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Negative mode–mode coupling among spin fluctuations and the magneto-volume effect in an itinerant-electron ferromagnet

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Abstract. By making use of the Landau–Ginzburg energy expanded up to the sixth power of magnetization density, the spontaneous volume magnetostriction and dependence on pressure P of the Curie temperature T_c of an itinerant-electron ferromagnet with negative mode–mode coupling among spin fluctuations are discussed. The spontaneous moment is found to show first-order and second-order transitions at T_c, according to the values of the Landau coefficients. It is shown that the difference between the spontaneous volume magnetostrictions at $T = 0$ and T_c is large and the P-dependence of T_c becomes anomalously large when a certain condition is satisfied by the Landau coefficients. The results obtained in the present theory could explain the anomalous T-dependence of the thermal expansion and the strong P-dependence of T_c observed in Invar alloys, some Laves phase Co compounds and others.

Spin fluctuations are known to play an important role in the thermodynamical properties of an itinerant-electron system with strongly exchange-enhanced susceptibility [1]. Recently, Yamada [2] has applied the spin fluctuation theory to a phenomenon of the metamagnetic transition (MT) from the paramagnetic to ferromagnetic state induced by the magnetic field, which is observed for the pyrite compound $Co(S, Se)_2$ and some Laves phase Co compounds. Two characteristic properties, the MT at low temperature and the susceptibility maximum at high temperature, have been explained clearly by this theory.

The spin fluctuation theory is phenomenologically described by the Landau–Ginzburg free-energy density

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

where $\boldsymbol{m}(\boldsymbol{r})$ is a magnetization density. The integration of $\Delta f(\boldsymbol{r})$ over the whole volume gives the magnetic part of free energy ΔF. The equation of state for bulk moment M and magnetic field H is given by

$$H = \left\langle \frac{\partial \Delta F}{\partial M} \right\rangle \qquad (2)$$

where $\langle\ \rangle$ denotes a statistical average. One gets [2]

$$H = A(T)M + B(T)M^3 + C(T)M^5 \qquad (3)$$

where

$$A(T) = a + \frac{5}{3}b\xi(T)^2 + \frac{35}{9}c\xi(T)^4 \tag{4}$$

$$B(T) = b + \frac{14}{3}c\xi(T)^2 \tag{5}$$

$$C(T) = c \tag{6}$$

and $\xi(T)^2$ is the mean square amplitude of spin fluctuations, which is a monotonically increasing function of T. In this paper, we do not give an explicit expression for $\xi(T)^2$. Nevertheless, we can discuss qualitatively the T-dependence of magnetic quantities, by making use of the fact that $\xi(T)^2$ is a monotonically increasing function of T and by neglecting the T-dependences of the Landau coefficients a, b and c.

The inverse of susceptibility is the linear coefficient $A(T)$ with respect to M in the equation of state (3). The second term of $A(T)$ in the right-hand side of equation (4) gives the main contribution from spin fluctuations at low T, which denotes a coupling between the spin fluctuations with different q [1]. Then the Landau coefficient b is taken as a mode–mode coupling constant among spin fluctuations. Moriya [3] has discussed the ferromagnetic and metamagnetic states at finite temperature for the case where $b < 0$. He has shown that the thermally induced ferromagnetic state cannot be deduced even if the effect of spin fluctuations with negative mode–mode coupling is taken into account.

The values of a, b and c in equations (4)–(6) for actual materials can be calculated by the fixed-spin-moment method of band calculation. They may take either positive or negative values, depending on the electronic structure near the Fermi level. In the case where $a > 0$, $b < 0$, $c > 0$ and $3/16 < ac/b^2 < 9/20$, the paramagnetic state is stable at $H = 0$ but the ferromagnetic state becomes stable at high magnetic field. That is, the field-induced MT from the paramagnetic to the ferromagnetic state occurs at a critical field. When $5/28 < ac/b^2 < 3/16$, on the other hand, the ferromagnetic state is stable without H at low T and the bulk moment shows a first-order transition at T_c which was written as T_1 in [2]. Above T_c, the MT occurs in this case. The MT is shown to disappear at T_0. These phenomena occur in the case where $b < 0$, i.e. for negative mode–mode coupling among spin fluctuations. Thus the metamagnetic behaviour is classified as one of the characteristic phenomena for negative mode–mode coupling among spin fluctuations.

Now, we are going to discuss the case where $a > 0$, $b < 0$, $c > 0$ and $ac/b^2 < 5/28$; this is the main purpose of the present paper. The inverse of susceptibility or $A(T)$ given by equation (4) becomes zero at

$$\xi(T)^2 = \frac{3|b|}{14c}\left\{ 1 \pm 2\sqrt{\frac{7}{5}}\sqrt{\frac{5}{28} - \frac{ac}{b^2}} \right\}. \tag{7}$$

The solution with a positive sign in the right-hand side of equation (7) gives the Curie temperature T_c [3]. This is because the equation of state (3) gives only the solution $M = 0$ at $H = 0$ as $A(T) = 0$, $B(T) > 0$ and $C(T) > 0$ at this temperature. The negative root in equation (7) does not indicate T_c. This is because $B(T)$ is negative at this temperature and the ferromagnetic state with the moment $\{|B|/C\}^{1/2}$ is stable.

The spontaneous volume magnetostriction can be discussed by adding the magneto-volume coupling energy given by [4, 5]

$$\Delta f_{mv}(\boldsymbol{r}) = -C_{mv}\rho(\boldsymbol{r})|\boldsymbol{m}(\boldsymbol{r})|^2 \tag{8}$$

to equation (1), where $\rho(\boldsymbol{r})$ and C_{mv} are the deviation of the volume density from the uniform one at $T = P = M = 0$ and the magneto-volume coupling constant. The spontaneous volume magnetostriction $\omega_m(T)$ is given by [4]

$$\omega_m(T) = \kappa C_{\mathrm{mv}} \left\{ M(T)^2 + \xi(T)^2 \right\} \tag{9}$$

where κ is the compressibility. The difference between $\omega_m(0)$ and $\omega_m(T_c)$ is then given by

$$\Delta\omega_m = \kappa C_{\mathrm{mv}} \left\{ M_0^2 - \xi(T_c)^2 \right\} \tag{10}$$

where M_0 is the spontaneous moment at $T = 0$ given by

$$M_0^2 = \frac{|b|}{2c} \left\{ 1 + \sqrt{1 - 4\frac{ac}{b^2}} \right\}. \tag{11}$$

At $ac/b^2 = 5/28$ $\Delta\omega_m/\omega_m(0)$ is 0.72 — larger than 0.4 obtained in the case where the positive mode–mode couplings, i.e. for $a < 0$, $b > 0$ and $c = 0$, by Moriya and Usami [4]. This means that a large reduction in the thermal expansion of the volume occurs in the case of negative mode–mode coupling among spin fluctuations.

The pressure effect on T_c can also be discussed using equations (1) and (8). The values of $A(T)$ and $B(T)$ are modified by the magneto-volume coupling. However, they are neglected for the sake of simplicity. In this case only $A(T)$ in the equation of state (3) depends on the pressure P through the P-dependence of the volume. That is, a term of $2\kappa C_{\mathrm{mv}} P$ is added to $A(T)$ [6]. In the case where $ac/b^2 < 5/28$ where $M(T)$ shows a second-order transition, the dependence of T_c on P is given by

$$\frac{\partial \xi(T_c)^2}{\partial P} = -\frac{3\kappa C_{\mathrm{mv}}}{\sqrt{35}\,|b|} \left\{ \frac{5}{28} - \frac{ac}{b^2} \right\}^{-1/2} \tag{12}$$

at $P = 0$. On the other hand, in the case where $3/16 > ac/b^2 > 5/28$ where $M(T)$ shows a first-order transition [6], the P-dependence of T_c is given by

$$\frac{\partial \xi(T_c')^2}{\partial P} = -\frac{6\kappa C_{\mathrm{mv}}}{\sqrt{7}\,|b|} \left\{ \frac{ac}{b^2} - \frac{5}{28} \right\}^{-1/2} \tag{13}$$

at $P = 0$, where T_c' is the Curie temperature of the first-order transition [2, 3] given by

$$\xi(T_c')^2 = \frac{3|b|}{14c} \left\{ 1 - 4\sqrt{7}\sqrt{\frac{ac}{b^2} - \frac{5}{28}} \right\}. \tag{14}$$

In figure 1, $\Delta\omega_m/\omega_m(0)$, $\xi(T_c)^2$ and $\partial\xi(T_c)^2/\partial P$ are plotted as functions of ac/b^2. In the region where $5/28 < ac/b^2 < 3/16$, $\xi(T_c')^2$ and $\partial\xi(T_c')^2/\partial P$ are plotted and $\Delta\omega_m/\omega_m(0)$ is that estimated by using equation (14). It is pointed out that an anomalously large and negative value of $\mathrm{d}T_c/\mathrm{d}P$ could be observed near the critical concentration between the ferromagnetic and metamagnetic states for pseudo-binary alloys and compounds, where $\Delta\omega_m/\omega_m(0)$ is also large. This is because the boundary between the two states is given by $ac/b^2 = 3/16$ which is very close to 5/28 [2]. The strong P-dependence of T_c is actually observed in Invar alloys and some cubic Laves phase Co compounds as discussed below.

The dependence of T_c on P has already been discussed intensively for the itinerant-electron ferromagnet; this was associated with the study on the Invar problem [7–9]. In such theories, however, the effect of spin fluctuations was not taken into account at all. The Invar properties are considered originate in the transition between the high-spin state with large volume and the low-spin state with small volume [10]. Recent band calculations for ordered Fe_3Ni and Fe_3Pt alloys support this assertion [11, 12]. Assuming that the low-spin state is paramagnetic, the present model with $a > 0$, $b < 0$ and $c > 0$ can be applied to the Invar alloys, as ΔF may take two minima at $M = 0$ and finite M. If the value of ac/b^2 is close to 5/28 at the Invar concentration of $Fe_{0.65}Ni_{0.35}$, $|\partial\xi(T_c)^2/\partial P|$ becomes anomalously

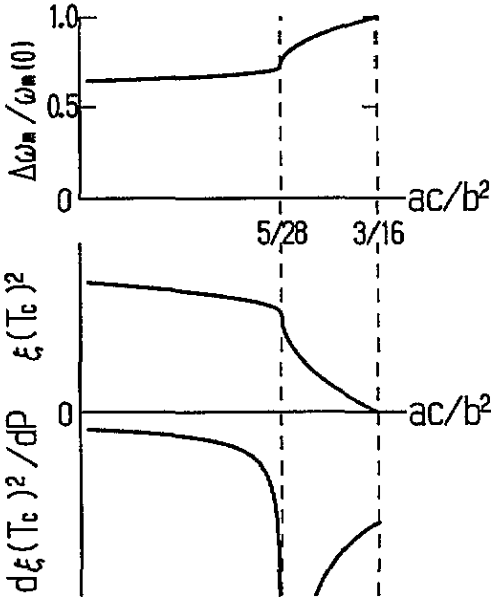

Figure 1. $\Delta\omega_m/\omega_m(0)$, $\xi(T_c)^2$ and $\partial\xi(T_c)^2/\partial P$ at $P=0$ as a function of ac/b^2.

large. However, the divergence of $\partial\xi(T_c)^2/\partial P$ shown in figure 1 is suppressed at finite P. This is because $\partial\xi(T_c)^2/\partial P$ and $\partial\xi(T_c')^2/\partial P$ are proportional to $P^{-1/2}$ for $ac/b^2 = 5/28$. Moreover, the inhomogeneity in the alloy will also suppress the divergence.

Entel and Schröter [11] have estimated the values of a, b and c for Fe_3Pt and Fe_3Ni by the fixed-spin-moment method. The value of a was found to depend strongly on the lattice constant or the Wigner–Seitz radius r_{WS}. It is found from their calculated values of a, b and c that the value of ac/b^2 becomes 5/28, where the the anomalous T-dependence of the thermal expansion and the strong P-dependence of T_c are derived, when the values of r_{WS} are 2.67 and 2.50 au for Fe_3Pt and Fe_3Ni, respectively. These values of r_{WS} are different by only about 1% or 2% from the calculated values.

The pseudo-binary compound $Zr(Fe_{1-x}Co_x)_2$ with cubic Laves phase structure also shows Invar-like properties of the anomalous thermal expansion of volume [13] and the strong P-dependence of T_c [14] near a critical concentration of $x \sim 0.5$. Around this concentration the compound is said to be in a mictomagnetic or in a spin glass state. However, the MT has been shown theoretically to occur in the narrow concentration range near the critical concentration [15]. As the value of ac/b^2 is equal to 3/16 at the critical concentration between the ferromagnetic and paramagnetic states, the value of $|\partial T_c/\partial P|$ becomes large in the ferromagnetic region near the critical concentration. Furthermore, it is pointed out that the Invar-like properties of the anomalous thermal expansion of volume and strong P-dependence of T_c will also be observed in $Hf(Fe, Co)_2$. This is because the MT has actually been observed clearly near the critical concentration [16, 17]. Moreover, the pyrite compound CoS_2 shows a strong P-dependence of T_c [18]. The pseudo-binary compound $Co(S, Se)_2$ with the same lattice structure shows the MT and the anomalous temperature dependence of the lattice constant [19]. Then the strong P-dependence of T_c could be also observed in this system near the critical concentration between the ferromagnetic and metamagnetic states.

References

[1] Lonzarich G G and Taillefer L 1985 *J. Phys. C: Solid State Phys.* **18** 4339
Moriya T 1985 *Spin Fluctuations in Itinerant Electron Magnetism* (Berlin: Springer)
[2] Yamada H 1991 *J. Phys.: Condens. Matter* **3** 4115; 1993 *Phys. Rev.* B **47** 11211
[3] Moriya T 1986 *J. Phys. Soc. Japan* **55** 357
[4] Moriya T and Usami K 1980 *Solid State Commun.* **34** 95
[5] Wagner D 1989 *J. Phys.: Condens. Matter* **1** 4635
[6] Yamada H 1994 *J. Magn. Magn. Mater* at press
[7] Wohlfarth E P 1983 *Physica* B **119** 203
[8] Terao K and Katsuki A 1974 *J. Phys. Soc. Japan* **37** 828
[9] Shimizu M 1981 *Rep. Prog. Phys.* **44** 145
[10] Wasserman E F 1990 *Ferromagnetic Materials* vol 5, ed K H J Buschow and E P Wohlfarth (Amsterdam: Elsevier Science) p 237
[11] Entel P and Schröter M 1989 *Physica* B **161** 160
[12] Entel P, Hoffmann E, Mohn P, Schwarz K and Moruzzi V L 1993 *Phys. Rev.* B **47** 8706
[13] Muraoka Y, Shiga M and Nakamura Y 1980 *J. Phys. F: Met. Phys.* **10** 127
[14] Alfieri G T, Banks E and Kanematsu K 1969 *J. Appl. Phys.* **40** 1322
[15] Yamada H and Shimizu M 1992 *J. Magn. Magn. Mater* **104–107** 1963.
[16] Sakakibara T, Goto T and Nishihara Y 1988 *J. Physique Coll.* **49** C8 263
[17] Yamada H and Shimizu M 1989 *J. Phys.: Condens. Matter* **1** 2597
[18] Sato K, Adachi K, Okamoto T and Tatsumoto E 1969 *J. Phys. Soc. Japan* **26** 639
[19] Adachi K, Matsui M and Kawai M 1979 *J. Phys. Soc. Japan* **46** 1474