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Metamagnetic transition and susceptibility maximum in an itinerant-electron system

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An itinerant-electron metamagnetism is discussed at finite temperature, by taking into account the effect of spin fluctuations on the Landau-Ginzburg theory. It is shown that the paramagnetic susceptibility always shows a maximum in its temperature dependence when the metamagnetic transition from the paramagnetic to the ferromagnetic state is induced by the external magnetic field at low temperature. This metamagnetic transition, associated with a hysteresis in the magnetization curve, is shown to disappear at high temperature. Moreover, the first-order transition in the temperature dependence of the spontaneous magnetization is shown to occur under a certain condition among the Landau coefficients. Three characteristic temperatures, at which the susceptibility reaches a maximum, the field-induced metamagnetic transition disappears, and the temperature-induced first-order transition of the magnetization occurs, are discussed. The present theory can explain qualitatively these anomalous magnetic properties observed in Co compounds $Co(S,Se)_2$, YCo_2, $LuCo_2$, and others.

I. INTRODUCTION

High magnetic fields of about 100 T are now being used in fundamental research on magnetism. One of them is on the metamagnetic transition (MT) induced by the external magnetic field in the d electron system. This is carried out intensively both from the theoretical and experimental points of view.[1,2] Co compounds with the cubic Laves phase structure, $ScCo_2$, YCo_2, and $LuCo_2$, are strongly exchange-enhanced paramagnets. A broad maximum in the temperature dependence of paramagnetic susceptibility $\chi_p(T)$ is observed at room temperature.[3–7] The field-induced MT from the paramagnetic to the ferromagnetic state, associated with a hysteresis in the magnetization curve, is observed at low temperature under an extremely high magnetic field of about 70 T for YCo_2 and $LuCo_2$.[8,9] The pyrite compound $Co(S,Se)_2$ also shows both the susceptibility maximum and the MT at a certain concentration of S.[10,11] These anomalous magnetic properties have theoretically been shown to relate to a sharp peak of the electronic density-of-states curve near the Fermi level.[1] It should be mentioned here that the present MT is associated with the change in the electronic structure of itinerant d electrons induced by the external magnetic field, and is distinguished from the classical one in the localized electron system. Thus, it is called the itinerant-electron metamagnetism (IEMM).

On the phenomenological Landau theory, Wohlfarth and Rhodes[12] have pointed out that the MT occurs if there exists a maximum in $\chi_p(T)$. Shimizu[13] has obtained a condition for the appearance of the MT. Here, the Wohlfarth-Rhodes-Shimizu (WRS) theory for the IEMM is briefly reviewed. The magnetic part of the free energy ΔF for the system with the magnetic moment M is written as

$$\Delta F(M)=\tfrac{1}{2}aM^2+\tfrac{1}{4}bM^4+\tfrac{1}{6}cM^6 , \tag{1}$$

where a, b, and c are the Landau expansion coefficients. By the fixed spin-moment method of the band calculation,[14] the total energy is obtained numerically as a function of M at $T=0$. Then, the values of a, b, and c for actual materials can be estimated from the first-principles calculation by this method, which is crucially discussed by Wagner.[15] Hathaway and Cullen[16] have actually estimated the values of a, b, and c for YCo_2, by using the results of the fixed spin-moment calculation of the electronic energy.[17] In the itinerant-electron system, the Landau coefficients are able to take either positive or negative values, depending on the electronic structure near the Fermi level. In fact, the values of a, b, and c estimated in the tight-binding approximation for any compounds of $ScCo_2$, YCo_2, and $LuCo_2$ are positive, negative, and positive, respectively.[1]

In the case of $a>0$, $b<0$, $c>0$, and $ac/b^2<1/4$, it can be easily seen from Eq. (1) that $\Delta F(M)$ has two minimums at $M=0$ and at a finite value of $M(=M_0)$, and a maximum between the two, which are schematically shown in Fig. 1. When $ac/b^2<3/16$, $\Delta F(M_0)$ is negative as shown by the curve (a). Then, the ferromagnetic state at $M=M_0$ is most stable without the external magnetic field. When $1/4>ac/b^2>3/16$, $\Delta F(M_0)$ is positive as shown by the curve (b). Then, the state at $M=M_0$ is metastable. However, this state can be stabilized by the external magnetic field H and the MT from the paramagnetic to the ferromagnetic state occurs at a critical field H_c. To see this fact more strictly, we discuss the magnetic equation of state for M and H given by

$$H=\frac{d}{dM}\Delta F(M)=aM+bM^3+cM^5 . \tag{2}$$

The magnetization curve $M(H)$ is obtained from Eq. (2). In the case of $a>0$, $b<0$, $c>0$, and $ac/b^2>9/20$, $M(H)$ increases monotonically with increasing H. On the other hand, when $9/20>ac/b^2>3/16$, $M(H)$ shows an anomalous curve as shown in Fig. 2. At $H_c'<H<H_c''$, $M(H)$ is a triple-valued function, where H_c' and H_c'' are defined

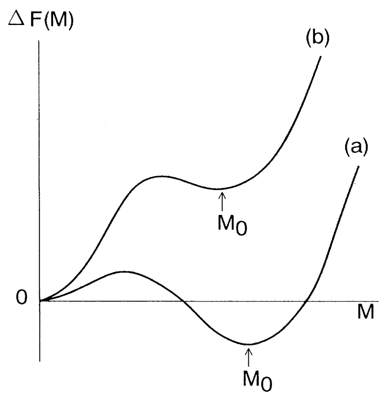

FIG. 1. Magnetic part of the free energy $\Delta F(M)$. Curves (a) and (b) show $\Delta F(M)$ for $ac/b^2 < 3/16$ and $1/4 > ac/b^2 > 3/16$, respectively.

by Shimizu,[13] as shown in the figure. That is, the hysteresis in the magnetization curve is associated with the MT. The MT occurs at a critical field H_c, where the total energies of two states of the upper and lower curves are equal to each other. Then, the condition for the appearance of the MT is given by[13]

$$a>0, b<0, c>0 \quad \text{and} \quad \frac{3}{16}<\frac{ac}{b^2}<\frac{9}{20} . \tag{3}$$

At $ac/b^2=9/20$, the equation $dH/dM=0$ has an equal solution. In this case, the hysteresis in the magnetization curve disappears. The condition of Eq. (3) has also been obtained by Moriya.[18] This is the brief review of the WRS theory for the IEMM.

On the Stoner model, the dependences on T of a, b, and c in Eq. (1) come from the Fermi distribution functions involved in their respective expressions.[19] However, the T dependences of a, b, and c are weak because the effective degenerate temperature in the Fermi distribution function is high. On the other hand, spin fluctuations (SF), which are not taken into account in the WRS theory, play an important role at finite T. It gives much stronger T dependence of $\Delta F(M)$ than a, b, and c.[20–22] In this paper, the effect on the MT of the longitudinal and transverse SF is discussed on the phenomenological

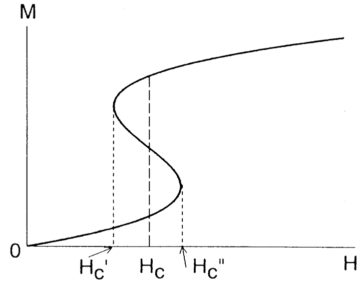

FIG. 2. Schematic curve of $M(H)$ for $9/20 > ac/b^2 > 3/16$.

Landau-Ginzburg theory by neglecting the T dependence of a, b, and c.

The present SF model for the IEMM is essentially based on the Murata and Doniach theory[22] for the weak itinerant-electron ferromagnet. A classical Gaussian approximation is made use of for the estimation of the mean amplitude of spin fluctuations, which is unable to be used for the critical phenomena near the Curie temperature.[23] However, the value of a in this paper is assumed not to depend on T but to be a positive finite value. In this point, our SF model is different from the usual Landau-Ginzburg theory for the critical phenomena. As the value of a is positive and finite at any T, the mean-field theory perturbed by the SF will be available for the present system. The SF model together with the volume fluctuations was developed by Wagner.[15] Mohn, Schwarz, and Wagner[24] have recently applied this model successfully to the calculation of the magnetoelastic anomalies in Fe-Ni Invar alloys at finite T. In this paper we apply this model to the IEMM excluding the effect of the volume fluctuations and including the terms up to the sixth power of the magnetization density in the free-energy density. In Sec. II, an expression of $\Delta F(M)$ is given by a functional of the amplitude of spatially fluctuating magnetizations. In Sec. III, the equation of state for M and H is obtained, including the effect of the SF. The conditions for the appearances of the susceptibility maximum and of the MT are discussed in Sec. IV and Sec. V, respectively. Conclusions and discussion are given in Sec. VI.

II. FREE ENERGY

The magnetic part of the free-energy density $\Delta f(r)$ is written on the Landau-Ginzburg theory as

$$\Delta f(r)=\tfrac{1}{2}a|m(r)|^2+\tfrac{1}{4}b|m(r)|^4+\tfrac{1}{6}c|m(r)|^6 + \tfrac{1}{2}D|\nabla\cdot m(r)|^2 , \tag{4}$$

where $m(r)$ is a magnetization density and a, b, c, and D are Landau-Ginzburg coefficients. By the Fourier transformation for the ith component of $m(r)$,

$$m_i(r)=M\delta_{i,z}+\frac{1}{\sqrt{V}}\sum_q m_i(q)\exp(iq\cdot r) , \tag{5}$$

the total free energy ΔF is obtained by

$$\Delta F=\frac{1}{V}\int d^3r\,\Delta f(r) , \tag{6}$$

where M and V are the bulk moment in the z direction and the volume, respectively.

The expression of ΔF is complicated and involves terms up to the sixth power with respect to $m_i(q)$. Out of them, we pick up only the terms with even power of $m_i(q)$ as[25]

$$\Delta F[M,\{|m_x(q)|^2\},\{|m_y(q)|^2\},\{|m_z(q)|^2\}]$$
$$=\frac{1}{2}aM^2+\frac{1}{4}bM^4+\frac{1}{6}cM^6+\frac{1}{2V}\sum_{i,q}\Omega_i^{(0)}(q)|m_i(q)|^2+\frac{1}{4V^2}\sum_{i,j}\sum_{q,q'}\{b+2cM^2(1+4\delta_{i,z})\}(1+2\delta_{i,j})|m_i(q)|^2|m_j(q')|^2$$
$$+\frac{c}{6V^3}\sum_{i,j,k}\sum_{q_1\sim q_3}\{15\delta_{i,j}\delta_{j,k}+9\delta_{i,j}(1-\delta_{j,k})+(1-\delta_{i,j})(1-\delta_{j,k})(1-\delta_{k,i})\}|m_i(q_1)|^2|m_j(q_2)|^2|m_k(q_3)|^2\ , \tag{7}$$

where i, j, and k denote x, y, or z and

$$\Omega_i^{(0)}(q)=Dq^2+a+(1+2\delta_{i,z})bM^2+(1+4\delta_{i,z})cM^4\ . \tag{8}$$

In this case, ΔF is taken as a functional of M and three sets of $\{|m_x(q)|^2\}$, $\{|m_y(q)|^2\}$, and $\{|m_z(q)|^2\}$.

The equation of state for the system with M and H is defined by

$$H=\left\langle\frac{\partial}{\partial M}\Delta F[M,\{|m_x(q)|^2\},\{|m_y(q)|^2\},\{|m_z(q)|^2\}]\right\rangle\ , \tag{9}$$

where $\langle\ \rangle$ denotes the thermal average discussed in the next section. Here, the following approximations are made use of:

$$\begin{aligned}&\langle[|m_i(q_1)|^2-\langle|m_i(q_1)|^2\rangle][|m_j(q_2)|^2-\langle|m_j(q_2)|^2\rangle]\rangle=0\ ,\\&\langle[|m_i(q_1)|^2-\langle|m_i(q_1)|^2\rangle][|m_j(q_2)|^2-\langle|m_j(q_2)|^2\rangle][|m_k(q_3)|^2-\langle|m_k(q_3)|^2\rangle]\rangle=0\ ,\end{aligned} \tag{10}$$

which derive

$$\begin{aligned}&\langle|m_i(q_1)|^2|m_j(q_2)|^2\rangle=\langle|m_i(q_1)|^2\rangle\langle|m_j(q_2)|^2\rangle\ ,\\&\langle|m_i(q_1)|^2|m_j(q_2)|^2|m_k(q_3)|^2\rangle=\langle|m_i(q_1)|^2\rangle\langle|m_j(q_2)|^2\rangle\langle|m_k(q_3)|^2\rangle\ .\end{aligned} \tag{11}$$

As the z axis is taken in the direction of M, x and y directions are equivalent to each other. Putting

$$\begin{aligned}&\langle|m_\parallel(q)|^2\rangle=\langle|m_z(q)|^2\rangle\ ,\\&\langle|m_\perp(q)|^2\rangle=\langle|m_x(q)|^2\rangle=\langle|m_y(q)|^2\rangle\ ,\end{aligned} \tag{12}$$

one gets

$$H=\widetilde{A}(M)M+\widetilde{B}(M)M^3+\widetilde{C}(M)M^5\ , \tag{13}$$

where

$$\begin{aligned}&\widetilde{A}(M)=a+b\{3\langle(\delta m_\parallel)^2\rangle+2\langle(\delta m_\perp)^2\rangle\}+c\{15\langle(\delta m_\parallel)^2\rangle^2+12\langle(\delta m_\parallel)^2\rangle\langle(\delta m_\perp)^2\rangle+8\langle(\delta m_\perp)^2\rangle^2\}\ ,\\&\widetilde{B}(M)=b+2c\{5\langle(\delta m_\parallel)^2\rangle+2\langle(\delta m_\perp)^2\rangle\}\ ,\\&\widetilde{C}(M)=c\ ,\end{aligned} \tag{14}$$

and

$$\langle(\delta m_i)^2\rangle=\frac{1}{V}\sum_q\langle|m_i(q)|^2\rangle\ . \tag{15}$$

The dependences on M of $\widetilde{A}$, $\widetilde{B}$, and $\widetilde{C}$ come from those of $\langle(\delta m_i)^2\rangle$, which will be discussed in the next section.

III. MAGNETIC EQUATION OF STATE

We define the energy $\Omega_i(q)$ associated with the spatially fluctuating magnetization $m_i(q)$ as

$$\Omega_i(q)=V\left\langle\frac{1}{m_i(-q)}\frac{\partial}{\partial m_i(q)}\Delta F[M,\{|m_x(q)|^2\},\{|m_y(q)|^2\},\{|m_z(q)|^2\}]\right\rangle\ . \tag{16}$$

By making use of the approximations [Eq. (11)], one gets from Eqs. (7) and (16) that

$$\Omega_{\parallel}(q)=\Omega_z(q)$$
$$=\Omega_z^{(0)}(q)+3(b+10cM^2)\langle(\delta m_{\parallel})^2\rangle+2(b+6cM^2)\langle(\delta m_{\perp})^2\rangle$$
$$+c\{15\langle(\delta m_{\parallel})^2\rangle^2+12\langle(\delta m_{\parallel})^2\rangle\langle(\delta m_{\perp})^2\rangle+8\langle(\delta m_{\perp})^2\rangle^2\}\ ,$$
$$\Omega_{\perp}(q)=\Omega_x(q)=\Omega_y(q)$$
$$=\Omega_x^{(0)}(q)+(b+6cM^2)\langle(\delta m_{\parallel})^2\rangle+4(b+2cM^2)\langle(\delta m_{\perp})^2\rangle$$
$$+c\{3\langle(\delta m_{\parallel})^2\rangle^2+8\langle(\delta m_{\parallel})^2\rangle\langle(\delta m_{\perp})^2\rangle+24\langle(\delta m_{\perp})^2\rangle^2\}\ . \quad (17)$$

By the classical Gaussian approximation,[26] $\langle(\delta m_{\parallel})^2\rangle$ and $\langle(\delta m_{\perp})^2\rangle$ are given by

$$\langle(\delta m_i)^2\rangle=\frac{1}{V}\sum_q{}' \frac{k_BT}{\Omega_i(q)}\ , \quad (18)$$

where the prime on the summation over q denotes the sum within a cutoff wave vector q_c for the fluctuating moment. Here, the difference between the values of q_c for the longitudinal and transverse fluctuations is neglected. Equation (18) is also derived by the functional integral method.[15,22]

From Eqs. (17) and (18), the simultaneous equations for $\langle(\delta m_{\parallel})^2\rangle$ and $\langle(\delta m_{\perp})^2\rangle$ are obtained. Expanding them in a power series of M^2, one gets

$$\langle(\delta m_i)^2\rangle=\tfrac{1}{3}\xi_p(T)^2+\alpha_i(T)M^2+\beta_i(T)M^4\ , \quad (19)$$

where $\xi_p(T)^2$ is the square of the amplitude of fluctuating moment at $M=0$ given by

$$\xi_p(T)^2=3k_BT\frac{N_{\mathrm{sf}}}{\hbar\omega_c}\left[1-\frac{1}{\sqrt{\hbar\omega_c\chi_p(T)}}\times\tan^{-1}\sqrt{\hbar\omega_c\chi_p(T)}\right]\ . \quad (20)$$

Here, $\chi_p(T)$, N_{sf}, and $\hbar\omega_c$ are the paramagnetic susceptibility, the number of fluctuating modes, and the cutoff energy, respectively. They are written by

$$\chi_p(T)^{-1}=a+\tfrac{5}{3}b\xi_p(T)^2+\tfrac{35}{9}c\xi_p(T)^4\ , \quad (21)$$

$$N_{\mathrm{sf}}=\frac{3}{V}\sum_q{}'=\frac{q_c^3}{2\pi^2}\ , \quad (22)$$

$$\hbar\omega_c=Dq_c^2\ . \quad (23)$$

The factor 3 in N_{sf} comes from the degree of freedom for the spatial fluctuations of magnetization. Explicit expressions of $\alpha_i(T)$ and $\beta_i(T)$ in Eq. (19) are given in the Appendix. Similar expansion of $\langle(\delta m_i)^2\rangle$ to Eq. (19) has been carried out by Wagner and Wohlfarth.[27]

Substituting Eq. (19) into Eq. (14), the magnetic equation of state given by Eq. (13) is rewritten as

$$H=A(T)M+B(T)M^3+C(T)M^5\ , \quad (24)$$

where

$$A(T)=\chi_p(T)^{-1}\ ,$$
$$B(T)=\Lambda(T)(1+3\alpha_{\parallel}+2\alpha_{\perp})\ , \quad (25)$$
$$C(T)=\Lambda(T)(3\beta_{\parallel}+2\beta_{\perp})+c\{1+10\alpha_{\parallel}+4\alpha_{\perp}+15\alpha_{\parallel}^2+12\alpha_{\parallel}\alpha_{\perp}+8\alpha_{\perp}^2\}\ ,$$

α_i and β_i mean $\alpha_i(T)$ and $\beta_i(T)$ in Eqs. (A1)–(A4) in the Appendix, respectively, and

$$\Lambda(T)=b+\tfrac{14}{3}c\xi_p(T)^2\ . \quad (26)$$

Lonzarich and Taillefer[20] and Moriya[21] have shown that $\langle(\delta m_i)^2\rangle$ can be estimated from the microscopic theory. Neglecting the zero-point fluctuations, they have obtained

$$\langle(\delta m_i)^2\rangle=\frac{4\hbar}{V}\sum_q\int_0^{\infty}\frac{d\omega}{2\pi}n(\omega)\mathrm{Im}\chi_i(q,\omega)\ , \quad (27)$$

where $n(\omega)$ is a Bose distribution function and $\chi_i(q,\omega)$ is a dynamical longitudinal or transverse susceptibility given by

$$\chi_i(q,\omega)=\chi_i(q)/[1-i\hbar\omega/\Gamma_i(q)]\ ,$$
$$\chi_i(q)^{-1}=\chi_i(0)^{-1}+Dq^2\ , \quad (28)$$
$$\Gamma_i(q)^{-1}=\gamma\chi_i(q)/q\ .$$

By the comparison between Eqs. (18) and (27), q_c in the classical theory can be obtained. The expression of q_c thus obtained is very complicated. However, at low T, q_c is approximately given by[20,21]

$$q_c=(\pi\gamma k_BT/3D)^{1/3}\ . \quad (29)$$

Here, the difference between the values of γ in $\Gamma_i(q)$ is neglected. Equation (18) has been derived by using the Gaussian approximation that can be used only in the high-temperature region. Nevertheless, we assume that the approximation of Eq. (29) at low T can still be made use of, as it is combined with Eq. (27). That is, the expression of Eq. (18) is assumed to be available even at low T as far as the value of q_c obtained from Eq. (27) is used.

At low T, q_c is proportional to $T^{1/3}$ as shown by Eq. (29). Then, in this case, N_{sf} and $\hbar\omega_c$ are proportional to T and $T^{2/3}$, respectively. Moreover, one gets

$$\xi_p(T)^2=\gamma k_B^2T^2/6\pi Da\ . \quad (30)$$

In the lowest order of T, $\alpha_i(T)$ and $\beta_i(T)$ given in the

Appendix are written as

$$\begin{aligned}\alpha_{\parallel}(T)&=-k_B T N_{sf}\Lambda(T)/a^2 ,\\ \alpha_{\perp}(T)&=-\tfrac{1}{3}k_B T N_{sf}\Lambda(T)/a^2 ,\\ \beta_{\parallel}(T)&=-\tfrac{5}{3}ck_B T N_{sf}/a^2+3k_B T N_{sf}\Lambda(T)^2/a^3 ,\\ \beta_{\perp}(T)&=-\tfrac{1}{3}ck_B T N_{sf}/a^2+\tfrac{1}{3}k_B T N_{sf}\Lambda(T)^2/a^3 .\end{aligned} \tag{31}$$

As shown in Sec. IV the temperature T_{max}, at which $\chi_p(T)$ reaches a maximum, is given by $\Lambda(T)=0$. Then $\Lambda(T)$ itself becomes small around T_{max}. By using the expressions of Eq. (31), $B(T)$ and $C(T)$ in Eq. (25) are given at low T and at small $\Lambda(T)$ as

$$\begin{aligned}B(T)&=\Lambda(T)[1-\tfrac{11}{3}k_B T N_{sf}\Lambda(T)/a^2] ,\\ C(T)&=c[1-17k_B T N_{sf}\Lambda(T)/a^2] .\end{aligned} \tag{32}$$

It is noted that $\alpha_i(T)$ is positive at low T when $\Lambda(T)<0$ at which the MT may occur. This means that, as far as $\Lambda(T)$ is negative, $\langle(\delta m_i)^2\rangle$ given by Eq. (19) increases with increasing M at small M. On the other hand, $\beta_i(T)$ is negative, when $\Lambda(T)$ is small, and then $\langle(\delta m_i)^2\rangle$ decreases at large M. At the critical field H_c of the MT, the value of M increases and then $\langle(\delta m_i)^2\rangle$ decreases discontinuously. That is, the spin fluctuations are quenched at H_c. The rapid decrease of the low-temperature specific-heat coefficient around H_c, which is observed in $Y(Co,Al)_2$,[28] will be due to the quenching of spin fluctuations. Shioda, Takahashi, and Moriya[29] have shown that the spin fluctuations are suppressed by H and the electronic specific heat is reduced.

IV. PARAMAGNETIC SUSCEPTIBILITY

From the equation of state given by Eq. (24), the differential susceptibility $\chi(T,H)$ $(=dM/dH)$ is obtained as a function of H. Expanding it with respect to H, we get in the paramagnetic state,

$$\chi(T,H)=\frac{1}{A(T)}-\frac{3B(T)}{A(T)^4}H^2+\frac{5B(T)^2}{A(T)^7}\left[3-\frac{A(T)C(T)}{B(T)^2}\right]H^4+\cdots . \tag{33}$$

As shown in Sec. V, the sign of $B(T)$ is negative and the value of $A(T)C(T)/B(T)^2$ is less than 3 when the MT occurs. In this case, the coefficients of H^2 and H^4 in Eq. (33) are both positive, then $\chi(T,H)$ increases with increasing H.

$\chi(T,0)$ at $H=0$ is equal to $\chi_p(T)$ given by Eq. (21). In the case of $a>0$, $b<0$, and $c>0$, $\chi_p(T)^{-1}$ shows a minimum at a finite value of $\xi_p(T)$. The minimum of $\chi_p(T)^{-1}$ is given by $\partial\chi_p(T)^{-1}/\partial\xi_p(T)=0$ and we get

$$\xi_p(T_{max})^2=-\frac{3}{14}\frac{b}{c} \tag{34}$$

and

$$\chi_p(T_{max})^{-1}=a-\frac{5}{28}\frac{b^2}{c} . \tag{35}$$

For $ac/b^2<5/28$, $\chi_p(T_{max})$ becomes negative. Then, ac/b^2 should be larger than 5/28 when the paramagnetic state is stable at any T. The condition for the appearance of the maximum in $\chi_p(T)$ is given by

$$a>0, b<0, c>0 \text{ and } \frac{ac}{b^2}>\frac{5}{28} . \tag{36}$$

When T_{max} is low enough, the approximation of Eq. (30) can be used for the estimation of T_{max}. One gets from Eq. (34) that

$$T_{max}^2=\frac{9}{7}\frac{|b|}{c}\frac{\pi D}{k_B^2\gamma}a . \tag{37}$$

That is, T_{max}^2 is proportional to the inverse of the enhancement factor of the susceptibility at $T=0$ as $a=\chi_p(0)^{-1}$.

It is pointed out that $B(T)$ in the equation of state given by Eq. (24) becomes zero at $T=T_{max}$. This is because $B(T)$ is proportional $\Lambda(T)$ and $\Lambda(T_{max})$ is zero as seen from Eqs. (26), (32), and (34). This means that $B(T)$ changes its sign from negative to positive at T_{max} when $a>0$, $b<0$, and $c>0$, as shown in Fig. 3(a). That is, the temperature T_B, at which $B(T)=0$, is equal to T_{max}. Then the Arrott plots (M^2 against H/M) will be observed like the curves (1), (2), and (3) shown schematically in Fig. 3(b) for $T<T_{max}$, $T=T_{max}$, and $T>T_{max}$, respectively. At $T=T_{max}$, χ_p^{-1} reaches a minimum and the coefficient $B(T)$ of M^2 in the Arrott plots becomes zero. For YCo_2, Bloch *et al.*[30] have actually found that $B(T)$ changes its sign from negative to positive at a little below 300 K, i.e., around the observed value of T_{max} (=250 K). Then, their observed result is consistent with the present theory.

However, Duc *et al.*[31] have recently pointed out that T_{max} for YCo_2 and $LuCo_2$ is higher than T_B from the analysis of their observed magnetic properties of RCo_2, where R is a heavy rare-earth element. They estimated the values of T_{max} and T_B for YCo_2 as 250 and 178 K, respectively. The difference between T_{max} and T_B may be attributed to the T dependence of a, which is neglected in the present theory. When the T dependence of a is given by

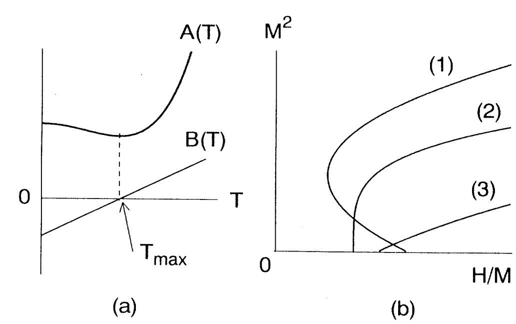

FIG. 3. Schematic curves of (a) the temperature dependences of $A(T)$ and $B(T)$ and (b) the Arrott plots. Curves (1), (2), and (3) in (b) are those for $T<T_{max}$, $T=T_{max}$, and $T>T_{max}$, respectively.

$$a(T)=a[1-(T/T_F)^2] , \tag{38}$$

we get

$$\left[\frac{T_{max}}{T_B}\right]^2=1+\frac{14}{5}\frac{ac}{b^2}\left[\frac{T_B}{T_F}\right]^2 . \tag{39}$$

For YCo_2, the value of T_F is roughly estimated as 500 K.[32] The value of ac/b^2 should be smaller than 9/20 as far as the MT occurs, as shown in Sec. V. Then the value of T_{max}/T_B for YCo_2 is shown to be smaller than 1.1. The difference between T_{max} and T_B estimated by Duc *et al.*[31] seems to be too large.

V. METAMAGNETIC TRANSITION

As mentioned in Sec. I, the condition for the appearance of the MT on the Landau theory[13] is given by Eq. (3). This result can be directly used at finite T by replacing a, b, an c in Eq. (3) with $A(T)$, $B(T)$, and $C(T)$ in Eq. (25), respectively. That is, $A(T)>0$, $B(T)<0$, $C(T)>0$, and

$$\frac{9}{20}>\frac{A(T)C(T)}{B(T)^2}>\frac{3}{16} . \tag{40}$$

When $A(T)C(T)/B(T)^2$ is smaller than 3/16, the system becomes ferromagnetic even at $H=0$, as shown by the curve (1) in Fig. 4. On the other hand, when $A(T)C(T)/B(T)^2$ is larger than 9/20, the MT does not occur at any H, as shown by the curve (3). At $A(T)C(T)/B(T)^2=9/20$, the equation $\partial H/\partial M=0$ has an equal solution. In this case the hysteresis in the magnetization curve disappears.

As shown in Sec. IV, $\Lambda(T)$ becomes zero at $T=T_{max}$. Then, $B(T)$ and $C(T)$ given by Eq. (32) at low T are written in the lowest order of $\Lambda(T)$ as

$$B(T)=\Lambda(T) \text{ and } C(T)=c . \tag{41}$$

One gets from Eq. (40) that the MT disappears at T_0 given by

$$\xi_p(T_0)^2=(|b|/c)(3/14-\sqrt{45/266}\sqrt{ac/b^2-5/28}) , \tag{42}$$

which is positive for $ac/b^2<9/20$. On the other hand, the ferromagnetic state becomes stable below T_1 given by

$$\xi_p(T_1)^2=(|b|/c)(3/14-\sqrt{36/7}\sqrt{ac/b^2-5/28}) . \tag{43}$$

For $ac/b^2<3/16$, $\xi_p(T_1)^2$ is positive and the first-order transition in the temperature dependence of the magnetization occurs at this temperature. Moriya[18] has also obtained Eq. (43) for the appearance of spontaneous magnetization.

These results of Eqs. (42) and (43) are the same as those obtained by the present author,[33] neglecting the dependence on M of the coefficients $\tilde{A}$, $\tilde{B}$, and $\tilde{C}$ in the equation of state given by Eq. (13). This means that the neglect of the dependences on M of $\tilde{A}$, $\tilde{B}$, and $\tilde{C}$ is valid when $\Lambda(T)$ is small, i.e., when $T\sim T_{max}$. When T_0 and T_1 are low enough, we get from Eqs. (30), (42), and (43)

$$T_0^2=T_{max}^2(1-\sqrt{70/19}\sqrt{ac/b^2-5/28}) ,$$
$$T_1^2=T_{max}^2(1-4\sqrt{7}\sqrt{ac/b^2-5/28}) , \tag{44}$$

where T_{max} is given by Eq. (37). It is found that

$$T_1<T_0<T_{max} . \tag{45}$$

For $5/28<ac/b^2<3/16$, the ferromagnetic state is stable at $T=0$. With increasing T, the temperature-induced first-order transition of the spontaneous magnetization occurs at $T=T_1$. And the field-induced MT occurs at $T_1<T<T_0$. At $T>T_0$, the system is paramagnetic and the susceptibility reaches a maximum at $T=T_{max}$. On the other hand, for $3/16<ac/b^2<9/20$, the system is paramagnetic at $H=0$ and the MT occurs at $T<T_0$. Above T_0, the susceptibility shows a maximum at $T=T_{max}$. For $9/20<ac/b^2$, the MT does not occur at any H and the susceptibility shows a maximum at $T=T_{max}$ as far as $a>0$, $b<0$, and $c>0$.

As seen from Eq. (44), the values of T_0/T_{max} and T_1/T_{max} are given only by a quantity ac/b^2. In Fig. 5 the calculated values of T_0/T_{max} and T_1/T_{max} are shown as a function of ac/b^2. The values of ac/b^2 can be estimated from the calculated result shown in Fig. 5 and from the observed values of T_0/T_{max} and T_1/T_{max} for YCo_2,[2] $Lu(Co,Al)_2$,[34] $(Y,Lu)(Co,Al)_2$,[35] and

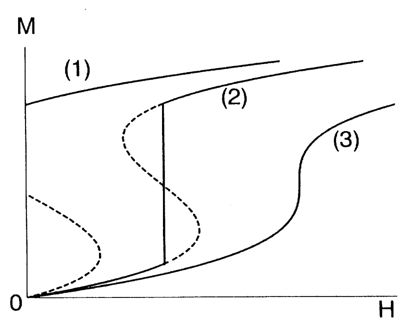

FIG. 4. Schematic curves of $M(H)$. Curves (1), (2), and (3) are those for $A(T)C(T)/B(T)^2=3/16$, $3/16<A(T)C(T)/B(T)^2<9/20$, and $A(T)C(T)/B(T)^2=9/20$, respectively.

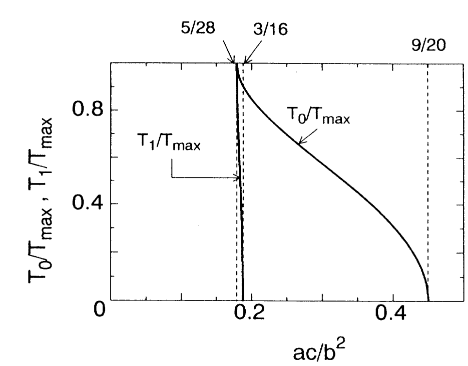

FIG. 5. Calculated values of T_0/T_{max} and T_1/T_{max} as a function of ac/b^2.

$Co(S_xSe_{1-x})_2$,[10] at $x=0.88$ and 0.9. In Table I, the observed values of T_0, T_1, and T_{max} and the estimated values of ac/b^2 are shown. The observed datum of T_0/T_{max} for $Co(S_{0.9}Se_{0.1})_2$ is very close to 1, which is consistent with our result as the value of ac/b^2 estimated from the observed value of T_1/T_{max} is close to 5/28, at which $T_0/T_{max}=1$.

The value of ac/b^2 can also be estimated from the observed values of $\chi_p(T)$ at $T=0$ and $T=T_{max}$. From Eq. (35), one gets

$$\frac{ac}{b^2}=\frac{5}{28}\left[1-\frac{\chi_p(0)}{\chi_p(T_{max})}\right]^{-1}, \tag{46}$$

where $\chi_p(0)=1/a$. From the observed data of $\chi_p(T_{max})$ and $\chi_p(0)$ for YCo_2,[4] $LuCo_2$,[6] and $ScCo_2$,[3] the values of ac/b^2 are estimated to be 0.40, 0.39, and 0.41, respectively, which satisfy the condition given by Eq. (3) for the appearance of the MT. In particular, the estimated value of ac/b^2 for YCo_2 is very close to that shown in Table I. It is noted that the values of ac/b^2 for YCo_2 estimated independently from the observed value of T_0/T_{max} and from that of $\chi_p(0)/\chi_p(T_{max})$ coincide with each other. This means that the present theory is reliable for the IEMM at finite temperature.

The values of T_0 for $LuCo_2$ and $ScCo_2$ have not been observed so far. However, by using the values of ac/b^2 estimated above and the observed values of $T_{max}=350$ and 600 K for $LuCo_2$ and $ScCo_2$, the values of T_0 are predicted as 120 and 170 K, respectively. The observation of T_0 is desirable for these compounds. It is pointed out that the observed values of $\chi_p(0)$ and $\chi_p(T_{max})$ depend strongly on H for the materials with a low critical field of the MT. Therefore, the bare data of $\chi_p(0)$ and $\chi_p(T_{max})$ at finite H for the Co compounds listed in Table I except YCo_2 are not available for the estimation of ac/b^2. For the materials with the low critical field χ_p should be estimated by the extrapolation of the Arrott plots onto the form of

$$H/M=A(T)+B(T)M^2+C(T)M^4$$

to eliminate the effect of the magnetic impurities or some secondary effects.

Finally, we will discuss the critical field H_c of the MT at finite T. The value of H_c is, strictly speaking, obtained by the comparison between the free energies in the ferromagnetic and paramagnetic states under the applied magnetic field. However, this cannot be carried out analytically. In the limiting case of $ac/b^2=3/16$ where $H_c=0$ at $T=0$, we get $H_c(T)$ in the lowest order of T as

$$H_c(T)=\frac{1}{2}\sqrt{|b|/3c}\left[a-\frac{3}{16}\frac{b^2}{c}\right]+\frac{1}{16}\sqrt{|b|/3c}\left[\frac{|b|\gamma k_B^2}{6\pi Da}\right]T^2. \tag{47}$$

As the coefficient of T^2 is positive, $H_c(T)$ is found to increase with increasing T as T^2. This is consistent qualitatively with the observed results for $Co(S,Se)_2$, YCo_2, $Y(Co,Al)_2$, $Lu(Co,Al)_2$, and $(Y,Lu)(Co,Al)_2$.[2,10,34,35]

TABLE I. Observed data of T_{max}, T_0, and T_1 and estimated values of ac/b^2. The value of ac/b^2 for $Co(S_{0.90}Se_{0.1})_2$ is estimated from the observed value of T_1/T_0.

	T_{max} (K)	T_0 (K)	T_1 (K)	ac/b^2
YCo_2	250	80		0.397
$Lu(Co_{0.94}Al_{0.06})_2$	$\lesssim$ 150	65		0.358
$(Y,Lu)(Co_{0.915}Al_{0.085})_2$	100	~90		0.188
$Co(S_{0.90}Se_{0.10})_2$	$\gtrsim$ 70	70	45	0.182
$Co(S_{0.88}Se_{0.12})_2$	80	60		0.231

VI. SUMMARY AND DISCUSSION

In this paper, the IEMM has been discussed at finite temperature. It has been shown on the Landau-Ginzburg theory that the spin fluctuations play an essential role in the susceptibility maximum, the field-induced MT at finite temperature and temperature-induced first-order transition of the spontaneous magnetization. The characteristics of the IEMM are summarized as follows. (i) The field-induced MT occurs from the paramagnetic to the ferromagnetic state, associated with a hysteresis in the magnetization curve, (ii) The critical field H_c of the MT increases with increasing T as T^2, (iii) The hysteresis in the magnetization curve, i.e., the MT disappears at a certain temperature T_0, (iv) $\chi(T)$ reaches a maximum at T_{max} which is higher than T_0, and (v) The temperature-induced first-order transition of M occurs at T_1, which is lower than T_0. In this case, the field-induced MT occurs at the temperature between T_1 and T_0. These characteristics of the IEMM were observed in Co compounds ACo_2 ($A=$Sc, Y, and Lu), $A(Co,Al)_2$, $(Y,Lu)(Co,Al)_2$, and $Co(S,Se)_2$ as mentioned in this paper and have been explained qualitatively by the present theory.

Finally, we discuss a linear relation between the values of $H_c(0)$ and T_{max}, which was first pointed out by Ishiyama[36] from the observed results for $Sc(Co,Al)_2$, $Y(Co,Al)_2$, and $Lu(Co,Al)_2$. From Eqs. (34), (35), and (47), $H_c(0)$ can be rewritten as

$$H_c(0)=\sqrt{7/18}\,\frac{\xi_p(T_{max})}{\chi_p(T_{max})}\left[\frac{21}{20}-\frac{1}{20}\frac{\chi_p(T_{max})}{\chi_p(0)}\right]. \tag{48}$$

As $\xi_p(T_{max})$ is proportional to T_{max} when T_{max} is low, the linear relation between $H_c(0)$ and T_{max} is obtained. Sakakibara *et al.*[2] have plotted the observed values of $H_c(0)$ against T_{max} for various kinds of compounds including the heavy Fermion system and found that the ratio between $H_c(0)$ and T_{max} seems not to depend on the material but to be a universal constant. However, in the present theory, the ratio between $H_c(0)$ and T_{max} is not a universal constant but depends on the material as shown in Eq. (48). It should be noted that the observed data of

$H_c(0)/T_{max}$ for $Co(S,Se)_2$ (Ref. 10) and $(Y,Lu)(Co,Al)_2$ (Ref. 35) are not actually the universal value given by Sakakibara *et al.*[2] Therefore, it is not concluded that there exists the universality between $H_c(0)$ and T_{max}.

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APPENDIX

The explicit expressions of $\alpha_\parallel(T)$, $\alpha_\perp(T)$, $\beta_\parallel(T)$, and $\beta_\perp(T)$ in Eq. (19) are given by

$$\alpha_\parallel(T)=-\frac{3Z+10Z^2}{(1+2Z)(1+5Z)}, \tag{A1}$$

$$\alpha_\perp(T)=-\frac{Z}{(1+2Z)(1+5Z)}, \tag{A2}$$

$$\beta_\parallel(T)=\frac{1}{(1+2Z)(1+5Z)}\left\{-k_BTF_2c\left[5+18Z-\frac{2Z}{(1+2Z)(1+5Z)}(51+328Z+540Z^2)+\frac{Z^2}{(1+2Z)^2(1+5Z)^2}(179+1586Z+5060Z^2+5400Z^3)\right]+\frac{k_BT\Lambda(T)^2F_3}{(1+2Z)^2(1+5Z)^2}(9+94Z+340Z^2+400Z^3)\right\}, \tag{A3}$$

$$\beta_\perp(T)=\frac{1}{(1+2Z)(1+5Z)}\left\{-k_BTF_2c\left[1-2Z-\frac{2Z}{(1+2Z)(1+5Z)}(13+18Z-60Z^2)+\frac{3Z^2}{(1+2Z)^2(1+5Z)^2}(25+102Z+20Z^2-200Z^3)\right]+\frac{k_BT\Lambda(T)^2F_3}{(1+2Z)^2(1+5Z)^2}(1-6Z-60Z^2-100Z^3)\right\}, \tag{A4}$$

where

$$Z=k_BTF_2\Lambda(T), \tag{A5}$$

$$F_2=\frac{N_{sf}\chi_p(T)}{2\hbar\omega_c}\left\{\frac{1}{\sqrt{\hbar\omega_c\chi_p(T)}}\tan^{-1}\sqrt{\hbar\omega_c\chi_p(T)}-\frac{1}{1+\hbar\omega_c\chi_p(T)}\right\}, \tag{A6}$$

$$F_3=\frac{N_{sf}\chi_p(T)^2}{8\hbar\omega_c}\left\{\frac{1}{\sqrt{\hbar\omega_c\chi_p(T)}}\tan^{-1}\sqrt{\hbar\omega_c\chi_p(T)}+\frac{1}{1+\hbar\omega_c\chi_p(T)}-\frac{2}{[1+\hbar\omega_c\chi_p(T)]^2}\right\}. \tag{A7}$$

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