

Chapter 2

Fluctuations and Magnetism

2.1 Fluctuations

Throughout this book, we are particularly concerned with the effects of fluctuations on various magnetic properties. As a brief introduction to the fluctuation phenomena, let us first take a system of a classical harmonic oscillator in equilibrium with its surroundings at temperature T . The Hamiltonian is given by

$$\mathcal{H}(q, p) = \frac{1}{2m} p^2 + V(q), \quad V(q) = \frac{1}{2} m \omega^2 q^2, \quad (2.1)$$

where q and p represent a coordinate and its conjugate momentum. The mass of the particle and the vibration frequency are denoted by m and ω , respectively. When it is in thermal equilibrium, both of its variables q and p show random motions around the origin in the phase space. Deviations or fluctuations of variables are defined by

$$\delta q \equiv q - \langle q \rangle, \quad \delta p \equiv p - \langle p \rangle, \quad (2.2)$$

where $\langle q \rangle$ and $\langle p \rangle$ are thermal averages of variables. Both of them are zero in this case. The variances are also defined for each variable by the average of fluctuation amplitude squared.

$$\langle \delta q^2 \rangle = \langle q^2 \rangle - \langle q \rangle^2, \quad \langle \delta p^2 \rangle = \langle p^2 \rangle - \langle p \rangle^2 \quad (2.3)$$

The above averages are easily evaluated as follows for the coordinate q :

$$\langle \delta q^2 \rangle = \frac{\int_{-\infty}^{\infty} dq dp q^2 e^{-\mathcal{H}(q,p)/k_B T}}{\int_{-\infty}^{\infty} dq dp e^{-\mathcal{H}(q,p)/k_B T}} = \frac{k_B T}{m \omega^2} \quad (2.4)$$

It corresponds to the law of equipartition of energy in classical statistical mechanics.

In the presence of external force F in a positive direction, the potential energy $V(q)$ of the system is then given by

$$V(q) = \frac{1}{2}m\omega^2 q^2 - Fq. \quad (2.5)$$

The stable position of the coordinate, shifted from the origin, is represented as follows.

$$\langle q \rangle = \chi F, \quad \chi = \frac{1}{m\omega^2} \quad (2.6)$$

The parameter χ defined as a coefficient of the F -linear term in the right hand side is generally called *susceptibility*. It characterizes the response of a system to the externally applied force. From the comparison of (2.4) and (2.6), it follows that the following relation is satisfied.

$$\langle \delta q^2 \rangle = k_B T \chi \quad (2.7)$$

The relation corresponds to the special case of the well-known fluctuation-dissipation theorem of statistical mechanics. It is the relation satisfied in general between the fluctuations and the response of the system to the external perturbation.

In quantum mechanical treatment, it is better to introduce the two new variables b and b^\dagger by

$$b = \sqrt{\frac{m\omega}{2\hbar}} q + i\sqrt{\frac{1}{2m\hbar\omega}} p, \quad b^\dagger = \sqrt{\frac{m\omega}{2\hbar}} q - i\sqrt{\frac{1}{2m\hbar\omega}} p. \quad (2.8)$$

Between them, the following commutation relation is satisfied.

$$[b, b^\dagger] = 1 \quad (2.9)$$

The Hamiltonian is then represented by

$$\mathcal{H} = \hbar\omega \left(\hat{n} + \frac{1}{2} \right), \quad \hat{n} \equiv b^\dagger b. \quad (2.10)$$

If we define the ground state by the condition $b\phi_0(q) = 0$, excited eigenstates of \hat{n} , $\phi_n(q)$, with integer eigenvalue n are successively generated by

$$b^\dagger \phi_n(q) = \sqrt{n+1} \phi_{n+1}(q). \quad (2.11)$$

Thermal expectations of \hat{n} and $\langle q^2 \rangle$ are evaluated as follows.

$$\langle \hat{n} \rangle = \frac{1}{e^{\hbar\omega/k_B T} - 1}, \quad \langle q^2 \rangle = \frac{\hbar}{2m\omega} \left(1 + \frac{2}{e^{\hbar\omega/k_B T} - 1} \right) \quad (2.12)$$

It is easy to see that at high temperatures where $\hbar\omega/k_B T \ll 1$ is satisfied, the above $\langle q^2 \rangle$ reduces to the classical limit (2.4). On the other hand at low temperatures, it remains to be finite and becomes the finite value, $\hbar/2m\omega$, called *zero point fluctuation*.

Let us next consider the system where its free energy is given by

$$\begin{aligned} F[\phi] &= F_0 + \int dr \phi^*(r) [a(T) - c\nabla^2] \phi(r) + \dots \\ &= F_0 + \sum_k [a(T) + ck^2] |\phi_k|^2 + \dots \end{aligned} \quad (2.13)$$

in terms of some field variable $\phi(r)$ defined as a function of spatial coordinate, r . The Fourier transform in the wave number space with variable k is shown in the second line.

We can also define fluctuation of the amplitude by

$$\delta\phi(r) \equiv \phi(r) - \langle \phi(r) \rangle. \quad (2.14)$$

The average of the amplitude squared in (2.2) is extended to the correlation function defined by

$$C(r - r') = \langle \delta\phi^*(r) \delta\phi(r') \rangle. \quad (2.15)$$

From the free energy (2.13), the following dependence is derived.

$$C(r - r') \propto e^{-\kappa|r-r'|}, \quad \kappa^2 = a(T)/c. \quad (2.16)$$

Its Fourier transform is then written in the following Lorentzian form.

$$C(k) \propto \frac{1}{k^2 + \kappa^2} \quad (2.17)$$

Correlations between fluctuation amplitudes in such systems are expected to play significant roles in the responses against the externally applied field.

2.2 Fluctuations and Responses

As a response to spatial modulated and temporally varying external magnetic field with wave vector q and frequency ω , the magnetic moment $M(r, t)$ is induced in the system. It is linear to the external field strength for weak external field. Such a response is called *linear response*. The susceptibility is defined as its coefficient. We can find the general expression of the susceptibility by using the following Hamiltonian of the system in the presence of the external magnetic field [1].

$$\mathcal{H} = \mathcal{H}_0 + V(t), \quad V(t) = -g\mu_B S_{-q}^\alpha B_e e^{-i\omega t} \quad (2.18)$$

The first and second terms represent the unperturbed Hamiltonian and the term of the Zeeman interaction, respectively. The α component of the spin operator is denoted by S_q^α and B_e is the magnitude of the external field.

The time evolution of the quantum mechanical state $|\Psi(t)\rangle$ of the system is obtained by solving the following equation.

$$-i \frac{\partial}{\partial t} |\Psi(t)\rangle = [\mathcal{H}_0 + V(t)] |\Psi(t)\rangle \quad (2.19)$$

To find the solution perturbatively, let us define a new state $|\Phi(t)\rangle$ by $|\Psi(t)\rangle = e^{-i\mathcal{H}_0 t} |\Phi(t)\rangle$ in the interaction representation. The time evolution of $|\Phi(t)\rangle$ is then written by

$$\begin{aligned} \mathcal{H}_0 e^{-i\mathcal{H}_0 t} |\Phi(t)\rangle + i e^{-i\mathcal{H}_0 t} \frac{\partial}{\partial t} |\Phi(t)\rangle &= [\mathcal{H}_0 + V(t)] e^{-i\mathcal{H}_0 t} |\Phi(t)\rangle, \\ \therefore i \frac{\partial}{\partial t} |\Phi(t)\rangle &= V_H(t) |\Phi(t)\rangle, \end{aligned} \quad (2.20)$$

where $V_H(t)$ is defined by

$$V_H(t) = e^{i\mathcal{H}_0 t} V(t) e^{-i\mathcal{H}_0 t} = -g\mu_B S_q^\alpha(t) B_e e^{-i\omega t}, \quad S_q^\alpha(t) \equiv e^{i\mathcal{H}_0 t} S_q^\alpha e^{-i\mathcal{H}_0 t} \quad (2.21)$$

The solution of (2.20) is formally given by

$$\begin{aligned} |\Phi(t)\rangle &= |\Phi(-\infty)\rangle - i \int_{-\infty}^t dt' V_H(t') |\Phi(t')\rangle = \left[1 - i \int_{-\infty}^t dt' V_H(t') \right. \\ &\quad \left. + (-i)^2 \int_{-\infty}^t dt' V_H(t') \int_{-\infty}^{t'} dt'' V_H(t'') + \dots \right] |\Phi(-\infty)\rangle, \end{aligned} \quad (2.22)$$

where we have assumed that the system is in the state $|\Phi(-\infty)\rangle$ at $t = -\infty$. In this representation, both the state and the operators become time dependent. After the time evolution of the system, the expectation value of the β component of the spin operator $\langle \Phi(t) | S_q^\beta(t) | \Phi(t) \rangle$ is therefore given by

$$\begin{aligned} g\mu_B \langle S_q^\beta \rangle(t) &\equiv g\mu_B \langle \Phi(t) | S_q^\beta(t) | \Phi(t) \rangle \\ &= g\mu_B \langle \Phi(-\infty) | \left[1 + i \int_{-\infty}^t dt' V_H(t') + \dots \right] S_q^\beta(t) \\ &\quad \times \left[1 - i \int_{-\infty}^t dt' V_H(t') + \dots \right] | \Phi(-\infty) \rangle \\ &= i g\mu_B \int_{-\infty}^t dt' \langle [V_H(t'), S_q^\beta(t)] \rangle + \dots \end{aligned} \quad (2.23)$$

We have assumed that the zeroth order expectation does not exist in the absence of the field. Within the first order of $V_H(t)$, it is rewritten in the form,

$$\begin{aligned} g\mu_B \langle S_q^\beta \rangle(t) &= -i(g\mu_B)^2 B_e \int_{-\infty}^t dt' e^{-i\omega t'} \langle [S_{-q}^\alpha(0), S_q^\beta(t-t')] \rangle \\ &= (g\mu_B)^2 \chi^{\beta\alpha}(q, \omega) B_e e^{-i\omega t}, \end{aligned} \quad (2.24)$$

where we have defined the dynamical magnetic susceptibility by

$$\begin{aligned} \chi^{\beta\alpha}(q, \omega) &= i \int_0^\infty d\tau e^{i\omega\tau} \langle [S_q^\beta(\tau), S_{-q}^\alpha(0)] \rangle \\ &= i \int_{-\infty}^\infty d\tau e^{i\omega\tau} \theta(\tau) \langle [S_q^\beta(\tau), S_{-q}^\alpha(0)] \rangle. \end{aligned} \quad (2.25)$$

In the second line of the above expression, the step function $\theta(\tau)$ is defined by

$$\theta(\tau) = \begin{cases} 1, & \text{for } 0 \leq \tau \\ 0, & \text{for } \tau < 0 \end{cases} \quad (2.26)$$

In the system in equilibrium at the temperature T at $t = -\infty$, the expectation is given by the canonical thermal average over the initial states.

For quantum mechanical systems, variables do not generally commute with each other. The correlation between variables $S_q^\beta(t)$ and $S_{-q}^\alpha(0)$ is defined by

$$\langle \{S_q^\beta(t), S_{-q}^\alpha(0)\} \rangle = \frac{1}{2} \left[\langle S_q^\beta(t) S_{-q}^\alpha(0) \rangle + \langle S_{-q}^\alpha(0) S_q^\beta(t) \rangle \right]. \quad (2.27)$$

According to the fluctuation-dissipation theorem of the inequilibrium statistical mechanics, the Fourier transform of (2.27) is represented in terms of the imaginary part of the dynamical magnetic susceptibility.

$$\begin{aligned} \int_{-\infty}^\infty \langle \{S_q^z(t), S_{-q}^z(0)\} \rangle e^{i\omega t} dt &= \coth\left(\frac{\beta\omega}{2}\right) \text{Im} \chi^{zz}(q, \omega) \\ \langle \{S_q^z(t), S_{-q}^z(0)\} \rangle &= \frac{1}{2\pi} \int_{-\infty}^\infty d\omega \coth\left(\frac{\beta\omega}{2}\right) \text{Im} \chi^{zz}(q, \omega) e^{-i\omega t} \end{aligned} \quad (2.28)$$

As the special case, the equal time correlation at $t = 0$ is written as follows.

$$\langle \{S_q^z(0), S_{-q}^z(0)\} \rangle = \frac{1}{2\pi} \int_{-\infty}^\infty d\omega \coth\left(\frac{\beta\omega}{2}\right) \text{Im} \chi^{zz}(q, \omega) \quad (2.29)$$

2.2.1 Kramers-Kronig Relation

An effect of an externally applied magnetic field at the time t' always appears in the system at later time t ($t > t'$). This is well-known as the causality in physic. For this reason, the integral of τ in (2.25) is restricted the positive range. If we define the Fourier transform of causality related functions, for instant the dynamical magnetic susceptibility in (2.25), i.e.,

$$\chi(q, \omega) = \text{Re}\chi(q, \omega) + i\text{Im}\chi(q, \omega).$$

their real and imaginary parts are related with each other by the following relations.

$$\begin{aligned} \text{Re}\chi(q, \omega) &= \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \frac{\text{Im}\chi(q, \omega')}{\omega' - \omega}, \\ \text{Im}\chi(q, \omega) &= -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \frac{\text{Re}\chi(q, \omega')}{\omega' - \omega} \end{aligned} \quad (2.30)$$

The above relation is known as the Kramers-Kronig relation. The static magnetic susceptibility $\chi(q, 0)$ is therefore given as

$$\text{Re}\chi(q, 0) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \frac{\text{Im}\chi(q, \omega')}{\omega'}. \quad (2.31)$$

2.3 SCR Spin Fluctuation Theory

It has been well-known that the Curie-Weiss law temperature dependence of the magnetic susceptibility is observed generally for itinerant electron ferromagnets in the paramagnetic phase. On the basis of the SW theory, however, it was difficult to explain the dependence, though other properties in the ordered phase seemed to be well accounted. The purpose of the self-consistent renormalization (SCR) spin fluctuation theory by Moriya and Kawabata (1973) [2, 3] was to find a solution of this difficulty. By taking into account the effect of nonlinear mode-mode coupling among spin fluctuation modes, they were successful in explaining the Curie-Weiss law dependence. In comparison with the SW theory, it has the following features.

- Temperature dependence of various magnetic properties is attributed to the boson-like magnetic excitations, i.e., spin fluctuations, in contrast to fermion excitations of conduction electrons in the SW theory.
- The effect of nonlinear coupling among these fluctuation modes plays predominant role as an origin of the Curie-Weiss law temperature dependence of the magnetic susceptibility.

For convenience of their theoretical treatments, the following assumptions have been also made.

1. Magnetic properties in the ground state are assumed to be well described by the band theoretical approach. It is therefore regarded as a revision of the finite temperature SW theory, for its applicability is exclusively restricted to the properties at finite temperatures.
2. Based on a perturbational method, nonlinear effects of thermal spin fluctuation amplitudes are treated by expanding in powers of their amplitudes. On the other hand, effects of zero-point fluctuations are neglected.
3. The effect of nonlinear couplings among spin fluctuation modes is mainly concerned with the renormalization of the lowest second expansion coefficient of the free energy with respect to the magnetization M .

In this way, effects of fluctuations on the higher order expansion coefficients are neglected in this theory, because they are regarded as higher order corrections. Values of them are to be estimated in the same way as the SW theory.

2.3.1 Free Energy of the SCR Theory

In the SCR theory, the effects of thermal spin fluctuations are incorporated into the free energy of the SW theory. In the following we show a brief outline of the theory. Since its detailed explanation is not an aim of the book, we rely on a phenomenological approach based on the following free energy functional.

$$\begin{aligned}
 \Psi(\{\mathbf{M}_q\}, M, T) &= F_{\text{SW}}(M, T) + \Phi(\{\mathbf{M}_q\}) \\
 \Phi(\{\mathbf{M}_q\}) &= \sum_{q \neq 0} \frac{1}{2\chi_0(\mathbf{q})} \mathbf{M}_q \cdot \mathbf{M}_{-\mathbf{q}} \\
 &\quad + \frac{1}{4}b \sum_{\sum_i \mathbf{q}_i = 0} \mathbf{M}_{\mathbf{q}_1} \cdot \mathbf{M}_{\mathbf{q}_2} \mathbf{M}_{\mathbf{q}_3} \cdot \mathbf{M}_{\mathbf{q}_4} + \cdots .
 \end{aligned} \tag{2.32}$$

It consists with two contributions, F_{SW} and Φ in the first line, which represent the SW free energy and the functional of spatially modulated magnetic fluctuations. The first and the second coefficients of Φ , $1/\chi_0(\mathbf{q})$ and b , are the wave-vector dependent magnetic susceptibility in the harmonic approximation and the coupling constant among magnetic fluctuation modes, respectively. The free energy is formally evaluated as the functional integral with respect to all the possible magnetization $\mathbf{M}(\mathbf{r})$ as a function of \mathbf{r} . Variables \mathbf{M}_q are the Fourier transform of $\mathbf{M}(\mathbf{r})$. The set of variables \mathbf{M}_q with wave-vector \mathbf{q} throughout the whole Brillouin zone are denoted by $\{\mathbf{M}_q\}$. The free energy of the system is then evaluated as follows.

$$\begin{aligned}
e^{-F(M,T)/k_B T} &= \sum_{\{\mathbf{M}_q\}} \exp[-\Psi(\{\mathbf{M}_q\})/k_B T] \\
&= e^{-F_{\text{SW}}(M,T)/k_B T} \sum_{\{\mathbf{M}_q\}} \exp[-\Phi(\{\mathbf{M}_q\})/k_B T] \quad (2.33)
\end{aligned}$$

Because of the presence of nonlinear terms in Φ , the rigorous treatment of the above integration is very difficult in general.

Variational Approach We employ a variational method to find the nonlinear correction to the SW theory. Let us first introduce the following approximate harmonic functional.

$$\Phi(\{\mathbf{M}_q\}) \simeq \Phi_0(\{\mathbf{M}_q\}) = \sum_{q \neq 0} (\Omega_q^{\parallel} |\mathbf{M}_q^{\parallel}|^2 + \Omega_q^{\perp} |\mathbf{M}_q^{\perp}|^2), \quad (2.34)$$

where Ω_q^{\parallel} and Ω_q^{\perp} are variational parameters to be determined later. From the comparison with Φ in (2.32), they correspond to the wave-vector dependent magnetic susceptibility.

$$\Omega_q^{\parallel} = \frac{1}{2\chi^{\parallel}(\mathbf{q})}, \quad \Omega_q^{\perp} = \frac{1}{2\chi^{\perp}(\mathbf{q})} \quad (2.35)$$

Superscripts \perp and \parallel means the transverse and parallel components, respectively, with respect to the static spontaneous magnetization. Note there exist two independent degrees of freedom in the transverse direction. The free energy F_0 from the harmonic functional Φ_0 in (2.34) is evaluated as follows.

$$\begin{aligned}
e^{-F_0/k_B T} &= \sum_{\{\mathbf{M}_q\}} e^{-\Phi_0(\{\mathbf{M}_q\})/k_B T} = \prod_q \int d\mathbf{M}_q e^{-\beta \Phi_0(\{\mathbf{M}_q\})} \\
&= \prod_q \left[\left(\frac{\pi k_B T}{\Omega_q^{\parallel}} \right)^{1/2} \left(\frac{\pi k_B T}{\Omega_q^{\perp}} \right) \right] \quad (2.36) \\
F_0 &= -k_B T \sum_{q \neq 0} \left[\frac{1}{2} \log \left(\frac{\pi k_B T}{\Omega_q^{\parallel}} \right) + \log \left(\frac{\pi k_B T}{\Omega_q^{\perp}} \right) \right]
\end{aligned}$$

Next, the nonlinear correction of the free energy, defined by $\Delta F \equiv F - F_{\text{SW}} - F_0$, is formally evaluated as follows.

$$\begin{aligned}
e^{-\Delta F/k_B T} &= e^{F_0/k_B T} \sum_{\{\mathbf{M}_q\}} \exp[-\Phi(\{\mathbf{M}_q\})/k_B T] \\
&= e^{F_0/k_B T} \sum_{\{\mathbf{M}_q\}} e^{-\Phi_0(\{\mathbf{M}_q\})/k_B T} e^{-[\Phi(\{\mathbf{M}_q\}) - \Phi_0(\{\mathbf{M}_q\})]/k_B T} \quad (2.37) \\
&= \langle e^{-(\Phi - \Phi_0)/k_B T} \rangle,
\end{aligned}$$

i.e., as a thermal average of $e^{-(\Phi - \Phi_0)/k_B}$. The statistical thermal average in (2.37) is defined by

$$\langle \dots \rangle = e^{F_0/k_B T} \sum_{\{\mathbf{M}_q\}} e^{-\Phi_0(\{\mathbf{M}_q\})/k_B T} \dots \quad (2.38)$$

The correction ΔF in (2.37) is written in the form of the moment expansion.

$$\begin{aligned} e^{-\Delta F/k_B T} &\equiv \langle e^{-\Delta\Phi/k_B T} \rangle = 1 - \frac{1}{k_B T} \langle \Delta\Phi \rangle + \frac{1}{2!(k_B T)^2} \langle \Delta\Phi^2 \rangle - \dots \\ &= \exp \left[-\langle \Delta\Phi \rangle / k_B T + (\langle \Delta\Phi^2 \rangle - \langle \Delta\Phi \rangle^2) / 2(k_B T)^2 - \dots \right]. \end{aligned} \quad (2.39)$$

From the comparison of both sides of (2.39), the following inequality is satisfied.

$$\langle \Delta\Phi \rangle - \Delta F \simeq \frac{1}{2k_B T} (\langle \Delta\Phi^2 \rangle - \langle \Delta\Phi \rangle^2) \geq 0. \quad (2.40)$$

It implies that the approximate free energy is estimated by minimizing the average given by

$$F = F_{SW} + F_0 + \langle \Phi - \Phi_0 \rangle, \quad (2.41)$$

with respect to the variational parameters in (2.34).

Variational SCR Free Energy For the last term in (2.41), the Gaussian average defined in (2.38) is easily evaluated. For instance, the average of Φ_0 is given by

$$\langle \Phi_0 \rangle = \sum_{q \neq 0} \left(\Omega_q^{\parallel} \langle |\mathbf{M}_q^{\parallel}|^2 \rangle + \Omega_q^{\perp} \langle |\mathbf{M}_q^{\perp}|^2 \rangle \right) = \frac{3}{2} k_B T \sum_{q \neq 0} 1 = \frac{3}{2} N_0 k_B T, \quad (2.42)$$

with using the following relations.

$$\langle |\mathbf{M}_q^{\parallel}|^2 \rangle = \langle \mathbf{M}_q^{\parallel} \cdot \mathbf{M}_{-q}^{\parallel} \rangle = \frac{k_B T}{2\Omega_q^{\parallel}}, \quad \langle |\mathbf{M}_q^{\perp}|^2 \rangle = \langle \mathbf{M}_q^{\perp} \cdot \mathbf{M}_{-q}^{\perp} \rangle = \frac{k_B T}{\Omega_q^{\perp}} \quad (2.43)$$

Let us next decompose the average $\langle \Phi \rangle$ into the sum of the harmonic and the nonlinear contributions, $\langle \Phi \rangle = \langle \Phi_a \rangle + \langle \Phi_b \rangle$, defined by

$$\begin{aligned} \langle \Phi_a \rangle &= \sum_{q \neq 0} \frac{1}{2\chi_0(q)} \langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle, \\ \langle \Phi_b \rangle &= \frac{1}{4} b \sum_{\{q_i\}} \langle \mathbf{M}_{q_1} \cdot \mathbf{M}_{q_2} \mathbf{M}_{q_3} \cdot \mathbf{M}_{q_4} \rangle + \dots \end{aligned} \quad (2.44)$$

The harmonic term is then simply evaluated as follows.

$$\langle \Phi \rangle_a = k_B T \sum_{q \neq 0} \frac{1}{2\chi_0(\mathbf{q})} \left(\frac{1}{2\Omega_q^\parallel} + \frac{1}{\Omega_q^\perp} \right) \quad (2.45)$$

A slightly complicated nonlinear term $\langle \Phi \rangle_b$ is also evaluated.

$$\begin{aligned} \langle \Phi \rangle_b = & \frac{b}{4} M_0^2 \sum_{q \neq 0} \left[2\langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle + 4\langle M_q^\parallel \cdot M_{-q}^\parallel \rangle \right] \\ & + \frac{b}{4} \sum_{q, q' \neq 0} \left[\langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle \langle \mathbf{M}_{q'} \cdot \mathbf{M}_{-q'} \rangle \right. \\ & \left. + 2 \sum_{\mu=\perp, \parallel} \langle M_q^\mu \cdot M_{-q}^\mu \rangle \langle M_{q'}^\mu \cdot M_{-q'}^\mu \rangle \right] \quad (2.46) \end{aligned}$$

The terms proportional to M_0^2 are derived in the case where either of the following two conditions are satisfied in (2.32).

- $\mathbf{q}_1 = \mathbf{q}_2 = \mathbf{0}$ or $\mathbf{q}_3 = \mathbf{q}_4 = \mathbf{0}$, for the first term.
- $\mathbf{q}_1 = \mathbf{q}_3 = \mathbf{0}$ or $\mathbf{q}_2 = \mathbf{q}_4 = \mathbf{0}$, for the second term.

The terms in the second and third lines are derived when none of q_i is equal to zero. By putting (2.43) into (2.46), the average $\langle \Phi \rangle_b$ is given by

$$\begin{aligned} \langle \Phi \rangle_b = & \frac{1}{2} b k_B T M_0^2 \left(\frac{3}{2\Omega_q^\parallel} + \frac{1}{\Omega_q^\perp} \right) \\ & + \frac{1}{4} b (k_B T)^2 \sum_{q, q' \neq 0} \left\{ \left(\frac{1}{2\Omega_q^\parallel} + \frac{1}{\Omega_q^\perp} \right) \left(\frac{1}{2\Omega_{q'}^\parallel} + \frac{1}{\Omega_{q'}^\perp} \right) \right. \\ & \left. + 2 \left(\frac{1}{4\Omega_q^\parallel \Omega_{q'}^\parallel} + \frac{1}{2\Omega_q^\perp \Omega_{q'}^\perp} \right) \right\}. \quad (2.47) \end{aligned}$$

The variational free energy is finally given in the form.

$$\begin{aligned} F(M, \{\Omega_q^\parallel\}, \{\Omega_q^\perp\}, T) &= F_{\text{SW}}(M_0) + F_0 + \langle \Phi_a + \Phi_b - \Phi_0 \rangle \\ F_{\text{SW}}(M) &= \frac{1}{2\chi_0(\mathbf{0})} M^2 + \frac{1}{4} b M^4 \quad (2.48) \end{aligned}$$

Minimum Conditions of the Free Energy It is now possible to determine the variational parameters Ω_q^\parallel and Ω_q^\perp in (2.34) as well as the spontaneous magnetization M_0 from the conditions to minimize the free energy in (2.48). They are determined from the following conditions.

- The condition for Ω_q^\perp ($\mathbf{q} \neq \mathbf{0}$), i.e.

$$\Omega_q^\perp = \frac{1}{2\chi_0(\mathbf{q})} + \frac{1}{2}bM^2 + \frac{1}{4}bk_B T \sum_{\mathbf{q}' \neq \mathbf{0}} \left(\frac{1}{\Omega_{q'}^\parallel} + \frac{4}{\Omega_{q'}^\perp} \right). \quad (2.49)$$

- The condition for Ω_q^\parallel ($\mathbf{q} \neq \mathbf{0}$), i.e.

$$\Omega_q^\parallel = \frac{1}{2\chi_0(\mathbf{q})} + \frac{3}{2}bM^2 + \frac{1}{4}bk_B T \sum_{\mathbf{q}' \neq \mathbf{0}} \left(\frac{3}{\Omega_{q'}^\parallel} + \frac{2}{\Omega_{q'}^\perp} \right). \quad (2.50)$$

- The condition for M , i.e.

$$\frac{H}{M} = \frac{1}{\chi_0(\mathbf{0})} + bM^2 + \frac{1}{2}bk_B T \sum_{\mathbf{q}' \neq \mathbf{0}} \left(\frac{3}{\Omega_{q'}^\parallel} + \frac{2}{\Omega_{q'}^\perp} \right). \quad (2.51)$$

For paramagnets or in the paramagnetic phase with no externally applied magnetic field, there appears no induced magnetic moment. The variational parameters then become isotropic, i.e., $\Omega_q \equiv \Omega_q^\parallel = \Omega_q^\perp$. The above two conditions (2.49) and (2.50) in this case reduce to the single condition,

$$\Omega_q = \frac{1}{2\chi_0(\mathbf{q})} + \frac{5}{4}bk_B T \sum_{\mathbf{q}' \neq \mathbf{0}} \frac{1}{\Omega_{q'}}. \quad (2.52)$$

In the uniform $\mathbf{q} = \mathbf{0}$ limit, $2\Omega_0 = H/M$ is satisfied. The final condition (2.51) also coincides with the above (2.52).

2.3.2 Curie–Weiss Law of Magnetic Susceptibility

In the SCR spin fluctuation theory, essentially the same equation in (2.52) is used to evaluate the temperature dependence of the magnetic susceptibility. It is however shown in a little bit different form.

Note that the isotropic spin fluctuation amplitude in the paramagnetic phase is given by

$$\langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle = \langle |\mathbf{M}_q^\parallel|^2 \rangle + \langle |\mathbf{M}_q^\perp|^2 \rangle = \frac{3k_B T}{2\Omega_q}. \quad (2.53)$$

Equation (2.52), in the uniform $\mathbf{q} = \mathbf{0}$ limit, is then written in the form,

$$\frac{1}{\chi(T)} = \frac{1}{\chi_0(\mathbf{0})} + \frac{5}{3}b \sum_q \langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle, \quad \chi(T) \equiv \chi(\mathbf{0}) = \frac{1}{2\Omega_0}, \quad (2.54)$$

where $\chi(T)$ is the uniform magnetic susceptibility. The second correction term to the SW theory, being proportional to b , results from the effect of nonlinear couplings among spin fluctuation modes. The temperature dependence of this term gives rise from the following two reasons.

1. The explicit temperature dependence in the thermal average in (2.53).

In our classical high temperature approximation, the thermal amplitude in (2.53) is proportional to the absolute temperature T . In quantum mechanical treatment, its dependence results from that of the Bose distribution function.

2. Implicit temperature dependence through the parameter, Ω_q .

In order to evaluate the mean thermal amplitude squared in (2.54), the parameter Ω_q is necessary. It is shown as a sum of two contributions.

$$\Omega_q = \Omega_0 + (\Omega_q - \Omega_0) = \frac{1}{2\chi(T)} + (\Omega_q - \Omega_0) \quad (2.55)$$

The first term of the inverse of the magnetic susceptibility is temperature dependent. The second term rather characterizes the dispersion of the parameters in wave-vector space.

The thermal amplitude therefore depend on temperature through the direct T -dependence of the statistical distribution function and also the indirect dependence from the magnetic susceptibility $\chi(T)$.

Self-Consistency Condition Magnetic susceptibility diverges at the critical temperature $T = T_c$. It can be used as the condition to determine the critical temperature, as given by

$$0 = \frac{1}{\chi_0(\mathbf{0})} + \frac{5}{3}b \sum_q \langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle(0, T_c), \quad (2.56)$$

where the thermal amplitude is explicitly shown as a function of χ^{-1} and T . In the SW theory without the second term, the temperature dependence of the first term $\chi^{-1}(0)$ determines T_c . Much higher T_c may be then obtained.

By subtracting both sides of (2.54) and (2.56), the following equation is derived.

$$\frac{1}{\chi(T)} = \frac{5}{3}b \sum_q [\langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle(\chi^{-1}, T) - \langle \mathbf{M}_q \cdot \mathbf{M}_{-q} \rangle(0, T_c)] \quad (2.57)$$

In quantum mechanical treatments, thermal amplitudes in the right hand side is evaluated in terms of the imaginary part of the dynamical magnetic susceptibility $\chi(\mathbf{q}, \omega)$, according to the fluctuation-dissipation theorem in (2.29). Since the solution χ^{-1} is also involved in the right hand side in (2.57), we have to find the solution that satisfies this equation *self-consistently*. Numerically, estimated solutions χ^{-1} of the

equation derived quantum mechanically show good linearity in a wide temperature range above T_c . From the comparison with experiments, its validity is confirmed even quantitatively [4].

2.4 Discontinuous Change of Magnetization

Although the SCR spin fluctuation theory was successful in the derivation of the Curie-Weiss law temperature dependence of the magnetic susceptibility in the paramagnetic phase, there exists seemingly a slight difficulty in the ordered phase. That is, its temperature dependence of the spontaneous magnetic moment always vanishes discontinuously at the Curie point. We show in this section, how the discontinuous change gives rise, and a possible prescription for the solution.

2.4.1 Temperature Dependence of Magnetization

To begin with, let us start from the following expansion of the free energy.

$$F(M, T) = F(0, T) + \frac{1}{2}a(T)M^2 + \frac{1}{4}b(T)M^4 + \dots \quad (2.58)$$

In the SCR theory, effects of spin fluctuations are mainly restricted to the coefficient $a(T)$ of the second term. In the presence of a finite magnetization in the system, the thermodynamic relation and magnetic susceptibilities are given by

$$\begin{aligned} H &= \frac{\partial F}{\partial M} = a(T)M + b(T)M^3 + \dots \\ \frac{1}{\chi_{\parallel}} &= \frac{\partial H}{\partial M} = a(T) + 3b(T)M^2 + \dots, \\ \frac{1}{\chi_{\perp}} &= \frac{H}{M} = a(T) + b(T)M^2 + \dots, \end{aligned} \quad (2.59)$$

where \perp and \parallel stand for the components perpendicular and parallel to the magnetization. The last two relations for inverse of magnetic susceptibilities are satisfied for rotationary invariant systems in spin space. The difference in these components results from the second M^2 -linear terms. In the absence of the magnetic field, the following results are obtained.

$$\frac{1}{\chi_{\perp}} = \frac{H}{M_0(T)} = a(T) + b(T)M_0^2(T) = 0, \quad \frac{1}{\chi_{\parallel}} = 2b(T)M_0^2(T) > 0 \quad (2.60)$$

Since the perpendicular component always remains in zero in this case, $M_0^2(T) = -a(T)/b(T)$ is satisfied. The parallel component χ_{\parallel}^{-1} , on the other hand, increases

with decreasing the temperature. The difference between them becomes finite and temperature dependent in the ordered phase, while in the paramagnetic phase it is very small even in the presence of an external magnetic field.

Let us next examine whether variational parameters Ω_q^\perp and M determined by (2.49) and (2.51) are consistent with the thermodynamic relation (2.59). If we notice the relation $2\Omega_0^\perp = \chi_\perp^{-1}$ in (2.35), (2.49) is written in the form,

$$\frac{1}{\chi_\perp} = \frac{1}{\chi_0(0)} + bM^2 + \frac{1}{2}bT \sum_{q' \neq 0} \left(\frac{1}{\Omega_{q'}^\parallel} + \frac{4}{\Omega_{q'}^\perp} \right), \quad (2.61)$$

in the limit $q = 0$. However, it does not satisfy the relation $\chi_\perp^{-1} = H/M$ in (2.59), since the subtraction of the right hand side of (2.61) and (2.51) for H/M gives the nonzero result,

$$\frac{1}{\chi_\perp} - \frac{H}{M} = bT \sum_{q' \neq 0} \left(\frac{1}{\Omega_{q'}^\perp} - \frac{1}{\Omega_{q'}^\parallel} \right) \neq 0. \quad (2.62)$$

To avoid the above inconsistency involved in (2.49), (2.50), and (2.51), let us simply assume the following wave-vector dependence of Ω_q^\perp and Ω_q^\parallel .

$$\begin{aligned} \Omega_q^\perp &= \Omega_0^\perp + (\Omega_q^\perp - \Omega_0^\perp) = \Omega_0^\perp + \frac{1}{2}Aq^2 = \frac{1}{2}Aq^2 \\ \Omega_q^\parallel &= \Omega_0^\parallel + (\Omega_q^\parallel - \Omega_0^\parallel) = \Omega_0^\parallel + \frac{1}{2}Aq^2 = bM_0^2 + \frac{1}{2}Aq^2, \end{aligned} \quad (2.63)$$

where the q^2 dependence of $\chi_0^{-1}(q)$ is extended throughout the whole of the Brillouin zone as given by

$$\frac{1}{\chi_0(\mathbf{q})} - \frac{1}{\chi_0(\mathbf{0})} = Aq^2 + \dots. \quad (2.64)$$

In the limit $H = 0$, (2.51) is then given by

$$\begin{aligned} \frac{1}{\chi_0(\mathbf{0})} + bM_0^2 + 3bT \sum_{q \neq 0} \frac{1}{Aq^2 + 2bM_0^2} + 2bT \sum_{q \neq 0} \frac{1}{Aq^2} &= 0, \\ \frac{1}{\chi_0(0)} + 5bT_c \sum_{q \neq 0} \frac{1}{Aq^2} &= 0, \end{aligned} \quad (2.65)$$

where the second equation corresponds to the first one at the critical point at $T = T_c$ and $M_0 = 0$. Subtraction of both sides of them finally give the following equation.

$$M_0^2 - 3T \sum_q \left(\frac{1}{Aq^2 + 2bM_0^2} - \frac{1}{Aq^2} \right) + 5(T - T_c) \sum_q \frac{1}{Aq^2} = 0. \quad (2.66)$$

As a solution, temperature dependence of the spontaneous moment $M_0(T)$ is estimated.

2.4.2 Origin of the Discontinuity

The wave-vector summation of the second term in (2.66) throughout the whole of the Brillouin zone is evaluated as follows.

$$\begin{aligned} \sum_q \left(\frac{1}{Aq^2 + 2bM_0^2} - \frac{1}{Aq^2} \right) &= -\frac{8\pi V b M_0^2}{(2\pi)^3 A^2} \int_0^{q_B} dq \frac{1}{q^2 + 2bM_0^2/A} \\ &= -\frac{bM_0^2 V}{\pi^2 A^2} \sqrt{\frac{A}{2bM_0^2}} \tan^{-1} \sqrt{\frac{Aq_B^2}{2bM_0^2}}, \end{aligned} \quad (2.67)$$

where the zone boundary wave-vector is denoted by q_B . It becomes proportional to M_0 as the magnitude of M_0 approaches to 0, i.e.,

$$\sum_q \left(\frac{1}{Aq^2 + 2bM_0^2} - \frac{1}{Aq^2} \right) \simeq -\frac{V}{2\pi A} \left(\frac{b}{2A} \right)^{1/2} M_0 \quad (2.68)$$

The temperature dependence of M_0 is therefore determined by solving the following quadratic equation.

$$\begin{aligned} M_0^2 - c_1(T)M_0 - c_2(T) &= 0, \\ c_1(T) &= \frac{3VT}{2\pi A} \left(\frac{b}{2A} \right)^{1/2}, \quad c_2(T) = 5(T_c - T) \sum_q \frac{1}{Aq^2} > 0 \end{aligned} \quad (2.69)$$

In the above definition, $c_1(t)$ and $c_2(t)$ are both positive for $T < T_c$.

The presence of the negative constant term $-c_2(T) \rightarrow 0$ (for $T \rightarrow T_c$) implies that both positive and negative solutions are present. The positive physical solution also remains finite at the critical temperature because of the finite value of $c_1(T_c)$. The M_0 -linear term results from the parallel component of the thermal amplitude. Reasons of the discontinuous jump of the magnetization are therefore stated as follows.

- It results from the critical behavior of the parallel component of thermal spin fluctuation amplitude with respect to the spontaneous magnetic moment. Because of this reason, only the transverse component of fluctuations is included in the SCR theory to avoid the difficulty in evaluating the temperature dependence of the spontaneous magnetization.
- The M_0^2 -linear dependence of the parallel component of the inverse magnetic susceptibility around the critical point, i.e., $\chi_{\parallel}^{-1} \propto M_0^2$.

There would be an argument that the above results derived from (2.66) are based on the classical high temperature approximation. However, even if we take quantum mechanical effects into consideration, our conclusion will remain unchanged. Phenomena of phase transitions at finite temperature are mostly governed by thermal fluctuations in the low energy region. There is nothing wrong with our approximation for these thermal excitations.

It may be interesting to find the magnetic isotherm just at the critical point. In the presence of the external magnetic field H , (2.66) is written by

$$\begin{aligned} \frac{H}{M} = bM^2 + cM^4 + \dots + 3bT_c \sum_{q \neq 0} \left(\frac{1}{Aq^2 + \partial H / \partial M} - \frac{1}{Aq^2} \right) \\ + 2bT_c \sum_{q \neq 0} \left(\frac{1}{Aq^2 + H/M} - \frac{1}{Aq^2} \right) \quad (2.70) \end{aligned}$$

As with (2.68), the wave-vector summations in the right hand side give terms proportional to $\sqrt{\partial H / \partial M}$ and $\sqrt{H/M}$ for a weak external magnetic field H . As a trial solution, let us assume the relation $H \propto M^\alpha$ with an odd integer exponent α . Then both of them become proportional to $M^{(\alpha-1)/2}$. It follows that a self-consistent solution of (2.70) has to satisfy the condition $\alpha \geq 5$, for even power terms, at least M^2 -linear or higher order terms, have to be present in the right hand side. It implies that $b(T_c) = 0$ has to be satisfied at the critical point, in contradiction to the assumption of the SCR theory.

Anyway, the discontinuous change of the spontaneous magnetization originates from the temperature independent fourth order coefficient $b(T)$ of the free energy. If we allow higher order expansion coefficients to be temperature dependent, more sophisticated treatments of the magnetic isotherm including higher order coefficients are necessary as will be shown in Chaps. 3 and 4. For convenience of later chapters, we show below in this chapter, properties of the thermal and the zero-point spin fluctuation amplitudes as functions of temperature and inverses of parallel and perpendicular magnetic susceptibilities.

2.5 Thermal and Zero-Point Spin Fluctuation Amplitudes

It may be well known that the spin fluctuation spectrum in low frequency (ω) and long wave number (q) regions is well described by the double Lorentzian distribution function. If various magnetic properties are influenced by these fluctuations, they will be described in terms of parameters that characterize spectral widths of the spin fluctuation amplitudes in q, ω space. First in this section, two spectral widths are defined. Then thermal and zero-point spin fluctuation amplitudes are represented in terms of these parameters. For convenience of later explanations, the following variables are introduced.

For crystals with N_0 magnetic ions, the dimensionless average of spin angular moment σ on a magnetic ion and the external magnetic field h in units of energy are defined by

$$\sigma = M/(N_0 g \mu_B), \quad h = g \mu_B H, \quad (2.71)$$

where M and H are the magnetization of the system and the externally applied magnetic field. We also define magnetic moment per atom by $p = g\sigma = M/(N_0 \mu_B)$. Magnetic susceptibilities are therefore measured in units of $(g \mu_B)^2$ and redefined by

$$\begin{aligned} \chi^{-1} &\equiv (g \mu_B)^2 \frac{H}{M} = \frac{h/g \mu_B}{N_0 g \mu_B \sigma} = \frac{1}{N_0} \frac{h}{\sigma}, \\ (g \mu_B)^2 \frac{\partial H}{\partial M} &= \frac{1}{N_0} \frac{\partial h}{\partial \sigma}. \end{aligned} \quad (2.72)$$

Hereafter we assume $g = 2$ for the gyro-magnetic ratio, and energies and temperatures are measured in units of \hbar and k_B , for simplicity.

According to the fluctuation-dissipation theorem (2.29), the following relation is satisfied between the average of the local spin amplitude squared and the imaginary part of the dynamical magnetic susceptibility in the paramagnetic phase and in the absence of external magnetic field.

$$\begin{aligned} \langle S_{\text{loc}}^2 \rangle &= \frac{1}{N_0} \sum_{\mathbf{q}} \langle \mathbf{S}_{\mathbf{q}} \cdot \mathbf{S}_{-\mathbf{q}} \rangle = \frac{3}{N_0^2} \sum_{\mathbf{q}} \int_0^\infty \frac{d\omega}{\pi} \coth(\omega/2T) \text{Im} \chi(\mathbf{q}, \omega), \\ \coth(\omega/2T) &= \frac{e^{\omega/T} + 1}{e^{\omega/T} - 1} = 1 + \frac{2}{e^{\omega/T} - 1} = 1 + 2n(\omega). \end{aligned} \quad (2.73)$$

With the use of the decomposition of $\coth(\omega/2T)$ in the above second line, let us define the thermal and zero-point local spin fluctuation amplitude, denoted by subscripts T and Z, by

$$\begin{aligned} \langle S_{\text{loc}}^2 \rangle &= \langle S_{\text{loc}}^2 \rangle_Z + \langle S_{\text{loc}}^2 \rangle_T, \\ \langle S_{\text{loc}}^2 \rangle_Z &= \frac{3}{N_0^2} \sum_{\mathbf{q}} \int_0^\infty \frac{d\omega}{\pi} \text{Im} \chi(\mathbf{q}, \omega), \\ \langle S_{\text{loc}}^2 \rangle_T &= \frac{6}{N_0^2} \sum_{\mathbf{q}} \int_0^\infty \frac{d\omega}{\pi} n(\omega) \text{Im} \chi(\mathbf{q}, \omega). \end{aligned} \quad (2.74)$$

In the integrand of the thermal amplitude, the Bose distribution function $n(\omega) = (e^{\omega/T} - 1)^{-1}$ is present.

For ferromagnets, the imaginary part of the dynamical magnetic susceptibility in the small q , ω regions is well described by the imaginary part of the dynamical magnetic susceptibility.

$$\begin{aligned} \text{Im}\chi(\mathbf{q}, \omega) &= \chi(\mathbf{q}, 0) \frac{\omega \Gamma_q}{\omega^2 + \Gamma_q^2}, \quad \chi(\mathbf{q}, 0) = \chi(\mathbf{0}, 0) \frac{\kappa^2}{\kappa^2 + q^2}, \\ \Gamma_q &= \Gamma_0 q (\kappa^2 + q^2), \quad q \equiv |\mathbf{q}| \end{aligned} \quad (2.75)$$

The damping constant Γ_q , the half width of the frequency distribution, has a meaning of the inverse of the life time of the fluctuation with wave-vector q . The correlation wave-number κ is defined by $\kappa = 2\pi/\lambda$ as the inverse of the magnetic correlation length λ . In this way, spectral shape of spin fluctuation amplitude depend on the value of κ . In the following, the parameter y defined below is also used in place of the inverse of magnetic susceptibility, for the relation $\kappa^2 \propto \chi(\mathbf{0}, 0)$ is satisfied.

$$y = \frac{\kappa^2}{q_B^2} = \frac{N_0}{2T_A \chi(\mathbf{0}, 0)} = \frac{h}{2T_A \sigma}. \quad (2.76)$$

Spectral distribution in q and ω spaces therefore depend on temperature and external magnetic field through the magnetic susceptibility.

2.5.1 Spectral Properties of Spin Fluctuation Amplitudes

For ferromagnets, the uniform component of the inverse magnetic susceptibility $\chi^{-1}(\mathbf{0}, 0)$ is very small, i.e., $y \ll 1$. Then the distribution of $\chi^{-1}(\mathbf{q}, 0)$ in wave-vector space is characterized by its zone-boundary value, $\chi^{-1}(\mathbf{q}_B, 0)$. Since the inverse of magnetic susceptibility in (2.72) is measured in units of energy, let us define the parameter T_A , in units of temperature, by

$$\frac{N_0}{\chi(\mathbf{q}_B, 0)} = \frac{N_0(1 + q_B^2/\kappa^2)}{\chi(\mathbf{0}, 0)} = \frac{N_0(1 + 1/y)}{\chi(\mathbf{0}, 0)} \simeq \frac{N_0}{\chi(\mathbf{0}, 0)y} \equiv 2k_B T_A, \quad (2.77)$$

as a measure of the spectral dispersion in the wave-vector space. The above definition of T_A has a close relationship with y in (2.76).

Likewise, we can define another parameter T_0 as a measure of the spectral distribution in the frequency space. It is defined by

$$\Gamma_{q_B} = \Gamma_0 q_B (\kappa^2 + q_B^2) = \Gamma_0 q_B^3 (y + 1) \simeq \Gamma_0 q_B^3 = 2\pi k_B T_0, \quad (2.78)$$

as the width Γ_q of the ω dependence at the zone-boundary wave vector $q = q_B$.

With these parameters, the wave-vector dependence of the magnetic susceptibility and the damping constant are written in the form,

$$\chi(\mathbf{q}, 0) = \chi(\mathbf{0}, 0) \frac{\kappa^2}{\kappa^2 + q^2} = \frac{N_0}{2T_A(y + x^2)},$$

$$\Gamma_q = \Gamma_0 q (q^2 + \kappa^2) = 2\pi T_0 x (x^2 + y), \quad x \equiv q/q_B, \quad (2.79)$$

where the reduced wave vector x is introduced. The wave-vector summation over the Brillouin zone is also written as follows.

$$\frac{1}{N_0} \sum_{\mathbf{q}} = \frac{4\pi V}{(2\pi)^3 N_0} \int_0^{q_B} dq q^2 = \frac{4\pi q_B^3 V}{(2\pi)^3 N_0} \int_0^1 dx x^2 = 3 \int_0^1 dx x^2 \quad (2.80)$$

Finally, the reduced temperature t defined below is used hereafter in place of the absolute temperature T .

$$t = \frac{T}{T_0}. \quad (2.81)$$

2.5.2 Thermal Spin Fluctuation Amplitude

The thermal spin fluctuation amplitude defined in the last line of (2.74) is regarded as a function of two independent variables, y and t . By introducing the reduced frequency $\xi = \omega/2\pi T$, the imaginary part of the dynamical susceptibility is written in the form,

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

$$= \frac{N_0}{2T_A} \frac{1}{y + x^2} \frac{\xi u(x)}{\xi^2 + u^2(x)}, \quad u(x) \equiv x(y + x^2)/t \quad (2.82)$$

where $u(x)$ is the reduced damping constant. The frequency and wave-vector integral over the variables ξ and x , after putting (2.82) into (2.74), is then written as follows.

$$\langle S^2 \rangle_T(y, t) = \frac{18T_0}{T_A} \int_0^1 dx x^3 \int_0^\infty d\xi \frac{\xi}{e^{2\pi\xi} - 1} \frac{1}{\xi^2 + u^2} = \frac{9T_0}{T_A} A(y, t)$$

$$A(y, t) \equiv \int_0^1 dx x^3 \left[\log(u) - \frac{1}{2u} - \psi(u) \right], \quad (2.83)$$

where $\psi(u)$ is the digamma function defined by the logarithmic derivative of the gamma function $\Gamma(u)$, i.e. $\psi(u) = d \log \Gamma(u)/du$. The function $u(x)$ defined in (2.82) is simply denoted by u in the above integrand.

Especially in the limit of low temperature and around the critical point, its dependence on y and t can be explicitly given as follows.

- Around the critical point

Reflecting the anomalous x dependence of the integrand in (2.83), the y dependence of the thermal amplitude $A(y, t)$ for $y \ll 1$ is dominated by the critical behavior. For $u \ll 1$ in the long wavelength limit, $\log u - 1/2u - \psi(u) \simeq 1/2u$ is satisfied. By putting this approximation into (2.83), the integral over x gives the following y dependence.

$$\begin{aligned} \Delta A(y, t) &\equiv A(y, t) - A(0, t) \simeq \frac{t}{2} \int_0^1 dx \left(\frac{x^2}{y + x^2} - 1 \right) \\ &= -\frac{1}{2}t y \int_0^1 dx \frac{1}{y + x^2} = -\frac{t}{2} \sqrt{y} \tan^{-1} \frac{1}{\sqrt{y}} \end{aligned} \quad (2.84)$$

In other words, the following nonanalytic behavior is derived around $y = 0$.

$$\Delta A(y, t) \simeq -\frac{\pi t}{4} \sqrt{y}, \quad (y \ll 1) \quad (2.85)$$

The dependence, that cannot be expanded in powers of y around the origin $y = 0$, is characteristic to the critical phenomena.

The t dependence of the thermal amplitude in this region is also evaluated as follows. By introducing a new variable $v = x^3/t$ in place of $u(x)$ for $y = 0$, the thermal amplitude for $t \ll 1$ is given by

$$\begin{aligned} A(0, t) &= \frac{1}{3} t^{4/3} \int_0^{1/t} dv v^{1/3} \left[\ln v - \frac{1}{2v} - \psi(v) \right] \simeq \frac{1}{3} C_{4/3} t^{4/3}, \\ C_\alpha &\equiv \int_0^\infty dv v^{\alpha-1} \left[\log v - \frac{1}{2v} - \psi(v) \right], \quad C_{4/3} = 1.00608 \dots \end{aligned} \quad (2.86)$$

The critical thermal amplitude is proportional to $t^{4/3}$ at the critical point for $y = 0$.

- Low temperature limit

Since $1 \ll u$ is satisfied in this limit, the following asymptotic expansion is satisfied for the integrand $\log u - 1/2u - \psi(u)$ in (2.83).

$$\log u - \frac{1}{2u} - \psi(u) \sim \frac{1}{12u^2} - \frac{1}{120u^4} + \frac{1}{252u^6} + \dots \quad (2.87)$$

If it is approximated by the first term $1/12u^2$, the amplitude shows the t^2 -linear dependence in this limit.

$$A(y, t) \simeq \frac{1}{12} \int_0^1 dx \frac{x^3}{u^2(x)} = \frac{t^2}{12} \int_0^1 dx \frac{x}{(y + x^2)^2} = \frac{t^2}{24} \frac{1}{y(1+y)}. \quad (2.88)$$

Its coefficient shows the tendency to diverge in proportion to $1/y$, as y approaches zero.

2.5.3 Zero-Point Spin Fluctuation Amplitude

The amplitude of zero-point fluctuations depends only on the variable y . In our treatment of various magnetic properties, the region around the origin $y = 0$ is particularly important. Although no explicit temperature dependence is involved in this amplitude, it implicitly depends on temperature through the variable y . To examine its detailed y dependence has a significant meaning for our purpose. Compared to the thermal amplitudes, anomalous behaviors do not like to give rise because of the absence of the Bose distribution function in the frequency integration in (2.74). As will be seen in (2.59), magnetic susceptibility is generally suppressed by the appearance of magnetization. As a result, the amplitude of fluctuations is also suppressed with increasing y .

By introducing the reduced frequency, $\eta = \omega/2\pi T_0$, the imaginary part of the dynamical susceptibility is written as follows.

$$\begin{aligned} \text{Im}\chi(\mathbf{q}, \omega) &= \frac{N_0}{2T_A} \frac{1}{y + x^2} \frac{\eta v(x)}{\eta^2 + v^2(x)} = \frac{N_0}{2T_A} \frac{\eta x}{\eta^2 + v^2(x)}, \\ v(x) &= x(y + x^2). \end{aligned} \quad (2.89)$$

The frequency integral of (2.74) is then written by

$$\begin{aligned} \langle S_{\text{loc}}^2 \rangle_Z(y) &= \frac{9T_0}{T_A} \int_0^1 dx x^3 \int_0^{\eta_c} d\eta \frac{\eta}{\eta^2 + v^2(x)} \\ &= \frac{9T_0}{2T_A} \int_0^1 dx x^3 \{ \log[\eta_c^2 + v^2(x)] - 2 \log v(x) \}, \end{aligned} \quad (2.90)$$

where η_c is the cut-off frequency to avoid logarithmic divergence. It follows that the following y -linear dependence is derived around the origin $y = 0$.

$$\langle S_{\text{loc}}^2 \rangle_Z(y) = \langle S^2 \rangle_Z(0) - \frac{9T_0}{T_A} c y + \dots \quad (2.91)$$

The numerical coefficient c is defined by extracting the factor $9T_0/T_A$, common to this case and (2.83) for the thermal amplitude.

The y -linear constant c defined in (2.91) can also be evaluated directly by the following derivative with respect to y .

$$\begin{aligned} \frac{\partial}{\partial y} \langle S^2 \rangle_Z(y) &= \frac{3}{N_0^2} \sum_{\mathbf{q}} \int_0^\infty \frac{d\omega}{\pi} \frac{\partial}{\partial y} \left\{ \chi(\mathbf{q}) \frac{\omega \Gamma(\mathbf{q}, \omega)}{\omega^2 + \Gamma^2(\mathbf{q}, \omega)} \right\} \\ &= \frac{3}{N_0^2} \sum_{\mathbf{q}} \int_0^\infty \frac{d\omega}{\pi} \left\{ \frac{\partial[\chi(\mathbf{q}) \Gamma(\mathbf{q}, \omega)]}{\partial y} \frac{\omega}{\omega^2 + \Gamma^2(\mathbf{q}, \omega)} \right\} \end{aligned}$$

$$-[\chi(\mathbf{q})\Gamma(q, \omega)] \frac{2\omega\Gamma(q, \omega)}{[\omega^2 + \Gamma^2(q, \omega)]^2} \frac{\partial \Gamma(q, \omega)}{\partial y} \Big\} \quad (2.92)$$

where the ω dependence is introduced for the damping constant $\Gamma(q, \omega)$ by taking account the spectral distribution, actually decaying faster than Lorentzian distribution, in the higher frequency region. The term in the second line is neglected, for the y dependence of $[\chi(\mathbf{q})\Gamma(q, \omega)]$ is neglected (see (2.79), for instance) at low frequencies, where the y dependence is particularly dominant. Finally, the ω dependence of $\Gamma(q, \omega)$ is also neglected, i.e., $\Gamma(q, \omega) \simeq \Gamma_q$, in the last line, because of the presence of the decaying factor, $\omega/[\omega^2 + \Gamma^2(q, \omega)]^2 \sim 1/\omega^3$, at high frequencies. The y derivative of (2.92) is therefore rewritten as follows.

$$\begin{aligned} \frac{\partial}{\partial y} \langle S^2 \rangle_Z(y) &\simeq -\frac{3}{N_0^2} \sum_{\mathbf{q}} \chi(\mathbf{q}) \frac{\partial \Gamma_q}{\partial y} \int_0^\infty \frac{d\omega}{\pi} \frac{2\omega\Gamma_q^2}{(\omega^2 + \Gamma_q^2)^2} \\ &= -\frac{3}{N_0^2} \sum_{\mathbf{q}} \frac{1}{\pi} \chi(\mathbf{q}) \frac{\partial \Gamma_q}{\partial y}. \end{aligned} \quad (2.93)$$

The coefficient c in (2.91) is then estimated by using

$$c = \frac{T_A}{3N_0^2 T_0} \sum_{\mathbf{q}} \frac{1}{\pi} \chi(\mathbf{q}) \left. \frac{\partial \Gamma_q}{\partial y} \right|_{y=0}. \quad (2.94)$$

Its value for the Lorentzian distribution function is given by

$$c = \int_0^1 dx \left. \frac{x^3}{y + x^2} \right|_{y=0} = \frac{1}{2}. \quad (2.95)$$

2.6 Spin Amplitude Conservation

We have shown in the preceding Sect. 2.5, that the zero-point spin fluctuation amplitude also depends on temperature and is suppressed by an externally applied magnetic field as with the thermal amplitude. With increasing temperature, the thermal amplitude monotonically increases in the paramagnetic phase, while the zero-point amplitude decreases because of its y dependence in (2.91). We feel therefore tempted to assume that the sum of both the amplitudes is conserved independent of temperature and/or irrespective of the presence of magnetic field.

There are several theoretical indications to date that seem to support the above idea. For example, Shiba and Pincus [5] have shown that the variation of the amplitude against temperature is only of the order of $(k_B T/W)^2$ in their study on the one-dimensional Hubbard model. Since W is the band width of the conduction electrons, the dependence is actually negligible in the range of temperature where magnetic

properties are usually observed experimentally. The same result is confirmed by the numerical Monte Carlo study by Hirsch [6] on the two-dimensional finite size Hubbard model. These are the examples where the dominant antiferromagnetic correlation is present. Recently, the occurrence of the partial ferromagnetism has been found by Nakano and Takahashi on the one-dimensional Hubbard model with next nearest hopping interaction. Almost temperature independent total spin amplitude is also confirmed in this case [7].

We show below in this section, results of several experimental studies that seem to support the spin amplitude conservation.

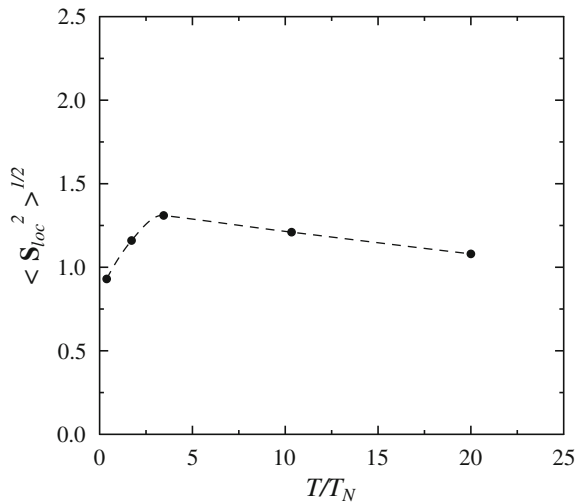
2.6.1 Neutron Scattering Experiment on MnSi

Inelastic thermal neutron scattering experiments on MnSi with polarized beam was made by Ziebeck et al. [8]. Observed results of intensities are plotted against the temperature in Fig. 2.1. In this experiment, scattered neutrons in all directions are collected. Energies of scattering neutrons are also not resolved as well. If we define the scattering amplitude of neutrons by $S(\mathbf{q}, \omega)$, the observed intensity I therefore amounts to the following integral.

$$I = \sum_{\mathbf{q}} \int_{-\omega_c}^{\omega_c} d\omega S(\mathbf{q}, \omega). \quad (2.96)$$

The cut-off frequency ω_c is determined by the upper limit of energies of incident thermal neutrons. The amplitude $S(\mathbf{q}, \omega)$ is related to the imaginary part of the

Fig. 2.1 Temperature dependence of the total spin amplitude observed by Ziebeck et al. [8]



dynamical magnetic susceptibility by

$$S(\mathbf{q}, \omega) \propto \bar{S}(\mathbf{q}, \omega) = \frac{1}{1 - e^{-\omega/T}} \text{Im} \chi(\mathbf{q}, \omega) \\ = \begin{cases} [1 + n(\omega)] \text{Im} \chi(\mathbf{q}, \omega), & \omega \geq 0 \\ n(|\omega|) \text{Im} \chi(\mathbf{q}, |\omega|), & \omega < 0 \end{cases} \quad (2.97)$$

In the paramagnetic phase, the imaginary part of the dynamical susceptibility is an odd function of ω . Because of the presence of the extra dependence on ω , the intensity is asymmetric with respect to the origin of ω . The intensity is also expressed as a sum of the thermal and zero-point components, $\bar{S}_T(\mathbf{q}, \omega)$ and $\bar{S}_Z(\mathbf{q}, \omega)$, respectively, i.e., by

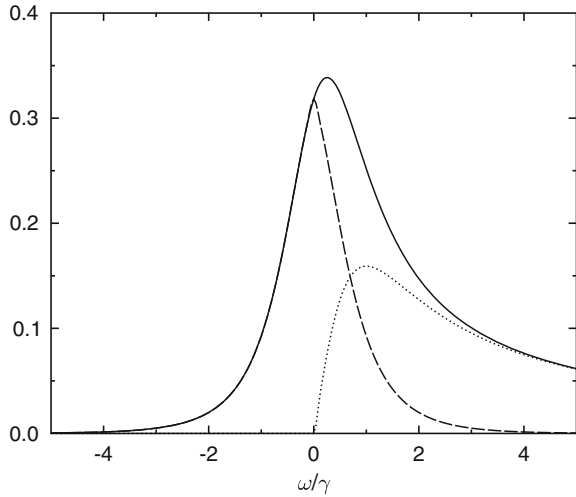
$$\bar{S}(\mathbf{q}, \omega) = \bar{S}_T(\mathbf{q}, \omega) + \bar{S}_Z(\mathbf{q}, \omega), \\ \bar{S}_T(\mathbf{q}, \omega) \equiv n(|\omega|) \text{Im} \chi(\mathbf{q}, |\omega|), \quad \bar{S}_Z(\mathbf{q}, \omega) \equiv \theta(\omega) \text{Im} \chi(\mathbf{q}, \omega) \quad (2.98)$$

where $\theta(\omega)$ is the step function with values, 1 for $0 \leq \omega$, or 0 for $\omega < 0$, depending on the sign of ω . Intensities for negative frequency originate only from the thermal component. We show in Fig. 2.2, the frequency dependence of the scattering amplitude as well as its components, evaluated by assuming the Lorentzian distribution function for some fixed wave-vector \mathbf{q} .

Let us define integrated thermal and zero-point intensities by

$$I_T = \sum_{\mathbf{q}} \int_0^{\omega_c} d\omega \bar{S}_T(\mathbf{q}, \omega), \quad I_Z = \sum_{\mathbf{q}} \int_0^{\omega_c} d\omega \bar{S}_Z(\mathbf{q}, \omega). \quad (2.99)$$

Fig. 2.2 Frequency dependence of the scattering intensity. *Solid, dashed, and thin dotted curves* represent the total intensity, zero-point and thermal components, respectively. The damping constant is denoted by γ



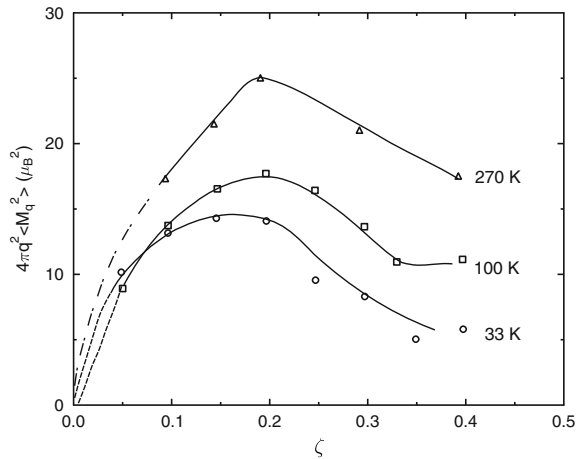
The observed intensity (2.96) by Ziebeck et al. is then written by

$$I = 2I_T + I_Z. \quad (2.100)$$

If the cut-off frequency ω_c is high enough, the above intensity corresponds to the total spin amplitude. Otherwise, it is the sum of the thermal amplitude $2I_T$ and a partial amplitude of the zero-point amplitude in the low frequency region. Observed intensities at several temperatures reported by Ziebeck et al. are plotted in Fig. 2.1. Almost temperature independent intensities by them is consistent with the spin amplitude conservation. The slight tendency of the decrease observed at high temperatures may results from the broadening of the spectral width with increasing temperature. A portion of the intensity at high frequencies will then shift beyond the upper bound frequency ω_c .

Soon after the report by Ziebeck et al., the another results of inelastic neutron measurements were published by Ishikawa et al. [9]. Their main purpose was to validate the assumption of the SCR theory, i.e., the increase of the thermal spin fluctuation amplitude with increasing temperature. They measured frequency and wave-vector decomposed scattering intensities. To extract the temperature dependence of thermal component of the fluctuation amplitudes I_T , only the observed intensity in the negative frequency range was numerically integrated. Their results at temperatures $T = 33$ K, 100 K, and 270 K are shown from the bottom in Fig. 2.3. The wave-number is denoted by ζ for the horizontal axis. These intensities increase with increasing temperature for almost all the wave number ζ . They insisted the validity of the SCR assumption based on their findings. We must be, however, a bit careful. Since only the thermal part of the amplitude is extracted, their results always make sense. At the same time, it does not necessarily contradict the total spin amplitude conservation, because they say nothing about the intensity estimated by the integral over the wide range of frequency including the positive side.

Fig. 2.3 Wave vector dependence of the scattering intensity at several temperatures, $T = 33$ K, 100 K, and 270 K by Ishikawa et al. [9]



2.6.2 Theoretical Explanation for Experiments on MnSi

For the explanation of the almost temperature independent scattering intensity of MnSi by Ziebeck et al., the corresponding intensity was theoretically evaluated by Takahashi and Moriya [10]. By assuming the double Lorentzian form of the spectrum for the imaginary part of the dynamical magnetic susceptibility, the following wave-vector summation and the frequency integral were performed numerically.

$$\bar{S}_L^2(T) \propto \sum_q \int_{-\omega_c}^{\omega_c} d\omega \bar{S}(q, \omega) \quad (2.101)$$

The temperature dependence of the magnetic susceptibility, being necessary for (2.101), is evaluated based on the SCR spin fluctuation theory. The result shown in Fig. 2.4 is fairly in agreement with the result of Fig. 2.1 by Ziebeck et al. They, however, argued that the observed temperature independent behavior would originate from the limited energy range of thermal neutron beams. If the cut-off frequency ω_c would become higher, the amplitude would show increase with increasing temperature as predicted by the SCR theory.

It seems that the effects of the temperature variation and the externally applied magnetic field are restricted within the lower frequency region. We show in Fig. 2.5 the spectral intensity $\bar{S}(q, \omega)$ for two different values of y against the frequency ω . The thermal components, $n(\omega)\text{Im}\chi(q, \omega)$, show steep increase toward the origin, while the zero-point components proportional to ω around the origin show broad peaks around $\omega/\gamma \sim 1$. Larger value of y is used for dashed curves than those for solid curves. Since there is no change in the Bose distribution function in this calculation, it amounts to the effect of external magnetic field.

Fig. 2.4 Temperature dependence of the theoretically calculated neutron scattering intensity of MnSi by Takahashi and Moriya [4]. Solid, dashed, and dash-dotted curves correspond to the total, the zero-point, and the thermal amplitudes, respectively

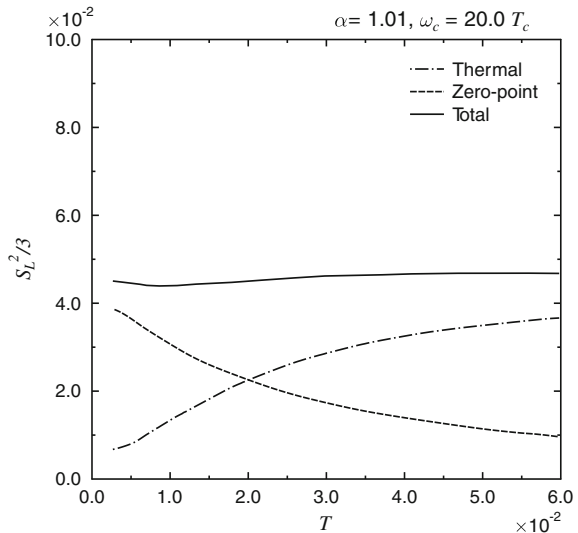
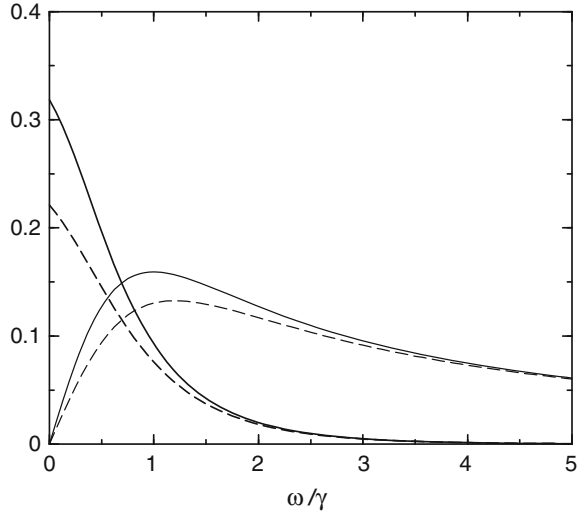


Fig. 2.5 Spectral shape change for thermal and zero-point amplitudes at some fixed wave-vector q caused by the variation of the inverse of the magnetic susceptibility γ



Although the thermal amplitude is notably suppressed at low frequencies, the effect on the zero-point amplitude should not also be ignored in the range of the frequency of the order of the damping constant γ . The effect on the intensities at high frequencies is equally neglected for both components. Even if measurements of the scattering intensity of MnSi becomes possible in higher frequency region, the temperature independence of the scattering amplitude by Ziebeck et al. will almost remain unchanged.

2.6.3 Giant Magnetic Fluctuations Observed in (Y,Sc)Mn₂

The presence of the zero-point amplitude is also demonstrated by the neutron scattering experiments on the the Laves phase compound YMn₂. It shows the first order like phase transition around $T = 100$ K from the antiferromagnetic to the paramagnetic state, accompanied by the huge magneto-volume striction. The antiferromagnetism is found to disappear with a slight substitution of Sc for Y. Nevertheless, the large thermal volume expansion coefficient is still observed at low temperatures, indicating the presence of magnetic fluctuations with large amplitude. Polarized inelastic neutron scattering experiments made by Shiga et al. [11] has clarified the following nature of spin fluctuations in this material.

- Antiferromagnetic fluctuations with large amplitude are actually present.
- Its amplitude increases with increasing temperature.
- Finite fluctuation amplitudes are present even at low temperatures. The frequency dependence of the scattering intensity is asymmetric with respect to the origin.

Particularly the above last behavior clearly indicates the presence of sizable zero-point fluctuation amplitudes at low temperatures.

2.7 Summary

In this chapter, we have shown that the Curie-Weiss law temperature dependence of the magnetic susceptibility of itinerant electron ferromagnets can be explained as an effect of nonlinear coupling among spin fluctuation modes. The same approach, however, inevitably gives an inappropriate discontinuous change of the spontaneous magnetization at the critical temperature. The reason is because the fourth expansion coefficient $b(T)$ of the free energy (2.58) is assumed to be independent of temperature. In order to solve the difficulty, it will be necessary to deal with higher order expansion coefficients of the magnetization curve, i.e., H as a function of M in (2.59).

We have also shown that the conservation of the total spin amplitude is also satisfied from both the theoretical and experimental point of views. It implies that the amplitude of the zero-point spin fluctuation also depends on temperature and is affected by the externally applied magnetic field.

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