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Theory of itinerant electron metamagnetism

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Abstract. We propose a phenomenological picture for the mechanism of the metamagnetic transition often observed in intermetallic alloy compounds near the paramagnetic side of the ferromagnetic instabilities, such as YCo_2 with Al doping. The present treatment assumes the magnetization process is described in terms of the spin fluctuation spectrum, in clear contrast with the previous approaches based on the Hartree–Fock approximation assuming the fine structure of the density of states curve around the Fermi energy.

1. Introduction

The phenomenon of metamagnetism has been observed in a wide variety of compounds such as insulator antiferromagnets, the heavy-fermion systems, exchange-enhanced paramagnets, and so on. However, depending on the systems there seem to be a lot of underlying mechanisms. The metamagnetism of insulator magnets, for example, can be understood based on the Heisenberg model with an antiferromagnetic exchange interaction. In contrast, in the case of itinerant electron systems, it has been so far explained based on quite different pictures. In the ground state, a naive picture is based on the Hartree–Fock approximation and the non-linear dependence of the free energy with respect to the uniform magnetization M is evaluated. From a suitable energy dependence of the density of states curve around the Fermi energy, metamagnetism is predicted (Wohlfarth and Rhodes 1962). The presence of logarithmic terms derived from the Fermi liquid effect has been also proposed as a possible explanation (Misawa 1994). In the present paper, we deal with the metamagnetic transition observed in itinerant electron systems very close to their ferromagnetic instabilities, such as $TiBe_2$ (Acker *et al* 1981, Gerhardt *et al* 1983, Takagi *et al* 1984), and YCo_2 systems doped with Al. Among them we especially treat the intermetallic compound $Y(Co_{1-x}Al_x)_2$, because a lot of experimental studies have been done on this system (Yoshimura *et al* 1988a, b). Let us summarize here the observed magnetic behaviours of $Y(Co_{1-x}Al_x)_2$ which is of particular interest from the point of view of the present study.

(i) The weak ferromagnetism is observed in the Al concentration range between $x = 0.11$ and $x = 0.19$, and the metamagnetism is observed only on the paramagnetic side of $x = 0.11$, but is not observed around $x = 0.19$. See figure 1 for the schematic phase diagram.

(ii) The transition is of the first order as will be seen from the presence of the hysteresis in the magnetization curves at low temperature.

(iii) Below and after the metamagnetic transition, a good linearity holds for the Arrott plot (M^2 against H/M relation, where H and M are the external magnetic field strength

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and the uniform magnetization, respectively) of the magnetization curve at low temperature (see figure 2).

(iv) The magnetic susceptibility shows a Curie–Weiss temperature dependence at high temperature. At low temperature, after showing the maximum around T_m, it slightly decreases with decreasing temperature. It has been revealed that T_m is well correlated with the metamagnetic transition field H_m (Sakakibara *et al* 1990).

(v) The spin fluctuation spectrum shows a sudden change when going from the ferromagnetic phase to the paramagnetic phase as observed by NMR relaxation measurements (Yoshimura *et al* 1988b).

In the ground state as stated above, a simple, conventional approach for this problem was to evaluate M as a function of H based on the Hartree–Fock approximation (Wohlfarth and Rhodes 1962). Then we get the following relation:

$$H = aM + bM^3 + cM^5 + \cdots$$

where the coefficients are assumed to be given in terms of the density of states and its derivatives near the Fermi energy. Depending on the shape of the density of states, if the coefficient b becomes negative, we expect the occurrence of metamagnetism.

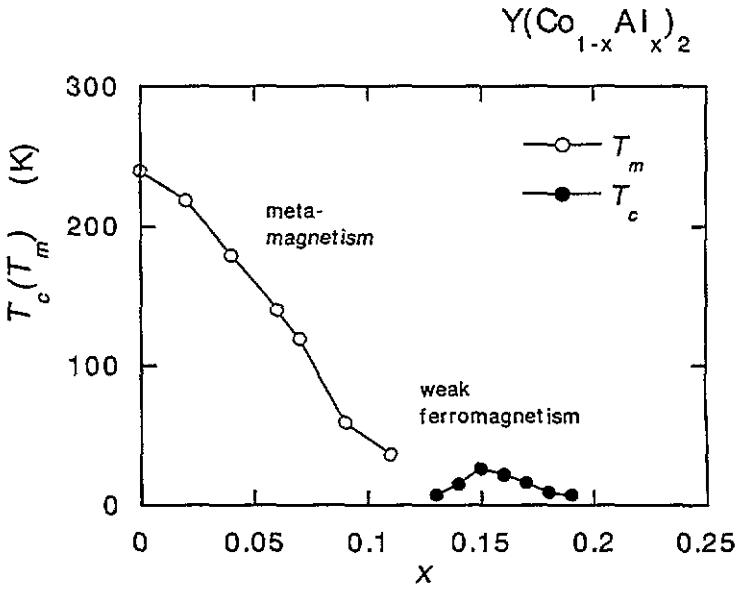

Figure 1. Phase diagram of $Y(Co_{1-x}Al_x)_2$.

The purpose of the present study is to propose a new mechanism for the phenomenon. The present work is motivated by the recent experimental investigations and the subsequent re-examination of the previous approaches. First the above Hartree–Fock approach does not seem to be consistent with the observed good linearity of the Arrott plot on the high-field side after the metamagnetic transition. A special form of the density of states curve may possibly explain the observed M against H relation. But it is not so easy to see why such a delicate form of the density of states happens to be realized. Another reason comes from the following observations. As for the finite-temperature properties of itinerant electron magnets, it has been well established that the thermal spin fluctuations play significant roles (Moriya 1979, Lonzarich 1984, Moriya 1985). The importance of the effects of

quantum (zero-point) spin fluctuations, however, has not yet been fully recognized. It is usually assumed that the effects of the quantum spin fluctuations are taken into account by the renormalization of parameters of the Hamiltonian such as the Coulomb interaction constant of the Hubbard Hamiltonian. Although the effects of thermal spin fluctuations on the metamagnetism have been discussed by Yamada (1993) and Yamada *et al* (1993), they still assumed the ground state would, in principle, be described by the Hartree–Fock approximation. It follows then, even in the framework of spin fluctuation theory, that we have to assume a suitable form of the density of states curve around the Fermi level to produce the field dependence of the magnetization curve. As for the effects of spin fluctuations in the ground state, the present study is based on a different point of view, i.e. all the magnetic properties should be determined by the nature of the spin fluctuations of the system, including ground state properties in quite the same way as at finite temperature (Takahashi 1986, 1990, 1992, 1994). It is natural to assume that even in the ground state, the magnetization process is determined by the behaviour of spin fluctuations in response to the external magnetic field. Actually the slope of the Arrott plot (M^2 against H/M plot) is determined by the spin fluctuation spectrum as will be seen below (Takahashi 1986), and its consequences are supported by recent experiments (Yoshimura *et al* 1988a, Shimizu *et al* 1990, Nakabayashi *et al* 1992).

Our preliminary treatment was presented in a preceding paper (Takahashi and Sakai 1995). Full details of our idea are presented here. The plan of the paper is as follows. In the next section, we briefly review the theoretical framework of the spin fluctuation theory. Then we show how to describe the metamagnetic transition of itinerant electron systems based on our new idea. The final section is devoted to conclusions and discussions.

2. Spin fluctuation theory in the ground state

In the present section we briefly review the theoretical framework of the spin fluctuation theory, though the discussions are mostly limited to the ground state properties. Our arguments are implicitly based on a model Hamiltonian for conduction electrons with strong intra-atomic electron–electron Coulomb repulsion among them, like the Hubbard Hamiltonian, as the origin of the magnetism. In the following, we show how to derive the magnetic equation of state with the use of a kind of sum rule without invoking any explicit perturbation expansion methods or mean-field-like approximation methods. As shown below, the present derivation does not require any explicit forms of the model Hamiltonian. We start with the condition that the squared local spin amplitude $\langle S_i^2 \rangle$ is almost independent of both the temperature and the external magnetic field variation as was indicated experimentally (Ziebeck *et al* 1982, Shiga *et al* 1988). In the presence of uniform spontaneous magnetization in the z-axis direction, the total spin fluctuation amplitude can be expressed in the following sum of contributions:

$$\langle S_i^2 \rangle = \langle \delta S_i \cdot \delta S_i \rangle + \sigma^2/4 \tag{1}$$

where σ is the spontaneous uniform magnetization per magnetic ion in units of μ_B, δS_i is the local spin deviation operator $S_i - \langle S_i \rangle$ on the ith lattice site (site index is neglected hereafter), and $\langle \ldots \rangle$ represents the thermal average. The equal-time spin correlation function in the right-hand side of equation (1) can be represented in terms of the dynamical spin susceptibility $\chi^{\alpha\beta}(q, \omega)$ with the use of the following fluctuation–dissipation theorem of statistical mechanics:

$$\left\langle \{\delta S_q^\alpha, \delta S_{-q}^\beta\}_+ \right\rangle = 2 \int_0^\infty \frac{\mathrm{d}\omega}{\pi} \coth\left(\frac{\omega}{2kT}\right) \operatorname{Im} \chi^{\alpha\beta}(q, \omega) \tag{2}$$

where $\{A, B\}_+$ represents the anticommutation relation between operators A and B, and δS_q^α stands for the α component of spin deviation operator with wave vector q. By decomposing the factor $\coth(\omega/2kT)$ in the integrand into the sum of the Bose factor $n(\omega)$ and a constant term, the local spin fluctuation amplitude $\langle \delta S^2 \rangle$ can be given as a sum of the quantum and thermal spin fluctuation parts as follows:

$$\begin{aligned} \langle \delta S^2 \rangle_{zp} &= \frac{1}{N_0^2} \sum_q \int_0^\infty \frac{d\omega}{\pi} \mathrm{Im}\{\chi^{xx}(q,\omega) + \chi^{yy}(q,\omega) + \chi^{zz}(q,\omega)\} \\ \langle \delta S^2 \rangle_{th} &= \frac{2}{N_0^2} \sum_q \int_0^\infty \frac{d\omega}{\pi} n(\omega) \mathrm{Im}\{\chi^{xx}(q,\omega) + \chi^{yy}(q,\omega) + \chi^{zz}(q,\omega)\} \\ n(\omega) &= [e^{\omega/kT} - 1]^{-1} \end{aligned} \tag{3}$$

where k is the Boltzmann constant and N_0 is the number of magnetic ions in the crystal. Hereafter we call the term $\langle \delta S^2 \rangle_{th}$ the thermal amplitude which has the Bose factor $n(\omega)$ in its integrand, and the other one $\langle \delta S^2 \rangle_{zp}$ the quantum fluctuation amplitude, which reduces to the *zero-point* fluctuations in the ground state. Although the quantum amplitude does not contain any explicit temperature dependence, note that it does depend on the external magnetic field and temperature through the variation of $\chi(q,\omega)$. In the case of weak ferromagnets, the low-energy and small-wave-vector parts of the spin fluctuation spectrum are known to be well described by the following Lorentzian form (Ishikawa *et al* 1985):

$$\mathrm{Im}\,\chi^{\alpha\alpha}(q,\omega) = \frac{\chi^\alpha}{1 + q^2/\kappa_\alpha^2} \frac{\omega \Gamma_q^\alpha}{\omega^2 + (\Gamma_q^\alpha)^2} \qquad \Gamma_q^\alpha = \Gamma_0 q(\kappa_\alpha^2 + q^2) \tag{4}$$

where χ^α represents the static uniform magnetic susceptibility for longitudinal and transverse components in units of $(g\mu_B)^2$ and κ_α^2 is the corresponding square of the inverse of the temperature-dependent correlation length proportional to χ_α^{-1}. The gyromagnetic ratio g is assumed to be two hereafter. In the presence of the static uniform magnetization, the above spectral form is slightly modified in the small-wave-vector region because of the appearance of the spin wave mode. However the effect can be neglected, because of the small phase volume, when performing the wave vector summation in equation (3). The form of equation (4) is also supported by the random phase approximation applied to the Hubbard model. In the following discussions, we assume equation (4) throughout whole the Brillouin zone.

According to our previous treatments on weak ferromagnets, $\chi(q,\omega)$ is characterized by the two energy scales kT_0 and kT_A defined by

$$kT_0 = \Gamma_0 q_B^3/2\pi \qquad kT_A = \frac{q_B^2}{2\kappa^2}\left(\frac{N_0}{\chi}\right) \tag{5}$$

where q_B is the effective zone boundary wave vector corresponding to the unit volume v_0 per magnetic ion defined by $(6\pi^2/v_0)^{1/3}$. These parameters T_0 and T_A give the measure of the width of the distribution of $\mathrm{Im}\chi(q,\omega)/\omega$ in q, ω space. Then in terms of the above parameters, $\mathrm{Im}\chi^{\alpha\alpha}(q,\omega)$ is rewritten in the following form:

$$\mathrm{Im}\,\chi^{\alpha\alpha}(q,\omega) = \frac{N_0}{2kT_A} \frac{1}{x^2 + y_\alpha} \frac{\omega[2\pi kT_0 x(y_\alpha + x^2)]}{\omega^2 + [2\pi kT_0 x(y_\alpha + x^2)]^2} \tag{6}$$

where x is the reduced wave vector defined by $x = q/q_B$. In the case where finite magnetization is present in the system, $\chi(q,\omega)$ becomes anisotropic owing to the anisotropy of the static uniform magnetic susceptibilities. We introduced the dimensionless parameters y_α (y and y_z for the transverse and the longitudinal components, respectively) in equation

(6). They are related to the transverse and the longitudinal static uniform susceptibilities by

$$y = \kappa_\alpha^2/q_B^2 = \frac{1}{kT_A}\frac{h}{\sigma} \qquad (\text{for } \alpha = x \text{ and } y)$$
$$y_z = \kappa_z^2/q_B^2 = \frac{1}{kT_A}\frac{\partial h}{\partial \sigma} \tag{7}$$

where we define $h = g\mu_B H$. Then it follows that the main origin of variation of $\chi(q, \omega)$ is those of the static and uniform susceptibilities.

From our definition (3), we see the thermal part $\langle \delta S^2 \rangle_{th}$ vanishes identically in the ground state. On the other hand the quantum fluctuation $\langle \delta S^2 \rangle_{zp}$ is finite and shows the field dependence through those of y and y_z. By substituting equation (6) into equation (3) and performing the integration and the summation with respect to ω and q, $\langle S^2 \rangle_{zp}$ can be expressed as a function of y and y_z. In the present paper, because we are particularly interested either in the highly exchange-enhanced paramagnetic cases or weakly ferromagnetic ones, both y and y_z are assumed to be very small. Therefore each component of $\langle S^2 \rangle_{zp}(y)$ is expanded with respect to y and y_z around their origins, resulting in the following y- and y_z-linear form (Takahashi 1986):

$$\langle \delta S_z^2 \rangle_{zp}(y_z) = \frac{1}{3}\langle S^2 \rangle_{zp}(0) - \frac{3T_0}{2T_A} y_z + \cdots$$
$$\langle S_x^2 + S_y^2 \rangle_{zp}(y) = \frac{2}{3}\langle S^2 \rangle_{zp}(0) - \frac{6T_0}{2T_A} y + \cdots. \tag{8}$$

According to Takahashi (1986), weakly ferro- and nearly ferromagnetic materials are characterized by their large zero-point amplitude $\langle S^2 \rangle_{zp}(0)$, which is almost comparable to the total amplitude. Depending on the sign of $\langle S^2 \rangle - \langle S^2 \rangle_{zp}(0)$, either the ferromagnetism or the near ferromagnetism is stabilized in the ground state, i.e., if we introduce the dimensionless parameter y_0 by

$$\langle S^2 \rangle_{zp}(0) - \langle S^2 \rangle = \frac{9T_0}{2T_A} y_0 \tag{9}$$

then we obtain the ferromagnetism in the ground state for negative y_0, whereas we get the exchange-enhanced paramagnetism for positive y_0. By substituting equation (8) into equation (1) and with the use of equation (9) we obtain

$$\frac{T_A}{9T_0}\frac{\sigma^2}{4} - \frac{1}{6}(y_z + 2y) = -\frac{y_0}{2} \qquad y_z = \sigma\frac{\partial y}{\partial \sigma} + y. \tag{10}$$

The second part of equation (10) simply comes from the definition of equation (7). The equation (10) serves as the basic equation in determining the magnetic equation of states by regarding it as a first-order differential equation of y with respect to σ. The magnetization process at $T = 0$ K is determined by solving y with respect to σ and the solution is easily found to be

$$y = y_0 + \frac{2T_A}{15T_0}\frac{\sigma^2}{4} = \frac{1}{kT_A}\frac{h}{\sigma}. \tag{11}$$

Note that in the case of weak ferromagnets with negative y_0, $h = 0$ in equation (11) gives a finite magnetization σ in the ground state. On the other hand, when y_0 is positive, σ is proportional to h for small h. Let us here introduce the third-order expansion coefficient $\bar{F}_1$ of h in the ground state with respect to σ (i.e., the fourth-order expansion coefficient of the free energy) by

$$h = kT_A y_0 \sigma + \bar{F}_1 \frac{\sigma^3}{8}. \tag{12}$$

Then from equation (11), the coefficient $\bar{F}_1$ is given by

$$\bar{F}_1 = \frac{4kT_A^2}{15T_0}. \tag{13}$$

Let us here summarize the results as follows. The significant consequence is that the magnetic equation of states is determined by the nature of spin fluctuations of the system. The good linearity of the Arrott plot (σ^2 against h/σ plot) is automatically derived in the ground state. The equation (12) may seem to have a similar form as the conventional expansion form of the free energy up to the fourth-order term with respect to the uniform magnetization. Note, however, equation (12) is not the result of the expansion, but the consequence of the y-linear form of equation (8). It terminates up to the third-order terms. From the above derivation, it is clear that coefficients of the expansion are determined by the nature of the spin fluctuation spectrum, i.e. in terms of T_0 and T_A. This means that even in the ground state the magnetization process is determined from the way the spin fluctuation will respond to the externally applied magnetic field. We are, of course, not saying that the density of states does not have any effect on the magnetization process of the system. What we actually mean is that the nature of the spin fluctuation spectrum will have the direct effects.

3. Origin of metamagnetism

Let us now discuss the metamagnetism in itinerant systems based on the results obtained in the preceding section. We first notice the results of NMR relaxation time measurements on $Y(Co_{1-x}Al_x)_2$ showing an abrupt change of the spin fluctuation spectrum as we vary Al concentration x through the ferromagnetic instability point, $x = 0.11$. We would also like to point out the fact that the Arrott plot for the compounds shows different slopes below and after the metamagnetic transition as shown in figure 2. Both the magnitudes of the slopes are well explained in terms of the spin fluctuation spectrum in respective para- and ferromagnetic phases as will be seen below. In order to be consistent with our standpoint based on the spin fluctuation theory, the above findings seem to suggest the existence of energetically almost degenerate electronic states with different spin fluctuation spectra. The metamagnetism is then interpreted as arising from the crossover of energies of these states in the presence of the external magnetic field, i.e., the paramagnetic state with spin fluctuation parameters T_0 and T_A becomes unstable against the ferromagnetic one with parameters T_0' and T_A'.

In order to describe the metamagnetic transition based on the above picture, let us assume the following free energies:

$$\begin{aligned} f_1(\sigma, 0) &= f_1(0, 0) + \tfrac{1}{2}a\sigma^2 + \tfrac{1}{4}b\sigma^4 \qquad \text{(for paramagnetic state)} \\ f_2(\sigma, 0) &= f_2(0, 0) - \tfrac{1}{2}a'\sigma^2 + \tfrac{1}{4}b'\sigma^4 \qquad \text{(for ferromagnetic state)} \end{aligned} \tag{14}$$

where $f_1(\sigma, T)$ and $f_2(\sigma, T)$ represent the free energies per magnetic ion as a function of the uniform magnetization σ and temperature T for the exchange-enhanced paramagnetic state and the nearly ferromagnetic state, respectively. We assume both the magnetization processes below and after the metamagnetic transition are well described by the above free energies except around the narrow metamagnetic transition region. From equation (13) the coefficients b and b' are determined by the spin fluctuation spectra through

$$b = \frac{kT_A^2}{60T_0} \qquad b' = \frac{kT_A'^2}{60T_0'}. \tag{15}$$

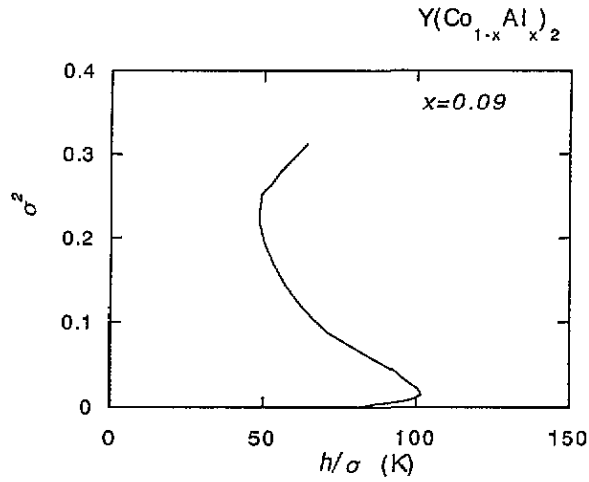

Figure 2. Arrott plot for $x = 0.09$.

We can check equation (15) experimentally as shown below. According to the NMR relaxation measurements and from the analysis of Takahashi (1994), the following inequalities hold:

$$T_0 < T_0' \qquad T_A > T_A'. \tag{16}$$

We show in table 1 calculated and observed values of b for $x = 0.09$. In the row labelled 'ferro', the observed b' (b^{obs}), estimated from the slope of the Arrott plot, is compared with b^{cal} estimated from equation (15). We employ the values of T_0' and T_A' for the ferromagnetic phase, $0.11 < x < 0.19$, evaluated by the NMR measurement. For the comparison of the low-field side of the Arrott plot in the second row, we employ the T_0 value from the NMR measurement, and the T_A value estimated by Takahashi (1994). Fair agreement seems to be obtained considering the ambiguity in estimating T_0 and T_A. Both the analyses give nearly the same ratio, $b/b' \simeq 10$, thus leading to the following inequality:

$$b > b'. \tag{17}$$

The present picture is also consistent with the observed linearity of the high-field side of the Arrott plot (figure 2) of $Y(Co_{0.91}Al_{0.09})_2$. In order to derive metamagnetism, we have to assume that the paramagnetic states are slightly lower in energy than the ferromagnetic ones for $x < 0.11$. Because of the inequality (17), the ferromagnetic state with higher magnetic moment, however, becomes stabilized in the presence of the external magnetic field, thus resulting in the metamagnetic transition.

Table 1. Calculated and observed coefficients b for $x = 0.09$.

	T_0 (K)	T_A (K)	b^{cal} (K)	b^{obs} (K)
Para	0.5×10^3	1.0×10^4	3.3×10^3	1.3×10^3
Ferro	2.0×10^3	0.7×10^3	0.41×10^3	0.12×10^3

By extending the previous discussion, we propose the following explanation for the overall phase diagram and the magnetic properties of this series of compounds. To begin with we assume that the higher-moment states for $x < 0.11$, stabilized by applying the

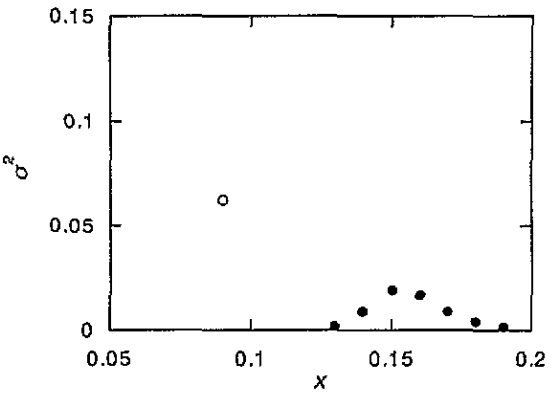

Figure 3. The observed squared saturation magnetization σ^2 as a function of Al concentration x. The open circle for $x = 0.09$ is estimated by extrapolating the Arrott plot (see the text).

high magnetic field, originate from the same electronic state with nearly the same spin fluctuation spectrum as the weak ferromagnetic states realized in the Al concentration range $0.11 < x < 0.19$. Supporting evidence comes from the analysis as shown in figure 3. We plot the values of σ^2 (solid circles) as a function of x for the ferromagnetic compounds. In the same figure, we also plot the value of σ^2 (an open circle) obtained by extrapolating the high-field part of the Arrott plot to the vanishing H/M limit for $x = 0.09$ (see figure 2). The open circle is clearly on the same straight line extrapolating from the observed σ^2 values for weak ferromagnets. This seems to suggest that the weak ferromagnetic state stabilized for $0.11 < x < 0.19$ still persists for $x < 0.11$ and a' in equation (14) behaves as

$$a' = \lambda'(x_l - x) \qquad (\text{with } x_l \simeq 0.19). \tag{18}$$

The paramagnetic state observed for $x < 0.11$, on the other hand, is assumed to originate from a different electronic state with a different spin fluctuation spectrum. The coefficient a in equation (14) is estimated experimentally from the inverse of the magnetic susceptibility at $T = 0$ K. As shown by Sakakibara *et al* (1990), the observed a decreases almost linearly in the paramagnetic phase towards $x = 0.11$ as represented by

$$a = \lambda(x_0 - x) \qquad (\text{with } x_0 \simeq 0.11). \tag{19}$$

Now the origin of the overall phase diagram is explained as follows. Two electronic states with different spin fluctuation spectra happen to show ferromagnetic instabilities in nearly the same concentration range of Al. A state becomes ferromagnetic around $x = 0.11$ with increasing x. Another one also becomes ferromagnetic around $x = 0.19$ but with decreasing x. The crossover of their relative energies seems to occur around $x = 0.11$. The state with the critical concentration around $x = 0.11$ becomes stabilized for $x < 0.11$ against the one with critical concentration $x = 0.19$. Spin fluctuation spectra for both states are assumed to give $b > b'$. It follows then that the ground state is paramagnetic for $x < 0.11$, and the metamagnetic transition occurs on applying the external magnetic field. For $0.11 < x < 0.19$, two ferromagnetic states coexist and one of them is stabilized in the ground state. Finally, for $0.19 < x$, the ground state again becomes paramagnetic. In this case we do not expect the occurrence of metamagnetism, because the ferromagnetic state is already higher in energy compared with the case around $x = 0.11$ and the relative magnitude of coefficients b and b' does not favour the ferromagnetic state any more. In this way, we can reasonably explain why the metamagnetic transition occurs only around

$x = 0.11$ and does not occur around the other side of the ferromagnetic instability point, $x = 0.19$.

Based on our picture, let us now estimate the metamagnetic transition field H_m simply by using the Maxwell condition, i.e., from energetic considerations. Then the transition field H_m is determined by solving the following set of equations, the condition that both the tangential lines of the function f_1 at σ_1 and of f_2 at σ_2 in equation (14) coincide with each other under the external field H_m:

$$\begin{aligned} h_m/2 &= a\sigma_1 + b\sigma_1^3 = -a'\sigma_2 + b'\sigma_2^3 \\ h_m/2 &= \{f_2(\sigma_2, 0) - f_1(\sigma_1, 0)\}/(\sigma_2 - \sigma_1) \end{aligned} \tag{20}$$

where we defined $h_m = 2\mu_B H_m$. The ferromagnetic free energy $f_2(\sigma, 0)$ has a minimum around $\sigma_0 = (a'/b')^{1/2}$. By linearizing equation (20) with respect to σ_1 and $\delta\sigma_2 = \sigma_2 - \sigma_0$, we obtain the following results:

$$a\sigma_1 = 2a'\delta\sigma_2 = h_m/2f_2(\sigma_0, 0) - f_1(0, 0) = \kappa(x_0 - x) \simeq a\sigma_1\sigma_0 \tag{21}$$

where we introduced a constant κ. From (18) and (21), we obtain the x dependence of h_m:

$$h_m = 2a\sigma_1 = \frac{2\kappa}{\sigma_0}(x_0 - x) = 2\kappa\left(\frac{b'}{\lambda'}\right)^{1/2}\frac{x_0 - x}{(x_1 - x)^{1/2}}. \tag{22}$$

We see that the metamagnetic transition field H_m decreases almost linearly towards $x = 0.11$ with increasing x, while σ_0 and σ_1 are nearly constant there in agreement with experiments. We show values of σ_1 for several values of x in table 2.

Table 2. Observed values of σ_1.

x	σ_1
0.00	0.17
0.03	0.14
0.06	0.19
0.07	0.21
0.09	0.22

Before concluding this section, we would like to make some comments on the finite-temperature magnetic properties. We first point out that the temperature T_m, where the magnetic susceptibility shows its maximum, seems to be closely related to the *ferromagnetic* Curie temperature T_C, evaluated by assuming that the ferromagnetic states would be stabilized still for $x < 0.11$. This is suggested by the following considerations. First the theoretically estimated T_C is well correlated with observed T_m. According to the spin fluctuation theory, T_C is related to the parameters σ_0, T_0 and T_A through

$$\sigma_0^2/4 = \frac{15T_0}{T_A}c\left(\frac{T_C}{T_0}\right)^{4/3} \qquad (c = 0.3353\ldots). \tag{23}$$

For $x = 0.09$, for instance, we can estimate $T_C = 67$ K with the use of $T_0 = 2.0 \times 10^3$ K, $T_A = 0.7 \times 10^4$ K and $\sigma_0 = 0.25$ evaluated by extrapolating the high-field side of the Arrott plot to the vanishing h/σ limit. The value compares well with the observed $T_m = 60$ K. We also see in figure 1 that plots of T_m in the paramagnetic phase are smoothly merged into the values of T_C in the weak ferromagnetic phase with increasing x, also indicating both quantities are closely related to each other. We do not have any convincing theoretical

explanation at present. Inhomogeneity-driven ferromagnetic small clusters may be a possible origin of T_m.

As for the x dependence of T_C, we get the following x dependence of σ^2 from equation (18) (see figure 3):

$$\sigma_0^2 = (\lambda'/b')(x_1 - x)$$

which is consistent with experiments shown in figure 3. By eliminating σ_0^2 in equation (23) with the use of the above equation, we get

$$\frac{T_C}{T_0} = \left(\frac{\lambda' T_A}{60 b' c T_0}\right)^{3/4} (x_1 - x)^{3/4}. \tag{24}$$

If we further eliminate x from equation (24) by using equation (22), we finally obtain

$$\frac{T_C}{T_0} = \left(\frac{\lambda' T_A}{240 b' c T_0}\right)^{3/4} \left\{\bar{h} + \sqrt{\bar{h}^2 + 4(x_1 - x_0)}\right\}^{3/2} \tag{25}$$
$$\bar{h} = (h_m/2\kappa)(\lambda'/b')^{1/2}.$$

The above results explain the observed general tendency between T_m and H_m, that is, H_m increases with increasing T_m. Contrary to the experiments, equation (25) is not a strict linear relation. The change of the spin fluctuation spectrum with varying x, which is not taken into account here, may improve the agreement. The main discrepancy of equation (25) lies in the small-T_m (i.e., T_C) region. The reason results from our overestimate of T_m. Our T_C is finite around the critical concentration $x = 0.11$, though T_m vanishes there. Around the critical concentration the temperature dependence of the susceptibility will be very sensitive to various perturbations. Because the actual system is not a clean system, the observed T_m is likely to be smeared out by various effects and smaller values will be obtained as a result. In order to make actual comparisons with experiments, we have to take various extrinsic effects into our considerations.

4. Conclusions and discussions

In the present paper, we have proposed a new mechanism of the metamagnetism for itinerant electron systems. We assumed the presence of the energetically almost degenerate electronic states, whose ferromagnetic instabilities are very close to each other. Based on the spin fluctuation theory, the magnetization processes are evaluated in terms of the spin fluctuation spectrum inherent to each of these states, except around the narrow metamagnetic transition region. The metamagnetism is brought about because of the crossover of these states in the presence of the external magnetic field. Based on this picture, we can explain the following properties:

(i) why the metamagnetism occurs only near the paramagnetic side of the critical concentration $x = 0.11$, but it does not occur around $x = 0.19$;

(ii) a good linearity of the Arrott plot for both the low- and high-field limits;

(iii) the tendency of the change of slopes from the low-field side to the high-field side, i.e. $b > b'$, is consistent with the observed change of the spin fluctuation parameters; sizes of inverses of slopes b and b' in (2) and (3) are roughly consistent with the spin fluctuation parameters;

(iv) the linear decrease of H_m with respect to the Al concentration x with increasing x towards $x = 0.11$;

(v) near x independence of σ_1.

The present mechanism is quite different from that of insulator magnets, for it is nothing to do with the antiferromagnetic correlation. In order to test the validity of the present mechanism, direct measurements of the spin fluctuation spectrum in the presence of the high magnetic field either by neutron diffraction experiments or by the NMR relaxation measurements are expected. Almost all the magnetization measurements have been so far focused on the presence of the metamagnetic transition, and therefore enough data showing the clear linearity of the high-field side of the Arrott plot are lacking. More detailed measurements in the high-field region are also awaited.

A remaining question is what are the microscopic origins of the energetically almost degenerate electronic state, a paramagnetic and a weak ferromagnetic states with different spin fluctuation spectra. In the present study we say nothing about this question. In this sense our approach remains phenomenological. The present study strongly suggests that the phenomenon has to be explained associated with the change of the spin fluctuation spectrum occurring at the transition. The discontinuous change of the electronic states associated with the appearance of the ferromagnetic moment in the system may lead to the change of the spin fluctuation spectrum as a result. In order to estimate the metamagnetic transition field, we have simply employed the Maxwell condition. For the discussion of the origin of the hysteresis, we need some sophistication to treat the transition region.

At finite temperature, the metamagnetic transition field increases with temperature, being proportional to T^2, as was observed experimentally by Goto *et al* (1994). According to the spin fluctuation theory, the temperature dependence of the magnetic free energy $f(0, T)$ at low temperature is represented in the following form (Konno and Moriya 1987):

$$f(0, T) - f(0, 0) = -\frac{3T^2}{T_0} \ln(1 + 1/y). \tag{26}$$

Because of the inequality (16) of T_0 for states with larger and smaller moment, we expect the additional positive T^2 term in the free energy difference, and therefore we predict $H_m \propto T^2$ from equation (21) in agreement with experiments. Discussions on finite-temperature properties will also be the subject of our future investigations.

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