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Spin Fluctuation Theory of Itinerant Electron Magnetism

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Spin Fluctuation Theory of Itinerant Electron Magnetism

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Preface

My research career in the field of itinerant electron magnetism started in 1977 when I got a job in Institute for Solid State Physics, the University of Tokyo. Since then as a member of Professor T. Moriya's laboratory, I have been involved in the development of the spin fluctuation theory of itinerant electron magnetism, well known now as the SCR spin fluctuation theory. The finite temperature Stoner-Wohlfarth theory has been improved by these studies as reviewed in the book, "Spin Fluctuations in Itinerant Electron Magnetism", by Moriya published in 1985. From the beginning, I was interested in a few difficulties still unresolved around 1980 and several assumptions of the theory. These have become the motivation for my subsequent studies. Most of them have been done after I moved to Faculty of Science, Himeji Institute of Technology.

The aim of this book is to review the new theoretical development of the spin fluctuation theory that began from around the middle of 1980s. It is based on the very simple ideas, i.e, the assumption of the total spin amplitude conservation and the explicit account the effect of zero-point spin fluctuations, etc. These allows us to deal with wide variety of phenomena ranging from the ground state to the paramagnetic phase at high temperatures. Various interesting predictions on the properties of itinerant electron ferromagnets have been also derived in agreement with a number of experimental observations.

The book does not attempt to cover a wide area of magnetism. What I would like to emphasize is that magnetic and thermal properties of itinerant magnets are even more determined under the influence of magnetic fluctuations than we have thought. Subjects are mainly confined in thermodynamic properties. It is valuable to capture the current status of the spin fluctuation theories for graduate students and researchers in the field of magnetism and is also helpful for the analyses of experimental data.

The author is grateful to Dr. Toru Moriya for inviting me in this field of research and various useful advice and instructions. I would like to thank Dr. Kazuyoshi Yoshimura, Dr. Takeshi Kanomata, Dr. Hironori Nishihara, Dr. Kazuaki Shimizu, Dr. Yuichi Dazuke, Dr. Masayuki Shiga, Dr. Tsuneaki Goto, Dr. Hideji Yamada, Dr. Keiichi Koyama, Dr. Hiroyuki Nakamura, Dr. Kazuaki Fukamichi, Dr. Asaya Fujita, Dr. Deguchi, and Dr. Takehide Koyama for collaborations and many fruitful suggestions and discussions. I also want to thank my staff and students in Himeji Institute of Technology, now renamed University of Hyogo.

Himeji, December 2012 Yoshinori Takahashi

Contents

Acronyms

CW	Curie–Weiss
GC	Global Consistency
MU	Moriya-Usami
QCP	Quantum Critical Point
SCR	Self-Consistent Renormalization
SEW	Stoner–Edwards–Wohlfarth
SW	Stoner–Wohlfarth
TAC	Total Amplitude Conservation

Chapter 1
Introduction

1.1 Historical Overview

The beginning of theoretical studies on itinerant electron magnets goes back to the theory by Stoner [1]. It associates the magnetism in metals with the band splitting of the conduction electron states. Temperature and external field dependence of various magnetic properties are understood in terms of the change in the occupation number of conduction electrons. Since then the development along this line has been carried through by Wohlfarth and co-workers. In the following, the theoretical framework on this line is called the Stoner-Wohlfarth (SW) theory. An introductory review is presented in the book by Mohn [2].

In 1973, two papers were published by Moriya and Kawabata [3, 4] on the effects of collective magnetic excitations, called spin fluctuations. The theory is now well known as the self-consistent renormalization (SCR) spin fluctuation theory. In contrast to the SW theory, rolls of thermal magnetic fluctuations are particularly emphasized in this theory in deriving the Curie–Weiss (CW) law temperature dependence of magnetic susceptibility, observed generally in itinerant electron ferromagnets. Since then intensive theoretical and experimental investigations have been done on various magnetic, thermal, and transport properties of itinerant electron magnets [5].

The SCR theory draws an exact line between properties in the ground state and those at finite temperatures. In the ground state, the SW theory is assumed to be justified. It means that the applicability of the theory is restricted to properties at finite temperatures. Only the effects of thermal spin fluctuations have been their main concerns. For instance, the magnetic isotherm, i.e., the relation between the magnetization M and the external magnetic field H, is generally given as

$$H = a(T)M + b(T)M^3 + \cdots, \tag{1.1}$$

where the coefficients, $a(T)$, $b(T)$, $\cdots$, on the right-hand side are functions of absolute temperature T. Their T-dependence is predominantly determined by the

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effect of nonlinear couplings among thermal fluctuation amplitudes. The first coefficient $a(T)$ corresponds to the inverse of the magnetic susceptibility.

The theory assumes that the relative importance of the effect is mainly restricted to the first coefficient $a(T)$, since its magnitude is very small for magnets and paramagnets close to their magnetic instability points. On the other hand, the T-dependence of higher coefficients has been usually neglected for simplicity. Their values are assumed to be well evaluated by band theoretical calculations. Particularly the coefficient $b(T)$, as a lowest order nonlinear coupling constant, has a significant role in deriving the temperature dependence of the first coefficient $a(T)$.

The theory seemed to be very successful in predicting various magnetic, thermal, and transport properties even quantitatively [5]. Nevertheless, subtle difficulties are involved in the theory as shown below.

1. Temperature dependence of spontaneous magnetization always discontinuously drops to zero at the critical temperature T_C.
2. Nonlinear relation between M^2 and H/M was sometimes observed in the magnetization curve for the compound MnSi [6]. It implies that higher order coefficient, $b(T)$, for instance, in (1.1) is also temperature dependent.
3. The temperature dependence of the specific heat shows a spurious negative peak just above the critical temperature [7].

No satisfactory treatments were, therefore, possible for properties in the magnetically ordered phase as well as effects of external magnetic field.

From the efforts to overcome the above difficulties, a new framework of the spin fluctuation theory was proposed by Takahashi [8]. In contrast to the SCR theory, it explicitly takes into account the effect of zero-point spin fluctuations. It follows that properties of the ground state have become the targets of the theory. The theory has the following characteristic features:

- Magnetic isotherm, i.e., to find the M dependence of H in (1.1), becomes our main theoretical concern.
- Both effects of temperature and external magnetic field can be treated consistently from a unified point of view.

These theoretical developments of the spin fluctuation theory starting from around 1985 are reviewed in this book. The basic ideas necessary to understand the various magnetic properties of itinerant electron magnetism are also presented in detail.

Later in this section, a brief introduction to the theory of magnetism is presented. Then band theoretical treatment of itinerant electron ferromagnetism is explained for the comparison with spin fluctuation theories in later chapters.

1.2 Localized Heisenberg Magnets

When localized moments are defined for atoms in an insulating crystal, interactions of these moments are sometimes described by the following Heisenberg Hamiltonian:

$$\mathscr{H} = -J \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \tag{1.2}$$

where $\boldsymbol{S}_i$ is the spin operator on i-th atom and $\langle i, j \rangle$ means that the summation is over the nearest neighbor pairs of i-th and j-th atoms. The coupling constant J, of the order of magnitude t_{ij}^2/U, is called the exchange interaction. Inter-atomic hopping energy and repulsive intra-atomic coulomb energy between electrons are denoted by t_{ij} and U, respectively. Owing to the presence of the above interactions, magnetic transition occurs at some critical temperature T_c. In the ordered phase below T_c, the spontaneous magnetization with some wave vector $\boldsymbol{q}$ appears in the absence of external magnetic field. To grasp a rough picture of magnetic phase transition derived from the above model, simple approximation methods are presented below.

1.2.1 Mean Field Approximation

Curie Law of the Isolated Single Atom System To begin with, let us deal with the single atom system with a finite total angular momentum $\boldsymbol{J}$ under the presence of the external magnetic field $\boldsymbol{H} = (0, 0, H)$ along the z-direction. The Hamiltonian is given by the following Zeeman energy:

$$\mathscr{H} = -\boldsymbol{\mu} \cdot \boldsymbol{H}. \tag{1.3}$$

In the presence of the spin–orbit interaction, only the total angular moment $\boldsymbol{J} = \boldsymbol{L} + \boldsymbol{S}$ is a conserved quantity, rather than the spin moment $\boldsymbol{S}$ in (1.3), and the magnetic moment $\boldsymbol{\mu}$ is effectively proportional to $\boldsymbol{J}$, i.e., $\boldsymbol{\mu} = -g\mu_{\mathrm{B}}\boldsymbol{J}$. The constant g is known as the gyro-magnetic ratio. The Hamiltonian (1.3) is therefore represented as

$$\mathscr{H}_z = g\mu_{\mathrm{B}} \boldsymbol{J} \cdot \boldsymbol{H}. \tag{1.4}$$

Although the thermal average of $\boldsymbol{\mu}$ is zero in equilibrium state, the finite magnetic moment m, proportional to the external magnetic field H, is induced.

$$m = \langle \mu_z \rangle = \chi(T)H. \tag{1.5}$$

According to the conventional method of statistical mechanics, the thermal average of the above moment can be evaluated. In the presence of the magnetic field H along the z-axis, the eigenenergies of (1.3) split into the equidistant $(2J + 1)$ levels. Thermodynamic properties of the system are derived from the free energy $F(H, T) = -k_{\mathrm{B}}T \log Z$, and therefore from the following partition function Z:

$$
\begin{aligned}
Z &= \sum_{m=-J}^{J} \mathrm{e}^{g\mu_\mathrm{B} m H/k_\mathrm{B}T} = \frac{\mathrm{e}^{-g\mu_\mathrm{B} J H/k_\mathrm{B}T}[1-\mathrm{e}^{g\mu_\mathrm{B}(2J+1)H/k_\mathrm{B}T}]}{1-\mathrm{e}^{g\mu_\mathrm{B} J H/k_\mathrm{B}T}} \\
&= \frac{\sinh[(2J+1)g\mu_\mathrm{B}H/2k_\mathrm{B}T]}{\sinh(g\mu_\mathrm{B}H/2k_\mathrm{B}T)} \qquad (1.6)\\
F(H,T) &= -k_\mathrm{B}\{\log\sinh[(2J+1)g\mu_\mathrm{B}H/2k_\mathrm{B}T] - \log\sinh(g\mu_\mathrm{B}H/2k_\mathrm{B}T)\}
\end{aligned}
$$

The magnetic moment is then derived from the free energy, as given by

$$
\begin{aligned}
m &= g\mu_\mathrm{B}\langle J_z\rangle = -\frac{\partial F}{\partial H} = g\mu_\mathrm{B} B_J(x) \\
B_J(x) &= \{(1+1/2J)\coth[(1+1/2J)x] - (1/2J)\coth(x/2J)\}, \qquad (1.7)
\end{aligned}
$$

where $x = g\mu_\mathrm{B} J H/k_\mathrm{B}T$, and $B_J(x)$ is known as the Brillouin function.

Depending on the magnitude of x, the following expansion is satisfied for $\coth x$ in (1.7):

$$
\coth x = \begin{cases} \dfrac{1}{x} + \dfrac{x}{3} + \cdots, & \text{for } |x| \ll 1 \\ 1 + 2\mathrm{e}^{-2x} + \cdots, & \text{for } |x| \gg 1 \end{cases} \qquad (1.8)
$$

At high temperatures, since $x \ll 1$ is satisfied, the first line of (1.7) is approximated by

$$
m \simeq \frac{(g\mu_\mathrm{B})^2 J(J+1)}{3k_\mathrm{B}T} H = \chi(T)H. \qquad (1.9)
$$

Even if the system consists of a large number of atoms, e.g., N atoms, its magnetic susceptibility becomes N times larger than that in the case of a single atom, as far as inter-atomic interactions are negligible like systems of dilute gas. The magnetic susceptibility is then given as

$$
\chi(T) = \frac{C}{T}, \quad C = \frac{N(g\mu_\mathrm{B})^2 J(J+1)}{3k_\mathrm{B}}. \qquad (1.10)
$$

The above temperature dependence is known as the Curie law, and the constant C is the Curie constant.

The total magnetic moment squared, $\mu^2 = \boldsymbol{\mu}\cdot\boldsymbol{\mu}$, is a conserved quantity of the system, for the commutation relation, $[J^2, \mathscr{H}] = 0$, is satisfied. Equation (1.10) means that the following relation is satisfied between the conserved amplitude squared, $J^2 = J_x^2 + J_y^2 + J_z^2$, and the magnetic susceptibility $\chi(T)$.

$$
\frac{1}{3}\boldsymbol{\mu}\cdot\boldsymbol{\mu} = (g\mu_\mathrm{B})^2 J(J+1) = \frac{1}{N} k_\mathrm{B}T\chi(T) \qquad (1.11)
$$

The above relation is a special case of the fluctuation–dissipation theorem of non-equilibrium statistical mechanics, i.e., the case where the high temperature approximation is justified. A brief explanation of the theorem will be given in Chap. 2. The Curie law temperature dependence of the magnetic susceptibility is closely related to the basic principle of the statistical mechanics. If we know the above relation (1.11) from the beginning, we will be able to derive the Curie law behavior of $\chi(T)$ straightforwardly.

Mean Field Approximation Let us next deal with a system, in which a large number of localized moments are included. Mutual interactions among them are described by the Heisenberg Hamiltonian of (1.2). By decreasing the temperature of such a system, the magnetic phase transition occurs at some critical temperature T_c. Below T_c, a finite spontaneous magnetization with some spatial modulation appears. As a simple approximate method for phase transitions, we will show below a treatment based on the molecular field approximation. We assume that the interaction in (1.2) is ferromagnetic, i.e., J is positive, for simplicity of the treatment.

In this approximation, interactions between a spin $\boldsymbol{S}_i$ on ith site with neighboring spins $\boldsymbol{S}_j$ on jth sites is approximated by an effective static external field.

$$\boldsymbol{H}_m = \frac{J}{g\mu_B} \sum_j{}' \langle \boldsymbol{S}_j \rangle \tag{1.12}$$

If the spontaneous moment is in the z-axis direction, it becomes equivalent to the problem in the presence of the following effective magnetic field H_{eff}:

$$H_{\mathrm{eff}} = H + H_m = H + \frac{\zeta J}{N(g\mu_B)^2} M, \quad M = \frac{g\mu_B N}{\zeta} \sum_j{}' \left\langle S_j^z \right\rangle \tag{1.13}$$

where j represents the summation over the nearest neighbor ζ magnetic ions. The magnetization is denoted by M in this treatment. If we put the effective field H_{eff} of (1.13) into the external magnetic field of the single spin problem, the magnetic susceptibility (1.9) at high temperatures is written as

$$M \simeq \frac{N(g\mu_B)^2 S(S+1)}{3k_B T} H_{\mathrm{eff}} = \frac{S(S+1)}{3k_B T}[N(g\mu_B)^2 H + \zeta J M]. \tag{1.14}$$

Now, from the definition of $\chi(T) = M/H$, the temperature dependence of the magnetic susceptibility is given as

$$\chi(T) = \frac{N(g\mu_B)^2 S(S+1)}{3k_B(T - T_C)}, \quad T_C = \frac{S(S+1)\zeta J}{3k_B} \tag{1.15}$$

It is called Curie–Weiss law temperature dependence. It diverges at the critical temperature T_c, called Curie temperature for ferromagnets. Below T_c, the spontaneous magnetization appears even in the absence of external magnetic field.

The temperature dependence of the magnetization in the ordered phase is obtained by solving the following equation:

$$M = N(g\mu_B)B_J(x), \quad x = \frac{\zeta JM}{N(g\mu_B)k_BT}. \tag{1.16}$$

It is the same equation as (1.7), if H in x is replaced by H_{eff} for $H = 0$. At low temperatures, the following exponential temperature dependence, characteristic of systems with finite energy gap, is derived:

$$\begin{aligned} M &\simeq \frac{N}{2} g\mu_B \left[(2S+1)(1 + 2e^{-g\mu_B(2S+1)H_{\text{eff}}/k_BT}) - (1 + 2e^{-g\mu_B H_{\text{eff}}/k_BT}) \right] \\ &= Ng\mu_B S \left(1 - \frac{1}{S} e^{-g\mu_B H_{\text{eff}}/k_BT} + \cdots \right), \end{aligned} \tag{1.17}$$

the same expansion as (1.8) justified for $1 \ll |x|$. In order to derive the more reasonable dependence, the effect of spin waves has to be included.

Note that in the above treatment of the magnetic susceptibility, the magnetization M is directly estimated by using the thermodynamic relation (1.7). The approximate free energy is first evaluated in this example. Then the magnetic susceptibility is obtained from its second derivative with respect to the magnetization M. Similar treatments are also employed by the SW and SCR theories.

1.2.2 Phase Transitions of Heisenberg Magnets and Spin Amplitude Conservation

We show in this section another different approach based on the spin amplitude conservation. Only the case of ferromagnetism is treated as well for simplicity. The spin operator $\boldsymbol{S}_j$ on a jth magnetic ion site commutes with the squared spin amplitude $\boldsymbol{S}_i^2$ on any ith site. The following commutation relation is then satisfied for each operator $\boldsymbol{S}_i^2$:

$$[\boldsymbol{S}_i^2, \mathscr{H}] = 0 \tag{1.18}$$

It implies that $\boldsymbol{S}_i^2$ is a constant of motion and its expectation value is always conserved. The following spin amplitude conservation is satisfied:

$$\sum_i \langle \boldsymbol{S}_i \cdot \boldsymbol{S}_i \rangle = \sum_{\boldsymbol{q}} \langle \boldsymbol{S}_{\mathbf{q}} \cdot \boldsymbol{S}_{-\mathbf{q}} \rangle = NS(S+1) \tag{1.19}$$

A number of magnetic ions in the crystal is denoted by N. In the case where a finite static magnetization is present, it is written as a sum of the mean spin amplitude squared and the average of squared fluctuation amplitudes.

$$|\langle \boldsymbol{S}_0 \rangle|^2 + \sum_{\boldsymbol{q}} \langle \delta\boldsymbol{S}_{\mathbf{q}} \cdot \delta\boldsymbol{S}_{-\mathbf{q}} \rangle = NS(S+1), \quad \delta\boldsymbol{S}_{\mathbf{q}} = \boldsymbol{S}_{\mathbf{q}} - \delta_{q,0} \langle \boldsymbol{S}_0 \rangle. \tag{1.20}$$

In Eqs. (1.19) and (1.20), fourier components of spin operators are defined as

$$\boldsymbol{S}_{\boldsymbol{q}} = \sum_i \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_i} \boldsymbol{S}_i, \quad \boldsymbol{S}_i = \frac{1}{N} \sum_{\boldsymbol{q}} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_i} \boldsymbol{S}_{\boldsymbol{q}}. \tag{1.21}$$

The Hamiltonian in (1.2) is also expressed in the wave number representation as

$$\begin{aligned} \mathscr{H} &= -\frac{J}{2N^2} \sum_{ij} \sum_{\boldsymbol{qp}} \mathrm{e}^{\mathrm{i}(\boldsymbol{q}\cdot\boldsymbol{R}_i - \boldsymbol{p}\cdot\boldsymbol{R}_j)} \boldsymbol{S}_{\boldsymbol{q}} \cdot \boldsymbol{S}_{-\boldsymbol{p}} \\ &= -\frac{J}{2N^2} \sum_{ij} \sum_{\boldsymbol{qp}} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{R}_i - \boldsymbol{R}_j) + \mathrm{i}(\boldsymbol{q}-\boldsymbol{p})\cdot\boldsymbol{R}_j} \boldsymbol{S}_{\boldsymbol{q}} \cdot \boldsymbol{S}_{-\boldsymbol{p}} = -\frac{1}{2N} \sum_{\boldsymbol{q}} J(\boldsymbol{q}) \boldsymbol{S}_{\boldsymbol{q}} \cdot \boldsymbol{S}_{-\boldsymbol{q}} \end{aligned} \tag{1.22}$$

where $J(\boldsymbol{q})$ is defined as a following sum of nearest neighbor sites j, as given by

$$J(\boldsymbol{q}) = J {\sum_j}' \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_{ij}}, \quad (\boldsymbol{R}_{ij} = \boldsymbol{R}_i - \boldsymbol{R}_j)$$

We show below that the magnetic properties of this model are derived from the above amplitude conservation (1.19). Note that the following fluctuation dissipation theorem is satisfied at high temperatures:

$$(g\mu_{\mathrm{B}})^2 \frac{1}{3} \langle \mathbf{S}_{\mathbf{q}} \cdot \mathbf{S}_{-\mathbf{q}} \rangle = k_{\mathrm{B}} T \chi(\boldsymbol{q}). \tag{1.23}$$

Equation (1.13) is then regarded as the condition that the wave vector dependence magnetic susceptibility $\chi(\boldsymbol{q})$ has to satisfy. What we have to do next is to find the wave vector dependence of $\chi(\boldsymbol{q})$.

Let us next deal with an effect of externally applied magnetic field $H_{\boldsymbol{q}}$ along z-axis with spatial modulation of a wave vector $\boldsymbol{q}$. The Zeeman energy of this effect is given as

$$\begin{aligned} \mathscr{H}_1 &= \frac{1}{2} g\mu_{\mathrm{B}} (H_{-\boldsymbol{q}} S_{\boldsymbol{q}}^z + H_{\boldsymbol{q}} S_{-\boldsymbol{q}}^z) \\ &= -g\mu_{\mathrm{B}} \sum_i [H_{\boldsymbol{q}}' \cos(\boldsymbol{q}\cdot\boldsymbol{R}_i) + H_{\boldsymbol{q}}'' \sin(\boldsymbol{q}\cdot\boldsymbol{R}_i)] \, S_i^z, \\ H_{\boldsymbol{q}} &= H_{\boldsymbol{q}}' - \mathrm{i} H_{\boldsymbol{q}}'' \end{aligned} \tag{1.24}$$

As an effect of this term, we expect the magnetic moments are induced in the system, that are proportional to $H_{\boldsymbol{q}}$ and $H_{-\boldsymbol{q}}$ with the same wave vectors $\pm\boldsymbol{q}$. The magnetic susceptibilities $\chi(\boldsymbol{q})$ and $\chi(-\boldsymbol{q})$ are obtained as their coefficients. Each spin operator with wave vector $\boldsymbol{q}$ or $-\boldsymbol{q}$ is then defined as a sum of the mean value and the fluctuation.

$$\boldsymbol{S}_{\boldsymbol{q}} = \langle \boldsymbol{S}_{\boldsymbol{q}} \rangle + \delta \boldsymbol{S}_{\boldsymbol{q}}, \quad \boldsymbol{S}_{-\boldsymbol{q}} = \langle \boldsymbol{S}_{-\boldsymbol{q}} \rangle + \delta \boldsymbol{S}_{-\boldsymbol{q}} \tag{1.25}$$

For other operators with different wave vectors, $\boldsymbol{p} \neq \pm\boldsymbol{q}$, their mean values are assumed to be zero ($\langle \boldsymbol{S}_{\pm\boldsymbol{p}} \rangle = 0$).

In the mean field approximation, only the linear terms with respect to fluctuations are retained, whereas the rest are neglected. The Hamiltonian is then written as follows:

$$\mathscr{H}_{MF} = -\frac{1}{N} J(q) \left(\langle \boldsymbol{S}_{\boldsymbol{q}} \rangle \cdot \boldsymbol{S}_{-q} + \langle \boldsymbol{S}_{-q} \rangle \cdot \boldsymbol{S}_{\boldsymbol{q}} \right) + \frac{1}{2} g\mu_{\mathrm{B}} (H_{-\boldsymbol{q}} S^z_{\boldsymbol{q}} + H_{\boldsymbol{q}} S^z_{-\boldsymbol{q}}) \tag{1.26}$$

It is easy to see that the above Hamiltonian is effectively equivalent with the model in the absence of the magnetic field, provided that the external magnetic field $\boldsymbol{H}_{\boldsymbol{q}}$ is replaced by

$$H_{\boldsymbol{q}} \to H_{\mathrm{eff},\boldsymbol{q}} = H_{\boldsymbol{q}} - \frac{2}{N g\mu_{\mathrm{B}}} J(q) \langle S^z_{\boldsymbol{q}} \rangle \tag{1.27}$$

The magnetic moment induced by the above spatially modulated effective magnetic field is given then by

$$M_{\boldsymbol{q}} = -g\mu_{\mathrm{B}} \langle \boldsymbol{S}_{\boldsymbol{q}} \rangle = \frac{1}{2} \chi_{\mathrm{loc}}(T) \boldsymbol{H}_{\mathrm{eff},\boldsymbol{q}}, \tag{1.28}$$

where $\chi_{\mathrm{loc}}(T)$ represents a local magnetic susceptibility defined against the local magnetic field acting on each atomic site in the crystal.

Substituting the effective field (1.27) into the right-hand side of (1.28), we are led to the expression

$$M_{\boldsymbol{q}} = \frac{\chi_{\mathrm{loc}}(T)}{1 - \chi_{\mathrm{loc}}(T) J(q)/N(g\mu_{\mathrm{B}})^2} \frac{1}{2} \boldsymbol{H}_{\boldsymbol{q}} = \chi(\boldsymbol{q}) \frac{\boldsymbol{H}_{\boldsymbol{q}}}{2}, \tag{1.29}$$

where the wave vector-dependent magnetic susceptibility $\chi(\boldsymbol{q})$ is defined as a coefficient of $H_{\boldsymbol{q}}/2$, given as

$$\begin{aligned} \chi(\boldsymbol{q}) &= \frac{\chi_{\mathrm{loc}}(T)}{1 - \chi_{\mathrm{loc}}(T) J(q)/N(g\mu_{\mathrm{B}})^2} \\ &= \frac{N(g\mu_{\mathrm{B}})^2}{[N(g\mu_{\mathrm{B}})^2/\chi_{\mathrm{loc}}(T) - J(0)] + J(0) - J(q)}. \end{aligned} \tag{1.30}$$

From the condition that both sides agree with each other for $\boldsymbol{q} = \boldsymbol{0}$, (1.30) is also rewritten in the form

$$\chi(\boldsymbol{q}) = \frac{1}{1/\chi(0) + [J(0) - J(\boldsymbol{q})]/N(g\mu_{\mathrm{B}})^2}, \tag{1.31}$$

where $\chi(0)$ is given by

$$\frac{1}{\chi(0)} = \frac{1}{\chi_{\rm loc}(T)} - \frac{J(0)}{N_0(g\mu_{\rm B})^2}. \tag{1.32}$$

By putting the above (1.31) into (1.23), the spin amplitude conservation (1.19) is finally given as

$$\frac{T}{N}\sum_{\boldsymbol{q}} \frac{1}{N/\chi(0) + [J(0) - J(q)]/(g\mu_{\rm B})^2} = \frac{1}{3}S(S+1). \tag{1.33}$$

Magnetic properties derived from the above equation are shown below.

- The critical temperature lower than that derived in the mean field approximation. In this formalism, T_c is determined by the condition,

$$k_{\rm B}T_c\frac{1}{N}\sum_{\boldsymbol{q}} \frac{1}{J(0) - J(q)} = \frac{1}{3}S(S+1). \tag{1.34}$$

  It is derived by assuming $\chi^{-1}(0) = 0$ at $T = T_c$ in (1.33). To compare the result with (1.15) for T_c in the mean field approximation, note that the following properties are satisfied for $J(q)$:

$$\sum_{\boldsymbol{q}} J(q) = 0, \quad J(0) = \zeta J, \quad \therefore\ \frac{1}{N}\sum_{\boldsymbol{q}}[J(0) - J(q)] = \zeta J \tag{1.35}$$

  Mathematically, it is also known that the following inequality is generally satisfied, as far as all the a_i are positive.

$$\frac{1}{n}\left(\frac{1}{a_1} + \frac{1}{a_2} + \cdots + \frac{1}{a_n}\right) \geq \frac{n}{(a_1 + a_2 + \cdots + a_n)} \tag{1.36}$$

  If we let $J(0) - J(q)$ and N correspond to a_i and n, we are led to the inequality

$$\frac{S(S+1)}{3k_{\rm B}T_c} = \frac{1}{N}\sum_{\boldsymbol{q}} \frac{1}{J(0) - J(\boldsymbol{q})} \geq \frac{N}{\sum_{\boldsymbol{q}}[J(0) - J(q)]} = \frac{1}{\zeta J} = \frac{S(S+1)}{3k_{\rm B}T_c^{\rm MF}},$$

  where the critical temperature in the mean field approximation is denoted by $T_c^{\rm MF}$. Owing to the effect of fluctuations, the critical temperature T_c is obtained lower than $T_c^{\rm MF}$, i.e., $T_c \leq T_c^{\rm MF}$ is satisfied.
- Temperature dependence of magnetic susceptibility $\chi(T)$ around the critical point. The temperature dependence of the magnetic susceptibility is evaluated by solving (1.33) for $\chi(0)$ as a function of temperature. At high temperatures, its inverse, $\chi^{-1}(0)$, has to increase proportional to T, because the right-hand side is constant.

To find the temperature dependence around the critical temperature, $T_c \lesssim T$, let us rewrite (1.33) as

$$\frac{T}{N}\sum_{\boldsymbol{q}}\frac{1}{N/\chi(0)+[J(0)-J(\boldsymbol{q})]/(g\mu_{\rm B})^2}=\frac{T_c}{N}\sum_{\boldsymbol{q}}\frac{1}{[J(0)-J(\boldsymbol{q})]/(g\mu_{\rm B})^2}, \tag{1.37}$$

where the right-hand side is replaced by the left-hand side of (1.34). By subtraction of the same value, $(T/N)\sum_{\boldsymbol{q}}\{[J(0)-J(\boldsymbol{q})]/(g\mu_{\rm B})^2\}^{-1}$, from both the sides, (1.37) is further written in the form

$$\frac{T}{N}\sum_{\boldsymbol{q}}\left(\frac{1}{N/\chi(0)+[J(0)-J(\boldsymbol{q})]/(g\mu_{\rm B})^2}-\frac{1}{[J(0)-J(\boldsymbol{q})]/(g\mu_{\rm B})^2}\right)$$
$$=\frac{(T_c-T)}{N}\sum_{\boldsymbol{q}}\frac{1}{[J(0)-J(\boldsymbol{q})]/(g\mu_{\rm B})^2}\equiv c(T_c-T) \tag{1.38}$$

where the constant c is defined as the summation over $\boldsymbol{q}$ on the right-hand side. The wave vector summation of the above left-hand side is easily evaluated by assuming the quadratic dependence, $[J(0)-J(q)]/(g\mu_{\rm B})^2 = Aq^2$, around the origin. The result is given as

$$T\frac{4\pi V}{(2\pi)^3N}\int_0^{q_B}{\rm d}q\, q^2\left(\frac{1}{y+Aq^2}-\frac{1}{Aq^2}\right)=-\frac{Tv_0}{2\pi^2A}y\int_0^{q_B}{\rm d}q\,\frac{1}{y+Aq^2}$$
$$=-\frac{Tv_0}{2\pi^2A}\sqrt{\frac{y}{A}}\tan^{-1}\left(\sqrt{\frac{A}{y}}q_B\right)\simeq-\frac{Tv_0}{4\pi A}\sqrt{\frac{y}{A}}=c(T_c-T), \tag{1.39}$$

where we have defined $y = N/\chi(0)$ and $v_0 = V/N$ for volume per magnetic ion. The following $(T-T_c)^2$-linear temperature dependence of $y \propto 1/\chi(0)$ is finally derived in this region.

$$y=\left(\frac{4\pi c}{v_0}\right)^2\frac{A^3}{T^2}(T-T_c)^2 \tag{1.40}$$

The behavior different from the result of the mean field theory is characteristic of the critical phenomena.

We have shown two different treatments of magnetic properties of localized Heisenberg magnets. The first treatment is particularly concerned only with the order parameter induced in the system. In the second approach, the effects of fluctuations are explicitly taken into account. As a result, the critical temperature is lowered and more reasonable temperature dependence of the magnetic susceptibility can be derived around $T = T_c$.

1.3 Band Theoretical Approach

In this section, we give a brief overview of the Stoner-Wohlfarth (SW) theory of itinerant electron magnetism. It is essentially an application of the electron theory of metals in solid state physics. The theory is based on the following theoretical model known as the Hubbard Hamiltonian:

$$\mathscr{H} = \mathscr{H}_0 + U \sum_i n_{i\uparrow} n_{i\downarrow}, \quad \mathscr{H}_0 = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} = \sum_{k\sigma} \varepsilon_{\boldsymbol{k}} c_{k\sigma}^\dagger c_{k\sigma}, \tag{1.41}$$

where $\mathscr{H}_0$ represents the single electron hopping between magnetic ions in a crystal. Symbols $c_{i\sigma}^\dagger$ and $c_{j\sigma}$ of this term are the creation and the annihilation operators, respectively, for conduction electrons of ith and jth sites with spin direction σ. Among them the following anti-commutation relations are satisfied:

$$\{c_{j\sigma}, c_{j\sigma'}^\dagger\} = \delta_{ij}\delta_{\sigma,\sigma'}, \quad \{c_{i\sigma}, c_{j\sigma'}\} = \{c_{i\sigma}^\dagger, c_{j\sigma}^\dagger\} = 0 \tag{1.42}$$

Operators and the single electron energy for the Bloch state with the wave number $\boldsymbol{k}$ are denoted by $c_{\boldsymbol{k}\sigma}^\dagger$, $c_{\boldsymbol{k}\sigma}$, and $\varepsilon_{\boldsymbol{k}}$. The second term, responsible for the origin of magnetism, is the repulsive Coulomb interaction between electrons with different spin directions on the same atomic site.

In the SW theory, the appearance of itinerant electron magnetism and its various properties are understood associated with the changes of occupation numbers of conduction electrons around the Fermi energy. The substantial difficulty of the theory stems from this idea. The difference in the numbers of conduction electrons with up and down spin directions is regarded as the origin of the magnetism. The magnetization and the total number of electrons are therefore given as

$$\begin{aligned} M &= -\frac{1}{2}\sum\nolimits_k \langle n_{k\uparrow} - n_{k\downarrow}\rangle = -\frac{N_0}{2}\langle n_\uparrow - n_\downarrow\rangle \\ N &= \sum\nolimits_k \langle n_{k\uparrow} + n_{k\downarrow}\rangle = N_0 \langle n_\uparrow + n_\downarrow\rangle, \end{aligned} \tag{1.43}$$

where we have defined M as a difference of the average numbers of electrons with up and down electrons. The magnetization is given by $2\mu_B M$. In terms of M and N, the average numbers of $\langle n_\uparrow\rangle$ and $\langle n_\downarrow\rangle$ are defined by

$$\begin{aligned} \langle n_\uparrow\rangle &= \frac{1}{2N_0}(N - 2M) \\ \langle n_\downarrow\rangle &= \frac{1}{2N_0}(N + 2M) \end{aligned} \tag{1.44}$$

1.3.1 Hartree–Fock Approximation

It is generally very difficult to deal with the system described by the Hubbard Hamiltonian with a huge number of mutually interacting electrons. In the SW theory, the second interaction term in (1.41) is approximated by an effective magnetic field as given by

$$\begin{aligned} U\sum_i n_{i\uparrow}n_{i\downarrow} &\Longrightarrow U\sum_i (n_{i\uparrow}\langle n_\downarrow\rangle + n_{i\downarrow}\langle n_\uparrow\rangle - \langle n_\downarrow\rangle\langle n_\uparrow\rangle) \\ &= U\sum_{k\sigma} n_{k\sigma}\langle n_{-\sigma}\rangle - N_0 U\langle n_\downarrow\rangle\langle n_\uparrow\rangle \\ &= I\sum_{k\sigma}\left(\frac{N}{2} - \sigma M\right)c^\dagger_{k\sigma}c_{k\sigma} - I\left(\frac{N^2}{4} - M^2\right), \quad (I = U/N_0) \end{aligned} \tag{1.45}$$

On the other hand, in the presence of a uniform external magnetic field H, the Zeeman energy is written in the form

$$-M^z H = -\sum_k \frac{h}{2}(c^\dagger_{i\uparrow}c_{i\uparrow} - c^\dagger_{i\downarrow}c_{i\downarrow}) = -\sum_{k\sigma}\sigma\frac{h}{2}c^\dagger_{k\sigma}c_{k\sigma}, \quad (h = 2\mu_\mathrm{B} H) \tag{1.46}$$

Comparing (1.45) with (1.46), the effect of the repulsive Coulomb interaction is regarded as the presence of extra magnetic field, $2IM$. The following effective Hamiltonian is derived, with substitution of (1.45) for (1.41), in the presence of the external magnetic field.

$$\begin{aligned} \mathscr{H} &= \textstyle\sum_{k\sigma}(\varepsilon_{k\sigma} - \mu)c^\dagger_{k\sigma}c_{k\sigma} - I\left(\frac{N^2}{4} - M^2\right) \\ \varepsilon_{k\sigma} &= \varepsilon_k + \frac{IN}{2} - \sigma\Delta, \quad \Delta = IM + \frac{h}{2} \end{aligned} \tag{1.47}$$

Thermodynamic properties of this system are now evaluated according to the conventional procedure of statistical mechanics.

The free energy of our system of non-interacting Fermions is given as

$$F(h,\mu,T) = IM^2 + F_0, \quad F_0(h,\mu,T) = -k_\mathrm{B}T\sum_{k\sigma}\ln(1 + \mathrm{e}^{-(\varepsilon_{k\sigma}-\mu)/k_\mathrm{B}T}) \tag{1.48}$$

The total electron number N and the magnetization M are related to the chemical potential μ and the external magnetic field h, respectively, by the following thermodynamic relations:

$$\begin{aligned}
-\frac{\partial F}{\partial \mu} &= N(h,\mu,T) = \sum_{k\sigma} f(\varepsilon_{k\sigma}) \\
&= \int \mathrm{d}\varepsilon \rho(\varepsilon)[f(\varepsilon+\Delta)+f(\varepsilon-\Delta)] \\
-\frac{\partial F}{\partial h} &= M(h,\mu,T) = -\frac{1}{2}\sum_{k\sigma} \sigma f(\varepsilon_{k\sigma}) \\
&= -\frac{1}{2}\int \mathrm{d}\varepsilon \rho(\varepsilon)[f(\varepsilon+\Delta)-f(\varepsilon-\Delta)]
\end{aligned} \tag{1.49}$$

where the Fermi distribution function is defined as

$$f(\varepsilon) = \frac{1}{\mathrm{e}^{(\varepsilon-\mu)/k_\mathrm{B}T}+1}. \tag{1.50}$$

1.3.2 Free Energy of Stoner-Wohlfarth Theory

In our treatment of systems showing magnetic phase transitions at some finite temperature, it is convenient to introduce the free energy $F(M,N)$, in place of $F(h,\mu,T)$, with respect to variables M and N. It is useful for our intuitive understanding of magnetic phase transitions. For example, the free energy then becomes minimum at finite spontaneous magnetization M for ferromagnet. These two free energies are related by the Legendre transformation,

$$F(M,N,T) = F(h,\mu,T) + hM + \mu N, \tag{1.51}$$

where variables h and μ on the right-hand side are eliminated by using (1.49) as functions of M and N. Then the following new thermodynamic relations are satisfied for new variables:

$$\begin{aligned}
\frac{\partial F(M,N,T)}{\partial N} &= \mu + \left[\frac{\partial F(h,\mu,T)}{\partial \mu} + N\right]\frac{\partial \mu}{\partial N} \\
&\quad + \left[\frac{\partial F(h,\mu,T)}{\partial h} + M\right]\frac{\partial h}{\partial N} = \mu \\
\frac{\partial F(M,N,T)}{\partial M} &= h + \left[\frac{\partial F(h,\mu,T)}{\partial \mu} + N\right]\frac{\partial \mu}{\partial M} \\
&\quad + \left[\frac{\partial F(h,\mu,T)}{\partial h} + M\right]\frac{\partial h}{\partial M} = h
\end{aligned} \tag{1.52}$$

As will be shown later, the new free energy can be expanded in powers of M as far as the M dependence of the free energy in (1.51) is concerned.

$$F(M,T) = F(0,T) + \frac{1}{2}a(T)M^2 + \frac{1}{4}b(T)M^4 + \cdots \tag{1.53}$$

The first term on the right-hand side is the free energy at $M = 0$. In the SW theory, the free energy is expanded in powers of M and temperature T by regarding them as small parameters. As for the temperature dependence, the Sommerfeld expansion is applied to the following integral:

$$\int_{-\infty}^{\infty} \mathrm{d}\varepsilon g(\varepsilon) f(\varepsilon) = \int_{-\infty}^{\mu} \mathrm{d}\varepsilon g(\varepsilon) + \sum_{n=1} a_n (k_\mathrm{B}T)^{2n} g^{(2n-1)}(\mu), \tag{1.54}$$

where the integrand consists of a product of the Fermi distribution function $f(\varepsilon)$ and an arbitrary function $g(\varepsilon)$. Precise values of expansion coefficients a_n are known, e.g., $a_1 = \pi^2/6$, in the second term.

For the derivation of the free energy (1.53), we need to evaluate (1.49) expanded in terms of small parameters, T, Δ, and $\delta\mu \equiv \mu - \varepsilon_\mathrm{F}$. The chemical potential in the non-magnetic ground state is denoted by ε_F. The energy of conduction electrons in the Fermi distribution function is then written as

$$\varepsilon_{\boldsymbol{k}\sigma} - \mu = (\varepsilon_{\boldsymbol{k}} - \sigma\Delta - \delta\mu) - \varepsilon_\mathrm{F} = \varepsilon_{\boldsymbol{k}} - (\delta\mu + \sigma\Delta).$$

If we always use the Fermi distribution function with fixed $\mu = \varepsilon_\mathrm{F}$, (1.49) is rewritten as follows:

$$\begin{aligned} N &= \int_{-\infty}^{\infty} \mathrm{d}\varepsilon\rho(\varepsilon)[f(\varepsilon - \Delta - \delta\mu) + f(\varepsilon + \Delta - \delta\mu)] \\ 2M &= \int_{-\infty}^{\infty} \mathrm{d}\varepsilon\rho(\varepsilon)[f(\varepsilon - \Delta - \delta\mu) - f(\varepsilon + \Delta - \delta\mu)] \end{aligned} \tag{1.55}$$

It is better to use the following equation, in place of the above first equation, being derived by subtracting both sides of the first line of (1.55) and the same but with $\Delta = 0$ and $\delta\mu = 0$.

$$\sum_{\sigma=\pm 1} \int_{-\infty}^{\infty} \mathrm{d}\varepsilon\rho(\varepsilon)[f(\varepsilon - \sigma\Delta - \delta\mu) - f(\varepsilon)] = 0. \tag{1.56}$$

Now the above (1.56) and the second equation of (1.55) are expanded in powers of $\delta\mu$, Δ, and $(k_\mathrm{B}T)^2$, as given by

$$\begin{aligned} &2\rho\delta\mu + \frac{2\pi^2}{3}\rho'(k_\mathrm{B}T)^2 + \rho'\Delta^2 + \cdots = 0 \\ &2M = 2\Delta\left[\rho + \frac{\pi^2}{3}\rho''(k_\mathrm{B}T)^2 + \cdots\right] + 2\rho'\Delta\delta\mu + \frac{1}{3}\rho''\Delta^3 + \cdots \\ &\quad = 2\rho\Delta\left[1 - \frac{\pi^2}{3}\left(\frac{\rho''}{\rho} - \frac{\rho'^2}{\rho^2}\right)(k_\mathrm{B}T)^2 + \cdots\right] + \left(\frac{\rho''}{3} - \frac{\rho'^2}{\rho}\right)\Delta^3 + \cdots \end{aligned} \tag{1.57}$$

The last line is obtained by putting $\delta\mu$ in terms of Δ^2 and $(k_B T)^2$ into the right-hand side of the second line. With the use of $\Delta = IM + h/2$, the last expression is finally converted in the form

$$h = \frac{\partial F}{\partial M} = a(T)M + b(T)M^3 + \cdots, \tag{1.58}$$

where the coefficients $a(T)$ and $b(T)$ are given by

$$\begin{aligned} a(T) &= \frac{2}{\rho} - 2I + \frac{\pi^2}{3}\left(\frac{\rho''}{\rho} - \frac{\rho'^2}{\rho^2}\right)(k_B T)^2 + \cdots, \\ b(T) &= \frac{1}{\rho^3}\left(\frac{\rho'^2}{\rho^2} - \frac{\rho''}{3\rho}\right) + \cdots. \end{aligned} \tag{1.59}$$

The M dependence of the free energy in (1.52) is obtained by integrating the thermodynamic relation (1.52) with respect to M.

The free energy (1.52) of the SW theory is given as a sum of two competitive contributions, i.e., the band energy resulting from the hopping of conduction electrons from an atomic site to site and the on-site repulsive Coulomb energy between electrons with opposite spin directions. The thermodynamic state is determined by its stability condition with respect to its variables. If it becomes stable for a state with finite magnetization M, ferromagnetism appears in the system. Since the variation of occupation numbers of conduction electrons is usually restricted within around the Fermi energy ε_F, magnetic properties are characterized in the form of the density of states around ε_F. The magnetic properties of the SW theory are therefore derived by the free energy (1.53) with coefficients $a(T)$ and $b(T)$ given by (1.59).

1.4 Magnetic Properties Derived from the SW Theory

Typical magnetic properties derived from the free energy (1.53) of the SW theory are summarized as follows:

- The condition of appearance of the spontaneous magnetization in the ground state is given as

$$a(0) < 0, \quad \text{or} \quad I\rho > 0. \tag{1.60}$$

  It is usually called the Stoner condition. When it is satisfied, the magnetism develops as the result of spin splitting of the conduction electron bands.
- Temperature dependence of the magnetic susceptibility is given as

$$\frac{1}{\chi_0(T)} \equiv \frac{\partial h}{\partial M} = \frac{1}{\chi_P(T)} - 2I, \tag{1.61}$$

where $\chi_{\rm P}(T)$ is magnetic susceptibility for Pauli paramagnets with $I = 0$, given as

$$\chi_{\rm P}(T) = \frac{\rho}{2}\left\{1 - \frac{\pi^2}{6}R(k_{\rm B}T)^2 + \cdots\right\}, \quad R = \frac{\rho''}{\rho} - \frac{\rho'^2}{\rho^2}. \tag{1.62}$$

The coefficient $a(T)$ in (1.58) corresponds to the inverse of magnetic susceptibility.

- The Curie temperature T_c is given as

$$kBT_c = \left[\frac{6(I\rho - 1)}{\pi^2 R}\right]^{1/2}. \tag{1.63}$$

It is given by the condition, $a(T_c) = 0$. The magnetic susceptibility shows divergence at $T = T_c$. With this T_c, the temperature dependence of the first coefficient $a(T)$ is written as

$$a(T) = a(0)\left(1 - \frac{T^2}{T_c^2}\right). \tag{1.64}$$

- The spontaneous magnetic moment in the ground state is given as

$$a(0)M + b(0)M^3 = 0, \ \therefore \ M_0 = \left[\frac{-a(0)}{b(0)}\right]^{1/2} = \left[\frac{2(I\rho - 1)}{\rho b(0)}\right]^{1/2} \propto T_c \tag{1.65}$$

It is determined by the magnetic isotherm (1.58) in the ground state for $h = 0$ by assuming $T = 0$. Comparing the result with (1.63) gives the relation $M_0 \propto T_c$.

- The temperature dependence of the spontaneous moment is given as

$$M(T) = \left[\frac{-a(T)}{b(T)}\right]^{1/2} \simeq \left[\frac{-a(0)}{b(0)}\right]^{1/2}\left[\frac{a(T)}{a(0)}\right]^{1/2} = M_0\left(1 - \frac{T^2}{T_c^2}\right)^{1/2} \tag{1.66}$$

Spontaneous magnetic moment at finite temperatures is also determined by the magnetic isotherm (1.58) for $h = 0$. The temperature dependence of the second coefficient $b(T)$ is assumed to be weak and is neglected here. It implies that the good linearity between M^2 and T^2 is satisfied in the wide range of temperature below T_c.

- Magnetic isotherm is given as

$$M^2(H, T) = -\frac{a(T)}{b(T)} + \frac{1}{b(T)}\frac{h}{M(H, T)}. \tag{1.67}$$

Therefore, the good linear relation between M^2 and H/M is satisfied in the case of small magnetic moment M. Observed results of M^2 from magnetization measurements are often plotted against H/M, called Arrott plot at present. It is also written in the form

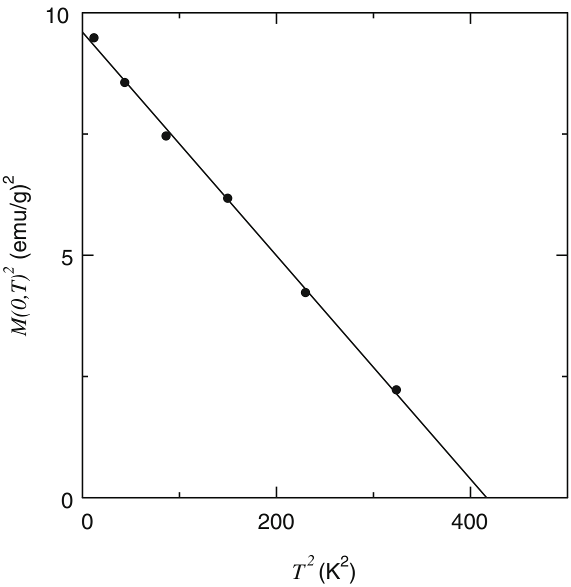

Fig. 1.1 Observed temperature dependence of the spontaneous magnetization square $M^2(0, T)$ of $ZrZn_2$, plotted against T^2, by Ogawa [9]

$$M^2(H, T) = M^2(0, 0)\left(1 - \frac{T^2}{T_c^2}\right) + M^2(0, 0)\frac{2\chi_0 H}{M(H, T)}, \tag{1.68}$$

where $\chi_0 = 1/[2b(T)M^2(0, 0)]$ is called differential magnetic susceptibility.

Stimulated by these theoretical investigations, a number of experimental studies were made around 1970 on itinerant electron ferromagnets with small induced magnetic moments such as $ZrZn_2$, Sc_3In, Ni_3Al, and MnSi. Good linearity of their Arrott plots have been actually confirmed for most of them except for MnSi. Typical T^2-linear dependence is also observed for spontaneous magnetic moments and fourth expansion coefficients $b(T)$ of the free energy in powers of M. As an example, the temperature dependence of the spontaneous magnetic moment of $ZrZn_2$ observed by Ogawa [9] is shown in Fig. 1.1, in agreement with the prediction of the SW theory. The T^2-linear behavior of $b(T)$ is also confirmed for $ZrTiZn_2$ and $ZrZn_{1.9}$ by Wohlfarth and de Chatel [10] and for Ni–Pt alloys by Beille et al. [11].

1.5 Summary

In Table 1.1, typical magnetic properties of itinerant ferromagnets predicted by the SW theory are compared with those of localized moment models. Differences in these properties are summarized as below.

Table 1.1 Characteristic magnetic properties of itinerant electron ferromagnets in comparison with local moment magnets

Magnetic properties	Local moment systems	Itinerant ferromagnets
$M/(N_0\mu_B)$	Integer	$\ll 1$
$M(H,T)$ versus H for $T/T_c \ll 1$	Saturated	Unsaturated
Arrott plot	Nonlinear	Linear
$M^2(0) - M^2(T)$	$\propto T^{3/2}$	$\propto T^2$
$\chi(T)$	Curie–Weiss law	Curie–Weiss law
p_{eff}/p_s	~ 1	$\gg 1$

1. The magnitude of magnetic moment per magnetic atom
   For insulator magnets in which the spin–orbit coupling is negligible, magnitude of atomic magnetic moment takes an integer or a half-integer in units of μ_B, because of the quantization of the angular momentum. For itinerant electron magnets, on the other hand, it can take any value, because it is determined by the spin splitting of conduction electron bands.
2. Magnetic isotherm in the ground state
   For localized moment magnets, magnetization is almost saturated at low temperatures. Therefore, it is little affected by external magnetic field. In the case of itinerant ferromagnets, it still shows increase with increass in the external magnetic field strength. It results from the increasing spin splitting, according to the SW theory.
3. Temperature dependence of spontaneous magnetization at low temperatures
   Spontaneous magnetization shows the $T^{3/2}$-linear decrease for localized moment ferromagnets at low temperatures, resulting from thermal spin-wave excitations. On the contrary, for weak itinerant ferromagnets, T^2-linear decreases are rather well observed, seeming to be in agreement with the SW theory.
4. Temperature dependence of magnetic susceptibility
   Curie–Weiss law temperature dependence is generally observed for both these magnets. The ratios of two magnetic moments, i.e., the effective moment p_{eff} estimated from the Curie constant of magnetic susceptibility and the spontaneous moment p_s, are of about 1 for localized moment magnets, while for itinerant ferromagnets, considerably larger values are obtained. In the SW theory, the different dependence proportional to $(T^2 - T_c^2)^{-1}$ is derived in disagreement with experiments.

To conclude, most magnetic properties seem to be well accounted by the SW theory, as long as they are in the ground state or in the magnetically ordered phase. An exceptional difficulty has been the Curie–Weiss law dependence of magnetic susceptibility. Efforts to overcome the difficulty have brought about a new theoretical development.

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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

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

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

$$\delta q \equiv q - \langle q \rangle, \quad \delta p \equiv p - \langle p \rangle, \tag{2.2}$$

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.

$$\langle \delta q^2 \rangle = \langle q^2 \rangle - \langle q^2 \rangle, \quad \langle \delta p^2 \rangle = \langle p^2 \rangle - \langle p^2 \rangle \tag{2.3}$$

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

$$\langle \delta q^2 \rangle = \frac{\int_{-\infty}^{\infty} \mathrm{d}q \mathrm{d}p \; q^2 \mathrm{e}^{-\mathscr{H}(q,p)/k_\mathrm{B}T}}{\int_{-\infty}^{\infty} \mathrm{d}q \mathrm{d}p \; \mathrm{e}^{-\mathscr{H}(q,p)/k_\mathrm{B}T}} = \frac{k_\mathrm{B}T}{m\omega^2} \tag{2.4}$$

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

Y. Takahashi, *Spin Fluctuation Theory of Itinerant Electron Magnetism*,
Springer Tracts in Modern Physics 253, DOI: 10.1007/978-3-642-36666-6_2,

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

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

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

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

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.

$$\langle \delta q^2 \rangle = k_\mathrm{B} T \chi \tag{2.7}$$

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

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

Between them, the following commutation relation is satisfied.

$$[b, b^\dagger] = 1 \tag{2.9}$$

The Hamiltonian is then represented by

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

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

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

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

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

It is easy to see that at high temperatures where $\hbar\omega/k_\mathrm{B}T \ll 1$ 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

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

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

$$\delta\phi(r) \equiv \phi(r) - \langle\phi(r)\rangle\,. \tag{2.14}$$

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

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

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

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

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

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

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].

$$\mathscr{H} = \mathscr{H}_0 + V(t), \quad V(t) = -g\mu_\mathrm{B} S^\alpha_{-q} B_\mathrm{e} \mathrm{e}^{-\mathrm{i}\omega t} \tag{2.18}$$

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^{α}_{-q} 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.

$$-\,\mathrm{i}\frac{\partial}{\partial t}\,|\,\Psi(t)\,\rangle = [\mathscr{H}_0 + V(t)]\,|\,\Psi(t)\,\rangle \tag{2.19}$$

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

$$\mathscr{H}_0\mathrm{e}^{-\mathrm{i}\mathscr{H}_0 t}\,|\,\Phi(t)\,\rangle + \mathrm{i}\mathrm{e}^{-\mathrm{i}\mathscr{H}_0 t}\frac{\partial}{\partial t}\,|\,\Phi(t)\,\rangle = [\mathscr{H}_0 + V(t)]\mathrm{e}^{-\mathrm{i}\mathscr{H}_0 t}\,|\,\Phi(t)\,\rangle\,,$$

$$\therefore\ \mathrm{i}\frac{\partial}{\partial t}\,|\,\Phi(t)\,\rangle = V_H(t)\,|\,\Phi(t)\,\rangle\,, \tag{2.20}$$

where $V_H(t)$ is defined by

$$V_H(t) = \mathrm{e}^{\mathrm{i}\mathscr{H}_0 t}V(t)\mathrm{e}^{-\mathrm{i}\mathscr{H}_0 t} = -g\mu_{\mathrm{B}}S^{\alpha}_q(t)B_e\mathrm{e}^{-\mathrm{i}\omega t}, \quad S^{\alpha}_q(t) \equiv \mathrm{e}^{i\mathscr{H}_0 t}S^{\alpha}_q\mathrm{e}^{-\mathrm{i}\mathscr{H}_0 t} \tag{2.21}$$

The solution of (2.20) is formally given by

$$\begin{aligned}|\,\Phi(t)\,\rangle &= |\,\Phi(-\infty)\,\rangle - \mathrm{i}\int_{-\infty}^{t}\mathrm{d}t'\,V_H(t')|\Phi_\nu(t')\rangle = \Bigg[1 - \mathrm{i}\int_{-\infty}^{t}\mathrm{d}t'\,V_H(t') \\ &\quad + (-\mathrm{i})^2\int_{-\infty}^{t}\mathrm{d}t'\,V_H(t')\int_{-\infty}^{t'}\mathrm{d}t''\,V_H(t'') + \cdots\Bigg]\,|\,\Phi(-\infty)\,\rangle\,,\end{aligned} \tag{2.22}$$

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 $\langle\Phi(t)|S^{\beta}_q(t)|\Phi(t)\rangle$ is therefore given by

$$\begin{aligned}g\mu_{\mathrm{B}}\langle S^{\beta}_q\rangle(t) &\equiv g\mu_{\mathrm{B}}\langle\Phi(t)|S^{\beta}_q(t)|\Phi(t)\rangle \\ &= g\mu_{\mathrm{B}}\langle\Phi(-\infty)|\left[1 + \mathrm{i}\int_{-\infty}^{t}\mathrm{d}t'\,V_H(t') + \cdots\right]S^{\beta}_q(t) \\ &\qquad \times\left[1 - \mathrm{i}\int_{-\infty}^{t}\mathrm{d}t'\,V_H(t') + \cdots\right]|\,\Phi(-\infty)\,\rangle \\ &= \mathrm{i}g\mu_{\mathrm{B}}\int_{-\infty}^{t}\mathrm{d}t'\,\langle[V_H(t'), S^{\beta}_q(t)]\rangle + \cdots\,.\end{aligned} \tag{2.23}$$

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,

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

where we have defined the dynamical magnetic susceptibility by

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

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

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

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

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

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.

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

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

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

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.,

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

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

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

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

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

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 electons 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.

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

It consists with two contributions, $F_{\rm SW}$ 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, $1/\chi_0(\boldsymbol{q})$ 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 $\boldsymbol{M}(\boldsymbol{r})$ as a function of $\boldsymbol{r}$. Variables $\boldsymbol{M}_{\boldsymbol{q}}$ are the Fourier transform of $\boldsymbol{M}(\boldsymbol{r})$. The set of variables $\boldsymbol{M}_{\boldsymbol{q}}$ with wave-vector $\boldsymbol{q}$ throughout the whole Brillouin zone are denoted by $\{\boldsymbol{M}_{\boldsymbol{q}}\}$. The free energy of the system is then evaluated as follows.

$$\begin{aligned} e^{-F(M,T)/k_BT} &= \sum_{\{M_q\}} \exp[-\Psi(\{M_q\})/k_BT] \\ &= e^{-F_{SW}(M,T)/k_BT} \sum_{\{M_q\}} \exp[-\Phi(\{M_q\})/k_BT] \end{aligned} \tag{2.33}$$

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.

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

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

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

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.

$$\begin{aligned} e^{-F_0/k_BT} &= \sum_{\{M_q\}} e^{-\Phi_0(\{M_q\})/k_BT} = \prod_q \int dM_q e^{-\beta\Phi_0(\{M_q\})} \\ &= \prod_q \left[\left(\frac{\pi k_BT}{\Omega_q^{\parallel}}\right)^{1/2}\left(\frac{\pi k_BT}{\Omega_q^{\perp}}\right)\right] \\ F_0 &= -k_BT\sum_{q\neq\mathbf{0}}\left[\frac{1}{2}\log\left(\frac{\pi k_BT}{\Omega_q^{\parallel}}\right) + \log\left(\frac{\pi k_BT}{\Omega_q^{\perp}}\right)\right] \end{aligned} \tag{2.36}$$

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

$$\begin{aligned} e^{-\Delta F/k_BT} &= e^{F_0/k_BT}\sum_{\{M_q\}} \exp[-\Phi(\{M_q\})/k_BT] \\ &= e^{F_0/k_BT}\sum_{\{M_q\}} e^{-\Phi_0(\{M_q\})/k_BT} e^{-[\Phi(\{M_q\})-\Phi_0(\{M_q\})]/k_BT} \\ &= \langle e^{-(\Phi-\Phi_0)/k_BT}\rangle, \end{aligned} \tag{2.37}$$

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

$$\langle \dots \rangle = e^{F_0/k_B T} \sum_{\{\boldsymbol{M_q}\}} e^{-\Phi_0(\{\boldsymbol{M_q}\})/k_B T} \dots \tag{2.38}$$

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

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

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

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

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

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

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

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

with using the following relations.

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

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

$$\begin{aligned} \langle\Phi_a\rangle &= \sum_{\boldsymbol{q}\neq 0}\frac{1}{2\chi_0(\boldsymbol{q})}\langle\boldsymbol{M_q}\cdot\boldsymbol{M_{-q}}\rangle, \\ \langle\Phi_b\rangle &= \frac{1}{4}b\sum_{\{\boldsymbol{q}_i\}}\langle\boldsymbol{M}_{\boldsymbol{q}_1}\cdot\boldsymbol{M}_{\boldsymbol{q}_2}\boldsymbol{M}_{\boldsymbol{q}_3}\cdot\boldsymbol{M}_{\boldsymbol{q}_4}\rangle + \cdots \end{aligned} \tag{2.44}$$

The harmonic term is then simply evaluated as follows.

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

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

$$\begin{aligned} \langle \Phi \rangle_b = &\frac{b}{4} M_0^2 \sum_{q \neq 0} \left[2\langle \boldsymbol{M_q} \cdot \boldsymbol{M_{-q}} \rangle + 4\langle M_{\boldsymbol{q}}^{\parallel} \cdot M_{-\boldsymbol{q}}^{\parallel} \rangle \right] \\ &+ \frac{b}{4} \sum_{q,q' \neq 0} \Bigg[\langle \boldsymbol{M_q} \cdot \boldsymbol{M_{-q}} \rangle \langle \boldsymbol{M_{q'}} \cdot \boldsymbol{M_{-q'}} \rangle \\ &\qquad + 2 \sum_{\mu = \perp, \parallel} \langle M_{\boldsymbol{q}}^{\mu} \cdot M_{-\boldsymbol{q}}^{\mu} \rangle \langle M_{\boldsymbol{q'}}^{\mu} \cdot M_{-\boldsymbol{q'}}^{\mu} \rangle \Bigg] \end{aligned} \tag{2.46}$$

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

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

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

The variational free energy is finally given in the form.

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

Minimum Conditions of the Free Energy It is now possible to determine the variational parameters $\Omega_q^{\parallel}$ and $\Omega_q^{\perp}$ 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 $\Omega_q^{\perp}$ ($\mathbf{q} \neq \mathbf{0}$), i.e.

$$\Omega_q^{\perp} = \frac{1}{2\chi_0(\boldsymbol{q})} + \frac{1}{2} b M^2 + \frac{1}{4} b k_{\mathrm{B}} T \sum_{\boldsymbol{q}' \neq 0} \left(\frac{1}{\Omega_{q'}^{\parallel}} + \frac{4}{\Omega_{q'}^{\perp}} \right). \tag{2.49}$$

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

$$\Omega_q^{\parallel} = \frac{1}{2\chi_0(\boldsymbol{q})} + \frac{3}{2} b M^2 + \frac{1}{4} b k_{\mathrm{B}} T \sum_{\boldsymbol{q}' \neq 0} \left(\frac{3}{\Omega_{q'}^{\parallel}} + \frac{2}{\Omega_{q'}^{\perp}} \right). \tag{2.50}$$

- The condition for M, i.e.

$$\frac{H}{M} = \frac{1}{\chi_0(\mathbf{0})} + b M^2 + \frac{1}{2} b k_{\mathrm{B}} T \sum_{\boldsymbol{q}' \neq 0} \left(\frac{3}{\Omega_{q'}^{\parallel}} + \frac{2}{\Omega_{q'}^{\perp}} \right). \tag{2.51}$$

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

$$\Omega_q = \frac{1}{2\chi_0(\boldsymbol{q})} + \frac{5}{4} b k_{\mathrm{B}} T \sum_{\boldsymbol{q}' \neq \mathbf{0}} \frac{1}{\Omega_{q'}}. \tag{2.52}$$

In the uniform $\boldsymbol{q} = \mathbf{0}$ 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

$$\langle \boldsymbol{M}_{\boldsymbol{q}} \cdot \boldsymbol{M}_{-\boldsymbol{q}} \rangle = \langle |\boldsymbol{M}_{\boldsymbol{q}}^{\parallel}|^2 \rangle + \langle |\boldsymbol{M}_{\boldsymbol{q}}^{\perp}|^2 \rangle = \frac{3 k_{\mathrm{B}} T}{2\Omega_q}. \tag{2.53}$$

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

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

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.

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

   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

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

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.

$$\frac{1}{\chi(T)} = \frac{5}{3}b\sum_{\boldsymbol{q}} \left[\langle \boldsymbol{M}_{\boldsymbol{q}} \cdot \boldsymbol{M}_{-\boldsymbol{q}} \rangle(\chi^{-1}, T) - \langle \boldsymbol{M}_{\boldsymbol{q}} \cdot \boldsymbol{M}_{-\boldsymbol{q}} \rangle(0, T_c)\right] \tag{2.57}$$

In quantum mechanical treatments, thermal amplitudes in the right hand side is evaluated in terms of the imaginary part of the dynamical magnetic susceptibility $\chi(\boldsymbol{q}, \omega)$, 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.

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

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

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

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.

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

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

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

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,

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

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

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

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

$$\frac{1}{\chi_0(\boldsymbol{q})} - \frac{1}{\chi_0(\boldsymbol{0})} = Aq^2 + \cdots . \tag{2.64}$$

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

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

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.

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

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.

$$\begin{aligned}\sum_q\left(\frac{1}{Aq^2+2bM_0^2}-\frac{1}{Aq^2}\right)&=-\frac{8\pi VbM_0^2}{(2\pi)^3A^2}\int_0^{q_\mathrm{B}}\mathrm{d}q\frac{1}{q^2+2bM_0^2/A}\\&=-\frac{bM_0^2V}{\pi^2A^2}\sqrt{\frac{A}{2bM_0^2}}\tan^{-1}\sqrt{\frac{Aq_\mathrm{B}^2}{2bM_0^2}},\end{aligned}\tag{2.67}$$

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.,

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

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

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

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 $-c_2(T)\to 0$ (for $T\to T_c$) 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

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

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

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

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

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

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.

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

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

$$\begin{aligned}\langle S_{\rm loc}^2\rangle &= \langle S_{\rm loc}^2\rangle_{\rm Z} + \langle S_{\rm loc}^2\rangle_{\rm T},\\ \langle S_{\rm loc}^2\rangle_{\rm Z} &= \frac{3}{N_0^2}\sum_{\boldsymbol{q}}\int_0^\infty\frac{d\omega}{\pi}{\rm Im}\chi(\boldsymbol{q},\omega),\\ \langle S_{\rm loc}^2\rangle_{\rm T} &= \frac{6}{N_0^2}\sum_{\boldsymbol{q}}\int_0^\infty\frac{d\omega}{\pi}n(\omega){\rm Im}\chi(\boldsymbol{q},\omega).\end{aligned} \tag{2.74}$$

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.

$$\mathrm{Im}\chi(\mathbf{q},\omega) = \chi(\mathbf{q},0)\frac{\omega\Gamma_q}{\omega^2+\Gamma_q^2}, \quad \chi(\mathbf{q},0) = \chi(\mathbf{0},0)\frac{\kappa^2}{\kappa^2+q^2},$$
$$\Gamma_q = \Gamma_0 q(\kappa^2+q^2), \quad q \equiv |\mathbf{q}| \tag{2.75}$$

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.

$$y = \frac{\kappa^2}{q_\mathrm{B}^2} = \frac{N_0}{2T_A\chi(\mathbf{0},0)} = \frac{h}{2T_A\sigma}. \tag{2.76}$$

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 $\chi^{-1}(\boldsymbol{q},0)$ 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

$$\frac{N_0}{\chi(\mathbf{q}_B,0)} = \frac{N_0(1+q_\mathrm{B}^2/\kappa^2)}{\chi(\mathbf{0},0)} = \frac{N_0(1+1/y)}{\chi(\mathbf{0},0)} \simeq \frac{N_0}{\chi(0,0)y} \equiv 2k_\mathrm{B}T_A, \tag{2.77}$$

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

$$\Gamma_{q_\mathrm{B}} = \Gamma_0 q_\mathrm{B}(\kappa^2+q_\mathrm{B}^2) = \Gamma_0 q_\mathrm{B}^3(y+1) \simeq \Gamma_0 q_\mathrm{B}^3 = 2\pi k_\mathrm{B}T_0, \tag{2.78}$$

as the width Γ_q of the ω dependence at the zone-boundary wave vector $q = q_\mathrm{B}$.

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

$$\chi(\mathbf{q},0)=\chi(\mathbf{0},0)\frac{\kappa^2}{\kappa^2+q^2}=\frac{N_0}{2T_A(y+x^2)},$$
$$\Gamma_q=\Gamma_0 q(q^2+\kappa^2)=2\pi T_0 x(x^2+y),\quad x\equiv q/q_{\mathrm{B}},\tag{2.79}$$

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

$$\frac{1}{N_0}\sum_{\boldsymbol{q}}=\frac{4\pi V}{(2\pi)^3N_0}\int_0^{q_{\mathrm{B}}}\mathrm{d}q\,q^2=\frac{4\pi q_{\mathrm{B}}^3V}{(2\pi)^3N_0}\int_0^1\mathrm{d}x\,x^2=3\int_0^1\mathrm{d}x\,x^2\tag{2.80}$$

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

$$t=\frac{T}{T_0}.\tag{2.81}$$

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,

$$\mathrm{Im}\chi(\mathbf{q},\omega)=\chi(\mathbf{q},0)\frac{\omega\Gamma_q}{\omega^2+\Gamma_q^2}$$
$$=\frac{N_0}{2T_A}\frac{1}{y+x^2}\frac{\xi u(x)}{\xi^2+u^2(x)},\quad u(x)\equiv x(y+x^2)/t\tag{2.82}$$

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.

$$\langle S^2\rangle_{\mathrm{T}}(y,t)=\frac{18T_0}{T_A}\int_0^1\mathrm{d}xx^3\int_0^\infty\mathrm{d}\xi\frac{\xi}{\mathrm{e}^{2\pi\xi}-1}\frac{1}{\xi^2+u^2}=\frac{9T_0}{T_A}A(y,t)$$
$$A(y,t)\equiv\int_0^1\mathrm{d}xx^3\left[\log(u)-\frac{1}{2u}-\psi(u)\right],\tag{2.83}$$

where $\psi(u)$ is the digamma function defined by the logarithmic derivative of the gamma function $\Gamma(u)$, i.e. $\psi(u)=\mathrm{d}\log\Gamma(u)/\mathrm{d}u$. 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.

$$\Delta A(y,t) \equiv A(y,t) - A(0,t) \simeq \frac{t}{2}\int_0^1 \mathrm{d}x \left(\frac{x^2}{y+x^2} - 1\right)$$
$$= -\frac{1}{2} t\, y \int_0^1 \mathrm{d}x \frac{1}{y+x^2} = -\frac{t}{2}\sqrt{y}\tan^{-1}\frac{1}{\sqrt{y}} \tag{2.84}$$

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

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

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

$$A(0,t) = \frac{1}{3}t^{4/3}\int_0^{1/t} \mathrm{d}v\, v^{1/3}\left[\ln v - \frac{1}{2v} - \psi(v)\right] \simeq \frac{1}{3}C_{4/3}t^{4/3},$$
$$C_\alpha \equiv \int_0^\infty \mathrm{d}v\, v^{\alpha-1}\left[\log v - \frac{1}{2v} - \psi(v)\right], \quad C_{4/3} = 1.00608\cdots \tag{2.86}$$

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).

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

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

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

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 signifancant 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.

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

The frequency integral of (2.74) is then written by

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

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

$$\langle S_{\mathrm{loc}}^2\rangle_{\mathrm{Z}}(y) = \langle S^2\rangle_{\mathrm{Z}}(0) - \frac{9T_0}{T_A}cy + \cdots . \tag{2.91}$$

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.

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

$$-[\chi(\boldsymbol{q})\Gamma(q,\omega)]\frac{2\omega\Gamma(q,\omega)}{[\omega^2+\Gamma^2(q,\omega)]^2}\frac{\partial\Gamma(q,\omega)}{\partial y}\Bigg\} \qquad (2.92)$$

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 $[\chi(\boldsymbol{q})\Gamma(q,\omega)]$ 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.

$$\begin{aligned}\frac{\partial}{\partial y}\langle S^2\rangle_{\mathrm{Z}}(y) &\simeq -\frac{3}{N_0^2}\sum_{\boldsymbol{q}}\chi(\boldsymbol{q})\frac{\partial\Gamma_q}{\partial y}\int_0^\infty\frac{\mathrm{d}\omega}{\pi}\frac{2\omega\Gamma_q^2}{(\omega^2+\Gamma_q^2)^2}\\ &= -\frac{3}{N_0^2}\sum_{\boldsymbol{q}}\frac{1}{\pi}\chi(\boldsymbol{q})\frac{\partial\Gamma_q}{\partial y}\,. \end{aligned}\qquad (2.93)$$

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

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

Its value for the Lorentzian distribution function is given by

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

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 $(k_{\mathrm{B}}T/W)^2$ 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 $S(\boldsymbol{q}, \omega)$, the observed intensity I therefore amounts to the following integral.

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

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

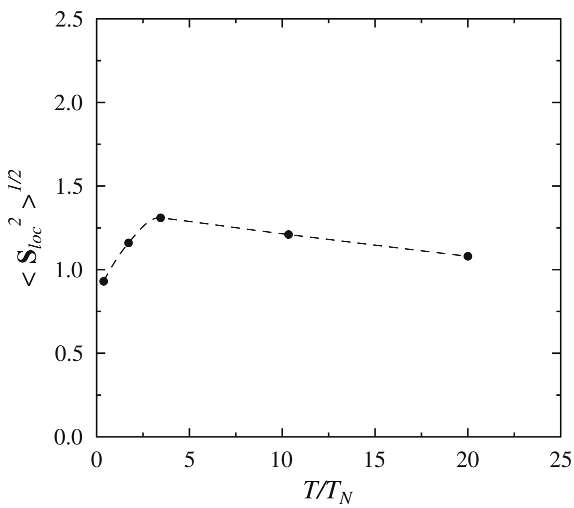

Fig. 2.1 Temperature dependence of the total spin amplitude observed by Ziebeck et al. [8]

dynamical magnetic susceptibility by

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

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, $\bar{S}_{\mathrm{T}}(\boldsymbol{q},\omega)$ and $\bar{S}_{\mathrm{Z}}(\boldsymbol{q},\omega)$, respectively, i.e., by

$$\begin{aligned} &\bar{S}(\boldsymbol{q},\omega) = \bar{S}_{\mathrm{T}}(\boldsymbol{q},\omega) + \bar{S}_{\mathrm{Z}}(\boldsymbol{q},\omega), \\ &\bar{S}_{\mathrm{T}}(\boldsymbol{q},\omega) \equiv n(|\omega|)]\mathrm{Im}\chi(\boldsymbol{q},|\omega|), \quad \bar{S}_{\mathrm{Z}}(\boldsymbol{q},\omega) \equiv \theta(\omega)\mathrm{Im}\chi(\boldsymbol{q},\omega) \end{aligned} \tag{2.98}$$

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 $\boldsymbol{q}$.

Let us define integrated thermal and zero-point intensities by

$$I_{\mathrm{T}} = \sum_{\boldsymbol{q}} \int_0^{\omega_c} \mathrm{d}\omega \bar{S}_{\mathrm{T}}(\boldsymbol{q},\omega), \quad I_{\mathrm{Z}} = \sum_{\boldsymbol{q}} \int_0^{\omega_c} \mathrm{d}\omega \bar{S}_{\mathrm{Z}}(\boldsymbol{q},\omega). \tag{2.99}$$

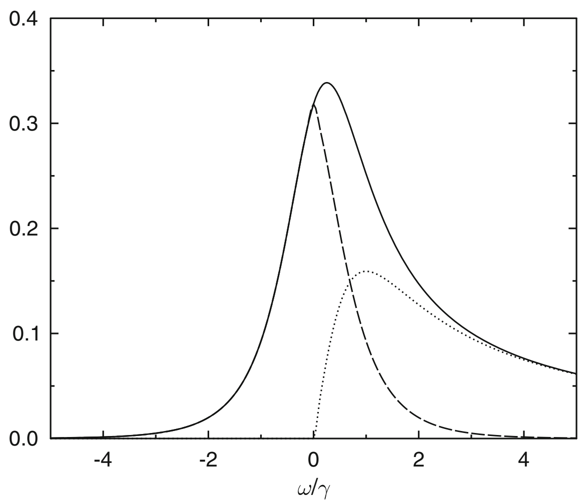

Fig. 2.2 Frequency dependence of the scattering intensity. *Solid*, *dashed*, and *thin dotted curves* represent the total intensity, zero-point and thermal components, respectively. The dampling constant is denoted by γ

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

$$I = 2I_T + I_Z. \tag{2.100}$$

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.

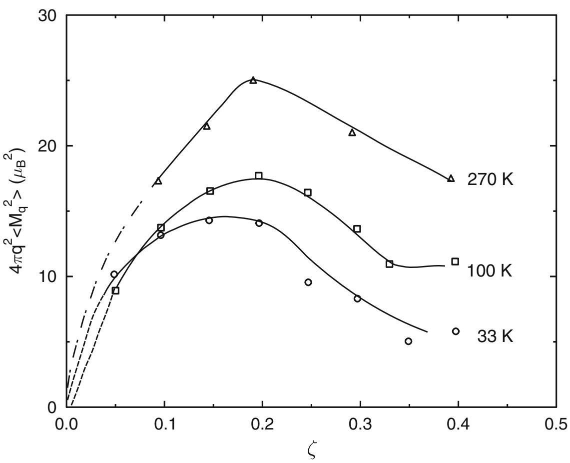

Fig. 2.3 Wave vector dependence of the scattering intensity at several temperatures, $T = 33$ K, 100 K, and 270 K by Ishikawa et al. [9]

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.

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

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, $n(\omega)\mathrm{Im}\chi(q,\omega)$, 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.

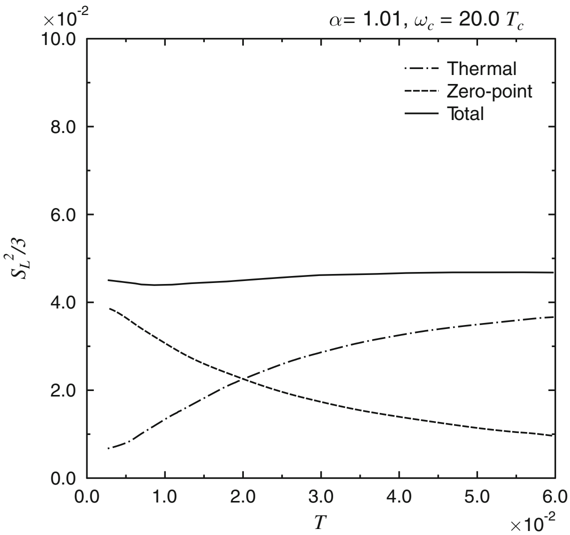

Fig. 2.4 Temperature dependence of the theoretically calculated neutron scattering intensity of MnSi by Takahashi and Moriya [4]. *Solid*, *dashed*, and *dash-dotted curves* correspond to the total, the zero-point, and the thermal amplitudes, respectively

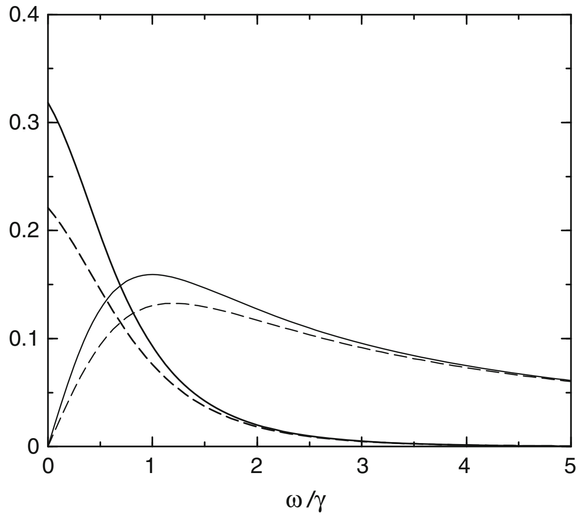

Fig. 2.5 Spectral shape change for thermal and zero-point amplitudes at some fixed wave-vector q caused by the variation of the inverse of the magnetic susceptibility y

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)Mn_2

The presence of the zero-point amplitude is also demonstrated by the neutron scattering experiments on the the Laves phase compound YMn_2. 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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Chapter 3
Effects of Spin Fluctuations on Magnetic Properties

3.1 Basic Idea of the Spin Fluctuation Theory

In our treatment of spin fluctuations of large amplitudes, it is inappropriate to employ a kind of expansion method with respect to their amplitudes. Instead it is better to rely on some general ideas justified independent of the magnitude of amplitudes. We propose the following ideas [1, 2] as the basis of our following discussions.

1. Total spin amplitude conservation (TAC)
   The total spin amplitude on each magnetic site of the crystal is conserved independent of temperature. It is also unaffected by external magnetic field.
2. Global consistency in the effect of magnetic field (GC)
   We mean by this that the magnetic isotherm, i.e., the functional relation between the external magnetic field H and the induced magnetization M, is globally consistent with the first condition.

Specifically, the condition of TAC is explicitly written in the form

$$\left\langle S_{\rm loc}^2 \right\rangle_{\rm tot} = \left\langle \delta S_{\rm loc}^2 \right\rangle_{\rm T} (y, y_z, T) + \left\langle \delta S_{\rm loc}^2 \right\rangle_{\rm Z} (y, y_z) + \sigma^2. \tag{3.1}$$

The left-hand side represents a conserved constant amplitude. The first two terms on the right-hand side are the thermal and the zero-point amplitudes, respectively, while the last term is a mean static local magnetic moment squared. In the presence of external magnetic field, both these amplitudes are determined by the reduced inverse of magnetic susceptibilities $y(\sigma, t)$ and $y_z(\sigma, t)$ as functions of variables σ and t. These functions are related to each other by

$$y_z(\sigma, t) = y(\sigma, t) + \sigma \frac{\partial y(\sigma, t)}{\partial \sigma}. \tag{3.2}$$

Y. Takahashi, *Spin Fluctuation Theory of Itinerant Electron Magnetism*, Springer Tracts in Modern Physics 253, DOI: 10.1007/978-3-642-36666-6_3,

Our GC requirement imposes on $y(\sigma, t)$ the condition that its σ dependence has always to be determined to satisfy (3.1). It means that (3.1) is regarded as *an ordinary differential equation* for $y(\sigma, t)$.

With use of the thermal amplitude $A(y, t)$ in (2.83), the total amplitude conservation is written as follows:

$$\begin{aligned} &2A(y,t) + A(y_z,t) - c(2y + y_z) + \frac{T_A}{3T_0}\sigma^2 = \frac{T_A}{3T_0}\Delta\left\langle S_{\text{loc}}^2\right\rangle_{\text{tot}} \\ &\Delta\left\langle S_{\text{loc}}^2\right\rangle_{\text{tot}} \equiv \left\langle S_{\text{loc}}^2\right\rangle_{\text{tot}} - \left\langle S_{\text{loc}}^2\right\rangle_{Z}(0,0) \end{aligned} \tag{3.3}$$

At the critical temperature in the absence of the external magnetic field, $y = y_z = \sigma = 0$ is satisfied for ferromagnets. The right-hand side of (3.3) is then given by $3A(0, t_c)$ on the left hand side. For paramagnets in the ground state ($T = 0$) with no thermal amplitude it is given by $-3cy_0$ because both y and y_z have the same finite value y_0. To summarize, the right-hand side of (3.3) is given by

$$\frac{T_A}{3T_0}\Delta\left\langle S_{\text{loc}}^2\right\rangle_{\text{tot}} = \begin{cases} 3A(0,t_c), & \text{for ferromagnets} \\ -3cy_0, & \text{for paramagnets} \end{cases} \tag{3.4}$$

In the following sections we will show that various magnetic properties are derived by solving the differential equation (3.3).

3.2 Magnetic Isotherm in the Ground State

To begin with, for convenience in later sections, we first show how the magnetic isotherm in the ground state is derived as a solution of Eq. (3.3). Since no thermal spin fluctuations are present in this case, it is written for ferromagnets in the form

$$-c\,(2y + y_z) + \frac{T_A}{3T_0}\sigma^2 = 3A(0, t_c), \tag{3.5}$$

and for paramagnets,

$$-c\,(2y + y_z) + \frac{T_A}{3T_0}\sigma^2 = -3cy_0. \tag{3.6}$$

These are regarded as ordinary differential equations for $y(\sigma) \equiv y(\sigma, 0)$ as a function of σ, since we can find the derivative $\mathrm{d}y/\mathrm{d}\sigma$ as a function of y and σ from them. In this section, the variable of the reduced temperature t is neglected. We can find their solutions as shown below.

3.2.1 Magnetic Isotherm for Ferromagnets

Since y and y_z are defined as dimensionless functions of H/M and $\partial H/\partial M$, they are both regarded as even functions of σ. If we notice that only zeroth and the first order terms of σ^2 are present in (3.5), it is reasonable to assume the following trial function with two unknown parameters, σ_0 and y_1.

$$y(\sigma) = y_1(\sigma^2 - \sigma_0^2). \tag{3.7}$$

The parameter σ_0 has a meaning of the spontaneous magnetic moment, for $y(\sigma) \propto H/M$ vanishes at $\sigma = \sigma_0$ for $H = 0$. It follows from (3.2) that the σ dependence of $y_z(\sigma)$ is also given by

$$y_z(\sigma) = y(\sigma) + \sigma \frac{\partial y(\sigma)}{\partial \sigma} = y(\sigma) + 2y_1\sigma^2. \tag{3.8}$$

Substitution of the above (3.7) and (3.8) for (3.5) then gives

$$\left(\frac{T_A}{3T_0} - 5cy_1\right)\sigma^2 + 3[cy_1\sigma_0^2 - A(0, t_c)] = 0. \tag{3.9}$$

The parameters y_1 and σ_0^2 are determined by the condition where the above (3.9) is identically satisfied.

$$y_1 = \frac{T_A}{15cT_0}, \tag{3.10}$$

$$\sigma_0^2 = \frac{1}{cy_1}A(0, t_c) = \frac{15T_0}{T_A}A(0, t_c) \simeq \frac{5T_0}{T_A}C_{4/3}\left(\frac{T_c}{T_0}\right)^{4/3}. \tag{3.11}$$

If we notice the expression of $A(y, t)$ in (2.54), the above second line is also represented in terms of the thermal spin fluctuation amplitude in the form

$$\left\langle S_{\text{loc}}^2 \right\rangle_T (0, 0, t_c) = \frac{9T_0}{T_A}A(0, t_c) = \frac{3}{5}\sigma_0^2 \tag{3.12}$$

The result for the parameter y_1 in (3.10) is attributed to the effect of zero-point fluctuations. It is therefore characteristic to the present treatment. As for (3.12), between the thermal amplitude at T_c and the spontaneous magnetic moment squared σ_0^2, the same relation was already derived by the SCR theory [3].

The magnetic isotherm, i.e., the functional relation between the external magnetic field h and the induced moment σ, is given by

$$h = 2T_A\sigma y = F_1\sigma(\sigma^2 - \sigma_0^2), \quad F_1 = \frac{2T_A^2}{15cT_0}. \tag{3.13}$$

In terms of original variables M and H in (2.71), it is also written as follows:

$$(g\mu_B)H = F_1\left[\frac{M^2}{(N_0 g\mu_B)^2} - \frac{M_0^2}{(N_0 g\mu_B)^2}\right]\frac{M}{N_0 g\mu_B}. \tag{3.14}$$

The M dependence of the magnetic free energy $F_m(M, T)$ is also obtained by integrating the thermodynamic relation, $H = \partial F_m(M, T)/\partial M$, with respect to M.

$$\begin{aligned} F_m(M) &= F_m(0) + \frac{1}{2}a(0)M^2 + \frac{1}{4}b(0)M^4 \\ &= F_m(0) + \frac{1}{2(g\mu_B)^2\chi}M^2 + \frac{F_1}{4(g\mu_B)^4 N_0^3}M^4, \end{aligned} \tag{3.15}$$

It is particularly interesting that even in the ground state, the magnetic isotherm is determined, being influenced by the effect of spin fluctuations. Its typical example is the fourth expansion coefficient F_1, given in terms of spectral parameters T_0 and T_A, of the free energy in (3.10). Since no thermal fluctuations are present in this case, the increasing magnetization is compensated by the suppression of the zero-point spin fluctuation amplitudes. In contrast, in the SW and SCR theories, it is determined by the density of state ρ at the Fermi energy ε_F, and its derivatives ρ', ρ'', and so on.

Experimental estimate of spectral parameters According to our present spin fluctuation theory, spectral parameters T_0 and T_A are involved in various magnetic properties derived theoretically. As examples, we show below (3.11) and (3.13) again in the form

$$p_s^2 = \frac{20T_0}{T_A}C_{4/3}\left(\frac{T_c}{T_0}\right)^{4/3}, \quad C_{4/3} = 1.006089\ldots, \tag{3.16}$$

$$F_1 = \frac{2T_A^2}{15cT_0}, \tag{3.17}$$

where $p_s = 2\sigma_0$ is the spontaneous magnetic moment in units of μ_B per magnetic ions. If these values are already known by some means, we can check the validity of these properties quantitatively. Both of them are estimated by inelastic neutron scattering measurements. Experimentally estimated values for MnSi by Ishikawa et al. [4] and Ni_3Al by Bernhoeft et al. [6] are shown in Table 3.1. We can therefore check the validity of (3.16) for these compounds. By putting the observed $T_c = 30$ K and those of T_0 and T_A into (3.16), we get the value $p_s = 0.38$, in good agreement with $p_s = 0.40$ from the magnetic measurements by Bloch et al. [5]. As with the case of MnSi, observed spectral parameters for Ni_3Al in Table 3.1 by Bernhoeft et al. [6] and the observed $T_c = 41$ K gives $p_s = 0.078$, being also in agreement with $p_s = 0.075$ by de Boer et al. [7]. Although there are not much examples thus far, these results seem to support the validity of (3.16).

Table 3.1 Comparison of the fourth expansion coefficient F_1 in (3.13) with experiments

Compounds	T_0(K)	T_A(K)	$4T_A^2/15T_0$(K)	F_1(K)	References
MnSi	231	2.08×10^3	5.0×10^3	9.7×10^3	Ishikawa et al. [4]
	171	2.11×10^3	6.94×10^3		Yasuoka et al.[a] [8]
Ni_3Al	3590	3.09×10^4	0.71×10^5		Bernhoeft et al. [6]
$Ni_{74.7}Al_{25.3}$	2860	4.05×10^4	1.53×10^5	1.0×10^5	Umemura et al.[a] [9]
Sc_3In	565	2.00×10^5	0.66×10^5	2.0×10^5	Hioki and Masuda[a] [10]
$ZrZn_2$	321	8.83×10^3	1.05×10^4	1.3×10^4	Kontani et al.[a] [11]
$Y(Co_{1-x}Al_x)_2$					Yoshimura et al.[a] [12]
$x = 0.13$	2290	1.16×10^4	1.57×10^4	2.1×10^4	
$x = 0.15$	2119	6.34×10^3	0.51×10^4	1.0×10^4	
$x = 0.17$	2093	7.03×10^3	0.63×10^4	1.6×10^4	

[a] NMR measurements

In the fourth column of Table 3.1, we show values of F_1 for those compounds estimated by using (3.17) with these parameters. In its fifth column, experimentally estimated F_1 from the slope of the Arrott plot are also shown for comparison. Fairy good agreement with these two values is evident. We can also estimate T_0 from the analysis of the temperature dependence of the NMR relaxation time measurements [3]. The parameter T_A is then estimated by (3.16) from the value of T_0, as well as the observed T_c and σ_0. In this case, only the validity of (3.17) is verified experimentally. In the fourth column, we show values of F_1 estimated by (3.17) by using the values of T_0 and T_A in the second and third columns. These results compare well with the values in the fifth column estimated from the slope of the Arrott plot.

Once the validity of (3.11) and (3.13) is recognized, we can estimate the spectral parameters T_0 and T_A of spin fluctuations only by using results of magnetization measurements. These equations provide two independent relations among five parameters, p_s, T_c, F_1, T_0, and T_A. It implies that any two parameters can be expressed in terms of the rest of the three parameters. Particularly for T_0/T_c and T_A/T_c, the following relations are satisfied:

$$\begin{aligned} \left(\frac{T_c}{T_0}\right)^{5/6} &= \frac{\sigma_0^2}{5C_{4/3}}\left(\frac{15cF_1}{2T_c}\right)^{1/2} \\ \left(\frac{T_c}{T_A}\right)^{5/3} &= \frac{\sigma_0^2}{5C_{4/3}}\left(\frac{2T_c}{15cF_1}\right)^{1/3} \end{aligned} \tag{3.18}$$

From (3.17), T_A is represented in terms of T_0 and F_1. The first relation is then obtained by putting it into (3.16) and eliminating T_A. The second relation is derived in the same way.

3.2.2 Magnetic Isotherm for Exchange Enhanced Paramagnets

For paramagnets near the ferromagnetic instability point, (3.6) is satisfied in the low temperature limit, in place of (3.5) for ferromagnets. Since the spontaneous magnetic moment is absent in this case, the induced magnetization M by an externally applied magnetic field H is proportional to H. Reciprocal magnetic susceptibilities, $y(t)$ and $y_z(t)$, thus remain finite, i.e., $y(0) = y_z(0) = y_0 > 0$, in the weak field limit. It is therefore possible to assume the following trial solution for $y(\sigma) \equiv y(\sigma, 0)$ in powers of σ^2 up to the linear term.

$$y(\sigma) = y_0 + y_1\sigma^2 = y_1[\sigma^2 + y_0/y_1] = y_1(\sigma^2 + \sigma_p^2) \tag{3.19}$$

The σ dependence of $y_z(\sigma)$ is given by (3.2). By putting these expressions into (3.2), the same (3.10) for y_1 is derived. To emphasize the similarity with ferromagnets, let us introduce a new parameter, $\sigma_{\rm p}^2$ defined by $\sigma_0^2 \equiv y_0/y_1$, that corresponds to the spontaneous moment squared, σ_0^2, for ferromagnets. The magnetic isotherm is then given by

$$h = 2T_A\sigma y = \frac{2T_A^2}{15cT_0}\sigma(\sigma^2 + \sigma_p^2), \tag{3.20}$$

in agreement with (3.13) except for the sign of $\sigma_{\rm p}^2$ on the right-hand side.

In (3.19), two unknown coefficients, y_0 and y_1 can be determined from the condition that (3.19) has to satisfy Eq. (3.6). It follows that the first σ^2-linear coefficient y_1 is given by the same (3.10) for ferromagnets, while the zeroth coefficient y_0 is given by

$$\sigma_p^2 = \frac{y_0}{y_1} = \frac{15cT_0}{T_A}y_0. \tag{3.21}$$

In terms of magnetic susceptibility $\chi(0)$ in the ground state, it is also written in the form

$$\sigma_p^2 = \frac{15cT_0}{T_A}\frac{N_0}{2T_A\chi(0)} = \frac{N_0}{\chi(0)F_1}. \tag{3.22}$$

Introduction of the following two new parameters t_p and T_p is useful for promoting the similarity further.

$$A(0, t_p) = cy_1\sigma_p^2 = cy_0, \quad T_p \equiv t_pT_0. \tag{3.23}$$

These correspond to $t_c = T_c/T_0$ and the critical temperature T_c for ferromagnets. With these parameters, (3.6) is written in the form

$$-c(2y + y_z) + \frac{T_A}{3T_0}\sigma^2 = -3A(0, t_p), \tag{3.24}$$

being similar to (3.5) for ferromagnets. Only the sign on the right-hand side is different.

3.3 Magnetic Properties in the Paramagnetic Phase

We show next in this section, how to solve the differential equation (3.3) in the paramagnetic phase. It is written as follows:

$$2A(y,t) + A(y_z,t) - c(2y+y_z) + \frac{T_A}{3T_0}\sigma^2 = 3A(0,t_c), \tag{3.25}$$

$$2A(y,t) + A(y_z,t) - c(2y+y_z) + \frac{T_A}{3T_0}\sigma^2 = -3A(0,t_p). \tag{3.26}$$

The upper Eq. (3.25) corresponds to (3.5) for ferromagnets, while the lower one (3.26) to (3.6) for paramagnets in the ground state. They are different from those in the ground state in the presence of the thermal spin fluctuation amplitudes on their left-hand sides.

3.3.1 Temperature Dependence of Magnetic Susceptibility

To begin with, let us discuss the temperature dependence of magnetic susceptibility. It is related to the initial condition of the differential equation (3.3). In the paramagnetic phase, the following magnetic isotherm is satisfied between the external magnetic field H and the induced magnetic moment M.

$$H = a(T)M + b(T)M^3 + c(T)M^5 + \cdots. \tag{3.27}$$

The first coefficient $a(T)$ is positive and finite in the paramagnetic phase. The above isotherm is also shown as the M dependence of inverse magnetic susceptibility H/M.

$$\frac{H}{M} = a(T) + b(T)M^2 + c(T)M^4 + \cdots. \tag{3.28}$$

The initial condition of $y(\sigma,t) \propto H/M$ at $\sigma = 0$, given by the first term $a(T)$, is always positive in this case. The first derivative with respect to M also vanishes as shown below.

$$\frac{\partial y}{\partial \sigma} \propto \frac{\partial(H/M)}{\partial M} = 2b(T)M \to 0, \quad (M \to 0) \tag{3.29}$$

These imply that to find the temperature dependence of magnetic susceptibility is equivalent to the initial condition of the magnetic isotherm. It is given by solving the following equation for $y(t) \equiv y(0,t)$.

$$y(t) = \begin{cases} \dfrac{1}{c}[A(y,t) - A(0,t_c)], & \text{for ferromagnets,} \\ \dfrac{1}{c}[A(y,t) + A(0,t_p)], & \text{for paramagnets.} \end{cases} \tag{3.30}$$

For purpose of comparison, we show below the similar equation in (2.57) again, derived by the SCR theory.

$$\frac{1}{\chi(T)} = \frac{5}{3}b(T)\left[\sum_{\mathbf{p}} \langle M_{\mathbf{p}} \cdot M_{-\mathbf{p}} \rangle (T) - \sum_{\mathbf{p}} \langle M_{\mathbf{p}} \cdot M_{-\mathbf{p}} \rangle (T_c)\right]. \tag{3.31}$$

If we notice the definitions of the reduced variable y and the thermal amplitude $A(y,t)$, i.e., $y = N_0/2T_A\chi(T)$ and $\langle S_{\text{loc}}^2 \rangle_{\text{T}} (y, y_z, t) = 9T_0 A(y,t)/T_A$, both (3.30) and (3.31) show close resemblance. As a subtle difference, the coefficient $b(t)$ in (3.31) will be possibly temperature dependent. Their origins are, however, quite different.

3.3.2 Magnetic Susceptibility in the Low Temperature Limit

At finite temperatures, the temperature dependence of the inverse of magnetic susceptibility y for paramagnets is determined by solving (3.30). From the temperature dependence of the thermal amplitude $A(y,t)$ in (2.88), it is approximated, at low temperatures, by

$$y(t) = y_0 + \frac{1}{c}A(y,t) = y_0 + \frac{t^2}{24cy(t)} + \cdots \simeq y_0 + \frac{t^2}{24cy_0}. \tag{3.32}$$

It is also shown as the relative change of $y(t)$ to its ground state value y_0.

$$\frac{\chi(0)}{\chi(T)} = \frac{y(t)}{y_0} \simeq 1 + \frac{1}{24cy_0^2}\left(\frac{T}{T_0}\right)^2 = 1 + \frac{75c}{8\sigma_p^4}\left(\frac{T}{T_A}\right)^2 = 1 + \alpha_2 T^2 \tag{3.33}$$

It follows from the above result that we can estimate the parameter T_A from the observed T^2-linear coefficient of $\chi^{-1}(T)$ in the low-T limit. It is estimated from the observed value of α_2 by using

$$T_A = \frac{1}{\sigma_p^2}\sqrt{\frac{75c}{8\alpha_2}}. \tag{3.34}$$

For paramagnets, there exist two independent relations among five parameters, σ_p, $T_{\rm p}$, F_1, T_0, and T_A. From the slope of the Arrott plot of magnetization curves at low temperatures, we can determine F_1 and σ_p. If T_p is also known by some means, we can determine T_0 and T_A in the same way as ferromagnets. Even if T_p is unknown, they will be determined by the following procedure:

1. Determine values of $\chi(0)$ and F_1 from the Arrott plot of the magnetization curve in the limit of low temperature. By putting these values into (3.22), σ_p^2 is evaluated.
2. The parameter T_A is then estimated from the temperature dependence of $\chi(T)$ at low temperatures by using (3.34) with the above σ_p^2 and the coefficient α_2.
3. Finally, T_0 is evaluated from values of F_1 and T_A by

$$T_0 = \frac{2T_A^2}{15cF_1}. \tag{3.35}$$

3.3.3 Magnetic Susceptibility Around the Critical Point

Reflecting the anomalous $\sqrt{y(t)}$ dependence of the thermal amplitude around the critical point, the spin amplitude conservation is written in the form

$$A(y,t) - cy(t) \simeq A(0,t) - \frac{\pi}{4}t\sqrt{y(t)} - cy(t) = A(0,t_c). \tag{3.36}$$

Since $y(t) \ll 1$ is satisfied very close to the critical point, $y(t)$ linear term originating from the zero-point amplitude can be neglected. The solution of (3.36) is thus determined in the form

$$\begin{aligned}&\frac{\pi}{4}t_c\sqrt{y(t)} = [A(0,t) - A(0,t_c)],\\ &\qquad y(t) = \left(\frac{4}{\pi t_c}\right)^2 [A(0,t) - A(0,t_c)]^2 = \left[\frac{4}{\pi}\frac{\partial A(0,t_c)}{\partial t_c}\right]^2 \left(\frac{t}{t_c} - 1\right)^2,\end{aligned} \tag{3.37}$$

where we have assumed $t \simeq t_c$. The $(T - T_c)^2$-linear dependence of y is therefore derived in this region. In cases where the condition $t_c \ll 1$ is further satisfied, $\partial A(0,t)/\partial t \simeq 4A(0,t)/3t$ is derived from (2.86). The above (3.37) is then rewritten as

$$y(t) \simeq \left[\frac{16A(0,t_c)}{3\pi t_c}\right]^2 \left(\frac{t}{t_c} - 1\right)^2 = \left(\frac{16}{45\pi}\right)^2 \left(\frac{T_A}{T_c}\right)^2 \sigma_0^4 \left(\frac{T}{T_c} - 1\right)^2, \tag{3.38}$$

with use of the relation $A(0,t_c) = cy_1\sigma_0^2$ in (3.11). The temperature dependence of the inverse of magnetic susceptibility per atom is finally represented in the form

$$\frac{N_0}{\chi(T)} = 2\left(\frac{16}{45\pi}\right)^2 \frac{T_A^3\sigma_0^4}{T_c^2}\left(\frac{T}{T_c}-1\right)^2. \tag{3.39}$$

3.3.4 Curie–Weiss Law of Magnetic Susceptibility

Even in itinerant electron ferromagnets, their observed magnetic susceptibilities usually obey the Curie-Weiss law temperature dependence. Theoretically, the temperature dependence of its inverse is evaluated by solving (3.30) for the variable $y(t)$. Numerically estimated $y(t)$ shows a good linearity in a wide range of reduced temperature t. However, it does not imply that the linearity is not strictly satisfied above T_c.

In relation to the Curie-Weiss law dependence of the magnetic susceptibility, We show in this section that (3.30) will lead to an interesting property. To check this property will then amount to confirming the validity of (3.30) experimentally.

Rhodes-Wohlfarth Plot and its Revision For insulator magnets, the effective magnetic moment $p_{\rm eff}$ is defined from the Curie constant by $C = N(\mu_{\rm B} p_{\rm eff})^2/3k_{\rm B}$. From the spontaneous magnetization M_0 in the ground state, the spontaneous magnetic moment p_s per magnetic is also defined by $p_s = M_0/N$. It is well known that they are given by

$$p_{\rm eff}^2 = g^2 S(S+1), \quad p_s = gS, \tag{3.40}$$

where S and g denote the magnitude of the spin and the gyro-magnetic ratio. They are thus related with each other by

$$p_{\rm eff}^2 = p_s(p_s+2), \tag{3.41}$$

for $g = 2$. If we define another magnetic moment $p_{\rm C}$ from $p_{\rm eff}$ by $p_{\rm eff}^2 = p_{\rm C}(p_{\rm C}+2)$, the relation $p_{\rm C}/p_{\rm s} = 1$ is always satisfied.

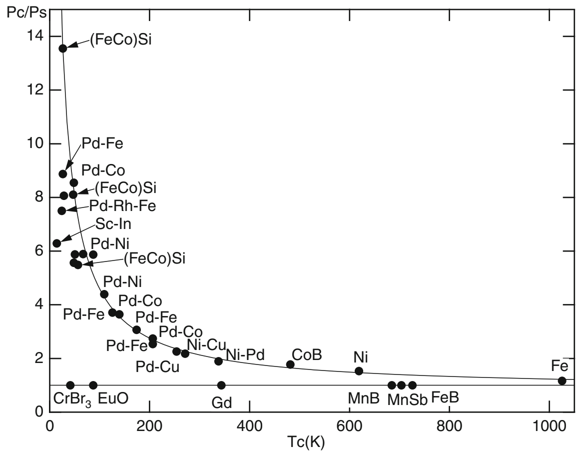
For most itinerant electron ferromagnets, the lower the critical temperature T_c is, the smaller spontaneous magnetic moment is usually observed. No such tendency is observed for $p_{\rm eff}$. To demonstrate these behaviors clearly, Rhodes and Wohlfarth proposed to plot the ratio $p_{\rm eff}/p_{\rm s}$ against T_c [18], called Rhodes-Wohlfarth plot. In Fig. 3.1, we show an example of the Rhodes-Wohlfarth plot for some itinerant electron ferromagnets as well as localized moment magnets. Later in 1986, another interesting plot was proposed by Takahashi [1], i.e., $p_{\rm eff}/p_{\rm s}$ versus T_c/T_0 plot, based on his spin fluctuation theory. We will show below the outline of his reasoning.

The Curie-Weiss law temperature dependence of the magnetic susceptibility in our units is written as

$$\frac{\chi(T)}{N_0} = \frac{p_{\rm eff}^2}{12(T-T_c)}. \tag{3.42}$$

The almost T-linear dependence of the inverse of the magnetic susceptibility $\chi^{-1}(T)$ is also equivalent with the relation,

Fig. 3.1 Rhodes-Wohlfarth plot. The ratio of magnetic moments $p_{\mathrm{eff}}/p_{\mathrm{e}}$ is plotted against the Curie temperature T_c of ferromagnets



$$\frac{N_0}{\chi(T)} = 2T_A y(t) \sim 2T_A \frac{\mathrm{d}y(t)}{\mathrm{d}t}(t - t_c) = 2\frac{T_A}{T_0}\frac{\mathrm{d}y(t)}{\mathrm{d}t}(T - T_c), \tag{3.43}$$

which is justified as far as the temperature dependence of the derivative $\mathrm{d}y/\mathrm{d}t$ is very weak. From the comparison of (3.42) and (3.43), the following relation is satisfied:

$$\frac{12}{p_{\mathrm{eff}}^2} = 2\frac{T_A}{T_0}\frac{\mathrm{d}y(t)}{\mathrm{d}t} \tag{3.44}$$

Notice that we have already derived in Sect. 3.2.1, the relation (3.14) between σ_0 and T_c/T_0, given by

$$\sigma_0^2 = \frac{p_s^2}{g^2} = \frac{15T_0}{T_A}A(0, t_c) \simeq \frac{5T_0}{T_A}C_{4/3}\left(\frac{T_c}{T_0}\right)^{4/3}. \tag{3.45}$$

By eliminating the ratio of T_A/T_0 in (3.44) with the use of (3.45), the following result is finally derived.

$$\left(\frac{p_{\mathrm{eff}}}{p_s}\right)^2 = \frac{1}{10\mathrm{d}y/\mathrm{d}t}\frac{1}{A(0, t_c)} \simeq \frac{3}{10C_{4/3}\mathrm{d}y/\mathrm{d}t}\left(\frac{T_0}{T_c}\right)^{4/3}. \tag{3.46}$$

The last expression on the right-hand side is justified for $t_c \ll 1$. Equation (3.46) implies that the observed ratio of p_{eff}/p_s is determined by the single parameter T_c/T_0. This is the reason for new plot, p_{eff}/p_s versus T_c/T_0, proposed by Takahashi [1]. The values of T_0 required for the plot have already been estimated from (3.18) for many itinerant electron ferromagnets by using observed σ_0, T_c, and F_1.

In order to demonstrate the validity of Takahashi's plot, two ways of plots were compared by Nakabayashi et al. for Y–Ni compounds [19]. Their result is shown

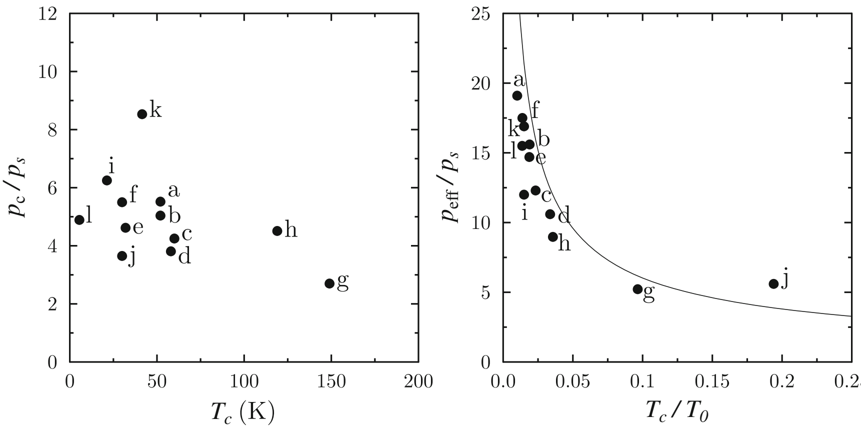


Fig. 3.2 Rhodes-Wohlfarth plot for Y_xNi_y (*left*), and Takahashi's revised plot (*right*): *a* Y_2Ni_7, *b* $Y_2Ni_{6.9}$, *c* $Y_2Ni_{6.8}$, *d* $Y_2Ni_{6.7}$, *e* $YNi_{2.9}$, *f* YNi_3, *g* Y_2Ni_{17}, *h* Y_2Ni_{15}, *i* $ZrZn_2$, *j* MnSi, *k* Ni_3Al, *l* Sc_3In

in Fig. 3.2. Experimentally estimated parameters for compounds in these plots are summarized in Table 3.3. Solid circles corresponding to compounds are found to fall on a narrow region around the theoretical curve given by

$$\frac{p_{\rm eff}}{p_{\rm s}} \simeq 1.4 \left(\frac{T_0}{T_c}\right)^{2/3}. \tag{3.47}$$

Since then, parameters T_0 and T_A have been estimated for a number of other itinerant ferromagnets such as La(Ni,Al)$_{13}$ by Fujita et al. [20] in 1995, for instance. These are also shown in Table 3.3. Having the theoretical result (3.47) in mind, $\log(p_{\rm eff}/p_{\rm s})$ versus $\log(T_c/T_0)$ plot in Fig. 3.3 is recently proposed by Deguchi. All the compounds shown in Tables 3.2 and 3.3 are shown in this figure. The tetragonal compound $LaCo_2P_2$ [21] in Table 3.3 has an easy-plane anisotropy perpendicular to the c-axis. Three uranium compounds by Deguchi (K. Deguchi, Private Commun.) at the bottom of the table have Ising-like anisotropy. Parameters of these compounds are estimated by assuming them as isotropic. It is known that (3.18) will then give slightly modified parameters of T_0 and T_A in the presence of magnetic anisotropy. Since the coefficient of (3.47) is also slightly modified, they will still fall near the same theoretical curve. These Figs. 3.2 and 3.3 clearly show that (3.47) is well supported by magnetization measurements on a number of itinerant electron ferromagnets.

Once we agree with the validity of (3.46), it is possible to estimate the parameter T_0 from these figures. From the ratio of $p_{\rm eff}/p_{\rm s}$ estimated experimentally, we can find the corresponding value of T_c/T_0 from the figures. With this ratio and the observed value of T_c, T_0 is estimated.

Table 3.2 Spectral parameters T_0, T_A estimated from magnetization measurements

Compound	T_c(K)	p_s	$p_{\rm eff}$	F_1(K)	T_0(K)	T_A (K)	T_c/T_0	$p_{\rm eff}/p_s$	References
MnSi	30	0.4	2.25	9.71×10^3	155	2180	0.194	5.6	[5]
Ni_3Al	41.5	0.075	1.3	1.30×10^5	2760	3.67×10^4	0.015	16.9	[7]
$Sc_{0.7575}In_{0.2425}$	5.5	0.045	0.7	2.00×10^5	286	1.46×10^4	0.019	15.6	[13]
$ZrZn_2$	21.3	0.12	1.44	1.05×10^4	1390	7.4×10^3	0.015	12.0	[14]
$Zr_{0.92}Ti_{0.08}Zn_2$	40	0.233	1.33	1.49×10^4	628	5.92×10^3	0.064	5.7	[15]
$Zr_{0.8}Hf_{0.2}Zn_2$	49.4	0.278	1.38	1.68×10^4	536	5.81×10^3	0.092	4.96	[15]
$Zr_{0.9}Hf_{0.1}Zn_2$	10.2	0.078	1.27	1.20×10^4	1110	7.07×10^3	0.0092	16.3	[15]
$Y(Co_{1-x}Al_x)_2$									[12]
$x = 0.13$	7	0.042	2.50	2.10×10^4	1920	1.23×10^4	0.0036	59.5	
0.14	15	0.094	2.24	1.10	1440	0.772	0.010	23.8	
0.15	26	0.138	2.15	1.00	1410	0.726	0.018	15.6	
0.16	22	0.130	2.14	0.95	1280	0.676	0.017	16.5	
0.17	16	0.095	2.13	1.56	1270	0.846	0.013	22.4	
0.18	9	0.063	2.08	2.77	984	1.01	0.0091	33.0	
0.19	7	0.040	2.04	4.11	1280	1.40	0.0055	51.0	
$Fe_xCo_{1-x}Si$									[16]
$x = 0.36$	23	0.11	1.12	5.79×10^4	640	11.79×10^3	0.0359	10.2	
0.48	48	0.19	1.32	3.16	841	9.98	0.0571	6.9	
0.67	55	0.22	1.39	3.82	680	9.87	0.0809	6.3	
0.77	40	0.18	1.13	9.76	399	12.09	0.100	6.3	
0.88	28	0.13	0.94	18.03	340	15.18	0.0824	7.2	
0.91	14	0.07	0.58	57.6	239	22.73	0.0586	8.3	
$Pt_{1-x}Ni_x$									[17]
$x = 0.429$	23	0.051	1.59	5.84×10^4	4370	3.07×10^4	0.0053	31.2	
$x = 0.452$	54.2	0.104	1.59	4.45	3670	2.46	0.0148	15.3	
$x = 0.476$	75	0.143	1.59	3.74	3120	2.08	0.0240	11.1	
$x = 0.502$	100	0.179	1.59	3.90	2870	2.04	0.0348	8.88	

3.3.5 Magnetic Isotherm in the Paramagnetic Phase

In the paramagnetic phase, magnetic isotherm is given as a solution $y(\sigma, t)$ of (3.25). It is generally evaluated by integrating this differential equation starting from the initial value, $y_0(t) = y(0, t)$ at $\sigma = 0$. The value of $y_0(t)$ is determined as a solution of (3.30).

In the region of weak external magnetic field, the analytic expression of the magnetic isotherm is available, for both y and y_z can be expanded in powers of σ^2.

$$\begin{aligned} y(\sigma, t) &= y_0(t) + y_1(t)\sigma^2 + \cdots, \\ y_z(\sigma, t) &= y_0(t) + 3y_1(t)\sigma^2 + \cdots. \end{aligned} \tag{3.48}$$

Putting them into (3.25) then gives

Table 3.3 Experimentally estimated values of p_{eff}/p_s and T_c/T_0 from magnetization measurements

Compound	p_{s}	p_{eff}	T_c (K)	T_0 (K)	T_A (K)	T_c/T_0	p_{eff}/p_s
Y_2Ni_x							
$x = 7.0$	0.033	0.631	52	5172	2.1×10^5	0.0101	19.1
6.9	0.047	0.728	52	3799	1.16×10^5	0.0137	15.5
6.8	0.064	0.786	60	2580	8.39×10^4	0.0233	12.3
6.7	0.078	0.826	58	1723	6.24×10^4	0.0337	10.6
$YNi_{2.9}$	0.047	0.693	32	1706	7.91×10^4	0.0188	14.7
YNi_3	0.04	0.70	30	2178	9.23×10^4	0.0138	17.5
Y_2Ni_{17}	0.27	1.41	149	1544	1.89×10^4	0.0965	5.22
Y_2Ni_{15}	0.15	1.35	119	3329	3.51×10^4	0.0357	8.97
$La(Ni_xAl_{1-x})_{13}$							
$x = 0.90$	0.024	0.55	13	3350	6.00×10^4	0.004	23
$x = 0.925$	0.14	1.36	87	2060	4.59	0.042	9.7
$x = 0.95$	0.25	1.43	169	1540	2.47	0.11	5.7
$x = 0.975$	0.31	1.55	218	1820	2.28	0.12	5.0
$LaCo_2P$	0.391	1.34	103	589	1.91×10^3	0.175	3.4
UCoGe	0.039	1.93	2.4	362	5.92×10^3	0.0065	49.5
URhGe	0.32	1.74	9.6	111	8.56×10^2	0.0865	5.44
UGe_2	1.44	2.58	53.5	61.5	4.93×10^2	0.870	1.79

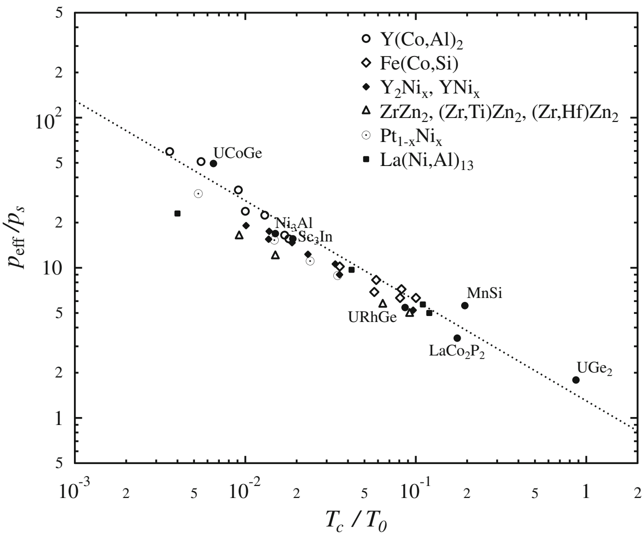


Fig. 3.3 Deguchi-Takahashi plot, i.e. $\log(p_{\mathrm{eff}}/p_s)$ versus $\log(T_c/T_0)$ plot for compounds in Tables 3.2 and 3.3. Theoretical result (3.47) is shown as a *dotted straight line*

$$3A(y_0,t) - 3cy_0(t) + 5[A'(y_0,t) - c]y_1(t)\sigma^2 + \frac{T_A}{3T_0}\sigma^2 + \cdots = 3A(0,t_c), \quad (3.49)$$

where the $A'(y,t)$ is the partial derivative $\partial A(y,t)/\partial y$. From the condition that (3.49) is satisfied identically, the temperature dependence of $y_1(t)$ is obtained.

$$y_1(t) = \frac{T_A}{15T_0}\frac{1}{c - A'(y_0,t)} = \frac{y_1(0)}{1 - A'(y_0,t)/c}. \quad (3.50)$$

The coefficient $y_1(t)$ has the meaning of the reduced fourth expansion coefficient of the free energy with respect to the magnetization M. The above result shows that this coefficient depends on temperature. Especially around the critical point, $y_0(t)$ is given by

$$\frac{y_1(t)}{y_1(0)} \simeq \frac{8c}{\pi t_c}\sqrt{y_0(t)} \to 0, \quad \text{for} \quad t \to t_c, \quad (3.51)$$

because of the critical dependence of $A'(y_0,t) \simeq -\pi t/(8\sqrt{y_0})$ around $y_0 = 0$. It follows that both $y_0(t)$ and $y_1(t)$ in (3.48) approach zero toward the critical temperature T_c.

In the same way as (3.49), (3.30) is expanded in powers of σ^2 for paramagnets.

$$3A(y_0,t) - 3cy_0(0) + 5[A'(y_0,t) - c]y_1(t)\sigma^2 + \frac{T_A}{3T_0}\sigma^2 + \cdots = -3A(0,t_p) \quad (3.52)$$

The same (3.50) is also derived for $y_1(t)$ in this case from the comparison of σ^2-linear coefficients. The derivative of the thermal amplitude $A'(y_0,t)$ at low temperatures is proportional to t^2 as given by

$$A'(y_0,t) \sim -\frac{t^2}{24y_0^2(0)}. \quad (3.53)$$

By putting it into (3.50), the following temperature dependence of $y_1(t)$ is derived:

$$y_1(t) \simeq \frac{T_A}{15cT_0}\left(1 - \frac{t^2}{24cy_0^2(0)} + \cdots\right) \quad (3.54)$$

It is also written in the following form with the use of (3.21),

$$\frac{y_1(t)}{y_1(0)} \simeq 1 - \frac{1}{24cy_0^2(0)}t^2 = 1 - \frac{c}{24\sigma_p^4}\frac{T^2}{T_A^2}, \quad (3.55)$$

where σ_p^2 is used in place of $y_0(0)$. The above T^2-linear coefficient shows tendency to diverge as systems approach the magnetic instability point, $y_0(t) \to 0$ (i.e. $\sigma_p^2 \to 0$).

As described in the above, the temperature dependence of the fourth expansion coefficient $y_1(t)$ of the free energy shows non-negligible temperature dependence in general. It cannot be neglected even in the paramagnetic phase. It decreases to zero for ferromagnets as temperature approaches the critical point, while for paramagnets the coefficient of its T^2-linear dependence shows divergence toward the ferromagnetic instability point. They are regarded as precursor phenomena of the magnetic isotherm as will be shown the next section. The temperature dependence of this coefficient is, however, neglected in the SCR spin fluctuation theory.

3.4 Critical Magnetic Behaviors

In the SW theory, the magnetic free energy is assumed to be expanded in powers of the magnetization M. Spontaneous magnetic moment appears below the Curie temperature T_c where the second expansion coefficient becomes negative. Since the fourth expansion coefficient $b(T)$ in (1.53) remains finite at $T = T_c$, the following relation between H and M is satisfied at the critical point:

$$H = b(T_c)M^3. \tag{3.56}$$

The same is applied for the SCR spin fluctuation theory. In our treatment, however, we do not need to make such an assumption on $b(T)$ and have shown in (3.51) that $b(T)$, i.e., $y_1(t)$, vanishes at the critical point. Instead of making such an assumption, we show below in this section how the magnetic isotherm at the critical temperature is determined as a solution of our differential equation (3.25).

3.4.1 Critical Magnetic Isotherm

Because of the divergence of the magnetic susceptibility, i.e. $y = y_z = 0$ satisfied at the critical point, the thermal amplitude at $t = t_c$ can be written as

$$A(y, t_c) = A(0, t_c) - \frac{\pi t_c}{2}\sqrt{y(\sigma)} + \cdots, \tag{3.57}$$

where $y(\sigma, t_c)$ is denoted by $y(\sigma)$. By putting the above dependence, (3.25) is written as follows:

$$\frac{1}{4}\pi t_c[2\sqrt{y(\sigma)} + \sqrt{y_z(\sigma)}] + c[2y(\sigma) + y_z(\sigma)] = \frac{T_A}{3T_0}\sigma^2. \tag{3.58}$$

In the region of weak magnetic field, both $y(\sigma)$-linear and $y_z(\sigma)$-linear terms, originating from zero-point fluctuations, are neglected in the above, because $y(\sigma, t) \ll 1$

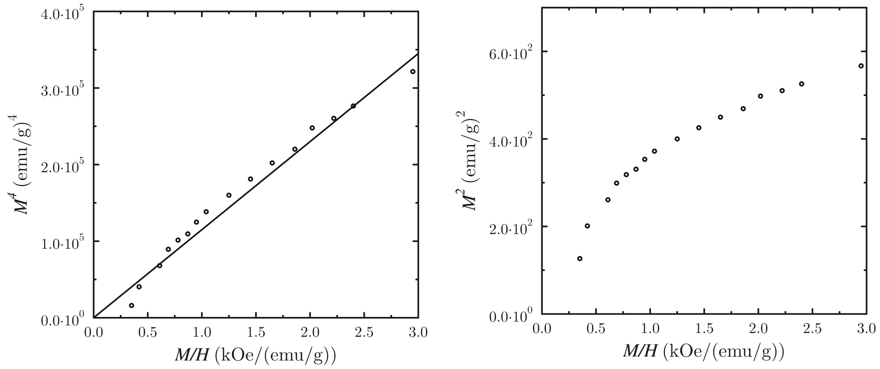


Fig. 3.4 M^4 versus H/M plot (*left*) and Arrott plot (*right*) for MnSi at $T = 29$ K by Bloch et al. [5]

and $y_z(\sigma, t) \ll 1$ are satisfied. It implies that the critical magnetic isotherm is predominantly influenced by the thermal amplitudes.

Note that no constant term is present in (3.58) at the critical point. Let us therefore assume a trial solution $y(\sigma) = y_c\sigma^{\delta-1}$. Then y_z is given by $y_z(\sigma) = \delta \cdot y_c\sigma^{\delta-1}$. By substituting them, (3.58) is now written as

$$\sigma^2 = \frac{3\pi t_c T_0}{4T_A}\sqrt{y_c} \cdot (2 + \sqrt{\delta})\sigma^{(\delta-1)/2}$$

From the comparison of both the sides, parameters δ and y_c of our trial solutions are determined as follows:

$$\delta = 5, \quad y_c = \left[\frac{4T_A}{3\pi T_c(2 + \sqrt{5})}\right]^2 \tag{3.59}$$

The value $\delta - 1 = 4$ is obtained as the critical index of the σ dependence of $y(\sigma)$.

In the theory of critical phenomena, the critical index δ is defined by the critical magnetic isotherm, i.e., $M \propto H^{1/\delta}$. The parameter δ in this section therefore corresponds to the critical index of magnetic isotherm. From the definition of the reduced inverse magnetic susceptibility, $y(\sigma) = h/2T_A\sigma$, the following relation is derived:

$$h = 2T_A\sigma y(\sigma) = 2T_A y_c\sigma^5, \quad H = \frac{T_A^3}{2[3\pi T_c(2 + \sqrt{5})]^2}\frac{M^5}{N_0^5\mu_B^6} \tag{3.60}$$

The first equation in the original units is given by the second.

It has long been believed that Arrott plot of magnetic isotherm will show good linearity independent of temperature. Among them MnSi was regarded as an exceptional. As will be seen in the right figure of Fig. 3.4, no good linearity is observed except at low temperatures. Later, the same good linearity is observed in $Fe_xCo_{1-x}Si$

Table 3.4 Comparison of two experimentally estimated values of T_A.

Compound	T_A (10^4 K)	$T_A^{(c)}$ (10^4 K)
MnSi	0.218	0.129
$Fe_xCo_{1-x}Si$		
$x = 0.36$	1.179	0.727
0.48	0.998	0.727
0.67	0.987	0.725
0.77	1.209	0.824
0.88	1.518	0.917
0.91	2.273	1.268

Those in the second and the third columns represent values from the isotherm in the ground state and from the critical isotherm, respectively

at the critical point by Shimizu et al. [16]. We should now regard the magnetic isotherm of MnSi as normal. It is known theoretically [2] that the temperature range of the critical magnetic isotherm becomes narrower for smaller t_c,

For convenience of comparison with experiments, the critical isotherm of (3.60) is rewritten as

$$\left(\frac{M}{M_s}\right)^4 = 1.20 \times 10^6 \frac{T_c^2}{T_A^3 p_s^4} \frac{H}{M}, \tag{3.61}$$

where $M_s = N_0 \mu_B p_s$ is the spontaneous magnetization in the ground state. Magnetic field H and induced magnetization M are measured in units of kOe and emu/mole, respectively. With the use of the relation (3.61), we can estimate the parameter T_A from the slope of M^4 versus H/M plot with known observed values of p_s and T_c. The values of T_A estimated in this way are compared in Table 3.4 with those estimated from the slope of Arrott plot in the ground state. From such an analysis, the linear relation $(M/M_s)^4 = 0.234 H/M_g$ (in units of kOe and emu/g for H and M_g) has been derived for MnSi [1]. By using $p_s = 0.4$, $M_g = 26.9$ (emu/g), $T_c = 30$ K, and $w_A = 83.024$, the value of $T_A = 0.129 \times 10^4$ is derived from (3.61), in fair good agreement with the value 2.1×10^3 from the neutron scattering experiment and the value given in Table 3.4.

Recently, magnetization measurements on itinerant ferromagnets with sizable spontaneous magnetic moments have been made around the critical temperature. They all seem to have relatively wide critical regions. For instance, magnetic isotherms of Ni and Ni_2MnGa at high temperatures have been measured by Nishihara et al. [22]. The good linearity is observed in M^4 versus H/M plot at the critical point for Ni as shown in Fig. 3.5. The critical index of the isotherm, estimate by assuming the relation $H \propto M^{\delta}$, is given by $\delta = 4.78$, which is close to our $\delta = 5$. From the slope of this figure, $T_A = 1.76 \times 10^4$ K is also estimated, being in good agreement with $T_A = 1.26 \times 10^4$ K [1] from the observed spin wave dispersion relation by neutron scattering experiment.

The magnetic isotherm of Fe with still larger spontaneous magnetic moment was measured by Hatta and Chikazumi [23] up to temperatures higher than the Curie

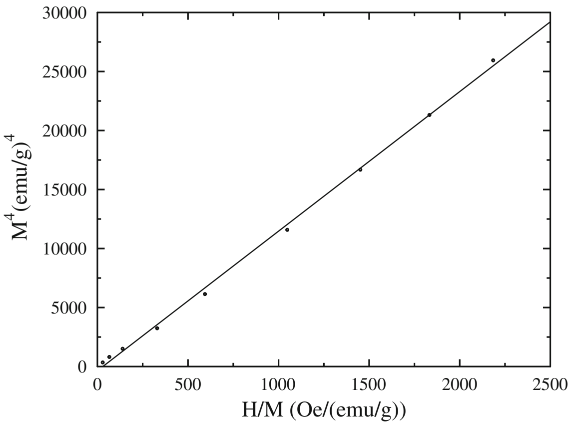


Fig. 3.5 M^4 versus H/M plot for Ni ($T = 623.2$ K) by Nishihara et al. [22]

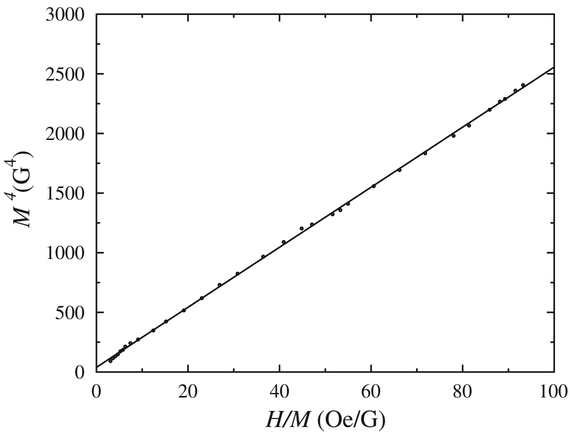


Fig. 3.6 M^4 versus H/M plot for Fe at the Curie temperature observed by Hatta and Chikazumi

temperature T_c. The Arrott plot at the critical point ($T = 1033$ K) is shown in Fig. 3.6, but in the form of M^4 versus H/M plot. The good linearity is also seen in this figure. As an optimal value of the critical index for Fe, $\delta = 4.6$ is obtained. Successive studies on critical magnetic isotherms have been made by Nishihara et al. on CoS_2 [24] and ferromagnetic Heusler alloys, Co_2VGa [25], Co_2CrGa [26]. Critical indexes estimated by them are summarized in Table 3.5.

In most cases hitherto, the critical temperature has been determined by using the Arrott plot. By extrapolating the linear part of graphs to the zero of the external magnetic field, it was estimated by the temperature of the graph, passing through the origin. The analysis is based on the implicit assumption that the linearity is always satisfied. Its revision is necessary, because it is not justified any more.

Table 3.5 Critical indexes of magnetic isotherms for various itinerant electron ferromagnets

	T_c (K)	p_s (μ_B)	p_{eff} (μ_B)	δ	T_A (K)
Ni	623	0.6	1.6	4.73	1.76×10^4
Fe	1366	2.2	3.2	4.6	
Ni_2MnGa	363	4.5/f.u.	3.4	4.77	
CoS_2	119.5	0.84		5.2	
Co_2CrGa	488	3.01/f.u.		4.93	1.0×10^4
Co_2VGa	351-358	2.1/f.u.		4.15	

3.4.2 Scaling Law Relations Among Critical Indexes

As temperature approaches the critical temperature, various magnetic properties show the following anomalous behaviors, called critical phenomena:

- Temperature dependence of magnetic susceptibility shows divergent behavior, following $\chi^{-1}(T) \propto (T - T_c)^{\gamma}$.
- Spontaneous magnetic moment approaches zero toward T_c according to $M_0(T) \propto (T - T_c)^{\beta}$.
- At the critical point, the relation $M \propto H^{1/\delta}$ is satisfied between the external magnetic field H and the induced magnetization M of the system.

Indexes γ, β, and δ are called *critical indexes*. It is known that the scaling law relation, $\gamma = \beta(\delta - 1)$, is satisfied among them. It is not the purpose of this book to determine these indexes as precisely as possible. We are rather interested in the internal consistency of these indexes, predicted by various theoretical studies. Let us first show in Table 3.6 values of indexes derived by the SW, the SCR, and the TAC-GC theories. In the following, we will briefly show the temperature dependence of the magnetic susceptibility and the spontaneous magnetization, as well as the critical magnetic isotherm predicted by these theories to see how the above indexes are derived.

- SW theory
  Indexes $\gamma = 1$ and $\beta = 1/2$ follow from the temperature dependence of magnetic susceptibility and spontaneous moment, $\chi^{-1} \propto (T^2 - T_c^2) \propto (T - T_c)$ and $M_0^2 \propto (T_c^2 - T^2) \propto (T_c - T)$. The disappearance of the coefficient $a(T_c)$ of the magnetic isotherm, $H = a(T)M + b(T)M^3 + \cdots$, at the critical point leads to $H \propto M^3$, giving $\delta = 3$.

Table 3.6 Theoretically derived values of critical indexes γ, β, and δ

Theory	γ	β	δ	$\beta(\delta - 1)$	$\gamma - \beta(\delta - 1)$
SW	1	1/2	3	1	0
SCR	2	1/2	3	1	1
TAC-GC	2	1/2	5	2	0

- SCR theory
  Temperature dependence of $\chi^{-1}(T)$ and $M_0^2(T)$ are proportional to $(T - T_c)^2$ and $(T_c^{4/3} - T^{4/3})$, respectively, around the critical point, giving the indexes $\gamma = 2$ and $\beta = 1/2$. Because the coefficient $b(T_c)$ remains finite at $T = T_c$, the same magnetic isotherm is derived as the SW theory, i.e., $\delta = 3$.
- TAC-GC theory
  Both $\chi^{-1}(T)$ and $M_0^2(T)$ show the same temperature dependence as those of the SCR theory, and therefore the same indexes $\gamma = 2$ and $\beta = 1/2$ are derived. On the contrary, $H \propto M^5$ is satisfied at the critical point, giving $\delta = 5$. The detailed treatment of the spontaneous magnetic moment below T_c is given in the next Chap. 4.

In the last column of Table 3.6, values of the difference $\gamma - \beta(\delta - 1)$ is shown. The consistency of the scaling law relation can be checked by whether this column is null or not. We can see the relation is violated in the SCR theory. It results from the inappropriate treatment of the magnetic isotherm.

3.5 Crossover Behavior Around the Quantum Critical Point

The phenomena observed in the limit of the vanishing critical temperature ($T_c \to 0$), called quantum critical point (QCP), have attracted much interest. These are usually called quantum critical phenomena. Two kinds of crossovers seem to be involved in this phenomena observed for itinerant electron ferromagnets, i.e., those between the critical and the low temperature behaviors, and between the classical and quantum critical behaviors.

In the following, we show the crossover behavior of itinerant electron ferromagnets around the QCP by paying particular attention to the temperature dependence of magnetic susceptibility.

3.5.1 Scaling Function

In three dimensions, the anomalous critical behavior is dominated by the thermal spin fluctuation amplitude, $\langle \mathbf{S}_i \cdot \mathbf{S}_i \rangle_{\mathrm{T}} (y, t) \propto A(y, t)$. Its y and t dependence around their origins plays a predominant role. The amplitude $A(y, t)$ defined in (2.83) is rewritten as

$$\begin{aligned} A(y,t) &= \int_0^1 \mathrm{d}x x^3 [\log u - 1/2u - \psi(u)], \quad u = x(y + x^2)/t \\ &= t^{4/3} \int_0^{t^{-1/3}} \mathrm{d}s s^3 [\log v - 1/2v - \psi(v)], \quad v = s(z + s^2) \end{aligned} \tag{3.62}$$

where we have defined a new variable $z = y/t^{2/3}$ and the inverse magnetic susceptibility $y(\sigma, t)$ is regarded as a function of temperature t and magnetic moment σ. At low temperatures where $t \ll 1$ is satisfied, it is written in the form

$$\begin{aligned} A(y,t) &= t^{4/3}F(z), \quad z = y/t^{2/3}, \\ F(z) &= \int_0^\infty \mathrm{d}s s^3[\log v - 1/2v - \psi(v)], \end{aligned} \tag{3.63}$$

where $F(z)$ is usually called the scaling function. In the case of conventional classical critical phenomena with finite critical temperature ($t_c > 0$), $y = 0$ at the critical point is equivalent to $z = 0$. On the other hand, the QCP, where both $y = 0$ and $t_c = 0$ are satisfied at the same time, corresponds to an ambiguous situation $0/0$ for z. In such a case, we must distinguish the order of the limiting processes, e.g., first take the limit $t \to 0$ while keeping y finite ($z \gg 1$), and vice versa.

Depending on the order of limiting processes, let us define the two regions, the low temperature and the critical regions. In these regions, the thermal spin fluctuation amplitude is expressed in the following different forms:

- Low temperature region for $z \gg 1$

$$A(y,t) \simeq \frac{t^2}{24y} = t^{4/3}\frac{t^{2/3}}{24y} = t^{4/3}\frac{1}{24z} \tag{3.64}$$

- Critical region for $z \ll 1$

$$\begin{aligned} A(y,t) &\simeq A(0,t) - \frac{\pi t}{4}\sqrt{y} \\ &= t^{4/3}\left(\frac{1}{3}C_{4/3} - \frac{\pi}{4}\sqrt{\frac{y}{t^{2/3}}}\right) = t^{4/3}\left(\frac{1}{3}C_{4/3} - \frac{\pi}{4}\sqrt{z}\right) \end{aligned} \tag{3.65}$$

The scaling function $F(z)$ in these regions is therefore given as

$$F(z) = \begin{cases} \dfrac{1}{24z}, & \text{for } z \gg 1 \\ \dfrac{C_{4/3}}{3} - \dfrac{\pi}{4}\sqrt{z}, & \text{for } z \ll 1 \end{cases} \tag{3.66}$$

We show in Fig. 3.7, the z dependence of the function $F(z)$. We can see that the critical region is restricted to the range, $z \lesssim 0.1$, around the origin.

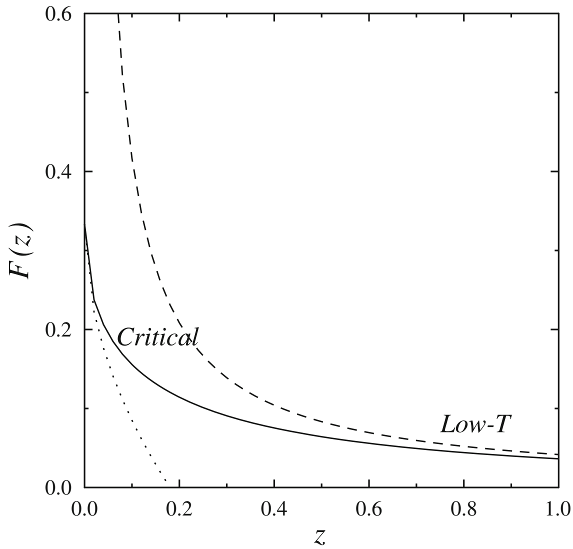


Fig. 3.7 The dependence of the scaling function $F(z)$ on the variable z. *Dotted curve* around the origin and *dashed curve* for $1 \lesssim z$ correspond to the critical and low temperature limits, respectively

3.5.2 Temperature Induced Crossover Between Classical and Quantum Critical Phenomena

As a typical magnetic property, we are particularly concerned with the inverse of the magnetic susceptibility $y(\sigma, t)$ in this section. Its temperature dependence will then draw a trajectory in $y - t$ space as shown in Fig. 3.8. Depending on the magnitude of z, the $y - t$ space is divided into two regions, the critical (for $z \ll 1$) and the low temperature regions (for $z \gg 1$), as will be shown in Fig. 3.8. Temperature dependence of y in each of these regions is determined, being influenced by the characteristic z dependence of the scaling function as given by (3.66). With variation of temperature or by the externally applied magnetic field, the trajectory of the system makes the transition from one region to another, giving rise to the crossover behavior. For instance, the temperature dependence of y for $t_c = 0$ is given by $y \propto t^{4/3}$ in the critical region. It will make the transition to the low temperature region in this y versus t plane in Fig. 3.8, with increasing temperature or by the externally applied magnetic field.

3.5.3 Temperature Dependence of Magnetic Susceptibility of Paramagnets Near the QC Point

Within the critical region, another crossover phenomena is expected to occur between the classical and the quantum critical phenomena. It results from the competition between the thermal and the quantum spin fluctuation amplitudes as will be described below.

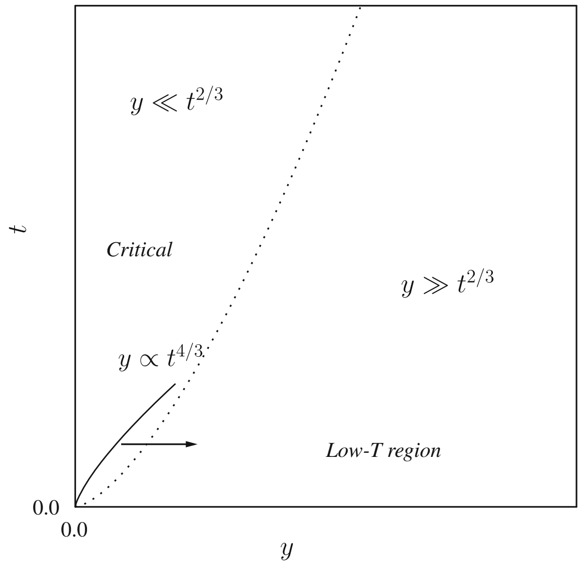


Fig. 3.8 Crossover behavior of the inverse of the magnetic susceptibility in the y-t space, divided in two regions, the critical and the low temperature regions. The *horizontal arrow* represents the effect of external magnetic field

Competition between Thermal and Zero-point Amplitudes at the Critical Point
Just at the critical point ($t_c = 0$) in the critical region, the temperature dependence of the magnetic susceptibility is determined as

$$A(y,t) - cy = t^{4/3}\left[F(z) - c\frac{z}{t^{2/3}}\right] = t^{4/3}\left[F(0) - \frac{\pi}{4}\sqrt{z} - c\frac{z}{t^{2/3}}\right] = 0. \quad (3.67)$$

We can simply find its solution as given by

$$\begin{aligned}\frac{F(0)}{c}t^{2/3} &= z + \frac{\pi}{4c}t^{2/3}\sqrt{z} = \left(\sqrt{z} + \frac{\pi}{8c}t^{2/3}\right)^2 - \left(\frac{\pi}{8c}\right)^2 t^{4/3},\\ z &= \left[\sqrt{\frac{F(0)}{c}t^{2/3} + \left(\frac{\pi}{8c}\right)^2 t^{4/3}} - \frac{\pi}{8c}t^{2/3}\right]^2 \\ &= \frac{F(0)}{c}t^{2/3} + 2\left(\frac{\pi}{8c}\right)^2 t^{4/3} - \frac{\pi}{4c}t\sqrt{\frac{F(0)}{c} + \left(\frac{\pi}{8c}\right)^2 t^{2/3}}.\end{aligned} \quad (3.68)$$

In the region of temperature where $(\pi/8c)^2 t^{2/3} \lesssim F(0)/c$ is satisfied, temperature dependence of z or y in this limit, $t_c = 0$, is given as

$$z \simeq \frac{F(0)}{c}t^{2/3}, \quad y \simeq \frac{F(0)}{c}t^{4/3}. \quad (3.69)$$

The thermal amplitude given by $t\sqrt{y} \propto t^{5/3} (\ll y)$ is neglected compared to the y linear dependence from the zero-point amplitude.

In the original variable y, (3.67) is also written as

$$cy + \frac{\pi t}{4}\sqrt{y} \simeq A(0,t) \propto t^{4/3}. \tag{3.70}$$

If we assume the dependence $y \propto t^{\alpha}$, the temperature dependence of the thermal amplitude is proportional to $t\sqrt{y} \propto t^{1+\alpha/2}$. As long as $\alpha < 2$ and $y \ll 1$ are both satisfied, $t\sqrt{y} \ll y$ is always satisfied. It implies that the $\sqrt{y}$ dependence of the thermal amplitude is neglected for $\alpha = 4/3$, compared to the y linear dependence of the zero-point amplitude. To summarize, the temperature dependence of y is determined, so as to balance the $t^{4/3}$-linear increase of the thermal amplitude at $y = 0$ by the y-linear suppression of the zero-point amplitude rather than the thermal amplitude.

Crossover Near the Critical Point In place of (3.67), the following equation is satisfied for ferromagnets with finite t_c.

$$\begin{aligned} A(y,t) - cy - A(0,t_c) &= F(0)(t^{4/3} - t_c^{4/3}) - t^{4/3}\left(\frac{\pi}{4}\sqrt{z} + c\frac{z}{t^{2/3}}\right) = 0, \\ \frac{F(0)}{c}g(t) &= \zeta + \frac{\pi}{4c}\sqrt{\zeta}, \quad g(t) = \frac{1}{t^2}(t^{4/3} - t_c^{4/3}), \end{aligned} \tag{3.71}$$

where we have defined a new variable, $\zeta = z/t^{4/3} = y/t^2$. By using the function $g(t)$, the temperature dependence of ζ in this case is represented as

$$\begin{aligned} \sqrt{\zeta} &= \sqrt{\frac{F(0)}{c}g(t) + \left(\frac{\pi}{8c}\right)^2} - \frac{\pi}{8c}, \\ \zeta &= \frac{F(0)}{c}g(t) + 2\left(\frac{\pi}{8c}\right)^2\left[1 - \sqrt{1 + \frac{64cF(0)}{\pi^2}g(t)}\right] \\ &= \frac{F(0)}{c}g(t)\left[1 - \frac{2}{1 + \sqrt{1 + \frac{64cF(0)}{\pi^2}g(t)}}\right] \\ &= \left[\frac{8F(0)}{\pi}g(t)\right]^2 \frac{1}{2 + \frac{64cF(0)}{\pi^2}g(t) + 2\sqrt{1 + \frac{64cF(0)}{\pi^2}g(t)}}. \end{aligned} \tag{3.72}$$

The crossover of the behavior of ζ is therefore controlled by the magnitude of the function $g(t)$. Depending on its magnitude, the above result can be written as follows:

$$\zeta \simeq \begin{cases} \left[\frac{4F(0)}{\pi}\right]^2 g^2(t) \simeq \left(\frac{16F(0)}{3\pi t^2}\right)^2 (t - t_c)^2, & g(t) \ll c/F(0) \\ \frac{F(0)}{c}g(t) = \frac{F(0)}{ct^2}(t^{4/3} - t_c^{4/3}), & g(t) \gg c/F(0) \end{cases} \tag{3.73}$$

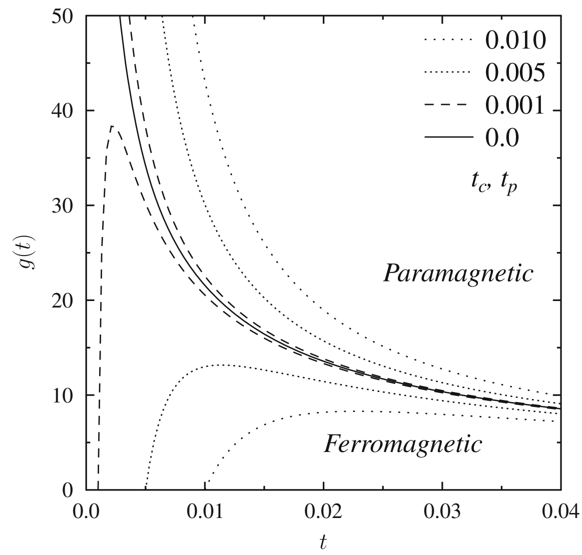


Fig. 3.9 Effect of the magnitude of parameters t_c and t_p on the temperature dependence of the function $g(t)$

It results from the difference whether we put $\zeta \simeq F(0)g(t)/c$ in (3.71) or $\sqrt{\zeta} \simeq F(0)g(t)/c$. For $F(0)/c \lesssim \zeta$, the thermal fluctuations are neglected because $\sqrt{\zeta} \lesssim \zeta$ is satisfied, while for $\zeta \lesssim F(0)/c$ the opposite is satisfied. The temperature dependence of $g(t)$ is shown in Fig. 3.9.

The crossover within the critical region is therefore expected to occur around the temperature t^*, determined by the condition $g(t) \sim c/F(0)$. The temperature t^* is estimated as follows:

$$g(t^*) \simeq \frac{4}{3t_c^2}(t^* - t_c) \simeq \frac{c}{F(0)}, \quad t^* = t_c + \frac{3c}{4F(0)}t_c^2 \tag{3.74}$$

For cases where $t_c \ll 1$ is satisfied, the temperature dependence of y is then given as

$$y \simeq \begin{cases} \left(\dfrac{16F(0)}{3\pi t}\right)^2 (t - t_c)^2, \; t_c \le t \lesssim t^* \\ \dfrac{F(0)}{c}(t^{4/3} - t_c^{4/3}), \qquad t^* \lesssim t \end{cases} \tag{3.75}$$

Around the critical point, it first starts from the classical $(t - t_c)^2$-linear dependence, and then crossovers to the quantum $t^{4/3}$-dependence with increasing temperature. It seems that the classical critical behavior has just intervened in the close vicinity of the critical temperature where the quantum $t^{4/3}$-linear temperature dependence becomes dominant everywhere. Such a $t^{4/3}$-linear dependence has long been observed in the temperature dependence of magnetic susceptibilities and spontaneous

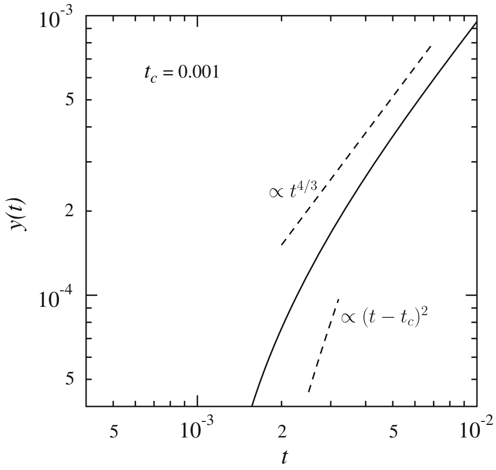


Fig. 3.10 Crossover from $(t - t_c)^2$ to $t^{4/3}$ behavior of temperature dependence of the inverse of magnetic susceptibility $y(t)$ in log-log plot

magnetizations of weak itinerant electron ferromagnets with small t_c. We show in Fig. 3.10, numerically calculated t dependence of $y(t)$ around the critical point $t = t_c$.

3.5.4 *Magnetic Susceptibility of Paramagnets Around the Quantum Critical Point*

Since y is always finite for paramagnets, they are regarded to be in the low temperature region at low temperatures. Their magnetic susceptibilities then obey the following equation:

$$c(y - y_0) - A(y, t) \simeq t^{4/3}\left(c\frac{z}{t^{2/3}} - \frac{1}{24z}\right) - cy_0 = 0, \quad z \to t^{-2/3}y_0, \quad y \to y_0, \quad (t \to 0) \tag{3.76}$$

The solution of (3.76) at low temperatures is given as

$$z \simeq t^{-2/3}y_0 + \frac{t^{4/3}}{24cy_0} + \cdots, \quad y \simeq y_0 + \frac{t^2}{24cy_0} + \cdots. \tag{3.77}$$

In the critical region where $y \lesssim t^{2/3}$ is satisfied, we expect a solution for y which satisfies the following inequality:

$$y = y_0 + \frac{1}{c}A(y, t) \simeq y_0 + \frac{t^2}{24cy_0} \lesssim t^{2/3}. \tag{3.78}$$

The temperature dependence of y in (3.77) has a contact with the critical boundary $y \simeq t^{2/3}$ at $y_0 = y_0^*$ and $t = t^*$ determined by the conditions given by

$$t^{2/3} \simeq y_0 + \frac{t^2}{24cy_0}, \quad \frac{2}{3t^{1/3}} = \frac{t}{12cy_0}. \tag{3.79}$$

As solutions of them, $y_0^* = 32c/9$ and $t^* = (8cy_0^*)^{3/4}$ are obtained. We therefore expect that the trajectory of y at low temperatures makes the transition to the critical region with increasing temperature, in cases where $y_0 \lesssim y_0^*$ is satisfied.

As with the case for ferromagnets within the critical region, the temperature dependence of the magnetic susceptibility is described as

$$c(y - y_0) - A(y, t) \simeq ct^2 \left[\zeta + \frac{\pi}{4c}\sqrt{\zeta} - \frac{F(0)}{c} g(t) \right] = 0. \tag{3.80}$$

In this case, however, the function $g(t)$ is defined as

$$g(t) = \frac{1}{t^2}(t^{4/3} + t_p^{4/3}), \quad t_p^{4/3} = \frac{cy_0}{F(0)}, \tag{3.81}$$

and therefore $g(t) \gg 1$ is always satisfied. The t dependence of $g(t)$ for various values of t_p is already shown in Fig. 3.9. The temperature dependence of ζ and y in this case is then given as

$$\zeta \simeq \frac{F(0)}{c} g(t) \simeq \frac{F(0)}{ct^2}(t^{4/3} + t_p^{4/3}), \quad y \simeq \frac{F(0)}{c}(t^{4/3} + t_p^{4/3}). \tag{3.82}$$

To conclude, the inverse of magnetic susceptibility of itinerant electron paramagnets near the QCP makes the transition

$$y_0 + \frac{t^2}{24cy_0} \Longrightarrow y_0 + \frac{F(0)}{c} t^{4/3}, \tag{3.83}$$

from the low temperature region to the quantum critical region with increasing temperature. In the limit of $y_0 \to 0$, the low-T region disappears and the critical $t^{4/3}$-linear dependence always becomes dominant at low temperatures.

3.6 Summary

We have shown in this chapter that a number of magnetic properties in the paramagnetic phase, as listed below, have been derived.

1. The magnetic isotherm in the ground state as determined under the influence of zero-point spin fluctuations.

2. The universal relation satisfied between p_{eff}/p_s versus T_c/T_0.
3. The vanishing of the fourth expansion coefficient $b(T)$ at the critical point.
4. The critical magnetic isotherm, $H \propto M^5$.

They are all related to the magnetic isotherm, i.e., the relation between the external magnetic field H and the induced magnetization M, and have been derived for the first time by the spin fluctuation theory presented in this chapter. Their validity has also been confirmed by many experimental studies.

The theory is based on the idea of the total spin amplitude conservation and the requirement that the dependence of the induced moment σ of the inverse of magnetic susceptibility $y(\sigma, t)$ has always to satisfy the TAC condition. It is also assumed that zero-point component of fluctuations is not to be neglected. As results, the magnetic isotherm becomes more flexible in this framework. All the expansion coefficients of the external magnetic field H in odd powers of the magnetization M will then become temperature dependent in principle. In contrast, the M dependence of the isotherm in the SCR theory is restrictive, since only the first expansion coefficient is assumed to depend on temperature. It is equivalent to assume that the linearity of the Arrott plot of magnetization curves is always satisfied. Because of this difference, the scaling law relation of critical phenomena is violated in the SCR theory. For our unified understanding of magnetic properties of itinerant electron magnets, both the effects of temperature and the external magnetic field have to be treated on equal footing.

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Chapter 4
Magnetic Properties in the Ordered Phase

4.1 Initial Value Problem

Even in the magnetically ordered phase, the magnetic isotherm is evaluated by solving the same ordinary differential equation as (3.25) given by

$$\Phi(\sigma^2, y, y_z, t) = 0, \quad y_z = y + \sigma \frac{\partial y}{\partial \sigma}, \tag{4.1}$$
$$\Phi(\sigma^2, y, y_z, t) \equiv 2A(y, t) + A(y_z, t) - c(2y + y_z) + \frac{T_A}{3T_0}\sigma^2 - 3A(0, t_c).$$

As a solution, inverse magnetic susceptibility $y(\sigma, t)$ is obtained as a function of σ. In reference to the magnetic isotherms in the ground state and in the paramagnetic phase, i.e., (3.7) and (3.48), respectively, it is reasonable to introduce the σ dependence of the inverses of magnetic susceptibilities in the ordered phase. They are given by

$$y(\sigma, t) = y_0(t) + y_1(t)\sigma^2 + \cdots = y_1(t)[\sigma^2 - \sigma_0^2(t)] + \cdots, \tag{4.2}$$
$$y_z(\sigma, t) = 2y_1(t)\sigma_0^2(t) + 3y_1(t)[\sigma^2 - \sigma_0^2(t)] = y_{z0}(t) + 3y(\sigma, t),$$

where $y(\sigma, t)$ and $y_z(\sigma, t)$, corresponding to H/M and $\partial H/\partial M$, are the perpendicular and the parallel components, respectively, to the uniform spontaneous magnetization. The value of $y_z(\sigma, t)$ in the absence of the external field for $\sigma = \sigma_0(t)$ is denoted by $y_{z0}(t) \equiv 2y_1(t)\sigma_0^2(t)$. In the ordered phase, $y_0(t)$ in the first line becomes negative, and the spontaneous moment squared, $\sigma_0^2(t) = -y_0(t)/y_1(t)$, appears. Both of these values, $\sigma_0(t)$ and $y_1(0)$, are therefore expected to be in agreement with σ_0 and y_1 in (3.10) and (3.11) in the ground state at $t = 0$. All the parameters, $y_0(t)$, $\sigma_0(t)$, and $\sigma_0(t)$, are regarded as functions of the reduced temperature t.

To find the initial conditions of (4.1), let us first consider the case, $H = 0$, in the absence of the magnetic field. Since $y(\sigma, t) = 0$ is then satisfied, $\sigma = \sigma_0(t)$ has to be satisfied in the ordered phase. Initial values of $y(\sigma, t)$ and $y_z(\sigma, t)$ at $H = 0$, after putting these values into (4.1), are shown in Table 4.1. Values in

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Table 4.1 Initial values of σ and inverse of magnetic susceptibilities, y and y_z, at $H = 0$ in the ordered phase

	σ	$y(\sigma, t)$	$y_z(\sigma, t)$
Ordered phase	$\sigma_0(t)$	0	$2y_1(t)\sigma_0^2(t)$
Paramagnetic phase	0	$y_0(t)$	$y_0(t)$

Same corresponding values in the paramagnetic phase are also shown

the paramagnetic phase are also shown in the same table for reference. Number of independent parameters is determined by putting these values into (4.1). In the ordered phase, two independent parameters, $\sigma_0(t)$ and $y_1(t)$, are involved in the initial values, whereas in the paramagnetic phase, only the single parameter $y_0(t)$ is present. An extra condition seems to be necessary to determine these parameters simultaneously, other than the single equation of (4.1).

To solve the problem, note that variables σ^2 and $y_z(\sigma, t)$ in the weak field limit are expanded with respect to the small parameter $y(\sigma, t)$ up to the linear term by

$$\sigma^2 = \sigma_0^2(t) + \frac{1}{y_1(t)} y(\sigma, t), \quad y_z(\sigma, t) = 2y_1(t)\sigma_0^2(t) + 3y(\sigma, t). \tag{4.3}$$

Equation (4.1) is also expanded up to the y-linear term as follows.

$$\begin{aligned} &\Phi(\sigma_0^2 + y/y_1, y, 2y_1\sigma_0^2 + 3y, t) = \Phi(\sigma_0^2, 0, 2y_1\sigma_0^2, t) \\ &\quad + y\left(\frac{1}{y_1}\frac{\partial}{\partial\sigma_0^2} + \frac{\partial}{\partial y} + 3\frac{\partial}{\partial y_{z0}}\right)\Phi(\sigma_0^2, y, y_{z0}, t)\Big|_{y=0} + \cdots = 0. \end{aligned} \tag{4.4}$$

Now our GC requirement on the external magnetic field effect implies that two independent conditions have to be satisfied in the limit, $\sigma = \sigma_0$, $y = 0$, and $y_z = y_{z0} = 2y_1\sigma_0^2$, as given by

$$\begin{aligned} &\Phi(\sigma, y, y_z, t) = 0, \\ &\left(\frac{1}{y_1}\frac{\partial}{\partial\sigma^2} + \frac{\partial}{\partial y} + 3\frac{\partial}{\partial y_z}\right)\Phi(\sigma^2, y, y_z, t) = 0. \end{aligned} \tag{4.5}$$

As solutions of these nonlinear simultaneous equations, we can uniquely determine both values of $\sigma_0(t)$ and $y_1(t)$.

4.1.1 *Analytic Property of Thermal Amplitude*

Another difficulty is still involved in the initial value problem in the ordered phase. The second condition of (4.5) implicitly assume that the condition $\Phi(\sigma^2, y, y_z) = 0$ can be expanded in powers of y around the origin $y = 0$. The definition (2.83) for the thermal amplitude of the transverse component is in contradiction to this analyticity. We briefly show below the non-analytic thermal amplitude leads to unphysical solution.

Let us assume anyway the following trial solution.

$$y(\sigma, t) = c[\sigma^2 - s^2(t)]^\gamma. \tag{4.6}$$

Then $y_z(\sigma, t)$ is given by

$$\begin{aligned} y_z(\sigma, t) &= y(\sigma, t) + 2c\gamma\sigma^2[\sigma^2 - s^2(t)]^{\gamma-1} \\ &\simeq 2c\gamma s^2(t)[\sigma^2 - s^2(t)]^{\gamma-1} \end{aligned} \tag{4.7}$$

where the lower order term $y(\sigma, t)$ in the right hand side is neglected in the weak field limit ($y \ll 1$). Since the $y(\sigma, t)$ dependence is also neglected in the TAC condition (4.1) as compared to y_z, it is given by

$$\frac{T_A}{3T_0}\sigma^2 - 3[A(0, t_c) - A(0, t)] - \frac{\pi t}{4}\sqrt{y_z(\sigma, t)} = 0. \tag{4.8}$$

As the solution, $y_z(\sigma, t)$ is given by

$$y_z(\sigma, t) \simeq \left(\frac{4T_A}{3\pi T_0 t}\right)^2 \left\{\sigma^2 - \frac{9T_0}{T_A}[A(0, t_c) - A(0, t)]\right\}^2. \tag{4.9}$$

By comparing (4.9) with (4.7), parameters γ, c, and $s^2(t)$ are determined as follows.

$$\gamma = 3, \quad c = \frac{1}{6}\left[\frac{4T_A}{3\pi T_0 t s(t)}\right]^2, \quad s^2(t) = \frac{9T_0}{T_A}[A(0, t_c) - A(0, t)] \tag{4.10}$$

The solution is, however, inappropriate because of the following reasons.

1. The spontaneous magnetic moment squared, $s^2(0) = 9T_0 A(t_c)/T_A = 3\sigma_0^2/5$ in the low temperature limit ($t \to 0$), is in disagreement with σ_0^2 in the ground state.
2. For the longitudinal component, $y_z = 0$ is always satisfied in the absence of the external magnetic field. It contracts with our magnetic isotherm in (4.2).

These inconvenient behaviors will suggest that we have to deal with analytic thermal spin fluctuation amplitudes except at the critical point.

4.1.2 Effect of the Presence of Spin Waves

Non-analyticity of the thermal spin fluctuation amplitudes is characteristic to the transverse component with respect to the static spontaneous magnetization.

No difficulty occurs in the paramagnetic phase, since inverse magnetic susceptibility y is always positive and finite. The same is true for the longitudinal amplitude in the ordered phase, for $y_z > 0$ is also satisfied. The difficulty is inherent only to the transverse thermal amplitude in the ordered phase, because $y = 0$ is *always* satisfied below T_c.

Inappropriate non-analytic $\sqrt{y}$ dependence at $y = 0$ originates from the wave-vector integration of the thermal amplitude in (2.83) around the origin $q = 0$. It is also known that there appear spin wave modes in this region. The range is, however, restricted within the narrow region ($0 \leq q \leq q_{\rm sw} \ll q_B$) for ferromagnets with small amplitude of spontaneous magnetization. Therefore, the effect has been usually neglected quantitatively. Its qualitative importance was first pointed by Takahashi [6] from the view of the analyticity. In the following, we show a simplified approach for recovering the analyticity of the thermal amplitude, by assuming that transverse component is given as a sum of the following two contributions:

$$A_\perp(y, t) \equiv A_{\rm sw}(t) + A_c(y, t). \tag{4.11}$$

The first and the second terms represent those of spin waves and spin fluctuations, respectively.

Contribution from Spin Fluctuations The second term of (4.11) is defined by

$$A_c(y, t) = \int_{x_c}^{1} {\rm d}x\, x^3 \left[\log u - \frac{1}{2u} - \psi(u)\right], \quad u = x(y + x^2)/t \tag{4.12}$$

where the lower bound $x_c = q_{\rm sw}/q_B$ is introduced to exclude the narrow spin-wave region, $0 \leq q \leq q_{\rm sw}$, around the origin. As will be shown below, the presence of the lower cut-off x_c in the above integral leads to the y-linear dependence of $A_c(y, t)$ around the origin $y = 0$, as long as $y \ll 1$ and $x_c \ll 1$ are satisfied.

$$\begin{aligned} A_c(y, t) - A_c(0, t) &\sim \frac{t}{2}\int_{x_c}^{1} {\rm d}x x^3 \left[\frac{1}{x(y + x^2)} - \frac{1}{x^3}\right] \\ &= -\frac{t}{2}\sqrt{y}\left[\tan^{-1}\left(\frac{1}{\sqrt{y}}\right) - \tan^{-1}\left(\frac{x_c}{\sqrt{y}}\right)\right] \\ &= -\frac{t}{2}\sqrt{y}\tan^{-1}\frac{\sqrt{y}(1 - x_c)}{y + x_c} \sim -\frac{t}{2} \times \begin{cases} \dfrac{y}{x_c}, & \text{for } y < x_c \\ \dfrac{\pi}{2}\sqrt{y}, & \text{for } x_c < y \end{cases} \end{aligned} \tag{4.13}$$

The integrand in (4.12) is, in the above derivation, approximated by $x^2/2u$. The analyticity is thus recovered around the origin $y = 0$. Phenomenologically, let us assume that both the transverse and the longitudinal spin fluctuation amplitudes suffer qualitatively the same suppression from the external magnetic field. In other word, the numerical parameter x_c is then written in the form

$$x_c = \frac{2}{\pi\xi}\sqrt{y_{z0}(t)}. \tag{4.14}$$

It is proportional to σ_0 at low temperatures, while it decreases, being in proportional to σ_0^2 as will be shown later. We still need to determine the parameter ξ. Later in Chap. 5, $\xi = 1$ is obtained from the continuity condition of the magnetic entropy at the critical temperature.

The result of Eq. (4.13) is equivalent with the assumption that the transverse amplitude of the thermal fluctuations is suppressed almost as much as the longitudinal one as the result of the appearance of spontaneous magnetization. The y dependence of $A_{\rm sw}$ is assumed to be neglected compared to that of $A_c(y, t)$. To summarize, thermal spin amplitudes around the origin is now written as follows:

$$\begin{aligned} A_\perp(y, t) &= A_\perp(0, t) - \frac{t}{2x_c}y + \cdots, \\ A(y_z, t) &= A(y_{z0}, t) - \frac{3\pi t}{8\sqrt{y_{z0}(t)}}y, + \cdots, \end{aligned} \tag{4.15}$$

where (4.3) is used for $y_z(\sigma, t)$ as a $y(\sigma, t)$-linear dependence.

Contribution from Spin Waves In the presence of the uniform and static magnetization, $\mathrm{Im}\chi(q, \omega)$ shows narrow high intensity peaks in the low-frequency and the long wave-length region in the q, ω space. They correspond to the long-lived damped eigen-oscillations, called spin-waves. The dispersion relation, $\omega_q = g\mu_{\rm B}H + Dq^2$, is satisfied between the peak frequency ω and the wave-vector q, in the long wave-length limit. The spin-wave part $A_{\rm sw}(t)$ in (4.11) is roughly estimated as a sum of such an intensity, which is well approximated by the following delta-function:

$$\mathrm{Im}\chi_\perp(q, \omega) \propto \sigma\delta(\omega - \omega_q), \tag{4.16}$$

at low temperatures. The thermal amplitude from spin-waves in the absence of external field is then evaluated by

$$A_{sw}(\sigma, t) = \frac{T_A\sigma}{2T_0\sigma_0}\int_0^{x_c}\frac{x^2}{\mathrm{e}^{\omega_q/T} - 1}\mathrm{d}x, \quad \omega_q = Dq_B^2x^2 = T_A(\sigma/\sigma_0)x^2, \tag{4.17}$$

where $Dq_{\rm B}^2$ is assumed to be equal to T_A. It characterizes the spin-wave dispersion in the wave-vector space. By assuming the continuity of integrands around the origin, the factor before the integral in (4.17) can be determined. The integrand of (4.17) around $x = 0$ given by

$$\frac{T_A\sigma}{2T_0\sigma_0}\frac{x^2}{\mathrm{e}^{T_A\sigma x^2/\sigma_0 T} - 1} \simeq \frac{T}{2T_0},$$

agrees with the lower bound limit of the integrand in (4.12) for $y = 0$, i.e.,

$$x^3[\ln u - 1/2u - \psi(u)] \simeq \frac{x^3}{2u} = \frac{Tx^3}{2T_0 x(y+x^2)} \to \frac{T}{2T_0}, \quad (y \to 0) \tag{4.18}$$

Let us next examine the influence of the temperature dependence of the thermal spin-wave amplitude. The upper bound of the integral is effectively restricted within the region, $x \lesssim (T/T_A)^{1/2}$, because of the Bose distribution function in (4.17). Depending on the relative magnitudes of x_c and $(T/T_A)^{1/2}$, the spin wave amplitude is estimated as follows:

$$\begin{aligned} A_{sw}(t) &\simeq \frac{T_A}{2T_0}\int_0^{x_c} \frac{x^2}{e^{T_A x^2/T}-1}\mathrm{d}x \\ &\simeq \frac{T_A}{2T_0} \times \begin{cases} \frac{\sqrt{\pi}}{4}\zeta(3/2)\left(\frac{T}{T_A}\right)^{3/2}, & (T/T_A)^{1/2} \lesssim x_c \\ \frac{T}{T_A}x_c, & x_c \lesssim (T/T_A)^{1/2} \end{cases} \end{aligned} \tag{4.19}$$

where the upper bound is assumed to be infinite when $(T/T_A)^{1/2} \lesssim x_c$ is satisfied. The result shows the crossover behavior from the characteristic $T^{3/2}$-linear dependence for spin-waves at low temperatures to the classical T-linear dependence with increasing temperature.

The condition for the appearance of the above $T^{3/2}$-linear dependence is also rewritten in the form

$$\frac{T}{T_A} < x_c^2 = \left(\frac{2}{\pi\xi}\right)^2 y_{z0}(0),$$

by putting the value of x_c in (4.14) at $t = 0$. With using the relation in (3.10), $cy_{z0}(0) = 2A(0, t_c) = 2C_{4/3}t_c^{4/3}/3$ satisfied in the ground state, it is further written as follows:

$$\frac{T}{T_c} < \frac{2T_A}{3cT_0}C_{4/3}\left(\frac{1}{\pi\xi}\right)^2 t_c^{1/3}.$$

Since $y_{z0}(t)$ actually depends on temperature, the above condition becomes more severe. Neverthless, it seems to be satisfied for wide region of temperature below T_c, considering that ratios of T_A/T_0 estimated for most itinerant weak ferromagnets have values of around 10. Its relative ratio to the total thermal amplitude, however, at the critical temperature, for instance, is given by

$$\frac{A_{sw}(t_c)}{A(0,t_c)} \sim \frac{x_c t_c/2}{t_c^{4/3}/3} = \frac{3}{2}\frac{x_c}{t_c^{1/3}} \ll 1, \quad \text{for } x_c \ll 1.$$

As long as the spin-wave is restricted within the very narrow region around the origin, its effect is neglected quantitatively.

4.2 Temperature Dependence of Spontaneous Moment

With the use of the analytic transverse thermal spin amplitude in the preceding section, we can derive the following set of simultaneous equations for initial conditions:

$$\begin{aligned} &2A_{\perp}(0,t) + A(y_{z0},t) - cy_{z0}(t) + 5cy_1(0)\sigma_0^2(t) - 3A(0,t_c) = 0, \\ &2A'_{\perp}(0,t) + 3A'(y_{z0},t) - 5c + 5c\frac{y_1(0)}{y_1(t)} = 0. \end{aligned} \tag{4.20}$$

They correspond to those in (4.5). In the following, the notation $A(y,t)$ is also used for $A_{\perp}(y,t)$ in the ordered phase to simplify the expression. Let us also introduce the following reduced parameters, $U(t)$ and $V(t)$, that correspond to $\sigma_0^2(t)$ and $y_{z0}(t)$.

$$U(t) = \frac{\sigma_0^2(t)}{\sigma_0^2(0)}, \quad V(t) = \frac{y_{z0}(t)}{y_{z0}(0)} = \frac{2y_1(t)\sigma_0^2(t)}{2y_1(0)\sigma_0^2(0)} = \frac{y_1(t)}{y_1(0)}U(t). \tag{4.21}$$

With these parameters, (4.1) is rewritten in the form

$$\begin{aligned} U(t) - \frac{2}{5}V(t) - \frac{3}{5} + \frac{1}{5A(0,t_c)}[2A(0,t) + A(y_{z0},t)] = 0, \\ V(t)\left[1 - \frac{2}{5c}A'(0,t) - \frac{3}{5c}A'(y_{z0},t)\right] - U(t) = 0, \end{aligned} \tag{4.22}$$

where $y_{z0}(t)$ is given by $y_{z0}(0)V(t)$. In the above derivation, $\sigma_0^2(0)$ and $y_1(t)$ are replaced by $A(0,t_c)$ and $V(t)/U(t)$, respectively, by using the relations, $A(0,t_c) = cy_1(0)\sigma_0^2(0)$ and $y_1(0)/y_1(t) = U(t)/V(t)$ in (4.21). As the solutions of (4.22), both of $\sigma_0(t)$ and $y_1(t)\sigma_0^2(t)$ are obtained simultaneously.

As a simplest example, variables $U(0)$ and $V(0)$ in the ground state satisfy the equations,

$$U(0) - \frac{2}{5}V(0) - \frac{3}{5} = 0, \quad V(t) - U(t) = 0,$$

because of the vanishing thermal amplitudes. Their solutions, $U(0) = V(0) = 1$, are in agreement with their definitions. To find general solutions at any temperatures, we have to resort to some numerical methods of calculation. The temperature dependence of solutions at low temperatures and around the critical temperature will follow in the following section.

4.2.1 Magnetic Properties at Low Temperatures

Reflecting the T^2 dependence of the thermal amplitude, the same temperature dependence is expected for various magnetic properties in this region. According to (2.88) in Chap.2, the thermal amplitudes are represented by

$$\begin{aligned} A(y,t) &\simeq A_{sw}(t) + \tfrac{t^2}{24(y+x_c^2)}, \quad A(y_z,t) \simeq \tfrac{t^2}{24y_z}, \\ A'(y,t) &\simeq -\tfrac{t^2}{24(y+x_c^2)^2}, \quad A'(y_z,t) \simeq -\tfrac{t^2}{24y_z^2}, \end{aligned} \tag{4.23}$$

where x_c is the lower bound of the wave-vector integral for the transverse thermal amplitude. By putting these expressions into the second equation of (4.22), the temperature dependence of $y_1(t)$ is derived:

$$\begin{aligned} \frac{y_1(t)}{y_1(0)} &= \frac{V(t)}{U(t)} = \left[1 - \frac{2}{5c}A'(0,t) - \frac{3}{5c}A'(y_{z0},t)\right]^{-1} \\ &= 1 - \frac{1}{5c}\frac{2(\pi\xi/2)^4+3}{24y_{z0}^2(0)}t^2 + \cdots \\ &= 1 - \frac{c[2(\pi/2)^4+3]}{480A^2(0,t_c)}\left(\frac{T}{T_0}\right)^2 + \cdots = 1 - \frac{b_0}{p_s^4}\left(\frac{T}{T_A}\right)^2 + \cdots, \\ b_0 &= \frac{15c}{2}[2(\pi/2)^4+3] = 56.91\ldots, \quad \text{for } c = 1/2 \end{aligned} \tag{4.24}$$

where the following relation satisfied in the ground state is used.

$$cy_{z0}(0) = 2A(0,t_c) = \frac{2T_A}{60T_0}p_s^2, \quad p_s = 2\sigma_0(0). \tag{4.25}$$

The above result implies that the expansion coefficient of the M^4 term of the free energy shows the decrease proportional to T^2 at low temperatures. In other words, the slope of the Arrott plot of the magnetization curve increases in proportion to T^2.

Then, to evaluate the temperature dependence of the spontaneous magnetic moment, let us first rewrite the first equation of (4.20) in the form

$$U(t)\left[1 - \frac{2}{3}\left(\frac{V(t)}{U(t)} - 1\right)\right] = 1 - \frac{2A(0,t) + A(y_{z0},t)}{3A(0,t_c)}.$$

By putting (4.24) and (4.23) for the thermal amplitudes into the left and the right hand sides of the above equation, respectively, the following result of $U(t)$ is derived:

$$\begin{aligned} U(t) &= \frac{1 - \dfrac{1}{3A(0,t_c)}\left(2A_{sw}(t) + \dfrac{c[2(\pi\xi/2)^2+1]}{48A(0,t_c)}t^2\right) + \cdots}{1 + \dfrac{2}{3}\dfrac{c[2(\pi\xi/2)^4+3]}{480A^2(0,t_c)}t^2 + \cdots} \\ &= 1 - \frac{2A_{sw}(t)}{3A(0,t_c)} - \frac{c[(\pi/2)^4 + 5(\pi/2)^2 + 4]}{360A^2(0,t_c)}\left(\frac{T}{T_0}\right)^2 + \cdots \end{aligned} \tag{4.26}$$

$$= 1 - \frac{2A_{sw}(t)}{3A(0,t_c)} - \frac{a_0}{p_s s^4}\left(\frac{T}{T_A}\right)^2 + \cdots,$$

$$a_0 = 10c[(\pi/2)^4 + 5(\pi/2)^2 + 4] = 112.1\ldots$$

If we neglect the effect of the spin wave, the spontaneous magnetic moment decreases in proportion to T^2 at low temperatures and its coefficient is expressed in terms of the parameters T_A and p_s.

4.2.2 Comparison with Experiments at Low Temperatures

Looking back on early experimental investigations, they are more or less affected by theoretical predictions and assertions. Methods of analysis are also sometimes deeply influenced by theories. Around the beginning of 1970s a number of experiments were made on magnetic properties in the ordered phase. Their aims were to confirm the temperature dependence predicted by the SW theory.

According to Wohlfarth and de Chatel [4], the magnetic isotherm is represented in the following form of expansion in powers of magnetization M and temperature T.

$$\frac{H}{M(H,T)} = -\frac{1}{2\chi_0}\left(1 - \alpha^{-1}A_1T^2 - \alpha^{-1}A_2T^4 - \cdots\right) + \frac{1}{2\chi_0}\frac{M^2(H,T)}{M^2(0,0)}(1 + B_1T^2 + B_2T^4 + \cdots). \quad (4.27)$$

The temperature dependence of the spontaneous magnetization is simply derived by the condition, $H = 0$.

$$\frac{M^2}{M_0^2} = 1 - (\alpha^{-1}A_2 + B_1)T^2 - (\alpha^{-1}A_2 - \alpha^{-1}A_1B_1 + B_2 - B_1^2)T^4 - \cdots. \quad (4.28)$$

As a slope of the Arrott plot, the following parameter $F(T)$ is introduced by them.

$$F(T) = \frac{\partial M^2(H,T)}{\partial[H/M(H,T)]} = \frac{2\chi_0 M^2(0,0)}{1 + B_1T^2 + B_2T^4 + \cdots}. \quad (4.29)$$

The value of $F(T)$ corresponds to the inverse of the coefficient $b(T)$ of the free energy (1.53). The T^2-linear dependence is therefore predicted by the above result. Experimentally observed this T^2 dependence of $F(T)$ and the spontaneous magnetic moment M in the ordered phase had been long regarded as the confirmation of the SW theory.

Fourth Order Expansion Coefficient of the Free Energy Temperature dependence of the observed slope of the Arrott plot of magnetization curves was analyzed by

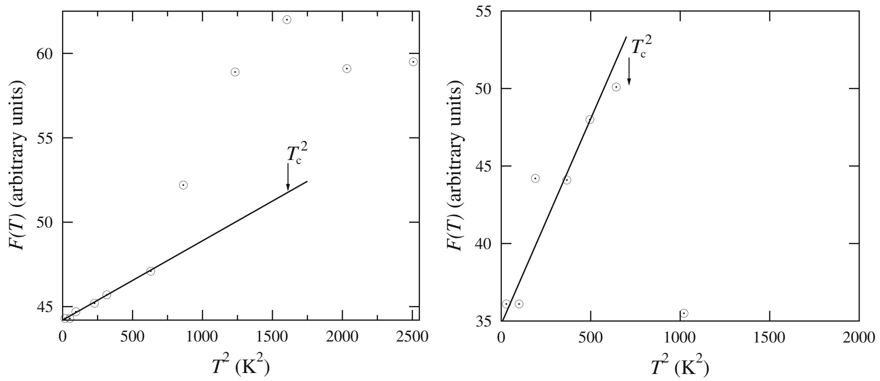

Fig. 4.1 Fourth expansion coefficients of $ZrTiZn_2$, $ZrZn_{1.9}$

Wohlfarth and de Chatel [4]. In Fig. 4.1, we show their estimated values of $F(T)$ against T^2 for $ZrZn_{1.9}$ by Knapp et al. [9] and for $Zr_{0.92}Ti_{0.08}Zn_2$ by Ogawa [3]. At low temperatures, slopes of these compounds fall well on straight lines with positive slopes. In Fig. 4.2, the inverse of the fourth order coefficient $1/b(T)$ for Ni–Pt alloys observed by Beille et al. [5] are plotted against T^2. These figures clearly demonstrate the presence of T^2-linear dependence in this coefficient. Temperature dependence shown in these figures corresponds to the negative coefficient B_1 in (4.27). It is also shown in Fig. 4.1 that there exists a tendency to deviate from the T^2-linear dependence with increasing temperature. Whereas the temperature dependence of the spontaneous magnetization seems to be well accounted by the single T^2-linear dependence throughout the wide range of temperature, the higher order terms are necessary for the fourth expansion coefficient $b(T)$. It is not so easy to realize this difference in the framework of the SW theory, since it is caused from the thermal smearing of the Fermi distribution function.

Later in the SCR theory, the temperature dependence of the coefficient $b(T)$ has been neglected. On the basis of the nonlinear mode–mode coupling mechanism, even higher order terms of the coupling among spin fluctuation modes are necessary to derive its temperature dependence. Inclusion of them is then likely to contradict to the observed linearity of the Arrott plot of the magnetization curve. The dependence has, therefore, become less and less interested after the appearance of the papers of Moriya and Kawabata [1, 2]. Since then it is usually assumed to be independent of temperature. Most Arrott plots of magnetization curves of many itinerant electron weak ferromagnets observed experimentally seem to show good linearity in the wide range of temperature.

In the preceding Sect. 4.2.1, we have shown that the fourth order coefficient $b(T)$ shows the T^2-linear dependence given by (4.24) based on the spin fluctuation mechanism. The behavior, restricted within a narrow range at low temperatures, is in agreement with the observed deviation from the T^2-linear behavior. For the quantitative comparison of (4.24) with experiments, let us define the constant β_0 by

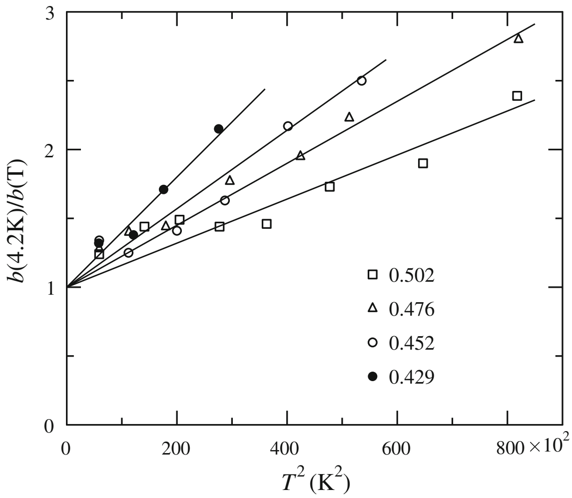

Fig. 4.2 Temperature dependence of $b(4.2K)/b(T)$ for Ni–Pt alloys observed by Beille et al.

Table 4.2 Comparison of values of the parameter T_A estimated from (4.31) and from the slope of the Arrott plot in the ground state

Compound	β_0 (K^{-2})	p_s (μ_B)	T_A (K)	$T_A{}^{(0)}$ (K)
$ZrZn_{1.9}$	7.4×10^{-4}	0.16	1.4×10^4	5.85×10^3
$Zr_{0.92}Ti_{0.08}Zn_2$	1.13×10^{-4}	0.233	1.6×10^4	5.92×10^3
$Pt_{1-x}Ni_x$				
$x = 0.429$	3.1×10^{-5}	0.051	4.5×10^5	3.07×10^4
0.452	2.9	0.104	1.3×10^5	2.46×10^4
0.476	2.3	0.143	7.7×10^4	2.08×10^4
0.502	1.6	0.179	5.8×10^4	2.04×10^4

$$\frac{b(T)}{b(0)} = \frac{y_1(t)}{y_1(0)} = 1 - \beta_0 T^2. \tag{4.30}$$

Values of β_0 estimated from the slopes of Figs. 4.1 and 4.2 are shown in Table 4.2. By comparing (4.24) with (4.30), the parameter T_A is represented in the form

$$T_A = \frac{1}{p_s^2}\sqrt{\frac{b_0}{\beta_0}}, \tag{4.31}$$

that enables us to estimate the parameter T_A experimentally. Values of T_A evaluated in this way are shown in the fourth column of Table 4.2. They are roughly in agreement with those in the last column shown as $T_A^{(0)}$ estimated from the slope of the Arrott plot of the magnetization curve in the ground state.

Spontaneous Magnetization It had long been believed that the temperature dependence of M^2 in most of weak itinerant electron ferromagnets is understood based on (4.27) of the SW theory. Observed values of M^2 against T^2 actually seem to be well fitted with a straight line. If however we see them more carefully, the dependence is not so simple. The results in Sect. 4.2.1 actually differ from the SW theory in the following respects.

- Temperature range of observed T^2-linear dependence
  While the T^2-linear behavior is satisfied in the wide range of temperature in the SW theory, it is restricted only within a narrow range at low temperatures in our view.
- Dependence of the slope of $\sigma^2(t)/\sigma^2(0)$ versus $(T/T_c)^2$ on the ratio $t_c = T_c/T_0$
  While it is almost independent of t_c in the SW theory, we predicts $t_c^{-2/3}$ dependence, i.e., the smaller the value of t_c the steeper the slope.

At low temperatures, observed spontaneous magnetic moment squared are well fitted with the T^2-linear decrease with steep negative slopes, in agreement with (4.26). To check this behavior experimentally, let us introduce another constant α_0 by

$$U(t) = 1 - \alpha_0 T^2 + \cdots \tag{4.32}$$

Then the following result is derived.

$$T_A = \frac{1}{p_s^2}\sqrt{\frac{a_0}{\alpha_0}} \tag{4.33}$$

The spectral parameter T_A can be also estimated from the T^2-linear coefficient of $\sigma_0^2(t)$ at low temperatures.

So far, temperature dependence of spontaneous magnetic moments of a number of weak itinerant electron ferromagnets have been measured; on Ni_3Al by de Boer et al. [7] and by Sasakura and Masuda [10], on $ZrZn_2$ by Ogawa [11], for instance. In addition, results on alloy systems (Fe,Co)Si are reported by Shimizu et al. [8] as well as on Pt–Ni alloys by Beille et al. [12], and Y_2Ni_{15}, YNi_3 by Gignoux et al. [13, 14]. In these references, values of $M_0^2(T)$ are plotted against T^2. We can estimate α_0 from the initial slope of these graphs in the limit of low temperature. The values of α_0 and T_A, estimated by using (4.33), are shown in Table 4.3. Recently, measurements of spontaneous magnetization of Ni–Pt has been made to check the validity of (4.26) by Koyama et al. [15]. Their observed results of M^2 are plotted against T^2 in Fig. 4.3. Only the results at low temperatures are fitted by them with a straight line. The value of T_A evaluated by (4.33) are also shown in Table 4.3, in agreement with the value $T_A^{(0)}$ estimated from the magnetic isotherm in the ground state. Depending on magnets, the T^2-linear dependence seems to be satisfied in a wide range of temperature below T_c. In many cases, however, we will find another restricted low temperature region where the same dependence is satisfied with a

Table 4.3 Values of T_A estimated from T^2-linear slope of σ^2

	p_s	α_0 (K^{-2})	T_A (K)	$T_A^{(0)}$ (K)	Refs.
$Ni_{100-x}Al_x$					Sasakura and Masuda [10]
$x = 25.3$	0.0474	2.77×10^{-3}	6.00×10^4	03.85×10^4	
$x = 25$	0.077	0.874×10^{-3}	4.08×10^4		
$x = 24.8$	0.0917	0.589×10^{-3}	3.48×10^4		
$x = 24.5$	0.110	0.386×10^{-3}	2.99×10^4		
Ni_3Al	0.075	0.784×10^{-3}	4.51×10^4	03.09×10^4	de Boer et al. [7]
$Ni_{75.5}Al_{24.5}$	0.104	0.372×10^{-3}	3.40×10^4		de Boer et al. [7]
$Ni_{76}Al_{24}$	0.125	0.246×10^{-3}	2.90×10^4		de Boer et al. [7]
$ZrZn_2$	0.12	2.69×10^{-3}	9.51×10^3	07.40×10^3	[11]
$Fe_{1-x}Si_x$					Shimizu et al. [8]
$x = 0.33$	0.22	0.400×10^{-3}	7.33×10^3	09.87×10^3	
$x = 0.23$	0.18	0.833×10^{-3}	7.59×10^3	12.09×10^3	
$x = 0.17$	0.13	1.49×10^{-3}	10.9×10^3	15.18×10^3	
$x = 0.09$	0.07	5.13×10^{-3}	20.2×10^3	22.73×10^3	
$Pt_{0.53}Ni_{0.47}$	0.121	1.30×10^{-4}	4.25×10^4		Beille et al. [12]
Y_2Ni_{15}	0.15	8.54×10^{-5}	3.41×10^4	03.51×10^4	Gignoux et al. [13]
YNi_3	0.04	1.20×10^{-3}	1.28×10^5	09.23×10^4	Gignoux et al. [14]
$Ni_{0.45}Pt_{0.55}$	0.182		1.0×10^4	00.69×10^4	Koyama et al. [15]

Values of $T_A^{(0)}$ estimated from the magnetic isotherm in the ground state

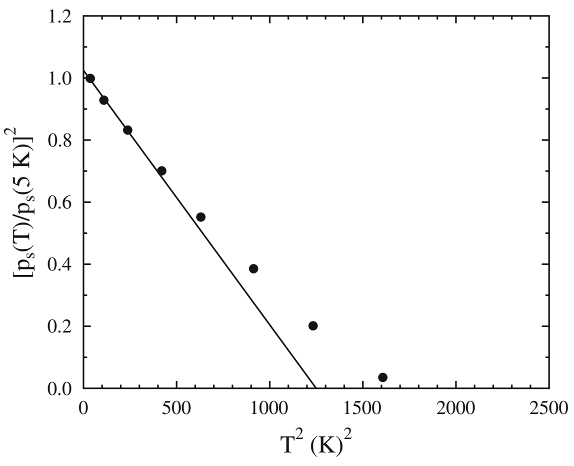

Fig. 4.3 Temperature dependence of the spontaneous moment of Ni–Pt by Koyama et al. [15]

steeper slope. The fair agreement of the values of T_A and $T_A^{(0)}$ in Table 4.3 clearly demonstrate the validity of (4.26).

4.2.3 Magnetic Properties Around the Critical Temperature

Analytical treatment is also available for properties around the critical point. By putting the critical thermal amplitudes (4.15) into the second equation of (4.22), it is rewritten in the form

$$\begin{aligned}\frac{U(t)}{V(t)} &\simeq \left(1 + \frac{2}{5c}\cdot\frac{\xi\pi t}{4\sqrt{y_{z0}(t)}} + \frac{3}{5c}\cdot\frac{\pi t}{8\sqrt{y_{z0}(t)}}\right) \\ &= 1 + \frac{(4\xi+3)\pi t}{40c}\,\frac{1}{\sqrt{y_{z0}(t)}} \simeq \frac{7\pi t_c}{40c}\left[\frac{c}{2A(0,t_c)V(t)}\right]^{1/2}, \end{aligned} \qquad (4.34)$$

where $\xi = 1$ is assumed as before. In the second line, the parameter $V(t)$ and $A(0, t_c)$ are substituted for $y_{z0}(t)$ and $y_{z0}(0)$, respectively, with the use of the definition (4.21) and $y_{z0}(0) = 2A(0, t_c)/c$ in (4.25). The following relation is therefore satisfied in this limit.

$$U(t) = \frac{7\pi t_c}{40c}\left[\frac{c}{2A(0,t_c)V(t)}\right]^{1/2} V(t) = \frac{7\pi t_c}{40c}\left[\frac{cV(t)}{2A(0,t_c)}\right]^{1/2}. \qquad (4.35)$$

It means that $V(t) \propto U^2(t)$ is satisfied for $U(t) \ll 1$.

In the first equation of (4.22), the variable $V(t)$ is then negligible since it is higher order than $U(t)$. With using the critical thermal amplitude, it is written as follows.

$$\begin{aligned} U(t) - \frac{\pi t_c}{20A(0,t_c)}\sqrt{y_{z0}(t)} &\simeq U(t) - \frac{\pi t_c}{10c}\sqrt{\frac{cV(t)}{2A(0,t_c)}} = \left(1 - \frac{4}{7}\right)U(t) \\ &= \frac{3}{5}\left[1 - \frac{A(0,t)}{A(0,t_c)}\right]. \end{aligned} \qquad (4.36)$$

Putting (4.35) into (4.34) also gives the ratio, $V(t)/U(t)$, given by

$$\begin{aligned} \frac{V(t)}{U(t)} &\simeq \frac{40c}{7\pi t_c}\left[\frac{2A(0,t_c)}{c}\right]^{1/2}\sqrt{V(t)} = \left[\frac{40c}{7\pi t_c}\right]^2 \frac{2A(0,t_c)}{c}U(t) \\ &= \frac{640cA(0,t_c)}{7(\pi t_c)^2}\left[1 - \frac{A(0,t)}{A(0,t_c)}\right]. \end{aligned} \qquad (4.37)$$

In the case of $t_c \ll 1$, (4.36) and (4.37) are also shown in the form

$$U(t) = \frac{\sigma_0^2(t)}{\sigma_0^2(0)} \simeq a_c[1 - (T/T_c)^{4/3}], \quad \frac{V(t)}{U(t)} = \frac{y_1(t)}{y_1(0)} \simeq b_c[1 - (T/T_c)^{4/3}], \qquad (4.38)$$

where $A(0, t) \propto t^{4/3}$ is used for the thermal amplitude. Coefficients a_c and b_c are defined by

$$a_c = \frac{7}{5}, \quad b_c = \frac{640cA(0,t_c)}{7(\pi t_c)^2} \simeq \frac{640cC_{4/3}}{21\pi^2 t_c^{2/3}}, \quad (t_c \ll 1) \tag{4.39}$$

To conclude, both $\sigma_0^2(t)$ and $y_1(t)$ show $(T - T_c)$-linear dependence around the critical point. The linear coefficient of the latter becomes steeper for cases with smaller t_c. It implies that the temperature dependence of the fourth expansion coefficient of the free energy with respect to M is actually restricted within a narrow range of temperature close to the critical point.

4.2.4 Numerical Results of Temperature Dependence

As an example of the general solutions of (4.22), numerically estimated results are shown in Fig. 4.4. In this figure, both the spontaneous magnetic moment squared $\sigma_0^2(t)$ and the coefficient $y_1(t)$ of the magnetization curve are plotted as functions of T/T_c. Both are proportional to $(T_c - T)$ around the critical point. Especially the steep slope of the curve observed for $y_1(t)$ around the critical point is due to the small parameter of $t_c = 0.05$, in agreement with (4.39). Both the values vanish simultaneously at the critical point.

4.2.5 Magnetization Curve

Once the initial conditions of the magnetic isotherm have been determined, what we need next is to solve the simultaneous differential equation (4.1). For solutions at arbitrary temperatures, we have to rely on numerical calculations. In this section, we show first that analytical solutions are available in the presence of weak external magnetic field and at low temperatures. Then, results of numerical studies are shown as an example of solutions at finite temperatures.

Magnetic Isotherm under the Weak External Magnetic Field at Low Temperatures The aim of this section is to extend our treatment of magnetic properties at low temperatures in Sect. 4.2.1 to the case in the presence of the external magnetic field. Substitution of $2y_1(t)\sigma^2 + y(\sigma,t)$ for $y_z(\sigma,t)$ in (4.1), justified in the weak field limit, then gives the following equation of the TAC condition of spin amplitude in the ordered phase.

$$\frac{1}{3A(0,t_c)}[2A(y,t) + A(y_z,t)] + \frac{c}{3A(0,t_c)}[5y_1(0) - 2y_1(t)]\sigma^2 - \frac{c}{A(0,t_c)}y = 1. \tag{4.40}$$

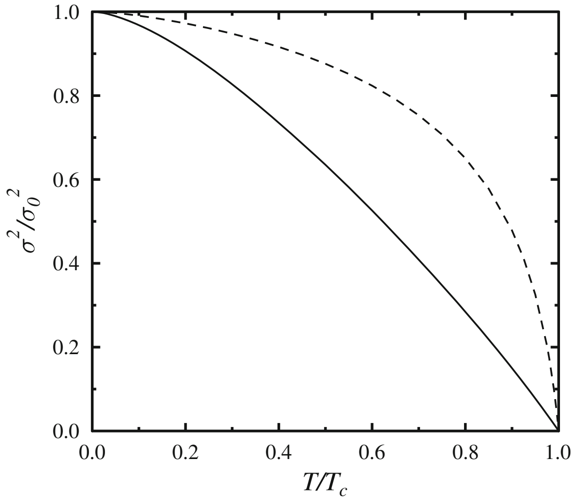

Fig. 4.4 Numerical results of the temperature dependence of $\sigma_0^2(t)/\sigma_0^2(0)$ and $y_1(t)/y_1(0)$ for $t_c = T_c/T_0 = 0.05$, $\sigma_s = \sigma_0(0) = 0.1$

Both sides are divided by $3A(0, t_c)$. If we further transpose terms in the left hand side to the right except for the σ^2-linear term, it is written in the form

$$\left[1 - \frac{2}{3}\left(\frac{y_1(t)}{y_1(0)} - 1\right)\right]\frac{\sigma^2}{\sigma_0^2(0)} = 1 + \frac{2y}{y_{z0}(0)} - \frac{2}{3cy_{z0}(0)}[2A(y,t) + A(y_z,t)]$$
$$= \left(1 + \frac{2y}{y_{z0}(0)}\right)\left(1 - \frac{4}{3c}\frac{2A(y,t) + A(y_z,t)}{y_{z0}(0) + 2y}\right), \tag{4.41}$$

with using the relation, $A(0, t_c) = cy_1(0)\sigma_0^2(0) = cy_{z0}(0)/2$, in (3.11). Finally, by putting the low temperature expressions of (4.23) and (4.24) for thermal amplitudes and $y_1(t)$, respectively, the following result is derived:

$$\left(1 + \frac{2y}{y_{z0}(0)}\right)^{-1}\frac{\sigma^2}{\sigma_0^2(0)}$$
$$= \left[1 - \frac{2}{3}\left(\frac{y_1(t)}{y_1(0)} - 1\right)\right]^{-1}\left(1 - \frac{4}{3c}\frac{2A(y,t) + A(y_z,t)}{y_{z0}(0) + 2y}\right)$$
$$= 1 - \frac{ct^2}{720A^2(0,t_c)}$$
$$\times \left\{2(\pi\xi/2)^4 + 3 - \frac{5}{1 + 2y/y_{z0}(0)}\left[\frac{2(\pi\xi/2)^2}{1 + y/x_c^2} + \frac{1}{1 + 3y/y_{z0}}\right]\right\}$$
$$= 1 - \frac{a_0(H)}{p_s^4}\left(\frac{T}{T_A}\right)^2. \tag{4.42}$$

Table 4.4 Field suppression of the T^2-linear coefficient of the spontaneous magnetization squared

H (kOe)	$\alpha_0(H)$ (K^{-2})	$\alpha_0(H)/\alpha_0(0)$
0.0	6.5 ± 1	1.0
25.0	4.8 ± 1	0.74
50.0	4.1 ± 0.5	0.63
75.0	3 ± 0.3	0.46
100.0	2.8 ± 0.3	0.43

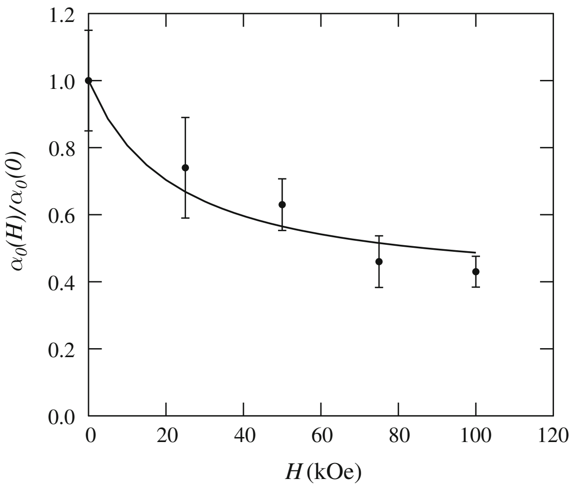

Fig. 4.5 Field dependence of T^2-linear coefficient of the spontaneous magnetization squared for Pt–Ni alloy systems

It corresponds to the extension of (4.26) to the case in the presence of the external magnetic field. Actually the coefficient $a_0(H)$ defined below reduces to a_0 in the limit of $y = 0$ ($H = 0$), i.e., $a_0(0) = a_0$ is satisfied.

$$a_0(H) = 5c\left\{2(\pi/2)^4 + 3 - \frac{5}{1 + 2y/y_{z0}(0)}\left[\frac{2(\pi/2)^2}{1 + y/x_c^2} + \frac{1}{1 + 3y/y_{z0}}\right]\right\} \quad (4.43)$$

The field effect on this coefficient $a_0(H)$ results from the σ dependence of $y(\sigma, t)$ in the right hand side. To evaluate its dependence at low temperatures, the value of σ has to be determined first from the magnetic isotherm (3.13) in the ground state. Then $y(\sigma, 0)$ is given by the ratio $h/(2T_A\sigma)$.

The effect of the external magnetic field on $a_0(H)$ of Ni–Pt alloys was experimentally investigated by Beille [12]. The observed results are shown in Table 4.4. For comparison with experiments, the values in this table are plotted against H in Fig. 4.5 with those estimated from (4.43) numerically. Although the number of data is limited, they are in good agreement with the theory even quantitatively. The same measurements have been also reported by Semwal and Kaul on Ni_3Al [16].

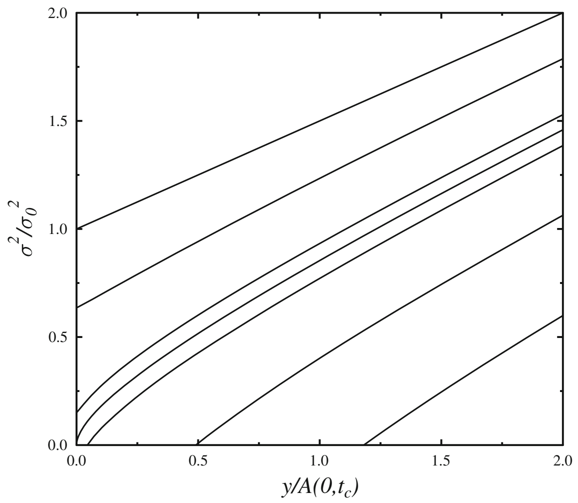

Fig. 4.6 Arrott plot of magnetic isotherms evaluated numerically

Numerical Results of Magnetization Curve We show in Fig. 4.6 the Arrott plot of magnetization curves, evaluated numerically by solving the differential equation (4.1). Linearity of the plot is satisfied at low temperatures. The initial slopes of curves in the weak field limit tend to increase with increasing temperature. Particularly around the critical temperature, they become very steep. These behaviors correspond to the temperature dependence of the M^4 term coefficient of the free energy. It is also clear that the critical isotherm, $y \propto \sigma^4$, is well justified around the critical region, in place of the σ^2-linear relation. The smaller the value of t_c, the narrower the critical region. Confirmation of the critical magnetic isotherm will be therefore very difficult for samples with $t_c \ll 1$.

4.3 Summary

In this chapter, it is shown that magnetic properties in the ordered phase are well described by the spin fluctuation theory. Up to around the middle of 1980s, the temperature dependence of $b(T)$ of the M^4 term of the free energy had been neglected in spin fluctuation theories, the SCR theory for instance. It contradicts, however, the scaling law relation of critical phenomena as shown in Chap. 3. We have shown that the temperature dependence of the spontaneous magnetic moment $\sigma_0(t)$ and the fourth expansion coefficient $y_1(t)$ are coupled with each other, and they simultaneously vanish at the critical temperature. The linearity of the Arrott plot of the magnetization curve is not generally satisfied except at low temperatures. These properties predicted by the theory have been confirmed by many experiments even quantitatively.

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Chapter 5
Thermal Properties of Itinerant Magnets

5.1 Difficulties Involved in the Spin Fluctuation Theory of Specific Heat

Temperature dependence of the specific heat of weak itinerant electron ferromagnets in a wide range of temperature was treated by Makoshi and Moriya [1]. The free energy used by them is written by

$$F(M,T) = F_{SW}(M,T) + F_{sf}(M,T,\chi^{-1}(T)). \tag{5.1}$$

It consists of the Stoner-Wohlfarth free energy F_{SW} and the contribution F_{sf} from thermal spin fluctuations. At low temperatures for exchange-enhanced paramagnets, it reduces to that of paramagnon theories for them. Moreover for ferromagnets, it can also be applied to properties at higher temperatures in the paramagnetic phase where the Curie-Weiss law temperature dependence of magnetic susceptibility is observed. Nevertheless, there exist the following difficulties:

1. As shown in the left figure of Fig. 5.1, a curious negative steep decrease of the specific heat appears just above the critical temperature with decreasing temperature.
2. It is based on the free energy that violates rotational invariance in the spin space. This is because only the transverse components of spin fluctuations are included in their treatment. Otherwise, spontaneous magnetic moment shows discontinuous change at the critical temperature.
3. Effects of zero-point spin fluctuations are neglected from the beginning.
4. The effect of the external magnetic field has not been treated by them. Their theory was later simply extended by Takeuchi and Masuda [2] to include the external magnetic field effect. Their numerically estimated changes of specific heat under the presence of magnetic fields of Sc_3In are compared with their experiments in Fig. 5.1.

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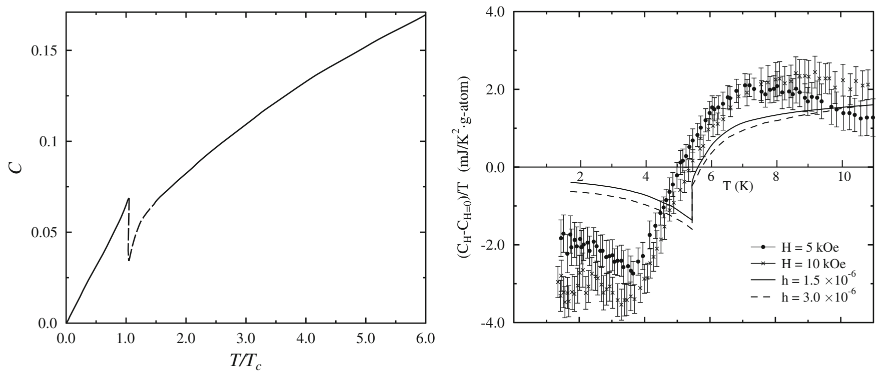

Fig. 5.1 Temperature dependence of the specific heat by Makoshi and Moriya derived from the SCR theory (*left*) and the effect of magnetic field on the specific heat of Sc_3In by Takeuchi and Masuda (*right*)

We will show in the following, how the temperature dependence and the external field effects of entropy and specific heat are derived based on our spin fluctuation theory presented in Chaps. 3 and 4.

5.2 Free Energy of Spin Fluctuations

In order to be consistent with our treatments of various magnetic properties, it will be better for the free energy to satisfy the following requirements:

- It is consistent with the total spin amplitude conservation (TAC). Then, the effect of zero-point spin fluctuations has to be included.
- The rotationally invariant treatment in the spin space has to be made. Thus, both the effects of transverse and the perpendicular components of spin fluctuations have to be included in the free energy.
- As a thermodynamically consistent treatment, the Maxwell relation on the external field effect of the magnetic entropy has to be satisfied.

5.2.1 Free Energy in the Presence of Magnetic Moment

For our treatments of properties in the magnetically ordered phase as well as effects of the external magnetic field, let us assume the following free energy:

$$F(y, \sigma, t) = F_0(y, \sigma, t) + \Delta F(\sigma, t)$$

$$
\begin{aligned}
F_0(y,\sigma,t) = F_{\mathrm{sw}} + \frac{2}{\pi}\Bigg[\sum_q \int_0^{\nu_c} \mathrm{d}\nu\, \frac{\nu}{2}\frac{\Gamma_q}{\Gamma_q^2+\nu^2} \\
+ \sum_{q_{\mathrm{sw}}<q}\int_0^{\infty}\mathrm{d}\nu\, T\ln(1-\mathrm{e}^{-\nu/T})\frac{\Gamma_q}{\Gamma_q^2+\nu^2}\Bigg] \\
+ \frac{1}{\pi}\sum_q\int_0^{\nu_c}\mathrm{d}\nu\,\left[\frac{\nu}{2}+T\ln(1-\mathrm{e}^{-\nu/T})\right]\frac{\Gamma_q^z}{(\Gamma_q^z)^2+\nu^2} + N_0T_A y\sigma^2 \\
\Delta F(\sigma,t) = -\frac{1}{3}N_0T_A\langle S_{\mathrm{loc}}^2\rangle_{\mathrm{tot}}[2y+y_z] + \Delta F_1(\sigma,t)
\end{aligned}
\tag{5.2}
$$

Aside from the additional contribution from zero-point spin fluctuations, the term F_0 in the first line corresponds to the free energy of the SCR theory. Both the perpendicular and parallel components of fluctuations with respect to the induced static moment are also included in F_0. The correction of the free energy ΔF, consisting of two contributions, will play significant role to satisfy the spin amplitude conservation, as will be shown in later subsections.

5.2.2 Stability Conditions of the Free Energy

In the following, let us assume that the free energy in (5.2) is a function of independent variables of σ, y, and the reduced temperature t. Since $\Delta y_z(\sigma,t) = y_z(\sigma,t) - y(\sigma,t)$ is regarded as a function of σ and t, it should not be regarded as an independent variable. These parameters are also assumed to be determined by the following conditions:

- From the stability condition of the free energy with respect to the variation of y, i.e., from $\partial F(y,\sigma,t)/\partial y = 0$, the following total spin amplitude conservation is derived.

$$
N_0T_A\left[\langle\delta S_{\mathrm{loc}}^2\rangle_{\mathrm{Z}}(y,y_z) + \langle\delta S_{\mathrm{loc}}^2\rangle_{\mathrm{T}}(y,y_z) + \sigma^2 - \langle S_{\mathrm{loc}}^2\rangle_{\mathrm{tot}}\right] = 0. \tag{5.3}
$$

The thermal and zero-point components of spin amplitudes are written in the form

$$
\begin{aligned}
\langle S_{\mathrm{loc}}^2\rangle_{\mathrm{T}}(y,y_z) &= \frac{3T_0}{T_A}[2A(y,t)+A(y_z,t)], \\
\langle S_{\mathrm{loc}}^2\rangle_{\mathrm{Z}}(y,y_z) &= \langle S_{\mathrm{loc}}^2\rangle_{\mathrm{Z}}(0,0) - c\frac{3T_A}{T_0}(2y+y_z).
\end{aligned}
\tag{5.4}
$$

The y dependence of the free energy in (5.2) results mainly from the implicit dependence through that of damping constants Γ_q and Γ_q^z defined in (2.79).

- The thermodynamic relation, $\partial F/\partial M = H$, has to be satisfied. Under the condition where the stability condition $\partial F(y, \sigma, t)/\partial y = 0$ is satisfied, its σ-derivative is given by

$$\frac{\partial F(y,\sigma,t)}{\partial \sigma} = 2N_0 T_A y\sigma + N_0 T_A \left[\left\langle (S_i^z)^2 \right\rangle (y_z, t) - \frac{1}{3} \left\langle S_{\rm loc}^2 \right\rangle_{\rm tot} \right] \frac{\partial \Delta y_z}{\partial \sigma} + \frac{\partial \Delta F_1}{\partial \sigma}. \quad (5.5)$$

The first term in the right hand side is equal to the external magnetic field $N_0 h$. The second term results from the Δy_z dependence of the parallel component of the spin fluctuations and the correction ΔF. With using (5.3), it can also be written as follows:

$$\begin{aligned} \left\langle (S_i^z)^2 \right\rangle (y_z, t) - \frac{1}{3} \left\langle S_i^2 \right\rangle_{\rm tot} &= \frac{1}{3} \left[2\left\langle (S_i^z)^2 \right\rangle (y_z, t) - \left\langle (S_i^\perp)^2 \right\rangle (y, t) - \sigma^2 \right] \\ &= \frac{2T_0}{T_A} [A(y_z, t) - A(y, t) - c\Delta y_z] - \frac{1}{3}\sigma^2. \quad (5.6) \end{aligned}$$

For $\sigma = 0$ in the absence of the magnetization, the above right hand side vanishes identically. Then the thermodynamic relation,

$$\frac{\partial F}{\partial \sigma} = 2N_0 T_A y\sigma = N_0 h, \quad (5.7)$$

is satisfied without introducing the correction term ΔF_1 in this case. Whereas for $\sigma \neq 0$, ΔF_1 is necessary, so that the last two terms in (5.5) cancel with each other. The correction ΔF_1 is thus defined by

$$\frac{1}{N_0 T_A} \frac{\partial \Delta F_1}{\partial \sigma} + \lambda(\sigma, t) \frac{\partial \Delta y_z}{\partial \sigma} = 0, \quad (5.8)$$

where $\lambda(\sigma, t)$ as the function of σ and t is also defined by

$$\lambda(\sigma, t) = \frac{2T_0}{T_A} [A(y + \Delta y_z, t) - A(y, t) - c\Delta y_z] - \frac{1}{3}\sigma^2. \quad (5.9)$$

5.2.3 Free Energy Corrections

Before proceeding further, we will show below how the σ dependence of $\Delta F_1(\sigma, t)$ is determined from its definition of (5.8) and (5.9) for $\lambda(\sigma, t)$ in the case of weak external magnetic field.

In the Paramagnetic Phase From the σ dependence of $y(\sigma, t)$ and $y_z(\sigma, t)$ in (3.48), $\Delta y_z(\sigma, t)$ is given by

$$\Delta y_z(\sigma, t) = 2y_1(t)\sigma^2 + \cdots . \tag{5.10}$$

By substituting the above result for (5.9), we obtain the σ dependence of $\lambda(\sigma, t)$ given by

$$\lambda(\sigma, t) = -\frac{4}{15}\left[1 - \frac{1}{c}A'(y, t)\right]\frac{y_1(t)}{y_1(0)}\sigma^2 - \frac{1}{3}\sigma^2 + \cdots = -\frac{3}{5}\sigma^2 + \cdots , \tag{5.11}$$

with use of the relations $T_A/T_0 = 15cy_1(0)$ in (3.10), and $y_1(t) = y_1(0)/[1 - A'(y, t)/c]$ in (3.50). By putting these results into (5.8), the correction $\Delta F_1(\sigma, t)$ is evaluated as follows:

$$\frac{1}{N_0 T_A}\Delta F_1(\sigma, t) = -4y_1(t)\int_0^\sigma \sigma'\lambda(\sigma', t)\mathrm{d}\sigma' = \frac{3}{5}y_1(t)\sigma^4 + \cdots . \tag{5.12}$$

In the Magnetically Ordered Phase If we notice the σ dependence of $y(\sigma, t)$ and $y_z(\sigma, t)$ in (4.2), $\Delta y_z(\sigma, t)$ is given by

$$\Delta y_z(\sigma, t) = 2y_1(t)\sigma^2 = 2y_1(t)\sigma_0^2(t) + 2y(\sigma, t).$$

As with the derivation of (5.11), the substitution of the above result for (5.9) gives the following expression of $\lambda(\sigma, t)$:

$$\begin{aligned}
\lambda(\sigma, t) &= \lambda(\sigma_0, t) + \delta\lambda(\sigma, t) \\
\lambda(\sigma_0, t) &= -\left[\frac{1}{3} + \frac{4y_1(t)}{15y_1(0)}\right]\sigma_0^2(t) + \frac{2}{15cy_1(0)}\left[A(2y_1\sigma_0^2, t) - A(0, t)\right] \\
\delta\lambda(\sigma, t) &= -\left\{\frac{1}{3} + \frac{4y_1(t)}{15y_1(0)}\left[1 - \frac{3}{2c}A'(2y_1\sigma_0^2, t) + \frac{1}{2c}A'(0, t)\right]\right\} \\
&\quad \times[\sigma^2 - \sigma_0^2(t)] + \cdots
\end{aligned} \tag{5.13}$$

The first term $\lambda(\sigma_0, t)$ represents the effect of the appearance of $\sigma_0(t)$, while the second term $\delta\lambda(\delta, t)$ is induced by external magnetic field. In the limit of zero-temperature, they reduce to

$$\lambda(\sigma_0, 0) = -\frac{3}{5}\sigma_0^2(0), \quad \delta\lambda(\sigma, 0) = -\frac{3}{5}[\sigma^2 - \sigma_0^2(0)]. \tag{5.14}$$

The correction $\Delta F_1(\sigma, t)$ is then evaluated by the following integration of (5.8) with respect to σ.

$$\frac{1}{N_0 T_A}\Delta F_1(\sigma, t) = -\int[\lambda(\sigma_0, t) + \delta\lambda(\sigma', t)]\frac{\partial\Delta y_z}{\partial\sigma'}\mathrm{d}\sigma'$$

$$= -\lambda(\sigma_0, t) \int \mathrm{d}\Delta y_z - 4y_1(t) \int \sigma' \delta\lambda(\sigma', t)\mathrm{d}\sigma', \tag{5.15}$$

where the approximation, $\partial \Delta y_z / \partial \sigma \simeq 4y_1(t)\sigma$, is used in the last line. The result is written in the form,

$$\begin{aligned} \frac{1}{N_0 T_A} \Delta F_1(\sigma, t) &= -\lambda(\sigma_0, t) \Delta y_z(\sigma, t) \\ &+ y_1(t) \left\{ \frac{1}{3} + \frac{4y_1(t)}{15y_1(0)} \left[1 - \frac{3}{2c} A'(2y_1\sigma_0^2, t) + \frac{1}{2c} A'(0, t) \right] \right\} \\ &\times [\sigma^2 - \sigma_0^2(t)]^2. \end{aligned} \tag{5.16}$$

Note the presence of the first correction term even in the absence of the external magnetic field. In the limit $\sigma_0(t) = 0$, it agrees with (5.12) in the paramagnetic phase.

At the Critical Temperature In this case, substitution of the $\sqrt{y}$ linear dependence for the thermal amplitude in (5.9) leads to the following expression:

$$\begin{aligned} \lambda(\sigma, t_c) &\simeq -\frac{2T_0}{T_A} \frac{\pi t_c}{4} (\sqrt{y_z} - \sqrt{y}) - \frac{1}{3}\sigma^2 = -\left[\frac{\pi T_c}{2T_A} \sqrt{y_c} (\sqrt{5} - 1) + \frac{1}{3} \right] \sigma^2 \\ &= -\frac{\sqrt{5}}{\sqrt{5} + 2} \sigma^2, \end{aligned} \tag{5.17}$$

with using the critical magnetic isotherms $y(\sigma, t_c) = y_c\sigma^4$ and $y_z(\sigma, t_c) = 5y_c\sigma^4$. The critical free energy correction is therefore given by

$$\Delta F_1(\sigma, t_c) \simeq -16N_0 T_A y_c \int_0^\sigma \sigma'^3 \lambda(\sigma', t_c)\mathrm{d}\sigma' = N_0 T_A \frac{8\sqrt{5} y_c}{3(2 + \sqrt{5})} \sigma^6. \tag{5.18}$$

The coefficient $y_1(t)$ of the σ^4 term of the free energy vanishes at the critical point. The correction ΔF_1 also becomes proportional to σ^6.

We have shown that the free energy in (5.2) is consistent with the TAC condition. The variational condition of the free energy with respect to the variable y agrees with the TAC condition. In the case of systems with the finite induced magnetization σ, we need to introduce the extra correction term $\Delta F_1(\sigma, t)$ in the free energy. Otherwise the thermodynamic relation is violated.

5.3 Temperature Dependence of Entropy and Specific Heat

In this section, the magnetic entropy is derived from the derivative of the free energy in (5.2) with respect to temperature T. The temperature dependence of the specific heat is then derived by differentiating the entropy again with respect to T.

5.3.1 *Temperature Dependence of Paramagnetic Entropy*

In the paramagnetic phase, the effect of spin waves, the difference between y and y_z, and the free energy correction ΔF_1 are all neglected in (5.2). Under the condition that $\partial F(y,t)/\partial y = 0$ is satisfied, the entropy is evaluated by differentiating the free energy with respect to temperature.

$$
\begin{aligned}
S_m(y,t) = -\frac{\partial F(y,t)}{\partial T} &= \frac{3}{\pi}\sum_q \left[-\int_0^\infty \mathrm{d}\nu \log(1-\mathrm{e}^{-\nu/T})\frac{\Gamma_q}{\nu^2+\Gamma_q^2}\right. \\
&\qquad \left. + \frac{1}{T}\int_0^\infty \mathrm{d}\nu \frac{1}{\mathrm{e}^{\nu/T}-1}\frac{\Gamma_q \nu}{\nu^2+\Gamma_q^2}\right] \\
&= 6\sum_q \left[-\frac{1}{2\pi}\int_0^\infty \mathrm{d}s \log(1-\mathrm{e}^{-2\pi s})\frac{u}{s^2+u^2}\right. \\
&\qquad \left. + u\int_0^\infty \mathrm{d}s \frac{s}{\mathrm{e}^{2\pi s}-1}\frac{1}{s^2+u^2}\right],
\end{aligned}
\tag{5.19}
$$

where new variables $s = \nu/2\pi T$ and $u(q) = \Gamma_q/2\pi T$ are introduced. In more simplified form, it is also written by

$$
\begin{aligned}
\frac{1}{N_0}S_m(y,t) &= -\frac{1}{N_0 T_0}\frac{\partial F(y,t)}{\partial t}, \\
&= -9\int_0^1 \mathrm{d}x x^2[\Phi(u) - u\Phi'(u)], \quad u = x(y+x^2)/t,
\end{aligned}
\tag{5.20}
$$

by introducing the new function $\Phi(z)$. A brief explanation of this function is given below.

Integral expression of $\Phi(z)$ The function $\Phi(z)$ is related to the logarithm of the gamma function $\Gamma(z)$ and is expressed in the following integral form:

$$
\begin{aligned}
\Phi(z) &= \log\sqrt{2\pi} - z + \left(z - \frac{1}{2}\right)\ln z - \log\Gamma(z) \\
&= \frac{1}{\pi}\int_0^\infty \mathrm{d}s \log(1-\mathrm{e}^{-2\pi s})\frac{z}{s^2+z^2}
\end{aligned}
\tag{5.21}
$$

The derivative of $\Phi(z)$ by z is equivalent with the integral expression of the digamma function $\psi(z)$.

$$
\begin{aligned}
\Phi'(z) &= \frac{1}{\pi}\int_0^\infty \mathrm{d}t \log(1-\mathrm{e}^{-2\pi t})\frac{\partial}{\partial z}\left(\frac{z}{t^2+z^2}\right) \\
&= -\frac{1}{\pi}\int_0^\infty \mathrm{d}t \log(1-\mathrm{e}^{-2\pi t})\frac{\partial}{\partial t}\left(\frac{t}{t^2+z^2}\right)
\end{aligned}
$$

$$= \int_0^\infty \mathrm{d}t \frac{2}{\mathrm{e}^{2\pi t} - 1} \frac{t}{t^2 + z^2} = \log z - \frac{1}{2z} - \psi(z)$$

From our expression of the entropy (5.20), the following interesting consequences are derived:

- The following term in the theory of Makoshi and Moriya is absent in (5.20).

$$-\frac{T_A}{T_0} \langle S_{\mathrm{loc}}^2 \rangle_{\mathrm{T}}(t) \frac{\mathrm{d}y}{\mathrm{d}t} \tag{5.22}$$

  The reason is because it disappears from the stability condition (5.3) of the free energy with respect to y. For the same reason, the effect of zero-point fluctuations does not appear.
- If the above term is present in the entropy, its temperature derivative gives the term proportional to $\mathrm{d}^2 y/\mathrm{d}t^2$, resulting in the negative peak in the temperature dependence of the specific heat just above the critical point.

5.3.2 Temperature Dependence of the Specific Heat

The paramagnetic specific heat is derived by the temperature derivative of the entropy in (5.20). It is given by

$$\begin{aligned}\frac{1}{N_0 t} C_m(y,t) &= \frac{1}{N_0 T_0} \frac{\partial S(y,t)}{\partial t} = 9 \int_0^1 \mathrm{d}x x^2 \left(-\frac{u}{t} + \frac{x}{t} \frac{\mathrm{d}y}{\mathrm{d}t} \right) u \Phi''(u) \\ &= -\frac{9}{t} \int_0^1 \mathrm{d}x x^2 u^2 \Phi''(u) - 9 \frac{\partial A(y,t)}{\partial t} \frac{\mathrm{d}y}{\mathrm{d}t}, \quad u = x(y + x^2)/t\end{aligned} \tag{5.23}$$

The coefficient of the second $\mathrm{d}y/\mathrm{d}t$ linear term is derived as follows.

If we notice the definition of the thermal amplitude $A(y,t)$ in (2.83), it can also be written in the form

$$A(y,t) = \int_0^1 \mathrm{d}x x^3 \Phi'(u). \tag{5.24}$$

Under the constant y condition, the partial t derivative of (5.24) is given by

$$-\frac{\partial A(y,t)}{\partial t} = -\frac{\partial}{\partial t} \int_0^1 \mathrm{d}x x^3 \Phi'(u) = \frac{1}{t} \int_0^1 \mathrm{d}x x^3 u \Phi''(u), \tag{5.25}$$

with use of the relation $\partial u/\partial t = -u/t$. Both the integrands, u linear term in (5.23) and the other one in (5.25), are in agreement with each other.

In the Low Temperature Limit In this range of temperature, both the inverse of the magnetic susceptibility $y(t)$ and the thermal amplitude are proportional to t^2. Since their t derivatives are both proportional to t, the second term of (5.23) is proportional to t^2 and is therefore negligible. Main contribution results from the following integral:

$$I(y,t) = -\frac{1}{t}\int_0^1 x^2u^2\Phi''(u)\mathrm{d}x, \quad \Phi''(u) = \left[\frac{1}{u} + \frac{1}{2u^2} - \psi'(u)\right] \tag{5.26}$$

Reflecting to the property of the digamma function, the integrand of (5.26) is approximated by

$$-\frac{1}{t}x^2u^2\Phi''(u) \sim \begin{cases} \dfrac{1}{2t}x^2, & \text{for } u \ll 1 \\ \dfrac{1}{6tu}x^2 = \dfrac{x}{6(y+x^2)}, & \text{for } u \gg 1 \end{cases} \tag{5.27}$$

To find the temperature dependence of the function $I(y,t)$, let us introduce the new variable $x' = x/t^{1/3}$ and represent u by

$$u = x'(y/t^{2/3} + x'^2) = x'(x_0^2 + x'^2), \quad x_0 \equiv y^{1/2}/t^{1/3}.$$

Then only the single parameter x_0 is involved in the integrand. The range of the integration is modified to be $0 \le x' \le 1/t^{1/3}$. Depending on the relative magnitude of x_0 and 1, the integral is estimated as follows:

1. In the case where $x_0 \lesssim 1$ ($y \lesssim t^{2/3}$) is satisfied
   In the range, $x_0 \le x' \le 1/t^{1/3}$, u is approximated by $u \simeq x'^3 = x^3/t$. The integration over the range, $1 \le x' \le 1/t^{1/3}$ within this region, gives

$$I(y,t) \simeq \frac{1}{6}\int_{t^{1/3}}^1 \frac{1}{x}\mathrm{d}x = \frac{1}{12}\log(1/t^{2/3}). \tag{5.28}$$

   Integration from the other region only gives a finite result.
2. In the case, $1 \lesssim x_0$ ($t^{2/3} \lesssim y$).
   The asymptotic expansion in this case is justified for $u \sim x_0^2x' = yx/t > 1$, for $u \sim x_0^2x'$ is satisfied around $x' = 0$. In terms of the original variable x, the integral in this region is evaluated by

$$I(y,t) \simeq \frac{1}{6}\int_{t/y}^1 \frac{x}{y+x^2}\mathrm{d}x = \frac{1}{12}\log\left(\frac{1+y}{y+t^2/y^2}\right) \simeq \frac{1}{12}\log(1/y), \tag{5.29}$$

   where $t^2/y^3 \ll 1$ (i.e., $t^{2/3} \ll y$) is assumed to be satisfied. The integral over the small range $0 \le x \le t/y$ around the origin is also negligible in this case.

To summarize, for exchange-enhanced paramagnets where $y \ll 1$ is satisfied, their temperature dependence of the specific heat at low temperatures is given by

$$\frac{1}{N_0 t} C_m \simeq \begin{cases} \dfrac{1}{2}\log(1/t), & (y \ll t^{2/3}, \text{ or } y/t^{2/3} \ll 1) \\ \dfrac{3}{4}\log(1/y). & (t^{2/3} \ll y, \text{ or } y/t^{2/3} \gg 1). \end{cases} \tag{5.30}$$

These are regarded as characteristic behaviors in the critical region for $z = y/t^{2/3} \ll 1$ in (3.65), and in the low temperature region for $z = y/t^{2/3} \gg 1$ around the QCP.

Temperature Dependence Around the Critical Temperature Around the critical temperature in the paramagnetic phase, we need to deal with the limit $y \to 0$ at finite temperature. In this case, the second term in (5.23) plays a predominant role on the temperature dependence of specific heat as will be shown below.

To begin with, the derivative of (3.30) with respect to temperature t is given as

$$[A'(y,t) - c]\frac{\mathrm{d}y(t)}{\mathrm{d}t} + \frac{\partial A(y,t)}{\partial t} = 0. \tag{5.31}$$

If we note (3.50) for $y_1(t)$ in Chap. 3, (5.31) is also written in the form

$$\frac{\mathrm{d}y(t)}{\mathrm{d}t} = \frac{1}{c - A'(y,t)}\frac{\partial A(y,t)}{\partial t} = \frac{y_1(t)}{c y_1(0)}\frac{\partial A(y,t)}{\partial t}. \tag{5.32}$$

The second term in (5.23) can be therefore given in the form

$$\frac{\partial A(y,t)}{\partial t}\frac{\mathrm{d}y(t)}{\mathrm{d}t} = c\frac{y_1(0)}{y_1(t)}\left[\frac{\mathrm{d}y(t)}{\mathrm{d}t}\right]^2 \simeq \frac{\pi t_c}{8\sqrt{y(t)}}\left[\frac{\mathrm{d}y(t)}{\mathrm{d}t}\right]^2,$$

with using $y_1(t) \propto \sqrt{y(t)}$ in (3.51). Substitution of the dependence of $y(t)$, proportional to $(t - t_c)^2$, for the above expression finally leads to the following dependence:

$$\frac{\partial A(y,t)}{\partial t}\frac{\mathrm{d}y(t)}{\mathrm{d}t} = \frac{\pi t_c}{4\sqrt{2}}(y_c'')^{3/2}(t - t_c), \quad y_c'' = \left.\frac{\mathrm{d}^2 y(t)}{\mathrm{d}t^2}\right|_{t=t_c} = 2\left[\frac{16A(0,t_c)}{3\pi t_c^2}\right]^2,$$

where (3.38) is used to evaluate the second derivative y_c''. The temperature dependence of the specific heat is thus given by

$$\frac{1}{N_0 t} C_m \simeq \frac{1}{2}\log(1/t_c) - \frac{9\pi t_c}{4\sqrt{2}}(y_c'')^{3/2}(t - t_c). \tag{5.33}$$

It increases proportional to $(T_c - T)$ with decreasing temperature toward T_c. For $t_c \ll 1$, since $A(0, t_c) \propto t_c^{4/3}$ is satisfied, the above $(y_c'')^{3/2}$ is proportional to $1/t_c^2$. Then, $[C_m(T) - C_m(T_c)] \propto (T_c - T)/T_0$ is satisfied with a numerical proportional constant.

5.3.3 Temperature Dependence of the Entropy and the Specific Heat in the Ordered Phase

Temperature dependence of the entropy and the specific heat in the magnetically ordered phase is treated in this section. As with the paramagnetic phase, they are given by differentiating the free energy in (5.2) with respect to temperature. Unlike the paramagnetic phase, the correction ΔF_1 of the free energy is necessary.

Temperature Dependence of the Entropy The entropy is derived from the partial temperature derivative of the free energy as given by

$$\begin{aligned} S_m(\sigma, t) &= S_{m0}(\sigma, t) + \Delta S_m(\sigma, t) \\ \frac{1}{N_0} S_{m0}(\sigma, t) &= -6 \int_{x_c}^{1} \mathrm{d}x x^2 [\Phi(u) - u\Phi'(u)] - 3 \int_0^1 \mathrm{d}x x^2 [\Phi(u_z) - u\Phi'(u_z)] \\ u &= x(y + x^2)/t, \quad u_z = x(y_z + x^2)/t. \end{aligned} \tag{5.34}$$

It consists of two contributions, S_{m0} corresponding to (5.21) in the paramagnetic phase and $\Delta S_m(\sigma, t)$ resulting from the t-dependence of $\Delta y_z(\sigma, t)$ and $\Delta F_1(\sigma, t)$. The effect of spin waves is neglected for simplicity. In the same way as (5.4) for the σ derivative of the free energy, the second term is evaluated by the partial t-derivative of $\Delta y_z(\sigma, t)$ and $\Delta F_1(\sigma, t)$ as given below.

$$\begin{aligned} T_0 \Delta S_m &= -N_0 T_A \left[\langle (S_i^z)^2 \rangle (y_z, t) - \frac{1}{3} \langle S_i^2 \rangle_{\text{tot}} \right] \frac{\partial \Delta y_z}{\partial t} - \frac{\partial \Delta F_1}{\partial t} \\ &= -N_0 T_A \lambda(\sigma, t) \frac{\partial \Delta y_z}{\partial t} - \frac{\partial \Delta F_1}{\partial t}. \end{aligned} \tag{5.35}$$

In the region of weak external magnetic field, the correction ΔF_1 in (5.16) can be approximated by

$$\Delta F_1(\sigma, t) \simeq -N_0 T_A \lambda(\sigma_0, t) \Delta y_z(\sigma, t). \tag{5.36}$$

Substitution of (5.36) for ΔF_1 in (5.35) gives the entropy correction given by

$$\begin{aligned} \Delta S_m(\sigma, t) &= -N_0 \frac{T_A}{T_0} \lambda(\sigma, t) \frac{\partial \Delta y_z}{\partial t} + N_0 \frac{T_A}{T_0} \frac{\partial}{\partial t} [\lambda(\sigma_0, t) \Delta y_z(\sigma, t)] \\ &= N_0 \frac{T_A}{T_0} \left[\frac{\mathrm{d}\lambda(\sigma_0, t)}{\mathrm{d}t} \Delta y_z(\sigma, t) - \delta\lambda(\sigma, t) \frac{\partial \Delta y_z(\sigma, t)}{\partial t} \right], \end{aligned} \tag{5.37}$$

where $\delta\lambda(\sigma, t) = \lambda(\sigma, t) - \lambda(\sigma_0, t)$. The second term proportional to $\delta\lambda(\sigma, t)$ in the second line is neglected in the absence of external magnetic field, since $\delta\lambda(\sigma, t) = 0$ is satisfied for $\sigma = \sigma_0$. The parameter $\lambda(\sigma_0, t)$ defined in (5.9) and its t-derivative are given by

$$\begin{aligned}
\lambda(\sigma_0, t) &= \frac{2T_0}{T_A}[A(y_{z0}, t) - A(y, t) - cy_{z0}(t)] - 5cy_1(0)\sigma_0^2(t) \\
\frac{T_A}{T_0}\frac{\mathrm{d}\lambda(\sigma_0, t)}{\mathrm{d}t} &= 2[A'(y_{z0}, t) - c]\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t} + 2\frac{\partial A(y_{z0}, t)}{\partial t} \\
&\quad -5cy_1(0)\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t} - 2\frac{\partial A(0, t)}{\partial t}
\end{aligned} \tag{5.38}$$

With the use of the TAC condition, the above t-derivative can be written in two different forms. Notice the t-derivative of the condition (3.3) is given by

$$\frac{\partial A(y_{z0}, t)}{\partial t} + [A'(y_{z0}, t) - c]\ \frac{\mathrm{d}y_{z0}}{\mathrm{d}t} + 2\frac{\partial A(0, t)}{\partial t} + 5cy_1(0)\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t} = 0. \tag{5.39}$$

Then $\mathrm{d}\lambda/\mathrm{d}t$ in (5.38) is written in the form

$$\frac{T_A}{T_0}\frac{\mathrm{d}\lambda(\sigma_0, t)}{\mathrm{d}t} = \begin{cases} -6\dfrac{\partial A(0, t)}{\partial t} - 15cy_1(0)\dfrac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}, & \text{(I)} \\ 3\dfrac{\partial A(y_{z0}, t)}{\partial t} + 3[A'(y_{z0}, t) - c]\dfrac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}, & \text{(II)} \end{cases} \tag{5.40}$$

depending on either the terms related to $y_{z0}(t)$ or $\sigma_0^2(t)$ are eliminated. The entropy correction is also expressed in two alternative forms:

$$\frac{1}{N_0}\Delta S_m(t) = \begin{cases} -3y_{z0}(t)\left[2\dfrac{\partial A(0, t)}{\partial t} + 5cy_1(0)\dfrac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}\right], & \text{(I)} \\ 3y_{z0}(t)\left\{\dfrac{\partial A(y_{z0}, t)}{\partial t} + [A'(y_{z0}, t) - c]\dfrac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}\right\}. & \text{(II)} \end{cases} \tag{5.41}$$

Temperature Dependence of the Specific Heat In the ordered phase, the specific heat is given by the sum of the temperature derivatives of S_{m0} and ΔS_m.

$$\begin{aligned}
C_m(t) &= C_{m0}(t) + \Delta C_m(t) \\
\frac{1}{N_0 t}C_{m0}(t) &= 6I_c(0, t) + 3I(y_{z0}, t), \quad I_c(y, t) = -\frac{1}{t}\int_{x_c}^1 \mathrm{d}x x^2 u^2 \Phi''(u) \\
\frac{1}{N_0 t}\Delta C_m(t) &= -3\frac{\partial A(y_{z0}, t)}{\partial t}\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t} + \frac{1}{N_0}\frac{\mathrm{d}\Delta S_m(t)}{\mathrm{d}t} \\
&= 3y_{z0}(t)\left[\frac{\partial^2 A(y_{z0}, t)}{\partial t^2} + A''(y_{z0}, t)\left(\frac{\mathrm{d}y_{z0}}{\mathrm{d}t}\right)^2\right. \\
&\qquad \left. +2\frac{\partial A'(y_{z0}, t)}{\partial t}\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}\right] \\
&\quad + 3[A'(y_{z0}, t) - c]\left[\left(\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}\right)^2 + y_{z0}(t)\frac{\mathrm{d}^2 y_{z0}(t)}{\mathrm{d}t^2}\right]
\end{aligned} \tag{5.42}$$

The first term $C_{m0}(t)$ results from the direct t derivative of $S_{m0}(t)$. The function $I(y_{z0}, t)$ in the second line is already defined in (5.26). The correction $\Delta C_m(t)$ consists of the sum of two contributions, i.e., the implicit temperature dependence through that of $y_{z0}(t)$ included in $S_{m0}(t)$ and the t derivative of the correction $\Delta S_m(t)$. It is derived by using the expression (II) in (5.41). If (I) is used, $\Delta C_m(t)$ is written in the form

$$\frac{1}{N_0 t}\Delta C_m(t) = -3\left[\left(2\frac{\partial A(0,t)}{\partial t} + \frac{\partial A(y_{z0},t)}{\partial t}\right)\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t} + 2y_{z0}(t)\frac{\partial^2 A(0,t)}{\partial t^2} + 5cy_1(0)\frac{\mathrm{d}}{\mathrm{d}t}\left(y_{z0}(t)\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}\right)\right] \quad (5.43)$$

The temperature dependence of $C_m(t)$ shows the following two characteristic features derived from the presence of $\Delta C_m(t)$.

- There exits another new enhancement in the T-linear coefficient of the specific heat in the limit of low temperature.
- A sharp peak appears at the critical temperature.

Numerically calculated results of (5.42) are shown in Fig. 5.2.

Dependence in the Limit of Low Temperature In the limit where $t^{3/2} \ll y_{z0}(t)$ is satisfied, $I(y_{z0}, t)$ is given by (5.29). The transverse contribution $I_c(0, t)$ is of the same size because of the presence of lower cut-off of the integral x_c. The T-linear coefficient of C_{m0} shows the logarithmic behavior:

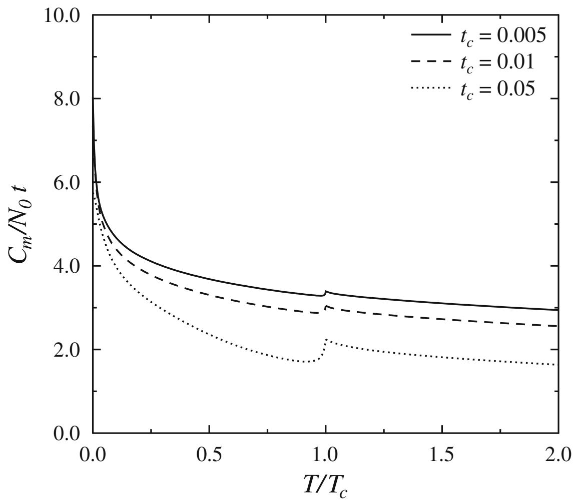

Fig. 5.2 Numerically calculated examples of the temperature dependence of the specific heat for $t_c = 0.005, 0.01, 0.05$ from the top

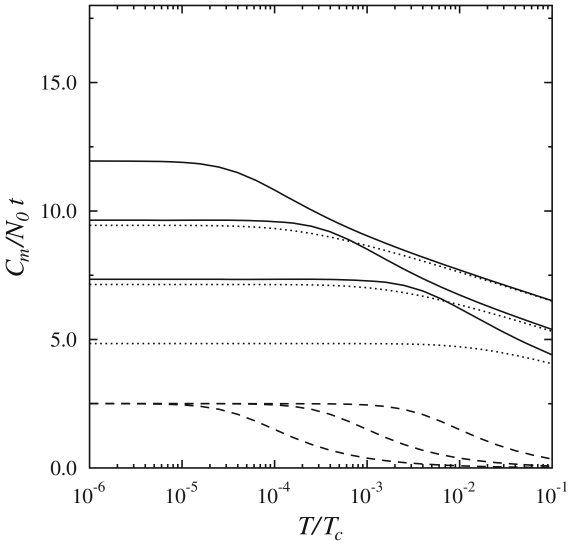

Fig. 5.3 Temperature dependence of the specific heat in the low-T limit for $t_c = 0.0001, 0.001, 0.01$ from the top in logarithmic temperature scale. The dependence of C_m, C_{m0}, and ΔC_m is denoted by *solid*, *dotted*, and *dashed curves*, respectively

$$\frac{1}{N_0 t} C_{m0} \simeq \frac{1}{4}\left[2\log(1/x_c^2) + \log(1/y_{z0})\right] \simeq \frac{3}{2}\log[1/\sigma_0(0)], \tag{5.44}$$

as the spontaneous moment tends to disappear, $\sigma_0(t) \to 0$. Another contribution of considerable size also results from ΔC_m, as given by

$$\begin{aligned}\frac{1}{N_0 t}\Delta C_m &\simeq -3cy_{z0}(t)\frac{\mathrm{d}^2 y_{z0}(t)}{\mathrm{d}t^2} + 3y_{z0}\frac{\partial^2 A(y_{z0}, t)}{\partial t^2} \\ &= \frac{1}{6}\left[\left(\frac{\pi}{4}\right)^4 + 2\left(\frac{\pi}{4}\right)^2 + 4\right].\end{aligned} \tag{5.45}$$

Though it is not divergent in the limit $\sigma_0(t) \to 0$, its size is nonnegligible in the limit of low temperature. The temperature dependence of these two contributions is shown in Fig. 5.3.

Dependence Around the Critical Temperature Around the critical temperature, the opposite condition $y \ll t^{3/2}$ is satisfied for $I(y_{z0}, t)$ in (5.42). The first term $C_{m0}(t)$ is then given by

$$\frac{1}{N_0 t} C_{m0}(t) \simeq 9I(0, t) \simeq \frac{1}{2}\log(1/t). \tag{5.46}$$

As with the case of the paramagnetic phase, the correction ΔC_m shows the $(t - t_c)$-linear dependence from the following dominant contributions:

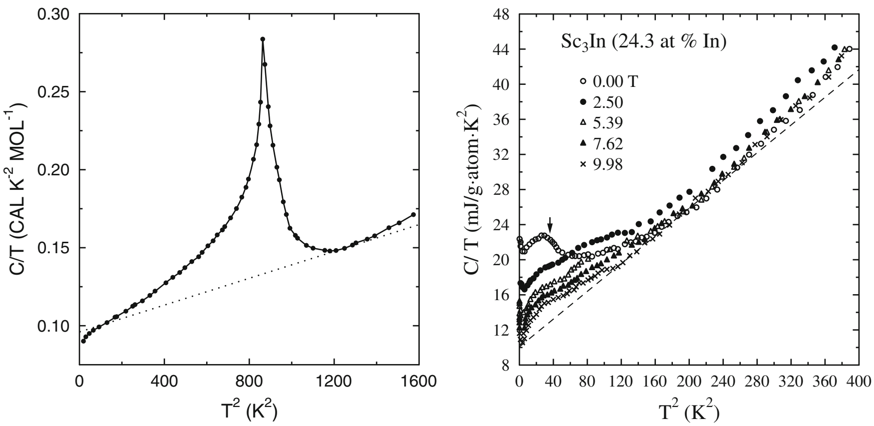

Fig. 5.4 Temperature dependence of the specific heat of MnSi by Fawcett et al. (*left*) and Sc_3In by Ikeda and Gschneidner (*right*)

$$\frac{1}{N_0 t}\Delta C_m(t) \simeq 3\left(\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}\right)^2 \left[y_{z0}(t)A''(y_{z0},t) + 3A'(y_{z0},t)\right] + 3y_{z0}(t)A'(y_{z0},t)\frac{\mathrm{d}^2 y_{z0}(t)}{\mathrm{d}t^2}. \quad (5.47)$$

With the use of the dependence $y_{z0}(t) \propto (t_c - t)^2$ and the critical $\sqrt{y_{z0}}$ dependence of the thermal amplitude $A(y_{z0}, t)$, the above right hand side is estimated as follows:

$$\left(\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}\right)^2 [y_{z0}(t)A''(y_{z0},t) + A'(y_{z0},t)] \simeq (y''_{zc})^2\left(\frac{\pi t}{16\sqrt{y_{z0}}} - \frac{\pi t}{8\sqrt{y_{z0}}}\right)(t_c - t)^2 = -\frac{\pi t}{\sqrt{2}}(y''_{zc})^{3/2}(t_c - t), \quad (5.48)$$

$$y_{z0}(t)A'(y_{z0},t)\frac{\mathrm{d}^2 y_{z0}(t)}{\mathrm{d}t^2} \simeq -\frac{\pi t y''_{zc}}{2}\sqrt{y_{z0}} \simeq -\frac{\pi t}{\sqrt{2}}(y''_{zc})^{3/2}(t_c - t).$$

The second derivative $\mathrm{d}^2 y_{z0}(t)/\mathrm{d}t^2$ at $t = t_c$ is denoted by y''_{zc} in the above. The correction ΔC_m also shows the $(t - t_c)$ linear dependence but with positive slope in this case.

$$\frac{1}{N_0 t}\Delta C_m(t) \simeq 3\sqrt{2}\pi (y''_{zc})^{3/2}(t - t_c). \quad (5.49)$$

If we combine (5.49) with (5.33) in the paramagnetic phase, the slope of the T dependence of ΔC_m shows a discontinuous change from positive to negative, resulting in the peak at the critical point. The behavior is observed in numerically calculated results in Fig. 5.2.

As examples of observed temperature dependence of specific heat, the results of the measurements on MnSi by Fawcett et al. [3] and Sc_3In by Ikeda and Gschneidner

[4] are shown in Fig. 5.4. In this figure, values of C/T are plotted against T^2. The dotted (left) and dashed (right) lines plotted by them are regarded as the contribution from lattice vibrations. A clear and definite peak is observed for MnSi with fairly large spontaneous magnetic moment ($t_c \sim 0.13$). Whereas for Sc_3In with tiny spontaneous moment ($t_c \sim 0.01$), the peak is not so clear. The tendency is consistent with the theoretical prediction that the larger the value of t_c, the larger and distinct peak appears as shown in Fig. 5.2 numerically.

5.4 Specific Heat Under the External Magnetic Field

We next show in this section how the temperature dependence of the magnetic entropy and the specific heat is determined in the presence of the external magnetic field. We will particularly deal with the following two subjects.

1. The σ dependence of the entropy and the specific heat under constant temperature.
2. Their temperature dependence under constant static external magnetic field.

As for the first one, we need to confirm that the Maxwell relation is satisfied for the field-induced change of the entropy. Concerning the second one, we need to know how to evaluate the temperature dependence under constant magnetic field in the treatment where σ is regarded as independent variable.

For convenience of our later explanation, note that the σ dependence of $y(\sigma, t)$ and $y_z(\sigma, t)$, and their variations, $\delta y(\sigma, t)$ and $\delta y_z(\sigma, t)$, induced by the external magnetic field are given by

$$\begin{aligned} y(\sigma, t) &= y_0(t) + y_1(t)\sigma^2, \quad y_z(\sigma, t) = y_0(t) + 3y_1(t)\sigma^2, \\ \delta y(\sigma, t) &= y(\sigma, t) - y(0, t) = y_1(t)\sigma^2, \\ \delta y_z(\sigma, t) &= y_z(\sigma, t) - y_z(0, t) = 3y_1(t)\sigma^2, \end{aligned} \tag{5.50}$$

in the paramagnetic phase, and by

$$\begin{aligned} y(\sigma, t) &= y_1(t)[\sigma^2 - \sigma_0^2(t)], \quad y_z(\sigma, t) = 2y_1(t)\sigma_0^2(t) + 3y(\sigma, t), \\ \delta y(\sigma, t) &= y(\sigma, t) - y(\sigma_0, t) = y(\sigma, t), \\ \delta y_z(\sigma, t) &= y_z(\sigma, t) - y_z(\sigma_0, t) = 3y(\sigma, t), \end{aligned} \tag{5.51}$$

in the magnetically ordered phase ($T < T_c$).

5.4.1 Maxwell Relation

For the free energy $F(M, T)$ with independent variables M and T, the total differential $\mathrm{d}F$ is written in the form

$$\begin{aligned} \mathrm{d}F(M,T) &= -S_m(M,T)\mathrm{d}T + H(M,T)\mathrm{d}M, \\ -S_m(M,T) = \frac{\partial F(M,T)}{\partial T}, &\quad H(M,T) = \frac{\partial F(M,T)}{\partial M}. \end{aligned} \tag{5.52}$$

The entropy S_m and the magnetic field H are derived by the first derivatives of F with respect to T and M, respectively. Their further derivatives with respect to M and T given by

$$-\frac{\partial S_m}{\partial M} = \frac{\partial^2 F}{\partial M \partial T}, \quad \frac{\partial H}{\partial T} = \frac{\partial^2 F}{\partial T \partial M}, \tag{5.53}$$

agree with each other. It means that the following Maxwell relation is satisfied.

$$\begin{aligned} \frac{\partial S_m}{\partial M} &= -\frac{\partial H}{\partial T} = -M\frac{\partial}{\partial T}\left(\frac{H}{M}\right)\Big|_M, \\ \frac{1}{N_0}\frac{\partial S_m(\sigma,t)}{\partial \sigma} &= -\frac{2T_A\sigma}{T_0}\frac{\partial y(\sigma,t)}{\partial t}. \end{aligned} \tag{5.54}$$

The second line is the dimensionless form of the first equation in terms of dimensionless parameters, $\sigma = M/2N\mu_\mathrm{B}$, $h = 2\mu_\mathrm{B}H$, $y(\sigma,t) = h/2T_A\sigma$, and $t = T/T_0$. According to (5.50) and (5.51), $\partial y(\sigma,t)/\partial t$ is written in the form

$$\frac{\partial y(\sigma,t)}{\partial t} \simeq \begin{cases} \dfrac{\mathrm{d}y_0(t)}{\mathrm{d}t}, & \text{for } \sigma \simeq 0 \\ -y_1(t)\dfrac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}, & \text{for } \sigma \simeq \sigma_0(t), \end{cases} \tag{5.55}$$

in the paramagnetic (above) and ordered (below) phases. In what follows, we will show the entropy in (5.34) actually satisfies the relation.

In the Paramagnetic Phase With using (5.10), (5.11), and (5.12) for $\Delta y_z(\sigma,t)$, $\lambda(\sigma,t)$, and $\Delta F_1(\sigma,t)$, respectively, the σ dependence of ΔS_m is given by

$$\begin{aligned} T_0\Delta S_m(\sigma,t) &= -N_0T_A\lambda(\sigma,t)\frac{\partial \Delta y_z(\sigma,t)}{\partial t} - \frac{\partial \Delta F_1}{\partial t} \\ &= \frac{3}{5}N_0T_A\frac{\mathrm{d}y_1(t)}{\mathrm{d}t}\sigma^4 + \cdots. \end{aligned} \tag{5.56}$$

This term of higher order correction can be neglected in this case. On the other hand for $S_{m0}(\sigma,t)$, effects of magnetic field on $u(\sigma,t)$ and $u_z(\sigma,t)$ are given by $\delta u(\sigma,t) = x\delta y(\sigma,t)/t$ and $\delta u_z(\sigma,t) = x\delta y_z(\sigma,t)/t$ in terms of variations δy and δy_z. Substituting them into (5.34), the entropy change is therefore represented in the form

$$\begin{aligned}\frac{1}{N_0}\delta S_m(\sigma,t) &= \frac{3}{t}\int_0^1 \mathrm{d}x x^3 u\Phi''(u)[2\delta y(\sigma,t)+\delta y_z(\sigma,t)] \\ &= -3\frac{\partial A(y_0,t)}{\partial t}[2\delta y(\sigma,t)+\delta y_z(\sigma,t)] \\ &= -15y_1(t)\frac{\partial A(y_0,t)}{\partial t}\sigma^2,\end{aligned} \tag{5.57}$$

by using the relation $2\delta y(\sigma,t)+\delta y_z(\sigma,t) = 5y_1(t)\sigma^2$ in the last line. If we notice the relation (5.32), then (5.57) is finally written as

$$\frac{1}{N_0}\frac{\partial \delta S_m}{\partial \sigma} = -\frac{2T_A\sigma}{T_0}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}. \tag{5.58}$$

It implies that the Maxwell relation in (5.54) is satisfied for the entropy in the paramagnetic phase.

In the Ordered Phase In this case, the entropy change induced by the applied magnetic field is given by

$$\begin{aligned}\delta S_m(\sigma,t) &= \frac{3N_0}{t}\int_0^1 \mathrm{d}x\, x^3[2u\Phi''(u)\delta y(\sigma,t)+u_z\Phi''(u_z)\delta y_z(\sigma,t)] \\ &\quad + \delta\Delta S_m(\sigma,t), \quad u=\frac{x^3}{t}, \quad u_z=\frac{x}{t}(y_{z0}+x^2),\end{aligned} \tag{5.59}$$

where deviations $\delta y(\sigma,t)$ and $\delta y_z(\sigma,t)$ are defined in (5.51). Because $\sigma=\sigma_0$ and $y(\sigma_0,0)=0$ are satisfied in the absence of the field, $\delta y(\sigma,0)=y(\sigma,0)$ is satisfied. If we denote the first term by $\delta S_{m0}(\sigma,t)$, (5.59) is also written in the form

$$\frac{1}{N_0}\delta S_{m0}(\sigma,t) = -3\left[2\frac{\partial A(0,t)}{\partial t}\delta y + \frac{\partial A(y_{z0},t)}{\partial t}\delta y_z\right]. \tag{5.60}$$

To evaluate the field effect on $\Delta S_m(\sigma,t)$, let us substitute (5.40) for $\mathrm{d}\lambda(\sigma_0,t)/\mathrm{d}t$ in (5.37). Then the correction is given by

$$\begin{aligned}\frac{1}{N_0}\delta\Delta S_m(\sigma,t) &= 3\left\{\frac{\partial A(y_{z0},t)}{\partial t} + [A'(y_{z0},t)-c]\frac{\mathrm{d}y_{z0}}{\mathrm{d}t}\right\}\delta y_z \\ &\quad + \left\{6\frac{\partial A(0,t)}{\partial t} + 15cy_1(0)\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}\right\}\delta y \\ &\quad - \frac{T_A}{T_0}\frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}\delta\lambda(\sigma,t) \\ &= 3\left[\frac{\partial A(y_{z0},t)}{\partial t}\delta y_z + 2\frac{\partial A(0,t)}{\partial t}\delta y\right] + \frac{T_A}{T_0}\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}\delta y,\end{aligned} \tag{5.61}$$

by using the definition $\Delta y_z \equiv \delta y_z - \delta y$ in (5.37). In the above derivation, we employ the expression (II) for $\mathrm{d}\lambda(\sigma_0,t)/\mathrm{d}t$ in (5.40) in the first line, and (I) in the second line. The following relation for $\delta\lambda(\sigma, t)$ in the third line is also used in the above derivation:

$$\frac{T_A}{T_0}\delta\lambda(\sigma, t) = 2\left\{[A'(y_{z0}, t) - c]\delta y_z - [A'(0, t) - c]\delta y\right\} - \frac{T_A}{3T_0}\delta\sigma^2$$
$$= 3[A'(y_{z0}, t) - c]\delta y_z, \tag{5.62}$$

which is derived from the definition of $\lambda(\sigma, t)$ in (5.9) and the deviation of the condition of TAC, given by

$$2[A'(0, t) - c]\delta y + [A'(y_{z0}, t) - c]\delta y_z + \frac{T_A}{3T_0}\delta\sigma^2 = 0. \tag{5.63}$$

By putting (5.60) and (5.61) into (5.59), the following entropy change is finally obtained:

$$\frac{1}{N_0}\delta S_m(\sigma, t) = \frac{T_A}{T_0}\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}\delta y(\sigma, t). \tag{5.64}$$

Partial derivative of the above both sides with respect to σ gives the Maxwell relation:

$$\frac{1}{N_0}\frac{\partial S_m(\sigma, t)}{\partial\sigma} = \frac{2T_A\sigma}{T_0}y_1(t)\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t}, \tag{5.65}$$

by using $\partial y(\sigma, t)/\partial\sigma = 2y_1(t)\sigma$. As the last term, the right hand side in (5.64) is involved in the entropy correction $\delta\Delta S_m(\sigma, t)$ in (5.61). This clearly means that we need to include this term to satisfy the Maxwell relation in the ordered phase.

5.4.2 Temperature Derivatives in the Static External Magnetic Field

To evaluate the temperature dependence of the specific heat in a constant external magnetic field h, we need temperature derivatives of $y(\sigma, t)$ and $y_z(\sigma, t)$ in this condition. These values are related with derivatives in a constant magnetization σ, as shown below.

To begin with, the derivative of the definition, $y(\sigma, t) = h/2T_A\sigma$, with respect to temperature in a constant σ gives the relation:

$$\left.\frac{\partial y(\sigma, t)}{\partial t}\right|_h = -\frac{h}{2T_A\sigma^2}\left.\frac{\partial\sigma}{\partial t}\right|_h = -\frac{y(\sigma, t)}{\sigma}\left.\frac{\partial\sigma}{\partial t}\right|_h. \tag{5.66}$$

It is also rewritten in the form

$$\left.\frac{\partial y(\sigma, t)}{\partial t}\right|_h = \frac{\partial y(\sigma, t)}{\partial t} + \frac{\partial y(\sigma, t)}{\partial\sigma}\left.\frac{\partial\sigma}{\partial t}\right|_h. \tag{5.67}$$

In the condition of constant h, $\sigma(h,t)$ is regarded as a function of h and t. By equating these relations, (5.66) and (5.67), the following relation between $\partial\sigma/\partial t|_h$ and $\partial y(\sigma,t)/\partial t$ is derived:

$$\left[\frac{y(\sigma,t)}{\sigma}+\frac{\partial y(\sigma,t)}{\partial\sigma}\right]\frac{\partial\sigma}{\partial t}\bigg|_h=\frac{y_z(\sigma,t)}{\sigma}\frac{\partial\sigma}{\partial t}\bigg|_h=-\frac{\partial y(\sigma,t)}{\partial t},$$
$$\therefore\ \frac{\partial\sigma}{\partial t}\bigg|_h=-\frac{\sigma}{y_z(\sigma,t)}\frac{\partial y(\sigma,t)}{\partial t}. \tag{5.68}$$

Then, the temperature derivative of any function $f(\sigma,t)$ in a constant h is generally written as follows:

$$\begin{aligned}\frac{\partial f(\sigma,t)}{\partial t}\bigg|_h&=\frac{\partial f(\sigma,t)}{\partial t}+\frac{\partial f(\sigma,t)}{\partial\sigma}\frac{\partial\sigma}{\partial t}\bigg|_h\\&=\frac{\partial f(\sigma,t)}{\partial t}-\frac{\sigma}{y_z(\sigma,t)}\frac{\partial y(\sigma,t)}{\partial t}\frac{\partial f(\sigma,t)}{\partial\sigma}.\end{aligned} \tag{5.69}$$

Substituting $y(\sigma,t)$ or $y_z(\sigma,t)$ for $f(\sigma,t)$, as special cases we obtain the relations:

$$\begin{aligned}\frac{\partial y(\sigma,t)}{\partial t}\bigg|_h&=\left[1-\frac{\sigma}{y_z(\sigma,t)}\frac{\partial y(\sigma,t)}{\partial\sigma}\right]\frac{\partial y(\sigma,t)}{\partial t}=\frac{y(\sigma,t)}{y_z(\sigma,t)}\frac{\partial y(\sigma,t)}{\partial t},\\\frac{\partial y_z(\sigma,t)}{\partial t}\bigg|_h&=\frac{\partial y_z(\sigma,t)}{\partial t}-\frac{\sigma}{y_z(\sigma,t)}\frac{\partial y(\sigma,t)}{\partial t}\frac{\partial y_z(\sigma,t)}{\partial\sigma}.\end{aligned} \tag{5.70}$$

By using these relations, various temperature derivatives in a constant h can be written in terms of derivatives with respect to σ.

5.4.3 Entropy and Specific Heat in the Paramagnetic Phase

Let us now show the temperature and the external dependence of the entropy and the specific heat.

Effect of Magnetic Field in the Paramagnetic Phase As we have already shown, the entropy change $\delta S_m(\sigma,t)$, induced by the external magnetic field h, is given by (5.57). Actually to evaluate the change in a constant h, we are required to find the value of σ as a function of h.

The magnetic field effect on the specific heat is also given by the temperature derivative of (5.57) under the constant h condition:

$$\begin{aligned}\frac{1}{N_0t}\delta C_m(\sigma,t)&=\frac{1}{N_0}\frac{\partial\delta S_m(\sigma,t)}{\partial t}\\&=-3\frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{\partial A(y_0,t)}{\partial t}\right](2\delta y+\delta y_z)-3\frac{\partial A(y_0,t)}{\partial t}\left[2\frac{\partial\delta y}{\partial t}\bigg|_h+\frac{\partial\delta y_z}{\partial t}\bigg|_h\right]\end{aligned}$$

$$= -3\frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{\partial A(y_0,t)}{\partial t}\right](2\delta y + \delta y_z)$$
$$-3\frac{\partial A(y_0,t)}{\partial t}\left[\left(2\frac{\partial \delta y}{\partial t} + \frac{\partial \delta y_z}{\partial t}\right) - \left(2\frac{\partial \delta y}{\partial \sigma} + \frac{\partial \delta y_z}{\partial \sigma}\right)\frac{\sigma}{y_z}\frac{\partial y}{\partial t}\right], \tag{5.71}$$

by using the relation (5.69) for $f(\sigma,t) = 2\delta y(\sigma,t) + \delta y_z(\sigma,t)$. In what follows, temperature and external field dependence of the entropy and the specific heat are examined in more detail in some particular temperature regions.

Exchange Enhanced Paramagnets at Low Temperatures For paramagnets in the vicinity of the ferromagnetic instability point, the inverse of the magnetic susceptibility (see Sect. 3.3.2) is given by

$$y_0(t) = y_0(0) + \frac{1}{24cy_0(0)}t^2 + \cdots . \tag{5.72}$$

Then, the following result is derived because of the relation between $y_0(t)$ and $\partial A(y_0,t)/\partial t$ in (5.32).

$$y_1(t)\frac{\partial A(y_0,t)}{\partial t} = cy_1(0)\frac{\mathrm{d}y_0(t)}{\mathrm{d}t} \simeq \frac{y_1(0)}{12y_0(0)}t. \tag{5.73}$$

By substituting (5.73) into (5.57), the entropy change is finally given by

$$\frac{1}{N_0}\delta S_m(\sigma,t) = -\frac{5y_1(0)}{4y_0(0)}t\sigma^2 + \cdots . \tag{5.74}$$

In this range of temperature, the entropy $S_m(0,t)$ in the absence of magnetic field shows the same temperature dependence as (5.30) for the specific heat. The sum of these contributions is written as follows:

$$S_m(\sigma,t) = \frac{N_0}{4}t\left[3\log\left(\frac{1}{y_0(0)}\right) - 5\frac{y_1(0)}{y_0(0)}\sigma^2\right] + \cdots . \tag{5.75}$$

It can be also expressed as the T-linear coefficient of the specific heat:

$$\gamma_m(\sigma) = \lim_{t\to 0}\frac{C_m(\sigma,t)}{T} = \frac{1}{T_0}\lim_{t\to 0}\frac{S_m(\sigma,t)}{t} = \frac{3N_0}{4T_0}\left[\log\frac{1}{y_0(0)} - \frac{5y_1(0)}{3y_0(0)}\sigma^2\right], \tag{5.76}$$

or in the form of the relative change of its magnitude.

$$\frac{\Delta C_m(\sigma,0)}{C_m(0,0)} = \frac{C_m(\sigma,0) - C_m(0,0)}{C_m(0,0)} = -\frac{5y_1(0)}{3y_0(0)\log[1/y_0(0)]}\sigma^2$$
$$= -\frac{5}{3}\frac{y_1(0)/y_0(0)}{\log(1/y_0(0))}\left[\frac{h}{T_A y_0(0)}\right]^2 = -\frac{5}{3}\frac{(\chi_0/N_0)^3 F_1}{\log(2T_A\chi_0/N_0)}h^2. \tag{5.77}$$

It is equivalent with the following result by Béal-Monod et al. [5].

$$\frac{\Delta C_m(\sigma,0)}{C_m(0,0)} = -0.1\frac{S}{\log S}\left(\frac{H}{T_{\rm sf}}\right)^2, \quad (S=(1-\alpha)^{-1},\ 1/T_{\rm sf} \propto S\chi^0_{\rm Pauli}),$$

where S and $H/T_{\rm sf}$ correspond to $1/y_0$ and $h/T_A y_0$, respectively, and $\alpha = I\rho$ which appears in the Stoner condition.

In the Region at High Temperatures Except for the region around the critical temperature, the field effect on the inverse of the magnetic susceptibility in (5.71) is well approximated by $\delta y(\sigma,t) + \delta y_z(\sigma,t) \simeq 5y_1(t)\sigma^2$. If we assume that the temperature dependence of the coefficient $y_1(t)$ of the σ^4 term of the free energy is neglected, the following approximation is satisfied.

$$\frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{\partial A(y_0,t)}{\partial t}\right] \simeq \frac{T_A}{15T_0 y_1(t)}\frac{\mathrm{d}^2 y_0(t)}{\mathrm{d}t^2}$$

Because the higher order effect of magnetic field is neglected in this case,

$$\frac{1}{y_z(\sigma,t)}\frac{\partial y(\sigma,t)}{\partial t} \simeq \frac{1}{y_0(t)}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t} \tag{5.78}$$

is justified in the last line. We can therefore obtain the following approximation for (5.71).

$$\begin{aligned}\frac{\delta C_m}{N_0 t} &\simeq -\frac{T_A}{T_0}\frac{\mathrm{d}^2 y_0(t)}{\mathrm{d}t^2}\sigma^2 - \frac{T_A}{5T_0 y_1(t)}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\left(5\frac{\mathrm{d}y_1(t)}{\mathrm{d}t} - 10\frac{y_1(t)}{y_0(t)}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\right)\sigma^2 \\ &\simeq -\frac{T_A}{T_0}\sigma^2\left[\frac{\mathrm{d}^2 y_0(t)}{\mathrm{d}t^2} - \frac{2}{y_0(t)}\left(\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\right)^2\right] = \frac{T_A}{4T_0}\frac{\mathrm{d}^2 y_0^{-1}(t)}{\mathrm{d}t^2}\frac{h^2}{T_A^2},\end{aligned} \tag{5.79}$$

where $\sigma \simeq h/[2T_A y_0(t)]$ is assumed in the last line.

In this region, the field effect gives the positive deviation δC_m proportional to h^2. In the range where the Curie-Weiss law behavior is observed, its coefficient shows the dependence, $t/(t-t_c)^3$. The external field generally suppresses the entropy, and its deviation δS_m is negative. However, its temperature dependence shows the positive slope, giving the positive δC_m.

Around the Critical Temperature In this case, the entropy change $\delta S_m(\sigma, t_c)$ is also evaluated by using the second line of (5.57) with $y_0 = 0$, i.e.,

$$\frac{1}{N_0}\delta S_m(\sigma,t_c) = -3\left.\frac{\partial A(0,t)}{\partial t}\right|_{t=t_c}[2\delta y(\sigma,t_c) + \delta y_z(\sigma,t_c)]. \tag{5.80}$$

Since $\delta y(\sigma,t_c) = y(\sigma,t_c)$ and $\delta y_z(\sigma,t_c) = y_z(\sigma,t_c)$ are satisfied, substituting the critical isotherm, $\delta y(\sigma,t_c) = y_c\sigma^4$ and $\delta y_z(\sigma,t_c) = 5y_c\sigma^4$, gives the following σ

dependence of the entropy:

$$\frac{1}{N_0}\delta S_m(\sigma, t_c) = -21y_c \frac{\partial A(0,t)}{\partial t}\bigg|_{t=t_c} \sigma^4 = -\frac{28A(0,t_c)}{t_c} y_c \sigma^4, \tag{5.81}$$

by using the relation, $\partial A(0,t)/\partial t = 4A(0,t)/3$, derived from the t dependence, $A(0,t) \propto t^{4/3}$, in (2.86).

The specific heat is evaluated as the critical limit of the expression (5.71) in the paramagnetic phase. We then need to evaluate the σ dependence of derivatives $\partial y(\sigma,t)/\partial t$ and $\partial y_z(\sigma,t)/\partial t$. They are determined by solving the equation:

$$2\frac{\partial A(y,t)}{\partial t} + \frac{\partial A(y_z,t)}{\partial t} + 2[A'(y,t) - c]\frac{\partial y}{\partial t} + [A'(y_z,t) - c]\frac{\partial y_z}{\partial t} = 0, \tag{5.82}$$

which is derived from the temperature derivative of the TAC condition. Because of the predominant $1/\sqrt{y}$ dependence of $A'(y,t)$, it can be approximated by

$$-\frac{\pi t_c}{4\sqrt{y}}\frac{\partial y}{\partial t} - \frac{\pi t_c}{8\sqrt{y_z}}\frac{\partial y_z}{\partial t} + \frac{4}{t_c}A(0,t_c) \simeq 0, \tag{5.83}$$

where the $\sqrt{y}$ and $\sqrt{y_z}$-linear dependence resulting from the first two terms in (5.83) are discarded in the limit, $y \to 0$ and $y_z \to 0$. From the σ^4-linear behavior of $\delta y(\sigma,t)$ and $\delta y_z(\sigma,t)$, the following results are obtained:

$$\frac{\partial \delta y(\sigma,t)}{\partial t} \propto \sigma^2, \quad \frac{\partial \delta y_z(\sigma,t)}{\partial t} = \frac{\partial \delta y(\sigma,t)}{\partial t} + \sigma\frac{\partial \delta y(\sigma,t)}{\partial \sigma} \simeq 3\frac{\partial \delta y(\sigma,t)}{\partial t}. \tag{5.84}$$

The σ^2-linear coefficient determined by (5.83) is given by

$$\frac{\partial \delta y(\sigma,t)}{\partial t} \simeq \frac{32\sqrt{5y_c}}{(2\sqrt{5}+3)\pi t^2}A(0,t)\sigma^2. \tag{5.85}$$

Substituting these results for $\partial\delta y/\partial t$ and $\partial \delta y_z/\partial t$ into (5.71), the effect of the magnetic field on the specific heat is written as follows:

$$\begin{aligned}\frac{\delta C_m}{N_0 t_c} &= -3\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial A(y_0,t)}{\partial t}\bigg|_{y_0=0}(2y+y_z) + \frac{9}{5}\frac{\partial A(0,t)}{\partial t}\frac{\partial y(\sigma,t)}{\partial t}\bigg|_{t=t_c} \\ &= \frac{384\sqrt{5y_c}}{5(2\sqrt{5}+3)\pi t_c^3}A^2(0,t_c)\sigma^2 + \frac{56}{3t_c^2}A(0,t_c)y_c\sigma^4 + \cdots \\ &= \frac{8A^3(0,t_c)}{t_c^4}\left[\frac{20}{\pi(2+\sqrt{5})}\right]^2\frac{12(5+2\sqrt{5})}{25(2\sqrt{5}+3)}\left(\frac{\sigma}{\sigma_s}\right)^2 + \cdots,\end{aligned} \tag{5.86}$$

where the coefficient of the first term and the σ derivative of the last term in (5.71) are estimated by

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial A(y_0,t)}{\partial t}\bigg|_{y_0=0} = \frac{\partial A'(y_0,t)}{\partial t}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\bigg|_{y_0=0} + \frac{\partial^2 A(0,t)}{\partial t^2}$$
$$\simeq -\frac{\pi}{8\sqrt{y_0(t)}}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\bigg|_{y_0=0} + \frac{4}{9t^2}A(0,t) = -\frac{8}{9t^2}A(0,t)$$
$$\frac{\sigma}{y_z}\left(2\frac{\partial y}{\partial\sigma} + \frac{\partial y_z}{\partial\sigma}\right) = \frac{4(2y+y_z)}{y_z} = \frac{28}{5}.$$

5.4.4 External Field Effect in the Ordered Phase

In the ordered phase, since $\delta y(\sigma,t) = y(\sigma,t)$ is satisfied in (5.64), the field effect on the entropy is given by

$$\frac{1}{N_0}\delta S_m(\sigma,t) = \frac{T_A}{T_0}y(\sigma,t)\frac{\mathrm{d}\sigma_0^2(t)}{\mathrm{d}t} = 15A(0,t_c)\frac{\mathrm{d}U(t)}{\mathrm{d}t}y(\sigma,t), \tag{5.87}$$

by using the relation (3.12) between the thermal amplitude $A(0,t_c)$ and σ_0^2. The same parameter $U(t) = \sigma_0^2(t)/\sigma_0^2(0)$ defined in (4.21) is also used.

The field-induced change of the specific heat is derived by the derivative of (5.87) with respect to temperature, i.e., as the sum of two contributions:

$$\begin{aligned}
\frac{1}{t}\delta C_m(\sigma,t) &= \frac{\partial\delta S_m(\sigma,t)}{\partial t}\bigg|_h = \frac{1}{t}[\delta C_{m1}(\sigma,t) + \delta C_{m2}(\sigma,t)]\\
\frac{1}{N_0 t}\delta C_{m1}(\sigma,t) &= 15A(0,t_c)y(\sigma,t)\frac{\mathrm{d}^2U(t)}{\mathrm{d}t^2}\\
\frac{1}{N_0 t}\delta C_{m2}(\sigma,t) &= 15A(0,t_c)\frac{\mathrm{d}U(t)}{\mathrm{d}t}\frac{\partial y(\sigma,t)}{\partial t}\bigg|_h\\
&= 15A(0,t_c)\frac{\mathrm{d}U(t)}{\mathrm{d}t}\frac{y(\sigma,t)}{y_z(\sigma,t)}\frac{\partial y(\sigma,t)}{\partial t},
\end{aligned} \tag{5.88}$$

where (5.70) is used as the temperature derivative, $\partial y(\sigma,t)/\partial t|_h$, in a constant h for $\delta C_{m2}(\sigma,t)$.

Field Effect on the Specific Heat at Low Temperatures According to (4.2) and (4.26) in Chap. 4, the σ dependence of the inverse of the magnetic susceptibilities and the temperature dependence of $U(t)$ are given by

$$\begin{aligned}
y(\sigma,t) &= y_1(t)[\sigma^2 - \sigma_0^2(t)] \simeq y_1(0)[\sigma^2 - \sigma_0(0)^2],\\
&= \frac{1}{c}A(0,t_c)\left[\frac{\sigma^2}{\sigma_0(0)^2} - 1\right],\\
y_z(\sigma,t) &= y_{z0}(t) + 3y_1(t)[\sigma^2 - \sigma_0^2(t)],
\end{aligned} \tag{5.89}$$

$$\simeq 2y_{z0}(0) + \frac{3}{c}A(0, t_c)\left[\frac{\sigma^2}{\sigma_0(0)^2} - 1\right],$$

$$U(t) = 1 - \frac{\alpha_0 t^2}{360A^2(0, t_c)} + \cdots, \quad \alpha_0 = c[(\pi/2)^4 + 5(\pi/2)^2 + 4].$$

Substituting these results for $y(\sigma, t)$ and $U(t)$ into (5.87), the entropy change is written in the form

$$\frac{1}{N_0}\delta S_m(\sigma, t) = -\frac{\alpha_0 t}{12A(0, t_c)}y(\sigma, t) = -\frac{\alpha_0 t}{12c}\left[\frac{\sigma^2}{\sigma_0^2(0)} - 1\right]. \tag{5.90}$$

As for the specific heat, the second contribution δC_{m2} in (5.88) is neglected. The reason is because both $\mathrm{d}U(t)/\mathrm{d}t$ and $\partial y(\sigma, t)/\partial t|_h$ in (5.88) are proportional to t. As a whole, it is proportional to t^2. If we define the T-linear coefficient of the specific heat $\gamma(\sigma) = C_m(\sigma, t)/T$, as with the case of the paramagnetic phase, its change $\delta\gamma_m(\sigma) = \gamma_m(\sigma) - \gamma_m(0)$ is given by

$$\frac{1}{N_0}\delta\gamma_m(\sigma) = -\frac{\alpha_0}{12T_0A(0, t_c)}y(\sigma, t) = -\frac{\alpha_0}{12cT_0}\left[\frac{\sigma^2}{\sigma_0^2(0)} - 1\right], \tag{5.91}$$

by using the relation, $A(0, t_c) = cy_1(0)\sigma_0^2(0)$ in (3.12). In the region of weak magnetic field, the following relation is satisfied between $y(\sigma, t)$ and h.

$$y(\sigma, t) \equiv \frac{h}{2T_A\sigma} \simeq \frac{h}{2T_A\sigma_0(0)}. \tag{5.92}$$

It follows then that $\delta\gamma_m(\sigma)$ is proportional to h, and its coefficient is given by

$$\frac{1}{N_0}\frac{\partial\gamma_m}{\partial h} = \frac{15A(0, t_c)}{T_0}\frac{\mathrm{d}^2U(t)}{\mathrm{d}t^2}\frac{\partial y(\sigma, t)}{\partial h} = -\frac{5\alpha_0}{8T_A^2\sigma_0^3(0)}. \tag{5.93}$$

Around the Critical Temperature According to (4.38) in Chap. 4, the temperature dependence of the reduced spontaneous magnetization squared $U(t)$ is given by

$$\begin{aligned} U(t) &\simeq a_c\left[1 - \left(\frac{t}{t_c}\right)^{4/3}\right], \\ \frac{\mathrm{d}U(t)}{\mathrm{d}t} &\simeq -\frac{4a_c}{3t}\left(\frac{t}{t_c}\right)^{4/3} \to -\frac{4a_c}{3t_c}, \quad (t \to t_c). \end{aligned} \tag{5.94}$$

By putting the above derivative $\mathrm{d}U(t)/\mathrm{d}t$ and $y(\sigma, t_c) = y_c\sigma^4$ into (5.87), the entropy change induced by external magnetic field is given by

$$\frac{1}{N_0}\delta S_m(\sigma, t_c) = 15A(0, t_c)\left(-\frac{4a_c}{3t_c}\right) y_c\sigma^4 = -\frac{20a_c}{t_c} y_c A(0, t_c)\sigma^4. \tag{5.95}$$

From the continuity condition of the entropies, (5.95) and (5.81) in the paramagnetic phase in the limit $t \to t_c$, we have to assume $a_c = 7/5$ in (5.94). It implies $\xi = 1$ for the parameter introduced in (4.14) related to the presence of spin waves in Chap. 4.

In the deviation of the specific heat $\delta C_m(\sigma, t_c)$, the second temperature derivative $\mathrm{d}^2U(t)/\mathrm{d}t^2$ is necessary. We can find its value by expanding $U(t)$ and $V(t)$ in powers of $(t - t_c)$.

$$\begin{aligned} U(t) &= -u_1(t - t_c) - \frac{u_2}{2}(t - t_c)^2 + \cdots, \\ V(t) &= \frac{v_2}{2}(t - t_c)^2 + \frac{v_3}{6}(t - t_c)^3 + \cdots. \end{aligned} \tag{5.96}$$

The above coefficients u_1, u_2, v_2, and so on, are obtained by putting the above expansion into (4.22) and comparing coefficients of terms with the same powers of $(t - t_c)$. For instant, the first coefficient u_1 is given by $4a_c/3t_c$. Then from (5.88) with these parameters, $\delta C_m(h, t_c)$ is evaluated as follows:

$$\frac{\delta C_m(h, t)}{N_0 t} = -15A(0, t_c)\left[u_2 y(\sigma, t) + u_1 \left.\frac{\partial y(\sigma, t)}{\partial t}\right|_h\right]. \tag{5.97}$$

As with the case in the paramagnetic phase, both $y(\sigma, t)$ and $\partial y(\sigma, t)/\partial t$ at $t = t_c$ are positive, and proportional to σ^4 and σ^2, respectively. The above $\delta C_m(h, t)$ thus becomes negative.

We show in Fig. 5.5, numerically calculated temperature dependence of the entropy change $\delta S_m(\sigma, t)$ induced by external magnetic field. The field-induced change of the specific heat is always negative below t_c, for the slope of the entropy is negative as shown in Fig. 5.5. Whereas in the paramagnetic phase, it is positive. Therefore, $\delta C_m(h, t)$ shows the discontinuous change at the critical point $t = t_c$.

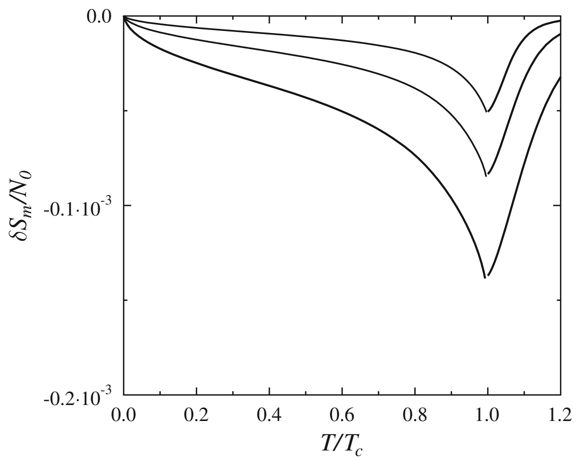

Fig. 5.5 Numerical estimated entropy change for $t_c = 0.1$ under the presence of magnetic field, $h = 0.05, 0.1$, and 0.2 ($\times 10^{-5}$) from the top

5.4.5 Numerical Estimate

To evaluate the entropy and the specific heat at any temperature and in the presence of the external magnetic field of any magnitude h, it is necessary to estimate the values of σ and those of $y(\sigma, t)$ and $y_z(\sigma, t)$ numerically, as well as their temperature derivatives. In the following, we will briefly show how to estimate these values.

Magnetization σ as Independent Variable In this method, we need to evaluate temperature derivatives of various variables as functions of σ. They are evaluated according to the explanation in Sect. 5.4.2. To estimate the value $\partial y(\sigma, t)/\partial t|_h$, for instance, first obtain the value of $\partial y(\sigma, t)/\partial t$, and then by (5.70). The derivative $\partial y(\sigma, t)/\partial t$ is estimated by solving the following simultaneous differential equation for $y(\sigma, t)$ and $\partial y(\sigma, t)/\partial t$ as functions of σ:

$$2A(y, t) + A(y_z, t) - c(2y + y_z) + 5cy_1(0)\sigma^2 = 3A(0, t_c) \tag{5.98}$$

$$2[A'(y, t) - c_z]\frac{\partial y}{\partial t} + [A'(y_z, t) - c_z]\frac{\partial y_z}{\partial t} + 2\frac{\partial A(y, t)}{\partial t} + \frac{\partial A(y_z, t)}{\partial t} = 0. \tag{5.99}$$

The first and the second lines correspond to the TAC condition and its temperature derivative. The functions $y_z(\sigma, t)$ and $\partial y_z(\sigma, t)/\partial t$ are related to $y(\sigma, t)$ and $\partial y(\sigma, t)/\partial t$ by

$$\begin{aligned} y_z(\sigma, t) &= y(\sigma, t) + \sigma\frac{\partial y(\sigma, t)}{\partial \sigma}, \\ \frac{\partial y_z(\sigma, t)}{\partial t} &= \frac{\partial y(\sigma, t)}{\partial t} + \sigma\frac{\partial}{\partial \sigma}\left(\frac{\partial y(\sigma, t)}{\partial t}\right). \end{aligned} \tag{5.100}$$

First σ derivatives of $y(\sigma, t)$ and $\partial y(\sigma, t)/\partial t$ in (5.98) and (5.99) are, therefore, determined by values of σ, $y(\sigma, t)$, and $\partial y(\sigma, t)/\partial t$. The magnetic field h corresponding to σ is determined by $h = 2T_A\sigma y(\sigma, t)$.

Magnetic Field h as Independent Variable On the other hand, it is possible to treat the problem by regarding h as independent variable. In this case, the magnetization $\sigma(h, t)$ is evaluated as a function of h, in place of finding $y(\sigma, t)$ as a function of σ. We then need to evaluate the derivative, $\partial\sigma/\partial t$. They are also evaluated as functions of h by using the same simultaneous equation (5.98) and (5.99).

Note that from the definition of $y(\sigma, t)$ and $y_z(\sigma, t)$, following relations are satisfied among σ, h, and these functions:

$$y(\sigma, t) = \frac{h}{2T_A\sigma}, \quad y_z(\sigma, t) = \frac{1}{2T_A\partial\sigma/\partial h}. \tag{5.101}$$

We can eliminate $y(\sigma, t)$ and $y_z(\sigma, t)$ by substituting (5.101) in (5.98). It is then regarded as the differential equation of σ as the function of h.

For the derivative $\partial\sigma/\partial t$, the t derivative of $y_z(\sigma, t)$ in the above definition (5.101) in a constant h can be written in the form

$$\left.\frac{\partial y_z(\sigma,t)}{\partial t}\right|_h = \frac{1}{2T_A}\frac{\partial}{\partial t}\left(\frac{\partial\sigma}{\partial h}\right)^{-1} = -\frac{1}{2T_A}\left(\frac{\partial\sigma}{\partial h}\right)^{-2}\frac{\partial}{\partial t}\left(\frac{\partial\sigma}{\partial h}\right)$$
$$= -2T_A y_z^2 \frac{\partial}{\partial h}\left(\frac{\partial\sigma}{\partial t}\right). \tag{5.102}$$

It is also written by

$$\left.\frac{\partial y_z(\sigma,t)}{\partial t}\right|_h = \frac{\partial y_z(\sigma,t)}{\partial t} + \frac{\partial y_z(\sigma,t)}{\partial \sigma}\left.\frac{\partial\sigma}{\partial t}\right|_h, \quad \left.\frac{\partial\sigma}{\partial t}\right|_h \equiv \frac{\partial\sigma(h,t)}{\partial t} \tag{5.103}$$

by regarding $y_z(\sigma,t)$ as a function of σ and t. By equating the right-hand sides of (5.102) and (5.103), $\partial y_z(\sigma,t)/\partial t$ is given by

$$\frac{\partial y_z(\sigma,t)}{\partial t} = -2T_A y_z^2 \frac{\partial}{\partial h}\left[\frac{\partial\sigma(h,t)}{\partial t}\right] - \frac{\partial y_z}{\partial\sigma}\frac{\partial\sigma(h,t)}{\partial t}. \tag{5.104}$$

By substituting (5.68) for $\partial y(\sigma,t)/\partial t$ and (5.104) for $\partial y_z(\sigma,t)/\partial t$, the first and second terms of (5.99) are written in the form,

$$2[A'(y,t)-c]\frac{\partial y(\sigma,t)}{\partial t} = -2[A'(y,t)-c]\frac{y_z}{\sigma}\frac{\partial\sigma}{\partial t}, \tag{5.105}$$

$$\begin{aligned}[A'(y_z,t)-c_z]\frac{\partial y_z(\sigma,t)}{\partial t} &= [A'(y_z,t)-c_z]\left[-2T_A y_z^2\frac{\partial}{\partial h}\left(\frac{\partial\sigma}{\partial t}\right) - \frac{\partial y_z}{\partial\sigma}\frac{\partial\sigma}{\partial t}\right] \\ &= -2T_A y_z^2[A'(y_z,t)-c_z]\frac{\partial}{\partial h}\left(\frac{\partial\sigma}{\partial t}\right) \\ &\quad + \left\{2[A'(y,t)-c]\frac{\partial y}{\partial\sigma} + 10cy_{10}\sigma\right\}\frac{\partial\sigma}{\partial t},\end{aligned} \tag{5.106}$$

with using the relation,

$$2[A'(y,t)-c_z]\frac{\partial y}{\partial\sigma} + [A'(y_z,t)-c_z]\frac{\partial y_z}{\partial\sigma} + 10c_z y_{10}\sigma = 0, \tag{5.107}$$

for $\partial y_z/\partial\sigma$, derived from the σ derivative of the TAC condition (5.98). Equation (5.106) is therefore finally written in the form

$$\begin{aligned}2T_A y_z^2[A'(y_z,t)-c_z]\frac{\partial}{\partial h}\left(\frac{\partial\sigma}{\partial t}\right) &= \left\{-2[A'(y,t)-c_z]\frac{y_z-y}{\sigma} + 10c_z y_{10}\sigma\right\}\frac{\partial\sigma}{\partial t} \\ &\quad + 2\frac{\partial A(y,t)}{\partial t} + \frac{\partial A(y_z,t)}{\partial t}.\end{aligned} \tag{5.108}$$

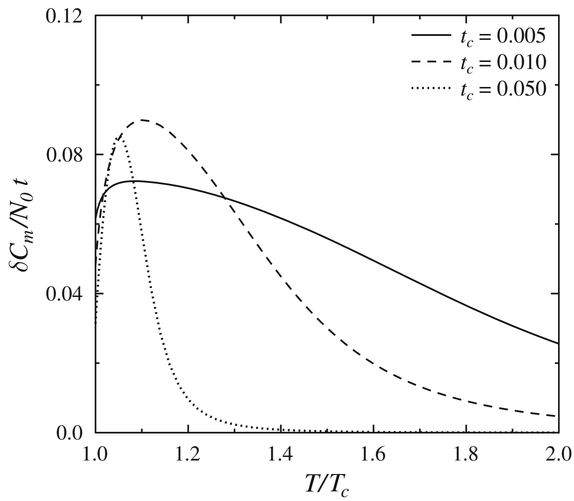

Fig. 5.6 Numerical examples of the temperature dependence of the specific heat change for $t_c = T_c/T_0 = 0.0005$ (*solid*), 0.01 (*dashed*), 0.05 (*dotted*) in the presence of magnetic field

We can now regard (5.98) with (5.101) and (5.108) as the simultaneous differential equation for σ and $\partial\sigma/\partial t$ as functions of h. The initial condition at $\sigma = 0$ in the paramagnetic phase, for instance, is given by

$$2T_A y_0(t)^2 \frac{\partial}{\partial h}\left(\frac{\partial\sigma}{\partial t}\right) = -\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}. \tag{5.109}$$

In this way, we can evaluate $y(\sigma, t)$ and its temperature derivative in (5.88) in a constant h as the functions of h.

Results of Numerical Calculations We have already shown in Fig. 5.5, the temperature dependence of the entropy, i.e., (5.57) and (5.87), in the presence of static external magnetic field. The entropy is always suppressed at any temperature by externally applied magnetic field. It results from the development of the magnetic ordering as the result of the field suppressed fluctuation amplitudes. The temperature dependence of the specific heat is evaluated as the derivative of the entropy with respect to temperature. Characteristic behaviors of Fig. 5.5 are therefore reflected in the temperature dependence of the specific heat. We expect from the steep decreases at low temperatures and around the critical point with increasing temperature in this figure, that the specific heats will show sizable increases of their magnitudes in these regions.

In Fig. 5.6, numerical results of the temperature dependence of the specific heat change in the paramagnetic phase in the constant magnetic field $h = 1.0 \times 10^{-4}$. The values of $\delta C_m/N_0$ are plotted against T/T_c for $t_c = 0.005$, 0.01, and 0.05 by solid, dashed, and dotted lines, respectively. There appear peaks above the critical temperature. Be aware that they are not plotted against T but the reduced temperature T/T_c. Such a peak is actually observed in Sc_3In by Takeuchi and Masuda [2] as shown

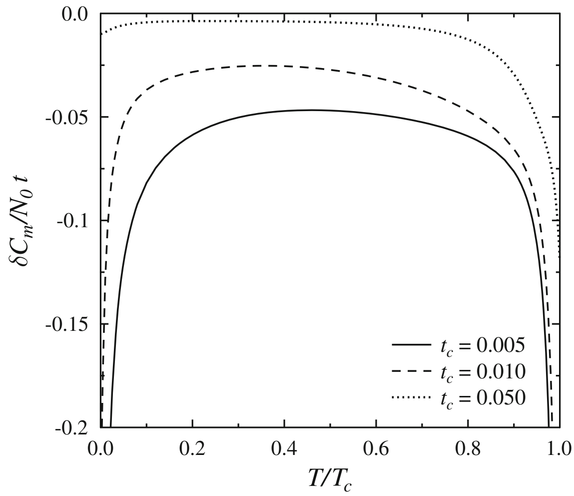

Fig. 5.7 Numerically estimated examples of the temperature dependence of the specific heat change in the ordered phase for $t_c = 0.005$ (*solid*), 0.01 (*dashed*), and 0.05 (*dotted*)

in Fig. 5.1 (right). The peak value is about 2 mJ/K^2 g-atom for $H = 2$ T, estimated by assuming that all atoms are magnetic. If we assume only Sc is magnetic and $T_0 = 500$ K, the value $T_0(\delta C_m/N_0 T)_{\max} \simeq 0.16$ is obtained. Numerical result by Takahashi and Nakano [6] gives a peak value of 0.1 by using the same values of T_0 and T_A.

Numerically estimated examples in the ordered phase are also shown in Fig. 5.7. They show steep decreases at low temperatures and near the critical temperature reflecting to the corresponding changes of entropies. Widths of them tend to become narrower for smaller t_c. These behaviors result from the second derivative $\mathrm{d}^2U(t)/\mathrm{d}t^2$ in δC_{m1}.

5.4.6 External Field Effect on Paramagnets Near the QCP

According to (5.34), the magnetic entropy of exchange-enhanced paramagnets in the presence of external magnetic field is given by

$$\begin{aligned}\frac{1}{N_0}S_m(\sigma, t) = & -3\int_0^1 \mathrm{d}x x^2\{2[\Phi(u) - u\Phi'(u)] + [\Phi(u_z) - u_z\Phi'(u_z)]\}\{\cdots\} \\ & + \frac{1}{N_0}\Delta S_m(\sigma, t), \quad u = x(y + x^2)/t, \quad u_z = x(y_z + x^2)/t.\end{aligned} \tag{5.110}$$

Since the correction $\Delta S_m(\sigma, t) \propto \sigma^4$ for paramagnets is neglected in the weak-field region, the specific heat in the presence of external field h is given by

$$\frac{1}{N_0 t} C_m(\sigma, t) = \frac{1}{N_0} \frac{\partial S_m}{\partial t} = -\frac{3}{t} \int_0^1 \mathrm{d}x x^2 [2u^2 \Phi''(u) + u_z^2 \Phi''(u_z)] \\ - 6 \frac{\partial A(y, t)}{\partial t} \frac{\partial y}{\partial t}\bigg|_h - 3 \frac{\partial A(y_z, t)}{\partial t} \frac{\partial y_z}{\partial t}\bigg|_h, \quad (5.111)$$

where t derivatives of $y(\sigma, t)$ and $y_z(\sigma, t)$ in a constant magnetic field are evaluated by (5.70). The induced magnetization σ involved in $y(\sigma, t)$ and $y_z(\sigma, t)$ in the right-hand side of (5.111) is determined by solving the magnetic isotherm,

$$y(\sigma, t) = \frac{h}{2T_A \sigma} \simeq y_0(t) + y_1(t)\sigma^2 + \cdots . \quad (5.112)$$

Around the quantum critical point (QCP), the effect of external magnetic field on the specific heat is understood associated with the cross-over between the critical and the low-temperature regions defined in Sect. 3.5.1. Just at the QCP, $t_p = 0$, the T-linear coefficient of the specific heat exhibits the $\log(1/t)$ increase with decreasing temperature toward $t = 0$, as shown in (5.30). It is the characteristic behavior for the critical region, $y/t^{2/3} \ll 1$, because the temperature evolution of $y(0, t) \propto t^{4/3}$ remains unchanged within this region at low temperatures. By applying the external magnetic field, the system will make the transition from the critical to the low-temperature region for $y/t^{2/3} \gg 1$, since the values of $y(\sigma, t)$ and $y_z(\sigma, t)$ become finite, according to (5.112). We will then expect that the critical $\log(1/t)$ behavior of $C_m/N_0 t$ will change into the $\log(1/y)$ behavior at low temperatures. However, with increasing temperature, the system makes the transition into the critical region again, because of the temperature evolution of $y(\sigma, t)$. Therefore, the $\log(1/t)$ behavior will also be recovered.

The above cross-over behavior of the specific heat can be confirmed by numerical studies. In the limit of low temperature, the effect of external field on the last two terms in (5.111) is negligible, since they are higher order corrections with respect to temperature. From the same reason, the magnetic isotherm is approximated by that in the ground state. Numerically estimated temperature dependence of the t-linear coefficient of the specific heats for various external magnetic fields are shown in Fig. 5.8.

5.5 Summary

In this chapter, we have proposed the free energy of the spin fluctuation degrees of freedom, that is consistent with the TAC condition. Based on the free energy, we have shown that the temperature and the external field dependence of the entropy and the specific heat are derived from the unified point of view as summarized below.

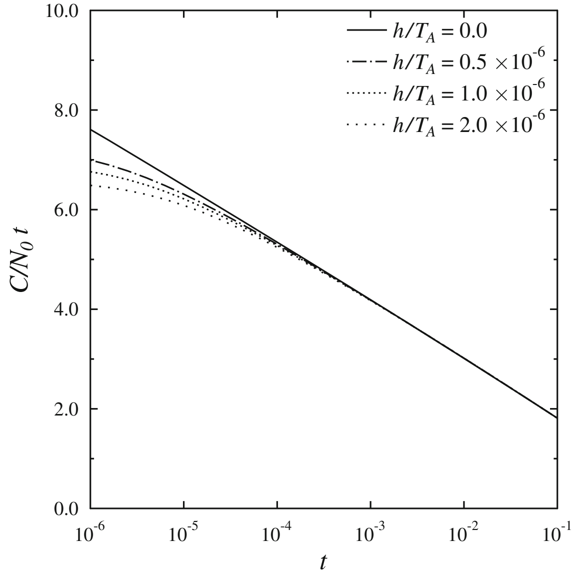

Fig. 5.8 Temperature dependence of the specific heat of a paramagnet at $t_p = 1.0 \times 10^{-4}$ near the QCP in the presence of the external magnetic field

1. Systematic treatment of the entropy and the specific heat becomes possible in predicting various properties even quantitatively through the wide temperature range that can be compared with experiments.
2. Field dependence of our entropy is consistent with the Maxwell relation of the thermodynamics in both the paramagnetic and the ordered phases. As a consequence, the term proportional to $\mathrm{d}^2\sigma_0^2(t)/\mathrm{d}t^2$ is involved in the change of the specific heat in the ordered phase as the effect of externally applied magnetic field.

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Chapter 6
Magneto Volume Effect

6.1 Introduction

Magneto-volume effect is the phenomena resulting from the interplay between magnetism and volume change of crystals. For instance, the volume contraction by applying external pressure will change the magnitude of spontaneous moment as well as the critical temperature of magnetic transition. Conversely, the appearance of spontaneous magnetization also results in the volume expansion of crystals.

Volume change δV of crystals of magnetic origins is usually described by

$$\delta V = \kappa C_s M_0(T)^2 + \kappa C_h [M^2 - M_0^2(T)], \tag{6.1}$$

where $M_0(T)$ is the spontaneous magnetization. In this chapter, the compressibility of crystals is denoted by κ. The first term in (6.1) is called spontaneous magneto-striction. The second term, called forced magneto-striction, represents the volume change induced by applying external magnetic field. The ratio of the volume change δV to the volume V, i.e., $\omega \equiv \delta V/V$, is generally called volume-strain. In the theory of elasticity, the volume-strain ω is used rather than the volume change itself. Coefficients C_s and C_h in (6.1) are magneto-volume coupling constants (or magneto-elastic constant).

Among others, the invar alloys are known as an example in which the magneto-volume effect appears outstandingly. In these alloys, the thermal expansion arising from lattice vibrations is compensated by the volume contraction from this effect. As a consequence, they show almost no thermal expansion in some range of temperature. The property is utilized in various area of technological applications. Weak itinerant electron ferromagnets usually have large magneto-volume coupling constants, though their spontaneous magnetic moments are very small. For such reasons, a large number of pressure effect experiments had been made from the mid 1960s to 1980s.

The purpose of this chapter is to clarify the effects of spin fluctuations on the volume change of crystals based on the free energy in the preceding chapter.

Y. Takahashi, *Spin Fluctuation Theory of Itinerant Electron Magnetism*, Springer Tracts in Modern Physics 253, DOI: 10.1007/978-3-642-36666-6_6,

6.1.1 Thermal Expansion Due to Lattice Vibrations

Lattice vibrations is a typical example of boson excitations in crystals. It is known that the anharmonicity of lattice vibrations bring about the thermal expansion of crystals. Prior to our discussion on the magneto-volume effect and the involvement of spin fluctuations in this effect, it will be helpful for us to understand how the thermal expansion is derived from the lattice vibrations.

Thermodynamically, thermal expansion of crystals is derived from the volume derivative of the free energy. The free energy of the Debye model of the lattice vibrations is given by

$$\begin{aligned} \mathscr{F}(T,V) &= \frac{V}{2\kappa}\omega^2 + F_{\mathrm{lat}}(T,V), \\ F_{\mathrm{lat}}(T,V) &= \sum_{qs}\left[\frac{1}{2}\nu_{qs} + T\log(1-\mathrm{e}^{-\nu_{qs}/T})\right], \end{aligned} \tag{6.2}$$

as the sum of the elastic energy of the first term and the free energy $F_{\mathrm{lat}}(T,V)$ of the Debye mode. The anharmonicity is included as the volume dependence of the frequency ν_{qs} of phonons with wave vector q for component s. From the thermodynamic relation for the pressure p, the temperature dependence of the volume striction is given by

$$-p = \frac{\partial\mathscr{F}(T,V)}{\partial V} = \frac{1}{V}\frac{\partial\mathscr{F}(T,V)}{\partial\omega} = \frac{1}{\kappa}\omega + \frac{1}{V}\frac{\partial F_{\mathrm{lat}}(T,V)}{\partial\omega}, \tag{6.3}$$

$$\omega(T) = -\kappa p + \omega_{\mathrm{lat}}(T), \quad \omega_{\mathrm{lat}}(T) = -\frac{\kappa}{V}\frac{\partial F_{\mathrm{lat}}(T,V)}{\partial\omega}, \tag{6.4}$$

where the first term in (6.4) represents the volume contraction by pressure p. The second term of ω_{lat} is the thermal volume expansion originating from lattice vibrations. The volume dependence of phonon frequencies is usually defined by $\nu_{qs} \propto V^{-\gamma_{qs}}$. As the average of exponents γ_{qa}, the following Grüneisen parameter γ is defined by

$$\gamma = -\frac{\mathrm{d}\log\Theta_D}{\mathrm{d}\log V}, \tag{6.5}$$

where Θ_D is the Debye temperature.

According to the definition (6.3), the volume thermal expansion $\omega_{\mathrm{lat}}(T)$ is given by

$$\omega_{\mathrm{lat}}(T) = \kappa\sum_{qs}\frac{\partial\nu_{qs}}{\partial V}\left[\frac{1}{2}+n(\nu_{qs})\right] = \frac{\kappa\gamma}{V}\sum_{qs}\nu_{qs}\left[\frac{1}{2}+n(\nu_{qs})\right], \tag{6.6}$$

where $n(\nu_{qs}) = [\mathrm{e}^{\nu_{qs}/T}-1]^{-1}$. The volume thermal expansion coefficient is then evaluated by further differentiating (6.6) with respect to the temperature T:

$$\beta(T) = \frac{d\omega_{lat}(T)}{dT} = \frac{\kappa\gamma}{V}\sum_{qs} \nu_{qs}\frac{\partial}{\partial T}\left[\frac{1}{2} + n(\nu_{qs})\right] = \frac{\kappa\gamma}{V}\sum_{qs}\nu_{qs}\frac{\partial n(\nu_{qs})}{\partial T}$$
$$c_V(T) = \frac{1}{V}\sum_{qs}\nu_{qs}\frac{\partial n(\nu_{qs})}{\partial T} \tag{6.7}$$

For isotropic crystals, the relation $\alpha(T) = \beta(T)/3$ is satisfied between the linear and volume thermal expansion coefficients, $\alpha(T)$ and $\beta(T)/3$. Thus the following Grüneisen relation is satisfied between the thermal expansion coefficient and the specific heat at constant volume:

$$\alpha(T) = \frac{1}{3}\kappa\gamma c_V(T) \propto T^3 \quad \text{for } T/\Theta_D \ll 1 \tag{6.8}$$

6.2 Stoner-Edwards-Wohlfarth Theory and its Correction

At the beginning, the magneto-volume effect is mainly understood by the Stoner-Edwards-Wohlfarth (SEW) theory. It is based on the Stoner-Wohlfarth (SW) free energy (1.53) in Chap. 1. Later in 1980, the theory was revised by Moriya and Usami [1] phenomenologically by including the contribution of spin fluctuations into the SEW free energy. We first show a brief outline of these theories.

6.2.1 SEW Theory of Magneto-Volume Effect

In the SEW theory, the following free energy is used for the derivation of the magneto-volume effect:

$$\mathscr{F}(M, T, V) = \frac{V}{2\kappa}\omega^2 + F_0(T, V) + F_m(M, T, V), \tag{6.9}$$

$$F_m(M, T, V) = F_m(0, T, V) + \frac{1}{2}a(T, V)M^2 + \frac{1}{4}b(T, V)M^4 + \cdots . \tag{6.10}$$

The second term $F_0(T, V)$ of (6.9) represents the contribution from the nonmagnetic degrees of freedom such as lattice vibrations. The third one $F_m(M, T, V)$ is the Stoner-Wohlfarth free energy (1.53), resulting from the band splitting of the conduction electron states. The SEW theory assumes that the coefficient of $a(T, V)$ in the SEW free energy (6.10) depends on the volume. The volume dependence of the higher coefficients, $b(T, V)$ for instance, are usually neglected. As is shown in (1.59), $a(T, V)$ in SW theory is given in terms of the single electron density of state $\rho(\varepsilon)$ at the Fermi energy and their energy derivatives, $\rho'(\varepsilon)$ and $\rho''(\varepsilon)$, as well as the intra-atomic electron-electron interaction I. The volume dependence of $a(T, V)$ is therefore determined by these quantities.

The volume strain is evaluated by the volume derivative of the free energy (6.9),

$$\begin{aligned}\omega(M,T) &= -\kappa p + \omega_0(T) + \omega_m(M,T),\\ \omega_0(T) &= -\frac{\kappa}{V}\frac{\partial F_0(T,V)}{\partial\omega},\\ \omega_m(M,T) &= -\frac{\kappa}{V}\frac{\partial F_m(T,V)}{\partial\omega} = -\frac{\kappa}{2V}\frac{\partial a(T,V)}{\partial\omega}M^2 + \cdots,\end{aligned} \tag{6.11}$$

where the terms $\omega_0(T)$ and $\omega_m(M,T)$ represent the nonmagnetic and magneto-volume contributions, respectively. The following consequences are derived from (6.11).

1. The spontaneous magneto-striction in the ordered phase
   In the absence of the external magnetic field, the magnetization M in (6.11) is replaced by the spontaneous moment $M_0(T)$. The first term of (6.1) is written by

$$\omega_m(T) = \frac{\kappa C}{V}M_0(T)^2, \quad C = -\frac{1}{2}\frac{\partial a(T,V)}{\partial\omega}. \tag{6.12}$$

   No spontaneous magneto-striction is present in the paramagnetic phase, because of the absence of the spontaneous magnetization $M_0(T)$. The magneto-volume coupling constant is given by the negative of the derivative of the coefficient $a(T,V)$ with respect to the strain ω.
2. The forced magneto-striction
   Increase of the magnetization induced by the external magnetic field also contributes to the volume expansion. An extra volume change from this effect in addition to (6.12) gives the second term of (6.1), i.e.,

$$\Delta\omega_m(M,T) = \frac{\kappa C}{V}[M^2 - M_0^2(T)]. \tag{6.13}$$

   Since the same coupling constant C appears, $C_s = C_h$ is satisfied. It can be applied in the paramagnetic phase, but with $M_0(T) = 0$.
3. Effects of volume change on the spontaneous magnetic moment and the critical temperature
   The conditions of (1.65) in Chap. 1 for the spontaneous magnetization in the ground state and its volume derivative give the following two relations:

$$\begin{aligned}a(0,V) + b(0,V)M_0^2(0,V) = 0,\\ \frac{\partial a(0,V)}{\partial\omega} + b(0,V)\frac{\partial M_0^2(0,V)}{\partial\omega} = 0\end{aligned} \tag{6.14}$$

   With the use of the definition of the coupling constant C in (6.12), the effect of the volume strain on the spontaneous magnetization is written in the form

$$\frac{\partial M_0^2(0, V)}{\partial \omega} = \frac{2C}{b(0, V)}. \tag{6.15}$$

We can also find the effect on the critical temperature T_c from the condition of $a(T_c, V) = 0$. The variation of this condition with respect to the change of volume strain $\delta\omega$ is given by

$$\left.\frac{\partial a(T, V)}{\partial T}\right|_{T=T_c} \delta T_c + \frac{\partial a(T, V)}{\partial \omega}\delta\omega = \frac{2a(0, V)}{T_c}\delta T_c - 2C\delta\omega = 0, \tag{6.16}$$

where we assume the volume dependence of $a(T, V) = a(0, V)[1 - T^2/T_c^2(V)]$. The temperature dependence of C is neglected. The ω derivative of $\log T_c$ is thus given by

$$\frac{1}{T_c}\frac{\partial T_c}{\partial \omega} = \frac{\partial \log T_c}{\partial \omega} = \frac{C}{a(0, V)} = \frac{C}{b(0, V)M_0^2(0, V)}. \tag{6.17}$$

From the comparison of (6.15) and (6.17), we are finally led to the following relation:

$$\frac{\partial \log M_0}{\partial \omega} = \frac{\partial \log T_c}{\partial \omega}. \tag{6.18}$$

4. Temperature dependence of the magneto-volume coupling constant
   In this theoretical framework, the value of C is expressed in the form

$$C = \frac{1}{4N\rho(\varepsilon_F)\mu_B^2}\left[\frac{\partial \rho(\varepsilon_F)}{\partial \omega} + \bar{I}\frac{\partial I}{\partial \omega} + \frac{T^2}{T_F^2}\left(\frac{\partial \rho(\varepsilon_F)}{\partial \omega} + 2\frac{\partial T_F}{\partial \omega}\right)\right], \tag{6.19}$$

where $\bar{I} \equiv I\rho(\varepsilon_F)$. As shown in Chap. 1, $\rho(\varepsilon_F)$ and I represent the density of states at the Fermi energy and the repulsive Coulomb energy among conduction electrons. From the temperature dependence the Fermi distribution function, the above T^2-linear dependence is derived [2–4]. The volume dependence of the parameters $\rho(\varepsilon_F)$ and I are actually estimated numerically based on band structure calculations. In such studies, the $V^{-5/3}$ dependence of the d-electron band width by Heine [5] has been usually employed.

6.2.2 *Correction of the Free Energy of Spin Fluctuations*

Whereas the volume effect on the SW free energy is only taken in consideration in the SEW theory, Moriya and Usami [1] proposed its revision by including the contribution of spin fluctuations into their free energy. In place of the free energy $F_m(M, T, V)$ in (6.10), the following free energy is employed by them.

$$F_m(M, T, V) = \frac{1}{2}a(T, V)M^2 + \frac{1}{4}bM^4 + \frac{1}{2}\sum_{q\neq 0}\frac{1}{\chi(q)}\langle M_q \cdot M_{-q}\rangle + \cdots. \quad (6.20)$$

In addition to the coefficient $a(T, V)$ for the uniform ($q = 0$) component of the magnetization, they assumed the volume dependence of $\chi^{-1}(\mathbf{q})$ for the spatially modulated magnetization. Then the volume derivative of the free energy is given by

$$\begin{aligned}\frac{\partial F_m(M, T, V)}{\partial \omega} &\simeq -CM^2 + \frac{1}{2}\sum_{q\neq 0}\frac{\partial \chi^{-1}(q)}{\partial \omega}\langle M_q \cdot M_{-q}\rangle \\ &= -CM^2 - \sum_{q\neq 0} C_q \langle M_q \cdot M_{-q}\rangle. \end{aligned} \quad (6.21)$$

Only the thermal components of fluctuations are included as before. Then the wave-vector dependence of the coupling C_q is neglected, since thermal fluctuations around $q = 0$ are mainly excited usually. Thus the following result of the spontaneous volume striction is derived.

$$\omega_m(T) = \frac{\kappa C}{V}[M_0^2(T) + \xi^2(T)], \quad \xi^2(T) = \sum_q \langle \delta M_q \cdot \delta M_{-q}\rangle, \quad (6.22)$$

where $\xi^2(T)$ stands for the average of the thermal spin fluctuation amplitude squared.

According to Moriya and Usami, their theory of magneto-volume effect is different from the SEW theory in the following respects.

1. The presence of the spontaneous magneto-striction at the critical temperature. In the SEW theory, the spontaneous volume striction $\omega_m(0) = \kappa C M_0^2(0)/V$ below T_c disappears at $T = T_c$, i.e., $\omega_m(T_c) = 0$. Though both theories predict the same spontaneous volume striction $\omega_m(0)$ in the ground state, the volume striction in the MU theory remains finite, and its value is given by $\omega_m(T_c) = \kappa C\xi^2(T_c)/V$. If we notice the relation, $\xi^2(T_c) = 3M_0(0)^2/5$ satisfied between the thermal spin amplitude squared at $T = T_c$ and the spontaneous magnetization squared in the ground state, the volume contraction in the MU theory from the ground state to the critical point remains $2/5$ of the value in the SEW theory.
2. The presence of the magnetic thermal expansion in the paramagnetic phase. Although no thermal volume expansion of the magnetic origin is present in the SEW theory, the MU theory predicts the presence of the thermal volume expansion in the paramagnetic phase that results from the thermal spin fluctuation amplitude $\xi^2(T)$ in (6.22). It shows the T-linear increase with increasing temperature in the region where the Curie–Weiss law behavior of magnetic susceptibility is observed.

To exhibit a qualitative difference, the temperature dependence of thermal volume expansions predicted by these two theories is shown in Fig. 6.1. From the analysis of thermal expansion measurements on MnSi, Matsunaga et al. [6] reported the presence

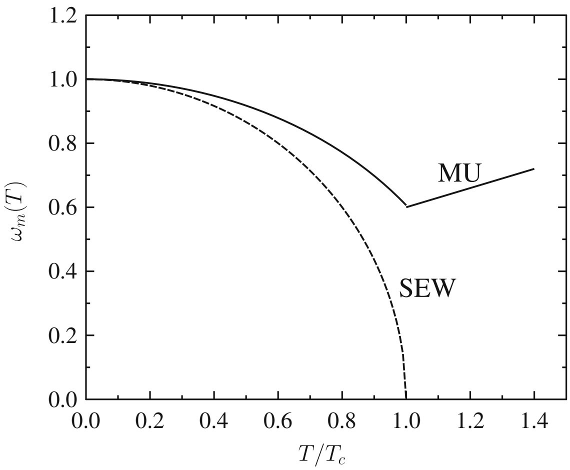


Fig. 6.1 Temperature dependence of the spontaneous volume magneto-striction by the SEW and the MU theories: the difference results from whether the effect of the thermal spin fluctuation amplitude $\xi^2(T)$ is present or not

of these two effects. The same analyses were also reported by Ogawa [7] on $ZrZn_2$, by Suzuki and Masuda [34, 35] on Ni_3Al, and by Shimizu et al. [50] on (Fe,Co)Si.

Stimulated by the MU theory, various theoretical investigations have been made since then. For instance, Hasegawa [8] has treated the magneto-volume effect of Hubbard model in the case with larger amplitudes of spin fluctuations based on the functional integral method by using the static single-site approximation. Results of the temperature dependence of the spontaneous magneto-striction were reported. Volume dependence of the model is included by the $V^{-5/3}$-dependence of d-band width by Heine [5]. The same numerical method was applied on the temperature dependence of the magneto-volume striction by Kakehashi [9] based on the Liberman-Pettifor's Virial theorem. These authors also reported their results of numerical studies on the pressure effect on the Curie temperature [10] the elastic constant of Fe at finite temperature [11] as well as Invar effect [12, 13]. On the other hand, the following result of magnetic pressure,

$$V_0 P_{\rm mag}(T) \simeq \frac{5}{3}[U(T) + Im^2(T)/4], \tag{6.23}$$

was derived by Holden [14] to show that so much drastic change of the volume magneto-striction does not happen above T_c with no spontaneous moment. In (6.23), $U(T)$ and $m(T)$ represent the internal energy and the amplitude of the local magnetic moment, respectively. Along the line of this theory, the magneto-volume effect of Fe–Co alloy is theoretically treated by Joynt [15].

The purpose of most of these theories was to understand the magneto-volume effect associated with electronic band structure of magnetic materials. This book rather sticks to the predominant roles of collective magnetic excitation on various magnetic phenomena. Then Grüneisen's approach to the thermal expansion will be very helpful. We are also required to cope with the following questions.

- What is the origin of the magneto-volume effect?
  If we insist on the predominant roles of the spin fluctuations, it is better to deal with the magneto-volume effect based on the same free energy, which is used in our previous discussions on various magnetic and thermal properties. The magneto-volume effect is to be related with the direct volume dependence of the free energy. Originally, the electron gas model was assumed for the dispersion of the conduction electrons in the spin fluctuation theory by Moriya and Kawabata [16, 17]. Based on the same model and approach, the magneto-volume effect was treated later by Edwards and Macdonard [18]. By assuming the volume dependence for the dispersion of the electron gas model, they have derived the volume strain and the pressure effect on the critical temperature T_c. Since only the perpendicular components of fluctuations are included with respect to the induced magnetization, it is inconsistent with the rotational symmetry of the system. Their volume expansion gives the ratio $\eta(T_c) = \omega_m(T_c)/\omega_m(0) > 1$ at $T = T_c$, in disagreement with 3/5 predicted by the MU theory. It may result from the symmetry breaking treatment, according to their arguments.
- Are there any contributions from zero-point spin fluctuation amplitudes?
  The presence of the zero-point amplitude is likely to be neglected from the beginning. The reason to neglect only one out of artificially divided two components is not so clear. Solontsov and Wagner (1995) argued that because of the nonlinear effect of zero-point spin amplitudes, the right hand of (6.22) can be rewritten by [19]
  $$\omega_m(T) = \rho\kappa C M^2 + \rho\kappa \sum_{\nu} [C(\delta m_\nu^2)_T + C_0(\delta m_\nu^2)_Z], \qquad (6.24)$$
  where ν denotes the transverse and the longitudinal components with respect to the spontaneous magnetization. The last term represents the contribution from zero-point spin fluctuations. The same origin is assumed for the magneto-volume coupling constants as those of the SEW and MU theories.
- What the relation between the pressure effects on the spontaneous magnetization and the critical temperature is satisfied?
- Is there any relation between the magneto-volume effect and the magnetic specific heat?
  If the same free energy as that for the specific heat is used for the magneto-volume effect, some kind of Grüneisen's relation should be satisfied between them.
- Are there any differences between the spontaneous and forced magneto-striction?

6.3 Volume Dependence of the Free Energy

In our view, the magneto-volume effect should be treated in the same way as the entropy and the specific heat in Chap. 5. It is then better to employ the free energy (5.2) as the magnetic contribution F_m in (6.9) [20]. Let us divide it into two parts, i.e., the thermal and the other components, F_{th} and F_{zp}, respectively.

$$
\begin{aligned}
F_m(y,\sigma,t,\omega) &= F_{th}(y,y_z,\sigma,t,\omega) + F_{zp}(y,y_z,\sigma,t,\omega) \\
F_{th} &= \frac{2}{\pi}\sum_q{}' \int_0^\infty \mathrm{d}\nu\, T \log(1-\mathrm{e}^{-\nu/T}) \frac{\Gamma_q}{\nu^2+\Gamma_q^2} \\
&\quad + \frac{1}{\pi}\sum_q \int_0^\infty \mathrm{d}\nu\, T \log(1-\mathrm{e}^{-\nu/T}) \frac{\Gamma_q^z}{\nu^2+(\Gamma_q^z)^2} + \Delta F_{th} \\
F_{zp} &= \frac{1}{\pi}\sum_q \int_0^{\nu_c} \mathrm{d}\nu\, \frac{\nu}{2}\left\{ 2\frac{\Gamma_q}{\nu^2+\Gamma_q^2} + \frac{\Gamma_q^z}{\nu^2+(\Gamma_q^z)^2} \right\} \\
&\quad + N_0 T_A y\sigma^2 - \frac{1}{3} N_0 T_A \left\langle S_i^2 \right\rangle_{\mathrm{tot}} (3y + \Delta y_z) + \Delta F_{zp}
\end{aligned} \tag{6.25}
$$

The corrections ΔF_{th} and ΔF_{zp} represent the thermal and all the rest components of ΔF_1 in (5.2), respectively. Since the effect of spin waves is neglected here, for simplicity, the summation $\sum'$ means that the spin-wave region around the origin is excluded.

Notice that two spectral parameters T_0 and T_A are included in the above free energy. They correspond to the Debye temperature Θ_D in the model of lattice vibrations. It is therefore reasonable to assume that these parameters are volume dependent. On the other hand, variables y, Δy_z, and σ should be determined by the extremum conditions of the free energy as well as to satisfy the thermodynamic relations. In the following, we are particularly concerned with the explicit volume dependence of the free energy. Its explicit volume deviation is then denoted by

$$
\delta_{\mathrm{v}} F_m = \delta_{\mathrm{v}} F_{th} + \delta_{\mathrm{v}} F_{zp}, \quad \delta_{\mathrm{v}} f(y,\sigma,t,\omega) \equiv \frac{\partial f(y,\sigma,t,\omega)}{\partial\omega}\delta\omega. \tag{6.26}
$$

To begin with, let us first examine how the thermal component of the free energy F_{th} is affected by the volume change of crystals. From the volume dependence of the spectral parameter T_0, the volume change will give rise to following deviation of the damping constant in (6.25):

$$
\delta_{\mathrm{v}}\Gamma_q = 2\pi\delta T_0 x(y+x^2) = \frac{\delta T_0}{T_0}\Gamma_q, \quad \delta_{\mathrm{v}}\Gamma_q^z = \frac{\delta T_0}{T_0}\Gamma_q^z \tag{6.27}
$$

Consequently, the variation of the thermal component of the free energy is written in the form

$$
\begin{aligned}
\delta_{\mathrm{v}} F_{th} &= \frac{\delta T_0}{T_0}\frac{1}{\pi}\sum_q \int_0^\infty \mathrm{d}\nu T \log(1-\mathrm{e}^{-\nu/T}) \\
&\quad \times \left\{ 2\Gamma_q \frac{\partial}{\partial\Gamma_q}\left(\frac{\Gamma_q}{\nu^2+\Gamma_q^2}\right) + \Gamma_q^z \frac{\partial}{\partial\Gamma_q^z}\left(\frac{\Gamma_q^z}{\nu^2+(\Gamma_{q^z})^2}\right)\right\} + \delta_{\mathrm{v}}\Delta F_{th} \\
&= \frac{\delta T_0}{T_0}\frac{1}{\pi}\sum_q \int_0^\infty \mathrm{d}\nu n(\nu) \left\{ 2\frac{\nu\Gamma_q}{\nu^2+\Gamma_q^2} + \frac{\nu\Gamma_q^z}{\nu^2+(\Gamma_q^z)^2}\right\} + \delta_{\mathrm{v}}\Delta F_{th},
\end{aligned} \tag{6.28}
$$

by using integration by parts and the following relation:

$$\frac{\partial}{\partial \nu}\frac{\nu}{\nu^2+\Gamma^2} = -\frac{\partial}{\partial \Gamma}\frac{\Gamma}{\nu^2+\Gamma^2}. \tag{6.29}$$

The last line of (6.28) is rewritten by using the derivative of the function $\Phi(u)$ defined in (5.21). The wave-vector summation and the frequency integral is given by

$$\frac{T}{N_0}\sum_q \frac{\Gamma_q}{2\pi T}\cdot 2\int_0^\infty \mathrm{d}\nu \frac{\nu}{\mathrm{e}^{\nu/T}-1}\frac{1}{\nu^2+\Gamma_q^2} = 3T_0 t\int_0^1 \mathrm{d}x x^2 u(x)\Phi'[u(x)],$$

where $x = q/q_B$ is the reduced wave-number and $u(x) = \Gamma_q/2\pi T$. With this result, the first term in (6.28) is further rewritten as

$$\delta_{\mathrm{v}} F_{th} = 3N_0 T_0 \frac{\delta T_0}{T_0} t\left[2\int_{x_c}^1 \mathrm{d}x x^2 u\Phi'(u) + \int_0^1 \mathrm{d}x x^2 u_z \Phi'(u_z)\right] \tag{6.30}$$

where $u = x(y+x^2)/t$ and $u_z = x(y_z+x^2)/t$. The derivative of the thermal component ΔF_{th} with regards to $\Delta y_z \equiv y_z - y$ is given by

$$\delta_{\mathrm{v}}\left(\frac{\partial \Delta F_{th}}{\partial \Delta y_z}\right) = -2N_0\delta_{\mathrm{v}}\{T_0[A(y_z,t)-A(y,t)]\}$$

Let us first evaluate the variation of the right hand side. Then its integral with respect to Δy_z gives

$$\delta_{\mathrm{v}}\Delta F_{th} \simeq -2N_0 T_0 \frac{\delta T_0}{T_0}\Delta y_{z0}\left\{A(y_{z0},t) - A(0,t) - t\left[\frac{\partial A(y_{z0},t)}{\partial t} - \frac{\partial A(0,t)}{\partial t}\right]\right\}\delta\omega \tag{6.31}$$

where $y_z = y_{z0}$ and $y = 0$ are assumed since we need the spontaneous striction here.

As for the component F_{zp}, it can be expanded with respect to y and Δy_z around their origins. The deviation $\delta_{\mathrm{v}} F_{zp}$ is then expanded as follows:

$$\delta_{\mathrm{v}} F_{zp}(y,\Delta y_z,\omega) = \delta_{\mathrm{v}} F_{zp}(0,0,\omega) + \frac{\partial \delta_{\mathrm{v}} F_{zp}(0,0,\omega)}{\partial y} y + \frac{\partial \delta_{\mathrm{v}} F_{zp}(0,0,\omega)}{\partial \Delta y_z}\Delta y_z + \cdots. \tag{6.32}$$

To evaluate the above linear coefficients with respect to y and Δy_z, note the relations (5.3), (5.5), and (5.8) in Chap. 5 are satisfied. In (6.32), the derivatives of $F_{zp}(y, y_z, \omega)$ with respect to these variables are then given by

$$\begin{aligned}
\frac{\partial F_{zp}(y,\Delta y_z,\omega)}{\partial y} &\to N_0 T_A\left[\left\langle \delta S_{\rm loc}^2\right\rangle_Z(0,0)+\sigma^2-\left\langle S_{\rm loc}^2\right\rangle_{\rm tot}\right],\\
\frac{\partial F_{zp}(y,\Delta y_z,\omega)}{\partial \Delta y_z} &\to N_0 T_A\left[\left\langle (\delta S_{\rm loc}^z)^2\right\rangle_Z(0)-\frac{1}{3}\left\langle S_{\rm loc}^2\right\rangle_{\rm tot}-\lambda_{zp}(\sigma,t)\right],\\
\lambda_{zp}(\sigma,t) &\to -\frac{\sigma^2}{3},\quad \text{for } y\to 0 \text{ and } \Delta y_z\to 0,
\end{aligned} \tag{6.33}$$

where $\lambda_{zp}(\sigma,t)$ represents a portion of $\lambda(\sigma,t)$ in (5.9) excluding the thermal contributions. By exchanging the order of the variation $\delta_{\rm v}$ and the differentiation with respect to y or Δy_z, the right hand side of (6.32) is rewritten in the form

$$\begin{aligned}
\frac{\partial \delta_{\rm v} F_{zp}}{\partial y} &= \delta_{\rm v}\left(\frac{\partial F_{zp}}{\partial y}\right) = -N_0\delta_{\rm v}\left[T_A\Delta\left\langle S_{\rm loc}^2\right\rangle_{\rm tot}\right]+N_0\delta T_A\sigma^2\\
\frac{\partial \delta_{\rm v} F_{zp}}{\partial \Delta y_z} &= \frac{1}{3}\frac{\partial \delta_{\rm v} F_{zp}}{\partial y},\quad \Delta\left\langle S_{\rm loc}^2\right\rangle \equiv \left\langle S_{\rm loc}^2\right\rangle_{\rm tot}-\left\langle S_{\rm loc}^2\right\rangle_Z(0).
\end{aligned} \tag{6.34}$$

After all, the variation of the free energy due to the volume change is given by

$$\begin{aligned}
&\delta_{\rm v} F_{zp}(y,y_z,\sigma,t,\omega) = -N_0 C_{zp}(2y+y_z)\delta\omega+\cdots,\\
&3C_{zp}\delta\omega = \delta_{\rm v}\left[T_A\Delta\left\langle S_{\rm loc}^2\right\rangle\right]-\sigma^2\delta T_A.
\end{aligned} \tag{6.35}$$

For ferromagnets, since $\Delta\left\langle S_{\rm loc}^2\right\rangle$ and $\sigma_0^2(0)$ are of the same order of magnitude, the term $\sigma^2\delta T_A$ in the above second line cannot be neglected. On the other hand, the first term $\delta_{\rm v} F_{zp}(0,0,\omega)$ in (6.32) is neglected, for it is constant independent of temperature.

With these free energy variations given in (6.30) and (6.35), the volume magnetostriction is evaluated by their derivatives with respect to the volume stain ω, i.e., as the sum of two components,

$$\begin{aligned}
\omega_m(t) &= -\frac{\kappa}{V}\frac{\partial F_m}{\partial\omega} = \omega_{th}(t)+\omega_{zp}(t),\\
\omega_{th}(t) &= -\frac{\kappa}{V}\frac{\partial F_{th}}{\partial\omega},\quad \omega_{zp}(t) = -\frac{\kappa}{V}\frac{\partial F_{zp}}{\partial\omega}.
\end{aligned} \tag{6.36}$$

6.3.1 Magnetic Grüneisen Parameters

Let us next introduce magnetic Grüneisen parameters in place of magneto-volume coupling constants. If we note the expression of the variations of free energies (6.28) and (6.32), it will be appropriate to define the following three Grüneisen parameters [20].

- Two parameters, γ_0 and γ_A, that characterize the volume dependence of spectral parameters T_0 and T_A.
  These spectral parameters are defined as distribution widths of the imaginary part of the dynamical magnetic susceptibility $\mathrm{Im}\,\chi(q,\omega)$ in frequency and wave-vector spaces, respectively. They therefore correspond to the Debye temperature Θ_D in lattice vibrations and the exchange interaction constant J in the Heisenberg model of localized spin systems. The following two magnetic Grüneisen parameters are defined as strain derivatives of logarithm of these values.

$$\gamma_0 = -\frac{\mathrm{d}\log T_0}{\mathrm{d}\omega}, \quad \gamma_A = -\frac{\mathrm{d}\log T_A}{\mathrm{d}\omega}, \tag{6.37}$$

  In terms of these parameters, variations of δT_0 and δT_A are represented by

$$\frac{\delta T_0}{T_0} = \frac{\mathrm{d}\log T_0}{\mathrm{d}\omega}\delta\omega = -\gamma_0\delta\omega, \quad \frac{\delta T_A}{T_A} = -\gamma_A\delta\omega.$$

- Parameter γ_m that characterize the volume dependence of the spin fluctuation amplitude, $\Delta\left\langle S_{\mathrm{loc}}^2\right\rangle$ defined in (6.34).
  This difference of the amplitudes is supposed to depend on the volume of the system. From the strain derivative of its logarithm, the third Grüneisen parameter is defined by

$$\gamma_m = \frac{\mathrm{d}\log\Delta\left\langle S_{\mathrm{loc}}^2\right\rangle}{\mathrm{d}\omega}. \tag{6.38}$$

  Because of the spin amplitude conservation, the value $\Delta\left\langle S_{\mathrm{loc}}^2\right\rangle$ is equivalent to the critical thermal amplitude squared $\left\langle S_{\mathrm{loc}}^2\right\rangle_T(0,t_c)$, i.e., the value $3\sigma_0^2(0)/5$ according to (3.12) in Chap. 3. Thus the above definition is also written in the form

$$\frac{\mathrm{d}\Delta\left\langle S_{\mathrm{loc}}^2\right\rangle}{\mathrm{d}\omega} = \frac{3}{5}\sigma_0^2(0)\gamma_m. \tag{6.39}$$

With these definitions, the coefficient C_{zp} in (6.35) is given by

$$\begin{aligned} C_{zp} &= \frac{1}{3}T_A\left\{\frac{\mathrm{d}\log T_A}{\mathrm{d}\omega}\left[\langle S_{\mathrm{loc}}^2\rangle - \sigma^2\right] + \frac{\mathrm{d}\log\Delta\langle S_{\mathrm{loc}}^2\rangle}{\mathrm{d}\omega}\Delta\langle S_{\mathrm{loc}}^2\rangle\right\} \\ &= \frac{1}{5}T_A\sigma_0^2(0)\left[\gamma_m + \gamma_A\left(\frac{5}{3}\frac{\sigma^2}{\sigma_0^2(0)} - 1\right)\right]. \end{aligned} \tag{6.40}$$

For later convenience, let us also introduce the following ratios g_A and g_0 defined by

$$g_A = \frac{\gamma_A}{\gamma_m}, \quad g_0 = \frac{\gamma_0}{\gamma_m} \tag{6.41}$$

According to Fawcett [21], Grüneisen parameters are defined as negatives of volume-strain derivatives of the logarithm of characteristic energy scales of phenomena. The first two parameters are introduced according to this criterion. It represents the variation of the spectral widths caused by the volume contraction by external pressure. They are equivalent of the volume dependence, $T_0 \propto V^{-\gamma_0} \propto e^{-\gamma_0\omega}$, $T_A \propto V^{-\gamma_A} \propto e^{-\gamma_A\omega}$. For the analysis of thermal expansion of heavy fermion systems, the Grüneisen parameter is introduced into the SCR spin fluctuation theory by Kambe et al. [22]. However, the volume dependence of parameter T_0 and T_A was assumed to be neglected.

6.3.2 *Forced Magneto-Striction and Maxwell Relation*

In our treatment of the magnetic specific heat in Chap. 5, we show that the Maxwell relation is satisfied for our free energy, i.e., (5.58) and (5.65) in the paramagnetic and ordered phases, respectively. Since the same free energy is used in this chapter, we assume from the beginning that the relation is satisfied.

For the free energy with independent variables σ and pressure p, its total derivative is given by

$$\mathrm{d}F(\sigma, p) = V\mathrm{d}p + N_0 h \mathrm{d}\sigma. \tag{6.42}$$

The following Maxwell relation is then satisfied.

$$\frac{1}{V}\frac{\partial V}{\partial \sigma}\bigg|_p = \frac{\partial \log V}{\partial \sigma}\bigg|_p = \frac{\partial \omega}{\partial \sigma}\bigg|_p = \frac{N_0}{V}\frac{\partial h}{\partial p}\bigg|_\sigma. \tag{6.43}$$

With the use of the compressibility κ, the pressure derivative is replaced by the ω derivative by

$$\frac{\partial}{\partial p} = -\kappa\frac{\partial}{\partial \omega}, \quad \kappa \equiv -\frac{\partial \omega}{\partial p}\bigg|_\sigma. \tag{6.44}$$

The relation in (6.43) is therefore written in the form

$$\frac{\partial \omega(\sigma, t)}{\partial \sigma} = \frac{N_0}{V}\sigma\frac{\partial (2T_A y)}{\partial p} = -2\kappa\rho\sigma\frac{\partial (T_A y)}{\partial \omega}, \tag{6.45}$$

where $\rho = N_0/V$ and $y = h/2T_A\sigma$. After substituting the Grüneisen parameter γ_A into the volume dependence of T_A, (6.45) is finally given by

$$\begin{aligned}\frac{\partial \omega_h(\sigma, t)}{\partial \sigma} &= 2\rho\kappa C_h(\sigma, t)\sigma,\\ C_h(\sigma, t) &= -T_A\left(\frac{1}{T_A}\frac{\partial T_A}{\partial \omega}y + \frac{\partial y}{\partial \omega}\right) = T_A\left[\gamma_A y(\sigma, t) - \frac{\partial y(\sigma, t)}{\partial \omega}\right].\end{aligned} \tag{6.46}$$

Hereafter, the forced magneto-striction is denoted by $\omega_h(\sigma, t)$ to avoid confusion.

Equation (6.46) is regarded as the general expression for the forced magneto-striction. To evaluate the value of $\omega_h(\sigma, t)$ at arbitrary value of σ, we need to find the solution of (6.46) by regarding its first line as a differential equation in σ. Because of the σ dependence of the coupling constant $C_h(\sigma, t)$, we have to know the σ dependence of $y(\sigma, t)$ and its volume derivative $\partial y(\sigma, t)/\partial\omega$.

6.4 Volume Magneto-Striction for Ferromagnets

Spontaneous and forced magneto-strictions are treated in this section based on the volume dependence of the free energy in Sect. 6.3. Let us first deal with systems of ferromagnets.

6.4.1 Magneto-Volume Effect in the Ground State

In the ground state with no thermal spin fluctuation amplitudes, inverses of reduced magnetic susceptibilities are given by $y(\sigma_0, 0) = 0$ and $y_z(\sigma_0, 0) = y_{z0}(0) = 2y_1(0)\sigma_0^2(0)$. The spontaneous magnetic moment is denoted by $\sigma_0(0)$. In this case, the spontaneous and forced magneto-strictions, $\omega_{zp}(0)$ and $\omega_h(\sigma, 0)$, are evaluated as follows.

- Spontaneous magneto-striction
  Since $\sigma = \sigma_0(0)$ is satisfied in (6.40) in the absence of the external field, $C_{zp}(0)$ is given by
  $$C_{zp}(0) = \frac{1}{5}\left(\gamma_m + \frac{2}{3}\gamma_A\right) T_A \sigma_0^2(0). \tag{6.47}$$
  From (6.35) and (6.36), the spontaneous magneto-striction is given by
  $$\begin{aligned} &\omega_{zp}(0) = \rho\kappa C_{zp}(0) y_{z0}(0) = \rho\kappa C_s(0)\sigma_0^2(0), \quad y_{z0}(0) = 2y_1(0)\sigma_0^2(0), \\ &C_s(0) = 2C_{zp}(0) y_1(0) = \frac{2}{5}\left(\gamma_m + \tfrac{2}{3}\gamma_A\right) T_A y_1(0)\sigma_0^2(0). \end{aligned} \tag{6.48}$$
  The function $C_s(0)$ has a meaning of the magneto-volume coupling constant for the spontaneous striction.
- Forced magneto-striction
  In the region of weak external magnetic field, the σ dependence of $C_h(\sigma, t)$ in (6.46) is neglected. The forced striction is given by
  $$\omega_h(\sigma, t) = \rho\kappa C_h(\sigma_0, 0)[\sigma^2 - \sigma_0^2(0)]. \tag{6.49}$$

The magneto-volume coupling constant $C_h(\sigma_0, 0)$ is also evaluated by putting the σ dependence of $y(\sigma, t) \simeq y_1(0)[\sigma^2 - \sigma_0^2(0)]$ into (6.46).

$$\begin{aligned} C_h(\sigma_0, 0) &= -T_A \frac{\partial}{\partial \omega} \{y_1(0)[\sigma^2 - \sigma_0^2(0)]\}\Big|_{\sigma=\sigma_0} \\ &= -T_A \left\{ \frac{\partial y_1(0)}{\partial \omega}[\sigma^2 - \sigma_0^2(0)] - y_1(0)\frac{\partial \sigma_0^2(0)}{\partial \omega} \right\}\Bigg|_{\sigma=\sigma_0} \\ &= T_A y_1(0) \gamma_m \sigma_0^2(0). \end{aligned} \tag{6.50}$$

Thus it depends only on the parameter γ_m in the ground state.
If we define $C_h(t) \equiv C_h(\sigma_0, 0)$, the comparison of two magneto-volume coupling constants, (6.48) and (6.50), for spontaneous and forced strictions leads to the relation:

$$\frac{C_s(0)}{C_h(0)} = \frac{2}{5}\left(1 + \frac{2}{3} g_A\right), \tag{6.51}$$

where g_A is defined in (6.41). The quite different result is derived from $C_s = C_h$ by the SEW and the MU theories.

- Effect of pressure on spontaneous magnetic moment
  From the definition of γ_m, the ω derivative of $\sigma_0^2(0, \omega)$ is given by

$$\frac{1}{\Delta \langle S_{\text{loc}}^2 \rangle} \frac{\mathrm{d}\Delta \langle S_{\text{loc}}^2 \rangle}{\mathrm{d}\omega} = \frac{1}{\sigma_0^2(0)} \frac{\mathrm{d}\sigma_0^2(0)}{\mathrm{d}\omega} = \gamma_m. \tag{6.52}$$

  It follows that the pressure dependence of the spontaneous moment is given by

$$\sigma_0^2(0, \omega) = \sigma_0^2(0, 0)(1 + \gamma_m \omega) = \sigma_0^2(0)(1 - \kappa \gamma_m p). \tag{6.53}$$

  The parameter γ_m can be therefore estimated by the change of the spontaneous magnetization at low temperatures induced by external pressure.

In conclusion, the magneto-volume effect in the ground state is described by

$$\omega_m(M, 0) = \frac{\rho\kappa C_s(0)}{(2N_0\mu_{\text{B}})^2} M_0^2(0) + \frac{\rho\kappa C_h(0)}{(2N_0\mu_{\text{B}})^2}[M^2 - M_0^2(0)], \tag{6.54}$$

with two different coupling constants.

6.4.2 *Ferromagnets at Finite Temperatures*

Temperature dependence of thermal volume expansion Spontaneous magneto-striction in the ordered phase is also obtained according to the general expression of the volume striction (6.36). It consists of two components, $\omega_{th}(t)$ in (6.30) and $\omega_{zp}(t)$ in (6.35), derived by the volume derivatives of corresponding components of the free energy. They are given by

$$\begin{aligned}
\omega_{th}(t) &= 3\rho\kappa T_0\gamma_0 t\left[2\int_{x_c}^1 \mathrm{d}x x^2 u\Phi'(u) + \int_0^1 \mathrm{d}x x^2 u_z\Phi'(u_z)\right] + \Delta\omega_{th}(t), \\
\omega_{zp}(t) &= \rho\kappa C_{zp}(t)y_{z0}(t) = \rho\kappa C_s(t)\sigma_0^2(t), \quad y_{z0}(t) = 2y_1(t)\sigma_0^2(t), \qquad (6.55) \\
C_s(t) &= \frac{2}{5}C_h(0)\frac{V(t)}{U(t)}\left[1 + g_A\left(\frac{5}{3}U(t) - 1\right)\right], \quad \frac{y_1(t)}{y_1(0)} = \frac{V(t)}{U(t)},
\end{aligned}$$

where $U(t)$ and $V(t)$ are defined in (4.21). The coefficient $C_{zp}(t)$ defined in (6.40) is given by

$$C_{zp}(t) = \frac{1}{5}T_A\sigma_0^2(0)\left[\gamma_m + \gamma_A\left(\frac{5}{3}U(t) - 1\right)\right], \tag{6.56}$$

for $\sigma = \sigma_0(t)$ in the absence of external field at finite temperatures. In the same way, the thermal expansion derived from the free energy correction ΔF_{th} is given by

$$\Delta\omega_{th}(t) = 2\rho\kappa T_0\gamma_0\Delta y_{z0}\left\{t\left[\frac{\partial A(y_{z0},t)}{\partial t} - \frac{\partial A(0,t)}{\partial t}\right] - A(y_{z0},t) + A(0,t)\right\}. \tag{6.57}$$

Thermal component $\omega_{th}(t)$ in (6.55) results from the thermal component of the free energy. It therefore increases monotonically with increasing temperature. In the paramagnetic phase, $u = u_z$ and $y = y_z$ are satisfied, as well as $x_c = 0$ since no spin-waves are present. The thermal correction $\Delta\omega_{th}(t)$ is also absent. The component $\omega_{zp}(t)$ from zero-point fluctuations is proportional to $\sigma_0^2(t)$, in the ordered phase. In the paramagnetic phase, it becomes proportional to the inverse of the magnetic susceptibility $y_0(t)$, since $3y_0(t)$ appears in place of $y_{z0}(t)$ for $T < T_c$. Its temperature dependence is similar to that of the MU theory. Using the correspondence between $y_0(t)$ and $y_1(t)\sigma_0^2(t)$ in the paramagnetic and the ordered phases, the definition (4.21) can be extended to the paramagnetic phase by

$$U(t) = \frac{y_0(t)}{y_1(t)\sigma_0^2(0)}, \quad V(t) = \frac{y_0(t)}{y_1(0)\sigma_0^2(0)} = \frac{y_1(t)}{y_1(0)}U(t). \tag{6.58}$$

In the paramagnetic phase, the temperature dependence of $\omega_{zp}(t)$ is then written by

$$\begin{aligned}
\omega_{zp}(t) &= \rho\kappa C_{zp}(t)[3y_0(t)] = \rho\kappa C_s(t)\frac{y_0(t)}{y_1(t)}, \\
C_{zp}(t) &= \frac{1}{5}T_A\sigma_0^2(0)(\gamma_m - \gamma_A), \quad C_s(t) \equiv 3y_1(t)C_{zp}(t)
\end{aligned} \tag{6.59}$$

In terms of reduced parameters, (6.59) is finally represented by

$$\omega_{zp}(t) = \rho\kappa C_s(t)\sigma_0^2(0)U(t)$$
$$C_s(t) = \frac{3}{5}C_h(0)(1-g_A)\frac{y_1(t)}{y_1(0)} = \frac{3}{5}C_h(0)(1-g_A)\frac{V(t)}{U(t)}. \tag{6.60}$$

Hereafter, let us introduce the constant ω_0 by

$$\omega_0 = \rho\kappa C_h(0)\sigma_0^2(0), \tag{6.61}$$

as a unit of volume-strain. The component $\omega_{zp}(t)$ in (6.59) is then given in more simplified form

$$\omega_{zp}(t) = \frac{3}{5}\omega_0(1-g_A)V(t). \tag{6.62}$$

The ratios of each component of thermal expansions in (6.55) to the unit strain ω_0 are also written by

$$\begin{aligned}\frac{\omega_{th}(t)}{\omega_0} &= \frac{g_0 t}{5c[y_1(0)\sigma_0^2(0)]^2} \\ &\quad \times \left\{2\int_{x_c}^1 \mathrm{d}x x^2 u\Phi'(u) + \int_0^1 \mathrm{d}x x^2 u_z \Phi'(u_z)\right\} + \frac{\Delta\omega_{th}(t)}{\omega_0}, \\ \frac{\Delta\omega_{th}(t)}{\omega_0} &= \frac{2g_0 y_{z0}}{15c[y_1(0)\sigma_0^2(0)]^2} \\ &\quad \times \left\{t\left[\frac{\partial A(y_{z0},t)}{\partial t} - \frac{\partial A(0,t)}{\partial t}\right] - A(y_{z0},t) + A(0,t)\right\}, \\ \frac{\omega_{zp}(t)}{\omega_0} &= \frac{2}{5}V(t)\left[1 + g_A\left(\frac{5}{3}U(t) - 1\right)\right].\end{aligned} \tag{6.63}$$

Likewise, thermal expansion coefficients are also given as the sum of reduced components:

$$\begin{aligned}\beta(t) &= \frac{\mathrm{d}\omega_s(t)}{\mathrm{d}T} = \frac{\omega_0}{T_0}\bar\beta(t), \\ \bar\beta(t) &= \frac{\mathrm{d}\omega_s(t)}{\mathrm{d}t} = \bar\beta_{th}(t) + \Delta\bar\beta_{th}(t) + \bar\beta_{zp}(t).\end{aligned} \tag{6.64}$$

Each of them are given by

$$\begin{aligned}\bar\beta_{th}(t) = \frac{cg_0}{5A^2(0,t_c)}&\left\{-2\int_{x_c}^1 \mathrm{d}x x^2 u^2 \Phi''(u) - \int_0^1 \mathrm{d}x x^2 u_z^2 \Phi''(u_z)\right. \\ &+ \frac{\mathrm{d}V(t)}{\mathrm{d}t}\left[-\frac{tx_c}{V(t)}x_c^2 u_c\left(\log u_c - \frac{1}{2u_c} - \psi(u_c)\right)\right. \\ &\left.\left. + 2y_1(0)\sigma_0^2(0)\left(A(y_{z0},t) - t\frac{\partial A(y_{z0},t)}{\partial t}\right)\right]\right\},\end{aligned}$$

$$
\begin{aligned}
\Delta\bar{\beta}_t(t) = \frac{4g_0}{15A(0,t_c)}\bigg\{ V'(t)\bigg[t\left(\frac{\partial A(y_{z0},t)}{\partial t} - \frac{\partial A(0,t)}{\partial t} + y_{z0}\frac{\partial A'(y_{z0},t)}{\partial t}\right) \\
- A(y_{z0},t) + A(0,t) - y_{z0}A'(y_{z0},t)\bigg] \\
+ tV(t)\left[\frac{\partial^2 A(y_{z0},t)}{\partial t^2} - \frac{\partial^2 A(0,t)}{\partial t^2}\right]\bigg\}, \\
\bar{\beta}_{zp}(t) = \frac{2}{5}\left\{ (1-g_A)V'(t) + \frac{5}{3}g_A[V'(t)U(t)+V(t)U'(t)]\right\}, \quad u_c = x_c^3/t,
\end{aligned}
\tag{6.65}
$$

where $A'(y,t) \equiv \partial A(y,t)/\partial y$.

These results derived above are different from those of the MU theory in the following respects.

1. The presence of an extra thermal volume expansion, $\omega_{th}(t)$, in (6.55).
   Its temperature dependence is quite different from the one derived by Moriya and Usami, although both are associated with thermal spin fluctuation amplitudes.
2. The magneto-volume coupling constants do depend on temperature.
   The reason is because Grüneisen parameters are not defined as the expansion coefficient with respect to $\sigma_0^2(t)$, but $\Delta y_z = y_{z0}(t)$. Hence, there appears in $C_s(t)$ the temperature dependent proportionality factor $y_1(t)$ contained in $y_{z0}(t)$. In addition for finite γ_A, another dependence proportional to $\sigma_0^2(t)$ also appears. At the critical point, it vanishes, i.e., $C_s(t_c) = 0$, reflecting the temperature dependence of $y_1(t)$.
   The dependence of $C_h(t)$ for the forced magneto-striction will be explained later.
3. Spontaneous and forced magneto-volume coupling constants, C_s and C_h, are different in their magnitudes.

Volume Expansion below T_c The ratio of spontaneous magneto-volume strictions at $T = 0$ and $T = T_c$, i.e., $\eta = \omega_m(t_c)/\omega_m(0)$, was introduced by the MU theory, as a measure of the volume contraction from the ground state to the critical point with increasing temperature. They claimed that the value of η is different for the SEW and MU theories. Because the magneto-volume coupling constants are different for the spontaneous and the forced magneto-strictions in our theory, the same comparison is impossible. Therefore, it seems rather preferable to introduce a new definition of η by

$$
1-\eta = \frac{\Delta\omega_m(0)}{\omega_0}, \quad \Delta\omega_m(t) = \omega_m(t) - \omega_m(t_c). \tag{6.66}
$$

In place of $\omega_m(0)$ in the denominator, we employ our unit strain ω_0 defined in (6.61) evaluated by using the forced magneto-volume coupling constant $C_h(0)$.

In the SEW theory with no thermal amplitudes at the critical point, $1-\eta = 1$ (i.e., $\eta = 0$) is derived, for $\omega_m(t_c) = 0$ is satisfied. According to the MU theory, on the other hand, η is given by

$$1-\eta=\frac{1}{\omega_0(0)}[\omega_m(0)-\omega_m(t_c)]=\frac{\sigma_0^2(0)-\xi^2(t_c)}{\sigma_0^2(0)}=\frac{2}{5}, \quad (6.67)$$

for $\omega_m(0)=\omega_0$ and $\xi^2(t_c)=3\sigma_0^2(0)/5$ are satisfied. The same ratio of η is derived for each of the SEW and the MU theories independent of definitions. The difference between them originates only from the presence of the thermal amplitude $\xi^2(T)$ in (6.22). Whereas in our treatment, the value of $\Delta\omega_m(0)$ is estimated by

$$\begin{aligned}\Delta\omega_m(0)&=[\omega_{th}(0)+\omega_{zp}(0)]-[\omega_{th}(t_c)+\omega_{zp}(t_c)]\\&=-\omega_{th}(t_c)+\rho\kappa C_s(0)\sigma_0^2(0)=\frac{2}{5}\left(1+\frac{2}{3}g_A\right)\omega_0-\omega_{th}(t_c), \quad (6.68)\end{aligned}$$

since $\omega_{th}(0)=0$ and $\omega_{zp}(t_c)=0$. The value of $1-\eta$ is given by

$$1-\eta=\frac{\Delta\omega_m(0)}{\omega_0}=\frac{2}{5}\left(1+\frac{2}{3}g_A\right)-\frac{\omega_{th}(t_c)}{\omega_0}. \quad (6.69)$$

Nearly the same value as the MU theory is therefore derived, as long as the thermal component $\omega_{th}(t_c)$ is negligible. However, it results from the different origin, i.e., from the different magneto-volume coupling constants, $C_s(0)/C_h(0)\simeq 2/5$. Since the effect of thermal amplitudes is generally involved in (6.69), the value of $1-\eta$ is not restricted to the single value $2/5$ but will take a variety of values.

Forced Magneto-Striction To estimate the forced magneto-striction for an arbitrary magnetization σ, numerical integration of (6.46) with respect to σ is necessary. Then $\omega_h(\sigma,t)$ is given by

$$\omega_h(\sigma,t)=2\rho\kappa T_A\int_{\sigma_0(t)}^{\sigma}\mathrm{d}\sigma'\sigma'\left[\gamma_A y(\sigma',t)-\frac{\partial y(\sigma',t)}{\partial\omega}\right], \quad (6.70)$$

where $\sigma_0(t)=0$ in the paramagnetic phase. The derivative $\partial y(\sigma,t)/\partial\omega$ in the above integrand is estimated as a solution of the following simultaneous differential equation:

$$2A(y,t)+A(y_z,t)-c(2y+y_z)+5cy_1(0)\sigma^2=3A(0,t_c), \quad (6.71)$$

$$\begin{aligned}&2[A'(y,t)-c_z]\frac{\partial y}{\partial\omega}+[A'(y_z,t)-c_z]\left[\frac{\partial y}{\partial\omega}+\sigma\frac{\partial}{\partial\sigma}\left(\frac{\partial y}{\partial\omega}\right)\right]\\&\quad+5cy_1(0)(-\gamma_A+\gamma_0)\sigma^2=3A(0,t_c)\gamma_m(1-g_A+g_0). \quad (6.72)\end{aligned}$$

Equation (6.71) represents the TAC condition (3.3). The second Eq. (6.72) is its partial derivative with respect to ω. The following relation, derived from $A(0,t_c)=cy_1(0)\sigma_0^2$ in (3.11) and $y_1(0)=T_A/15cT_0$ in (3.10), is used in the above derivation.

$$\frac{\partial \log A(0,t_c)}{\partial \omega} = \frac{\partial \log y_1(0)}{\partial \omega} + \frac{\partial \log \sigma_0^2(0)}{\partial \omega},$$
$$\therefore \frac{\partial A(0,t_c)}{\partial \omega} = (\gamma_m - \gamma_A + \gamma_0)A(0,t_c). \tag{6.73}$$

Equation (6.70) is also written in the form of the derivative with respect to ω as given by

$$\frac{\partial}{\partial \sigma}\left(\frac{\omega_h(\sigma,t)}{\omega_0}\right) = \frac{2c\sigma}{A(0,t_c)}\left[g_A y(\sigma,t) - \frac{1}{\gamma_m}\frac{\partial y(\sigma,t)}{\partial \omega}\right]. \tag{6.74}$$

The forced magneto-striction $\omega_h(\sigma,t)$ is then obtained as the solution of the simultaneous differential equation consisting of (6.71), (6.72), and (6.74).

Initial value of $y(\sigma,t)$ is given by $y_0(t)$ for $\sigma = 0$ in the paramagnetic phase, and 0 for $\sigma = \sigma_0(t)$ in the ordered phase. Initial value of the derivative $\partial y(\sigma,t)/\partial\omega$ in (6.72) and (6.74) is related to the forced magneto-striction in the weak external magnetic field limit. In this limit, (6.46) in the paramagnetic phase is written as

$$\omega_h(\sigma,t) = \rho\kappa C_h(0,t)\sigma^2, \quad C_h(0,t) = T_A y_0(t)\left[\gamma_A - \frac{\partial \log y_0(t)}{\partial \omega}\right], \tag{6.75}$$

for $y(\sigma,t) \simeq y_0(t) + y_1(t)\sigma^2 \to y_0(t)$ in (5.50) is satisfied. The temperature dependence of $y_0(t)$ is determined as the solution of (3.30). Its ω derivative is then given by

$$\left[A'(y_0,t) - c\right]\frac{\partial y_0(t)}{\partial \omega} = \frac{\partial A(0,t_c)}{\partial \omega} = c(\gamma_m - \gamma_A + \gamma_0)y_1(0)\sigma_0(0). \tag{6.76}$$

Thus the initial condition of the derivative $\partial y(\sigma,t)/\partial\omega \to \partial y_0(t)/\partial\omega$ (for $\sigma \to 0$) is estimated by

$$\begin{aligned}\frac{\partial y_0(t)}{\partial \omega} &= -\frac{y_1(t)}{c y_1(0)}\frac{\partial A(0,t_c)}{\partial \omega} = -(\gamma_m - \gamma_A + \gamma_0)\frac{\sigma_0^2(0)}{\sigma_0^2(t)}y_0(t),\\ \therefore \frac{\partial \log y_0(t)}{\partial \omega} &= -\frac{1}{U(t)}\gamma_m(1 - g_A + g_0),\end{aligned} \tag{6.77}$$

with using (3.50) for $y_1(t)$. By putting these results of initial conditions into (6.75), the temperature dependence of $C_h(t)$ is given by

$$\begin{aligned}C_h(t) &\equiv C_h(0,t) = T_A y_1(t)\sigma_0^2(t)\gamma_m\left[g_A + (1 - g_A + g_0)\frac{1}{U(t)}\right],\\ \frac{C_h(t)}{C_h(0)} &= \frac{V(t)}{U(t)}\left\{1 - g_A[1 - U(t)] + g_0\right\},\end{aligned} \tag{6.78}$$

with the use of $C_h(0)$ defined in (6.50).

In the case of the ordered phase, the initial condition of the derivative is given by

$$\begin{aligned}\frac{\partial y(\sigma,t)}{\partial\omega} &= -y_1(t)\frac{\partial\sigma_0^2(t)}{\partial\omega}, \quad \text{for } \sigma\to\sigma_0(t),\\ \frac{\partial\log\sigma_0^2(t)}{\partial\omega} &= \frac{\partial\log\sigma_0^2(0)}{\partial\omega}+\frac{\partial\log U(t)}{\partial\omega} = \gamma_m+\frac{\partial\log U(t)}{\partial\omega},\end{aligned} \tag{6.79}$$

since $y(\sigma,t)\simeq y_1(t)[\sigma^2-\sigma_0^2(t)]\to 0$ is satisfied. Equation (6.46) is therefore given by

$$\begin{aligned}\omega_h(\sigma,t) &= \rho\kappa C_h(\sigma_0(t),t)[\sigma^2-\sigma_0^2(t)],\\ \frac{C_h(t)}{C_h(0)} &= \frac{C_h(\sigma_0(t),t)}{T_A\gamma_m y_1(0)\sigma_0^2(0)} = \frac{1}{\gamma_m}V(t)\frac{\partial\log\sigma_0^2(t)}{\partial\omega}\\ &= \frac{V(t)}{U(t)}\left[U(t)+\frac{1}{\gamma_m}\frac{\partial U(t)}{\partial\omega}\right].\end{aligned} \tag{6.80}$$

To evaluate the initial value of the σ derivative of $\partial y/\partial\omega$ in (6.72), notice the following expression satisfied in the weak field limit:

$$\frac{\partial y(\sigma,t)}{\partial\sigma} = 2y_1(t)\sigma = 2\sigma y_1(0)\frac{V(t)}{U(t)}. \tag{6.81}$$

By exchanging the order of differentiation, its initial value is evaluated by

$$\begin{aligned}\left.\frac{\partial}{\partial\sigma}\frac{\partial y}{\partial\omega}\right|_{\sigma=\sigma_0(t)} &= 2\sigma_0(t)y_1(0)\frac{V(t)}{U(t)}\frac{\partial\log[y_1(0)V(t)/U(t)]}{\partial\omega}\\ &= 2y_1(0)\sigma_0(0)\frac{V(t)}{\sqrt{U(t)}}\left[-\gamma_A+\gamma_0+\frac{1}{V(t)}\frac{\partial V}{\partial\omega}-\frac{1}{U(t)}\frac{\partial U}{\partial\omega}\right],\end{aligned} \tag{6.82}$$

where $y_1(0)\propto T_A/T_0$. In the ordered phase, we need to know the derivatives, $\partial U(t)/\partial\omega$ and $\partial V(t)/\partial\omega$ in (6.79) and (6.82). These values are evaluated by solving the simultaneous differential equations for variables $U(t)$ and $V(t)$ in Chap. 4 and their ω derivatives.

6.5 Magneto-Volume Effect in Some Temperature Ranges

According to our general expressions of the spontaneous and forced magneto-strictions, we show in this section how the effects are described in more detail at low temperatures, around the critical temperature, and at higher temperatures in the paramagnetic phase.

6.5.1 Magneto-Volume Effect at Low Temperature and Grüneisen Relation

At low temperatures where $t \ll 1$ and $y_{z0}(0) \ll 1$ are satisfied, thermal components of the thermal expansion and its temperature coefficient show the following t^2-linear and t-linear dependences, respectively:

$$\begin{aligned}\omega_{th}(t) &\simeq \frac{1}{8}T_0\rho\kappa\gamma_0\{2\log(1/x_c^2) + \log[1/y_{z0}(0)]\}t^2 \\ &\simeq \frac{3}{4}T_0\rho\kappa\gamma_0 t^2\log[1/\sigma_0(0)], \\ \beta_{th}(t) &\simeq \frac{3}{2}\rho\kappa\gamma_0 t\log[1/\sigma_0(0)],\end{aligned} \tag{6.83}$$

where both x_c^2 and $y_{z0}(0)$ are proportional to $\sigma_0^2(0)$. As was already shown in Chap. 5, the magnetic specific heat (5.44) at low temperatures is given by

$$\frac{C_{m0}(t)}{V} \simeq \frac{3}{2}\frac{N_0}{V}t\log[1/\sigma_0(0)] = \frac{3}{2}\rho t\log[1/\sigma_0(0)]. \tag{6.84}$$

It corresponds to the T^2-linear dependence of the free energy:

$$F_m(T) = F_m(0) - \frac{3}{4}N_0\frac{T^2}{T_0}\log[1/\sigma_0(0)] + \cdots, \tag{6.85}$$

for its temperature derivative gives the specific heat in (6.84). The thermal expansion (6.83) is given by the derivative of the above free energy with respect to the strain ω.

$$\omega_{th}(t) = -\frac{\kappa}{V}\frac{\partial F_m(t)}{\partial\omega} = \frac{3}{4}\rho\kappa\gamma_0 t^2\log[1/\sigma_0(0)].$$

From the comparison of (6.83) and (6.84), the following Grüneisen relation between the magnetic specific heat and the thermal volume expansion coefficient is thus satisfied at low temperatures.

$$\beta_{th}(t) = \kappa\gamma_0\frac{C_{m0}(t)}{V} = \frac{3}{2}\rho\kappa\gamma_0 t\log\frac{1}{\sigma_0(0)}. \tag{6.86}$$

The component $\omega_{zp}(t)$ shows similar behavior to those of the SEW theory and the MU theory. It is given in this limit by (6.55), i.e.,

$$\omega_{zp}(t) = \rho\kappa C_s(t)\sigma_0^2(t), \quad \frac{C_s(t)}{C_h(0)} = \frac{2V(t)}{5U(t)}\left[1 + g_A\left(\frac{5}{3}U(t) - 1\right)\right]. \tag{6.87}$$

After substituting the results (4.24) and (4.26) for $V(t)/U(t)$ and $U(t)$ in the above constant $C_s(t)$, we obtain the following temperature dependence.

$$C_s(t) = C_s(0)\left\{1 - \frac{ct^2}{120A^2(0,t_c)} \times \left[\frac{3+2r^2}{4} + \frac{5g_A}{3+2g_A}\frac{4+5r+r^2}{3}\right] + \cdots\right\}, \qquad (6.88)$$

where $r = (\pi/2)^2$.

As the sum of these two contributions, the temperature dependence of the spontaneous volume-striction at low temperatures is finally given by

$$\frac{\omega_m(t)}{\omega_0} = \frac{cg_0}{120A^2(0,t_c)}\left[2\log x_c^{-2} + \log y_{z0}^{-1}\right]t^2 + \frac{cg_0(1-r^2)}{180A^2(0,t_c)}t^2 + \frac{2}{5}\left(1+\frac{2}{3}g_A\right)\left\{1 - \frac{c\,t^2}{120A^2(0,t_c)} \times \left(\frac{3+2r^2}{4} + \frac{3+7g_A}{3+2g_A}\frac{4+5r+r^2}{3}\right) + \cdots\right\}, \qquad (6.89)$$

where the second term in the right hand side results from the thermal free energy correction ΔF_{th}. Because of the above second and the third terms, the t^2-linear coefficient usually becomes negative. The volume change from this origin shows contraction with increasing temperature. For weak itinerant ferromagnets with tiny spontaneous magnetization ($\sigma_0(0) \ll 1$), the positive first term will be also non-negligible. The presence of this $\log[1/\sigma_0(0)]$-linear term is, however, not yet verified experimentally.

Thermal expansion measurements on Ni_3Al and Ni-Pt alloys at low temperatures was made by Kortekaas et al. [23] over the composition ranging from the paramagnets close to the magnetic instability and to the weak ferromagnets. According to their report [23], the temperature dependence of the thermal expansion can be fitted with a sum of T^2-linear term and the T^4-linear term of the lattice vibrations, as given by

$$\Delta\ell/\ell = AT^2 + BT^4, \qquad (6.90)$$

where the length of the sample is denoted by ℓ. In the paramagnetic phase, the coefficient A increases toward the magnetic instability point. Its sign changes from positive to negative across the para- to ferromagnetic transition. They simply assumed that conduction electrons are responsible for the above T^2-linear dependence. However, the observed enhancement of A in the paramagnetic phase seems to suggest that it is caused by the magnetic origin (i.e., by the term $t^2\log[1/y_0(0)]$ to be explained later).

Forced Magneto-Striction at Low Temperatures In the case of weak external magnetic field where $\sigma \simeq \sigma_0(t)$ is satisfied, the temperature dependence of the constant $C_h(t)$ for the forced magneto-striction is generally given by (6.80). The analytic expression of its temperature dependence is available at low temperatures. According to (4.26), $U(t)$ decreases proportional to $T^2/[T_A\sigma_0^2(0)]^2$ with increasing temperature. The temperature dependence of the derivative $\partial U(t)/\partial\omega$ is also given by

$$\frac{\partial U(t)}{\partial \omega} = \frac{4+5r+r^2}{180cA^2(0,t_c)}(\gamma_m - \gamma_A)t^2 + \cdots . \tag{6.91}$$

Substituting (6.91), (4.24) for $V(t)/U(t)$, and (4.26) for $U(t)$ into (6.80), the t^2-linear dependence of $C_h(t)$ is given by

$$\frac{C_h(t)}{C_h(0)} = 1 + \frac{ct^2}{120A^2(0,t_c)}\left[(1-2g_A)\frac{4+5r+r^2}{3} - \frac{3+2r^2}{4}\right] + \cdots . \tag{6.92}$$

6.5.2 Around the Critical Point

The thermal component of the volume expansion in (6.55) at the critical temperature is given by

$$\frac{\omega_{th}(t_c)}{\omega_0} = 3\rho\kappa T_0\gamma_0 t_c^2 \int_0^{1/t_c} \mathrm{d}u\, u\Phi'(u) \simeq \frac{1}{4}\rho\kappa T_0\gamma_0 t_c^2 \log(1/t_c), \quad (t_c \ll 1) \tag{6.93}$$

where $u = x^3/t$. The temperature dependence of $\omega_{th}(t)$ is less affected by those of $y_0(t)$ and $y_{z0}(t)$ around $t = t_c$, as with the case of the specific heat.

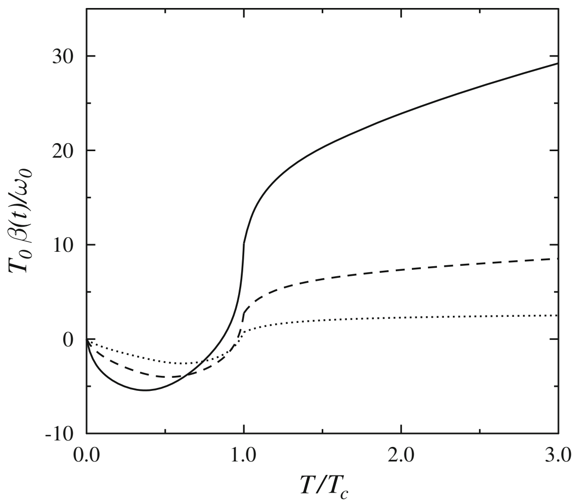
On the other hand, the temperature dependence of the coupling constant $C_s(t)$ and the volume-striction $\omega_{zp}(t)$ are estimated by substituting the t dependence of $U(t)$ and $V(t)/U(t)$ in (4.38) for $t \lesssim t_c$ into (6.87).

$$\begin{aligned}
\frac{C_s(t)}{C_h(0)} &= \frac{14}{25c}(1-g_A)A(0,t_c)\left(\frac{40\sqrt{2}c}{7\pi t_c}\right)^2\left[1-\left(\frac{t}{t_c}\right)^{4/3}\right] + \cdots , \\
\frac{\omega_{zp}(t)}{\omega_0} &= \frac{98}{125c}(1-g_A)A(0,t_c)\left(\frac{40\sqrt{2}c}{7\pi t_c}\right)^2\left[1-\left(\frac{t}{t_c}\right)^{4/3}\right]^2 + \cdots .
\end{aligned} \tag{6.94}$$

They both vanish at the critical point in proportion to $(T - T_c)$ and $(T - T_c)^2$. The thermal expansion coefficient $\beta_{zp}(t)$ is therefore proportional to $(T - T_c)$. Contrary to this result, both the SEW and MU theories give a finite negative value of $\beta(t)$ in the limit $t \to t_c$, reflecting the temperature dependence of $M_0^2(T) \propto (T_c - T)$.

The temperature dependence is also estimated by (6.60) around t_c in the paramagnetic phase. The dependence of $U(t)$ and $V(t)/U(t)$ are then given by

Fig. 6.2 Temperature dependence of the thermal expansion coefficient



$$U(t) = \frac{1}{2}[(t/t_c)^{4/3} - 1], \quad \frac{V(t)}{U(t)} = 2c\left(\frac{4}{\pi t_c}\right)^2 A(0, t_c)[(t/t_c)^{4/3} - 1]. \tag{6.95}$$

Substituting these results into (6.60) gives

$$\begin{aligned} \frac{\omega_{zp}(t)}{\omega_0} &= \frac{6c}{10}(1 - g_A)\left(\frac{4}{\pi t_c}\right)^2 A(0, t_c)[(t/t_c)^{4/3} - 1]^2, \\ \frac{C_s(t)}{C_h(0)} &= \frac{6c}{5}(1 - g_A)\left(\frac{4}{\pi t_c}\right)^2 A(0, t_c)[(t/t_c)^{4/3} - 1]. \end{aligned} \tag{6.96}$$

Both the MU and SEW theories predict the discontinuous change in the slope of the temperature dependence of the spontaneous magneto-striction $\omega_m(t)$, from the negative to the positive value (MU) and from the negative to zero (SEW), with increasing temperature. The above results of (6.94) and (6.96) predict the continuous change. The difference results from the temperature dependence of our magneto-volume coupling constant $C_s(t)$. Both the experiments of thermal expansion coefficient on $ZrZn_2$ by Ogawa, Kasai [24] and by Creuzet et al. [25] seem to support the continuous change. We show in Fig. 6.2, numerical results of the thermal expansion coefficient in the wide range of temperature from the order phase to the paramagnetic phase. The solid, dashed, and doted lines correspond to $t_c = 0.05, 0.1, 0.2$, respectively, for $g_0 = 0.1$ and $g_A = 0.1$.

Forced Magneto-Striction Around the Critical Point The temperature dependence of the forced magneto-volume coupling constant $C_h(t)$ is also evaluated by (6.80). The first term proportional to $V(t) \propto (t_c - t)^2$ is neglected since it is higher order than the second. The derivative $\partial U(t)/\partial\omega$ at the critical point is evaluated by using the temperature dependence of (4.38) for $U(t)$.

$$\left.\frac{\partial U(t)}{\partial \omega}\right|_{T=T_c} = \frac{28}{15}\left(\frac{T}{T_c}\right)^{4/3}\frac{\mathrm{d}\log T_c}{\mathrm{d}\omega} \simeq \frac{28}{15}\frac{\mathrm{d}\log T_c}{\mathrm{d}\omega}. \tag{6.97}$$

Putting the above result and (4.38) for $V(t)/U(t)$ into (6.80), the temperature dependence of $C_h(t)$ is given by

$$\begin{aligned}\frac{C_h(t)}{C_{h0}} &\simeq \frac{V(t)}{\gamma_m U(t)}\frac{\partial U(t)}{\partial \omega} \\ &= \frac{32c}{3}\left(\frac{4}{\pi t_c}\right)^2 A(0,t_c)\frac{1}{\gamma_m}\frac{\mathrm{d}\log T_c}{\mathrm{d}\omega}\left[1-\left(\frac{t}{t_c}\right)^{4/3}\right]+\cdots.\end{aligned} \tag{6.98}$$

It means that we can estimate the value of $\mathrm{d}\log T_c/\mathrm{d}\omega$ experimentally from the observed slope of the coupling constant $C_h(t)$ against $(T - T_c)$ around the critical temperature. We will show later, the value is represented in terms of γ_m, γ_0, and γ_A.

In the paramagnetic phase, the temperature dependence of $C_h(t)$ is evaluated by (6.78). Higher order term proportional to $V(t)$ is also neglected in this case. By putting (6.95) for $V(t)/U(t)$ into (6.78), the temperature dependence of $C_h(t)$ is given by

$$\frac{C_h(t)}{C_{h0}} = 2c(1-g_A+g_0)\left(\frac{4}{\pi t_c}\right)^2 A(0,t_c)\left[\left(\frac{t}{t_c}\right)^{4/3}-1\right]. \tag{6.99}$$

To summarize, the forced magneto-volume coupling constant $C_h(t)$ also decreased in proportion to $|T - T_c|$ toward the critical point in the same way as $C_s(t)$ for the spontaneous striction.

Critical Forced Magneto-Striction We have already shown in Chap. 3 that the magnetic isotherm at the critical point exhibits the anomalous behavior under the influence of critical spin fluctuations. The same behavior is also expected for the forced magneto-striction, because it is given by the volume derivative of the same free energy. The critical forced magneto-striction can be treated according to the general formula in (6.70). Both the σ dependence of $y(\sigma,t)$ and the ω-derivative $\partial y(\sigma,t)/\partial\omega$ are then necessary. These are determined by solving the simultaneous differential equations (6.71) and (6.72).

Substituting the critical behaviors, $A'(y,t) \propto 1/\sqrt{y}$ and $A'(y_z,t) \propto 1/\sqrt{y_z}$, for the thermal fluctuation amplitudes, (6.72) is written by

$$-\frac{\pi t_c}{8}\left(\frac{2}{\sqrt{y}}\frac{\partial y}{\partial \omega}+\frac{1}{\sqrt{y_z}}\frac{\partial y_z}{\partial \omega}\right) = 3A(0,t_c)\gamma_m(1-g_A-g_0), \tag{6.100}$$

where the higher order terms with respect to σ^2 are neglected. At the critical point, both $y(\sigma,t_c)$ and $y_z(\sigma,t_c)$ are proportional to σ^4, as was already shown in Chap. 3. Then the derivative $\partial y(\sigma,t_c)/\partial\omega$ has to be proportional to σ^2, and therefore $\partial y_z(\sigma,t_c)/\partial\omega = 3\partial y(\sigma,t_c)/\partial\omega$ is derived from the relation between $y(\sigma,t)$ and

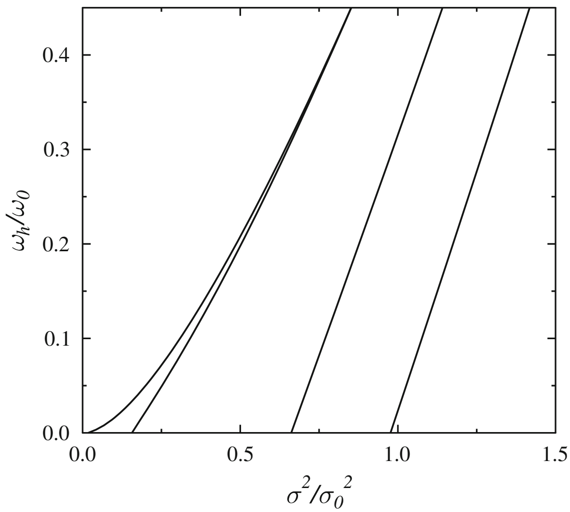


Fig. 6.3 Numerically estimated forced magneto-striction at temperatures $T/T_c = 0.10, 0.50, 0.90, 0.99$ from the right for $T_c/T_0 = 0.05$

$y_z(\sigma, t)$. The σ^2-linear coefficient of $\partial y(\sigma, t_c)/\partial\omega$ is determined as follows:

$$\frac{1}{\gamma_m}\frac{\partial y(\sigma, t_c)}{\partial \omega} = -\frac{24\sqrt{5}}{3+2\sqrt{5}}(1-g_A-g_0)\frac{\sqrt{y_c}}{\pi t_c}A(0, t_c)\sigma^2.$$

Substituting the result into (6.70) finally leads to the critical forced magneto-striction given by

$$\frac{\omega_h(\sigma, t_c)}{\omega_0} = \frac{12\sqrt{5}}{3+2\sqrt{5}}(1-g_A-g_0)\frac{\sqrt{y_c}}{\pi t_c y_1(0)}A(0, t_c)\frac{\sigma^4}{\sigma_0^4(0)}. \qquad (6.101)$$

We show in Fig. 6.3, the numerically estimated σ dependence of the forced magneto-striction in the ordered phase by solving the simultaneous differential equations (6.71), (6.72), and (6.74). Relative volume-strictions $\omega_h(\sigma, t)/\omega_0$ at temperatures, $T/T_c = 0.10,\ 0.50,\ 0.90,\ 0.99$, are plotted against $\sigma^2/\sigma_0^2(0)$. At low temperatures, good linearity is observed because of the weak σ dependence of the coupling constant $C_h(\sigma, t)$. Since the coupling constant $C_h(t)$ decreases to zero toward the critical point in accordance with (6.98), the σ^4-linear behavior is expected to emerge around the critical temperature. The behavior is actually observed in Fig. 6.3 as the result at $T/T_c = 0.99$. It is evident from this figure that the σ^2-linear behavior at low temperatures changes to the critical σ^4-linear behavior with increasing temperature.

Forced Magneto-Striction Observed in MnSi In the field of itinerant electron magnetism, not enough attention have long been payed on the concept of the critical magnetic isotherm. The same is true for the critical forced magneto-striction. Although the anomalous forced magneto-striction seemed to be observed in MnSi at the critical temperature, it did not attract much attention until Takahashi [26] pointed

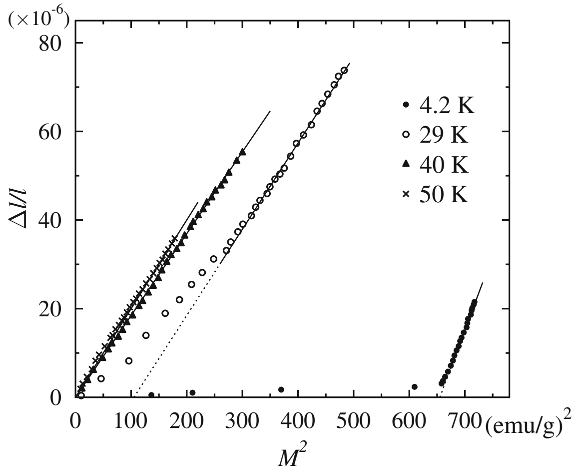


Fig. 6.4 Observed forced magneto-striction in MnSi (Matsunaga et al. [6])

out its relevance to the critical forced magneto-striction. We show in Fig. 6.4 the forced magneto-striction of MnSi observed by Matsunaga et al. cited from Fig. 8 of [6]. In this figure, observed forced-strictions (relative changes of the length of the sample, $\Delta\ell/\ell$) are plotted against M^2. The plot considerably deviates from the linearity around the critical temperature $T_c \simeq 30$ K. The good linearity is, however, confirmed by plotting the data against M^4 at $T = 29$ K. There seem to be no other observed critical forced magneto-striction at present.

6.5.3 In the Paramagnetic Phase

Spontaneous Magneto-Striction The magneto-volume effect observed at higher temperatures in the paramagnetic phase, where the Curie-Weiss law temperature dependence of the magnetic susceptibility is observed, is discussed in this section. In the region where the Curie-Weiss law of the inverse of the magnetic susceptibility in (3.44), i.e., $y_0(t) \simeq 2(t-t_c)/[5cy_1(0)p_{\rm eff}^2]$, is satisfied, the temperature dependence of $y_1(t)$ is negligible. Then $V(t)/U(t) = y_1(t)/y_1(0)$ is almost independent of temperature and $V(t)$ is given by

$$V(t) = \frac{y_0(t)}{y_1(0)\sigma_0^2(0)} \simeq \frac{c}{10A^2(0,t_c)}\frac{p_s^2}{p_{\rm eff}^2}(t-t_c), \tag{6.102}$$

by using $A(0,t_c) = cy_1(0)\sigma_0^2(0)$. According to (6.60), the ratio $C_s(t)/C_h(0)$, as given below, is about $3(1-g_A)/5$.

$$\frac{C_s(t)}{C_h(0)} = \frac{3}{5}(1-g_A)\frac{y_1(t)}{y_1(0)} \simeq \frac{3}{5}(1-g_A). \tag{6.103}$$

The temperature dependence of the thermal expansion $\omega_{zp}(t)$ in the same (6.60) is given by

$$\begin{aligned} \frac{\omega_{zp}(t)}{\omega_0} &= \frac{C_s(t)}{C_h(0)}\frac{y_0(t)}{y_1(t)\sigma_0^2(0)} = \frac{3}{5}(1-g_A)V(t) \\ &\simeq \frac{3(1-g_A)c}{50A^2(0,t_c)}\frac{p_s^2}{p_{\text{eff}}^2}(t-t_c). \end{aligned} \tag{6.104}$$

The thermal expansion coefficient then becomes almost temperature independent as given by

$$\frac{T_c\beta_{zp}(t)}{\omega_0} = \frac{T_c}{\omega_0 T_0}\frac{\mathrm{d}\omega_{zp}(t)}{\mathrm{d}t} \simeq \frac{3}{50}\frac{c(1-g_A)t_c}{A^2(0,t_c)}\frac{p_s^2}{p_{\text{eff}}^2} = \frac{27c(1-g_A)}{50(C_{4/3})^2 t_c^{5/3}}\frac{p_s^2}{p_{\text{eff}}^2}. \tag{6.105}$$

Note that the close relation is satisfied between the ratio of moments p_{eff}/p_s and $t_c = T_c/T_0$ as shown in Sect. 3.3.4. The right hand side of (6.105) is determined by the single parameter t_c.

The validity of (6.105) can be confirmed experimentally. The value of $\beta_{zp}(t)$ in the paramagnetic phase is determined by extracting the temperature independent component from the observed thermal expansion coefficient. The value of ω_0 is estimated from the observed forced magneto-volume constant $C_h(0)$ at low temperatures and the spontaneous magnetization squared $\sigma_0^2(0)$. It is, however, not so easy to extract the magnetic contribution from the total volume expansion by subtracting those from the lattice vibrations and etc. The value of $T_c\beta/\omega_0$ estimated in this way by using available data from references are plotted against the ratio T_c/T_0 in Fig. 6.5. In the same figure, numerically estimated values of the right hand side of (6.105) is plotted by the solid curve. Though the factor $(1-g_A)$ is not included in the plot, raw experimental data from references are employed.

The figure shows that solid circles of experiments fall fairly close to the theoretical curve. According to (6.103) and (6.104), the ratio $T_c\beta/\omega_0$ is closely related to the coupling ratio $C_s(t)/C_h(0)$. The observed data in the figure also support the theoretical prediction for the ratio smaller than 1.

Forced Magneto-Striction We have already shown in Sect. 6.4 that the forced magneto-striction in the paramagnetic phase is given by $\omega_h(t) = \rho\kappa C_h(t)\sigma^2$, and the temperature dependence of the coupling constant $C_h(t)$ is described by (6.78). The value of $C_h(t)$ has the general tendency to saturate with increasing temperature in the paramagnetic phase. In cases with non-negligible size of g_A, however, it will show a slight increase, because of the presence of $(t-t_c)$-linear term of $U(t)$ in this (6.78).

According to (6.75), the volume derivative $\partial y_0(t)/\partial\omega$ is necessary to evaluate the coupling constant $C_h(t)$. The value of this derivative is also closely related to

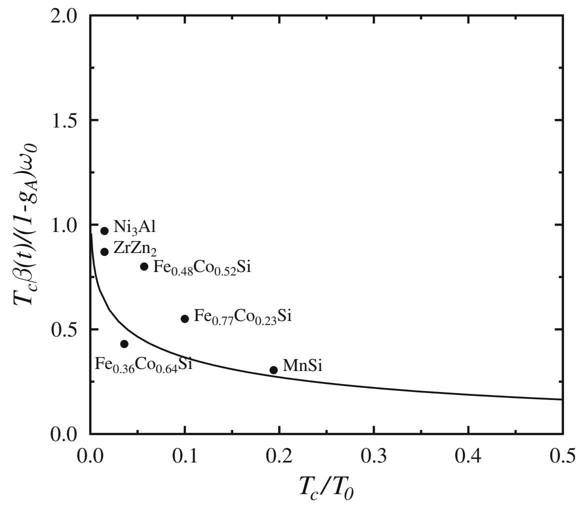


Fig. 6.5 Observed thermal expansion coefficients in the paramagnetic phase versus T_c/T_0 by Takahashi and Nakano [20]

the pressure effect measurements of the paramagnetic susceptibility by Brommer et al. [27]. They reported that the temperature dependence of the derivative $\mathrm{d}\log\chi(T)/\mathrm{d}\omega$ for Ni_3Al and TiCo is proportional to the magnetic susceptibility $\chi(T)$, i.e., $\mathrm{d}\log\chi(T)/\mathrm{d}\omega \propto \chi(T)$. In other words, the value of $\chi^{-2}(T)\mathrm{d}\chi(T)/\mathrm{d}\omega$, and therefore $\mathrm{d}\chi^{-1}(T)/\mathrm{d}\omega$ is independent of temperature, being in agreement with (6.77). Values of $\mathrm{d}\log\chi(T)/\mathrm{d}\log V$ for Ni_3Al observed by them at three temperatures are shown in Fig. 6.6 against $\chi(T)$. They fall on a straight line with a positive slope as shown in this figure. The slope of the figure is also represented in our

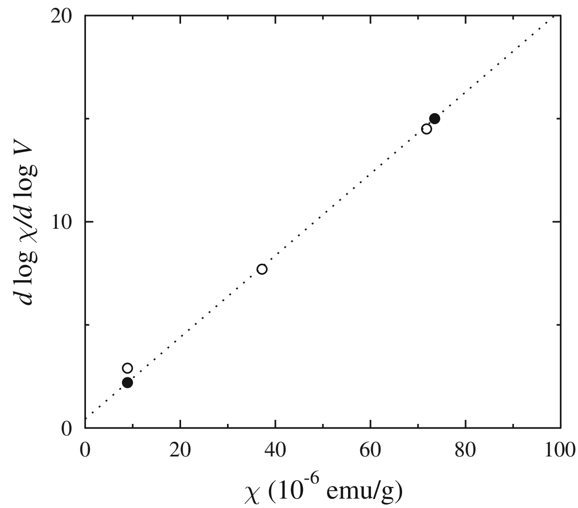


Fig. 6.6 Pressure effect on paramagnetic susceptibility of Ni_3Al by Brommer et al. (*solids circles* are results by levitation method)

theoretical notations as

$$\frac{N_0}{2\chi}\frac{\mathrm{d}\log\chi}{\mathrm{d}\ln V} = -T_A y_0(t)\frac{\partial \log y_0(t)}{\partial\omega} = -T_A\frac{\partial y_0(t)}{\partial\omega} = T_A y_1(t)\sigma_0^2(0)\frac{\mathrm{d}\log A(0,t_c)}{\mathrm{d}\omega} \tag{6.106}$$

The value of the above left hand side is estimated to be 2.73×10^3 K for $Ni_{74.8}Al_{25.2}$ from the observed data by Brommer et al. Spectral parameters of spin fluctuations in this compound are already estimated to be $T_0 \simeq 3\times 10^3$ K and $T_A \simeq 3\times 10^4$ K, giving $y_1(t) \simeq y_1(0) \simeq 1/3$. The volume-striction in the right-hand side is also estimated to be

$$\frac{\mathrm{d}\log A(0,t_c)}{\mathrm{d}\omega} = -B\frac{\mathrm{d}\log A(0,t_c)}{\mathrm{d}p} = -B\frac{\mathrm{d}\log\sigma_0^2(0)}{\mathrm{d}p} \simeq 46.2, \tag{6.107}$$

where $B = 1.7$ M bar as a bulk modulus and $\mathrm{d}\log\sigma_0^2(0)/\mathrm{d}p = 27.2$. Effects of γ_0 and γ_A are neglected as a rough estimate. If we finally assume $\sigma_0(0) = 0.05$ or 0.07 as the spontaneous magnetization, the right hand side of (6.106) is given by 1.15×10^3 K or 2.26×10^3 K, respectively, in nearly close agreement with the estimate by Brommer et al.

6.5.4 Numerical Results on Volume-Strictions

Numerical results of the temperature dependence of spontaneous magneto-volume striction by Takahashi and Nakano [20] are shown in Fig. 6.7. Dashed, dotted, and solid lines correspond to the components $\omega_{th}(t)$, $\omega_{zp}(t)$ of the thermal expansion, and the sum of the both, respectively, for $t_c = 0.01, 0.05, 0.1$, in descending order from the top. It is interesting to notice that the relative ratio of the thermal fluctuation component to the total thermal expansion becomes larger for smaller value of t_c. It will cancel the increase of $\omega_{zp}(t)$ below the critical temperature with decreasing temperature. Thermal expansion will then become monotonically increasing function. Note that the relative volume-striction divided by ω_0 is plotted in this figure. The smaller the value of t_c, the value of ω_0 becomes smaller. The magnitude of this figure is nothing to do with the absolute value of the thermal expansion.

The enhancement of the thermal expansion coefficients at low temperatures is shown in Fig. 6.8. The t-linear coefficient of the thermal expansion coefficient, $[\beta_t(t) + \Delta\beta(t)]/3\rho\kappa\gamma_0 T$, is plotted against T/T_c in this figure. Solid, dashed, dot-dashed, and dotted curves from the top corresponds to $t_c = 0.005, 0.01, 0.05, 0.1$, respectively. The value of $\sigma_0(0)$ increases in this order, whereas the enhancement decreases inversely. We finally show in Fig. 6.9, the temperature dependence of the spontaneous (thin lines) and the forced (thick lines) magneto-coupling constants,

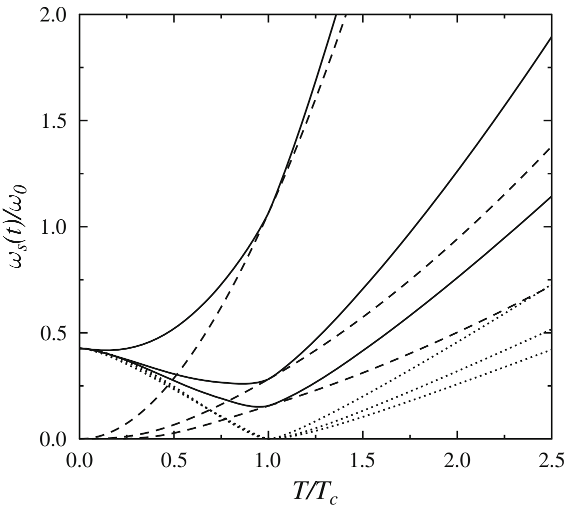


Fig. 6.7 Numerically estimated temperature dependence of spontaneous magneto-striction, for $g_0 = g_A = 0.1$ and $T_A/T_0 = 10$

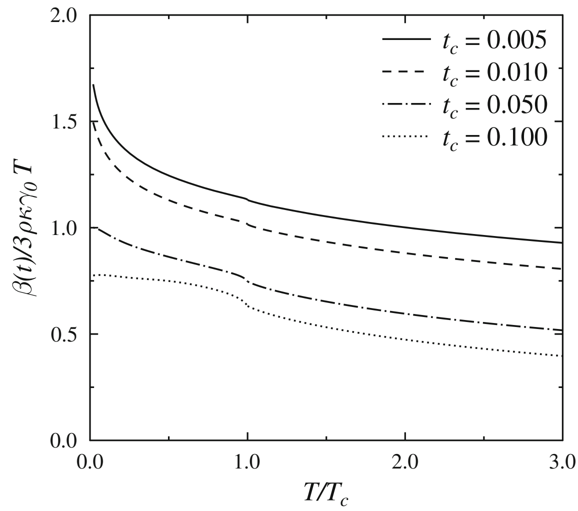


Fig. 6.8 Enhancement of the t-linear coefficient of the thermal expansion coefficient at low temperatures [20]

$C_s(t)$ and $C_h(t)$. Solid, dashed, and dotted curves correspond to $t_c = 0.01, 0.05, 0.1$, respectively.

6.6 Magneto-Volume Effect for Paramagnets

From the similarity between magnetic isotherms for ferromagnets and paramagnets near the magnetic instability point, we show in Chap. 3, that the value of $\sigma_p^2(0) \equiv y_0(0)/y_1(0)$ defined in (3.21) for paramagnets corresponds to the spontaneous

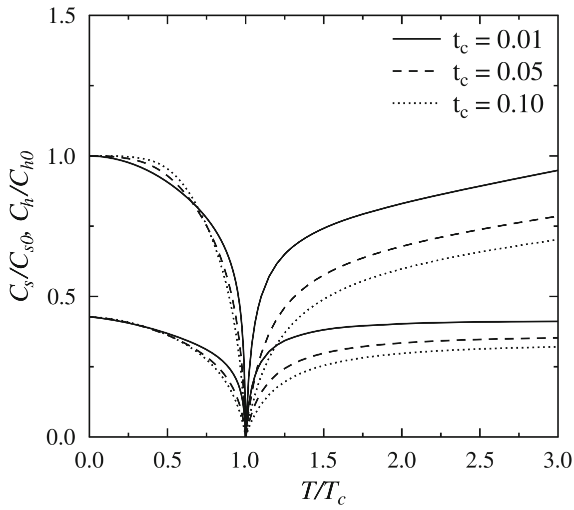


Fig. 6.9 Temperature dependence of magneto-volume coupling constants, $C_s(t)$ and $C_h(t)$, for $T_A/T_0 = 10$ [20]

magnetic moment squared $\sigma_0^2(0)$ for ferromagnets. From the same analogy, the Grüneisen parameter γ_m for paramagnets is also defined by

$$\Delta\left\langle S_{\text{loc}}^2\right\rangle = -\frac{3}{5}\sigma_p^2(0), \quad \frac{\mathrm{d}\Delta\left\langle S_{\text{loc}}^2\right\rangle}{\mathrm{d}\omega} = \frac{3}{5}\gamma_m\sigma_p^2(0). \tag{6.108}$$

The negative value of $\Delta\left\langle S_{\text{loc}}^2\right\rangle$ is characteristic to paramagnets. Corresponding to the definitions of the coupling constant $C_h(0)$ and ω_0 for ferromagnets, (6.50) and (6.61), the same parameters can be defined by

$$C_{h0} = T_A y_0(0)\gamma_m, \quad \omega_0 = \rho\kappa C_{h0}\sigma_p^2(0). \tag{6.109}$$

Note, however, the above forced magneto-volume coupling C_{h0} is slightly different from the value $C_h(0)$ in the ground state ($t = 0$), as will be shown later. We also define the reduced parameters $V(t)$ and $U(t)$ by

$$V(t) = \frac{y_0(t)}{y_0(0)}, \quad U(t) = \frac{y_0(t)}{y_0(0)}\frac{y_1(0)}{y_1(t)} = \frac{\sigma_p^2(t)}{\sigma_p^2(0)} \tag{6.110}$$

as scaled values of $y_0(t)$ and $\sigma_p^2(t)$. In the next subsection, we first deal with the temperature dependence of the spontaneous magneto-striction, followed by the forced magneto-volume striction.

6.6.1 Spontaneous Magneto-Striction for Paramagnets

Along with the case of ferromagnets, the thermal component of the volume-strain in this case is also obtained by (6.55), except for $u_z = u$ because of the absence of the spontaneous magnetization. The component $\omega_{zp}(t)$ is also evaluated, according to the general definition (6.35) and (6.36). The coefficient C_{zp} is evaluated by the volume derivative of the free energy F_{zp}, the volume dependence of which is characterized by the Grüneisen parameters defined in (6.108) and (6.37). They are given by

$$\begin{aligned}
\frac{\omega_{th}(t)}{\omega_0} &= \frac{3g_0 t}{5cy_0^2(0)} \int_0^1 \mathrm{d}x x^2 u \Phi'(u), \\
\omega_{zp}(t) &= 3\rho\kappa C_{zp} y_0(t) = \frac{3}{5}\rho\kappa C_{h0}\sigma_p^2(0)(1+g_A)\frac{y_0(t)}{y_0(0)} \\
&= \frac{3}{5}\omega_0(1+g_A)V(t), \\
C_{zp} &= \frac{1}{3}\frac{\partial}{\partial\omega}\left[T_A \Delta \left\langle S_{\mathrm{loc}}^2 \right\rangle\right] = \frac{1}{5}T_A\sigma_p^2(0)(\gamma_m + \gamma_A).
\end{aligned} \tag{6.111}$$

These results in (6.111) correspond to (6.55) for ferromagnets. We cannot define the magneto-volume coupling constant literally for paramagnets with no spontaneous magnetic moment. We have, however, intentionally defined the coefficient $C_s(t)$ from the similarity with ferromagnets.

$$\omega_{zp}(t) = \rho\kappa C_s(t)\sigma_p^2(t), \quad \frac{C_s(t)}{C_{h0}} = \frac{3V(t)}{5U(t)}(1+g_A). \tag{6.112}$$

The thermal expansion coefficient is also given by the temperature derivative of (6.111).

$$\begin{aligned}
\frac{1}{\omega_0}\frac{\mathrm{d}\omega_m(t)}{\mathrm{d}t} &= \bar{\beta}(t) = \bar{\beta}_{th}(t) + \bar{\beta}_{zp}(t), \\
\bar{\beta}_{th}(t) &= \frac{g_0}{5c_z y_0^2(0)}\left\{-3\int_0^1 \mathrm{d}x x^2 u^2 \Phi''(u)\right. \\
&\quad \left. + 2y_0(0)\frac{\mathrm{d}V(t)}{\mathrm{d}t}\left[A(y_0,t) - t\frac{\partial A(y_0,t)}{\partial t}\right]\right\}, \\
\bar{\beta}_{zp}(t) &= \frac{3}{5}(1+g_A)V'(t).
\end{aligned} \tag{6.113}$$

In analogy with (6.83) for ferromagnets in the ordered phase, the thermal component of the volume expansion $\omega_{th}(t)$ at low temperatures is approximated by

$$\omega_{th}(t) = \frac{3}{8}\rho\kappa\gamma_0 t^2 \log y_0^{-1}(0) + \cdots . \tag{6.114}$$

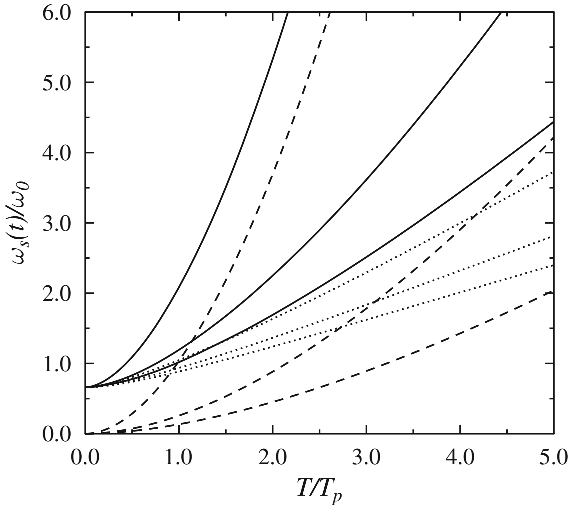


Fig. 6.10 Temperature dependence of magneto-volume strictions of paramagnets

On the other hand, $\omega_{zp}(t)$ is given by

$$\frac{\omega_{zp}(t)}{\omega_0} = \frac{3}{5}(1 + g_A)\left[1 + \frac{t^2}{24cy_0^2(0)} + \cdots\right]. \tag{6.115}$$

by putting the temperature dependence of $y_0(t)$ into (6.111). The total magneto-volume striction is therefore given by

$$\frac{\omega_m(t)}{\omega_0} = \frac{3}{5}(1 + g_A) + \frac{t^2}{40cy_0^2(0)}[g_0 \log y_0^{-1}(0) + 1 + g_A] + \cdots. \tag{6.116}$$

Nearly the same behavior is thus expected at higher temperatures, independent of ferro- and paramagnets, where the Curie-Weiss law of the magnetic susceptibility is observed.

In Fig. 6.10, numerically estimated temperature dependence of the magneto-volume strictions of (6.111) is shown. The results for components, $\omega_{th}(t)$ and $\omega_{zp}(t)$, and the sum of them are shown by dashed, dotted, and solid curves, respectively, for $t_p = 0.01, 0.05, 0.10$ from the top in descending order. The numerical results for the t-linear coefficient of the thermal expansion coefficient, i.e., $\beta(t)/3\rho\kappa\gamma_m T$, are also shown in Fig. 6.11. The enhancement of this t-linear coefficient at low temperatures in this figure results from the factor $\log y_0^{-1}(0)$ in (6.114).

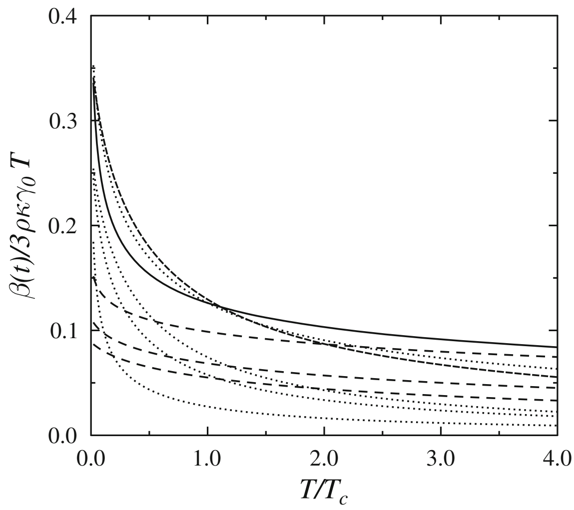


Fig. 6.11 Temperature dependence of $\beta(t)/3\rho\kappa\gamma_m T$ with the same parameters t_P as Fig. 6.10

6.6.2 Forced Magneto-Striction for Paramagnets

Forced magneto-volume striction $\omega_h(\sigma, t)$ is generally given by the σ derivative of (6.46). In the weak external magnetic field limit, $\omega_h(\sigma, t) = \rho\kappa C_h(t)\sigma^2$ is satisfied with coupling constant $C_h(t)$ in (6.75). The ω-derivative $\partial y_0(t)/\omega$ in this equation is evaluated by differentiating (3.30) with respect to ω, i.e.,

$$A(y_0, t) - c_z y_0(t) = -cy_0(0) = -A(0, t_p), \tag{6.117}$$

for paramagnets. It is given by

$$\begin{aligned}[A'(y_0, t) - c]\frac{\partial y_0(t)}{\partial\omega} &= -\frac{cy_1(0)}{y_1(t)}\frac{\partial y_0(t)}{\partial\omega} \\ &= -c\frac{\partial y_0(0)}{\partial\omega} = c(\gamma_m + \gamma_A - \gamma_0)y_0(0),\end{aligned} \tag{6.118}$$

with the use of (3.50) for $y_1(t)$. The right hand side is derived from the ω derivative of the relation $y_0(0) = T_A\sigma_p^2(0)/15T_0$ in (3.21).

$$\frac{1}{y_0(0)}\frac{\mathrm{d}y_0(0)}{\mathrm{d}\omega} = \frac{\mathrm{d}\log y_0(0)}{\mathrm{d}\omega} = -\gamma_m - \gamma_A + \gamma_0. \tag{6.119}$$

The derivative $\partial y_0(t)/\omega$ is therefore finally given by

$$\frac{\partial y_0(t)}{\partial\omega} = -\frac{y_1(t)}{y_1(0)}(\gamma_m + \gamma_A - \gamma_0)y_0(0). \tag{6.120}$$

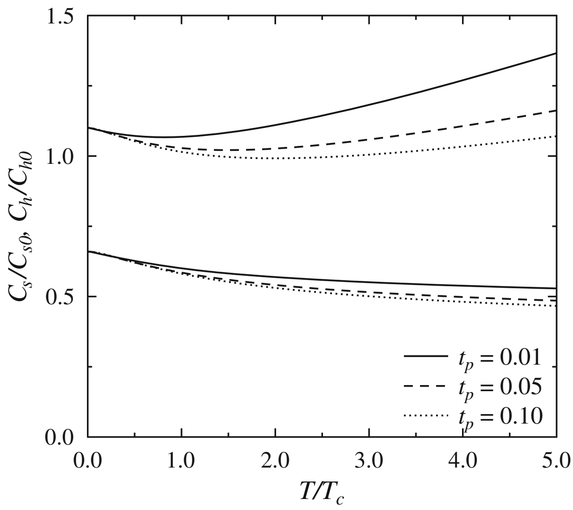


Fig. 6.12 Numerically estimated temperature dependence of the magneto-volume coupling constants $C_s(t)$ and $C_h(t)$ for paramagnets for $t_p = 0.01,\ 0.05$, and 0.10

It corresponds to (6.77) for ferromagnets. Substitution of (6.120) into (6.75) gives

$$\frac{C_h(t)}{C_{h0}} = V(t)\left[g_A + (1 + g_A - g_0)\frac{1}{U(t)}\right] = \frac{V(t)}{U(t)}\{1 + g_A[1 + U(t)] - g_0\} \tag{6.121}$$

Numerically estimated results of (6.112) for $C_s(t)$ and (6.121) for $C_h(t)$ are shown in Fig. 6.12.

6.7 Pressure Effects on Spontaneous Magnetic Moment and the Critical Temperature

We mentioned, at the beginning of this chapter, that the volume change of magnets induces changes of their spontaneous magnetic moment $\sigma_0(0)$ in the ground state and the Curie temperature T_c. According to the definition of the Grüneisen parameter in (6.39), the volume change of $\sigma_0(0)$ is characterized by the parameter γ_m. In this last section, we first show how the volume dependence of the critical temperature T_c is described in terms of Grüneisen parameters.

6.7.1 Effect of Pressure on the Critical Temperature

The critical temperature is determined by the condition, $y_0(t_c) = 0$, for the inverse of the magnetic susceptibility. Along with the SEW theory in Sect. 6.3.1, the change of the critical temperature δT_c against the volume change can be determined by this condition. Let us first note the following relation between $\delta\omega$ and δT_c derived by the condition:

$$\left.\frac{\partial y_0(t)}{\partial t}\right|_{t=t_c}\left(\frac{\delta T_c}{T_0}-\frac{T_c}{T_0^2}\delta T_0\right)+\left.\frac{\partial y_0(t)}{\partial \omega}\right|_{t=t_c}\delta\omega=0. \tag{6.122}$$

With the use of (3.30) for $y_0(t)$, the above two partial derivatives of $y_0(t)$ can be represented as (5.32) and (6.77), i.e.,

$$\frac{\partial y_0(t)}{\partial t}=\frac{y_1(t)}{cy_1(0)}\frac{\partial A(y_0,t)}{\partial t},\quad \frac{\partial y_0(t)}{\partial \omega}=-\frac{y_1(t)}{cy_1(0)}\frac{\partial A(0,t_c)}{\partial \omega}. \tag{6.123}$$

They are given by the partial derivatives of (3.30) with respect to t and ω, respectively. Equation (6.122) is then written in the form

$$\frac{\partial A(0,t_c)}{\partial t_c}\frac{T_c}{T_0}\left(\frac{\delta T_c}{T_c}-\frac{\delta T_0}{T_0}\right)-\frac{\partial A(0,t_c)}{\partial \omega}\delta\omega=0,$$

where the limit $t\to t_c$ is taken after dividing the both sides by $y_1(t)/cy_1(0)$. Substituting (6.73) for the derivative $\partial A(0,t_c)/\partial\omega$, (6.122) is given by

$$t_c\frac{\partial A(0,t_c)}{\partial t_c}\left(\frac{\mathrm{d}\log T_c}{\mathrm{d}\omega}+\gamma_0\right)=(\gamma_m-\gamma_A+\gamma_0)A(0,t_c).$$

In the above left-hand side, the following relation is satisfied, because of $A(0,t_c)\propto t_c^{4/3}$ in (3.21) for $t_c\ll 1$.

$$t_c\frac{\partial A(0,t_c)}{\partial t_c}=\frac{4}{3}A(0,t_c).$$

As a result, the following relation is satisfied for the volume effect on the critical temperature T_c.

$$\frac{4}{3}\frac{\mathrm{d}\log T_c}{\mathrm{d}\omega}=\gamma_m-\gamma_A-\frac{1}{3}\gamma_0,\quad \frac{\mathrm{d}\log\sigma_0^2(0)}{\mathrm{d}\omega}=\gamma_m. \tag{6.124}$$

The definition of the parameter γ_m is also shown for reference.

The above result (6.124) is equivalent to the relation (3.11) in Chap. 3, i.e.,

$$\sigma_0^2(0)=\frac{5C_{4/3}T_0}{T_A}\left(\frac{T_c}{T_0}\right)^{4/3}, \tag{6.125}$$

which is satisfied between $t_c=T_c/T_0$ and $\sigma_0^2(0)$, irrespective of the volume change. The same relation as (6.124) is derived from the volume derivative of the both sides of (6.125). Note that multiple Grüneisen parameters are involved in (6.124). The result is reasonable, because phase transitions at finite temperatures are affected by spin fluctuations, the time dependence and the spatial variation of which are characterized by parameters γ_0 and γ_A.

As the effect of external pressure, (6.124) can be written in the form

$$\begin{aligned}
\frac{4}{3}\frac{\mathrm{d}\log T_c}{\mathrm{d}p} &= -\frac{4}{3}\kappa\frac{\mathrm{d}\log T_c}{\mathrm{d}\omega} = -\kappa(\gamma_m - \gamma_A - \gamma_0/3),\\
\frac{\mathrm{d}\log\sigma_0^2(0)}{\mathrm{d}p} &= -\kappa\gamma_m.
\end{aligned} \tag{6.126}$$

by introducing the compressibility κ. It is also rewritten as

$$\frac{\mathrm{d}\log T_c}{\mathrm{d}p} - \frac{3}{4}\frac{\mathrm{d}\log\sigma_0^2(0)}{\mathrm{d}p} = \frac{\kappa}{4}(3\gamma_A + \gamma_0) \equiv \kappa\gamma_{0,A}, \tag{6.127}$$

by eliminating the parameter γ_m from them. We have already shown in (3.13), the fourth expansion coefficient F_1 of the free energy in powers of the magnetization M is expressed in terms of spectral parameters T_0 and T_A. The pressure effect on F_1 is then given by

$$\frac{\mathrm{d}\log F_1}{\mathrm{d}p} = 2\kappa\gamma_A - \kappa\gamma_0. \tag{6.128}$$

We can estimate the value of F_1 experimentally from the slope of the Arrott plot of the observed magnetization curve. From the slope of its pressure dependence against the pressure, the pressure derivative of F_1 is estimated. As solutions of a simultaneous equation of (6.127) and (6.128), parameters γ_0 and γ_A are now represented as follows:

$$\begin{aligned}
\kappa\gamma_A &= \frac{4}{5}\frac{\mathrm{d}\log T_c}{\mathrm{d}p} - \frac{3}{5}\frac{\mathrm{d}\log\sigma_0^2(0)}{\mathrm{d}p} + \frac{1}{5}\frac{\mathrm{d}\log F_1}{\mathrm{d}p},\\
\kappa\gamma_0 &= \frac{8}{5}\frac{\mathrm{d}\log T_c}{\mathrm{d}p} - \frac{6}{5}\frac{\mathrm{d}\log\sigma_0^2(0)}{\mathrm{d}p} - \frac{3}{5}\frac{\mathrm{d}\log F_1}{\mathrm{d}p}.
\end{aligned} \tag{6.129}$$

In order to evaluate the magnetic Grüneisen parameters experimentally, the value of γ_m in (6.126) is estimated from the slope of the variation of $\sigma_0^2(0)$ against the pressure p. For the rest of parameters, γ_A and γ_0 in (6.129), additional pressure effect measurements of T_c and F_1 are needed.

One of the distinct features of the theory of magneto-volume effects in this book, compared to the SEW and MU theories, is that spectral parameters T_0 and T_A are volume dependent. It is reflected in the relation (6.127) between the pressure effects on $\sigma_0(0)$ and T_c. The SEW theory predicts the relation, $\mathrm{d}\log\sigma_0(0)/\mathrm{d}p = \mathrm{d}\log T_c/\mathrm{d}p$, since $\sigma_0^2(0) \propto T_c^2$ is satisfied. In the MU theory, on the other hand, the same relation (6.127) is satisfied, but with $\gamma_{0,A} = 0$ in the right hand side. Validity of them are verified by the pressure effect measurements of $\sigma_0(0)$ and T_c.

Many experiments have been done on the pressure effects on $\sigma_0(0)$ and T_c. According to Kanomata (T. Kanomata, Private Commun.), the observed results show variety of signs dependent on each itinerant electron magnets against the applied pressure. Most of them are, however, classified into the following three categories:

1. Both the change of $\sigma_0(0)$ and T_c have the same signs.
   This case is characteristic to itinerant electron magnets.
2. Though T_c changes, the value of $\sigma_0(0)$ remains almost unchanged.
   It is usually observed for localized electron magnets.
3. Each of them show changes with different signs.

These properties can be understood by introducing multiple Grüneisen parameters, and in some cases by assuming that they are of comparable magnitude. They will be interpreted associated with signs and relative magnitudes of these parameters.

6.7.2 Pressure Effect Measurements of Spontaneous Magnetic Moment and Critical Temperature

A large number of experiments on the magneto-volume effects had been reported from the late 1960s to the beginning of 1980s. Their aim was to verify the SEW theory experimentally. Analyses of experiments were also based on the theory. These were reviewed by Franse [28, 29]. Later, magneto-volume effects on $ZrZn_2$, MnSi, and Ni_3Al were reported by Brommer and Franse [30]. Results of analyses based on the MU theory were also found here. These authors also published the handbook on the magneto-volume effects in 1990 [31]. Most of these experiments belong to the first category of the Kanomata's classification. The observed large T^2-linear thermal expansions for para- and ferromagnets near the magnetic instability points should be rather associated with magnetic origins. They were, however, regarded as the effect of conduction electrons from the conventional view. Many magneto-volume properties reported up to the present need to be re-examined.

The following is a brief summary of observed magneto-volume effects on weak itinerant electron ferromagnets where weak spontaneous magnetization are observed.

Ni_3Al

So far, a number of magneto-volume measurements have been done on this compound. The M^2-linear coefficients of the free energy were estimated by Buis et al. [32] from the observed magnetic isotherms under the pressure up to 5 kbar. The pressure dependence of the critical temperature T_c and the value of the magneto-volume coupling constant C are then evaluated by their temperature and magnetic field dependence. The critical temperature was determined as the temperature at which the Arrott plot of the magnetization curve passes through the origin. These values vary within the range, $\partial T_c/\partial p = -0.58 \sim -0.36$ K/kbar and $C \times 10^{-6} = 0.12 \sim 0.16$ (g/cm^3), depending on the composition of Ni and Al, according to their report. As a compressibility, $\kappa = 4.2 \times 10^{-13}$ cm^2/dyne was employed. The forced magneto-volume coupling constant C was also estimated by Kortekaas and Franse [4] from the magneto-striction measurements in the ordered phase. From the observed constants at different temperatures, they showed that C is temperature dependent, that presumably originates from the T^2/T_F^2 dependence of the SEW theory. The value of the coupling $\rho\kappa C \times 10^6 \sim 0.6$ ($G^{-2}g^2cm^{-6}$) at 4.2 K is reduced by 0.4 at T_c. As the compressibility, $\kappa = 4.18 \times 10^{-13}$ cm^2/dyne was used to estimate the value of C.

On the other hand, Buis et al. [33] made magnetization measurements on samples under pressure with different Al composition of the compounds. From the analysis of the composition dependence of the M^2 expansion coefficient (i.e., the inverse of the magnetic susceptibility) of the free energy, they predicted the value of the spontaneous magnetic moment and the pressure dependence of the critical temperature of the ideal Ni_3Al compound [33] with $\sigma_0 = 0.077\ \mu_B$/at and $T_c = 63$ K as given by

$$\frac{\partial \log \sigma_0(0)}{\partial p} = -5.29\ \text{Mbar}^{-1}, \quad \frac{\partial \log T_c}{\partial p} = -6.35\ \text{Mbar}^{-1}.$$

The pressure dependence of the magnetic susceptibility in the paramagnetic phase was reported Brommer et al. [27] as was already shown in Sect. 6.5.3 in this chapter.

Measurements of forced magneto-strictions and thermal expansions were done by Suzuki and Masuda [34, 35] to check the validity of the MU theory. They showed that the forced magneto-volume coupling constant C decreases with increasing temperature, according to the $T^{4/3}$-linear dependence [34, 35]. In their analysis they assume the presence of the following thermal expansion from the nonmagnetic origin:

$$\alpha_{nm} = aT + bT^3,$$

where the second term results from the lattice vibrations. In the paramagnetic phase at high temperatures, they extract the magnetic contribution by subtracting the Debye part. They concluded that the magneto-volume thermal expansion is present even in the paramagnetic phase that tends to saturate with increasing temperature.

$ZrZn_2$

The forced magneto-striction of this compounds was reported by Ogawa and Waki [36] as given by

$$\omega = 1.02 \times 10^{-10} M^2, \ (M \text{ in emu/mole}),$$

based on their measurements over the temperature range from 4.2 to 40 K under the external field up to 10 kOe. Around the same time, Meincke et al. [37] also reported their measurements of the thermal expansion $\omega(T)$ in the range up to 6.8 K, and the forced magneto-volume striction at 4.2 K under the external field up to 35 kOe. Their results are summarized by

$$\omega(T) = -10.6 \times 10^{-8}\, T^2, \quad \omega = 1.80 \times 10^{-10} M^2, \ (M \text{ in emu/mole}).$$

There exists almost two times difference between the above forced magneto-volume coupling constants.

As for the pressure effect on T_c, Wayne and Edwards [38] reported the value, $-1.95\,\mathrm{K\ kbar^{-1}}$, for samples with $T_c = 21.5\,\mathrm{K}$. Then nearly the same pressure decrease of the critical temperature, $T_c = 22.2 - 1.9P\,\mathrm{K}$ (P in units of kbar), was later reported by Smith [39] under the pressure up to 25 kbar. A slightly different $\mathrm{d}T_c/\mathrm{d}p = -1.29$ K/kbar ($T_c = 27.6\,\mathrm{K}$) was also reported by Huber et al. [40].

MnSi

The results of measurements of the thermal volume expansion and the forced magneto-striction were reported by Fawcett et al. [41]. According to them, $\partial\sigma/\partial\omega = 8.5$ was obtained as a volume dependence of the spontaneous magnetization. Bloch et al. [42] reported the values, $\mathrm{d}\log M/\mathrm{d}p = -1.15 \times 10^{-2}\,\mathrm{kbar^{-1}}$ and $\mathrm{d}\log T_c/\mathrm{d}p = -3.9 \times 10^{-2}\,\mathrm{kbar^{-1}}$, as the pressure dependence of the spontaneous magnetization at 4.2 K and the pressure effect on T_c, respectively. They amount to $\mathrm{d}\log M/\mathrm{d}\omega = 16$ and $\mathrm{d}\log T_c/\mathrm{d}\omega = 53$, if the observed value of the compressibility $\kappa^{-1} = -1.36 \times 10^6\,\mathrm{kbar^{-1}}$ is used. Thessieu et al. [43] also independently measured the pressure dependence of $M_0(0)$ and T_c, and estimated the pressure dependence of spectral parameters T_0 and T_A. The pressure effect on both $M_0(0)$ and T_c are also reported by Koyama et al. [44], recently.

Meanwhile, the temperature dependence of the magneto-volume expansion and the forced magneto-striction were measured by Matsunaga et al. [6] up to the temperature 200 K for the purpose to confirm the prediction of the MU theory. They reported the following value as the coupling constant of the forced striction at 4.2 K.

$$\omega = 1.49 \times 10^{-10} M^2, \quad (M \text{ in emu/mole})$$

As its temperature dependence, values $\rho\kappa C = 10.25$, 5.88, 5.63, and 6.08×10^{-7} (g/emu)^2 are estimated at temperatures, $T = 4.2$, 29, 40, and 50 K, respectively. The critical temperature of this compound is around 30 K. On the other hand, the observed coupling constant of the thermal expansion is given by $\rho\kappa C_T = 6.33 \times 10^{-7}$ (g/emu)^2. They also concluded that there exists a definite component of the thermal expansion in the paramagnetic phase other than the effect of lattice vibrations.

Sc_3In

As the pressure effect on the Curie temperature, $\mathrm{d}T_c/\mathrm{d}p = 0.19\,\mathrm{kbar^{-1}}$ ($\mathrm{d}\log T_c/\mathrm{d}\omega = -13$) was estimated by Gardner et al. [45] for sample with $T_c = 6.1$ K. Later, Grewe et al. [46] made the same experiments under the pressure up to 6 kbar by applying the magnetic field up to 57 kOe in the range of temperature from 3 to 300 K. The pressure dependence of their report is shown below.

$$\frac{dT_c}{dp} = \begin{cases} 0.15 \ \ (\mathrm{K/kbar}), & T_c = 5.5\mathrm{K}, \ \text{for 24.1 at \% In} \\ 0.195 \ \ (\mathrm{K/kbar}), & T_c = 6.0\mathrm{K}, \ \text{for 24.3 at \% In} \end{cases}$$

They correspond to $d \log T_c/dp = 2.7$, and $3.25\,\%\,\mathrm{kbar}^{-1}$, respectively. As the pressure effect on the spontaneous magnetization at 3 K for the same. In concentrations, $d \log M_0/dp = 0.85, 0.94\,\%\,\mathrm{kbar}^{-1}$ were reported.

$Y(Co,Al)_2$

The Al-substituted Laves phase compounds $Y(Co_{1-x}Al_x)_2$ have attracted much interest since they show metamagnetic transitions. The magneto-volume effect of this compound with $x \sim 0.15$ was measured by Armitage et al. [47]. They reported the values, $d \log T_c/d\omega = d \log \sigma_0(0)/d\omega = 120 \pm 17$. Later, the measurements of magnetization, magneto-volume expansion, and magneto-volume striction had been made by Duc et al. [48] in the presence of high magnetic field under the high pressure. In these studies, the value of the compressibility, $\kappa = 9.4 \times 10^{-4}\,(\mathrm{kbar})^{-1}$ in Yamada and Shimizu [49] were used.

Ni-Pt Alloys and Other Compounds

The forced magneto-striction measurements were made by Kortekaas et al. [4] on Ni-Pt alloys (of density $\rho = 17\,\mathrm{g/cm^3}$). According to them, $\rho\kappa C \times 10^6 = 4.50\,(\mathrm{G^{-2}g^2cm^{-6}})$ was obtained as a coupling constant of the alloy at 36.6 at % Ni concentration at 4.2 K. The value decreases with increasing the Ni concentration, reaching the value 3.32 at the concentration, 45.2 at % Ni. These values tend to decrease with increasing temperature. In addition to this, thermal volume expansion measurements on (Fe, Co)Si and YNi_3 were reported by Shimizu et al. [50] and Parviainen, Lehtinen [51], respectively. Oraltay et al. [52] reported their thermal expansion, specific heat, and forced magneto-striction measurements on Y_9Co_7.

Heusler Alloys

Recently, the pressure effect on the critical temperature and the spontaneous magnetic moment of ferromagnetic heusler alloys have been measured on Co_2ZrAl by Kanomata et al. [53] and Rh_2NiGe by Adachi et al. [54], for instance.

To summarize, many observations described above show that forced magneto-volume coupling constants are temperature dependent. At first, its dependence was regarded as resulting from the T^2/T_F^2-linear dependence of the SEW theory.

Table 6.1 Grüneisen parameters estimated from the pressure effects on T_c and M_0

Compounds	$-\dfrac{\mathrm{d}\log M_0}{\mathrm{d}p}$	$-\dfrac{\mathrm{d}\log T_c}{\mathrm{d}p}$	$\kappa\gamma_{0A}$	γ_{0A}/γ_m	References
$TiFe_{0.5}Co_{0.5}$	13.8	19.3	1.4	0.051	[55]
$Ni_{75}Al_{25}$	8.7	11.6	1.45	0.083	[33]
$Y(Co_{0.85}Al_{0.15})_2$	120	113	67	0.279	[47]
Co_2ZrAl	1.8	2.2	0.5	0.139	[53]
$Fe_{67}Ni_{33}$	6.9	8.9	1.45	0.105	[56]
$ZrZn_{1.9}$	44	46.7	19.3	0.219	[40]
$Ni_{45}Pt_{55}$	21	18	13.5	0.321	Kanomata[a]
$Fe_{0.3}Co_{0.7}Si$	16	12	12	0.375	[57, 58]
MnSi	12.2	38	−19.7	−0.807	[44]
Co_2TiGa	2.9	9.5	−5.2	−0.897	[59]
$Sc_{75.7}In_{24.3}$	−9.4	−32.5	18.4	−0.979	[46]
Rh_2NiGe	1.5	5.3	−3.1	−1.033	[54]

[a] Private commun.

However, the dependence has soon become of little interest. Results of the pressure effect on the spontaneous magnetic moment and the critical temperature are summarized in Table 6.1. The value of $\kappa\gamma_{0A}$ estimated from (6.116) and its ratio to $\kappa\gamma_m$ are also shown in fourth and fifth columns of the table, respectively. From this table, we will find that the parameters γ_0 and γ_A are not negligible compared to γ_m. According to the SEW theory, values of the second and third columns of this table would be in agreement with each other. Values of the fourth column are assumed to be zero in the MU theory. Experimentally estimated values of this table do seem to support neither of them. In the case of MnSi, for example, the larger suppression of the critical temperature T_c by the external pressure than that of $M_0^2(0)$ can be accounted by neither of them. The problem is easily solved by introducing two new parameters, γ_0 and γ_A.

For confirmation of some mutual correlations among the magnetic Grüneisen parameters, the values of γ_m for magnets in Table 6.1 are plotted against $\gamma_{0,A}$ in Fig. 6.13. No definite correlations seem to be present in the figure. They are all regarded as significant parameters to be used to characterize the magneto-volume effects of itinerant electron magnets.

6.8 Summary of Magneto-Volume Effects

In this chapter, we have shown that the magneto-volume effect is derived from the explicit volume dependence of the free energy that is used in our treatment of the magnetic specific heat in the preceding chapter. It enables our unified understanding of the magneto-volume effect, as well as the thermal and magnetic properties of magnetic susceptibility, magnetic isotherms, and magnetic specific heat. For this

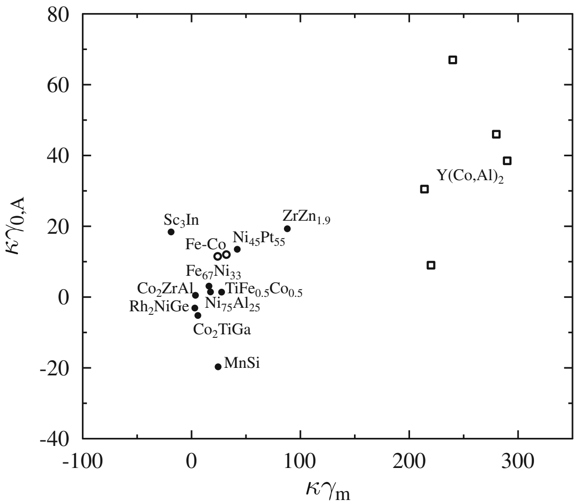


Fig. 6.13 Correlation between Grüneisen parameters, $\gamma_{0,A}$ and γ_m

purpose, three non-traditional magnetic Grüneisen parameters, γ_m, γ_0, and γ_A are introduced, that characterize the interactions between the magnetism and the volume of magnets. As a result, the following novel properties have been derived as summarized below.

- The magneto-volume expansion $\omega_m(t)$ that consists of two kinds of components
  The thermal component $\omega_{th}(t)$, resulting from the finite parameter γ_0, has long been neglected. The presence of this term is evident from the thermodynamic relation between the thermal volume expansion and the magnetic specific heat at low temperatures. The other one, $\omega_{zp}(t)$, related with the parameter γ_m corresponds to the conventional contribution predicted by the SEW and MU theories.
- The new magneto-volume coupling constants defined for the component $\omega_{zp}(t)$
  Two magneto-volume coupling constants C_s and C_h are necessary for spontaneous and forced magneto-strictions, respectively. They have different values ($C_s \sim 2C_h/5$) and are both temperature dependent.
- The anomalous critical forced magneto-striction observed at the critical temperature
  At the critical temperature, the forced magneto-volume expansion $\omega_h(\sigma, t_c)$ becomes proportional to σ^4.
- The revised relation satisfied between $\mathrm{d}\log T_c/\mathrm{d}p$ and $\mathrm{d}\log \sigma_0^2(0)/\mathrm{d}p$
  Because of the presence of multiple Grüneisen parameters, a somewhat different relation is satisfied between the above two pressure effects.

There seem to be many observed magneto-volume measurements that will support the above theoretical predictions.

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