\gamma\, T_m^2 = 0.4$ and $AT_m^2/\chi_2(0) = 0.05$.

whether the observed behaviour of the magnetic susceptibility is reproduced by (22), we numerically calculated the temperature dependence of $\chi(T)$ with the use of the Lorentzian form of the distribution function for $D(u)$, given by

$$D(u) = \frac{1}{\pi}\frac{\delta}{u^2+\delta^2}$$

where δ represents a measure of the standard deviation of $D(u)$. The probabilities in (21) are in this case given by

$$\begin{aligned} p_1(T) &= \frac{1}{2} - \frac{1}{\pi}\tan^{-1}\left(\frac{\Delta\gamma\, T_m(T-T_m)}{\delta}\right) \\ p_2(T) &= 1 - p_1(T) = \frac{1}{2} + \frac{1}{\pi}\tan^{-1}\left(\frac{\Delta\gamma\, T_m(T-T_m)}{\delta}\right). \end{aligned} \tag{23}$$

For illustration, we show in figure 1 the temperature dependence of $\chi(T)$ evaluated by using (22). In this calculation we have simply assumed the following temperature dependences of $\chi_1(T)$ and $\chi_2(T)$:

$$\chi_1(T) = \chi_1 - A_1T^2 \qquad \chi_2(T) = \chi_2 - A_2T^2 \tag{24}$$

which is justified at low temperature. We have also assumed that $\Delta A = 0$ for simplicity. It is easy to see that the temperature scale for the transition from the paramagnetic to the pseudo-magnetic state is given by $\delta/\Delta\gamma\, T_m$.

The positive T^2-dependence of the transition field H_m, observed for YCo_2 for instance, suggests a positive value of ΔA when $\Delta\gamma$ is positive. Then $\Delta\chi(T)$ would appear to vanish at some temperature if (24) holds over the wide temperature range, because the following relation holds for $\Delta\chi(T)$:

$$\Delta\chi(T) = \Delta\chi - \Delta A\, T^2 = T_m^2(\Delta\gamma/H_m^2 - \Delta A) + \Delta A\,(T_m^2 - T^2). \tag{25}$$

In the preceding section we saw that the first term is negative for YCo_2 from the condition (14), indicating that $\Delta\chi(T)$ changes sign at some temperature T below T_m. If this was the case, it would contradict our scenario for the susceptibility maximum as mentioned above. We think, however, that this will not happen and that $\Delta\chi(T)$ always remains positive, because the magnetic susceptibilities soon obey a Curie–Weiss-like temperature dependence with nearly the same Curie constants on increasing temperature. Note that the

T^2-dependence with a positive coefficient is only observed for YCo_2 at low temperature, far below T_m.

For the purpose of making comparisons between the theory and experiments, the intermetallic compound $Y(Co_{1-x}Al_x)_2$ is the best system to consider. A lot of experimental studies have been done on this system, using various experimental techniques. With the use of (7) and (8) we can estimate the free-energy difference ΔF from the observed values of $\Delta\chi$ and H_m, or $\Delta\gamma$ and T_m. We give in table 1 values of ΔF and $\Delta\gamma$ obtained from (9) by using the experimentally estimated values of $\Delta\chi$ and H_m. We see that the free-energy difference decreases monotonically on increasing the Al concentration towards the critical concentration for the appearance of the weak ferromagnetism, while $\Delta\chi$ and $\Delta\gamma$ remain almost constant.

Table 1. The metamagnetic behaviour and the susceptibility maximum of $Y(Co_{1-x}Al_x)_2$, after Sakakibara *et al* (1990).

x	$\Delta\chi$ (10^{-3} emu mol^{-1})	H_m (T)	ΔF (K)	T_m (K)	$\Delta\gamma$ (mJ mol^{-1} K^{-2})
0.00	1.2	69	3.3	240	1.0
0.02	1.3	62	3.0	220	1.0
0.04	1.2	51	1.9	180	1.1
0.06	1.0	39	0.94	140	0.8
0.07	0.83	32	0.53	110	0.7
0.09	0.56	22	0.16	60	0.7
0.11	1.2	17	0.21	35	2.9

Though there are no direct measurements of values of $\Delta\gamma$ available at present, we can get rough estimates of these values from the Al concentration dependence of the observed γ-values (Wada *et al* 1989, 1990). From the γ-value reported for the ferromagnetic sample with the highest T_c, we estimate $\Delta\gamma \simeq 10$ mJ K^{-2} mol^{-1}, which will serve as an upper bound for the actual values. The values of $\Delta\gamma$ of several mJ mol^{-1} K^{-2} listed in table 1 are comparable with this value. The analysis suggests that our equation (9) agrees well with experiments, including this factor, considering our crude estimates of the values of $\Delta\chi$ and $\Delta\gamma$.

With the use of the value of $\Delta\gamma$ estimated above, we can also get a rough picture of the temperature width of the susceptibility maximum, given by $\delta/\Delta\gamma\, T_m$. If we assume that $\delta \simeq 0.5$ K per magnetic ion, this gives about 10 K for the width of the magnetic susceptibility peak for $x = 0.11$, consistent with experiments. It is quite reasonable to assume that the free-energy difference is as small as this for the samples. This also suggests that we are dealing with realistic situations.

4. Discussion

In the present paper, we have proposed a mechanism for the occurrence of the susceptibility maximum observed universally, associated with itinerant-electron metamagnetism. According to our picture, its origin is closely related to the first-order phase transition. It is nothing to do with the fine structure of the density-of-states curve around the Fermi energy, though that may be responsible for the occurrence of the double-minimum structure for the classical Hartree–Fock contribution to the free energy. In our view, both the temperature dependence of the magnetic susceptibility and the magnetization process have to be derived as effects of the spin fluctuations.

On the basis of the spin-fluctuation mechanism, however, we can only predict a quite normal temperature dependence of the magnetic susceptibility χ, i.e. a monotonic decrease with temperature proportional to T^2 at low temperature. No peak structure in its temperature dependence occurs in general. The free energy is also expanded very well up to the fourth-order term in M. No higher-order terms need to appear. A possible way to produce the observed temperature dependence of χ that is consistent with the above restrictions is to assume the existence of almost degenerate nearly ferromagnetic states, the existence of which is explained by our spin-fluctuation mechanism. The transition between these states triggered by the thermal activations or by applying an external magnetic field gives rise to the metamagnetic transition. Note, however, that the linearity of the Arrott plot of the magnetization process before and after the transition is a natural consequence of our mechanism. Otherwise, we would need unrealistic higher-order expansion terms in order to reproduce the behaviour.

According to our mechanism, the susceptibility maximum appears as an extrinsic effect associated with the first-order transition. For MnSi it has recently be shown from NMR measurements (Thessieu *et al* 1998) that the magnetic and the paramagnetic states coexist around the critical region in which the magnetism disappears under pressure. For CoS_2 with the pyrite crystal structure, it was also reported from NMR measurements that the local environmental effect around magnetic ions is important and that non-magnetic and magnetic states coexist around the metamagnetic transition. Moreover, recent pressure dependence measurements made on MnSi (Thessieu 1997) show that its fourth-order coefficient b remains finite and does not seem to vanish as the critical pressure at which the magnetism disappears is approached. It would be very difficult to interpret this behaviour on the basis of a the model free energy of the form (1) and assuming a negative value of b. These experimental observations are, on the other hand, quite consistent with our idea.

With the use of the Maxwell relation for the thermodynamics, we can show that the magnetic field dependence of the specific heat is related to the curvature of the temperature dependence of the magnetic susceptibility. The appearance of the susceptibility maximum has been analysed in terms of the relation associated with the field dependence of the electronic specific heat measurements (Béal-Monod 1981, Shioda *et al* 1988). Our present arguments, however, indicate that we have to be very careful when applying the relation to the itinerant-electron metamagnetic systems. If the susceptibility maximum at low temperature is not intrinsic but is derived from extrinsic effects, as suggested here, such a relation need not hold. In checking our picture, it is of interest to see whether or not the Maxwell relation holds at low temperature and in the weak-field region. The dependence of the sample preparation on the temperature width of the susceptibility may also be valuable. Magnetization measurements at still higher magnetic fields after the metamagnetic transition has taken place, to check the linearity of the Arrott plot, are, of course, crucial for testing our mechanism.

In the present treatment, we have assumed that the free energy is well represented by the expansion up to the second-order terms in M in order to clarify the central point of our treatment in a simple manner. Thus its use will be very suitable for cases in which the magnetization always remains very small in magnitude. In some cases, MnSi under pressure (Thessieu *et al* 1995) and CoS_2 with Se substitution (Adachi *et al* 1979) for instance, a sizable moment is induced during the transition. In such cases, slight modifications may be needed, such as taking into account the effects of the higher-order M^4-term as was done by Takahashi and Sakai (1995).

Note also that the reason that we feel that we can employ the term '*pseudo-magnetic* state' lies in the following observation. The higher-magnetic-field side of the Arrott plot of

the magnetization process for $Y(Co_{1-x}Al_x)_2$ extrapolates linearly to a finite M^2-value in the $H/M = 0$ limit (Takahashi and Sakai 1995), suggesting the presence of a magnetic state after the metamagnetic transition. This is why we associated one of the degenerate states with a *magnetic* state in our previous treatment. The present paper thus takes the opposite view, and regards both of the states as *paramagnetic*, with the reservation that one of them is very close to being a *magnetic* state. Whichever is closer to reality, our central point of the assumption of the coexistence of degenerate thermodynamic states remains unchanged.

Acknowledgment

The authors would like to thank Dr C Thessieu for valuable comments and discussions.

References

Acker F, Fisk Z, Smith J L and Huang C Y 1981 *J. Magn. Magn. Mater.* **22** 250
Acker F, Huguenin R, Pelizzone M and Smith J L 1984 *J. Magn. Magn. Mater.* **46** 11
Adachi K, Matsui M, Ohshima Y, Mollymoto M, Motokawa M and Date M 1979 *J. Phys. Soc. Japan* **47** 675
Béal-Monod M T 1981 *Phys. Rev.* B **24** 261
Gerhardt W, Schilling J S, Olijnyk H and Smith J L 1983 *Phys. Rev.* B **28** 5814
Goto T, Katori H A, Sakakibara T, Mitamura H, Fukamichi K and Murata K 1994 *J. Appl. Phys.* **76** 6682
Hathaway K and Cullen J 1991 *J. Phys.: Condens. Matter* **3** 8911
Lonzarich G G and Taillefer 1985 *J. Phys. C: Solid State Phys.* **18** 4339
Monod P, Felner I, Chouteau and Shaltiel D 1980 *J. Physique* **41** L511
Moriya T 1985 *Spin Fluctuations in Itinerant Electron Magnetism* (Berlin: Springer)
——1986 *J. Phys. Soc. Japan* **55** 357
Nakabayashi R, Tazuke Y and Maruyama S 1992 *J. Phys. Soc. Japan* **61** 774
Sakakibara T, Goto T, Yoshimura K and Fukamichi K 1990 *J. Phys.: Condens. Matter* **2** 3381
Schwarz K and Mohn P 1984 *J. Phys. F: Met. Phys.* **14** L129
Shimizu M 1981 *Rep. Prog. Phys.* **44** 145
——1982 *J. Physique* **43** 155
Shimizu K, Maruyama H, Yamazaki H and Watanabe H 1990 *J. Phys. Soc. Japan* **59** 305
Shioda S, Takahashi Y and Moriya T 1988 *J. Phys. Soc. Japan* **57** 3146
Takahashi Y 1986 *J. Phys. Soc. Japan* **55** 3553
——1990 *J. Phys.: Condens. Matter* **2** 8405
——1992 *J. Phys.: Condens. Matter* **4** 3611
——1994 *J. Phys.: Condens. Matter* **6** 7063
——1997 *J. Phys.: Condens. Matter* **9** 2593
Takahashi Y and Sakai T 1995 *J. Phys.: Condens. Matter* **7** 6279
Thessieu C 1997 private communication
Thessieu C, Flouquet J, Lapertot G, Stepanov A N and Jaccard D 1995 *Solid State Commun.* **95** 707
Thessieu C, Ishida K, Kitaoka Y, Asayama K and Lapertot G 1998 *J. Magn. Magn. Mater.* **177–181** 609
Wada H, Hada M, Ishihara K N, Shiga M and Nakamura Y 1990 *J. Phys. Soc. Japan* **59** 2956
Wada H, Shiga M and Nakamura Y 1989 *Physica* B **161** 197
Wohlfarth E P and Rhodes P 1962 *Phil. Mag.* **7** 1817
Yamada H 1993 *Phys. Rev.* B **47** 11 211
Yoshimura K, Mekata M, Takigawa M, Takahashi Y, Yasuoka H and Nakamura Y 1988a *J. Phys. Soc. Japan* **56** 1138
Yoshimura K, Takigawa M, Takahashi Y and Yasuoka H 1988b *Phys. Rev.* B **37** 3593