Magnetic Isotherm of Itinerant Electron Magnets – A New Approach to Itinerant Electron Metamagnetism

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Abstract. Theoretical development on the magnetic isotherm in itinerant electron ferromagnets is briefly reviewed in comparison with observed magnetization measurements. Itinerant electron metamagnetic transitions originate from the magnetization curve derived from the negative fourth expansion coefficient of the free energy in magnetization. In our view, however, it is difficult to expect the negative coefficient of the magnetic origin. We therefore propose a new mechanism of the metamagnetic transition, by taking an explicit account of volume dependence of the free energy, which is consistent with our spin fluctuation theory of magnetic isotherms.

1. Introduction
Metamagnetism is usually observed in highly exchange enhanced itinerant electron paramagnets close to the magnetic instability point. It exhibits as a sudden discontinuous jump of the induced magnetic moment at some critical magnetic field \( H_c \). According to the widely accepted theoretical idea, the phenomenon originates from the distinct magnetic isotherm associated with the concave shape of density of states of single-electron excitations around the Fermi energy. At finite temperatures, transitions are affected by thermal spin fluctuations. On the other hand, magnetic isotherms are exclusively determined by the effect of spin fluctuations influenced by the externally applied magnetic field. This can be applied even in the ground state with no thermal spin fluctuation amplitudes. This means that the theoretical bases of metamagnetic transitions conflicts with the spin fluctuation theory of magnetic isotherms.

The motivation of this study is, therefore, first to make a brief review of the current theoretical ideas on the magnetic isotherm of itinerant electron magnets. We also show how the theoretical predictions are verified experimentally. We next propose a new idea for the metamagnetic transition observed in itinerant electron magnets, that is consistent with the spin fluctuation theory.

2. Theories of magnetic isotherm of itinerant electron magnets
If we apply magnetic field \( H \) to magnets, the magnetic moment \( M \) is induced. The relation between \( H \) and \( M \), called magnetic isotherm, is usually described by the following thermodynamic relation:

\[
H = \frac{\partial F(M, T)}{\partial M} = a(T)M + b(T)M^3 + \cdots .
\]
This is a fundamental property of magnetic materials, for we can derive a number of magnetic properties from them. One of the main concerns of theoretical studies has been therefore to determine the magnetic isotherm as the effect of spin fluctuations in the presence of the magnetic field. It has long been believed that observed magnetization curves in itinerant ferromagnets are well-fitted with the above formula with almost temperature independent $b(T)$ as far as the induced static moment is very small. Only the temperature dependence of the coefficient $a(T)$ of the first term in the right hand side was a subject of our main interest. However, the idea is now regarded as too simple and more elaborate treatments are necessary.

Theory of magnetic isotherms is crucial for our understanding of the mechanism of metamagnetic transitions. At present there are following two opposite theoretical explanations for them.

(i) Band splitting mechanism
According to this picture, the splitting of energies of each conduction electron states induced by the applied magnetic field is the origin of the induced magnetic moment in the crystal. The well-known self-consistent renormalization (SCR) theory of spin fluctuations is also classified as this, because it is equivalent with the single-particle band picture as far as the isotherm in the ground state is concerned.

(ii) Spin fluctuation mechanism
In this picture, amplitudes of spin fluctuations suppressed by the external magnetic field is regarded as the origin of induced magnetic moment.

Depending on mechanisms, we expect a different temperature dependence of $b(T)$. The first view gives the very weak dependence, that can be usually neglected. On the other hand, $b(T)$ becomes temperature dependent in the second view. Most of experiments to date support the second view.

2.1. Spin fluctuation theory of itinerant electron magnets
What follow are basic mechanisms how magnetic properties are determined by the effects of spin fluctuations. They are based on the following basic two ideas [1, 2].

(i) The total spin amplitude conservation (TAC)
This means that the sum of the squared quantum and thermal spin fluctuation amplitudes and the squared static moment remains constant independent of temperature and/or in the presence of the external magnetic field.

(ii) The global consistency (GC) in the $H$-$M$ space
The SCR spin fluctuation theory is mainly concerned with the temperature dependence of $a(T)$ of the first term of the isotherm (1), i.e., the inverse of magnetic susceptibility. We mean by GC, on the other hand, that we should rather deal with the relation between $H$ and $M$ over a wide range of their variables.

For paramagnets, for simplicity, the equal-time local spin correlation is given by

$$\langle S_{loc}^2 \rangle = \frac{3}{N_0^2} \sum_q \int_0^\infty \frac{d\omega}{\pi} \coth(\omega/2k_BT) \text{Im}\chi(q,\omega),$$

where the imaginary part of the dynamical magnetic susceptibility is given in the following double Lorentzian form,

$$\text{Im}\chi(q,\omega) = \chi(q,0) \frac{\omega \Gamma_q}{\omega^2 + \Gamma_q^2},$$

$$\chi(q,0) = \chi(0,0) \frac{\kappa^2}{q^2 + \kappa^2}, \quad \Gamma_q = \Gamma_0 q (\kappa^2 + q^2).$$
The number of magnetic atoms, the correlation wave-number squared, and the damping constant are, respectively, denoted by $N_0$, $\kappa^2$, and $\Gamma_q$. From the decomposition, $\coth(\omega/2k_BT) = 1 + 2n(\omega)$, the amplitude in (2) is split into thermal and zero-point amplitudes as follows.

\[
3A_t(\kappa^2, T) \equiv \langle S_{loc}^2 \rangle_T = \frac{6}{N_0^2} \sum_q \int_0^\infty \frac{d\omega}{\pi} n(\omega) \text{Im} \chi(q, \omega),
\]

\[
3A_z(\kappa^2) \equiv \langle S_{loc}^2 \rangle_Z = \frac{3}{N_0^2} \sum_q \int_0^\infty \frac{d\omega}{\pi} \text{Im} \chi(q, \omega).
\]

Both amplitudes depend on $\kappa^2$ through its dependence of $\chi(q, 0)$ and $\Gamma_q$ in (3). The thermal amplitude has an extra explicit temperature dependence through the Bose distribution function $n(\omega)$. In the presence of the spontaneous or induced magnetic moment due to external magnetic field, $\kappa^2$ becomes anisotropic, as given by $\kappa^2 \propto \partial H/\partial M$ and $\kappa^2 \propto H/M$ for fluctuations parallel or perpendicular to the direction of the static moment. The amplitude conservation can now be written in the form,

\[
2A_t(\kappa^2, T) + A_t(\kappa^2, T) + [2A_z(\kappa^2) + A_z(\kappa^2)] + m^2 = 3[A_t(0, T_c) + A_z(0)],
\]

where $m = M/N_0$ is the magnetic moment per magnetic atom. The right hand side represents the value of the amplitude at the critical point.

We require that (5) should be regarded as a differential equation of $H$ as a function of $M$, for we can find the derivative, $\partial H/\partial M$, for given values of $M$ and $H$ from (5). Because $M$ dependence of $H$ is obtained as a solution of (5), our main concerns are not restricted to the first term of (1) but the global behavior of $M$ dependence over the wide range of the external magnetic field. This is what we mean by GC.

For example, the thermal amplitude increases with temperature in the paramagnetic phase. To satisfy the amplitude conservation,

\[
A_t(\kappa^2, T) + A_z(\kappa^2) - A_z(0) = A(0, T_c).
\]

the zero-point amplitude has to be suppressed to compensate for the increase of the thermal amplitude by the temperature dependent magnetic susceptibility, $\chi \propto \kappa^{-2}$. As a solution of (6), the temperature dependence of $\kappa^2$, i.e. the Curie-Weiss law behavior of the magnetic susceptibility, is derived. In the same way, the spin amplitude suppression by the external magnetic field induces a static magnetic moment to satisfy the TAC requirement, that determines the magnetic isotherm. The mechanism is quite different from that of the band picture.

2.2. Theory of Magnetic isotherm

According to the spin fluctuation mechanism, various interesting magnetic properties have been derived. We are particularly concerned with only three of them, which are all related to the magnetic isotherm.

(i) Magnetic isotherm in the ground states

The first example is related to the magnetization curve in the ground state. It can be determined by solving the differential equation,

\[
2[A_z(\kappa^2) - A_z(0)] + [A_z(\kappa^2) - A_z(0)] + m^2 = A_t(0, T_c).
\]

Since $A_z(\kappa^2) - A_z(0) \propto \kappa^2$ for $\kappa^2 \ll 1$ and from the relations $\kappa^2 \propto H/M$, $\kappa^2 \propto \partial H/\partial M$, we can easily find the solution in the form,

\[
H = a_0 M + b_0 M^3
\]
The substitution into (7) gives the following $b_0$ of the second term in the right hand side given by 

$$b_0 = \frac{4k_B T_2^2}{15(2\mu_B)^4 N_0^3 T_0},$$

(9)

where $T_0$ and $T_A$, defined by $\Gamma_0 q_B^2 / 2\pi$ and $q_B^2 / (2\chi(0, 0) \kappa^2)$ with zone boundary wave-vector $q_B$, represent the dispersions of spin fluctuation spectra in frequency and wave-vector spaces.

(ii) Critical magnetic isotherms

In the critical limit, since $\kappa^2 \to 0$ as $T \to T_c$, (5) is written in the form,

$$2[A_t(\kappa^2, T_c) - A_t(0, T_c)] + [A_t(\kappa_0^2, T_c) - A_t(0, T_c)] + m^2 = 0,$$

(10)

because the effects of zero-point fluctuations become negligible in this limit. From the critical behavior of the thermal amplitude, $A_t(\kappa^2, T) - A_t(0, T) \propto \sqrt{\kappa^2}$, we can find the solution of the critical magnetic isotherm of the form,

$$H \propto M^5.$$  

(11)

The moment $M$ is induced in proportional to $H^{1/5}$ at $T = T_c$, instead of $H^{1/3}$ by the band picture.

(iii) Positive mode-mode coupling in semiconducting paramagnets

The coefficient $b(T)$ in (1) is regarded as the nonlinear mode-mode coupling among spin fluctuation modes. While the negative sign is derived by the band splitting mechanism, we can predict the positive value even in cases of semiconducting paramagnets with concave form of the density of states around the Fermi energy [3].

We now show how these properties are verified experimentally in the following subsections.

2.3. Magnetic isotherm in the ground state

In place of the Rhodes-Wohlfarth plot ($p_C / p_s$ vs $T_c$), a new revised plot, $p_{\text{eff}} / p_s$ vs $T_c / T_0$, was proposed by Takahashi [1, 2], where $p_s$, $p_{\text{eff}}$ and $p_C$ represent the spontaneous moment in the low temperature limit, the effective moment estimated from the Curie constant of the magnetic susceptibility, and $p_C(p_C + 2) = p_{\text{eff}}^2$. It is based on the universal relation between these two ratios that can be derived from (9) in the ground state. As shown in Figure 1, almost all the experimentally estimated ratios fall near the solid curve determined theoretically. The good agreement of the plot is equivalent with the validity of (9). It is very difficult to figure out its reason based on the band picture.

2.4. Critical magnetic isotherm

The idea of critical magnetic isotherm had been long absent in the theories of itinerant magnetism. Since the relation (11) was first predicted by Takahashi [1], the observed linear relations satisfied between $M^4$ and $H/M$ have now accumulated. As an old example, we show in Figure 2 (above) the observed values of $M^4$ almost at the critical point, $T = 29$ K, plotted against $H/M$. The conventional Arrott plot [4] is also shown in the same figure (below). Recently the good linear relations are observed by Nishihara et al. in Ni [5] and the Heusler compound Co$_2$CrGa [6]. These results clearly shows that coefficient $b$ of the free energy must be temperature dependent and has to vanish at the critical point, in contradiction to the band picture and the assumption of the SCR theory.
2.5. Sign of mode-mode coupling constant in semiconducting paramagnets
The last example is closely related to the metamagnetic transitions. According to the band picture, the coefficient $b(T)$ is given by

$$b = \frac{1}{\rho^3} \left[ \left( \frac{\rho'}{\rho} \right)^2 - \frac{\rho''}{3\rho} \right].$$

(12)

For semiconducting materials with the concave shape of the density of states curve $\rho$ around the Fermi energy, $b$ becomes negative, i.e., in cases of $\rho'' > 0$ and $\rho' \sim 0$. Analyses of the magnetization process of semiconducting compounds, FeSi by Koyama [7] and FeSb$_2$ by Koyama [8], show that the couplings $b$ determined from their slopes in the Arrott plots are rather positive, that favors again the spin fluctuation mechanism even qualitatively.

3. Theory of metamagnetic transitions
Let us now turn to the subject of metamagnetic transitions. The current dominant idea of itinerant metamagnetic transition is based on the following magnetic isotherm [9],

$$H = a(T)M + b(T)M^3 + cM^5.$$  (13)

The coefficient $b(T)$ in the second term is assumed to be negative because of the concave form of the density of states around the Fermi energy. However as we have shown in the preceding section, the negative $b(T)$ of the purely magnetic origin is very difficult from the spin fluctuation mechanism.

In addition, the third $M^5$ term with finite positive $c$ is necessary in (13) for the stability of the free energy. Only because of this higher order nonlinear mode-mode coupling, $b(T)$ becomes temperature dependent and changes its sign at high temperatures. There however
Figure 2. Magnetization curve of MnSi at $T = 29$ K reploted in the form, $M^4 H/M$, (upper) and in Arrott plot (lower).

exist unfavorable experiments for this term. We show the Arrott plot of high field magnetic measurements on $Y(Co_{0.91}Al_{0.09})_2$ by Goto and Bartashevich (1998) [11] in Figure 3. In their original figure, induced moment $M$ is plotted against $H$. The linear relation seems to be satisfied between $M^2$ and $H/M$ in the ferromagnetic state after the transition. The same behavior is also observed recently in the magnetization curve of $LaCo_9Si_4$ by Michor et al [12] in even wider range of $H/M$. All of these observations clearly require a new idea for more plausible understanding of the phenomena.

3.1. Volume dependence of the magnetic isotherm

As a new mechanism, it seems necessary to introduce another degrees of freedom that couples with magnetism. The volume will be one of the best candidates, for the increase or the appearance of the magnetization usually accompanies the volume expansion of crystals. The metamagnetic transitions will then occur between two almost degenerate free energies, $F_p(M, V)$ and $F_f(M, V)$, from the paramagnetic state, stable around the volume $V_0$, to the ferromagnetic
state around the volume $V_1$ in the presence of magnetic field. The magneto-volume effect [10] is assumed to be involved in the transition. In the paramagnetic ground state for $V \sim V_0$, the free energy is expanded in $M$ as follows.

$$F_p(M, V) = F(0, V) + \frac{1}{2} a(V) M^2 + \frac{1}{4} b(V) M^4 \quad (14)$$

In the absence of $H$, both $a(V_0) > 0$ and $b(V_0) > 0$ are satisfied for volume $V = V_0$. For ferromagnetic state, metastable around $V \sim V_1$, it is given by

$$F_f(M, V) = F_f(M_0, V) + \frac{1}{2} a(V)[M^2 - M_0^2] + \frac{1}{4} b(V)[M^4 - M_0^4]$$

$$= F_f(M_0, V) - a(V)[M - M_0]^2 + b(V) M_0[M - M_0]^3 + \cdots, \quad (15)$$

$$F_f(M_0, V) = F(0, V) + \frac{1}{2} a(V) M_0^2 + \frac{1}{4} b(V) M_0^4,$$

where $M_0^2 = -a(V)/b(V)$ and $a(V_1) < 0$.

Because of the volume dependence of expansion coefficients around $V_0$, $a(V)$ is given by

$$a(V) \simeq a(V_0) + a'(V_0) \delta V = a(V_0) - \frac{1}{2} \gamma_p M^2,$$

$$\delta V = V - V_0 = V_0 C_p M^2, \quad \gamma_p = -2V_0 a'(V_0) C_p$$

where $C_p$ is the forced magneto-volume coupling constant. Because of the forced magneto-volume effect, the volume change is equivalent with the induced magnetic moment squared. This means that $b(V)$ in (14) effectively becomes negative if

$$b(V) = b(V_0) - \gamma_p < 0,$$

is satisfied. The point is that the negative $b$ becomes effectively possible because of the magneto-volume effect.
3.2. Condition of the transition

Once $b$ becomes negative, metamagnetic transitions in the ground state can be realized as follows. For field-increasing processes, the transition occurs at the field $H_c$ of divergent differential magnetic susceptibility, $\partial M/\partial H = \infty$, or zero of its reciprocal, $\partial H/\partial M = a + b M^3 = 0$. It follows then,

$$M_{c1}^2 = \frac{a}{3|b|}, \quad H_c = -2bM_{c1}^3 = \frac{2a}{3^{3/2}} \sqrt{\frac{a}{|b|}} = \frac{2}{3} a M_{c1}.$$  

The transition occurs from the state with the moment $M_{c1}$ and the volume $V_0$ slightly shifted by $\delta V = C_p M_{c1}^2$. The ratio of the critical field $H_c$ and the moment $M_{c1}$ is given by

$$\frac{H_c}{M_{c1}} = \frac{2}{3} a |H| \left| \frac{M_{c1}}{M} \right|_0.$$  

(16)

The reverse process with decreasing field is also realized in the same way. If the transition occurs from the moment $M_{c1}$ to $M_{c2}$ at the critical field $H = H_c$, the magnetic isotherm of the ferromagnetic state can be expanded as follows,

$$H = H_c + \alpha (M - M_{c2}) + \beta (M - M_{c2})^2 + \cdots,$$  

(17)

where $\alpha$ and $\beta$ are coefficients renormalized by the effect of magneto-volume effects. The volume $V_1$ and $M_{c2}$ are evaluated by the simultaneous equations,

$$\frac{\partial F(M_{c2}, V_1)}{\partial V_1} = \frac{\partial F(0, V_1)}{\partial V_1} + H_c \frac{\partial M_{c2}}{\partial V_1} + \frac{1}{2} a'(V_1) M^2 + \frac{1}{4} b'(V_1) M_{c2}^4 = 0.$$  

(18)

The magnetic isotherm of the system with the volume around $V_1$ is then given by

$$H = H_c + \left[ a(V_1) + 3M_{c2}^2 b(V_1) \right] M + 3M_{c2} b(V_1) \delta M \quad + (a' M_{c2} + b' M_{c2}^3) \delta V + (a' + 3M_{c2} b') \delta V M + \cdots$$

$$\frac{\partial}{\partial V} \left[ F(0, V) + \frac{1}{2} a(V) M_{c2}^2 + \frac{1}{4} b(V) M_{c2}^4 \right] \delta V \quad \frac{\partial}{\partial V} \left[ F(0, V) + \frac{1}{2} a(V) M_{c2}^2 + \frac{1}{4} b(V) M_{c2}^4 \right] \delta V$$

$$- H_c \frac{\partial M}{\partial V} + H_c \frac{\partial M_{c2}}{\partial V} - \frac{1}{2} a'(V) [M^2 - M_{c2}^2] - \frac{1}{4} b'(V) [M^4 - M_{c2}^4],$$

(19)

where $\delta M = M - M_{c2}$. From the second equation of (19), the forced magnetostriction $\delta V$, and therefore the volume dependence of $a(V)$ are given by

$$\delta V = V_1 C_f (M^2 - M_{c2}^2) \simeq 2V_1 C_f M_{c2} \delta M,$$

$$a(V) \simeq a(V_1) + a'(V_1) \delta V = a(V_1) - \gamma f M_{c2} \delta M,$$  

(20)

where the first two terms and the last term are neglected. The magneto-volume coupling in the ferromagnetic state is denoted by $C_f$ and $\gamma_f = -2V_1 a'(V_1) C_f$. The substituting (20) into the first line of (19) leads to values of $\alpha$ and $\beta$ in (17) given by

$$\alpha = a(V_1) + 3M_{c2}^2 b(V_1) - \gamma_f M_{c2}^2, \quad \beta = M_{c2} [3b(V_1) + \gamma_f].$$  

(21)

The critical field for the reverse transition is now determined by

$$\frac{\partial H}{\partial M} = \alpha + 2\beta (M - M_{c2}) = 0.$$  

(22)
The critical field and the moment are given by

\[ M_{c2}' = M_{c2} - \frac{\alpha}{2\beta}, \quad H_{c}' = H_{c} - \frac{\alpha^2}{4\beta} \]  

(23)

Because of the magneto-volume effect, smaller \( \alpha \) and larger \( \beta \) lead to the smaller difference between the critical fields, \( H_{c} \) and \( H_{c}' \).

**Figure 4.** Free energies and magnetic isotherms, \( H \) vs \( M \), of metamagnetic transitions.

The proposal of my idea of metamagnetic transition is clearly shown in Figure 4. In the left part, the magnetization curve and the paramagnetic free energy around the volume \( V_0 \) are shown. Those around \( V_1 \) shown in the right hand side are for the ferromagnetic state. These are effective magnetic isotherms and free energies. Transitions occur at the maximum and the minimum of \( H \) for increasing magnetic field. In our mechanism, we only need the conditions for the transition:

- \( b \) becomes effectively negative due to the volume effect, for instance.
- Systems must be very close to the instability point for transitions in lower magnetic field range.

The above idea is actually supported by the high field experiment of the metamagnetic transition on \( \text{Y(Co,Al)}_2 \) [11]. Simultaneously observed discontinuous volume changes at the transitions clearly show that both the volume change and the metamagnetic transition are intimately correlated with each other.

**4. Summary**

In this study we made a brief review on predominant rolls of spin fluctuations on the magnetic isotherm of itinerant electron magnets by paying particular attention on the following properties:
Magnetization curves in the ground state
Critical magnetic isotherms
Positive fourth expansion coefficient of the free energy in magnetization observed in semiconducting paramagnets

Agreements between theoretical predictions and experiments demonstrate that magnetization curves of itinerant electron magnets are exclusively determined by the effect of spin fluctuations. The above view imposes a stringent restriction to the magnetization curves. It becomes very difficult to assume a negative coefficient $b(T)$ of the isotherm in (1). We therefore propose a new idea as a basic mechanism of itinerant metamagnetic transition. It consists of the assumption that the negative $b$ becomes possible only because of the coupling between magnetism and other degrees of freedom, a change of the crystal volume, for instance. The absence of the higher $M^6$ term in the free energy expansion observed in the magnetization curves of Y(Co,Al)$_2$ and LaCo$_9$Si$_4$ is consistent with our picture.

We are particularly concerned with the metamagnetic transition in the ground state by taking the forced magneto-volume effects into account. At finite temperature we also need to take account of the effect of thermal volume expansions. The temperature dependence of the magnetization curve and magnetic susceptibility will be the subject of our future studies including the detailed comparison with experiments as well.

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