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Invar-like behaviours in an itinerant-electron antiferromagnet

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Abstract. By making use of the Landau–Ginzburg energy expansion up to the sixth power of the sublattice magnetization density, the spontaneous volume magnetostriction and the dependence on pressure of the Néel temperature T_N in an itinerant-electron antiferromagnet are discussed. The antiferromagnetic moment is found to show first-order and second-order transitions at T_N—according to the values of the Landau coefficients. It is shown that the difference between the spontaneous volume magnetostrictions at $T = 0$ and T_N is large and the P-dependence of T_N becomes anomalously large when a certain condition is satisfied by the Landau coefficients.

Recently, the Invar properties of Fe alloys have been discussed using the Landau–Ginzburg free energy [1–3]. The model is based on the coexistence of two states: non-magnetic and magnetic. The free energy with respect to the magnetic moment M has two local minima at $M = 0$ and finite M, and is very similar to that in the two-γ-states model given by Weiss [4]. On taking into account the magnetovolume coupling energy, Invar properties—the large spontaneous volume magnetostriction and strong pressure dependence of the Curie temperature T_C—have been obtained. These Invar anomalies have been shown to become significant near the critical point between the first-order and second-order transitions at T_C [3].

The Invar anomalies are observed even in the antiferromagnetic FeMn and Fe(Ni, Mn) alloys [5]. A large spontaneous volume magnetostriction is observed for the intermetallic compound YMn_2 [6]. In this paper, the Invar anomalies are discussed by extending the model mentioned above to the antiferromagnetic system. For the sake of simplicity, the antiferromagnetic moment is assumed to be described by a single wavevector $\boldsymbol{Q}$:

$$\boldsymbol{m}_Q(\boldsymbol{r}) = \boldsymbol{m}_s(\boldsymbol{r})\ \exp(\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{r}). \tag{1}$$

Here, $\boldsymbol{m}_s(\boldsymbol{r})$ is the staggered or sublattice magnetization density and is given by the sum of the antiferromagnetic moment M_Q in the z-direction and the fluctuating staggered magnetization density $m_Q(\boldsymbol{r})$:

$$\boldsymbol{m}_s(\boldsymbol{r}) = M_Q\boldsymbol{e}_z + \frac{1}{\sqrt{V}}\sum_q \boldsymbol{m}_Q(\boldsymbol{q})\exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}). \tag{2}$$

The Landau–Ginzburg free energy in the present case is written up to the sixth power of the staggered magnetization density $\boldsymbol{m}_s(\boldsymbol{r})$ as

$$f(\boldsymbol{r}) = \frac{1}{2}a_Q|\boldsymbol{m}_s(\boldsymbol{r})|^2 + \frac{1}{4}b_Q|\boldsymbol{m}_s(\boldsymbol{r})|^4 + \frac{1}{6}c_Q|\boldsymbol{m}_s(\boldsymbol{r})|^6 + \frac{1}{2}D_Q|\nabla\boldsymbol{m}_s(\boldsymbol{r})|^2. \tag{3}$$

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Here, the coefficients a_Q, b_Q, c_Q and D_Q are different from those in the ferromagnetic state. They should be obtained by band calculation in the antiferromagnetic state but have not been estimated so far for any antiferromagnetic materials. However, such a form of the expansion is very useful for discussion of antiferromagnetic properties, taking into account the effect of spin fluctuations. It is noted that the last term in the right-hand side of equation (3) denotes the non-local energy associated with the antiferromagnetic spin fluctuations.

The integration of $f(\boldsymbol{r})$ over the whole volume gives the magnetic part of the free energy which is written as a functional of M_Q and the mean square amplitude of antiferromagnetic spin fluctuations. The free energy thus obtained is written mathematically in the same form as that in the ferromagnetic state [3]. Therefore, we get the same results as in [3]. The weakly antiferromagnetic state is stabilized when $a_Q < 0$ and $b_Q > 0$. However, the spontaneous volume magnetostriction is too small in this case and the Invar anomalies cannot be obtained. On the other hand, the antiferromagnetic state becomes stable also when the following relations are satisfied:

$$a_Q > 0 \qquad b_Q < 0 \qquad c_Q > 0 \qquad a_Q c_Q / b_Q^2 < 3/16. \tag{4}$$

In this case the mode–mode coupling b_Q among spin fluctuations is negative. The first-order transition occurs at T_N when $5/28 < a_Q c_Q / b_Q^2 < 3/16$. On the other hand, the second-order transition occurs when $a_Q c_Q / b_Q^2 < 5/28$.

The magnetic equation of state for the staggered magnetic field H_Q and sublattice magnetization M_Q is obtained as

$$H_Q = A_Q(T) M_Q + B_Q(T) M_Q^3 + C_Q(T) M_Q^5 \tag{5}$$

where

$$A_Q(T) = \chi_Q(T)^{-1} = a_Q + \frac{5}{3} b_Q \xi_Q(T)^2 + \frac{35}{9} c_Q \xi_Q(T)^4 \tag{6}$$

$$B_Q(T) = b_Q + \frac{14}{3} c_Q \xi_Q(T)^2 \tag{7}$$

$$C_Q(T) = c_Q \tag{8}$$

and the mean square amplitude of antiferromagnetic spin fluctuations $\xi_Q(T)^2$ is given by

$$\xi_Q(T)^2 = \frac{1}{V} \sum_q \langle |m_Q(\boldsymbol{q})|^2 \rangle. \tag{9}$$

Here, $\chi_Q(T)$ is the staggered susceptibility and $\langle \cdots \rangle$ denotes a thermal average. As $\xi_Q(T)^2$ is a monotonically increasing function of T, then $A_Q(T)$ or $\chi_Q(T)^{-1}$ has a minimum at a certain temperature as $a_Q > 0$, $b_Q < 0$ and $c_Q > 0$. This means that the staggered susceptibility in the paramagnetic state shows a maximum in its temperature dependence, which can be observed via the NMR measurements as the nuclear spin–lattice relaxation time T_1 is proportional to $T^{-1} \chi_Q(T)^{-1/2}$ [7].

By taking into account the magnetovolume energy and elastic energy, the spontaneous volume magnetostriction can be obtained as [8]

$$\omega_{\mathrm{m}}(T) = \kappa C_{\mathrm{mv}} \left\{ M_Q(T)^2 + \xi_Q(T)^2 \right\} \tag{10}$$

where κ and C_{mv} are the compressibility and magnetovolume coupling constant. Here, the volume dependences of b_Q, c_Q and D_Q are, for simplicity, neglected. The difference $\Delta\omega_{\mathrm{m}}$ ($= \omega_{\mathrm{m}}(0) - \omega_{\mathrm{m}}(T_N)$) is given by

$$\Delta\omega_{\mathrm{m}} / \omega_{\mathrm{m}}(0) = 1 - \eta_Q \tag{11}$$

where

$$\eta_Q = \xi_Q(T_N)^2/M_Q(0)^2 \tag{12}$$

$$M_Q(0)^2 = \frac{|b_Q|}{2c_Q}\left\{1+\sqrt{1-4a_Qc_Q/b_Q^2}\right\} \tag{13}$$

$$\xi_Q(T_N)^2 = \frac{3|b_Q|}{14c_Q}\left\{1+2\sqrt{7/5}\sqrt{5/28-a_Qc_Q/b_Q^2}\right\} \tag{14}$$

for $a_Qc_Q/b_Q^2 < 5/28$ and

$$\xi_Q(T_N')^2 = \frac{3|b_Q|}{14c_Q}\left\{1-4\sqrt{7}\sqrt{a_Qc_Q/b_Q^2-5/28}\right\} \tag{15}$$

for $5/28 < a_Qc_Q/b_Q^2 < 3/16$. T_N and T_N' are the Néel temperatures of the second-order transition and of the first-order one, respectively. The calculated value of η_Q is shown in figure 1 as a function of a_Qc_Q/b_Q^2. The value of η_Q is about 0.28 for $a_Qc_Q/b_Q^2 = 5/28$ which is the critical point between the first-order and second-order phase transitions.

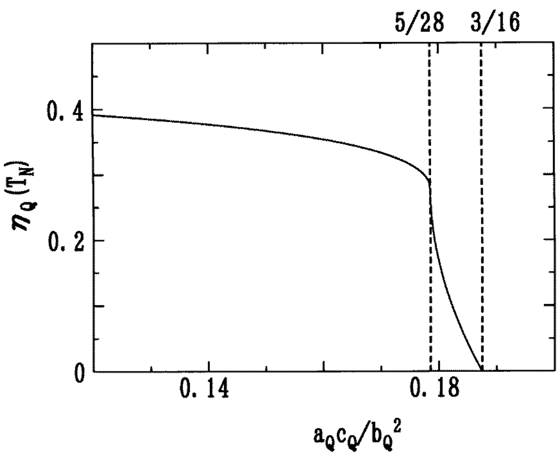

Figure 1. η_Q as a function of a_Qc_Q/b_Q^2 obtained by calculation.

The pressure (P-) dependence of T_N is also obtained as $a_Q(P) = a_Q + 2\kappa C_{\mathrm{mv}}P$, while b_Q and c_Q do not depend explicitly on P as in the ferromagnetic state [3]. We get

$$\partial\xi(T_N)^2/\partial P = -\frac{3\kappa C_{\mathrm{mv}}}{\sqrt{35}\ |b_Q|}\left\{5/28-a_Qc_Q/b_Q^2\right\}^{-1/2} \tag{16}$$

$$\partial\xi(T_N')^2/\partial P = -\frac{6\kappa C_{\mathrm{mv}}}{\sqrt{7}\ |b_Q|}\left\{a_Qc_Q/b_Q^2-5/28\right\}^{-1/2} \tag{17}$$

which are the same curves as those shown in figure 1 of [3]. T_N and T_N' are proportional to $P^{-1/2}$ at $a_Qc_Q/b_Q^2 = 5/28$ because a_Q is linear in P. $\partial T_N/\partial P$ and $\partial T_N'/\partial P$ diverge at this critical point. Moreover, the curve for $\xi_Q(T_N)$ against a_Qc_Q/b_Q^2, which is the same as that of $\xi(T_C)^2$ against ac/b^2 shown in figure 1 of [3], can be seen qualitatively as the P-dependence of T_N because only a_Q is linear in P. The material which shows the second-order transition at T_N without applied pressure may show the first-order transition under pressure if $a_Q c_Q/b_Q^2 \sim 5/28$. The large spontaneous volume magnetostriction will also be observed for such a material.

Finally we summarize our results obtained in the present paper for $a_Q > 0$, $b_Q < 0$ and $c_Q > 0$.

(i) The first-order transition occurs at T_N for $3/16 > a_Q c_Q/b_Q^2 > 5/28$.

(ii) The second-order transition occurs at T_N for $5/28 > a_Q c_Q/b_Q^2$.

(iii) The staggered susceptibility shows a maximum in its temperature dependence for $a_Q c_Q/b_Q^2 > 5/28$.

(iv) Near $a_Q c_Q/b_Q^2 = 5/28$ Invar-like behaviours, i.e., large spontaneous volume magnetostriction and strong P-dependence of T_N, are derived.

These results have been obtained for the Landau–Ginzburg energy (3). Band calculations for estimating the coefficients a_Q, b_Q and c_Q are desired for actual antiferromagnetic materials.

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