Quantum spin fluctuation theory of the magnetic equation of state of weak itinerant-electron ferromagnets

2001 J. Phys.: Condens. Matter 13 6323

(http://iopscience.iop.org/0953-8984/13/29/305)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 6323–6358 PII: S0953-8984(01)23106-3

Quantum spin fluctuation theory of the magnetic equation of state of weak itinerant-electron ferromagnets

Y Takahashi

Faculty of Science, Himeji Institute of Technology, 3-2-1 Kouto, Kamigori-cho, Ako-gun, 678-1297, Japan

E-mail: takahash@sci.himeji-tech.ac.jp

Received 20 March 2001
Published 6 July 2001
Online at stacks.iop.org/JPhysCM/13/6323

Abstract

On the basis of the spin fluctuation mechanism, magnetic properties of itinerant-electron weak ferromagnets are discussed, for the wide temperature range from the ground state to the paramagnetic state, explicitly taking into account the effects of zero-point quantum spin fluctuations. Particular attention is focused on properties of the ordered phase. The temperature dependence of the spontaneous magnetic moment, for instance, is quantitatively analysed in close comparison with experiments. It is also shown that the fourth-order expansion coefficient of the free energy in powers of the static magnetic moment is temperature dependent, and therefore magnetic isotherms are not so simple as was originally anticipated in the Stoner–Wohlfarth theory.

We explicitly examine the effect of the spin-wave mode on the transverse spin fluctuation amplitude, and show that this effect is crucial for the proper theoretical description of magnetic behaviours in the ordered state.

1. Introduction

Collective spin fluctuations play predominant roles in the magnetic properties of itinerant-electron magnets, while the single-particle excitations have minor roles. Spin fluctuation theories based on the above picture have been quite successful in explaining and predicting various interesting magnetic properties of the system (Moriya 1985, Lonzarich and Taillefer 1985, Takahashi 1986). However, there are still unsolved problems. One long-standing problem is related to the description of the ordered phase below the Curie temperature T_c. The difficulty arises because it is sometimes not so easy to take into account the restriction caused by the rotational invariance of the system in the spin space. A naive treatment (see, for instance, Murata and Doniach (1972) and Yamada (1975)) leads to a fictitious first-order phase transition. In the self-consistent renormalization (SCR) spin fluctuation theory (see,

for instance, Moriya 1985), the temperature dependence of the spontaneous magnetization M for weak itinerant magnets was originally derived by taking into account only the transverse modes of the thermal spin fluctuations with respect to the direction of the static uniform moment (Moriya and Kawabata 1973a, b). As the typical temperature dependence of the squared spontaneous moment, a qualitative T^2-dependence at low temperature and a $(T_c^{4/3} - T^{4/3})$-dependence around the critical temperature T_c were predicted and were later confirmed by experiments. On the other hand, based on the single-particle picture, the Stoner–Wohlfarth theory (Stoner 1936, Wohlfarth 1968) predicted an overall $(T_c^2 - T^2)$-dependence below T_c (Edwards and Wohlfarth 1968). The observed temperature dependence was sometimes simply fitted with the Stoner–Wohlfarth theory. No quantitative comparison with experiments, however, has yet been made on the basis of the spin fluctuation mechanism.

If we take into account both the transverse and the longitudinal components of the fluctuations required by the rotational invariance, we have to solve two coupled integro-differential equations simultaneously, each of which is related to the condition determining the transverse and the longitudinal components of the magnetic susceptibilities. Numerical solutions of these equations show that the first-order discontinuity is reduced or will one hopes be eliminated (Lonzarich and Taillefer 1985). The nature of the solution has not been fully discussed yet. To save computational effort, an interpolation scheme for the mode-coupling effect was also proposed by Lonzarich and Taillefer, giving a smooth evolution between the ground state and the critical point without showing a first-order transition. Its theoretical basis is, however, not too clear.

The spin fluctuation theories mentioned above only take into account the effects of thermal spin fluctuations. Particular interest has been shown in the effects of renormalization on the second expansion coefficient of the magnetic free energy in terms of the uniform magnetization M, i.e. the temperature dependence of the magnetic susceptibility. Therefore the magnetic equation of state has not so far been discussed very thoroughly. Although the effects of spin fluctuations are included in the description of finite-temperature properties, the magnetic field dependence of M is simply assumed to be given by using the Stoner–Wohlfarth theory in the ground state and it is determined by the density-of-states curve around the Fermi energy. The SCR theory particularly emphasizes the significance of the self-consistent treatment in deriving the Curie–Weiss-like temperature dependence of the magnetic susceptibility. This means that we have to take into account the change of the spin fluctuation spectrum against temperature and the external field variations. The same spectral change ought to have some effect on the quantum amplitude. However, it is simply assumed to be negligible. Though the spin fluctuation effects are included at finite temperature, the ground state is still assumed to be well described in terms of the single-particle density of states like in the Stoner–Wohlfarth theory.

I first pointed out the significant role of quantum spin fluctuations in the magnetic properties of weak itinerant-electron magnets. According to my idea, the ground-state and finite-temperature properties have to be treated within the same theoretical framework. In order to implement this idea, an interesting approach has been proposed. It is based on solving a single equation containing both the transverse and the longitudinal magnetic susceptibilities. Because the quantities are related to each other through their differentials with respect to the magnetization M, the magnetic equation of state is obtained by integrating the equation by regarding it as a first-order differential equation.

The purpose of this paper is to present a theoretical description of magnetic properties of weak itinerant-electron magnets by studying the magnetic equation of state throughout a wide temperature range. In this way we can discuss their temperature and magnetic field dependence from a unified point of view. Our main interest is in giving a consistent and satisfactory description of properties in the ordered state by solving the differential equation.

Although an attempt has been made to find solutions by using their continuation from the high-field side (Takahashi 1992), the situation is still not very clear compared with the paramagnetic cases because of the difficulty of finding the initial condition of the equation. In this regard, we explicitly examine the roles of the spin-wave modes in the ordered state. In most previous treatments this effect is simply neglected; the assumption is made that it is very small. We show here how to cope with the difficulty involved in determining the initial condition by taking explicit account of the presence of spin-wave modes.

The absence of a fully consistent theoretical treatment of the ordered state is the reason that quantitative comparisons between theoretical predictions and experiments have not so far been made on the basis of the spin fluctuation mechanism. In the present paper, various observed temperature and external field dependences are quantitatively analysed on the basis of the results of this paper. In our previous studies we have already given a prescription for dealing with magnetic properties in the paramagnetic phase. In order to achieve a self-contained presentation, we will duplicate some of our former arguments here in referring to experimental results.

In the following we represent the magnetization σ per magnetic atom in units of μ_B and the external magnetic field h in energy units, i.e.,

$$M = N_0\mu_B\sigma \qquad h = g\mu_B H$$

where N_0 is the number of magnetic atoms in the crystal and g is the gyromagnetic ration, assumed to be 2. The magnetic susceptibility χ is measured in units of $(g\mu_B)^2$, given in the present units by

$$\chi/N_0 = \sigma/(2h).$$

2. Theoretical framework based on the spin fluctuation mechanism

According to Takahashi (1986) the present study is based on the following sum rule:

$$\left\langle \boldsymbol{S}_i^2 \right\rangle = \left\langle \left\{ S_i^+, S_i^- \right\}/2 + \left(\delta S_i^z\right)^2 \right\rangle + \sigma^2/4 \tag{2.1}$$

where

$$\delta S_i^z = S_i^z - \left\langle S_i^z \right\rangle \qquad \sigma = 2\left\langle S_i^z \right\rangle$$

which indicates that the total spin fluctuation amplitude is almost constant (see also Takahashi 1990, 1992, 1994, 1997a, b, 1998, Takahashi and Sakai 1995, 1998). At the outset, it is worth recognizing that the spin fluctuation amplitude is not so small when the quantum component is included, as was confirmed experimentally. This is why we employ the sum rule and do not rely on the expansion in terms of the amplitude of the spin fluctuations. If we extract the thermal spin fluctuation amplitude, we find that it increases with increasing temperature above T_c as was actually observed in neutron scattering experiments on MnSi (Ishikawa *et al* 1985). The total amplitude, however, does not seem to show a monotonic increase. We show in figure 1 the temperature dependence of the total spin fluctuation amplitude for the same compound MnSi as was observed in the neutron scattering experiment (Ziebeck *et al* 1982) in comparison with the calculated thermal spin fluctuation amplitude. The discrepancy between experiments and the theoretical curve at low temperature clearly shows the presence of a sizable quantum amplitude. The weak temperature dependence of the total amplitude compared with the thermal component indicates that the quantum amplitude is also temperature dependent. The effect of zero-point spin fluctuations has also been considered by expanding the free energy in powers of the spin fluctuation amplitude up to the fourth-order term (Solontsov and Wagner

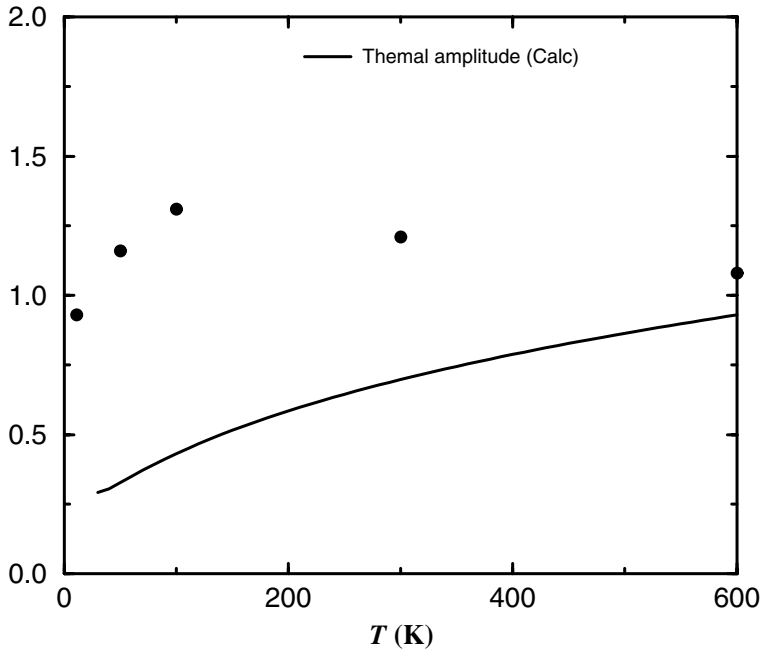

Figure 1. The temperature dependence of the observed spin fluctuation amplitude for MnSi (solid circles), using an energy resolution of 12 THz. The solid line represents the calculated thermal spin fluctuation amplitude.

1994, 1995). It is however not so clear why such an expansion in terms of large fluctuation amplitudes is justified when the quantum component is included.

We have already derived various interesting consequences of the conserved amplitude (2.1), which were confirmed by later experimental investigations (Yoshimura *et al* 1987, 1988, Shimizu *et al* 1990, Nakabayashi *et al* 1992). Before we present our theoretical framework, we describe the behaviour of the spin fluctuation spectrum and amplitudes for weak itinerant-electron magnets.

2.1. Spin fluctuation spectrum

From the fluctuation-dissipation theorem of statistical mechanics, the spin fluctuation amplitude is expressed in terms of the response of the system, i.e. in terms of the imaginary part of the dynamical magnetic susceptibility $\chi(q,\omega)$. For instance, in the paramagnetic phase, it is represented by

$$\left\langle \boldsymbol{S}_i^2 \right\rangle = \frac{3}{N_0^2}\sum_{\boldsymbol{q}} \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} \coth(\beta\omega/2)\,\mathrm{Im}\,\chi(\boldsymbol{q},\omega)$$
$$= \frac{3}{N_0^2}\sum_{\boldsymbol{q}} \int_{0}^{\infty} \frac{\mathrm{d}\omega}{\pi}\ \{1+2n(\omega)\}\,\mathrm{Im}\,\chi(\boldsymbol{q},\omega). \tag{2.2}$$

That is,

$$\left\langle \boldsymbol{S}_i^2 \right\rangle = \left\langle \boldsymbol{S}_i^2 \right\rangle_Z + \left\langle \boldsymbol{S}_i^2 \right\rangle_T \tag{2.3}$$

where we have defined quantum (zero-point) and thermal amplitudes, denoted by the subscripts Z and T, by decomposing the factor $\coth(\beta\omega/2)$ into a constant term and the Bose factor $n(\omega)$ as follows:

$$\coth(\beta\omega/2) = 1 + 2n(\omega) \qquad n(\omega) = \frac{1}{\mathrm{e}^{\beta\omega} - 1}.$$

In the case of weak itinerant-electron ferromagnets, the imaginary part of the dynamical magnetic susceptibility is given by the following double Lorentzian spectrum in the small-$\boldsymbol{q}, \omega$ space:

$$\mathrm{Im}\,\chi(\boldsymbol{q}, \omega) = \chi(q)\frac{\omega\Gamma_q}{\omega^2 + \Gamma_q^2} \tag{2.4}$$

where

$$\chi(q) = \frac{\chi(0)}{1 + q^2/\kappa^2} \qquad \Gamma_q = \Gamma_0 q(\kappa^2 + q^2)$$

where κ represents the inverse of the magnetic correlation length. The above q-linear dependence of the damping constant Γ_q results from the Landau damping mechanism. Its validity is confirmed experimentally for MnSi (Ishikawa *et al* 1985) and Ni_3Al (Bernhoeft *et al* 1983, 1986). On the basis of our previous studies, we represent the above spectrum in a parametrized form by introducing the energy scales T_0 and T_A defined by

$$T_0 = \Gamma_0 q_B^3/2\pi \qquad T_A = N_0 q_B^2/[2\chi(0)\kappa^2] \qquad \left(q_B^3 = 6\pi^2 N/V\right)$$

where q_B is the zone-boundary wave vector for the crystal with N magnetic atoms within the volume V. These parameters give measures of the spectral widths in the energy and wave-vector spaces, respectively, which correspond to the exchange coupling constant in the case of Heisenberg magnets. They are directly estimated from neutron scattering experiments (Ishikawa *et al* 1985, Bernhoeft *et al* 1983, 1986) or from analyses of the temperature dependence of NMR relaxation time measurements (Hioki and Masuda 1977, Kontani 1977, Yasuoka *et al* 1978, Umemura and Masuda 1983, Yoshimura *et al* 1987, 1988). The imaginary part, $\mathrm{Im}\,\chi(\boldsymbol{q}, \omega)$, the damping constant, Γ_q, and $\chi(q)$ are then given in the reduced form by

$$\begin{aligned} &\mathrm{Im}\,\chi(\boldsymbol{q}, \omega)/N_0 = \frac{T_0}{2T_A T}\frac{\xi x}{\xi^2 + u^2} \qquad u = x(y + x^2)/t \\ &\Gamma_q = 2\pi T_0 x(y + x^2) \\ &\chi(q) = \frac{N_0}{2T_A}\frac{1}{y + x^2} \end{aligned} \tag{2.5}$$

where $\xi = \omega/2\pi T$, $t = T/T_0$, $x = q/q_B$, and $y = \kappa^2/q_B^2$. The change in spectral form of the fluctuations is therefore taken into account through the parameter y. In this sense y plays a significant role in our following discussions. It has the meaning of the reciprocal of the dimensionless magnetic susceptibility.

2.2. Thermal and quantum spin fluctuation amplitudes

With the use of the parametrized form of the spectrum, we can now explicitly evaluate the thermal and the quantum spin fluctuation amplitudes as functions of t and y. After ω-integration, the thermal amplitude can be represented in the following form as a wave-vector integral:

$$\left\langle \boldsymbol{S}_i^2 \right\rangle_T (y, t) = \frac{2}{N_0^2}\sum_{\boldsymbol{q}} \int_0^\infty \frac{\mathrm{d}\omega}{\pi}\, n(\omega)\,\mathrm{Im}\,\chi(\boldsymbol{q}, \omega) = \frac{9T_0}{T_A} A(y, t) \tag{2.6}$$

where

$$A(y,t)=\int_0^1 \mathrm{d}x\; x^3\,[\ln u-1/2u-\psi(u)] \qquad (x=q/q_B)$$

where $\psi(u)$ is the digamma function defined by

$$\ln u-1/2u-\psi(u)=2\int_0^\infty \mathrm{d}t\;\frac{t}{\mathrm{e}^{2\pi t}-1}\frac{1}{t^2+u^2}.$$

Because of the presence of the Bose factor in its definition, the amplitude has dependence on both the temperature t and the reciprocal susceptibility y. The critical thermal amplitude for $y=0$ is, in the small-t limit, given by

$$\begin{aligned}A(0,t)&=\int_0^1 \mathrm{d}x\; x^3\,[\ln u_c-1/2u_c-\psi(u_c)] \qquad (u_c=x^3/t)\\ &=\frac{1}{3}t^{4/3}\int_0^{1/t}\mathrm{d}s\; s^{1/3}\,[\ln s-1/2s-\psi(s)]\simeq\frac{1}{3}C_{4/3}t^{4/3}\end{aligned} \tag{2.7}$$

where

$$\begin{aligned}C_\alpha&=\int_0^\infty \mathrm{d}s\; s^{\alpha-1}\,[\ln s-1/2s-\psi(s)]=\frac{\pi\zeta(\alpha)\Gamma(\alpha)}{(2\pi)^\alpha\sin(\pi\alpha/2)}\\ &=1.006089\cdots \qquad (\text{for } \alpha=4/3).\end{aligned}$$

On the other hand, its y-dependence around the origin is estimated as follows. Note that only the narrow region around the origin of the integrand of (2.7) is affected when y changes its magnitude for $y\ll 1$. Since $[\ln u-1/2u-\psi(u)]$ is approximated by $1/2u$ for $u\ll 1$ there, the amplitude is dominated by the following singular $\sqrt{y}$-dependence:

$$\begin{aligned}A(y,t)-A(0,t)&\simeq\int_0^1 \mathrm{d}x\; x^3\left(\frac{1}{2u}-\frac{1}{2u_c}\right)\\ &=\frac{t}{2}\int_0^1\mathrm{d}x\;\left(\frac{x^2}{y+x^2}-1\right)=-\frac{t\sqrt{y}}{2}\tan^{-1}\left(\frac{1}{\sqrt{y}}\right)\\ &\simeq-\frac{\pi t}{4}\sqrt{y} \qquad (\text{for } y\simeq 0).\end{aligned} \tag{2.8}$$

The critical dependence of $A(y,t)$ on y and t is therefore summarized by

$$A(y,t)\simeq\frac{1}{3}C_{4/3}t^{4/3}-\frac{\pi t}{4}\sqrt{y}+\cdots \qquad (\text{for } t,\,y\ll 1). \tag{2.9}$$

The quantum amplitude, on the other hand, has no explicit temperature dependence, though it still has an implicit y-linear dependence given by

$$\left\langle \boldsymbol{S}_i^2\right\rangle_Z(y)=\left\langle \boldsymbol{S}_i^2\right\rangle_Z(0)-\frac{9T_0}{T_A}c_z y. \tag{2.10}$$

The above y-linear coefficient can be estimated as follows. Since taking a Lorentzian shape for the spectrum is only justified in the low-frequency region, we have in general to introduce the ω-dependence of the damping constant $\Gamma(q,\omega)$ in the higher-frequency region. The first-order y-derivative of the quantum amplitude is then represented by

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

$$\simeq -\frac{3}{N_0^2}\sum_{\boldsymbol{q}}\chi(q)\frac{\partial\Gamma_q}{\partial y}\int_0^\infty\frac{\mathrm{d}\omega}{\pi}\frac{2\omega\Gamma_q^2}{\left[\omega^2+\Gamma_q^2\right]^2}$$

$$= -\frac{3}{N_0^2}\sum_{\boldsymbol{q}}\chi(q)\frac{\partial\Gamma_q}{\partial y}\bigg|_{y=0}.$$

We first note that the y-dependence mainly arises from the low-frequency region in the integrand where $\Gamma(q,\omega)$ is very small. The first term is then neglected since the product $[\chi(q)\Gamma(q,\omega)]$ is almost y-independent there, as we see by recalling the expressions for $\chi(q)$ and Γ_q in (2.5). The y-linear coefficient mainly comes from the second term, shown as the second line. Since its spectral intensity is now confined to within the low-frequency region, the ω-dependence of $\Gamma(q,\omega)$ of this term is well approximated by $\Gamma_q=\Gamma(q,0)$. After ω-integration it is given as an average of $\chi(q)\partial\Gamma_q/\partial y$ over the whole Brillouin zone. From the expressions for Γ_q and $\chi(q)$ in (2.5), we can see that it has a magnitude of the order of T_0/T_A. If we simply extrapolate the expressions of (2.5) throughout the whole Brillouin zone, we obtain $c_z=1/2$. Depending on the behaviours of $\chi(q)$ and Γ_q as functions of q over a wide area in wave-vector space, the value of c_z will show some slight variation, but its magnitude is of the order of unity. In this way, y-dependence of the quantum amplitude is present and is caused by the change of the spin fluctuation spectrum. The effect is in general not so small and is not neglected.

With the use of the above expressions for both of the spin fluctuation amplitudes, we obtain the following relation between them at the critical temperature $t=t_c=T_c/T_0$:

$$\left\langle \boldsymbol{S}_i^2\right\rangle_{\mathrm{tot}}=\left\langle \boldsymbol{S}_i^2\right\rangle_Z(0)+\left\langle \boldsymbol{S}_i^2\right\rangle_T(0,t_c)=\left\langle \boldsymbol{S}_i^2\right\rangle_Z(0)+\frac{9T_0}{T_A}A(0,t_c).$$

Weak itinerant-electron ferromagnets are characterized by their small t_c-ratios. From the expression for $A(0,t)$ in (2.7), the thermal amplitude for these magnets remains always very small because t, of the same order of t_c, is very small, whereas the quantum component $\langle \boldsymbol{S}_i^2\rangle_Z(y)$ has a large amplitude, of the same magnitude as the total spin fluctuation amplitude. In contrast, their size difference is reversed for Heisenberg magnets because of the localization of the spectral weight within the low-energy region. To show the clear distinction between these types of magnet, the magnitudes of the critical amplitudes for the two components are compared in table 1. It is important to realize from the table that weak itinerant-electron magnets are characterized by the presence of quantum spin fluctuations with large amplitude throughout the wide temperature range of interest. For this reason, the quantum amplitude has key significance in our understanding of the properties of these magnets.

Table 1. Comparison of the magnitudes of the thermal and the quantum amplitudes.

Compound	Heisenberg system	Itinerant weak ferromagnets
Thermal	$\langle \boldsymbol{S}_i^2\rangle_T(0,t_c)\sim \mathrm{O}(1)$	$\langle \boldsymbol{S}_i^2\rangle_T(0,t_c)\ll 1$
Quantum	$\langle \boldsymbol{S}_i^2\rangle_Z(0)\ll 1$	$\langle \boldsymbol{S}_i^2\rangle_Z(0)\sim \mathrm{O}(1)$

Our basic idea for deriving the magnetic equation of state is based on the following observations on the behaviour of the spin fluctuations in the presence of a static uniform moment:

- The magnitude of the local squared spin operator $\boldsymbol{S}_i^2$ has an almost conserved expectation value. This assertion seems to be well justified for Heisenberg magnets. What we assume is that it is also applicable to the itinerant-electron magnets.

When a spontaneous moment appears at low temperature below T_c or in the presence of an external magnetic field, the expectation has to be given by the sum of the squared static moment $|\langle \boldsymbol{S}_i \rangle|^2$ and the squared fluctuation amplitude $\langle \delta \boldsymbol{S}_i^2 \rangle$ ($\delta \boldsymbol{S}_i = \boldsymbol{S}_i - \langle \boldsymbol{S}_i \rangle$).

- The anisotropy of the spin fluctuation amplitude has to be taken into account.

  This effect arises from the anisotropy of the reciprocals of the magnetic susceptibilities. Depending on the direction of the external field applied to the system relative to the static moment, the magnitudes of the perpendicular and the longitudinal magnetic susceptibilities become different. They are, respectively, given by H/M and $\partial H/\partial M$, or in dimensionless units by y and y_z defined below:

$$\begin{aligned} y &= \kappa^2 \big/ q_B^2 = \frac{1}{T_A}\frac{h}{\sigma} \\ y_z &= \kappa_z^2 \big/ q_B^2 = \frac{1}{T_A}\frac{\partial h}{\partial \sigma} = y + \sigma \frac{\partial y}{\partial \sigma}. \end{aligned} \tag{2.11}$$

  The effect is included by assuming anisotropic inverse squared correlation lengths κ^2 and κ_z^2 in evaluating spin fluctuation amplitudes.

- The effect of the presence of spin-wave mode also has to be taken into account for evaluating the transverse amplitude.

  Because of the appearance of the spin-wave mode around the origin of the wave-vector space, the transverse component of the dynamical susceptibility is affected there. The effect has often been neglected in previous studies. Later we will discuss the importance of this effect in more detail.

If we take into account the first two of these effects, the sum rule condition in the paramagnetic state, for instance, is expressed by

$$\frac{3T_0}{T_A}[2A(y,t) + A(y_z,t) - c_z(2y + y_z)] + \frac{\sigma^2}{4} = \frac{9T_0}{T_A}A(0,t_c). \tag{2.12}$$

The magnetic equation of state is now derived as follows. According to Takahashi (1986), equation (2.12) can be regarded as a first-order ordinary differential equation for y as a function of σ if we substitute y_z in (2.11) into (2.12). By solving for y_z in terms of y and σ, we can determine the first derivative $\partial y/\partial \sigma$ as a function of y and σ. What we need to do is therefore to deal with a single first-order differential equation rather than two coupled integro-differential equations as in the case of the SCR theory. The rotational invariance requirement is automatically satisfied.

3. Equation of state in the ground state

As a simple illustration, let us first reproduce our previous results (Takahashi 1986) on the magnetic isotherm in the ground state. Because of the absence of the thermal spin fluctuation amplitude, equation (2.12) reduces to the following simplified form of a linear first-order differential equation for y:

$$\frac{\sigma^2}{4} - \frac{3c_zT_0}{T_A}(2y + y_z) = \frac{\sigma^2}{4} - \frac{3c_zT_0}{T_A}\left(3y + \sigma\frac{\partial y}{\partial \sigma}\right) = \frac{9T_A}{T_0}A(0,t_c). \tag{3.1}$$

We can easily find its solution by assuming that

$$y = y_{00} + y_{10}\sigma^2. \tag{3.2}$$

On substituting (3.2) into (3.1), the coefficients y_{00} and y_{10} are determined. They are given by

$$y_{00} = -\frac{A(0,t_c)}{c_z} = -\frac{T_A}{9c_zT_0}\left\langle S_i^2 \right\rangle_T (0,t_c) \qquad y_{10} = \frac{T_A}{60c_zT_0}. \tag{3.3}$$

The saturation moment σ_s is obtained from the condition $y = 0$ (i.e., $h = 0$):

$$\sigma_s^2 = -\frac{y_{00}}{y_{10}} = \frac{1}{c_z y_{10}} A(0, t_c) = \frac{60T_0}{T_A} A(0, t_c)$$
$$= \frac{20T_0}{T_A} C_{4/3} t_c^{4/3} \qquad (\text{for } t_c \ll 1) \tag{3.4}$$

where the t-dependence of the thermal amplitude of (2.7) is used in the last line. In terms of σ_s^2, the solutions for y and y_z are also expressed as follows:

$$y = y_{10}\sigma_s^2 \left(\frac{\sigma^2}{\sigma_s^2} - 1\right)$$
$$y_z = y + 2y_{10}\sigma^2 = y_{10}\sigma_s^2 \left(3\frac{\sigma^2}{\sigma_s^2} - 1\right) = 2y_{10}\sigma_s^2 + 3y. \tag{3.5}$$

The σ^2-linear dependence of y in (3.5) is equivalent to the free-energy expansion in powers of M up to the fourth-order term. Let us take the expansion and the magnetic equation of states in terms of the original thermodynamic variable M and H as follows:

$$F(M) = F(0) + \frac{1}{2(g\mu_B)^2\chi_0} M^2 + \frac{1}{4} F_{10} M^4$$
$$H = \frac{\partial F(M)}{\partial M} = \frac{1}{(g\mu_B)^2\chi_0} M + F_{10} M^3. \tag{3.6}$$

Then in reduced units the second line of (3.6) is also represented by

$$2h = \frac{N_0}{\chi_0}\sigma + \frac{\bar{F}_{10}}{4}\sigma^3 \qquad \bar{F}_{10} = (2\mu_B)^4 N_0^3 F_{10} = \frac{2T_A^2}{15c_z T_0}. \tag{3.7}$$

The above result is interesting because the fourth-order coefficient $\bar{F}_{10}$ is unrelated to the form of the single-particle density-of-states curve $\rho(\varepsilon)$ around the Fermi level ε_F, in contrast with the situation in Stoner–Wohlfarth theory and that in SCR theory. According to (3.7), it is related instead to the collective magnetic excitation spectrum, i.e. the spectral widths T_0 and T_A of spin fluctuations. The reason for this can be given as follows. Even in the ground state with no thermal fluctuation amplitude, the response of the system is still governed by the reaction of low-lying magnetic excitations to the external perturbation of the magnetic field. In this case, the quantum amplitude is suppressed and its reduced amplitude is transformed into a uniform magnetization since the total amplitude remains unchanged. The field dependence of the magnetization is therefore determined by the property of magnetic fluctuation. On the other hand, excitations associated with the redistribution of single-particle occupation numbers are higher in energy.

We can check the validity of (3.7) experimentally. The value of $\bar{F}_{10}$ is estimated from the magnetization measurements. If we plot the observed squared magnetization M^2 against the ratio H/M, i.e. in the form of an Arrott plot (Arrott 1957), $\bar{F}_{10}$ can be obtained from the slope of the curve. We show in the second column of table 2 values of $\bar{F}_{10}$ thus obtained experimentally for several compounds at low temperature. These values compare well with the values of $4T_A^2/15T_0$ in the third column calculated by using the parameters T_0 and T_A obtained from other independent measurements. The values of T_0 and T_A shown in the table are those estimated directly from neutron scattering experiments for MnSi (Ishikawa *et al* 1985) and Ni_3Al (Bernhoeft *et al* 1983, 1986). The values of T_0 can also be evaluated from the analysis of the temperature dependence of the NMR relaxation time; those listed in the table are estimated from the measurements by Yasuoka *et al* (1978), Umemura and Masuda (1983), Hioki and Masuda (1977), Kontani (1977), and Yoshimura *et al* (1987) for MnSi,

$Ni_{74.7}Al_{25.3}$, Sc_3In, $ZrZn_2$, and $Y(Co_{1-x}Al_x)_2$, respectively. The values of T_A in the table are then determined from these T_0 and the observed values of σ_s and T_c by using (3.4). It is quite difficult to envisage a close correlation between the observed $\bar{F}_{10}$ and the values obtained from (3.7) if the fourth expansion coefficient F_{10} was determined by the density-of-states curve around the Fermi energy ε_F; i.e.

$$F_{10} = [\rho'(\varepsilon_F)/\rho(\varepsilon_F)]^2 - \rho''(\varepsilon_F)/3\rho(\varepsilon_F).$$

Table 2. Comparison of the results from (3.7) with the observed coefficients $\bar{F}_{10}$ given by (a) Bloch *et al* (1975) (at 4.2 K), (b) Sasakura *et al* (1984), (c) Takeuchi and Masuda (1979), (d) Ogawa (1976), and (e) Yoshimura *et al* (1987). The values of T_0 and T_A marked with the superscripts † were obtained from neutron scattering experiments.

Compound	$\bar{F}_{10}$ (K)	$4T_A^2/15T_0$ (K)	T_0 (K)	T_A (K)
$MnSi^{(a)}$	9.71×10^3	5.0×10^3	$231^\dagger$	$2.08 \times 10^{3\dagger}$
		6.9×10^3	171	2.11×10^3
Ni_3Al		0.71×10^5	$3590^\dagger$	$3.09 \times 10^{4\dagger}$
$Ni_{74.7}Al_{25.3}{}^{(b)}$	1.03×10^5	1.53×10^5	2860	4.05×10^4
$Sc_3In^{(c)}$	2.00×10^5	0.66×10^5	565	1.18×10^4
$ZrZn_2{}^{(d)}$	1.05×10^4	6.5×10^4	321	8.83×10^3
$Y(Co_{0.87}Al_{0.13})_2{}^{(e)}$	2.1×10^4	1.57×10^4	2290	1.16×10^4
$Y(Co_{0.85}Al_{0.15})_2$	1.0×10^4	0.51×10^4	2119	6.34×10^3
$Y(Co_{0.83}Al_{0.17})_2$	1.6×10^4	0.63×10^4	2093	7.03×10^3

With the use of (3.7) we can also derive useful relations that enable us to estimate the microscopic spin fluctuation parameters T_0 and T_A. By eliminating either T_A or T_0 from both of (3.4) and (3.7), we can represent the ratios T_0/T_c and T_A/T_c in terms of T_c, σ_s, and $\bar{F}_{10}$, quantities readily available from macroscopic magnetic measurements:

$$\left(\frac{T_c}{T_0}\right)^{5/6} = \frac{\sqrt{30c_z}\sigma_s^2}{40C_{4/3}}\left(\frac{\bar{F}_{10}}{T_c}\right)^{1/2}$$
$$\left(\frac{T_c}{T_A}\right)^{5/3} = \frac{\sigma_s^2}{20C_{4/3}}\left(\frac{2}{15c_z}\right)^{1/3}\left(\frac{T_c}{F_{10}}\right)^{1/3}. \tag{3.8}$$

We show in table 3 the values of T_0 and T_A estimated from (3.8) for the compounds listed in table 2 by using observed values of T_c, σ_s, and $\bar{F}_1$. The table also includes results of the analysis for $(ZrTi)Zn_2$, $(ZrHf)Zn_2$ (Ogawa 1968), $Fe_xCo_{1-x}Si$ (Shimizu *et al* 1990), Y_2Ni_x (Nakabayashi *et al* 1992), Y_2Ni_{15}, Y_2Ni_{17}, YNi_3 (Gignoux *et al* 1980a, b), and $Pt_{1-x}Ni_x$ (Beille *et al* 1974, Beille 1975).

The values of T_0 and T_A in the last two columns (denoted by $T_0^\dagger$ and $T_A^\dagger$) are reproduced from table 2 for comparison. The values of $T_A^\dagger$ for $Fe_xCo_{1-x}Si$ are determined from the slope of the H/M versus M^4 plot at the critical temperature (Shimizu *et al* 1990) as will be explained in a later section.

4. Magnetic equation of state in the paramagnetic phase

In the paramagnetic state, the magnetic equation of state is obtained by solving the following equation for y as a function of σ:

$$\frac{3T_0}{T_A}[2A(y,t) + A(y_z,t) - c_z(2y + y_z)] + \frac{\sigma^2}{4} = \frac{9T_0}{T_A}A(0,t_c). \tag{4.1}$$

Table 3. Values of T_0, T_A estimated experimentally from the observed T_c, σ_s, and $\bar{F}_1$ by (a) Bloch *et al* (1975), (b) de Boer *et al* (1969), (c) Umemura and Masuda (1983) (powder), (d) Sasakura *et al* (1984), (e) Hioki and Masuda (1977) (powder), (f) Takeuchi and Masuda (1979), (g) Ogawa 1976, and (h) Knapp *et al* (1971).

Compound	T_c (K)	σ_s (μ_B)	$\bar{F}_1$ (K)	T_0 (K)	T_A (K)	$T_0^\dagger$ (K)	$T_A^\dagger$ (K)
$MnSi^{(a)}$	30	0.4	9.71×10^3	155	2.18×10^3	231	2.08×10^3
$Ni_3Al^{(b)}$	41.5	0.075	1.30×10^5	2760	3.67×10^4	3590	3.09×10^4
$Ni_3Al^{(c)}$	40	0.0692	1.68×10^5	2703	4.13×10^4		
$Ni_{74.7}Al_{25.3}{}^{(d)}$	23.2	0.047	1.03×10^5	3840	3.85×10^4		
$Sc_3In^{(e)}$	6	0.081	4.72×10^5	479	9.21×10^3	565	1.18×10^4
$Sc_{0.7575}In_{0.2425}{}^{(f)}$	5.5	0.045	2.00×10^5	286	1.46×10^4		
$ZrZn_2{}^{(g)}$	21.3	0.12	1.05×10^4	1390	7.40×10^3	321	8.83×10^3
$ZrZn_{1.9}{}^{(h)}$	26	0.16	8.21×10^3	1110	5.85×10^3		
$Zr_{0.92}Ti_{0.08}Zn_2$	40	0.233	1.49×10^4	628	5.92×10^3		
$Zr_{0.8}Ti_{0.2}Zn_2$	49.4	0.278	1.68×10^4	536	5.81×10^3		
$Zr_{0.9}Hf_{0.1}Zn_2$	10.2	0.078	1.20×10^4	1110	7.07×10^3		
$Y(Co_{1-x}Al_x)_2$							
$x = 0.13$	7	0.042	2.10×10^4	1.92×10^3	1.23×10^4	2.290×10^3	1.16×10^4
$x = 0.14$	15	0.094	1.10	1.44	0.772		
$x = 0.15$	26	0.138	1.00	1.41	0.726	2.119	0.634
$x = 0.16$	22	0.130	0.95	1.28	0.676		
$x = 0.17$	16	0.095	1.56	1.27	0.846	2.093	0.703
$x = 0.18$	9	0.063	2.77	0.984	1.01		
$x = 0.19$	7	0.040	4.11	1.28	1.40		
$Fe_xCo_{1-x}Si$							
$x = 0.36$	23	0.11	5.79×10^4	0.69×10^3	1.2×10^4		0.727×10^3
$x = 0.48$	48	0.19	3.16	0.87	1.0		0.727
$x = 0.67$	55	0.22	3.82	0.68	0.99		0.725
$x = 0.77$	40	0.18	9.76	0.38	1.2		0.824
$x = 0.88$	28	0.13	18.03	0.32	1.5		0.917
$x = 0.91$	14	0.07	57.60	0.23	2.2		1.268
Y_2Ni_x							
$x = 7.0$	52	0.033	22.7×10^5	5.172×10^3	21×10^4		
$x = 6.9$	52	0.047	9.45	3.799	11.6		
$x = 6.8$	60	0.064	7.28	2.580	8.39		
$x = 6.7$	58	0.078	6.03	1.723	6.24		
$x = 15$	119	0.15	9.87×10^4	3.33	3.51		
$x = 17$	140	0.27	6.79	1.54	1.98		
$YNi_{2.9}$	32	0.047	9.78×10^5	1.706	7.91		
YNi_3	30	0.04	10.4	2.18	9.23		
$Pt_{1-x}Ni_x$							
$x = 0.429$	23.0	0.051	5.84×10^4	4.37×10^3	3.07×10^4		
$x = 0.452$	54.2	0.104	4.45	3.67	2.46		
$x = 0.476$	74	0.143	3.74	3.12	2.08		
$x = 0.502$	100	0.179	3.90	2.87	2.04		

In this case, y always becomes finite even in the absence of an external magnetic field. We can determine its initial condition, the value of $y = y_0$ ($=y_z$) for $\sigma = 0$ ($h = 0$), by solving

$$A(y_0, t) - c_z y_0 = A(0, t_c). \tag{4.2}$$

Starting from $y = y_0$ at $\sigma = 0$, the magnetic isotherm is obtained by numerically integrating (4.1) at any given temperature.

The temperature dependence of the reciprocal magnetic susceptibility is determined as the solution of (4.2). For instance around $t = t_c$, with the use of the critical dependence on y and t, equation (2.9), of the thermal amplitude, equation (4.2) is approximated by

$$\frac{1}{3}C_{4/3}\left(t^{4/3} - t_c^{4/3}\right) - \frac{\pi t}{4}\sqrt{y_0} = 0. \tag{4.3}$$

The y_0-linear term due to the quantum amplitude can be neglected here compared with the square-root term because y_0 is very small. The critical temperature dependence of y_0 is therefore given by

$$y_0 = \left[\frac{4}{3\pi t}C_{4/3}\left(t^{4/3} - t_c^{4/3}\right)\right]^2. \tag{4.4}$$

At general temperature we need to solve (4.2) numerically. It is already known that a Curie–Weiss-like temperature dependence is derived over a wide temperature range from the equation.

4.1. Universal relation for the magnetic susceptibility

Equation (4.2) has an interesting property as will be explained below. We have already argued that the coefficient c_z will have a magnitude of order unity. If the temperature dependence of the reciprocal magnetic susceptibility is represented in the form of the t-dependence of y, equations for various compounds look the same independently of the material parameters specific to them. From this scaling property we can derive the relation between the ratio σ_e/σ_s and the critical temperature T_c.

In our present units, the Curie–Weiss law for the magnetic susceptibility is given by

$$(g\mu_B)^2\chi = \frac{N_0\mu_B^2\sigma_e^2}{3(T - T_c)}. \tag{4.5}$$

In reduced units, it is also represented by

$$\frac{\chi}{N_0} = \frac{1}{2T_A y} = \frac{\sigma_e^2}{12T_0(t - t_c)}. \tag{4.6}$$

We can now associate the effective moment σ_e with the slope of the t-dependence of y as follows:

$$\sigma_e^2 = \frac{6T_0(t - t_c)}{T_A y} = 6\frac{\sigma_s^2}{20C_{4/3}t_c^{4/3}}\frac{t - t_c}{y} \simeq \frac{3\sigma_s^2}{10C_{4/3}t_c^{4/3}}\frac{1}{\mathrm{d}y/\mathrm{d}t} \tag{4.7}$$

where from (3.4) we used the relation

$$\frac{T_0}{T_A} = \frac{\sigma_s^2}{20C_{4/3}t_c^{4/3}}.$$

On the other hand, the slope of the curve $\mathrm{d}y/\mathrm{d}t$ is a universal constant determined by solving (4.2). From these results, it follows that the ratio σ_e/σ_s is determined as a function of the single parameter $t_c = T_c/T_0$, i.e.,

$$\frac{\sigma_e^2}{\sigma_s^2} = \frac{3}{10C_{4/3}t_c^{4/3}}\frac{1}{\mathrm{d}y/\mathrm{d}t}. \tag{4.8}$$

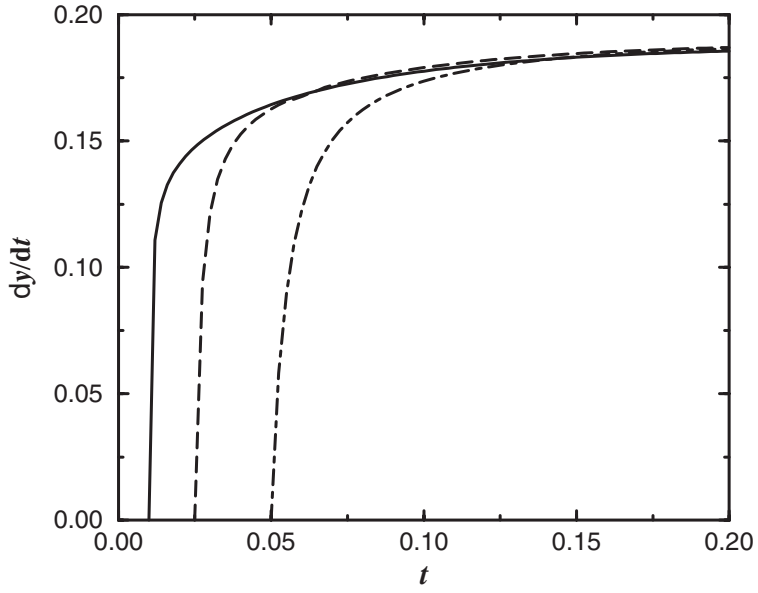

Figure 2. Numerical results for the temperature dependence of $\mathrm{d}y/\mathrm{d}t$ for $t_c = 0.01$ (solid line), 0.05 (dashed line), and 0.1 (dot–dashed line).

We show in figure 2 the numerically calculated t-dependence of the slope $\mathrm{d}y/\mathrm{d}t$ for several values of t_c. If we employ the numerical estimate of $\mathrm{d}y/\mathrm{d}t \simeq 0.17$, the following relation is derived:

$$\frac{\sigma_e^2}{\sigma_s^2} \simeq 1.75\, t_c^{-4/3}. \tag{4.9}$$

To test the above t_c-dependence, we plotted in figure 3 the observed σ_e/σ_s as a function of $t_c = T_c/T_0$ for various compounds listed in table 4. The solid curve in the figure stands for the above theoretical result (4.9). In order to show the validity of the explicit $t_c^{-4/3}$-dependence, values of $(\sigma_e/\sigma_s)^2$ are also plotted against $t_c^{-4/3}$ in figure 4 for $Y(Co_{1-x}Al_x)_2$. On the other hand, in the Rhodes–Wohlfarth plot (Rhodes and Wohlfarth 1963), σ_C/σ_s is plotted against T_c (σ_C is defined by $\sigma_C(\sigma_C + 2) = \sigma_e^2$). We reproduce in figure 5 the comparison between these two types of plot (Nakabayashi *et al* 1992) for the same compounds as are listed in table 4. From the figure we can clearly see the regularity when results are plotted according to our proposal for most of the itinerant weak ferromagnets. Heisenberg magnets fall on the straight line with the constant ratio $\sigma_C/\sigma_s = 1$ in the Rhodes–Wohlfarth plot, whereas in our plot they are all located around the narrow region in the limit of our theoretical curve with $\sigma_e/\sigma_s \simeq 1$ and $T_c/T_0 \simeq 1$.

4.2. Equation of state in the paramagnetic phase

In the presence of a magnetic field, we can solve the equation by assuming the following field dependence of y and y_z for small σ^2-values:

$$\begin{aligned} y &= y_0 + y_1\sigma^2 + \cdots \\ y_z &= y + \sigma\frac{\partial y}{\partial \sigma} = y_0 + 3y_1\sigma^2 + \cdots. \end{aligned} \tag{4.10}$$

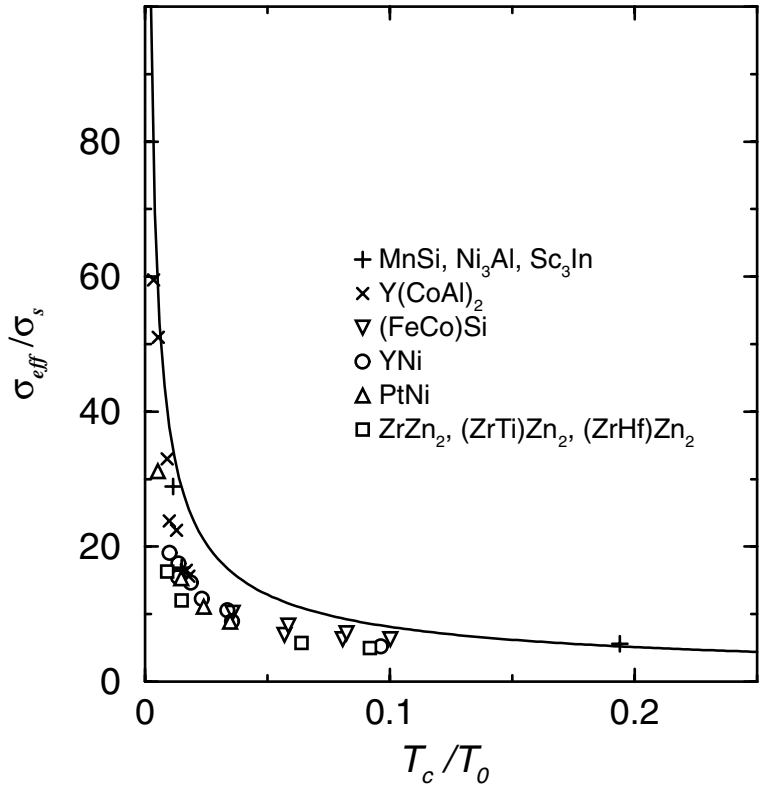

Figure 3. The universal relation between the ratio σ_e/σ_s and t_c $(=T_c/T_0)$.

The solution has the same expansion form as the ground-state one. It is equivalent to the free-energy expansion in powers of M^2. In the same way as for (3.7) in the ground state, let us assume that the σ-dependence of y is given as follows:

$$y = \frac{1}{T_A}\frac{h}{\sigma} = \frac{N_0}{2\chi(T)T_A}\sigma + \frac{\bar{F}_1}{8T_A}\sigma^2 + \cdots.$$

From comparison with (4.10), the relation $\bar{F}_1 = 8T_A y_1$ is then obtained. After substitution of (4.10), equation (4.1) can be expanded up to the σ^2-linear term as follows:

$$\frac{3T_0}{T_A}\left\{3A(y_0,t) + 5A'(y_0,t)y_1\sigma^2 - c_z\left(3y_0 + 5y_1\sigma^2\right)\right\} + \frac{\sigma^2}{4} = \frac{9T_0}{T_A}A(0,t_c)$$

where $A'(y,t)$ is the partial y-derivative of the amplitude $A(y,t)$ given by

$$A'(y,t) = \frac{1}{t}\int_0^1 \mathrm{d}x\, x^4[1/u + 1/2u^2 - \psi'(u)]. \tag{4.11}$$

Comparing the σ^2-linear coefficients, y_1 and therefore $\bar{F}_1$ are given as follows:

$$y_1 = \frac{T_A}{60T_0[c_z - A'(y_0,t)]} = \frac{y_{10}}{1 - A'(y_0,t)/c_z}$$

$$\bar{F}_1(T) = 8T_A y_1 = \frac{2T_A^2}{15T_0[c_z - A'(y_0,t)]} = \frac{\bar{F}_1(0)}{1 - A'(y_0,t)/c_z} \quad (F_1(0) = F_{10}).$$

Table 4. The observed moment ratio σ_e/σ_s and T_c/T_0. The values of T_0 are estimated from the slope of an Arrott plot according to (3.8). The effective moments for the first four compounds are from (a) Yasuoka *et al* (1978), (b) de Boer *et al* (1969), (c) Hioki and Masuda (1977), and (d) Kontani *et al* (1975).

Compound	$\sigma_s(\mu_B)$	σ_e (μ_B)	T_c (K)	T_0 (K)	σ_e/σ_s	T_c/T_0
MnSi	0.4	2.25[a]	30	155	5.6	0.194
Ni_3Al	0.077	1.3[b]	41.5	2760	16.9	0.015
Sc_3In	0.045	1.3[c]	5.5	479	28.9	0.0115
$ZrZn_2$	0.12	1.44[d]	21.3	1390	12.0	0.015
$Zr_{0.92}Ti_{0.08}Zn_2$	0.233	1.33	40.0	628	5.71	0.064
$Zr_{0.8}Ti_{0.2}Zn_2$	0.278	1.38	49.4	536	4.96	0.092
$Zr_{0.9}Hf_{0.1}Zn_2$	0.078	1.27	10.2	1110	16.3	0.0092
$Y(Co_{1-x}Al_x)_2$						
$x = 0.13$	0.042	2.50	7	1920	59.5	0.0036
$x = 0.14$	0.094	2.24	15	1440	23.8	0.010
$x = 0.15$	0.138	2.15	26	1410	15.6	0.018
$x = 0.16$	0.130	2.14	22	1280	16.5	0.017
$x = 0.17$	0.095	2.13	16	1270	22.4	0.013
$x = 0.18$	0.063	2.08	9	984	33.0	0.0091
$x = 0.19$	0.040	2.04	7	1280	51.0	0.0055
$Fe_xCo_{1-x}Si$						
$x = 0.36$	0.11	1.12	23	640	10.2	0.0359
$x = 0.48$	0.19	1.32	48	841	6.9	0.0571
$x = 0.67$	0.22	1.39	55	680	6.3	0.0809
$x = 0.77$	0.18	1.13	40	399	6.3	0.1002
$x = 0.83$	0.13	0.94	28	340	7.2	0.0824
$x = 0.91$	0.07	0.58	14	239	8.3	0.0586
Y_2Ni_x						
$x = 7$	0.033	0.631	52	5172	19.1	0.0101
$x = 6.9$	0.047	0.728	52	3799	15.5	0.0137
$x = 6.8$	0.064	0.786	60	2580	12.3	0.0233
$x = 6.7$	0.078	0.826	58	1723	10.6	0.0337
$x = 17$	0.27	0.729	149	1544	5.22	0.0965
$x = 15$	0.15	0.677	119	3329	8.97	0.0357
$YNi_{2.9}$	0.047	0.693	32	1706	14.7	0.0188
YNi_3	0.04	0.70	30	2178	17.5	0.0138
$Pt_{1-x}Ni_x$						
$x = 0.429$	0.051	1.59	23	4370	31.2	0.0053
$x = 0.452$	0.104	1.59	54.2	3670	15.3	0.0148
$x = 0.476$	0.143	1.59	75	3120	11.1	0.0240
$x = 0.502$	0.179	1.59	100	2870	8.89	0.0348

Numerical results for the temperature dependences of y and y_1/y_{10} are shown in figure 6. Because $A'(y_0, t)$ is always positive, $\bar{F}_1$ is slightly smaller than the ground-state value by 20 to 30% in the paramagnetic state except around the critical point. It is important to realize that the fourth expansion coefficient $\bar{F}_1(T)$ is also dependent on the temperature and external field, though its dependence is weak except around $t = t_c$.

As we approach the critical point, the derivative $A'(y_0, t)$ shows the divergent behaviour $-\pi t/8\sqrt{y_0}$ around $y_0 = 0$. From the $(t^{4/3} - t_c^{4/3})^2$-dependence of y_0 around $t = t_c$ in (4.4),

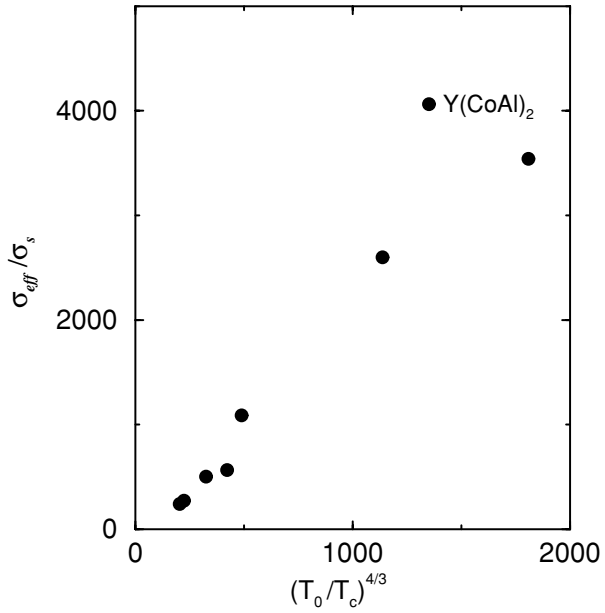

Figure 4. The linear relation between $(\sigma_e/\sigma_s)^2$ and $t_c^{4/3}$ for $Y(Co_{1-x}Al_x)_2$.

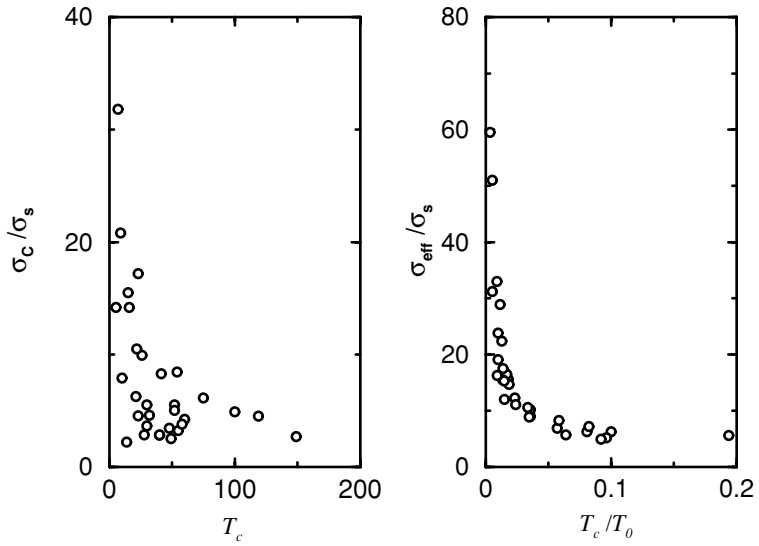

Figure 5. Comparison between two types of plot, σ_C/σ_s versus T_c (the Rhodes–Wohlfarth plot) and σ_e/σ_s versus T_c/T_0.

the following critical temperature dependence of y_1 is obtained:

$$y_1 \sim \frac{8c_z y_{10}}{\pi t_c}\sqrt{y_0} = \frac{8c_z y_{10}}{3\pi t_c}\frac{4C_{4/3}}{3\pi t_c}\left(t^{4/3} - t_c^{4/3}\right) = \frac{32c_z y_{10} C_{4/3}}{3\pi^2 t_c^2}\left(t^{4/3} - t_c^{4/3}\right). \tag{4.12}$$

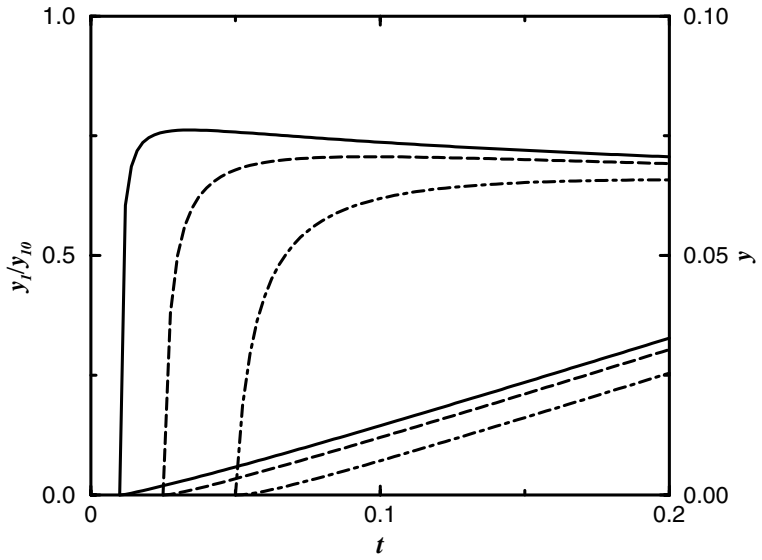

Figure 6. The t-dependence of y and y_1/y_{10} in the paramagnetic state for $t_c = 0.01$ (solid line), 0.05 (dashed line), and 0.1 (dot–dashed line).

Therefore $\bar{F}_1$ shows the same temperature dependence as that of $\sqrt{y_0}$ and decreases towards the critical point $t = t_c$ according to the following temperature dependence as shown in figure 6:

$$\frac{\bar{F}_1(T)}{\bar{F}_1(0)} = \frac{y_1}{y_{10}} = \frac{32 c_z C_{4/3}}{3\pi^2 t_c^{2/3}} \left[\left(\frac{T}{T_c} \right)^{4/3} - 1 \right]. \tag{4.13}$$

When we compare the result with experiments, we must be aware that we are discussing the inverse of the *initial* slope of the Arrott plot (M^2 versus H/M plot) in the weak-field limit. If we estimate the slope in the presence of the field, it is likely to be underestimated. With increasing field strength, the slope $\mathrm{d}\sigma^2/\mathrm{d}y$ of each magnetic isotherm decreases monotonically, and y_1 approaches the value y_{10}. Numerical results for the equation of state throughout the wide temperature range will be given later after we have discussed the magnetic properties for the ordered state below t_c.

4.3. Critical magnetization process

At the critical temperature $t = t_c$, both components y and y_z of the reciprocal susceptibility are very small around $\sigma = 0$ because both of them just vanish for $\sigma = 0$. Each of the thermal amplitudes is then dominated by the following square-root dependence:

$$A(y, t_c) \simeq A(0, t_c) - \frac{\pi t_c}{4}\sqrt{y} \quad A(y_z, t_c) \simeq A(0, t_c) - \frac{\pi t_c}{4}\sqrt{y_z}.$$

Hence the sum rule condition (2.12) is approximated by

$$\sigma^2 = \frac{3\pi T_c}{T_A}(2\sqrt{y} + \sqrt{y_z}) + \mathrm{O}(y, y_z). \tag{4.14}$$

In the right-hand side, both the y- and the y_z-linear terms, coming from the implicit magnetic field dependence of the quantum amplitude, are neglected compared with the first two square-root terms. We can easily see that (4.14) has the following solution in the $\sigma = 0$ limit:

$$y = y_c \sigma^\beta \quad y_z = y + \sigma \frac{\partial y}{\partial \sigma} = (1+\beta) y_c \sigma^\beta. \tag{4.15}$$

On substituting (4.15) into (4.14), the numerical constant y_c and the exponent β are determined as follows:

$$y_c = \left\{ \frac{T_A}{3\pi T_c (2+\sqrt{5})} \right\}^2 = \left\{ \frac{20 c_z y_{10}}{\pi (2+\sqrt{5}) t_c} \right\}^2 \quad \beta = 4. \tag{4.16}$$

We are thus led to the critical magnetization process:

$$y = \frac{h}{T_A \sigma} = \frac{1}{T_A} \frac{2\mu_B H}{M/(N_0 \mu_B)} = y_c \left(\frac{M}{N_0 \mu_B} \right)^4.$$

In a similar form to the Arrott plot, it is also represented as follows:

$$\left(\frac{\sigma}{\sigma_s} \right)^4 = \frac{3 t_c^{2/3}}{C_{4/3}} \left[\frac{\pi (2+\sqrt{5})}{20} \right]^2 \frac{y}{A(0, t_c)}. \tag{4.17}$$

In figure 7 numerical results from (4.1) at $t = t_c$ are shown for several values of t_c. The linear relation between $(\sigma/\sigma_s)^4$ and $y/A(0, t_c)$ is evident from the figure. We can also see the t_c-dependence of the slope of the plots, i.e. the slopes become steeper for larger t_c.

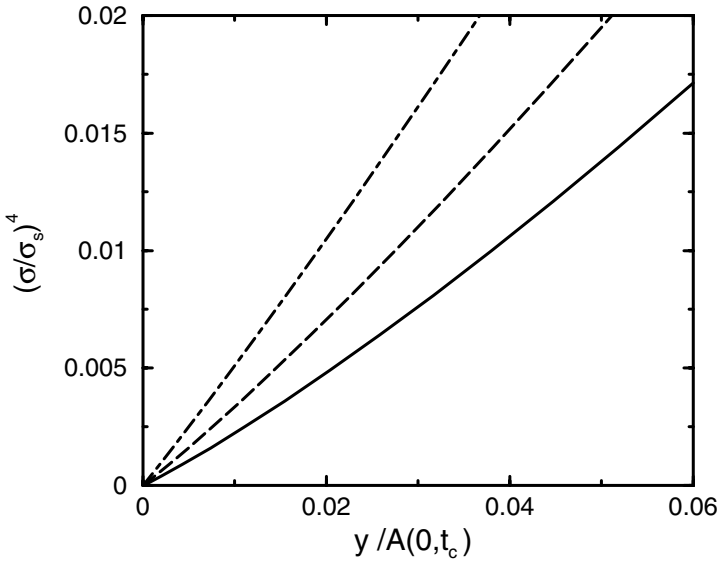

Figure 7. The critical magnetization process for $t_c = 0.05$ (solid line), 0.1 (dashed line), and 0.2 (dot–dashed line).

In terms of the original variables H and M, it is expressed by

$$M^4 = 2[3\pi (2+\sqrt{5})]^2 N_0^4 \mu_B^6 \frac{T_c^2}{T_A^3} \frac{H}{M}$$

or, in a more convenient form for experimental analyses,

$$\left(\frac{M}{M_s}\right)^4 = 1.20 \times 10^6 \frac{T_c^2}{T_A^3 \sigma_s^4} \frac{H}{M} \tag{4.18}$$

if H and M are measured in units of kOe and emu mol^{-1}, respectively. The above relation is useful in estimating the value of T_A. If the slope is determined experimentally from the M^4–H/M plot at $T = T_c$, T_A is evaluated from the observed values of σ_s and T_c. In the case of MnSi the Arrott plot of the observed M–H curve (Bloch *et al* 1975) shows a good linearity between M^2 and H/M at low temperature. At the critical temperature, on the other hand, it is rather well fitted with the relation $(M/M_s)^4 = 0.234\, H/M$ (kOe g emu^{-1}). With the use of $T_c = 30$ K and $\sigma_s = 0.4$ from table 3, we obtain $T_A = 1.29 \times 10^3$ K which compares well with $T_A = 2.08 \times 10^3$ K obtained from the neutron scattering experiments (Ishikawa *et al* 1985). Such an analysis was also performed for $Fe_xCo_{1-x}Si$ by Shimizu *et al* (1990). The values of T_A estimated in this way are shown in the last column of table 3. They also agree well with those estimated by using (3.8) within a factor of 2.

5. Magnetic equation of state in the ordered state

According to the Stoner–Wohlfarth theory (Stoner 1936, Wohlfarth 1968, Edwards and Wohlfarth 1968), the temperature and field dependence of the magnetization $M(H, T)$ obeys the following equation:

$$M^2(H, T) = M^2(0, 0)\left[1 - (T/T_c)^2\right] + \frac{2\chi_{z0} H}{M(H, T)} \tag{5.1}$$

where χ_{z0} is the longitudinal (differential) magnetic susceptibility. The same T^2-dependence of $M^2(0, T)$ has been predicted by the SCR theory at low temperature, based on the spin fluctuation mechanism. In the latter, however, $M^2(0, T)$ follows a $(T_c^{4/3} - T^{4/3})$-dependence at higher temperature around T_c. Although there have been lots of experiments on the temperature and field dependence of the magnetization in the ordered state, neither comprehensive nor quantitative analyses of them have yet been performed from the latter point of view. This is the purpose of this section.

According to our view, the magnetic properties of the system in its ordered state are treated on the basis of the following sum rule:

$$\frac{3T_0}{T_A}[2A_t(y, t) + A(y_z, t) - c_z(2y + y_z)] + \frac{\sigma^2}{4} = \frac{9T_0}{T_A} A(0, t_c) \tag{5.2}$$

where $A_t(y, t)$ is the transverse thermal amplitude in the presence of the spontaneous magnetic moment. With the use of the parameter y_{10} defined by (3.3), it is also given by

$$2A_t(y, t) + A(y_z, t) - c_z(2y + y_z) + 5c_z y_{10}\sigma^2 = 3c_z y_{10}\sigma_s^2 \tag{5.3}$$

where

$$y_z = y + \sigma \frac{\partial y}{\partial \sigma}.$$

To solve the above differential equation, we need an initial condition, the value of y at some starting σ-value. In the case of the paramagnetic state, we can determine the condition with the use of the same differential equation. In contrast, it is not so easy to find it from (5.3) in the ordered state for the following reason. From the continuity of solutions, it is reasonable to

assume that the solution y of (5.3) should have the same σ^2-dependence as that of the ground state:

$$\begin{aligned} y &= y_1\left[\sigma^2 - \sigma_0^2\right] + \cdots \\ y_z &= y_1\left[3\sigma^2 - \sigma_0^2\right] + \cdots = 3y_1\left[\sigma^2 - \sigma_0^2\right] + 2y_1\sigma_0^2 + \cdots = 3y + y_{z0} + \cdots \end{aligned} \tag{5.4}$$

where y_{z0} is the reduced longitudinal reciprocal susceptibility in the absence of the external field. The above form of the solution y is equivalent to the free-energy expansion in powers of M^2. It includes two independent parameters, σ_0 and y_1. The parameter σ_0 has the meaning of the spontaneous moment per magnetic atom at general temperature below t_c. On the other hand, y_1 is proportional to the fourth-order expansion coefficient F_1 of the free energy. Even if we put $y = 0$ in (5.3), for instance, we are left with only a single condition for two independent parameters σ_0 and y_1. It seems that we require another condition to determine them. The key to resolving this difficulty is to realize that the solution (5.4) imposes a strict restriction on the form of the differential equation. Equation (5.3) has to be compatible with the above solution.

For the longitudinal amplitude, there is no difficulty because y_z is always finite below t_c. It is a well-behaved function of y_z. The transverse mode is, on the other hand, influenced by the appearance of the spin-wave mode. At the outset, it is therefore necessary to examine its effect on the dependence on y and t of the amplitude $A_t(y, t)$.

5.1. The effect of the presence of the spin wave

Due to the presence of the spin-wave mode, the transverse thermal amplitude consists of a sum of contributions:

$$A_t(y, t) = A_{sw}(t) + A_c(y, t).$$

The spin-wave contribution, denoted by $A_{sw}(t)$, comes from the spin-wave modes arising from the well-defined spin-wave pole ω_q of the transverse dynamical susceptibility at low temperature around the origin of the wave-vector space:

$$\operatorname{Im}\chi(\boldsymbol{q}, \omega) \propto \sigma\delta(\omega - \omega_q).$$

The other one is given by the wave-vector integral over spin fluctuation modes with the finite damping Γ_q outside the spin-wave region, like the expression $A(y, t)$ in (2.7). It is explicitly given by

$$A_c(y, t) = \int_{x_c}^{1} \mathrm{d}x\, x^3\left[\ln u - 1/2u - \psi(u)\right] \quad u = x(y + x^2)/t \tag{5.5}$$

where the reduced cut-off wave vector x_c represents the boundary of the spin-wave region.

Although the spin-wave modes are confined within a restricted region around the origin of the wave-vector space, they have a significant effect on the analytic property of the transverse thermal amplitude, as will be shown below. If we do not take the effect into account, for instance, the $\sqrt{y}$-dependence of (2.8) becomes dominant around $y = 0$, and (5.3) is approximated by

$$-\frac{\pi t}{4}(2\sqrt{y} + \sqrt{y_z}) + 5c_z y_{10}\sigma^2 = 5c_z y_{10} s^2(t) \tag{5.6}$$

where

$$s^2(t) = \frac{3}{5c_z y_{10}}[A(0, t_c) - A(0, t)].$$

According to the same argument as before, it is easy to see that (5.6) has a solution $y \propto [\sigma^2 - s^2(t)]^3$. Both y and y_z then vanish for $h = 0$ and the spontaneous moment σ_0 is

simply given by $s(t)$, in disagreement with the value σ_s in the $t = 0$ limit ($\sigma_s^2 = 5s^2(0)/3$). The magnetization process in the weak-field limit also shows the following spurious behaviour:

$$\sigma^2 - s^2(t) \propto (h/\sigma)^{1/3}$$

which reflects the singular non-analytic $\sqrt{y}$-dependence of the thermal amplitude. From this example, it is clear that the naive replacement of $A_t(y, t)$ with the paramagnetic form $A(y, t)$ is not compatible with the solution (5.4). This strongly suggests the necessity of examining the effect of the spin-wave mode on the analytic behaviour of the transverse amplitude as a function of y.

5.1.1. The y-dependence of the transverse thermal amplitude. In the present treatment we assume that the explicit y-dependence of the thermal amplitude comes from that of $A_c(y, t)$. Its y-dependence around $y = 0$ is evaluated as follows. It mainly originates from the integral around the origin, where the integrand is well approximated by $x^3/2u$. It is therefore given as follows:

$$\begin{aligned} A_c(y,t) - A_c(0,t) &\simeq \int_{x_c}^{1} \mathrm{d}x\; x^3 \left(\frac{1}{2u} - \frac{1}{2u_c}\right) = \frac{t}{2}\int_{x_c}^{1} \mathrm{d}x\; \left(\frac{x^2}{y+x^2} - 1\right) \\ &= \frac{t\sqrt{y}}{2}\left(\tan^{-1}\sqrt{y} - \tan^{-1}\frac{\sqrt{y}}{x_c}\right) \\ &= -\frac{t\sqrt{y}}{2}\tan^{-1}\left(\frac{\sqrt{y}(1-x_c)}{y+x_c}\right) \simeq \begin{cases} -ty/(2x_c) & \text{for } y < x_c \\ -\pi t\sqrt{y}/4 & \text{for } x_c < y \end{cases} \end{aligned} \tag{5.7}$$

where $u_c = x^3/t$. If we assume $x_c = 0$ from the beginning, the $\sqrt{y}$-behaviour recovers around $y = 0$. For finite x_c, it becomes proportional to y and is expanded in powers of y around the origin. By expanding the amplitude in y around the origin $y = 0$, we obtain the same form of equation as that for the ground state. We can therefore find its solution by assuming (5.4). In view of this meaning, the introduction of the lower bound of the integral is quite important.

Phenomenologically, let us introduce the following form of the cut-off wave vector x_c:

$$x_c = \frac{4}{\pi\xi}\sqrt{y_{z0}} = \frac{4}{\pi\xi}\sqrt{2y_1\sigma_0^2}. \tag{5.8}$$

It is linearly proportional to σ_0 at low temperature, while it decreases in proportional to σ_0^2 around the critical point as will be clarified below. This is equivalent to an assumption that the transverse thermal amplitude is suppressed almost as much as the longitudinal one by the appearance of the static uniform moment. The y- and y_z-derivatives of $\Delta A_c(y, t)$ and $\Delta A(y_z, t)$ are then given by

$$\frac{\partial \Delta A_c(y,t)}{\partial y} \simeq -\frac{\xi\pi t}{8\sqrt{y_{z0}}} \quad \frac{\partial \Delta A(y_z,t)}{\partial y_z} \simeq -\frac{\pi t}{8\sqrt{y_z}} \quad (\text{for } y, y_z \simeq 0).$$

At present, we simply assume that the numerical factor ξ is slightly larger than 1, since the transverse amplitude is more susceptible to the external field than the longitudinal one.

5.1.2. The temperature dependence of the thermal amplitude. Let us next examine the temperature dependence of the transverse thermal amplitude $A_t(y, t)$. At low temperature, because of the presence of the finite lower bound x_c, $A_c(y, t)$ shows the following t^2-dependence:

$$A_c(0,t) = \int_{x_c}^{1} \mathrm{d}x\; x^3[\ln u - 1/2u - \psi(u)] \sim \int_{x_c}^{1} \mathrm{d}x\; \frac{x^3}{12u^2} = \int_{x_c}^{1} \mathrm{d}x \frac{t^2}{12x^3} = \frac{t^2}{24x_c^2} \tag{5.9}$$

on using the asymptotic expansion of the digamma function $\psi(u)$:

$$\ln u - 1/2u - \psi(u) \sim \frac{1}{12u^2} + \cdots \quad (u \gg 1). \tag{5.10}$$

If we let $x_c = 0$, the critical $t^{4/3}$-dependence of (2.7) is recovered for $y = 0$. The presence of the lower bound is therefore important in deriving the t^2-dependence of various magnetic properties at low temperature.

To evaluate the spin-wave amplitude $A_{sw}(t)$, we simply assume that its spectrum is always approximated by the delta function $\sigma\delta(\omega - \omega_q)$. What we need is the frequency-integrated value. In our present treatment, the effect of the lifetime broadening at finite temperature on the integrated intensity is assumed to be small. Its temperature dependence is then given by the following form of the wave-vector integral:

$$A_{\rm sw}(t) = \frac{\sigma T_A}{2T_0}\int_0^{x_c} \frac{x^2}{{\rm e}^{(h+T_A\sigma x^2)/T} - 1}\,{\rm d}x$$
$$\hbar\omega_q = h + Dq_B^2x^2 = h + T_A\sigma x^2 \quad \left(Dq_B^2 = T_A\sigma\right)$$

where D represents the spin-wave stiffness constant. We determined the constant D and the pre-factor $T_A\sigma/2T_0$ before the integral, so the integrand coincides with the long-wavelength limit of the integrands of $A_c(y,t)$ and $A_c(0,t)$. At low temperature it shows the well-known $T^{3/2}$-dependence:

$$A_{\rm sw}(t) \sim \frac{\sigma T_A}{2T_0}\left(\frac{T}{\sigma T_A}\right)^{3/2}\int_0^\infty \frac{s^2}{{\rm e}^{s^2}-1}\,{\rm d}s = \frac{\sqrt{\pi}\zeta(3/2)}{8}\frac{\sigma T_A}{T_0}\left(\frac{T}{\sigma T_A}\right)^{3/2} \tag{5.11}$$

when $\sigma T_A x_c^2/T > 1$ is satisfied. If we assume that $x_c \sim \sqrt{y_{z0}}$ and $\sigma \sim \sigma_s$, which is justified at low temperature, the condition can be expressed as

$$\frac{t}{t_c} < \frac{\sigma_s T_A}{T_c}x_c^2 = 2y_{10}\sigma_s^2\frac{\sigma_s T_A}{T_c} = \frac{2C_{4/3}}{3c_z}\frac{T_A}{T_0}\sigma_s t_c^{1/3}$$

where we have replaced $y_{10}\sigma_s^2$ with $[C_{4/3}t_c^{4/3}]/3c_z$ by using (3.3) and (3.4). For most weak itinerant ferromagnets the ratio T_A/T_0 has a magnitude of about 10 (see tables 2 and 3, for instance). The above inequality is therefore equivalent to $t/t_c < 10\sigma_s t_c^{1/3}$. The spin-wave $T^{3/2}$-dependence is therefore observed at low temperature for those magnets with small σ_s- and t_c-values.

5.1.3. Initial conditions of the equation. On the basis of the foregoing discussions on the transverse thermal amplitude, let us rewrite the condition (5.2) in the form

$$s^2(t) = \sigma^2 + \frac{1}{5c_z y_{10}}\left[2\Delta A_c(y,t) + \Delta A(y_z,t) - c_z(2y + y_z)\right] - \frac{2}{5c_z y_{10}}[A(0,t) - A_t(0,t)] \tag{5.12}$$

where

$$\Delta A_c(y,t) = A_c(y,t) - A_c(0,t) \quad \Delta A(y,t) = A(y,t) - A(0,t).$$

By expanding (5.12) in y and equating each of its zeroth- and first-order coefficients, the following coupled simultaneous equations for parameters σ_0^2 and y_1 are obtained:

$$\left(1 - \frac{2}{5}\frac{y_1}{y_{10}}\right)\sigma_0^2 + \frac{1}{5c_z y_{10}}\{\Delta A(y_{z0},t) - 2[A(0,t) - A_t(0,t)]\} = s^2(t)$$
$$y_1\left[1 - \frac{2}{5c_z}\Delta A_c'(0,t) - \frac{3}{5c_z}\Delta A'(y_{z0},t)\right] = y_{10} \tag{5.13}$$

where $A'(y,t)$ is the first derivative of $A(y,t)$ defined by (4.11) and $A'_c(y,t)$ is given by

$$A'_c(y,t) = \frac{1}{t}\int_{x_c}^{1} \mathrm{d}x\; x^4[1/u + 1/2u^2 - \psi'(u)].$$

The initial condition of (5.2) can be uniquely determined by solving (5.13) for two independent parameters σ_0^2 and y_1. It follows that the value of y_1 also shows some temperature dependence. The temperature dependences of both of these parameters are closely related to each other. We show in figure 8 a typical temperature dependence of σ_0^2/σ_s^2 and y_1/y_{10} for $t_c = 0.05$. Before we show numerical results for the magnetic equation of state at general temperature below t_c, let us next examine limiting behaviours of solutions, i.e. those around the critical region and in the low-temperature region.

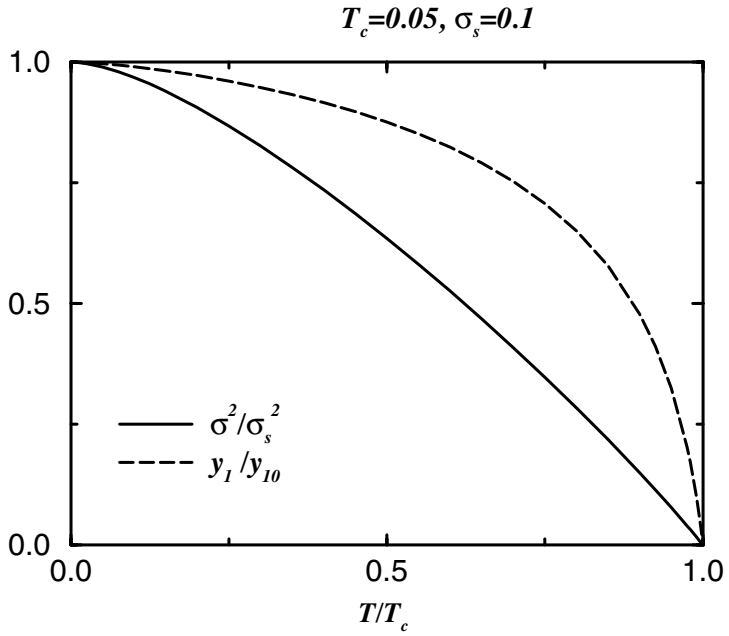

Figure 8. The temperature dependence of σ_0^2/σ_s^2 (solid line) and y_1/y_{10} (dashed line) below t_c for $t_c = 0.05$ and $\sigma_s = 0.1$.

5.2. Around the critical temperature

Because of our definition of the spin-wave amplitude, the t-dependence of the transverse thermal amplitude has the following property of continuity as we approach the critical temperature:

$$A_t(0,t) = A_{\mathrm{sw}}(t) + A_c(0,t) \to A(0,t_c) \quad (\text{for } t \to t_c).$$

For small values of y and y_z, the thermal amplitudes and their derivatives are approximated as follows:

$$\Delta A(y_z,t) \sim -\frac{\pi}{4} t\sqrt{y_z} \quad \Delta A'(y_z,t) \sim -\frac{\pi}{8}\frac{t}{\sqrt{y_z}}$$

$$\Delta A'_c(0,t) \sim -\frac{\xi\pi}{8}\frac{t}{\sqrt{y_z}}.$$

By substituting these results into (5.13), we obtain the following simultaneous equations:

$$\left(1-\frac{2}{5}\frac{y_1}{y_{10}}\right)\sigma_0^2-\frac{\pi t}{20c_z y_{10}}\sqrt{y_{z0}}=s^2(t)$$
$$y_1\left(1+\frac{\xi\pi t}{20c_z\sqrt{y_{z0}}}+\frac{3\pi t}{40c_z}\frac{1}{\sqrt{y_{z0}}}\right)=y_{10}. \tag{5.14}$$

If we recall the definition $y_{z0}=2y_1\sigma_0^2$, we can easily find a solution for y_1 of the form $y_1=y_c'\sigma_0^2$ from the second line. The coefficient y_c' is determined by

$$y_c'=\left[\frac{40\sqrt{2}c_z y_{10}}{\pi t_c(2\xi+3)}\right]^2=\left[\frac{2\sqrt{2}T_A}{3\pi T_c(2\xi+3)}\right]^2. \tag{5.15}$$

The same expression for $y_{z0}=2y_c'\sigma_0^4$ is already obtained just at the critical point, i.e. $y_z=5y_c\sigma^4$ in (4.15), if σ_0 is taken as the field-induced moment σ. In the critical limit, let us assume that y_z should have the same dependence on the moment σ irrespective of whether it is induced by the external field or appears spontaneously. We can then determine the numerical factor ξ of x_c defined in (5.8). On equating y_c in (4.16) with $2y_c'/5$, ξ is determined by the following condition:

$$\left(\frac{20}{2+\sqrt{5}}\right)^2=\frac{2}{5}\left(\frac{40\sqrt{2}}{2\xi+3}\right)^2$$

which leads to ξ_c being given by

$$\xi_c=\frac{5+8\sqrt{5}}{10}=2.288\ldots.$$

On substituting in the result, the first line of (5.14) is given by

$$\sigma_0^2\left(1-\frac{\pi t_c\sqrt{2y_c'}}{20c_z y_{10}}\right)=\sigma_0^2\left(1-\frac{4}{2\xi+3}\right)=s^2(t). \tag{5.16}$$

With the use of (2.7) for the critical thermal amplitude, the temperature dependence of $s^2(t)$ on the right-hand side is explicitly given by

$$s^2(t)=\frac{12T_0}{T_A}C_{4/3}\left(t_c^{4/3}-t^{4/3}\right)=\frac{3}{5}\sigma_s^2\left[1-(T/T_c)^{4/3}\right]. \tag{5.17}$$

The temperature dependence of σ_0^2 is therefore finally given by

$$\left(\frac{\sigma_0(t)}{\sigma_s}\right)^2\simeq\frac{3(2\xi+3)}{5(2\xi-1)}\left[1-(T/T_c)^{4/3}\right]=a_c\left[1-(T/T_c)^{4/3}\right] \tag{5.18}$$

where

$$a_c=\frac{3(2\xi+3)}{5(2\xi-1)}.$$

The numerical factor a_c is given by 1.27 for $\xi=\xi_c$. We show in table 5 the dependence of a_c on our various choices of ξ-value. The value of a_c is easily estimated experimentally as the extrapolation of the observed linear relation between σ_0^2 and $T^{4/3}$ to the $T=0$ limit. The values of a_c obtained in this way are slightly larger than 1 by 10 to 20% (Sasakura *et al* 1984, Shimizu *et al* 1990), in agreement with our estimate for $\xi=\xi_c$.

Table 5. The dependence on the numerical factor ξ.

ξ	a_c	a_F	a_0	a_T
1.0	3	12	14.1	37.3
1.5	1.8	5	25.7	64.3
2.0	1.4	2.857	56.9	112
ξ_c	1.27	2.21	89.6	153
2.5	1.2	1.875	122.7	191

According to the result given above, y_1 becomes proportional to σ_0^2 around $t = t_c$. Hence the fourth-order expansion coefficient $\bar{F}_1$ shows the same temperature dependence as σ_0^2 and is given by

$$\frac{F_1}{F_{10}} = 8T_A y_1 \frac{15c_z T_0}{2T_A^2} = \frac{y_1}{y_{10}} = \frac{y_c' \sigma_s^2}{y_{10}} \left(\frac{\sigma_0}{\sigma_s}\right)^2 = \frac{640 c_z C_{4/3}}{(2\xi - 1)(2\xi + 3)\pi^2 t_c^{2/3}} \left[1 - (T/T_c)^{4/3}\right]$$
$$= \frac{32 a_F c_z C_{4/3}}{3\pi^2 t_c^{2/3}} \left[1 - (T/T_c)^{4/3}\right] \quad a_F = \frac{60}{(2\xi - 1)(2\xi + 3)}. \tag{5.19}$$

The same temperature dependence of $\bar{F}_1$ is now derived as in the paramagnetic solution, i.e. (4.13), except for the numerical factor a_F ($a_F = 1$ for the paramagnetic case). These results for σ_0^2 and $\bar{F}_1$ prove the validity of our treatment of the ordered state as well as our expression for the cut-off vector x_c of (5.8). The t-dependence of $\bar{F}_1$ is also consistent with our critical solution, (4.18), i.e. the absence of $\bar{F}_1$ at $t = t_c$. In previous studies, the temperature dependence of $\bar{F}_1$ has not been seen as important. It would be quite interesting if the above t-dependence for $\bar{F}_1$ was confirmed experimentally. As an illustration, we show in figure 9 numerical results for the temperature dependences of $(\sigma/\sigma_s)^2$ and y_1/y_{10}

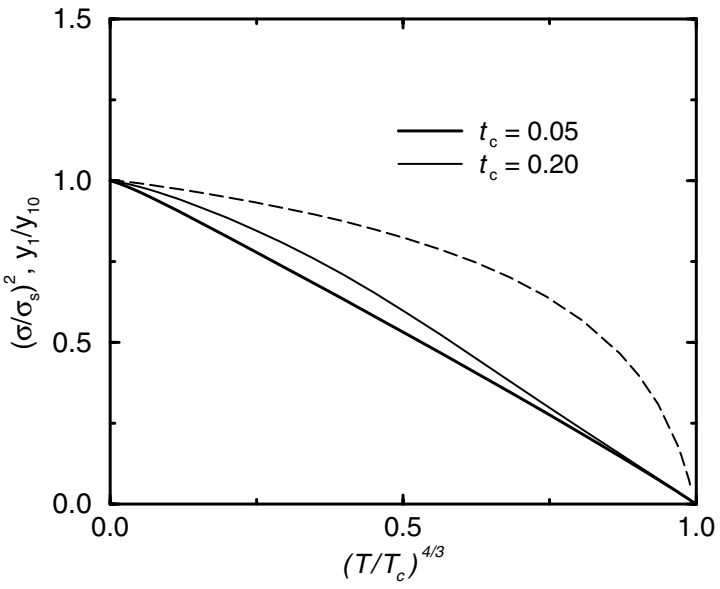

Figure 9. The $t^{4/3}$-dependence of σ^2 (thick and thin lines for $t_c = 0.05$ and 0.2) and y_1/y_{10} (dashed line for $t_c = 0.05$).

as functions of $(t/t_c)^{4/3}$. We can see from the figure that the linearity of the curve looks better for cases with smaller t_c. The calculated qualitative tendency is in accordance with the observed temperature dependence for Ni_3Al (Sasakura *et al* 1984) and (FeCo)Si (Shimizu *et al* 1990). It also looks as if the calculated slope of the curve depends slightly on t_c. According to our result (5.18), however, the slope a_c is a universal constant, independent of t_c. This means that the real critical $(T_c^{4/3} - T^{4/3})$-dependence of σ_0^2 is supported only in the very narrow region around t_c when t_c becomes smaller. The apparent overall good linearity of such a plot for compounds with small t_c is therefore not the result of critical behaviour and has a different cause.

5.3. Low-temperature behaviour

5.3.1. Temperature dependence of the spontaneous moment. At low temperature, from (5.9) the transverse thermal amplitude and its derivative show the following t^2-dependence:

$$\begin{aligned} A_t(0,t) &= A_{\mathrm{sw}}(t) + \frac{t^2}{24x_c^2} \\ \Delta A_c'(0,t) &= -\frac{t^2}{24x_c^4} = -\frac{t^2}{24}\left(\frac{\pi\xi}{4}\right)^4 \frac{1}{4y_1^2\sigma_0^4}. \end{aligned} \tag{5.20}$$

The longitudinal component and its derivative also show the same t^2-dependence:

$$A(y_z,t) = \frac{t^2}{24y_z} + \cdots \qquad A'(y_z,t) = -\frac{t^2}{24y_z^2}. \tag{5.21}$$

On substituting these expressions into the second line of (5.13), y_1 is given by

$$\begin{aligned} y_1/y_{10} &= 1 + \frac{1}{5c_z}[2\Delta A_c'(0,t) + 3\Delta A(y_{z0},t)] + \cdots \\ &= 1 - \frac{t^2}{480c_z\left(y_{10}\sigma_s^2\right)^2}\left[2\left(\frac{\pi\xi}{4}\right)^4 + 3\right] + \cdots \\ &= 1 - \frac{a_0 t_c^2}{\left(60c_z y_{10}\sigma_s^2\right)^2}\left(\frac{T}{T_c}\right)^2 + \cdots = 1 - \frac{a_0}{[20C_{4/3}]^2 t_c^{2/3}}\left(\frac{T}{T_c}\right)^2 + \cdots \\ &= 1 - \frac{a_0}{\sigma_s^4}\left(\frac{T}{T_A}\right)^2 + \cdots = 1 - b_0 T^2 + \cdots \end{aligned} \tag{5.22}$$

where

$$a_0 = \frac{15c_z}{2}\left[2\left(\frac{\pi\xi}{4}\right)^4 + 3\right]$$

where we have used $c_z y_{10} = T_A/60T_0 = C_{4/3}t_c^{4/3}/3\sigma_s^2$ from (3.4) and $y_{z0} = 2y_1\sigma_0^2 \simeq 2y_{10}\sigma_s^2$. If we assume $c_z = 1/2$ and $\xi = \xi_c$ for instance, we obtain $a_0 = 89.6$. The fourth expansion coefficient y_1 therefore decreases from its ground-state value y_{10} according to the t^2-dependence at low temperature. In spite of numerous experiments on the temperature dependence of the spontaneous moment, only a few analyses are available in the literature for the t-dependence of F_1. For example, Wohlfarth and de Chatel (1970) estimated the coefficient b_0 from the observed isotherms for $ZrZn_{1.9}$ (Knapp *et al* 1971) and $Zr_{0.92}Ti_{0.08}Zn_2$ (Ogawa 1968). t^2-dependence of F_1 was also reported for $Pt_{1-x}Ni_x$ (Beille *et al* 1974). The coefficients b_0 evaluated from linear fits to the observed results are shown in table 6. For $ZrZn_{1.9}$

Table 6. Experimentally estimated slopes of the t^2-dependences of y_1/y_{10} for $ZrZn_{1.9}$, $(ZrTi)Zn_2$, and $Pt_{1-x}Ni_x$ alloy. The values of $T_A^\dagger$ are reproduced from the sixth column of table 3 for comparison.

Compound	b_0 (K^{-2})	σ_s (μ_B)	t_c	T_A (K)	$T_A^\dagger$ (K)
$ZrZn_{1.9}$	7.4×10^{-4}	0.16	0.023	1.4×10^4	5.85×10^3
$Zr_{0.92}Ti_{0.08}Zn_2$	1.13×10^{-4}	0.233	0.064	1.6×10^4	5.92×10^3
$Pt_{1-x}Ni_x$					
$x = 0.429$	3.1×10^{-5}	0.051	0.0053	4.5×10^5	3.07×10^4
$x = 0.452$	2.9	0.104	0.015	1.3×10^5	2.46×10^4
$x = 0.476$	2.3	0.143	0.024	7.7×10^4	2.08×10^4
$x = 0.502$	1.6	0.179	0.035	5.8×10^4	2.04×10^4

and $Pt_{1-x}Ni_x$ for $x = 0.476$ and 0.502, the values of T_A estimated by using (5.22) compare relatively well with those shown in table 3 (denoted by $T_A^\dagger$) obtained from the T-dependence of the slope of the Arrott plot. For the lower Ni concentrations for $Pt_{1-x}Ni_x$ with smaller t_c, the agreement is rather poor. According to the conventional non-linear mode–mode coupling mechanism of the SCR theory, the t^2-dependence of F_1 will arise from the renormalization effect of the sixth-order non-linear mode–mode coupling among spin fluctuation modes. The observed good linearity of the Arrott plot, however, suggests the absence of higher-order non-linear terms. It will therefore be difficult to explain the above t^2-dependence on the basis of such a mechanism. The analysis for $Zr_{0.92}Ti_{0.08}Zn_2$ by Wohlfarth and de Chatel (1970) also indicates the presence of higher-order terms in the series of the T^2-expansion because of the deviation from the t^2-dependence at higher temperature. This is also in accordance with our preceding conclusion that y_1 will deviate from the t^2-dependence and finally follow the $(t_c^{4/3} - t^{4/3})$-dependence around the critical point.

In order to obtain the temperature dependence of σ_0^2, let us next rewrite the first line of (5.13) in the form

$$3c_z y_{10}\left(\sigma_0^2 - \sigma_s^2\right) = -2A_t(0,t) - A\left(2y_1\sigma_0^2, t\right) + 2c_z(y_1 - y_{10})\sigma_0^2.$$

It is also represented by

$$\sigma_0^2\left[1 - \frac{2}{3}\left(\frac{y_1}{y_{10}} - 1\right)\right] = \sigma_s^2 - \frac{1}{3c_z y_{10}}\left[2A_t(0,t) + A\left(2y_1\sigma_0^2, t\right)\right]. \quad (5.23)$$

With the use of the t^2-dependence of the thermal amplitudes, equations (5.20) and (5.21), and on substituting the expression (5.22) for y_1/y_{10}, we get the following result:

$$\begin{aligned}\frac{\sigma_0^2}{\sigma_s^2} &= \left\{1 - \frac{1}{3c_z y_{10}\sigma_s^2}\left[2A_t(0,t) + A\left(2y_1\sigma_0^2, t\right)\right]\right\}\left[1 - \frac{2}{3}\left(\frac{y}{y_{10}} - 1\right)\right]^{-1} \\ &= 1 - \frac{1}{3c_z y_{10}\sigma_s^2}\left\{2A_{\mathrm{sw}}(t) + \frac{t^2}{120y_{10}\sigma_s^2}\left[4 + 5(\pi\xi/4)^2 + (\pi\xi/4)^4\right] + \cdots\right\} \\ &= 1 - \frac{t^2}{360c_z y_{10}^2\sigma_s^4}\left[4 + 5(\pi\xi/4)^2 + (\pi\xi/4)^4\right] - \frac{2}{3c_z y_{10}\sigma_s^2}A_{\mathrm{sw}}(t) + \cdots \\ &= 1 - \frac{a_T}{\sigma_s^4}\left(\frac{T}{T_A}\right)^2 - \frac{2}{3c_z y_{10}\sigma_s^2}A_{\mathrm{sw}}(t) \end{aligned} \quad (5.24)$$

where

$$a_T = 10c_z[4 + 5(\pi\xi/4)^2 + (\pi\xi/4)^4].$$

It is also represented by

$$\frac{\sigma_0^2}{\sigma_s^2} = 1 - \frac{a_T}{400C_{4/3}^2 t_c^{2/3}}\left(\frac{T}{T_c}\right)^2 - \frac{2}{C_{4/3}t_c^{4/3}}A_{\rm sw}(t) + \cdots.$$

The spontaneous moment includes two kinds of contribution at low temperature, the spin-wave $t^{3/2}$-dependence and the t^2-dependence from the spin fluctuations. Their relative importance depends on the temperature range of interest. The $t^{3/2}$-dependence, for instance, becomes dominant when the following inequality:

$$\frac{a_T}{\sigma_s^2}\left(\frac{T}{T_A}\right)^2 < \frac{2}{3c_z y_{10}}A_{\rm sw}(t) \tag{5.25}$$

is satisfied. With the use of the expression for $A_{\rm sw}(t)$ in (5.11), equation (5.25) can be expressed as follows:

$$\frac{t}{t_c} < 5C_{4/3}\left\{\frac{10\sqrt{\pi}\zeta(3/2)}{a_T}\right\}^2 \sigma_s t_c^{1/3} \simeq 0.461\,\sigma_s t_c^{1/3} \tag{5.26}$$

where we have assumed $\xi = \xi_c$. Since both σ_s and t_c are very small for weak itinerant-electron ferromagnets, the spin-wave contribution becomes dominant only at very low temperature. This explains why the $t^{3/2}$-dependence of the spontaneous moment is not observed for most of the itinerant weak ferromagnets.

As long as the spin-wave contribution is neglected, the temperature dependence of σ_0^2 is simply given by

$$\frac{\sigma_0^2}{\sigma_s^2} = 1 - \frac{a_T}{\sigma_s^4}\left(\frac{T}{T_A}\right)^2 + \cdots = 1 - b_T T^2 + \cdots. \tag{5.27}$$

The T^2-coefficients have been estimated for several weak itinerant-electron ferromagnets experimentally. They are summarized in table 7. From the observed values of b_T and σ_s, we can estimate the parameter T_A by using

$$T_A = \frac{1}{\sigma_s^2}\sqrt{\frac{a_T}{b_T}}. \tag{5.28}$$

In the same table, table 7, values of T_A estimated from b_T are also shown in comparison with those reproduced from table 3 obtained from the slope of the Arrott plot of the magnetization measurements. The general agreements are satisfactory.

We have to be very careful in evaluating b_T from the observed slope of the σ^2 versus T^2 plot, for it depends on the temperature range used for the fit. As will be shown below, the t^2-dependence of σ_0^2 is sometimes confined within the very-low-temperature region. As we increase the temperature, σ_0^2 will soon deviate from the t^2-dependence and the critical $[t_c^{4/3} - t^{4/3}]$-behaviour follows. The crossover temperature is estimated as follows. The t^2-dependence originates from the asymptotic expansion of the digamma function $\psi(u)$, equation (5.10), permissible for $u \ll 1$. This is satisfied when $x_c^3/t \gg 1$ for the transverse amplitude. The same inequality also holds for the longitudinal amplitude. With the use of $x_c \sim \sqrt{y_{z0}}$, the condition can be represented by

$$t \ll \left(2y_{10}\sigma_s^2\right)^{3/2} = \left[\frac{2C_{4/3}}{3c_z}\right]^{3/2} t_c^2.$$

Table 7. The spin fluctuation parameter T_A estimated from the T^2-coefficient of the magnetization. The values of $T_A^\dagger$ are reproduced from table 3 for comparison.

Compound	σ_s	b_T (K^{-2})	T_A (K)	$T_A^\dagger$ (K)	References
$Ni_{74.7}Al_{25.3}$	0.0474	2.77×10^{-3}	1.05×10^5	3.85×10^4	Sasakura *et al* (1984)
$Ni_{75}Al_{25}$	0.0770	0.874×10^{-3}	7.06×10^4		Sasakura *et al* (1984)
$Ni_{75.2}Al_{24.8}$	0.0917	0.589×10^{-3}	6.06×10^4		Sasakura *et al* (1984)
$Ni_{75.5}Al_{24.5}$	0.110	0.386×10^{-3}	5.20×10^4		Sasakura *et al* (1984)
Ni_3Al	0.075	0.784×10^{-3}	7.9×10^4	3.09×10^4	de Boer *et al* (1969)
$Ni_{75.5}Al_{24.5}$	0.104	0.372×10^{-3}	5.9×10^4		de Boer *et al* (1969)
$Ni_{76}Al_{24}$	0.125	0.246×10^{-3}	5.0×10^4		de Boer *et al* (1969)
$ZrZn_2$	0.12	2.69×10^{-3}	1.66×10^4	7.40×10^3	Ogawa (1972)
$Fe_{0.67}Co_{0.33}Si$	0.22	0.400×10^{-3}	1.28×10^4	9.9×10^3	Shimizu *et al* (1990)
$Fe_{0.77}Co_{0.23}Si$	0.18	0.833×10^{-3}	1.32×10^4	1.2×10^4	Shimizu *et al* (1990)
$Fe_{0.83}Co_{0.17}Si$	0.13	1.49×10^{-3}	1.90×10^4	1.5×10^4	Shimizu *et al* (1990)
$Fe_{0.91}Co_{0.09}Si$	0.07	5.13×10^{-3}	3.52×10^4	2.2×10^4	Shimizu *et al* (1990)
$Pt_{0.53}Ni_{0.47}$	0.121	1.30×10^{-4}	7.4×10^4		Beille *et al* (1975)
Y_2Ni_{15}	0.15	8.54×10^{-5}	5.95×10^4	3.51×10^4	Gignoux *et al* (1980a)
YNi_3	0.04	1.20×10^{-3}	2.2×10^5	9.23×10^4	Gignoux *et al* (1980b)

Or it is simply given by $t/t_c \ll 1.5\,t_c$. To illustrate the situation, we show in figure 10 numerical results for σ_0^2/σ_s^2 as a function of $(t/t_c)^2 = T^2/T_c^2$. In the same figure, t^2-linear behaviour in (5.25) is also shown by dashed lines for reference. We can see that the slope of the real

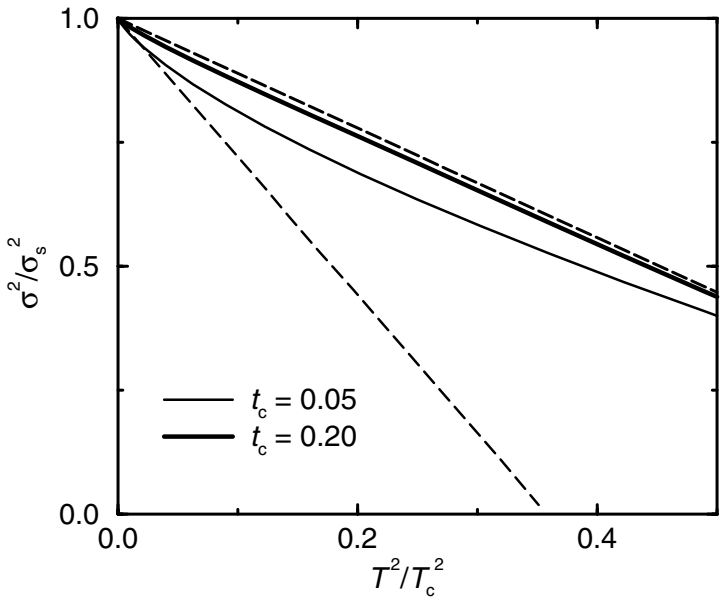

Figure 10. The t^2-dependence of σ^2 (solid thin and thick lines for $t_c = 0.05$ and 0.2). Dashed lines represent t^2-linear dependence at low temperature. The discrepancy between the solid and dashed lines at low temperature comes from the spin-wave contribution.

t^2-dependence becomes very steep and is limited to within the very-low-temperature region for cases with smaller t_c in accordance with the above criterion. Reliable estimation of b_T from experiments therefore becomes very difficult for magnets with small t_c-values. If the observed results are globally fitted with a t^2-relation, only a moderate slope is obtained and b_T is likely to be underestimated. The same argument is applicable to the t-dependence of y_1. This explains the discrepancy between the spin fluctuation parameters T_A estimated from the slope of the t^2-dependence of y_1 and those obtained from another measurements (see table 6) for small t_c.

5.3.2. Field dependence at low temperature. By extending our preceding arguments, the magnetic field dependence of the moment at low temperature can be also evaluated. If we substitute (5.4) into (5.3), our basic equation (5.3) can be expressed in the form

$$\left[1-\frac{2}{3}\left(\frac{y_1}{y_{10}}-1\right)\right]\sigma^2=\sigma_s^2-\frac{1}{3c_z y_{10}}[2A_t(y,t)+A(y_z,t)]+\frac{y}{y_{10}}.$$

The temperature and field dependence of σ^2 is then given by

$$\sigma^2=\left[1-\frac{2}{3}\left(\frac{y_1}{y_{10}}-1\right)\right]^{-1}\left\{\sigma_s^2+\frac{y}{y_{10}}-\frac{1}{3c_z y_{10}}[2A_t(y,t)+A(y_z,t)]\right\}. \tag{5.29}$$

Due to the presence of the external magnetic field, y becomes finite and the low-temperature expansion (5.9) of the transverse thermal amplitude is slightly modified as follows:

$$A_t(y,t)\simeq A_c(y,t)\simeq\int_{x_c}^1 \mathrm{d}x\,\frac{x^3}{12u^2}=\frac{t^2}{24}\frac{1-x_c^2}{1+y}\frac{1}{y+x_c^2}\simeq\frac{t^2}{24}\frac{1}{y+x_c^2}.$$

The longitudinal component $A(y_z,t)$ has already been given by (5.21). By using these expressions for the thermal amplitudes, the temperature and field dependence of the moment is finally given by

$$\begin{aligned}\sigma^2&=\left(\sigma_s^2+y/y_{10}\right)\left[1-\frac{t^2}{36c_z y_{z0}^2}\left\{\frac{1}{5}\left(2\frac{y_{z0}^2}{x_c^4}+3\right)\right.\right.\\&\qquad\left.\left.+\frac{1}{1+2y/y_{z0}}\left(\frac{2y_{z0}/x_c^2}{1+y/x_c^2}+\frac{1}{1+3y/y_{z0}}\right)\right\}+\cdots\right]\\&=\left(\sigma_s^2+y/y_{10}\right)\left[1-\frac{a_T(H)}{\sigma_s^4}\left(\frac{T}{T_A}\right)^2+\cdots\right]\end{aligned}$$

where

$$a_T(H)=5c_z\left[2\frac{y_{z0}^2}{x_c^4}+3+\frac{5}{1+2y/y_{z0}}\left(\frac{2y_{z0}/x_c^2}{1+y/x_c^2}+\frac{1}{1+3y/y_{z0}}\right)\right]. \tag{5.30}$$

The spin-wave contribution $A_{\mathrm{sw}}(t)$ is neglected here for simplicity. In the above derivation, we have used again the relation $c_z y_{10}=T_A/60T_0$ and $y_{z0}\simeq 2y_{10}\sigma_s^2$. In the weak-field limit, $a_T(H)$ shows the following H-linear dependence:

$$a_T(H)=a_T-25c_z\left[5+4\frac{y_{z0}}{x_c^2}+2\frac{y_{z0}^2}{x_c^4}\right]\frac{y}{y_{z0}}+\cdots\quad(y\simeq 2\mu_B H/\sigma_s T_A).$$

At low temperature the t-dependence of y on the right-hand side of (5.30) is neglected, as far as the t^2-dependence is concerned. The field dependence of y is therefore evaluated simply by solving the equation of state in the ground state. In the presence of the external field H, the induced moment $\bar{\sigma}=\sigma/\sigma_s$ in the ground state is determined as the solution of

$$\bar{\sigma}(\bar{\sigma}^2-1)=\frac{y\sigma}{y_{10}\sigma_s^3}=\frac{120c_z T_0}{\sigma_s^3 T_A^2}\mu_B H. \tag{5.31}$$

The field dependence of y is then given by

$$y/y_{z0} = (\bar{\sigma}^2 - 1)/2. \tag{5.32}$$

By solving $\bar{\sigma}$ as a function of H and putting the corresponding value of y/y_{z0} into (5.30), we can evaluate the field dependence of $a_T(H)$.

For $Pt_{0.53}Ni_{0.47}$ the field dependence of $a_T(H)$ was measured for H up to 100 kOe (Beille 1975). The observed relative suppression of $a_T(H)/a_T(0)$ is reproduced in figure 11. In the same figure our numerical result from (5.30) is shown for comparison. We assumed $T_A = 2 \times 10^4$ K, $T_0 = 3 \times 10^3$ K from table 3, and $\sigma_s = 0.12$. The agreement between the theory and experiments is very good.

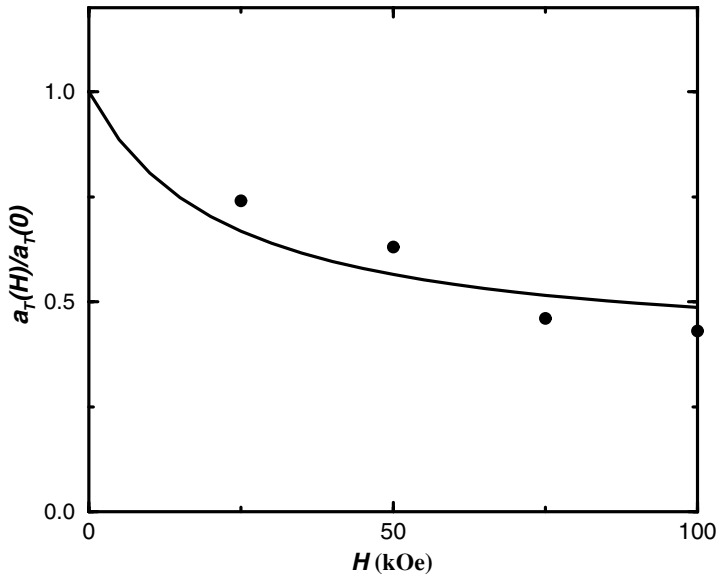

Figure 11. The magnetic field dependence of the T^2-coefficient of the magnetization. Solid circles represent the observed results for $Pt_{0.53}Ni_{0.47}$.

5.4. Magnetic isotherms at general temperature

Once the spontaneous moment σ_0^2 is determined by solving (5.13), we can obtain the magnetic isotherm by numerically integrating the first-order differential equation starting from the initial condition $y = 0$ at $\sigma = \sigma_0$ at any given temperature t. It is easy to see that the scaling property also holds for solutions thus obtained. In place of σ_0 and y, let us define reduced parameters, $\bar{\sigma}$ and $\bar{y}$, by

$$\sigma_0 = \sigma_s \bar{\sigma} \quad y = \left(y_{10}\sigma_s^2\right)\bar{y} = \frac{A(0,t_c)}{c_z}\bar{y} = \frac{C_{4/3}t_c^{4/3}}{3c_z}\bar{y}$$
$$y_z = \left(y_{10}\sigma_s^2\right)\bar{y}_z.$$

Then (5.3) can be represented by

$$2\bar{A}_t(y,t) + \bar{A}(y_z,t) - (2\bar{y} + \bar{y}_z) + 5\bar{\sigma}^2 = 3 \tag{5.33}$$

where $\bar{A}(y,t) = A(y,t)/A(0,t_c)$ and t is given by $t_c(T/T_c)$. The lower bound of x_c for $A_t(y,t)$ is also determined by the parameter $y_z = (y_{10}\sigma_s^2)\bar{y}_z$ obtained by solving (5.33) for $\bar{y} = 0$. The differential equation (5.33) for $\bar{y}$ as a function of $\bar{\sigma}$ at each reduced temperature

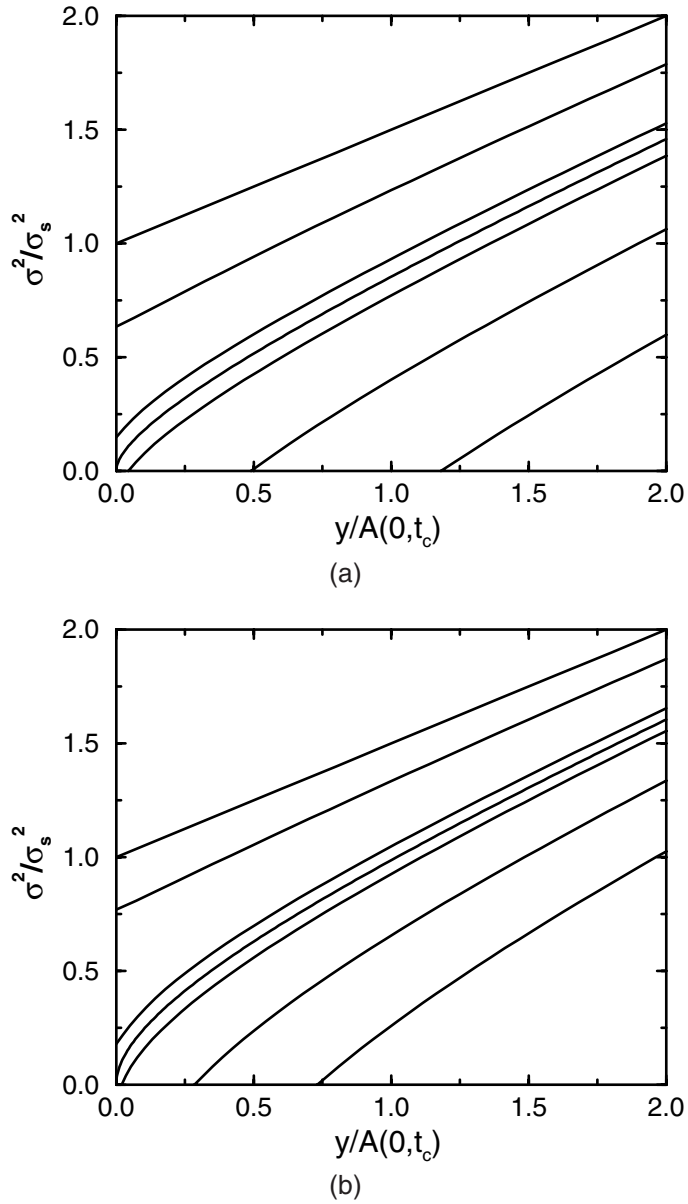

Figure 12. Arrott plots of calculated magnetic isotherms for $t_c = 0.05$ (a) and 0.2 (b). The curves, from the top, correspond to $T/T_c = 0$, 0.5, 0.9, 1, 1.1, 1.5, and 2, respectively.

T/T_c now depends only on a single parameter t_c. The behaviour of the solutions is therefore determined by the magnitude of t_c. This means that the temperature and the field dependences of σ for any weak ferromagnets look the same if they have the same t_c, and σ, T, and H are properly scaled. The result is consistent with our preceding results for the temperature and the field dependences of σ_0^2 and y_1, as well as our discussion on their ranges of validity. In the last figure, figure 12, the Arrott plots of numerically calculated magnetic isotherms are shown for $t_c = 0.05$ and 0.2 in reduced units.

6. Discussion

In this paper, we have discussed a lot of quantitative magnetic properties of weak itinerant ferromagnets on the basis of the spin fluctuation mechanism. We have studied both their temperature and magnetic field dependences throughout a wide temperature range by dealing with the magnetic equation of state. It seems to be widely accepted that the Arrott plot of magnetic isotherms shows good linearity with nearly the same slopes throughout the wide temperature and external field range for weak itinerant-electron magnets. They are sometimes globally fitted well with the simple Stoner–Wohlfarth formula (5.1). If we inspect the observed behaviours in more detail, we will however find slight deviations. For instance, the presence of the T^2-dependence on temperature of the fourth expansion coefficient F_1 was recognized almost 30 years ago (Wohlfarth and de Chatel 1970). Since then it has not been seen as very important either theoretically or experimentally. That is because the discrepancies are sometimes very small and not very clearly evident. Their ranges of deviations are also very limited. We have clarified in the present paper that the actual magnetic isotherms for weak ferromagnets are not so simple and do show a variety of behaviours. The slight deviations have their own meaning and we can draw valuable information from them on the spin fluctuation spectra.

The underlying motif of this study is the emphasis on the predominance of the roles of collective magnetic excitations over those of the single-particle ones in the magnetic properties of itinerant-electron magnets. This is true even in the low-temperature region including the ground state with no thermal spin fluctuation amplitude. Our approach is based on a very simple assumption on the conservation of the local spin amplitude and the requirements from the rotational symmetry of the system. In the Stoner–Wohlfarth theory most of the magnetic properties were explained as associated with single-particle excitations of the system, while the collective magnetic excitations are neglected. As an approach to extending the Stoner–Wohlfarth picture, the SCR theory tried to include the small-amplitude fluctuations around the mean-field free energy. It is therefore appropriate when the amplitude of the fluctuations is very small. Although the roles of thermal spin fluctuations are emphasized at finite temperature, this is still based on the single-particle picture in the ground state. The non-linear mode–mode coupling constants are therefore assumed to be given by the form of the density-of-states curve around the Fermi energy.

We start from the opposite limit. From our point of view, weak itinerant magnets are rather well characterized by the dominant spin fluctuation amplitudes. This is particularly due to the presence of the quantum component of fluctuations. The appearance of small uniform static moments does not indicate small fluctuation amplitudes. It results from the presence of dominant fluctuation amplitudes. Because of the total-amplitude conservation, the increase of spin fluctuation amplitudes leads to a reduction of the magnitude of the ordered moments. Collective Bose-like excitations therefore always play major roles in almost all aspects. In this respect the situation looks quite similar to the case of Heisenberg magnets. Then, what kinds of property will distinguish itinerant-electron systems from Heisenberg magnets? According to the SCR theory, for instance, the amplitude of the spin fluctuations increases with temperature in strong contrast with the case for Heisenberg magnets. A lot of magnetic properties of itinerant weak ferromagnets have been explained in association with this variation of the amplitude.

Our answer to the above question is stated as follows. One is the difference in damping mechanism of the magnetic fluctuations. Especially in the case of the itinerant ferromagnets, the Landau damping mechanism prevails in most of the wave-vector region. Its magnitude is in general larger for itinerant-electron systems. The spectral shape of the magnetic excitations

is also different. We have to introduce two independent parameters T_0 and T_A for the spectral widths in the energy and wave-vector spaces. They correspond to the single parameter J of the exchange coupling of the Heisenberg magnets. Another one is associated with the presence of large-amplitude quantum fluctuations. We have argued that the qualitative differences in properties between these two classes of magnets originate from the presence of quantum spin fluctuations. This means that taking explicit account of the quantum fluctuations is necessary for the proper theoretical description of weak itinerant-electron magnets. On the other hand, the amplitude conservation and the predominance of collective magnetic degrees of freedom are common to Heisenberg magnet and itinerant-electron systems.

In summary, the following consequences are all derived from our theoretical studies by taking explicit account of the effects as stated above:

- The magnetic equation of state is mainly determined by the response of the spin fluctuations to the external magnetic field. This is true even in the ground state. Therefore the non-linear expansion coefficients F_1 of the magnetic free energy are determined by the spectral properties of spin fluctuations.
- Scaling behaviour is predicted for the temperature dependence of the magnetic susceptibility as well as various properties in the ordered state. We showed for instance that the ratio σ_e/σ_s is almost uniquely determined by the single parameter T_c/T_0.
- At the critical temperature, we predicted the critical magnetization process $H \propto M^5$.
- We can establish a prescription for calculating the temperature dependence of the spontaneous magnetic moments below T_c. The magnetic equation of state is also calculated rather easily throughout a wide temperature range without dealing with complicated integro-differential equations.
- We derived an explicit t^2-dependence on temperature of the squared spontaneous magnetization σ^2 at low temperature and a $[1-(t/t_c)^{4/3}]$-dependence around the critical temperature, including their precise numerical coefficients, that agree well with experiments.
- We must be careful to note that the real critical $[1-(t/t_c)^{4/3}]$-behaviour occurs in a very restricted region around t_c for compounds with smaller t_c.
- We showed that the T^2-dependence of σ^2 at low temperature is observed over a wider temperature range for compounds with larger t_c-values.
- The $T^{3/2}$-dependence of σ^2 due to the spin wave is generally confined to the region of extremely low temperature for weak itinerant ferromagnets.
- The fourth-order expansion coefficient F_1 of the free energy shows a temperature dependence (t^2-dependence at low temperature and $[1-(t/t_c)^{4/3}]$-dependence around the critical temperature) that is of the same form as that on the magnetization. In our theoretical framework this parameter is therefore no longer a constant. On the other hand, it has the meaning of an important theoretical input parameter in the SCR theory that gives the non-linear mode–mode coupling among various thermal spin fluctuation modes.

In the present paper we are only concerned with the magnetic properties of the system. The results of the present paper will have some significant consequences for the various related properties. For instance, the metamagnetic transitions observed in itinerant-electron magnets are usually treated in terms of the magnetic equation of state obtained by assuming a single-particle density-of-states curve $\rho(\varepsilon)$. If our arguments are valid, we have instead to take into account the effects of magnetic excitations on the equation of states. Especially in describing the states with small moments before the metamagnetic transition, the M–H curve is instead governed by the spin fluctuation mechanism. The validity of the present picture on the magnetic equation of states is clearly exhibited by the recent non-linear magnetization

measurements on the semiconducting compound FeSi (Koyama *et al* 2000). The $M-H$ curve derived by using the density-of-states curve $\rho(\varepsilon)$ gives the wrong sign for the fourth expansion coefficient F_1 The observed qualitative behaviours are in accord with our present mechanism (Takahashi 1998, Takahashi *et al* 2000). Because of the Maxwell relation, our temperature and field dependences of the magnetic moments are also closely related to the field dependence of the magnetic specific heat.

Acknowledgments

The author thanks Professors T Kanomata, K Yoshimura, Y Tazuke, and Dr K Shimizu, for valuable discussions on experiments.

References

Arrott A 1957 *Phys. Rev.* **108** 1394
Beille J 1975 Thèse L'Universite Scientifique et Medicale et l'Institut National Polytechnique de Grenoble
Beille J, Block D and Besnus M J 1974 *J. Phys. F: Met. Phys.* **4** 1275
Bernhoeft N R, Lonzarich G G, Mitchell P W and Paul D M^{c}K 1983 *Phys. Rev.* B **18** 422
Bernhoeft N R, Lonzarich G G, Paul D M^{c}K and Mitchell P W 1986 *Physica* B **136** 443
Bloch D, Voiron J, Jaccarino V and Wernick J H 1975 *Phys. Lett.* A **51** 259
de Boer F R, Schinkel C J, Biesterbos J and Proost S 1969 *J. Appl. Phys.* **40** 1049
Edwards D M and Wohlfarth E P 1968 *Proc. R. Soc.* A **303** 127
Gignoux D, Lemaire R and Molho P 1980a *J. Magn. Magn. Mater.* **21** 119
Gignoux D, Lemaire R, Molho P and Tasset F 1980b *J. Magn. Magn. Mater.* **21** 307
Hioki T and Masuda Y 1977 *J. Phys. Soc. Japan* **43** 1200
Ishikawa Y, Noda Y, Uemura Y J, Majkrzak C F and Shirane G 1985 *Phys. Rev.* B **31** 5884
Knapp G S, Fradin F Y and Culbert H 1971 *J. Appl. Phys.* **42** 1341
Kontani M 1977 *J. Phys. Soc. Japan* **42** 83
Kontani M, Hioki T and Masuda Y 1975 *J. Phys. Soc. Japan* **39** 665
Koyama K, Goto T, Kanomata T, Note R and Takahashi Y 2000 *J. Phys. Soc. Japan* **69** 219
Lonzarich G and Taillefer G 1985 *J. Phys.* **18** 4339
Moriya T 1985 *Spin Fluctuations in Itinerant Electron Magnetism* (Berlin: Springer)
Moriya T and Kawabata A 1973a *J. Phys. Soc. Japan* **34** 639
Moriya T and Kawabata A 1973b *J. Phys. Soc. Japan* **35** 669
Murata K K and Doniach S 1972 *Phys. Rev. Lett.* **29** 285
Nakabayashi R, Tazuke Y and Maruyama S 1992 *J. Phys. Soc. Japan* **61** 774
Ogawa S 1968 *J. Phys. Soc. Japan* **25** 109
Ogawa S 1972 *Research of the Electrochemical Laboratory Report* 735
Ogawa S 1976 *J. Phys. Soc. Japan* **40** 1007
Rhodes P R and Wohlfarth E P 1963 *Proc. R. Soc.* **273** 247
Sasakura H, Suzuki K and Masuda Y 1984 *J. Phys. Soc. Japan* **53** 754
Shimizu K, Maruyama H, Yamazaki H and Watanabe H 1990 *J. Phys. Soc. Japan* **59** 305
Solontsov A Z and Wagner D 1994 *J. Phys.* M **6** 7395
Solontsov A Z and Wagner D 1995 *Phys. Rev.* B **51** 410
Stoner E C 1936 *Proc. R. Soc.* A **154** 656
Takahashi Y 1986 *J. Phys. Soc. Japan* **55** 3553
Takahashi Y 1990 *J. Phys.: Condens. Matter* **2** 8405
Takahashi Y 1992 *J. Phys.: Condens. Matter* **4** 3611
Takahashi Y 1994 *J. Phys.: Condens. Matter* **6** 7063
Takahashi Y 1997a *J. Phys.: Condens. Matter* **9** 2593
Takahashi Y 1997b *J. Phys.: Condens. Matter* **9** 359
Takahashi Y 1998 *J. Phys.: Condens. Matter* **10** L671
Takahashi Y, Kanomata T, Note R and Nakagawa T 2000 *J. Phys. Soc. Japan* **69** 4018
Takeuchi J and Masuda Y 1979 *J. Phys. Soc. Japan* **46** 468
Takahashi Y and Sakai T 1995 *J. Phys.: Condens. Matter* **7** 6279

Takahashi Y and Sakai T 1998 *J. Phys.: Condens. Matter* **10** 5373
Umemura T and Masuda Y 1995 *J. Phys. Soc. Japan* **52** 1439
Wohlfarth E P 1968 *J. Appl. Phys.* **39** 1061
Wohlfarth E P and de Chatel P F 1970 *Physica* **48** 477
Yamada H 1975 *Phys. Lett.* A **55** 233
Yasuoka K, Jaccarino V, Sherwood R C and Wernick J H 1978 *J. Phys. Soc. Japan* **44** 842
Yoshimura K, Mekata M, Takigawa M, Takahashi Y, Yasuoka H and Nakamura Y 1987 *J. Phys. Soc. Japan* **56** 1138
Yoshimura K, Takigawa M, Takahashi Y and Yasuoka H 1988 *Phys. Rev.* B **37** 3593
Ziebeck K R A, Capellmann H, Brown P J and Booth J G 1982 *Z. Phys.* **48** 241