

# Itinerant-electron metamagnetism and strong pressure dependence of the Curie temperature

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A temperature dependence of the Landau coefficients for an itinerant-electron metamagnet is discussed on the spin fluctuation model, by taking the magnetovolume effect into account. Both of the first- and second-order transitions at the Curie temperature  $T_C$  are possible to take place for an itinerant-electron ferromagnet with a negative mode-mode coupling among spin fluctuations. It is shown that  $T_C$  depends strongly on the pressure near the boundary between the first- and second-order transitions. On the other hand, the spontaneous magnetization at  $T=0$  does not depend so much on the pressure. These results are consistent with the observed results for the pyrite compound  $\text{CoS}_2$ , the Laves phase compound  $\text{Lu}(\text{Co,Al})_2$  and other ferromagnets which show a metamagnetic transition in the paramagnetic phase.

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## I. INTRODUCTION

Two peculiar phenomena in itinerant-electron metamagnetism (IEM), i.e., a field induced first-order phase transition from the paramagnetic to the ferromagnetic state at low temperature and a maximum in the temperature dependence of susceptibility at high temperature are the consequence of the special feature of the density of states near the Fermi level. This is well known to be described by the Landau-type expression for the free energy  $\Delta E(M)$  written as

$$\Delta E(M) = \frac{1}{2}aM^2 + \frac{1}{4}bM^4 + \frac{1}{6}cM^6, \quad (1.1)$$

where  $M$  is the magnetic moment. The Landau coefficients  $a$ ,  $b$ , and  $c$  are conventionally written in terms of the density of states and its derivatives with respect to the energy at the Fermi level. Wohlfarth and Rhodes<sup>1</sup> considered the IEM only up to  $M^4$  term in Eq. (1.1), whereas Shimizu<sup>2</sup> addressed it by including  $M^6$  term.

An equation-of-state for  $M$  and the magnetic field  $B$  is given by

$$B = aM + bM^3 + cM^5. \quad (1.2)$$

The Landau coefficients  $a$ ,  $b$ , and  $c$  in Eq. (1.2) are renormalized at finite temperature  $T$  by spin fluctuations as<sup>3,4</sup>

$$\begin{aligned} a(T) &= a_0 + \frac{5}{3}b_0\xi_T(T)^2 + \frac{35}{9}c_0\xi_T(T)^4, \\ b(T) &= b_0 + \frac{14}{3}c_0\xi_T(T)^2, \\ c(T) &= c_0, \end{aligned} \quad (1.3)$$

where  $\xi_T(T)^2$  is the mean square amplitude of *thermal* spin fluctuations. The coefficients  $a_0$ ,  $b_0$ , and  $c_0$  in the right-hand side of Eq. (1.3) are those renormalized by the *zero-point* spin fluctuations as<sup>5</sup>

$$\begin{aligned} a_0 &= a + \frac{5}{3}b\xi_0^2 + \frac{35}{9}c\xi_0^4, \\ b_0 &= b + \frac{14}{3}c\xi_0^2, \\ c_0 &= c, \end{aligned} \quad (1.4)$$

where  $\xi_0^2$  is the square amplitude of the zero-point spin fluctuations and  $a$ ,  $b$ , and  $c$  are Landau-Ginzburg coefficients defined in Eq. (4) of Ref. 4. In this way, the effect of zero-point spin fluctuations can be included in the previous theory<sup>4</sup> when the temperature dependence of  $\xi_0^2$  is neglected. The temperature dependence of  $\xi_T(T)^2$  is much stronger than that of  $\xi_0^2$ , as the fluctuation-dissipation theorem gives a Bose distribution function in the expression of  $\xi_T(T)^2$ .<sup>6</sup> Then, the neglect of the temperature dependence of  $\xi_0^2$  is a rather reasonable assumption.

It can be shown<sup>4</sup> for  $a_0 > 0$ ,  $b_0 < 0$  and  $c_0 > 0$  that the inverse of the spin susceptibility  $A(T)$  shows a maximum at  $T_{\max}$  given by  $\xi_T(T_{\max})^2 = 3|b_0|/14c_0$ , as  $\xi_T(T)^2$  is a monotonic increasing function of  $T$ .<sup>6,7</sup>  $b_0$  is the mode-mode coupling constant among thermal spin fluctuations. On the other hand,  $b(T)$  increases with increasing  $T$ . One gets  $b(T_{\max}) = 0$ . That is,  $b(T)$  should change its sign from negative to positive at  $T_{\max}$ .

It has been shown that the ferromagnetic state is stable when  $a(T) > 0$ ,  $b(T) < 0$ ,  $c(T) > 0$ , and  $a(T)c(T)/b(T)^2 < 3/16$ .<sup>3,4</sup> For  $a_0c_0/b_0^2 < 5/28$ , the Curie temperature  $T_C$ , where the second-order phase transition takes place, is given by

$$\xi_T(T_C)^2/\xi_T(T_{\max})^2 = 1 + 2\sqrt{7/5}\sqrt{5/28 - a_0c_0/b_0^2}. \quad (1.5)$$

A first-order transition occurs at the Curie temperature  $T_1$  when  $5/28 < a_0c_0/b_0^2 < 3/16$ , where  $T_1$  is given by

$$\xi_T(T_1)^2/\xi_T(T_{\max})^2 = 1 - 4\sqrt{7}\sqrt{a_0c_0/b_0^2 - 5/28}. \quad (1.6)$$

In this case, the MT takes place at  $T_1 < T < T_0$ , where  $T_0$  is given by

$$\xi_T(T_0)^2/\xi_T(T_{\max})^2 = 1 - \sqrt{70/19}\sqrt{a_0c_0/b_0^2 - 5/28}. \quad (1.7)$$

For  $3/16 < a_0c_0/b_0^2 < 9/20$ , the ferromagnetic state is not stable even at  $T=0$ , but the MT takes place at lower temperatures than  $T_0$ . These results based on the theory with the negative mode-mode coupling among spin fluctuations give a qualitative explanation of the observed results not only for Laves phase compounds  $\text{YCo}_2$ ,  $\text{LuCo}_2$  and pyrite compounds  $\text{CoS}_2$ ,  $\text{Co}(\text{S,Se})_2$  but also for  $\text{La}(\text{Fe,Si})_{13}$  and the itinerant  $5f$ -electron system  $\text{UCoAl}$ .<sup>8-11</sup> Moreover, it gives a universal linear relation between the critical field of the MT and the inverse of the susceptibility maximum for Co-based compounds.<sup>12</sup> The details of these results are given in our review article.<sup>13</sup>

Recently, magnetic phase diagrams for these compounds have been determined from the observed results under high pressures and high magnetic fields. The pyrite compound  $\text{CoS}_2$  shows a second-order ferromagnetic phase transition at  $T_C$  under low pressures. At higher pressures it shows the first-order transition. Above  $T_1$  the MT with the hysteresis has actually been observed.<sup>9</sup> Similar results were obtained for  $\text{La}(\text{Fe,Si})_{13}$ ,  $\text{Lu}(\text{Co,Ga})_2$ , and for  $\text{MnSi}$  at high pressure.<sup>10,14-16</sup> As a common property among these compounds, a strong pressure dependence of the Curie temperature is observed near the boundary between the first- and second-order transitions, while the spontaneous magnetization at low temperature does not depend so much on the pressure. In Sec. II, a survey of experimental results is given for these compounds which show the MT in the paramag-

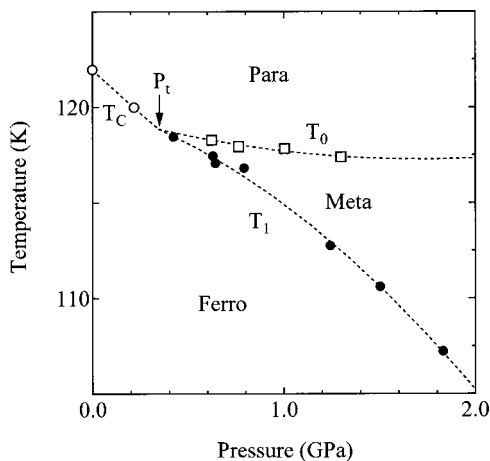


FIG. 1. Observed phase diagram of  $\text{CoS}_2$  in Ref. 9. Open and closed circles and open squares denote the observed  $T_C$ ,  $T_1$ , and  $T_0$ , respectively.

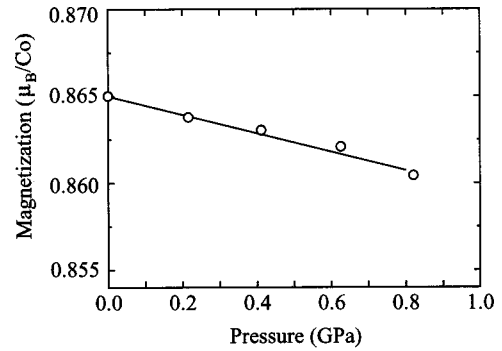


FIG. 2. Pressure dependence of the spontaneous magnetization for  $\text{CoS}_2$  observed at 4.2 K.

netic phase. In Sec. III the magnetic phase diagram is discussed theoretically on the spin fluctuation model, by taking into account the magnetovolume effect. In Sec. IV our conclusion and discussion are given.

## II. SURVEY OF OBSERVED RESULTS

A magnetic phase diagram for the pressure and temperature estimated from the observed results<sup>9</sup> for the pyrite compound  $\text{CoS}_2$  is shown in Fig. 1.  $P_t$  denotes the pressure of the triple point, where  $T_C = T_1 = T_0$ . Under the pressure less than  $P_t$  the second-order transition is observed at  $T_C$ . The first-order transition is observed at  $P > P_t$ . The temperature  $T_0$ , where the MT disappears, is also observed at  $P > P_t$ . The value of  $dT_C/dP$  shown by the dotted  $T_C$  line is  $-9.0$  K/GPa. Figure 2 denotes the pressure dependence of the spontaneous moment  $M_s$  for  $\text{CoS}_2$  observed at 4.2 K. The value of  $dM_s/dP$  is  $-0.5 \times 10^{-2} \mu_B/\text{Co GPa}$ .

Figure 3 shows the magnetic phase diagram<sup>9</sup> observed for  $\text{Co}(\text{S}_x\text{Se}_{1-x})_2$ .  $T_t$  denotes the temperature at the triple point, where  $T_C = T_1 = T_0$ . It can be seen that the observed values of  $T_{\max}$  shown by the closed circles are smaller than  $T_t$ . Figures 4 and 5 denote the pressure dependence of the Curie temperature  $T_1$  and  $M_s$  at 4.2 K. The open and closed circles

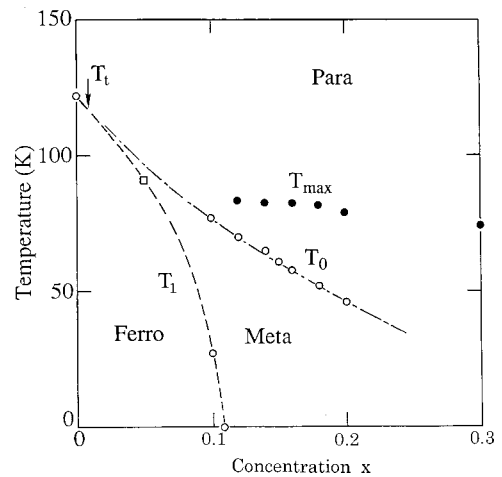


FIG. 3. Observed magnetic phase diagram of  $\text{Co}(\text{S}_{1-x}\text{Se}_x)_2$  in Ref. 9. Open and closed circles are observed values of  $T_1$ ,  $T_0$ , and  $T_{\max}$ . Open square is the observed  $T_1$  in Ref. 17.

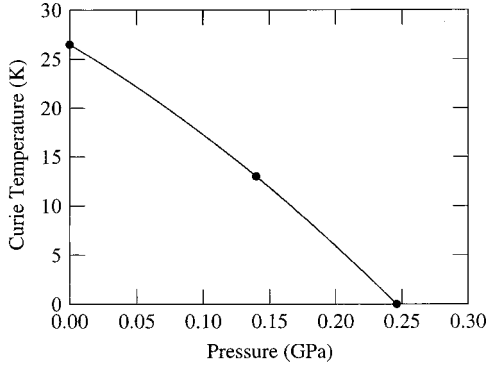


FIG. 4. Pressure dependence of the Curie temperature observed for  $\text{Co}(\text{S}_{0.9}\text{Se}_{0.1})_2$ .

in Fig. 5 indicate the observed  $M_s$  at low magnetic fields and the induced moments after the MT takes place. The values of  $dT_C/dP$  and  $dM_s/dP$  are  $-81.5$  K/GPa and  $-0.71 \times 10^{-2} \mu_B/\text{Co}$  GPa at  $P=0$ , respectively.

In Table I, the observed values of  $M_s$ ,  $-d \ln M_s/dP$ ,  $T_C$ , and  $-d \ln T_C/dP$  at  $P=0$  for Laves phase compounds  $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$  and  $\text{Lu}(\text{Co}_{1-x}\text{Ga}_x)_2$  are shown for the concentration  $x$  near the critical boundary of the onset of ferromagnetism  $x_C \sim 0.092$  and  $0.095$  for  $\text{Lu}(\text{Co},\text{Al})_2$  and  $\text{Lu}(\text{Co},\text{Ga})_2$ , respectively.<sup>14,18</sup> For  $x < x_C$  the ground state is paramagnetic. The MT is observed in this concentration range. For  $x > x_C$  the ferromagnetic state becomes stable. The first-order transition was observed near  $x_C$  and the second-order transition was observed at more Al-rich region. It is clearly seen in Table I that the value of  $-d \ln T_C/dP$  increases strongly with decreasing  $x$ , while the value of  $d \ln M_s/dP$  does not depend so much on  $x$ . For MnSi, similar pressure dependences of  $T_C$  and  $M_s$  were also observed under high pressure.<sup>15,16</sup> The Curie temperature decreases most strongly by applying pressure near the boundary between the first- and second-order transitions. These pressure dependences of  $T_C$  and  $M_s$  are considered to be characteristics of itinerant-electron ferromagnets which show the MT in the paramagnetic phase.

### III. THEORY

As discussed in Refs. 19–21, the magnetoelastic free energy density is written with the magnetization density  $\mathbf{m}(\mathbf{r})$  as

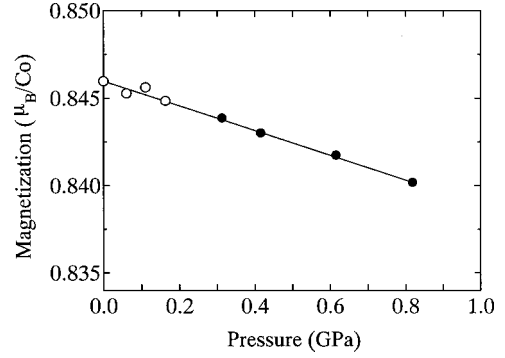


FIG. 5. Pressure dependence of the spontaneous moment for  $\text{Co}(\text{S}_{0.9}\text{Se}_{0.1})_2$  observed at 4.2 K, which is shown by open circles. Closed circles are the induced moments after the MT takes place.

$$\Delta f_{\text{mv}}(\mathbf{r}) = -C_{\text{mv}}\omega|\mathbf{m}(\mathbf{r})|^2, \quad (3.1)$$

where  $\omega = (V - V_0)/V_0$ ,  $V_0$  the volume at  $M=0$  without spin fluctuations,  $C_{\text{mv}}$  the magnetovolume coupling constant. The elastic energy is, on the other hand, written by  $\omega^2/2\kappa$ , where  $\kappa$  is the compressibility. With these energies the free energy  $\Delta F$  is written as a functional of  $M$ ,  $\omega$  and  $\{|\mathbf{m}(\mathbf{q})|^2\}$ , where  $\mathbf{m}(\mathbf{q})$  is a Fourier component of  $\mathbf{m}(\mathbf{r})$ . The pressure  $P$  and the magnetic field  $B$  are given by<sup>4</sup>

$$P = - \left\langle \frac{\partial}{\partial \omega} \Delta F[M, \omega, \{|\mathbf{m}(\mathbf{q})|^2\}] \right\rangle, \quad (3.2)$$

$$B = \left\langle \frac{\partial}{\partial M} \Delta F[M, \omega, \{|\mathbf{m}(\mathbf{q})|^2\}] \right\rangle, \quad (3.3)$$

where  $\langle \dots \rangle$  is a thermal average. From Eqs. (3.2) and (3.3) one gets

$$P = -\omega/\kappa + C_{\text{mv}}\{M^2 + \xi_0^2 + \xi_T(T)^2\}, \quad (3.4)$$

$$B = a(T)M + b(T)M^3 + c(T)M^5 - 2C_{\text{mv}}\omega M, \quad (3.5)$$

where  $a(T)$ ,  $b(T)$ , and  $c(T)$  are given in Eq. (1.3). Inserting Eq. (3.4) into Eq. (3.5) one gets a magnetic equation-of-state as

$$B = \{\tilde{a}(T) + 2\kappa C_{\text{mv}}P\}M + \tilde{b}(T)M^3 + \tilde{c}(T)M^5, \quad (3.6)$$

where

TABLE I. Observed results of  $M_s$ ,  $-d \ln M_s/dP$ ,  $T_C$ , and  $-d \ln T_C/dP$  at  $P=0$  for  $\text{Lu}(\text{Co}_{1-x}M_x)_2$  ( $M = \text{Al}$  and  $\text{Ga}$ ).

$\text{Lu}(\text{Co}_{1-x}M_x)_2$	Al				Ga				
	$x$	$M_s$ ( $\mu_B/\text{Co}$ )	$-d \ln M_s/dP$ ( $\text{GPa}^{-1}$ )	$T_C$ (K)	$-d \ln T_C/dP$ ( $\text{GPa}^{-1}$ )	$M_s$ ( $\mu_B/\text{Co}$ )	$-d \ln M_s/dP$ ( $\text{GPa}^{-1}$ )	$T_C$ (K)	$-d \ln T_C/dP$ ( $\text{GPa}^{-1}$ )
0.11						0.64	0.13	85	1.06
0.12	0.66		0.10	112	0.53	0.68	0.09	101	0.66
0.13						0.64	0.08	110	0.44
0.15	0.56		0.07	132	0.23	0.59	0.10	126	0.25

$$\begin{aligned}\tilde{a}(T) &= a(T) - 2\kappa C_{\text{mv}}^2 \{\xi_0^2 + \xi_T(T)^2\}, \\ \tilde{b}(T) &= b(T) - 2\kappa C_{\text{mv}}^2, \\ \tilde{c}(T) &= c(T).\end{aligned}\quad (3.7)$$

Similar expressions for  $\tilde{a}(T)$ ,  $\tilde{b}(T)$ , and  $\tilde{c}(T)$  without  $\xi_0^2$  have been obtained by Yamada.<sup>22</sup> It should be noted here that the estimated values of the Landau coefficients from the observed magnetization curves are not  $a(T)$ ,  $b(T)$ , and  $c(T)$  given by Eq. (1.3), but  $\tilde{a}(T)$ ,  $\tilde{b}(T)$ , and  $\tilde{c}(T)$  in Eq. (3.7) where the magnetovolume effect is taken into account.

The Landau coefficients estimated from the observed magnetization curves at  $T=0$  are those in Eq. (3.7) with  $\xi_T(0)^2=0$ . By using Eq. (1.3), Eq. (3.7) is rewritten as

$$\begin{aligned}\tilde{a}(T) &= \tilde{a}(0) + \left\{ \frac{5}{3}\tilde{b}(0) + \frac{4}{3}\kappa C_{\text{mv}}^2 \right\} \xi_T(T)^2 + \frac{35}{9}\tilde{c}(0)\xi_T(T)^4, \\ \tilde{b}(T) &= \tilde{b}(0) + \frac{14}{3}\tilde{c}(0)\xi_T(T)^2, \\ \tilde{c}(T) &= \tilde{c}(0).\end{aligned}\quad (3.8)$$

In this case,  $\xi_T(T_{\text{max}})^2$  is given by

$$\xi_T(T_{\text{max}})^2 = \frac{3}{14} \frac{|\tilde{b}(0)|}{\tilde{c}(0)} \left\{ 1 - \frac{14}{5}\eta \right\}, \quad (3.9)$$

where

$$\eta = \frac{2}{7} \kappa C_{\text{mv}}^2 |\tilde{b}(0)|. \quad (3.10)$$

On the other hand, the temperature  $T_b$ , where  $\tilde{b}(T)$  becomes zero, is given by

$$\xi_T(T_b)^2 = \frac{3}{14} \frac{|\tilde{b}(0)|}{\tilde{c}(0)}. \quad (3.11)$$

From Eqs. (3.9) and (3.11), it is found that  $T_{\text{max}}$  is lower than  $T_b$ . The Landau coefficient  $\tilde{b}(T)$  at  $T=T_{\text{max}}$  is written as

$$\tilde{b}(T_{\text{max}}) = -\frac{4}{5} \kappa C_{\text{mv}}^2. \quad (3.12)$$

That is,  $\tilde{b}(T)$  is still negative at  $T_{\text{max}}$  by the magnetovolume effects. This is consistent with our observed result for a typical itinerant-electron metamagnet  $\text{Y}(\text{Co}_{0.92}\text{Al}_{0.08})_2$ . The temperature dependences of the Landau coefficients  $\tilde{a}(T)$ ,  $\tilde{b}(T)$ , and  $\tilde{c}(T)$  are determined by fitting the observed magnetization curves<sup>23</sup> to the form of Eq. (3.6), which are shown in Fig. 6. In the analysis of the magnetization curve, the higher-order terms than  $M^5$  in Eq. (3.6) are neglected. The estimated temperature dependence of  $\tilde{c}(T)$  suggests that the higher-order terms are not negligible. However, from these estimations we can get some information about  $\tilde{a}(T)$  and  $\tilde{b}(T)$ . That is, a broad minimum of the inverse of suscepti-

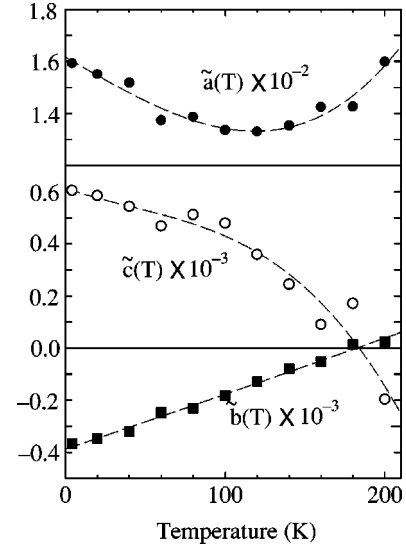


FIG. 6. Temperature dependences of the Landau coefficients  $\tilde{a}(T)$ ,  $\tilde{b}(T)$ , and  $\tilde{c}(T)$  for  $\text{Y}(\text{Co}_{0.92}\text{Al}_{0.08})_2$ , in the units of  $T/(\mu_B/\text{Co})$ ,  $T/(\mu_B/\text{Co})^3$  and  $T/(\mu_B/\text{Co})^5$ , respectively.

bility  $\tilde{a}(T)$  is seen at  $T_{\text{max}} \sim 120$  K, where  $\tilde{b}(T_{\text{max}})$  is still negative. The value of  $T_b$  is about 180 K. The difference between  $T_{\text{max}}$  and  $T_b$  is rather large. This is due to the strong magnetovolume coupling in this compound, as observed by Goto and Bartashevich.<sup>24</sup>

The ferromagnetic state is stabilized in the present system at  $B=P=0$  when the following conditions are satisfied:<sup>3,4</sup>

$$\tilde{a}(T) > 0, \quad \tilde{b}(T) < 0, \quad \tilde{c}(T) > 0, \quad \tilde{a}(T)\tilde{c}(T)/\tilde{b}(T)^2 < 3/16. \quad (3.13)$$

The second-order transition at  $T_C$  takes place when  $\tilde{a}(T_C) = 0$  and  $\tilde{b}(T_C) > 0$ . In the case of  $\tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 < 5/28 - \eta$ ,  $\xi_T(T_C)^2$  is given by

$$\begin{aligned}\xi_T(T_C)^2/\xi_T(T_b)^2 &= 1 - \frac{14}{5}\eta + 2\sqrt{\frac{7}{5}} \left\{ \frac{5}{28} - \eta \right. \\ &\quad \left. - \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 + \frac{7}{5}\eta^2 \right\}^{1/2},\end{aligned}\quad (3.14)$$

where  $\eta$  is given by Eq. (3.10). When  $5/28 - \eta < \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 < 3/16$ , the first-order transition takes place at  $T_1$  given by

$$\begin{aligned}\xi_T(T_1)^2/\xi_T(T_b)^2 &= 1 + 56\eta - 4\sqrt{7} \left\{ \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 - \frac{5}{28} \right. \\ &\quad \left. + \eta + 28\eta^2 \right\}^{1/2}.\end{aligned}\quad (3.15)$$

In this case, the MT occurs above  $T_1$ . When  $3/16 < \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 < 9/20$  the ferromagnetic state is not stable even at  $T=0$  but the MT takes place. The MT disappears at  $T_0$  given for  $5/28 - \eta < \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 < 9/20$  by

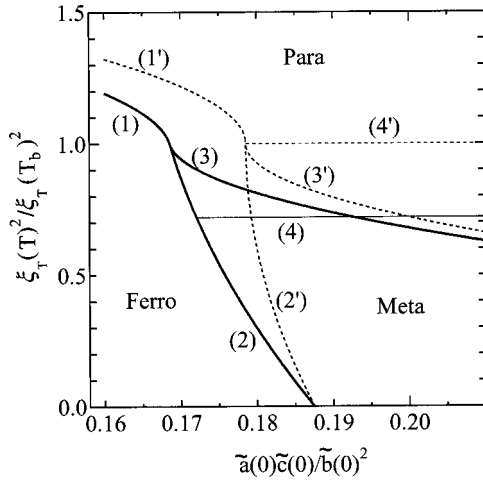


FIG. 7. Theoretical magnetic phase diagram for  $\tilde{a}(0) > 0$ ,  $\tilde{b}(0) < 0$ , and  $\tilde{c}(0) > 0$ . Curves (1), (2), (3), and (4) show  $\xi_T(T_C)^2$ ,  $\xi_T(T_1)^2$ ,  $\xi_T(T_0)^2$ , and  $\xi_T(T_{\max})^2$  reduced by  $\xi_T(T_b)^2$ , respectively, for  $\eta = 0.01$ . Dotted curves are those for  $\eta = 0$ .

$$\xi_T(T_0)^2 / \xi_T(T_b)^2 = 1 + \frac{35}{19} \eta - \sqrt{\frac{70}{19} \left[ \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 - \frac{5}{28} \right] + \eta + \frac{35}{38} \eta^2}^{1/2}. \quad (3.16)$$

The magnetic phase diagram is obtained from Eqs. (3.14), (3.15), and (3.16), as shown in Fig. 7. Here, the value of  $\eta$  is taken to be 0.01, which has recently been estimated from the observed results for  $\text{Lu}(\text{Co,Al})_2$  and  $\text{Lu}(\text{Co,Ga})_2$ .<sup>25</sup> As  $\xi_T(T)^2$  is a monotonic increasing function of  $T$ , the vertical axis corresponds to the temperature. On the other hand, the horizontal axis corresponds to the pressure as  $P$  is included only in the coefficient of  $M$  in the equation-of-state (3.6). The curves (1) and (2) denote  $\xi_T(T_C)^2$  and  $\xi_T(T_1)^2$ , where the second- and first-order transitions take place, respectively. The curves (3) and (4) denote  $\xi_T(T_0)^2$  and  $\xi_T(T_{\max})^2$ , where the MT disappears and the susceptibility reaches a maximum. The dotted curves (1'), (2'), (3'), and (4') are those for  $\eta = 0$ , respectively. It can be seen that the region of  $\tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2$ , where the first-order transition takes place, becomes wider than that without magnetovolume effects. In the very narrow range  $5/28 - \eta < \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 < 5/28 - \eta + 287\eta^2/100$ , the susceptibility does not show a maximum above  $T_1$ . The observed values of  $T_{\max}$  for  $\text{Co}(\text{S}_{1-x}\text{Se}_x)_2$  are smaller than  $T_1$ , as shown in Fig. 3. A small region of  $x$ , where the susceptibility does not show a maximum in its temperature dependence, seems to exist near the triple point even if the MT takes place above  $T_1$ .

The pressure dependences of  $\xi_T(T_C)^2$ ,  $\xi_T(T_1)^2$ , and  $\xi_T(T_0)^2$  are obtained from the equation-of-state (3.6). They are given at  $P = 0$  by

$$\frac{d}{dP} \xi_T(T_C)^2 = -\frac{3}{\sqrt{35}} \frac{\kappa C_{\text{mv}}}{|\tilde{b}(0)|} \left[ \frac{5}{28} - \eta - \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 + \frac{7}{5} \eta^2 \right]^{-1/2},$$

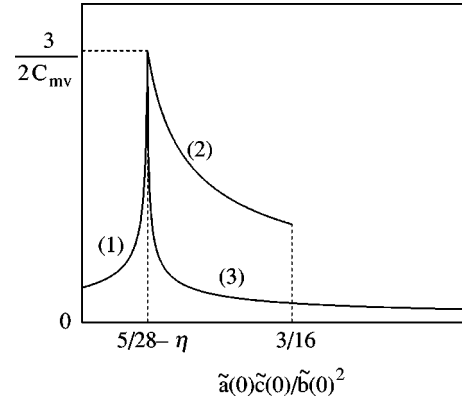


FIG. 8. Pressure dependences of  $\xi_T(T_C)^2$ ,  $\xi_T(T_1)^2$ , and  $\xi_T(T_0)^2$ . Curves (1), (2), and (3) denote  $-d\xi_T(T_C)^2/dP$ ,  $-d\xi_T(T_1)^2/dP$ , and  $-d\xi_T(T_0)^2/dP$ , respectively.

$$\frac{d}{dP} \xi_T(T_1)^2 = -\frac{6}{\sqrt{7}} \frac{\kappa C_{\text{mv}}}{|\tilde{b}(0)|} \left[ \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 - \frac{5}{28} + \eta + 28\eta^2 \right]^{-1/2},$$

$$\frac{d}{dP} \xi_T(T_0)^2 = -3 \sqrt{\frac{5}{266}} \frac{\kappa C_{\text{mv}}}{|\tilde{b}(0)|} \left[ \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 - \frac{5}{28} + \eta + \frac{35}{38} \eta^2 \right]^{-1/2}. \quad (3.17)$$

In Fig. 8,  $-d\xi_T(T_C)^2/dP$ ,  $-d\xi_T(T_1)^2/dP$ , and  $-d\xi_T(T_0)^2/dP$  are shown by curves (1), (2), and (3), respectively, as a function of  $\tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2$ . At the boundary between the first-order and second-order transitions [ $\tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 = 5/28 - \eta$ ],  $d\xi_T(T_C)^2/dP$  and  $d\xi_T(T_1)^2/dP$  become  $-3/2C_{\text{mv}}$ . In this way, the pressure dependence of the Curie temperature becomes very strong near the critical value of  $\tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2$ . The pressure dependence of spontaneous magnetization  $M_s$  at  $T = 0$  is also obtained from the equation-of-state (3.6). One gets

$$M_s^2 = \frac{|\tilde{b}(0)|}{2\tilde{c}(0)} \left[ 1 + \sqrt{1 - 4\{\tilde{a}(0) + 2\kappa C_{\text{mv}}P\}\tilde{c}(0)/\tilde{b}(0)^2} \right], \quad (3.18)$$

which is not anomalous near the critical value  $\tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 = 5/28 - \eta$ .

#### IV. DISCUSSION AND CONCLUSION

In this paper, the pressure dependence of the Curie temperature has been discussed for a ferromagnet with negative mode-mode couplings among spin fluctuations, by taking into account the magnetovolume effect. It has been shown that the Curie temperature depends strongly on pressure near the triple point  $T_t$ , while the magnetization at 0 K does not depend so much on the pressure. The present result is

consistent with the observed ones for  $\text{Co}_2$ ,  $\text{Co}(\text{S,Se})_2$ ,  $\text{Lu}(\text{Co},M)_2$  ( $M=\text{Al}$  and  $\text{Ga}$ ), and  $\text{MnSi}$  shown in Sec. II. These dependences of  $T_C$  and  $M_s$  on pressure are characteristics of itinerant-electron ferromagnets which show the MT in the paramagnetic phase.

The magnetic phase diagram has also been obtained in this paper for a ferromagnet with negative mode-mode couplings among spin fluctuations. The obtained phase diagram is very similar to the observed one for  $\text{CoS}_2$  (Fig. 1),  $\text{Co}(\text{S,Se})_2$  (Fig. 3), and  $\text{Lu}(\text{Co},M)_2$  ( $M=\text{Al}$  and  $\text{Ga}$ ). It has been explicitly shown that the susceptibility does not show a maximum above  $T_1$  in a narrow range near the triple point  $T_t$ . This is because temperature  $T_{\text{max}}$ , where the susceptibility reaches a maximum, is a little lower than the triple point  $T_t$  as shown in Fig. 7. This result is also consistent with the observed results for  $\text{Co}(\text{S,Se})_2$  shown in Fig. 3.

In this way, our theory can explain qualitatively the anomalously strong pressure dependence of the Curie temperature and the magnetic phase diagram observed in itinerant-electron ferromagnets which show the MT in the paramagnetic phase. However, only the mean amplitude of spin fluctuations at the Curie temperature, not the Curie temperature itself, was discussed in this paper. The mean amplitude of spin fluctuations  $\xi(T)^2$  is known to be proportional

to  $T^2$  at low  $T$  and to  $T$  at high  $T$ . These coefficients of  $T^2$  or  $T$  in  $\xi(T)^2$  would depend also on the pressure. Moreover, the pressure dependence of the zero-point spin fluctuation<sup>26</sup> was also neglected. Nevertheless, our results in this paper have explained very well the observed results of itinerant-electron ferromagnets which show the MT in the paramagnetic phase.

The essential results obtained in this paper rely on the occurrence of a negative mode-mode coupling  $b$  among spin fluctuations. Such a negative coupling is signified by the negative curvature of the density of states around the Fermi level and enhances the growth of spin fluctuations with temperature. The coupling between spin fluctuation modes itself is given by an integral equation for the dynamical susceptibility. Moriya<sup>6</sup> has shown in a static approximation that the mode-mode coupling is expressed in the same form as in the phenomenological description. Then, the phenomenological treatment for spin fluctuations in this paper is still available to discuss the magnetic properties at finite temperature.

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