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Temperature and magnetic field dependence of the heat capacity in itinerant electron ferromagnets

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Abstract
Based on the spin fluctuation mechanism we have succeeded in deriving formulae for the magnetic entropy and the specific heat of itinerant electron ferromagnets that cover the wide range of the temperature and the external field strength. We show that it is necessary to include an extra term into the free energy for a thermodynamically consistent treatment. We are able to predict several new features on the temperature and the external field dependence, an extra enhancement of the T-linear coefficient at low temperature and the presence of a critical peak anomaly of the specific heat, for instance. They result from terms proportional to the second-order temperature derivative of the spontaneous magnetization. The presence of the terms is connected with the Maxwell relation of thermodynamics.

1. Introduction

The enhancement of the heat capacity at low temperature and its suppression by the external magnetic field of exchange-enhanced paramagnets as well as itinerant electron weak ferromagnets already have a long history of intensive theoretical and experimental studies. There has been a revival of interest recently in relation to low-dimensional itinerant electron systems (Hatatani and Moriya 1995) and quantum critical phenomena (Hertz 1976, Millis 1993, Zülicke and Millis 1995, Ishigaki and Moriya 1996, Pfleiderer *et al* 1997).

In the 1960s most of the interests were focused on the temperature and the external field dependence of the highly exchange-enhanced Landau Fermi liquids. Based on the random phase approximation, low temperature behaviours were discussed by Doniach and Engelsberg (1966), Brinkmann and Engelsberg (1968), and Berk and Schrieffer (1966). These studies are now known as paramagnon theories. Within the same framework the external field suppression of the specific heat was also discussed by Béal-Monod *et al* (1968), Béal-Monod (1981), Hertel *et al* (1980), and Béal-Monod and Daniel (1983).

Around the beginning of the 1970s, a novel approach, now called the self-consistent renormalization (SCR) spin fluctuation theory, was proposed by Moriya and Kawabata (1973a, 1973b) for the explanation of the observed Curie–Weiss temperature dependence of the magnetic susceptibility of weak itinerant electron ferromagnets. It is based on the decoupling of nonlinear terms among spin fluctuation modes with various wavevectors. By the proper self-consistent account of the feedback effect of the mode–mode coupling, these authors succeeded in explaining many magnetic properties in the wide temperature range from the low temperature limit to the paramagnetic phase through the transition temperature (Moriya 1985). The temperature dependence of the specific heat was treated by Murata and Doniach (1972) for the first time from the simplified view of the mode–mode coupling idea. More elaborate treatment was given by Makoshi and Moriya (1975), and Hasegawa (1975) based on the SCR spin fluctuation theory. It was soon extended by Takeuchi and Masuda (1979) to include the external field dependence in order to compare their experiments on Sc_3In. For nearly ferromagnetic metals, the quantitative analysis of the temperature dependence was given by Konno and Moriya (1987).

The studies on the specific heat along this approach, however, had some shortcomings. The SCR theory predicts a spurious discontinuous dip just above the critical point in the temperature dependence (Makoshi and Moriya 1975). As for the external magnetic field dependence, no attention has been paid to the Maxwell relation of the thermodynamics, though its importance was recognized by Béal-Monod (1981) and Shioda *et al* (1988), for instance, in relation to the field dependence of the specific heat of exchange-enhanced paramagnets.

The original SCR spin fluctuation theory also assumes that the zero-point quantum spin fluctuation amplitude is rigid and will show neither temperature nor external magnetic field dependence. The effect is therefore discarded from the beginning. The self-consistent spectral change of spin fluctuation modes plays predominant roles in the Curie–Weiss like temperature dependence of the magnetic susceptibility. It is not so clear why its effect on the quantum amplitudes is neglected. It was not so easy to treat the magnetic field dependence of various properties, the magnetic isotherm for instance. In the presence of the induced magnetic moment, we have to deal with anisotropic spin fluctuation amplitudes. We are then forced to study coupled integro-differential equations simultaneously (Lonzarich and Taillefer 1985). Because of this difficulty, the SCR theory is mainly concerned with properties on the temperature axis of the phase diagram.

One of the present authors has shown that the spectral change also has a profound effect on the quantum spin fluctuations. The quantum fluctuations cannot be neglected. We then have to deal with the large amplitude fluctuations as the sum of both the thermal and quantum components. Based on the idea of the total spin amplitude conservation, he has succeeded in deriving various interesting theoretical consequences (Takahashi 1986, 1990, 1992, 1994, 1997a, 1997b) and Takahashi and Sakai (1995, 1998). They were supported by various later experimental studies (Yoshimura *et al* 1988a, 1988b, Shimizu *et al* 1990, Nakabayashi *et al* 1992, Fujita *et al* 1995). As an example, the M–H curve is generally determined by the effect of spin fluctuations. Even in the ground state with no thermal amplitude, it is determined by the response of zero-point fluctuations to the applied magnetic field, in distinct contrast with the Stoner–Wohlfarth single particle picture and the SCR theory. The positive mode–mode coupling constant of FeSi observed by nonlinear magnetization measurements (Koyama *et al* 2000) is consistent with this mechanism (Takahashi 1998), while the single particle picture gives an inappropriate negative constant. Weak temperature dependence of the total spin amplitude in partial ferromagnets has recently been confirmed in the one-dimensional Hubbard chain by the exact numerical diagonalization method (Nakano and Takahashi 2004). The effects of the quantum spin fluctuation amplitude have also been studied based on the

Landau–Ginzburg model of the second-order phase transition (Solontsov and Wagner 1994, 1995, Kaul 1999, Semwal and Kaul 1999). It is not so obvious how the free energy expansion is justified in terms of such large amplitude quantum fluctuations as were clearly observed by polarized neutron scattering experiments on MnSi (Ziebeck *et al* 1982) throughout the wide temperature range.

According to the quantum spin fluctuation theory mentioned above, the temperature dependence of the specific heat was treated by Takahashi (1999) based on a free energy, consistent with the conserved local spin amplitude. Although it is confined within the paramagnetic phase with no external magnetic field, he can give an answer that resolves the unfavourable temperature dependence of the specific heat just above T_c. As far as the paramagnetic phase is concerned, it is based on the entropy that satisfies the Maxwell relation of thermodynamics. It is extended to the case of FeSi, and compared well with experiments (Takahashi *et al* 2000).

The purpose of the present paper is therefore to extend our previous study to the ordered phase and to give the unified theoretical framework that enables us to treat the specific heat in the global H–T or M–T phase diagram. Almost no efforts have appeared since the work by Takeuchi and Masuda (1979) to overcome the above difficulties on the field dependence of the specific heat of itinerant electron ferromagnets. We propose a way to extend our previous free energy expression to the more general case in the presence of anisotropic spin fluctuation amplitudes below T_c. Based on the free energy, we will give a consistency check of our formalism by the Maxwell relation of thermodynamics.

In the next section, we propose a free energy due to the magnetic excitations. A brief review of our spin fluctuation theory is also given. The general expressions of the magnetic entropy and the specific heat are derived in section 3. Based on the formula, the temperature and the external magnetic field dependence are discussed in sections 4 and 5, respectively. The summary and some discussions are presented in the final section.

In what follows the uniform magnetization M is expressed in terms of the dimensionless parameter σ in units of Bohr magneton μ_B per magnetic atom, and the wavevector dependent external field H by h in energy units:

$$M = N_0\mu_B\sigma, \qquad h = 2\mu_B H$$

where N_0 is the number of magnetic atoms in the system. The static and uniform magnetic susceptibility $\chi(0,0)$ measured in units of $4\mu_B^2$ is in the present units given by

$$\chi(0,0)/N_0 = \sigma/2h.$$

2. Magnetic free energy in the ordered phase

Let us first assume that the free energy consists of the sum of the following two contributions.

$$F(y, y_z, \sigma, t) = F_{sf}(y, y_z, \sigma, t) + F_{sw}(y, \sigma, t). \tag{2.1}$$

The first term F_{sf} comes from the effect of spin fluctuations with finite damping given by

$$\begin{aligned} F_{sf}(y, y_z, t) = {} & \frac{1}{\pi}\sum_{\mathbf{q}}\int_0^\infty d\omega \left[\frac{\omega}{2} + T\ln(1 - e^{-\omega/T})\right]\frac{\Gamma_q^z}{(\Gamma_q^z)^2 + \omega^2} \\ & + \frac{2}{\pi}T\sum_{q_{sw}<q}\int_0^\infty d\omega\,\ln(1 - e^{-\omega/T})\frac{\Gamma_q}{\Gamma_q^2 + \omega^2} \\ & + \frac{1}{\pi}\sum_{\mathbf{q}}\int_0^\infty d\omega\,\frac{\omega\Gamma_q}{\Gamma_q^2 + \omega^2} + N_0T_A y\sigma^2/4 + \Delta F \end{aligned} \tag{2.2}$$

where Γ_q and Γ_q^z represent the damping constants. It consists of the sum of contributions from both the longitudinal and the transverse fluctuations, and the Zeeman energy due to the externally applied magnetic field. We assume that $F_{\rm sf}$ is a function of the inverse of the magnetic susceptibilities, y and y_z defined below, for transverse and longitudinal degrees freedom with respect to the external field direction. The wavevector summation of the transverse component starts from the lower bound $q_{\rm sw}$ because of the presence of spin-wave modes. In our previous study on the magnetic isotherm of the itinerant electron weak ferromagnets (Takahashi 2001), we pointed out the significance of the cut-off effect in dealing with the transverse thermal spin fluctuation amplitude below the critical temperature $T_{\rm c}$. Since the quantum spin fluctuations have widespread spectral widths in frequency space, spin-wave effects on them are not so serious. Therefore only the explicit account of the effect on the thermal modes is taken in this treatment. The third term of the transverse quantum component is thus given as the sum throughout the whole of the Brillouin zone. The fourth term represents the Zeeman energy $-MH$ expressed in terms of y and σ. The last correction term ΔF is discussed later in this section.

The above form of the free energy assumes the presence of highly exchange-enhanced spin fluctuation modes. Their energy spectrum, given in terms of the imaginary part of the dynamical magnetic susceptibility, is well approximated by the double Lorentzian form in the frequency and the wavevector space,

$$\mathrm{Im}\,\chi_\alpha(q,\omega) = \chi_\alpha(q,0)\frac{\omega\Gamma_q^\alpha}{\omega^2+\Gamma_q^{\alpha 2}} \tag{2.3}$$

where α represents the parallel and perpendicular polarization to the induced moment. The wavevector dependent static susceptibility $\chi_\alpha(q,0)$ and the damping constant Γ_q^α are given by

$$\begin{aligned} \chi_\alpha(q,0) &= \frac{\chi_\alpha(0,0)}{1+q^2/\kappa_\alpha^2} = \frac{N_0}{2T_A}\frac{1}{y_\alpha+x^2}, \qquad (x=q/q_B) \\ \Gamma_q^\alpha &= \Gamma_0 q(q^2+\kappa_\alpha^2) = 2\pi T_0 x(y_\alpha+x^2) \\ T_A &= N_0 q_B^2/2\chi_\alpha(0,0)\kappa_\alpha^2, \qquad T_0 = \Gamma_0 q_B^3/2\pi, \qquad y_\alpha = \kappa_\alpha^2/q_B^2. \end{aligned} \tag{2.4}$$

The inverse squared correlation length κ_α^2 is proportional to $\chi_\alpha^{-1}(0,0)$ defined by the second-order derivatives of the free energy in the parallel and perpendicular moments, $\delta M_\parallel$ and $M_\perp$, i.e.

$$\begin{aligned} \frac{N_0}{2\chi_\parallel(0,0)} &= \frac{\partial h}{\partial\sigma} = T_A y_\parallel \propto \frac{\partial^2 F}{\partial \delta M_\parallel^2}, \\ \frac{N_0}{2\chi_\perp(0,0)} &= \frac{h}{\sigma} = T_A y_\perp \propto \frac{\partial^2 F}{\partial M_\perp^2}. \end{aligned} \tag{2.5}$$

The constant parameter T_A introduced above stands for the measure of the distribution of $N_0/\chi_\alpha(q,0)$ in wavevector space, since the lower and the upper bounds of $N_0/\chi_\alpha(q,0)$ are given by $y_\alpha \ll 1$ for $q = 0$ and $2T_A(1+y_\alpha) \simeq 2T_A$ for $q = q_B$, respectively, while T_0 characterizes the spectral width in frequency space. The zone-boundary wavevector is denoted by q_B. The spin fluctuation amplitudes become anisotropic when the static magnetic moment is present. The effect is taken into account in terms of reduced anisotropic inverse susceptibilities y_α. Throughout the paper, the magnetic moment and the field are assumed to be along the z-axis. The subscript α (superscript for Γ) for the parallel modes are, therefore, denoted by y_z for instance, while those for transverse modes are suppressed.

In terms of the imaginary part of the dynamical susceptibility, the local spin amplitude on the ith lattice site can be given as the sum of thermal and quantum (zero-point) amplitudes as follows (in the paramagnetic phase).

$$\langle \mathbf{S}_i^2 \rangle = \langle \mathbf{S}_i^2 \rangle_T(y, y_z, t) + \langle \mathbf{S}_i^2 \rangle_Z(y, y_z)$$

$$\begin{aligned} \langle \mathbf{S}_i^2 \rangle_T(y, y_z, t) &= \frac{2}{N_0^2} \sum_{q\alpha} \int_0^\infty \frac{d\omega}{\pi} n(\omega)\, \mathrm{Im}\, \chi_\alpha(q, \omega) \\ &= \frac{3T_0}{T_A}[2A(y, t) + A(y_z, t)] \\ \langle \mathbf{S}_i^2 \rangle_Z(y, y_z) &= \frac{1}{N_0^2} \sum_{q\alpha} \int_0^\infty \frac{d\omega}{\pi} \mathrm{Im}\, \chi_\alpha(q, \omega) \\ &= \langle \mathbf{S}_i^2 \rangle_Z(0, 0) - \frac{3T_0}{T_A} c_z(2y + y_z) \end{aligned}$$

where $t = T/T_0$ is the reduced temperature and c_z is a positive numerical constant of the order of unity. The thermal amplitude $A(y, t)$ is defined by

$$A(y, t) = \int_0^1 dx\, x^3[\ln u - 1/2u - \psi(u)], \qquad u = x(y + x^2)/t. \tag{2.6}$$

The Bose factor and the digamma function are, respectively, denoted by $n(\omega)$ and $\psi(u)$.

The second term in (2.1) represents the contribution from the spin-wave excitations. It is given by

$$F_{\mathrm{sw}}(y, \sigma, t) = T \sum_{q<q_{\mathrm{sw}}} \ln(1 - e^{-\beta\omega_q}) + N_0 T_A[y\eta_1(\sigma, t) + \eta_0(\sigma, t)]$$

$$\omega_q = T_A \sigma y + D(\sigma) q^2.$$

The first term of the spin-wave frequency ω_q represents the external magnetic field h. Since our free energy is a function of y and σ, we use this notation. The functions $\eta_1(\sigma, t)$ and $\eta_0(\sigma, t)$ are variables introduced for the Legendre transformation to define the free energy as a function of σ instead of h. The spin-wave modes are confined within the small region around the origin of the wavevector space. The upper cut-off vector is denoted by q_{sw}.

To find the minimum of the free energy, we need its variation against the parameters σ and y. For the spin-wave modes, it is given by

$$\begin{aligned} \delta F_{\mathrm{sw}} = T_A \sum_{q<q_{\mathrm{sw}}} n(\omega_q) \left[\sigma \delta y + \left(y + \frac{1}{T_A} \frac{\partial D(\sigma)}{\partial \sigma} q^2 \right) \delta\sigma \right] \\ + N_0 T_A[\eta_1(\sigma)\delta y + y\eta_1'(\sigma)\delta\sigma] + N_0 T_A \eta_0'(\sigma)\delta\sigma \\ = N_0[6T_0 A_{\mathrm{sw}}(y, \sigma, t) + T_A \eta_1(\sigma)]\delta y \\ + N_0 T_A \left\{ \frac{1}{N_0} \sum_{q<q_{\mathrm{sw}}} \left(y + \frac{1}{T_A} \frac{\partial D}{\partial \sigma} q^2 \right) n(\omega_q) + y\eta_1'(\sigma) + \eta_0'(\sigma) \right\} \delta\sigma. \end{aligned}$$

The first term represents the thermal spin-wave amplitude for the transverse modes. The function A_{sw} is defined by

$$A_{\mathrm{sw}}(y, \sigma, t) = \frac{T_A \sigma}{6N_0 T_0} \sum_{q<q_{\mathrm{sw}}} n(\omega_q) = \frac{T_A \sigma}{2T_0} \int_0^{x_c} dx\, \frac{x^2}{e^{\beta\omega_q} - 1}, \qquad (x_c = q_{\mathrm{sw}}/q_B).$$

In the long-wavelength limit where $\beta\omega_q \ll 1$ is satisfied, the integrand of the above wavevector integration is given by

$$\frac{T_A \sigma}{2T_0} \frac{x^2}{e^{\beta\omega_q} - 1} \simeq \frac{T_A T \sigma}{2T_0 \omega_q} = \frac{t T_A \sigma x^2}{2[T_A \sigma y + D(\sigma) q^2]} = \frac{t}{2} \frac{x^2}{y + [D(\sigma) q_B^2 / T_A \sigma] x^2}.$$

If we assume the relation $D(\sigma)q_B^2/T_A\sigma \simeq 1$, it is easy to see that it smoothly tends to the long-wavelength limit, $x^3/2u$, of the integrand of the transverse thermal spin fluctuation amplitude $A(y,t)$ in (2.6). It is reasonable for us to assign this as the transverse spin amplitude due to spin-waves. We assume the above form of the spin-wave stiffness constant throughout this paper.

Now against the variations of σ and y, the change of the free energy δF is given by

$$\begin{aligned}\delta(F/N_0) = T_A\left\{3\frac{T_0}{T_A}[2A_t(y,t) + A(y_z,t) - c_z(2y+y_z)] + \frac{\sigma^2}{4} + \eta_1(\sigma) - \Delta\langle \mathbf{S}_i^2\rangle_{\text{tot}}\right\}\delta y \\ + 3T_0\left[A(y_z,t) - c_z y_z - \frac{T_A}{9T_0}\Delta\langle \mathbf{S}_i^2\rangle_{\text{tot}}\right]\delta\Delta y_z \\ + T_A\left[\frac{1}{N_0}\sum_{q<q_{\text{sw}}}(y+x^2)n(\omega_q) + y\eta_1'(\sigma) + \eta_0'(\sigma)\right]\delta\sigma \\ + T_A y\sigma\delta\sigma/2 + \delta(\Delta F_1/N_0)\end{aligned} \tag{2.7}$$

where $\Delta y_z = y_z - y$. We have introduced $\Delta\langle \mathbf{S}_i^2\rangle_{\text{tot}}$ and the transverse thermal amplitude $A_t(y,t)$ by

$$\begin{aligned}\Delta\langle \mathbf{S}_i^2\rangle_{\text{tot}} &= \langle \mathbf{S}_i^2\rangle_{\text{tot}} - \langle \mathbf{S}_i^2\rangle_Z(0) \\ A_t(y,t) &= A_{\text{sw}}(y,t) + A_{\text{c}}(y,t) \\ A_{\text{c}}(y,t) &= \int_{x_{\text{c}}}^1 \mathrm{d}x\, x^3[\ln u - 1/2u - \psi(u)].\end{aligned}$$

Due to the presence of the spin-wave modes, the transverse amplitude has to be modified in the long-wavelength limit. The above free energy expression is the straightforward generalization of our previous proposal for the paramagnetic case to the ordered phase that can afford the presence of the external magnetic field. The correction term is also extended as follows.

$$\Delta F(\sigma,t) = -\tfrac{1}{3}N_0 T_A\langle \mathbf{S}_i^2\rangle_{\text{tot}}(2y+y_z) + \Delta F_1(\sigma,t).$$

Its variation is given by

$$\delta\Delta F(\sigma,t) = -N_0 T_A\langle \mathbf{S}_i^2\rangle_{\text{tot}}\tfrac{1}{3}(3\delta y + \delta\Delta y_z) + \delta\Delta F_1(\sigma,t).$$

The first term of ΔF corresponds to the correction introduced by our previous study. The last new term ΔF_1 results from the appearance of the spontaneous moment. This term is necessary when we make sure that the Maxwell relation is justified in general cases under the presence of the finite induced magnetic moment. The parameter $\langle \mathbf{S}_i^2\rangle_{\text{tot}}$ has the same meaning as in the paramagnetic phase, i.e. the squared total spin amplitude.

It is reasonable to assume that the free energy is an extremum against the variation of y just like in the paramagnetic phase. We also impose the condition that the σ-derivative of the free energy agrees with the external magnetic field in consistence with the thermodynamic equation of state between H and M. From the σ-dependence of Δy_z, the above two conditions lead to the following two equations:

$$\Delta\langle \mathbf{S}_i^2\rangle_{\text{tot}} = \frac{\sigma^2}{4} + \frac{3T_0}{T_A}\big[2A_t(y,t) + A(y_z,t) - c_z(2y+y_z)\big] + \eta_1(\sigma) \tag{2.8}$$

$$\left\{\frac{2T_0}{T_A}\big[A(y_z,t) - A_t(y,t) - c_z\Delta y_z\big] - \frac{1}{3}\left[\frac{\sigma^2}{4} + \eta_1(\sigma)\right]\right\}\frac{\partial\Delta y_z}{\partial\sigma} + \frac{1}{N_0T_A}\frac{\partial\Delta F_1}{\partial\sigma} = 0. \tag{2.9}$$

Equation (2.9) can also be rewritten in the following form:

$$\begin{aligned}\lambda(\sigma,t) &= -\frac{1}{N_0T_A\,\partial\Delta y_z/\partial\sigma}\frac{\partial\Delta F_1}{\partial\sigma} \\ &= \frac{2T_0}{T_A}[A(y_z,t) - A_t(y,t) - c_z\Delta y_z] - \frac{1}{3}\left[\frac{\sigma^2}{4} + \eta_1(\sigma)\right].\end{aligned} \tag{2.10}$$

For the spin-wave modes, let us assume that the following condition is satisfied with respect to the σ variation:

$$\frac{1}{N_0}\sum_{q<q_{\rm sw}} n(\omega_q)(y+x^2) + y\eta_1'(\sigma) + \eta_0'(\sigma) = 0. \tag{2.11}$$

It will be replaced by the following two conditions:

$$\frac{1}{N_0}\sum_{q<q_{\rm sw}} n(\omega_q) = \frac{6T_0}{T_A\sigma}A_{\rm sw} = -\frac{\partial\eta_1(\sigma,t)}{\partial\sigma} \tag{2.12}$$

$$\frac{1}{N_0}\sum_{q<q_{\rm sw}} x^2 n(\omega_q) = 3\int_0^{x_c} {\rm d}x\, \frac{x^4}{{\rm e}^{\sigma T_A(y+x^2)/tT_0}-1} = -\frac{\partial\eta_0(\sigma,t)}{\partial\sigma}. \tag{2.13}$$

If the above conditions are all satisfied, the following well-known thermodynamic relation is derived as the minimum condition of the free energy:

$$\frac{1}{N_0}\frac{\partial F}{\partial\sigma} = T_A y\sigma = \frac{h}{2}.$$

Equation (2.8) represents the conservation of the total spin amplitude. It poses the implicit relation between the two functions, y and y_z, and the variable σ. It is a first-order differential equation, since y and y_z are related to each other by

$$y_z(\sigma,t) = y(\sigma,t) + \sigma\,\frac{\partial y(\sigma,t)}{\partial\sigma} \tag{2.14}$$

from their definitions (2.5). As the solution we can obtain the inverse magnetic susceptibility y as a function of σ. On the other hand, the free energy correction ΔF_1 is given as a function of σ with the use of (2.9) or (2.10). Two spin-wave related functions η_0 and η_1 are also obtained by solving (2.12) and (2.13). As their initial conditions, we can choose them to vanish at $\sigma = \sigma_0(t)$. Then we obtain $(\sigma-\sigma_0)$-linear solutions around $\sigma=\sigma_0$. In our previous study on the magnetic isotherm, we showed that the spin-wave effect is generally very small. Therefore they are neglected in this study. The following discussions are based on the above form of the free energy expression. All the parameters, y, y_z, and σ, are assumed to be determined by the above stability conditions.

2.1. The σ-dependence of the correction term

In order to find the explicit form of the free energy correction ΔF_1, let us study its σ-dependence in some particular temperature ranges below.

- *In the ground state at $T = 0$ K.* In this case, the σ-dependence of $y(\sigma,0)$ and $y_z(\sigma,0)$ is given by

$$y(\sigma,0) = y_{10}(\sigma^2-\sigma_{\rm s}^2), \qquad y_z(\sigma,0) = 2y_{10}\sigma_{\rm s}^2 + 2y_{10}(\sigma^2-\sigma_{\rm s}^2) \tag{2.15}$$

where y_{10} represents the ratio of the spin fluctuation parameters given by $60c_z y_{10} = T_A/T_0$ (Takahashi 2001). Because the thermal amplitudes $A(y,t)$ and $A(y_z,t)$ vanish identically, the σ-dependence of ΔF_1 can be simply determined as follows. From the definition of (2.10) the parameter $\lambda(\sigma,t)$ is given by

$$\lambda(\sigma,0) = -\frac{2T_0}{T_A}c_z\Delta y_z - \frac{1}{12}\sigma^2.$$

The correction ΔF_1 is then determined by solving the equation

$$\frac{1}{T_A}\frac{\partial}{\partial\sigma}\left(\frac{\Delta F_1}{N_0}\right) = \left(\frac{2T_0}{T_A}c_z\Delta y_z + \frac{1}{12}\sigma^2\right)\frac{\partial\Delta y_z}{\partial\sigma}$$
$$= \left(\tfrac{1}{15}+\tfrac{1}{12}\right)[(\sigma^2-\sigma_s^2)+\sigma_s^2](4y_{10}\sigma)$$
$$= \frac{3}{5}y_{10}\sigma(\sigma^2-\sigma_s^2) + \frac{3}{20}\sigma_s^2\frac{\partial\Delta y_z}{\partial\sigma}.$$

The solution is given by

$$\Delta F_1(\sigma,0) = \tfrac{3}{20}N_0T_A\sigma_s^2\Delta y_z + \tfrac{3}{20}N_0T_Ay_{10}(\sigma^2-\sigma_s^2)^2. \tag{2.16}$$

- *In the paramagnetic phase for $T > T_c$.* The following expansions for y and y_z are justified in the paramagnetic phase in the small σ^2 limit.

$$y(\sigma,t) = y_0(t) + y_1(t)\sigma^2 + \cdots, \qquad y_z(\sigma,t) = y_0(t) + 3y_1(t)\sigma^2 + \cdots$$

where $y_1(t)$ is defined by

$$y_1(t) = \frac{c_zy_{10}}{c_z - A'(y_0(t),t)}.$$

The partial y-derivative of $A(y,t)$ is hereafter denoted by $A'(y,t)$. By expanding $\lambda(\sigma,t)$ in powers of σ^2, $\Delta F_1(\sigma,t)$ can be determined by solving

$$\frac{1}{N_0T_A}\frac{\partial\Delta F_1(\sigma,t)}{\partial\sigma} = \left\{-\frac{2T_0}{T_A}[A(y_z,t)-A(y,t)-c_z\Delta y_z] + \frac{\sigma^2}{12}\right\}\frac{\partial\Delta y_z}{\partial\sigma}$$
$$\simeq -\left\{\frac{2T_0}{T_A}[A'(y_0,t)-c_z]\Delta y_z - \frac{\sigma^2}{12}\right\}4y_1(t)\sigma$$
$$= 4\sigma^3y_1(t)\left(\tfrac{1}{15}+\tfrac{1}{12}\right) = \tfrac{3}{5}y_1(t)\sigma^3.$$

The solution is given by

$$\Delta F_1(\sigma,t) \simeq \tfrac{3}{20}N_0T_Ay_1(t)\sigma^4 + \cdots, \qquad \lambda(\sigma,t) = -\tfrac{3}{20}\sigma^2 + \cdots.$$

The lowest-order term vanishes at the critical temperature, $t = t_c$, because $y_1(t_c) = 0$.

- *In the ordered phase for $T < T_c$.* In the presence of the spontaneous magnetic moment $\sigma_0(t)$, the σ^2-dependence of y and y_z is given by

$$y(\sigma,t) = y_1(t)[\sigma^2 - \sigma_0^2(t)],$$

$$y_z(\sigma,t) = y(\sigma,t) + 2y_1(t)\sigma^2 = 2y_1(t)\sigma_0^2(t) + 3y(\sigma,t)$$
$$= y_{z0}(t) + 3y(\sigma,t).$$

In the small y limit, the following expansion of the parameter $\lambda(\sigma,t)$ is well justified:

$$\lambda(\sigma,t) = \frac{2T_0}{T_A}\left[A(y_z,t) - A_t(y,t) - c_z(y_z-y)\right] - \frac{\sigma^2}{12} - \frac{\eta_1}{3}$$
$$= \lambda_0(t) + (\sigma^2-\sigma_0^2)$$
$$\times\left\{\frac{2T_0}{T_A}[3A'(y_{z0},t) - A_t'(0,t) - 2c_z]y_1(t) - \frac{1}{12}\right\} + \cdots$$

where $\lambda_0(t)$, the value in the absence of the magnetic field, is defined by

$$\lambda_0(t) = \frac{2T_0}{T_A}\left[A(y_{z0},t) - A_t(0,t) - c_zy_{z0}(t)\right] - \frac{\sigma_0^2(t)}{12}. \tag{2.17}$$

In the paramagnetic phase, $\lambda_0(t)$ vanishes identically because $\sigma_0(t) = 0$ and therefore $y_{z0}(t) = 0$ holds there. The correction term $\Delta F_1(\sigma, t)$ is determined by solving

$$\frac{1}{N_0 T_A}\frac{\partial \Delta F_1}{\partial \sigma} = -4y_1(t)\sigma(\sigma^2 - \sigma_0^2) \times \left\{\frac{2T_0}{T_A}[3A'(y_{z0}, t) - A'_t(0, t) - 2c_z]y_1(t) - \frac{1}{12}\right\} - \lambda_0(t)\frac{\partial \Delta y_z}{\partial \sigma} + \cdots.$$

Its solution is given by

$$\Delta F_1(\sigma, t) = -N_0 T_A \lambda_0(t)\Delta y_z + N_0 T_A y_1(t)[\sigma^2 - \sigma_0^2(t)]^2 \times \left\{\frac{1}{12} - \frac{2T_0}{T_A}[3A'(y_{z0}, t) - A'_t(0, t) - 2c_z]y_1(t)\right\}. \quad (2.18)$$

The above form of the free energy correction is quite general. It tends to the ground state expression in the $t = 0$ limit. For instance, the parameter $\lambda_0(t)$ is, in this limit, given by

$$\lambda_0(0) = \frac{4T_0}{T_A}c_z y_{10}\sigma_s^2 - \frac{1}{12}\sigma_s^2 = \frac{3}{20}\sigma_s^2.$$

It also agrees with the expression in the paramagnetic phase.

- *At the critical temperature,* $T = T_c$. Owing to the critical y-dependence of the thermal amplitude $A(y, t) \simeq A(0, t) - \pi t\sqrt{y}/4$, the parameter $\lambda(\sigma, t_c)$ can be given by

$$\lambda(\sigma, t_c) = -\frac{2\pi T_0 t_c}{4T_A}(\sqrt{y_z} - \sqrt{y}) - \frac{\sigma^2}{12}.$$

If we take the σ^4-dependence of y and y_z (Takahashi 2001) into account,

$$y(\sigma, t_c) = y_c\sigma^4, \qquad y_z(\sigma, t_c) = 5y_c\sigma^4, \qquad y_c = \left[\frac{20c_z y_{10}}{\pi(2+\sqrt{5})t_c}\right]^2 \quad (2.19)$$

we are led to the following σ-dependence:

$$\lambda(\sigma, t_c) \simeq -\left[\frac{2\pi T_0 t_c}{4T_A}(\sqrt{5}-1)\sqrt{y_c} + \frac{1}{12}\right]\sigma^2 = -\left[\frac{(\sqrt{5}-1)}{6(2+\sqrt{5})} + \frac{1}{12}\right]\sigma^2 = \frac{\sqrt{5}}{4(2+\sqrt{5})}\sigma^2.$$

From the above result, $\Delta F_1(\sigma, t)$ is given by

$$\frac{1}{N_0 T_A}\frac{\partial \Delta F_1(\sigma, t_c)}{\partial \sigma} = -\lambda(\sigma, t_c)\frac{\partial \Delta y_z}{\partial \sigma} = \frac{4\sqrt{5}}{(2+\sqrt{5})}y_c\sigma^5 + \cdots$$

$$\Delta F_1(\sigma, t_c) = \frac{2\sqrt{5}}{3(2+\sqrt{5})}N_0 T_A y_c\sigma^6.$$

To summarize, we have extended our paramagnetic form of the free energy correction to the general cases where finite static magnetization is present in the system, because of the presence of the spontaneous moment below T_c and/or due to the application of the external magnetic field. We have found that an additional correction term,

$$\Delta F_1(\sigma, t) = -N_0 T_A \lambda_0(t)\Delta y_z(\sigma, t) + \cdots, \quad (2.20)$$

is necessary. In the following sections, we will show how the above correction will give rise to new behaviours of the entropy and the specific heat in their temperature and magnetic field dependence that were disregarded in previous studies.

3. Magnetic entropy and specific heat

The temperature dependence of our free energy comes from two origins: the direct dependence related to the Bose factor, and the implicit dependence through the parameters $y(\sigma, t)$ and $y_z(\sigma, t)$. We assume that the t- and σ-dependence of $y(\sigma, t)$ and $y_z(\sigma, t)$ are determined by the following conditions: the stability of the free energy against the variation of y, and the validity of the thermodynamic relation derived from the σ-derivative. Under these constraints, the magnetic entropy can be obtained as the temperature derivative of the free energy (2.1) as follows.

$$
\begin{aligned}
S_{\mathrm{m}}(\sigma, t)/N_0 = &-6 \int_{x_c}^{1} \mathrm{d}x\, x^2 \left[\ln \sqrt{2\pi} - u + (u - 1/2)\ln u - \ln \Gamma(u)\right] \\
&+ 6 \int_{x_c}^{1} \mathrm{d}x\, x^2 u \left[\ln u - \frac{1}{2u} - \psi(u)\right] \\
&- 3 \int_{0}^{1} \mathrm{d}x\, x^3 \left[\ln \sqrt{2\pi} - u_z + (u_z - 1/2)\ln u_z - \ln \Gamma(u_z)\right] \\
&+ 3 \int_{0}^{1} \mathrm{d}x\, x^2 u_z \left[\ln u_z - \frac{1}{2u_z} - \psi(u_z)\right] \\
&- \frac{1}{N_0} \sum_{q<q_{\mathrm{sw}}} \left[\ln(1 - \mathrm{e}^{-\beta\omega_q}) - \beta\omega_q n(\omega_q)\right] \\
&- \frac{T_A}{T_0}\left(y \frac{\partial \eta_1}{\partial t} + \frac{\partial \eta_0}{\partial t}\right) + \Delta S_{\mathrm{m}}(\sigma, t)/N_0
\end{aligned}
\qquad (3.1)
$$

where $\Gamma(u)$ is the gamma function of the argument u.

The above expression is an extension of our previous formula in the paramagnetic phase (Takahashi 1999). The last specific term ΔS_{m} of this treatment comes from the free energy correction ΔF_1. We can associate its σ- and t-dependence with those of $\lambda(\sigma, t)$. If we note the relation $\delta\Delta y_z = \partial(\Delta y_z/\partial t)\delta t$ in (2.7) for the explicit t-dependence, it is easy to see that $\Delta S_{\mathrm{m}}(\sigma, t)$ should be defined by

$$
T_0 \Delta S_{\mathrm{m}}(\sigma, t) = -\frac{\partial \Delta F_1(\sigma, t)}{\partial t} - N_0 T_A \lambda(\sigma, t) \frac{\partial \Delta y_z(\sigma, t)}{\partial t}. \qquad (3.2)
$$

On substitution of (2.20), we obtain

$$
\begin{aligned}
\Delta S_{\mathrm{m}}(\sigma, t) &= \Delta S_{\mathrm{m}}(\sigma_0, t) + \delta \Delta S_{\mathrm{m}}(\sigma, t) \\
\Delta S_{\mathrm{m}}(\sigma_0, t) &= N_0 \frac{T_A}{T_0} \frac{\mathrm{d}\lambda_0(t)}{\mathrm{d}t} y_{z0}(t)
\end{aligned}
\qquad (3.3)
$$

$$
\delta \Delta S_{\mathrm{m}}(\sigma, t) \simeq N_0 \frac{T_A}{T_0} \left[\frac{\mathrm{d}\lambda_0(t)}{\mathrm{d}t} \Delta y_z(\sigma, t) - \delta\lambda(\sigma, t) \frac{\mathrm{d}y_{z0}(t)}{\mathrm{d}t}\right] \qquad (3.4)
$$

where $\delta\lambda = \lambda(\sigma, t) - \lambda(\sigma_0, t)$. In the absence of the external field, the temperature dependence of the entropy correction is evaluated by (3.3), since $\Delta y_z = y_{z0}(t)$ and $\delta\lambda = 0$ hold for $h = 0$. The field dependence is given by (3.4).

The temperature dependence of the specific heat is now derived as the t-derivative of the entropy in (3.1). It is shown below explicitly.

$$\frac{C_{\rm m}}{N_0 t} = \frac{6}{t}\int_{x_{\rm c}}^{1} {\rm d}x\, x^2 u\left\{-1 - 1/2u + u\psi'(u)\right\}$$
$$+\frac{3}{t}\int_0^1 {\rm d}x\, x^2 u_z\left\{-1 - 1/2u_z + u_z\psi'(u_z)\right\}$$
$$-3\left(2\frac{\partial A_t(y,t)}{\partial t}\frac{\partial y}{\partial t}\bigg|_h + \frac{\partial A(y_z,t)}{\partial t}\frac{\partial y_z}{\partial t}\bigg|_h\right)$$
$$+3\left(\frac{\sigma T_A}{tT_0}\right)^2\int_0^{x_{\rm c}} {\rm d}x\, x^2\frac{{\rm e}^{\beta\omega_q}(y+x^2)^2}{({\rm e}^{\beta\omega_q}-1)^2}\left(\frac{1}{t} - \frac{1}{\sigma}\frac{\partial\sigma}{\partial t}\bigg|_h\right) \tag{3.5}$$
$$-\frac{T_A}{T_0}\left(\frac{\partial y}{\partial t}\frac{\partial\eta_1}{\partial t} + y\frac{\partial^2\eta_1}{\partial t^2} + \frac{\partial^2\eta_0}{\partial t^2}\right) + \frac{\Delta C_{\rm m}}{N_0 t}$$
$$\frac{\Delta C_{\rm m}}{t} = \frac{\partial\Delta S_{\rm m}}{\partial t}.$$

We are now ready to discuss the temperature and the external field dependence of the entropy and the specific heat.

4. Temperature dependence of the entropy and specific heat

For the numerical analysis of the temperature dependence, let us rewrite our expression of $\Delta S_{\rm m}$ in (3.3) with the use of the explicit t-dependence of the parameter $\lambda_0(t)$ defined in (2.17). The t-derivative of $\lambda_0(t)$ is given by

$$\frac{T_A}{T_0}\frac{{\rm d}\lambda_0(t)}{{\rm d}t} = 2[A'(y_{z0},t) - c_z]\frac{{\rm d}y_{z0}}{{\rm d}t} - 5c_z y_{10}\frac{{\rm d}\sigma_0^2}{{\rm d}t}$$
$$+2\left[\frac{\partial A(y_{z0},t)}{\partial t} - \frac{\partial A_t(0,t)}{\partial t}\right]$$
$$= -6\frac{\partial A_t(0,t)}{\partial t} - 15c_z y_{10}\frac{{\rm d}\sigma_0^2(t)}{{\rm d}t}. \tag{4.1}$$

The last line is derived with the use of the following partial t-derivative of the amplitude sum rule (2.8), i.e.,

$$2\frac{\partial A_t(0,t)}{\partial t} + \frac{\partial A(y_{z0},t)}{\partial t} + [A'(y_{z0},t) - c_z]\frac{{\rm d}y_{z0}}{{\rm d}t} + 5c_z y_{10}\frac{{\rm d}\sigma_0^2}{{\rm d}t} = 0. \tag{4.2}$$

In the same way, (4.1) can take the different form

$$\frac{T_A}{T_0}\frac{{\rm d}\lambda_0(t)}{{\rm d}t} = 3\frac{\partial A(y_{z0},t)}{\partial t} + 3[A'(y_{z0},t) - c_z]\frac{{\rm d}y_{z0}}{{\rm d}t}. \tag{4.3}$$

On substitution of either (4.1) or (4.3) into (3.3), the entropy correction $\Delta S_{\rm m}$ can be given in two equivalent forms,

$$\frac{\Delta S_{\rm m}}{N_0} = -3y_{z0}\left[2\frac{\partial A_t(0,t)}{\partial t} + 5c_z y_{10}\frac{{\rm d}\sigma_0^2(t)}{{\rm d}t}\right] \tag{4.4}$$
$$= 3y_{z0}\left\{\frac{\partial A(y_{z0},t)}{\partial t} + [A'(y_{z0},t) - c_z]\frac{{\rm d}y_{z0}}{{\rm d}t}\right\}. \tag{4.5}$$

From the straightforward t-differentiation of (4.4), the specific heat correction $\Delta C_{\rm m}$ is given by

$$\frac{\Delta C_{\rm m}}{N_0 t} = -3\bigg[2\frac{\partial A_t(0,t)}{\partial t}\frac{{\rm d}y_{z0}}{{\rm d}t} + 2y_{z0}\frac{\partial^2 A_t(0,t)}{\partial t^2} + 5c_z y_{10}\frac{\rm d}{{\rm d}t}\left(y_{z0}\frac{{\rm d}\sigma_0^2(t)}{{\rm d}t}\right)\bigg] \tag{4.6}$$

in terms of thermal transverse spin fluctuation amplitude $A_t(0,t)$. From (4.5) it is also represented by

$$\frac{\Delta C_{\rm m}}{N_0 t} = 3y_{z0}\left[\frac{\partial^2 A(y_{z0},t)}{\partial t^2} + A''(y_{z0},t)\left(\frac{{\rm d}y_{z0}}{{\rm d}t}\right)^2 + 2\,\frac{\partial A'(y_{z0},t)}{\partial t}\frac{{\rm d}y_{z0}}{{\rm d}t}\right] + 3\,\frac{\partial A(y_{z0},t)}{\partial t}\frac{{\rm d}y_{z0}}{{\rm d}t} + 3[A'(y_{z0},t) - c_z]\left[\left(\frac{{\rm d}y_{z0}}{{\rm d}t}\right)^2 + y_{z0}\frac{{\rm d}^2 y_{z0}}{{\rm d}t^2}\right] \tag{4.7}$$

in terms of longitudinal amplitudes $A(y_{z0},t)$. On substituting these results in (3.5), the temperature dependence of the magnetic specific heat is finally given as the sum of two contributions,

$$\frac{C_{\rm m}}{N_0 t} = \frac{C_{\rm m0}}{N_0 t} + \frac{C_{\rm m1}}{N_0 t} \tag{4.8}$$

$$\frac{C_{\rm m0}}{N_0 t} = 6I + 3I_z$$
$$I = \frac{1}{t}\int_{x_{\rm c}}^1 {\rm d}x\, x^2 u[-1 - 1/2u + u\psi'(u)] \tag{4.9}$$
$$I_z = \frac{1}{t}\int_0^1 {\rm d}x\, x^2 u_z[-1 - 1/2u_z + u\psi'(u_z)]$$

$$\frac{C_{\rm m1}}{N_0 t} = -3\,\frac{\partial A(y_{z0},t)}{\partial t}\frac{\partial y_{z0}}{\partial t} + \frac{\Delta C_{\rm m}}{N_0 t} = -3\left\{2\,\frac{\partial A_t(0,t)}{\partial t} + \frac{\partial A(y_{z0},t)}{\partial t}\right\}\frac{{\rm d}y_{z0}}{{\rm d}t} - 6y_{z0}\,\frac{\partial^2 A_{\rm c}(0,t)}{\partial t^2} - 15c_z y_{10}\left(\frac{{\rm d}\sigma_0^2}{{\rm d}t}\frac{{\rm d}y_{z0}}{{\rm d}t} + y_{z0}\frac{{\rm d}^2\sigma_0^2}{{\rm d}t^2}\right). \tag{4.10}$$

We have neglected spin-wave contributions, the second-order T-derivative of $A_{\rm sw}$ for instance, by assuming them to be very small. The first term of (4.10) reduces to the similar isotropic expression in the paramagnetic phase. The rest of the terms vanish because of the absence of finite σ_0^2 and y_{z0} there. Equation (4.10) is therefore regarded as a natural extension of our paramagnetic result to the ordered phase. The term $C_{\rm m1}$ is also given by the following equivalent form:

$$\frac{C_{\rm m1}}{N_0 t} = 3y_{z0}\left[\frac{\partial^2 A(y_{z0},t)}{\partial t^2} + A''(y_{z0},t)\left(\frac{{\rm d}y_{z0}}{{\rm d}t}\right)^2 + 2\,\frac{\partial A'(y_{z0},t)}{\partial t}\frac{{\rm d}y_{z0}}{{\rm d}t}\right] + 3[A'(y_{z0},t) - c_z]\left[\left(\frac{{\rm d}y_{z0}}{{\rm d}t}\right)^2 + y_{z0}\frac{{\rm d}^2 y_{z0}}{{\rm d}t^2}\right]. \tag{4.11}$$

The presence of the second-order t-derivative terms in (4.10) and (4.11) is one of the distinct results derived first in this study. In contrast, only the first-order derivative ${\rm d}M/{\rm d}T$ appears in previous studies based on the SCR theory (Makoshi and Moriya (1975) for instance).

In order to derive numerical results, we need to know the t-dependence of $\sigma_0(t)$, $y_{z0}(t)$, and their t-derivatives. These quantities can be evaluated according to our previous study on the magnetic isotherm of itinerant electron magnets (Takahashi 2001). See appendix A for a brief explanation of how to evaluate them. We show in figure 1 numerical results of the t-dependence of $C_{\rm m}/N_0 t$ for several values of the parameter $t_{\rm c}$ in a wide temperature range. We particularly notice two characteristic features shown in the figure: the rapid enhancement in the low temperature limit and the appearance of the peak around the critical temperature.

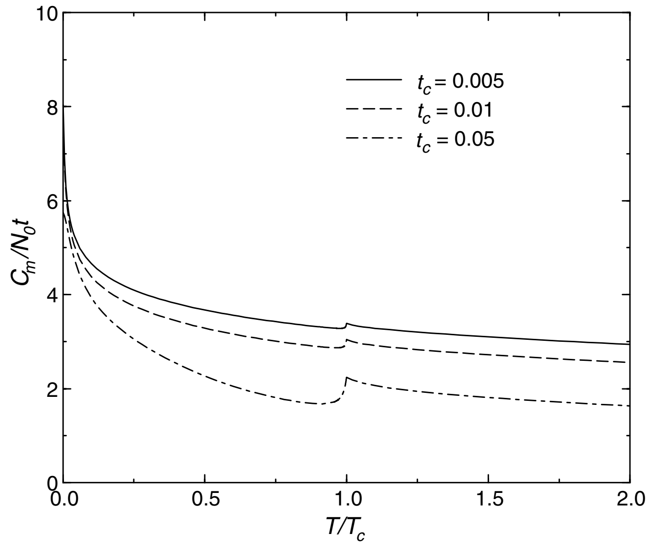

Figure 1. Temperature dependence of the specific heat $C_{\rm m}/N_0t$ for $t_{\rm c} = 0.005$, 0.01, and 0.05 from the top, respectively.

We show below the temperature dependence of them in more detail, paying particular attention on the regions in the low temperature limit as well as around the critical point.

4.1. In the low temperature limit

The first term $C_{\rm m0}/N_0t$ in (4.8) dominates in the low temperature limit. Actually, in this limit, the longitudinal part I_z in (4.9) shows the well-known logarithmic enhancement,

$$I_z \simeq \tfrac{1}{12}\ln(1 + 1/y_{z0}(0)),$$

as the longitudinal inverse magnetic susceptibility $y_{z0}(0)$ becomes very small. For the transverse part, we have to take account of the cut-off effect for the lower bound of the wavevector integration due to the presence of spin-wave modes. It is approximated by

$$I \simeq \frac{1}{6}\int_{x_{\rm c}}^{1} {\rm d}x\,\frac{x^2}{x(y+x^2)} = \frac{1}{12}\ln\left(\frac{1+y}{y+x_{\rm c}^2}\right) = \frac{1}{6}\ln\left(\frac{1}{x_{\rm c}}\right),$$

for $y = 0$ in the absence of the external field. The first term $C_{\rm m0}/N_0t$ therefore gives the enhancement,

$$C_{\rm m0}/N_0t \simeq \ln\frac{1}{x_{\rm c}} + \frac{1}{4}\ln\frac{1}{y_{z0}} \simeq \frac{3}{2}\ln\frac{1}{\sigma_{\rm s}} \tag{4.12}$$

as we approach the magnetic instability point, $\sigma_{\rm s} \to 0$.

On the other hand, among terms of $C_{\rm m1}/N_0t$ in (4.11), only the terms proportional to the second-order t-derivative of $A(y_{z0}, t)$ and $y_{z0}(t)$ remain finite in this limit as shown below:

$$\begin{aligned} &3y_{z0}\frac{\partial^2 A(y_{z0},t)}{\partial t^2} \simeq \frac{1}{4}, \\ &3[A'(y_{z0},t) - c_z]y_{z0}\frac{{\rm d}^2 y_{z0}}{{\rm d}t^2} \simeq \frac{1}{12}(5+4r+2r^2) \end{aligned} \tag{4.13}$$

where $r = y_{z0}/x_{\rm c}^2$ is a temperature-independent constant introduced by Takahashi (2001). All the other terms vanish in this limit, for they are proportional to t^2. For more explanation

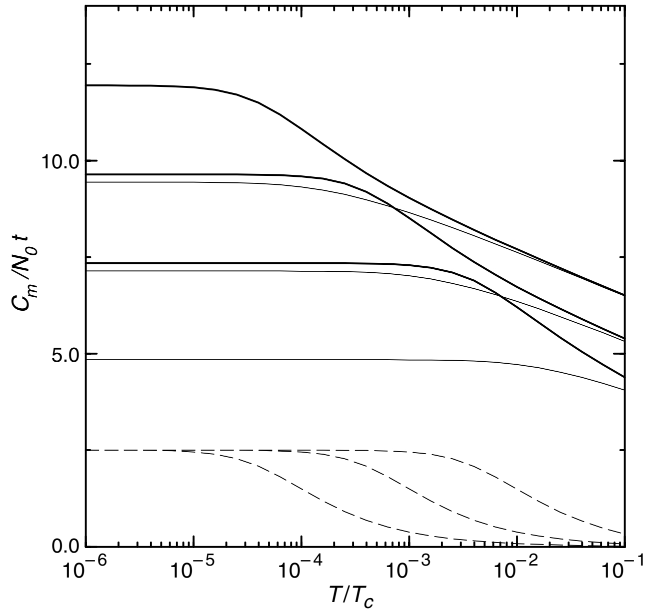

Figure 2. Temperature dependence of the specific heat C_m/N_0t against a logarithmic scale of the temperature for $t_c = 0.005$, 0.01, and 0.05 from the top, respectively, at low temperature. The thin and broken curves represent components C_{m0}/N_0t and C_{m1}/N_0t, respectively.

of the temperature dependence of y_{z0}, see appendix A, for instance. Therefore the second term C_{m1}/N_0t gives the following additional enhancement to the T-linear specific heat coefficient:

$$\frac{\Delta C_{m1}}{N_0 t} \simeq \frac{1}{6}(4 + 2r + r^2) = 2.5038\ldots, \qquad \text{for } r = \pi^2/4. \tag{4.14}$$

The reason of the choice of the above value of r will be given later. Although ΔC_{m1} does not show any divergence in the limit of the magnetic instability, $t_c \to 0$, it gives a sizable extra enhancement to the T-linear specific coefficient for cases with moderate t_c values. Its value is independent of the value of t_c. We show numerical results of C_m/N_0t and its components C_{m0}/N_0t and C_{m1}/N_0t in figure 2 in the low temperature region against a logarithmic scale of the temperature. The relative importance of the additional enhancement is evident for cases with larger t_c. It amounts to about 30% of the total enhancement for $t_c = 0.01$. From the figure we also see that its presence is limited to the very low temperature region depending on the value of t_c, the lower temperature region for the smaller t_c. Because of the presence of this term, the shoulders of the temperature dependence in figure 2 become a bit distinct. The dominance of the second-order t-derivative terms can be recognized from a comparison of figures 2 and 3, where only the sum of the first and the fifth terms of C_{m1}/N_0t is plotted against the temperature.

No such extra enhancement of the specific heat was mentioned previously. It results from the second-order t-derivative terms in the correction term ΔC_m. It is related to the t^2-dependence of magnetic properties, such as the saturation moment $\sigma_0^2(t)$, in the low temperature limit. For a quantitative analysis of experiments, we have to be careful of this additional effect involved in the observed enhancement.

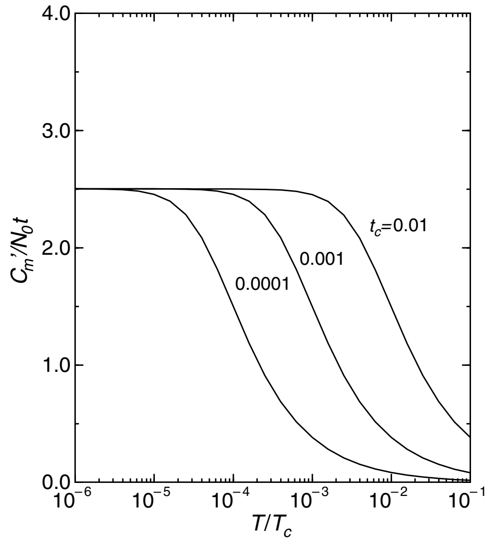

Figure 3. Temperature dependence of $C'_{\rm m}/N_0t$, the sum of two second order t-derivative terms of $C_{\rm m1}/N_0t$, against the logarithmic scale of the temperature for $t_{\rm c} = 0.005$, 0.01, and 0.05 from the top, respectively, at low temperature.

4.2. Around the critical temperature below t_c ($t \leqslant t_c$)

In order to clarify the distinction of our treatments between cases in the ordered and the paramagnetic phases around the critical region, let us next study the temperature dependence of the second term $C_{\rm m1}/N_0t$ in (4.8), the novel term in this treatment. In this limit it can be well approximated by the following sum of dominant terms,

$$\frac{C_{\rm m1}}{N_0t} = 3y_{z0}A''(y_{z0},t)\left(\frac{{\rm d}y_{z0}}{{\rm d}t}\right)^2 + 3A'(y_{z0},t)\left[\left(\frac{{\rm d}y_{z0}}{{\rm d}t}\right)^2 + y_{z0}\frac{{\rm d}^2y_{z0}}{{\rm d}t^2}\right]$$
$$= 3\left(\frac{{\rm d}y_{z0}}{{\rm d}t}\right)^2[y_{z0}A''(y_{z0},t) + A'(y_{z0},t)] + 3y_{z0}A'(y_{z0},t)\frac{{\rm d}^2y_{z0}}{{\rm d}t^2} \qquad (4.15)$$

if we take into account the critical behaviours, $A'(y_z,t) \propto t/\sqrt{y_z}$ and $y_{z0}(t) \propto (t - t_{\rm c})^2$. According to Takahashi (2001), the temperature dependence of $\sigma_0^2(t)$ and $y_{z0}(t)$ is given by

$$\sigma_0^2(t) \simeq a_{\rm c}\sigma_{\rm s}^2[1 - (t/t_{\rm c})^{4/3}]$$
$$y_{z0}(t) = 2y_1(t)\sigma_0^2(t), \qquad y_1(t) \simeq y'_{\rm c}\sigma_0^2(t) \qquad (4.16)$$

where $y'_{\rm c}$ and $a_{\rm c}$ are defined by

$$y'_{\rm c} = \left[\frac{2\sqrt{2}(2+\sqrt{5})}{2\xi+3}\right]^2 y_{\rm c}, \qquad a_{\rm c} = \frac{3(2\xi+3)}{5(2\xi-1)} = \frac{7}{5}, \qquad \xi = \frac{4r^{1/2}}{\pi} = 2.$$

From these results it follows that the t-derivatives in (4.15) are given by

$$\frac{{\rm d}y_{z0}(t)}{{\rm d}t} = 4y'_{\rm c}\sigma_0^2(t)\frac{\partial\sigma_0^2(t)}{\partial t} = 4y'_{\rm c}\sigma_0^2(t)\left(-\frac{4a_{\rm c}\sigma_{\rm s}^2}{3t_{\rm c}}\right)$$
$$\frac{{\rm d}^2y_{z0}(t)}{{\rm d}t^2} = 4y'_{\rm c}\left(\frac{4a_{\rm c}\sigma_{\rm s}^2}{3t_{\rm c}}\right)^2, \qquad \frac{{\rm d}\sigma_0^2(t)}{{\rm d}t} = -\frac{4a_{\rm c}\sigma_{\rm s}^2}{3t_{\rm c}}.$$

In addition, if we note the following critical behaviour of $A'(y_{z0}, t)$,

$$A'(y_{z0}, t) \simeq -\frac{\pi t_c}{8\sqrt{y_{z0}}} \simeq -\frac{\pi t_c}{8\sqrt{2y_c'\sigma_0^2(t)}}$$

we can get the critical t-dependence,

$$[y_{z0}A''(y_{z0}, t) + A'(y_{z0}, t)] \left(\frac{\mathrm{d}y_{z0}}{\mathrm{d}t}\right)^2 \simeq \left[\frac{\pi t_c}{16\sqrt{y_{z0}}} - \frac{\pi t_c}{8\sqrt{y_{z0}}}\right] \frac{\mathrm{d}y_{z0}}{\mathrm{d}t}$$
$$= -\frac{\pi t_c}{\sqrt{2}}(y_c')^{3/2} \left(\frac{4a_c\sigma_s^2}{3t_c}\right)^2 \sigma_0^2(t).$$

On substitution of the results into (4.15), the critical t-dependence of $C_{m1}/N_0 t$ is given as follows:

$$\begin{aligned}
\frac{C_{m1}}{N_0 t} &= -3\pi t_c y_c' \sqrt{2y_c'} \left(\frac{4a_c\sigma_s^2}{3t_c}\right)^2 \sigma_0^2(t) \\
&= 12(3 - 5a_c)y_c' \left(\frac{4a_c\sigma_s^2}{3t_c}\right)^2 A(0, t_c)[1 - (t/t_c)^{4/3}] \\
&= \frac{4}{3}(5a_c/3 - 1) \left[\frac{320\sqrt{2}a_c}{7\pi}\right]^2 \left(\frac{C_{4/3}}{3}\right)^3 \left(\frac{t}{t_c} - 1\right) \\
&= 55.656\ldots \times \left(\frac{t}{t_c} - 1\right)
\end{aligned} \tag{4.17}$$

where we have used the relation

$$A(0, t_c) = \frac{C_{4/3}}{3} t_c^{4/3}, \qquad \frac{\pi t_c \sqrt{2y_c'}}{4c_z y_{10}} = 5 - \frac{3}{a_c} = \frac{20}{7}.$$

It follows from (4.17) that the specific heat exhibits t-linear dependence with positive slope just below the critical temperature. The magnitude of the slope is independent of t_c, if we plot $C_m/N_0 t$ against the reduced temperature, $t/t_c = T/T_c$. Because the slope is negative just above t_c, it causes a cusp anomaly in its temperature dependence. No such peak anomaly was mentioned in previous theoretical studies. It originates from the second-order t-derivative terms. For reference, we show numerical results of the t-dependence of $C_{m1}/N_0 t$ in figure 4 for several values of t_c in the narrow temperature region below T_c. The sharp critical peak anomaly of the specific heat was observed by Fawcett *et al* (1970) on MnSi that has a moderate value of $t_c = 0.2$–0.3.

The results of this section on the temperature dependence are summarized as follows. The following new features are all related to the presence of the correction term ΔC_m. In particular, the second-order t-derivative terms are responsible for them.

- In the low temperature limit, an additional enhancement to the T-linear coefficient of the specific heat is present. The origin is related to the t^2 temperature dependence of various magnetic properties, such as $\sigma_0^2(t)$ and $y_{z0}(t)$. The dependence is specific to itinerant electron magnets near the magnetic instability point, and is limited to the very low temperature region.
- Around the critical temperature, the specific heat exhibits a critical peak anomaly because the slope of its t-dependence changes its sign. However, for those magnets with very small values of t_c, the behaviour is not so distinct and it will be hard to be observed experimentally, for the above t-linear dependence is limited to the very narrow region around t_c as shown in figure 4.

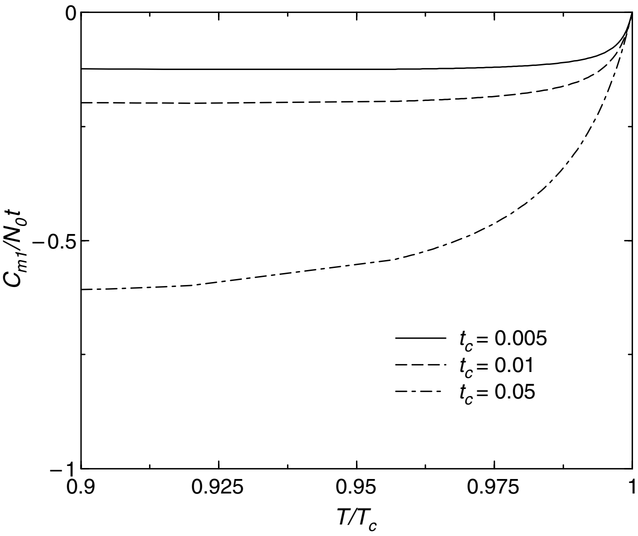

Figure 4. Temperature dependence of C_{m1}/N_0t around the critical region for $t_c = 0.005$, 0.01, and 0.05.

5. Magnetic field dependence of specific heat

The magnetic field dependence of the entropy and the specific heat is the subject of this section. The following discussion is based on the results (3.1) and (4.8)–(4.11). In order to find their temperature dependence in the presence of an external magnetic field H, we have first to determine the field-induced magnetic moment $\sigma(h, t)$. Then the inverse magnetic susceptibilities, $y(\sigma, t)$ and $y_z(\sigma, t)$, have to be determined corresponding to the σ-value. For the numerical estimate of σ, we can use the sum rule of the spin amplitude (2.8) derived as the stability condition of the free energy. On neglecting the tiny spin-wave term, it can be written as follows:

$$\left[2A_t(y, t) + A(y_z, t) - c_z(2y + y_z)\right] + 5c_z y_{10}\sigma^2 = 3A(0, t_c). \tag{5.1}$$

The reduced parameters y_z and y defined in (2.5) are proportional to $\partial H/\partial M$ and H/M. By solving (5.1), we can find y_z, i.e. the first-order derivative $\partial\sigma/\partial h$, as a function of $y = h/T_A\sigma$ and σ, i.e. as a function of σ and h. This means that (5.1) is regarded as an implicit form of the first-order differential equation for σ as a function of h. We have already shown how to determine its initial condition, i.e. the spontaneous moment $\sigma_0(t)$ below t_c in the absence of the field (Takahashi 2001). In the paramagnetic phase, the condition is given by $\sigma_0(t) = 0$ for $h = 0$. In the expression (3.5) we also need to know the t-derivatives of y and y_z in the presence of the magnetic field H for numerical estimates of the specific heat. They are evaluated with the use of the following equation that is derived by the partial t-derivative of (5.1):

$$2[A_t'(y, t) - c_z] \left.\frac{\partial y}{\partial t}\right|_h + [A'(y_z, t) - c_z] \left.\frac{\partial y_z}{\partial t}\right|_h + 10c_z y_{10}\sigma \left.\frac{\partial \sigma}{\partial t}\right|_h + 2\frac{\partial A_t(y, t)}{\partial t} + \frac{\partial A(y_z, t)}{\partial t} = 0. \tag{5.2}$$

Let us note that the t-derivatives of y and y_z under the constant H condition are related to the derivative $\partial\sigma/\partial t$ as follows:

$$\left.\frac{\partial y}{\partial t}\right|_h = -\frac{h}{T_A\sigma^2}\left.\frac{\partial\sigma}{\partial t}\right|_h = -\frac{y}{\sigma}\left.\frac{\partial\sigma}{\partial t}\right|_h$$
$$\left.\frac{\partial y_z}{\partial t}\right|_h = -\frac{1}{T_A}\frac{1}{(\partial\sigma/\partial h)^2}\frac{\partial}{\partial t}\left(\frac{\partial\sigma}{\partial h}\right) = -T_A y_z^2\frac{\partial}{\partial h}\left(\frac{\partial\sigma}{\partial t}\right). \tag{5.3}$$

On substitution of the results into (5.2) we obtain the following differential equation:

$$[A'(y_z,t) - c_z]T_A y_z^2\frac{\partial}{\partial h}\left(\frac{\partial\sigma}{\partial t}\right) = \left\{-2[A_t'(y,t) - c_z]\frac{y}{\sigma} + 10c_z y_{10}\sigma\right\}\left.\frac{\partial\sigma}{\partial t}\right|_h$$
$$+ 2\frac{\partial A_t(y,t)}{\partial t} + \frac{\partial A(y_z,t)}{\partial t}. \tag{5.4}$$

By solving (5.4) simultaneously with (5.1) under a suitable initial condition, we can numerically determine $\partial\sigma/\partial t|_h$ as a function of the external field h. The initial condition at $h = 0$ is given by

$$\frac{\partial\sigma(h,t)}{\partial t} = \begin{cases} 0 & \text{for } t > t_c \\ \dfrac{d\sigma_0(t)}{dt} & \text{for } t < t_c. \end{cases}$$

The partial t-derivatives of y and y_z are then estimated from the relation (5.3).

In what follows, let us discuss the field dependence of the magnetic entropy and the specific heat in more detail.

5.1. Exchange-enhanced paramagnets

The first example is the exchange-enhanced paramagnet just on the verge of the appearance of ferromagnetism. In this case, the spin amplitude sum rule can be written by

$$2A(y,t) + A(y_z,t) - c_z(2y + y_z) + 5c_z y_{10}\sigma^2 = -3c_z y_{00}$$
$$y_{00} = -\tfrac{20}{3}y_{10}\Delta\langle\mathbf{S}_i^2\rangle_{\text{tot}} \tag{5.5}$$

where $y_{00} = y_0(0)$. The temperature dependence of the inverse of the magnetic susceptibility, $y_0(t) = y(0,t)$, is given by solving

$$c_z y_0(t) - A(y_0(t),t) = c_z y_{00}. \tag{5.6}$$

In the weak field limit, the field-induced change of its transverse and longitudinal components are well represented by

$$\delta y(\sigma,t) = y(\sigma,t) - y_0(t) = y_1(t)\sigma^2 + \cdots$$
$$\delta y_z(\sigma,t) = y_z(\sigma,t) - y_0(t) = 3y_1(t)\sigma^2 + \cdots. \tag{5.7}$$

The above σ^2-linear dependence corresponds to the linearity of the Arrott plot (M^2 versus H/M plot) of the magnetic isotherm. The coefficient $y_1(t)$ stands for the fourth-order expansion coefficient of the free energy in powers of σ. On substitution of (5.7) into (5.5), it is explicitly given by

$$y_1(t) = \frac{c_z y_{10}}{c_z - A'(y_0,t)} \tag{5.8}$$

from the comparison of the σ^2-linear coefficients of (5.5).

We can easily confirm the thermodynamic Maxwell relation as follows. At first the partial t-derivative of $y(\sigma, t)$ for small σ is given by

$$\frac{\partial y(\sigma, t)}{\partial t} = \frac{\mathrm{d}y_0(t)}{\mathrm{d}t} = \frac{1}{c_z - A'(y_0, t)} \frac{\partial A(y_0, t)}{\partial t} = \frac{y_1(t)}{c_z y_{10}} \frac{\partial A(y_0, t)}{\partial t}. \tag{5.9}$$

On the other hand, from (3.1) the field-induced change of the magnetic entropy is given by

$$S_{\mathrm{m}}(\sigma, t) = S_{\mathrm{m}}(0, t) + \delta S_{\mathrm{m}}(\sigma, t) \tag{5.10}$$

$$\begin{aligned}\frac{\delta S_{\mathrm{m}}(\sigma, t)}{N_0} &= 3 \int_0^1 \mathrm{d}x\, x^2 u[1/u + 1/2u^2 - \psi'(u)] \frac{x}{t} (2\delta y + \delta y_z) \\ &= -3 \frac{\partial A(y_0, t)}{\partial t} [2\delta y(\sigma, t) + \delta y_z(\sigma, t)].\end{aligned} \tag{5.11}$$

The higher-order correction terms are neglected. The σ-derivative of the entropy is, therefore, given by

$$\frac{\partial}{\partial \sigma} \left(\frac{S_{\mathrm{m}}}{N_0} \right) = -30 \frac{\partial A(y_0, t)}{\partial t} y_1(t) \sigma. \tag{5.12}$$

Comparison of the above two expressions, (5.9) and (5.12), now verifies the Maxwell relation,

$$\begin{aligned}\frac{T_A \sigma}{2T_0} \frac{\partial y(\sigma, t)}{\partial t} &= \frac{T_A \sigma}{2T_0} \frac{y_1(t)}{c_z y_{10}} \frac{\partial A(y_0, t)}{\partial t} = 30 y_1(t) \sigma \frac{\partial A(y_0, t)}{\partial t} \\ &= -\frac{\partial}{\partial \sigma} \left(\frac{S_{\mathrm{m}}}{N_0} \right)\end{aligned} \tag{5.13}$$

where we used the relation $y_{10} = T_A/60 c_z T_0$, introduced in (2.15).

The above explanation is the finite temperature extension of the field dependence of the specific heat in paramagnon theories. It should be noted that its validity is not confined to the very low temperature region. As an example, the low temperature limit of the field dependence can be obtained as follows. In this limit, $y(\sigma, t)$ and $S_{\mathrm{m}}(\sigma, t)$ are given by

$$y_0(t) = y_{00} + \frac{1}{24 c_z y_{00}} t^2 + \cdots$$

$$\begin{aligned}S_{\mathrm{m}}(\sigma, t) &= \frac{1}{2} N_0 t \left[\ln\left(1 + \frac{1}{y(\sigma, 0)} \right) + \frac{1}{2} \ln\left(1 + \frac{1}{y_z(\sigma, 0)} \right) \right] + \cdots \\ &\simeq N_0 t \left[\frac{3}{4} \ln\left(\frac{1}{y_{00}} \right) - \frac{5}{4} \frac{y_{10}}{y_{00}} \sigma^2 \right] + \cdots.\end{aligned} \tag{5.14}$$

From these results we obtain the following external field suppression of the T-linear specific heat coefficient $\gamma_m(\sigma)$:

$$\begin{aligned}\gamma_m(\sigma) &= \lim_{t \to 0} \frac{C_{\mathrm{m}}(\sigma, t)}{T} = \frac{1}{T_0} \lim_{t \to 0} \frac{S_{\mathrm{m}}(\sigma, t)}{t} \\ &= N_0 \left[\ln\left(1 + \frac{1}{y(\sigma, 0)} \right) + \frac{1}{2} \ln\left(1 + \frac{1}{y_z(\sigma, 0)} \right) \right] \\ &= \frac{3N_0}{4T_0} \left[\ln \frac{1}{y_{00}} - \frac{5 y_{10}}{3 y_{00}} \sigma^2 \right] + \cdots.\end{aligned} \tag{5.15}$$

The relative suppression of the specific heat in the $t = 0$ limit is also expressed in the form

$$
\begin{aligned}
-\frac{\Delta C_{\mathrm{m}}(\sigma,0)}{C_{\mathrm{m}}(0,0)} &= -\frac{C_{\mathrm{m}}(0,0) - C_{\mathrm{m}}(\sigma,0)}{C_{\mathrm{m}}(0,0)} \\
&\simeq \frac{2\ln(1+y_1\sigma^2/y_0) + \ln(1+3y_1\sigma^2/y_0)}{3\ln(1/y_0)} \\
&= \frac{5y_{10}}{3y_0\ln(1/y_0)}\sigma^2 \simeq \frac{5y_{10}}{3y_0^3\ln(1/y_0)}\left(\frac{h}{T_A}\right)^2 \\
&= \frac{5}{3}\frac{(\chi_0/N_0)^3\bar{F}_{10}}{\ln(2T_A\chi_0/N_0)}h^2, \qquad (\sigma = h/T_A y_0)
\end{aligned}
\tag{5.16}
$$

where χ_0 and $\bar{F}_1$ are the magnetic susceptibility and the fourth expansion coefficient of the free energy expansion in the magnetization M in the ground state, respectively. The above result of the h^2-linear suppression of ΔC_{m} corresponds to the formula

$$
\frac{\Delta C_{\mathrm{m}}(\sigma,0)}{C_{\mathrm{m}}(0,0)} = -0.1\frac{S}{\ln S}\left(\frac{H}{T_{\mathrm{sf}}}\right)^2, \qquad (S = (1-\alpha)^{-1},\ 1/T_{\mathrm{sf}} \propto S\chi^0_{\mathrm{pauli}})
$$

derived by Béal-Monod *et al* (1968) for the electron gas model.

5.2. Field dependence in the paramagnetic phase ($T > T_c$)

Let us next discuss the temperature and magnetic field dependence in the paramagnetic phase. The preliminary results were reported by Takahashi *et al* (2004). The σ-dependence of the inverse magnetic susceptibility $y(\sigma,t)$ and the entropy $S_{\mathrm{m}}(\sigma,t)$ assumes the same forms as (5.9) and (5.11) in the preceding subsection. Therefore the Maxwell relation can be confirmed as well. The reduced inverse magnetic susceptibility $y_0(t)$ is, on the other hand, determined by solving

$$
A(y_0,t) - c_z y_0 = A(0,t_{\mathrm{c}}). \tag{5.17}
$$

The field-induced suppression of the entropy has the same expression as (5.11). The temperature dependence of the field-induced specific heat is simply obtained by the t-derivative of (5.11) under the constant h condition as follows:

$$
\delta C_{\mathrm{m}} = t\,\frac{\partial \delta S_{\mathrm{m}}}{\partial t} = \delta C_{\mathrm{m0}} + \delta C_{\mathrm{m1}} \tag{5.18}
$$

$$
\begin{aligned}
\frac{\delta C_{\mathrm{m0}}}{N_0 t} &= -3\,\frac{\mathrm{d}}{\mathrm{d}t}\,\frac{\partial A(y_0,t)}{\partial t}\bigg|_{y_0}(2\delta y + \delta y_z) \\
\frac{\delta C_{\mathrm{m1}}}{N_0 t} &= -3\,\frac{\partial A(y_0,t)}{\partial t}\left(2\,\frac{\partial y}{\partial t}\bigg|_h + \frac{\partial y_z}{\partial t}\bigg|_h - 3\,\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\right) \\
\delta y(\sigma,t) &= y(\sigma,t) - y_0(t), \qquad \delta y_z(\sigma,t) = y_z(\sigma,t) - y_0(t).
\end{aligned}
\tag{5.19}
$$

The above expression can be shown in a slightly different form. Note that the t-dependence of the thermal amplitude $A(y_0,t)$ is given by

$$
\frac{\partial A(y_0,t)}{\partial t} = -[A'(y_0,t) - c_z]\,\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}.
$$

On differentiation of the above result again, we obtain

$$
\frac{\mathrm{d}}{\mathrm{d}t}\,\frac{\partial A(y_0,t)}{\partial t} = -\left[A''(y_0,t)\,\frac{\mathrm{d}y_0(t)}{\mathrm{d}t} + \frac{\partial A'(y_0,t)}{\partial t}\right]\frac{\mathrm{d}y_0(t)}{\mathrm{d}t} - [A'(y_0,t) - c_z]\,\frac{\mathrm{d}^2 y_0(t)}{\mathrm{d}t^2}.
$$

On the other hand, the t-derivative of $y_1(t)$ defined in (5.8) gives

$$\left[\frac{\partial^2 A(y_0,t)}{\partial y_0^2}y_0'(t)+\frac{\partial^2 A(y_0,t)}{\partial y_0\,\partial t}\right]y_1(t)+[A'(y_0,t)-c_z]y_1'(t)=0.$$

From these results the second-order derivative of the thermal amplitude can be expressed in the form

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial A(y_0,t)}{\partial t}=\frac{c_z y_{10}}{y_1(t)}\frac{\mathrm{d}^2 y_0(t)}{\mathrm{d}t^2}-\frac{c_z y_{10}}{y_1^2(t)}\frac{\mathrm{d}y_1(t)}{\mathrm{d}t}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}.$$

In conclusion, the field dependence of the specific heat can be given in the following equivalent form

$$\begin{aligned}\frac{\delta C_{\mathrm{m}}(\sigma,t)}{N_0 t}&=-\frac{3c_z y_{10}}{y_1(t)}\left[\frac{\mathrm{d}y_0^2(t)}{\mathrm{d}t^2}-\frac{1}{y_1(t)}\frac{\mathrm{d}y_1(t)}{\mathrm{d}t}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\right](2\delta y+\delta y_z)\\&\quad-\frac{3c_z y_{10}}{y_1(t)}\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\left(2\left.\frac{\partial y}{\partial t}\right|_h+\left.\frac{\partial y_z}{\partial t}\right|_h-3\frac{\mathrm{d}y_0(t)}{\mathrm{d}t}\right)\end{aligned}\tag{5.20}$$

in terms of the t-derivatives of $y_0(t)$ and $y_1(t)$.

Based on the above expressions, the field dependence of the specific heat is discussed below in more detail, focusing on the region at higher temperature and the critical region above t_c.

5.2.1. Field dependence at high temperature. Except in the critical region, the σ^2-dependence of y and y_z is given by (5.7). At higher temperature where this dependence is well justified, the t-dependence of the field-induced specific heat change is therefore given by

$$\frac{\delta C_{\mathrm{m}}}{N_0 t}=-15c_z y_{10}\sigma^2\left[\frac{\partial^2 y_0(t)}{\partial t^2}-\frac{2}{y_0}\left(\frac{\partial y_0}{\partial t}\right)^2\right]=\frac{15c_z y_{10}}{T_A^2}\frac{\partial^2 y_0^{-1}(t)}{\partial t^2}h^2.\tag{5.21}$$

This gives the positive enhancement, and the magnitude of δC_{m} rapidly decreases according to the $t/(t-t_c)^3$-dependence with increasing the temperature, if $y_0(t)$ obeys the Curie–Weiss law behaviour, $y_0(t)\propto(t-t_c)$. The field-induced entropy change $\delta S_{\mathrm{m}}(\sigma,t)$ is always negative. Because its temperature dependence has a positive slope, it gives rise to the positive enhancement of δC_{m} at high temperature. From the above expression, it seems that the h^2-coefficient would diverge at $t=t_c$. This comes from our σ^2-linear approximation for y and y_z. Around the critical region, the effect of the critical magnetic isotherm has to be included for the proper treatment as shown below.

5.2.2. Field dependence around the critical temperature. In the critical limit, $t\to t_c$, we have already mentioned that $y(\sigma,t)$ and $y_z(\sigma,t)$ obey the critical σ^4-linear behaviour given in (2.19). According to (5.11), the field-induced change of the entropy is, therefore, given by

$$\frac{\delta S_{\mathrm{m}}}{N_0 t}=-\frac{4}{t_c}A(0,t_c)[2\delta y(\sigma,t_c)+\delta y_z(\sigma,t_c)]\simeq-\frac{28}{t_c}A(0,t_c)y_c\sigma^4.\tag{5.22}$$

With the use of (5.18) and (5.19), the critical limit of the specific heat is evaluated as follows. At first the t-derivative coefficients of the thermal amplitude $A(0,t)$ in (5.19) are estimated as follows:

$$\frac{\partial A(0,t)}{\partial t}\simeq\frac{4}{3t}A(0,t),\qquad\frac{\partial^2 A(0,t)}{\partial t^2}\simeq\frac{4}{9t^2}A(0,t),$$

$$\frac{\partial^2 A(y_0,t)}{\partial t\,\partial y_0}\frac{\partial y_0}{\partial t}\simeq-\frac{\pi}{8\sqrt{y_0}}\frac{\partial y_0}{\partial t}=-\frac{\pi}{4}\frac{\partial\sqrt{y_0}}{\partial t}=-\frac{4}{3t^2}A(0,t)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial A(y_0,t)}{\partial t}=\frac{\partial^2 A(y_0,t)}{\partial t^2}+\frac{\partial^2 A(y_0,t)}{\partial t\,\partial y_0}\frac{\partial y_0}{\partial t}\simeq-\frac{8}{9t^2}A(0,t)$$

where we have used the relation $\sqrt{y_0(t)}\simeq 4[A(0,t)-A(0,t_\mathrm{c})]/\pi t$, justified around $t\simeq t_\mathrm{c}$. Because the first term $C_{\mathrm{m}0}/t$ in (5.18) is proportional to σ^4, it is negligible in the weak field limit. In the presence of the applied field, the t-derivatives of y and y_z in (5.19) are evaluated from the following t-derivative of the sum rule (5.2):

$$-2\frac{\pi t}{8\sqrt{y}}\left.\frac{\partial y}{\partial t}\right|_h-\frac{\pi t}{8\sqrt{y_z}}\left.\frac{\partial y_z}{\partial t}\right|_h+10c_z y_{10}\sigma\frac{\partial\sigma}{\partial t}+3\frac{\partial A(0,t)}{\partial t}\simeq 0 \tag{5.23}$$

where we have used the critical y-dependence of the thermal spin fluctuation amplitude, $\partial A(y,t)/\partial y\simeq-\pi t/8\sqrt{y}$. The above equation (5.23) implies the σ-dependence, $\partial y/\partial t\propto\sigma^2$, in the $\sigma\to 0$ limit, if we take into account the critical magnetization process, $y\simeq y_\mathrm{c}\sigma^4$ and $y_z\simeq 5y_\mathrm{c}\sigma^4$.

We can now derive the relation between the t-derivatives of y_z and y, the first two terms of (5.23), by using the following relations:

$$\begin{aligned}\left.\frac{\partial y_z}{\partial t}\right|_h&=\left.\frac{\partial y_z}{\partial t}\right|_\sigma+\frac{\partial y_z}{\partial\sigma}\frac{\partial\sigma}{\partial t}=\left.\frac{\partial y_z}{\partial t}\right|_\sigma-\frac{\partial y_z}{\partial\sigma}\frac{\sigma}{y}\left.\frac{\partial y}{\partial t}\right|_h\\&=\left.\frac{\partial y_z}{\partial t}\right|_\sigma-20\left.\frac{\partial y}{\partial t}\right|_h\end{aligned} \tag{5.24}$$

$$\left.\frac{\partial y}{\partial t}\right|_h=\left.\frac{\partial y}{\partial t}\right|_\sigma+\frac{\partial y}{\partial\sigma}\frac{\partial\sigma}{\partial t}=\left.\frac{\partial y}{\partial t}\right|_\sigma-\frac{\sigma}{y}\frac{\partial y}{\partial\sigma}\left.\frac{\partial y}{\partial t}\right|_h=\left.\frac{\partial y}{\partial t}\right|_\sigma-4\left.\frac{\partial y}{\partial t}\right|_h \tag{5.25}$$

where we have taken into account (5.3) and the critical σ^4 behaviours of y and y_z. Because of the σ^2-linear dependence of $\partial y/\partial t|_\sigma$, $\partial y_z/\partial t|_\sigma$ in (5.24) is given by

$$\left.\frac{\partial y_z}{\partial t}\right|_\sigma=\left.\frac{\partial y}{\partial t}\right|_\sigma+\sigma\frac{\partial}{\partial\sigma}\left.\frac{\partial y}{\partial t}\right|_\sigma=3\left.\frac{\partial y}{\partial t}\right|_\sigma. \tag{5.26}$$

The constant-h derivative, $\partial y_z/\partial t|_h$, in (5.23) is therefore given as follows:

$$\left.\frac{\partial y_z}{\partial t}\right|_h=3\left.\frac{\partial y}{\partial t}\right|_\sigma-20\left.\frac{\partial y}{\partial t}\right|_h=-5\left.\frac{\partial y}{\partial t}\right|_h. \tag{5.27}$$

On the other hand, from the relation between the t-derivatives of σ and y in (5.3), we can find that the third term is also proportional to the first term as shown below:

$$\begin{aligned}10c_z y_{10}\sigma\frac{\partial\sigma}{\partial t}&=-10c_z y_{10}\frac{\sigma^2}{y}\left.\frac{\partial y}{\partial t}\right|_h=-10c_z y_{10}\frac{\sqrt{y/y_\mathrm{c}}}{y}\left.\frac{\partial y}{\partial t}\right|_h\\&=-\frac{\pi t_\mathrm{c}}{2\sqrt{y}}(2+\sqrt{5})\left.\frac{\partial y}{\partial t}\right|_h\end{aligned} \tag{5.28}$$

where y_c is defined in (2.19). On substitution of (5.27) and (5.28) into (5.23), the following result is derived:

$$\left.\frac{\partial y}{\partial t}\right|_h\simeq\frac{24\sqrt{y_\mathrm{c}}}{(10+3\sqrt{5})\pi t_\mathrm{c}}\frac{\partial A(0,t_\mathrm{c})}{\partial t_\mathrm{c}}\sigma^2. \tag{5.29}$$

We are thus finally led to the following critical limit of the magnetic specific heat in the paramagnetic phase:

$$\frac{\delta C_{\mathrm{m}0}}{N_0 t}=-3\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial A(0,t_\mathrm{c})}{\partial t_\mathrm{c}}(2y+y_z)=\frac{56}{3t^2}A(0,t)y_\mathrm{c}\sigma^4$$

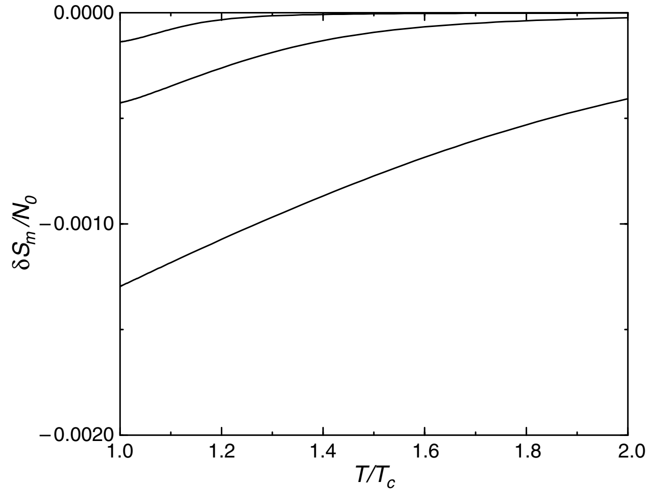

Figure 5. Temperature dependence of the entropy change $\delta S_{\rm m}/N_0$ above $T_{\rm c}$ under applied magnetic fields $h = 0.2 \times 10^{-5}$, 1.0×10^{-5}, and 5.0×10^{-5} from the top.

$$\begin{aligned}
\frac{\delta C_{\rm m1}}{N_0 t} &= -3\,\frac{\partial A(0,t_{\rm c})}{\partial t_{\rm c}}\left(2\,\frac{\partial y}{\partial t}\bigg|_h + \frac{\partial y_z}{\partial t}\bigg|_h\right)\\
&= \frac{216\sqrt{y_{\rm c}}}{(10+3\sqrt{5})\pi t_{\rm c}}\left(\frac{\partial A(0,t_{\rm c})}{\partial t_{\rm c}}\right)^2\sigma^2\\
\frac{\delta C_{\rm m}}{N_0 t} &= \frac{8A^3(0,t)}{t_{\rm c}^4}\left[\frac{20}{\pi(2+\sqrt{5})}\right]^2\left[\frac{7}{3}\left(\frac{\sigma}{\sigma_{\rm s}}\right)^4 + \frac{12(2+\sqrt{5})}{5(10+3\sqrt{5})}\left(\frac{\sigma}{\sigma_{\rm s}}\right)^2\right].
\end{aligned}$$

We show in figures 5 and 6 numerical results of the temperature dependence of the entropy and the specific heat, respectively, in the presence of an applied magnetic field. The field-induced entropy is always negative. It monotonically increases with temperature. As the slope of its t-dependence, the specific heat always becomes positive above $T_{\rm c}$ including its critical limit. The thin full and dashed curves in figure 6 represent $\delta C_{\rm m0}/N_0 t$ and $\delta C_{\rm m1}/N_0 t$, respectively. The component $\delta C_{\rm m0}/N_0 t$ is dominant only near the critical temperature $T_{\rm c}$. On the other hand, $\delta C_{\rm m1}/N_0 t$ shows a broad maximum slightly above $T_{\rm c}$. It then rapidly decreases proportional to t^{-2} with increasing the temperature. Because $\delta C_{\rm m0}/N_0 t$ is negligible under a weak applied magnetic field, the total change, $\delta C_{\rm m}/N_0 t$, shows a similar temperature dependence with $\delta C_{\rm m1}/N_0 t$. However, in the presence of a higher magnetic field it becomes monotonically decreasing owing to the rapid growth of $\delta C_{\rm m1}/N_0 t$ around $T_{\rm c}$. In figure 7, the temperature dependence of the specific heat change is shown for various values of $t_{\rm c}$ under the same applied magnetic field $h = 1.0 \times 10^{-5}$.

The external field effect on the magnetic specific heat of Sc_3In was measured by Takeuchi and Masuda (1979). Their observed $\delta C_{\rm m}/T$ shows a broad peak above the critical temperature. The peak value is around 2 mJ K^{-2} g-atom for $H = 10$ T, if we assume that all the atoms have the same magnetic moment. This corresponds to $T_0(\delta C_{\rm m}/N_0 T)_{\rm max} \simeq 0.16$ estimated by assuming that only Sc atoms have moments and $T_0 = 500$ K. On the other hand, our numerical result gives a peak value of about 0.1 for the same values of T_0 and T_A, in fairly good quantitative agreement with experiments. The applied field of 1 T corresponds to $h = 1.3 \times 10^{-4}$ for $T_A = 10^4$ K. Their numerical study based on the SCR theory showed the monotonous increase

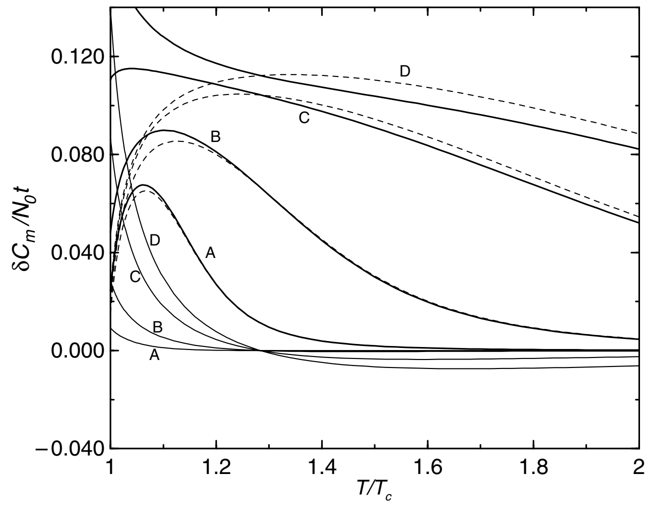

Figure 6. Temperature dependence of the specific heat change $\delta C_{\rm m}/N_0 t$ above $T_{\rm c}$ under applied magnetic fields. (A): $h = 0.2 \times 10^{-5}$, (B): $h = 1.0 \times 10^{-5}$, (C): $h = 5.0 \times 10^{-5}$, and (D): $h = 1.0 \times 10^{-4}$. The thin full and dashed curves represent $\delta C_{\rm m0}/N_0 t$ and $\delta C_{\rm m1}/N_0 t$, respectively.

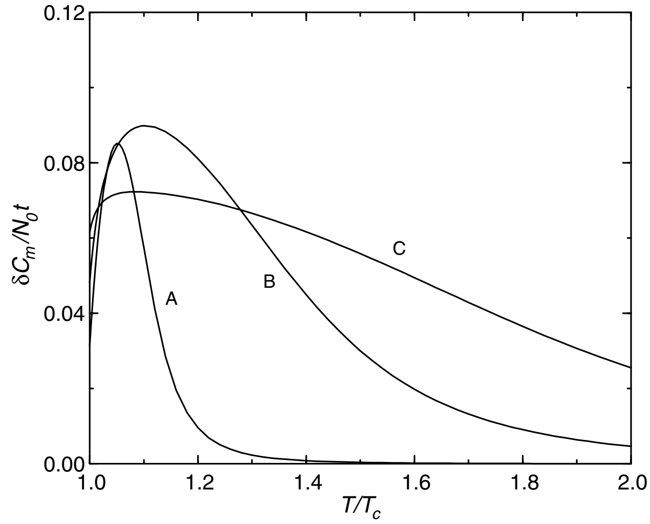

Figure 7. Temperature dependence of the specific heat change $\delta C_{\rm m}/N_0 t$ above $T_{\rm c}$ for $t_{\rm c} = 0.05$ (A), 0.01 (B), and 0.005 (C) under the applied magnetic field $h = 1.0 \times 10^{-5}$.

of $\delta C_{\rm m}/T$ in their calculated range of the temperature. They also predicted the negative value of the critical limit above $t_{\rm c}$ that seems to arise from the neglect of the critical behaviour of the magnetic isotherm.

5.3. Field dependence in the ordered phase

In the ordered phase the field dependence of the entropy and the specific heat can be treated in the same manner as in the paramagnetic phase except that an explicit account of $\lambda(\sigma, t)$ is necessary. From (3.1) and (3.3), the field-induced change of the entropy is given by

$$\begin{aligned} \frac{\delta S_{\rm m}(\sigma,t)}{N_0} &= -3\left[2\frac{\partial A_t(0,t)}{\partial t}y(\sigma,t)+\frac{\partial A(y_{z0},t)}{\partial t}\delta y_z(\sigma,t)\right]+\frac{\delta\Delta S_{\rm m}(\sigma,t)}{N_0} \\ \frac{\delta\Delta S_{\rm m}(\sigma,t)}{N_0} &= 60c_z y_{10}\left[\frac{{\rm d}\lambda_0(t)}{{\rm d}t}(\delta y_z-y)-\frac{{\rm d}y_{z0}(t)}{{\rm d}t}\delta\lambda(\sigma,t)\right]. \end{aligned} \tag{5.30}$$

The above expression can be written in a simplified form as shown below. Note the following field-induced change of the amplitude sum rule (5.1):

$$2[A_t'(0,t)-c_z]y(\sigma,t)+[A'(y_{z0},t)-c_z]\delta y_z(\sigma,t)+5c_z y_{10}(\sigma^2-\sigma_0^2)=0.$$

Using the result, the σ-variation of $\lambda(\sigma,t)$ defined in (2.10) is given by

$$\begin{aligned} 60c_z y_{10}\delta\lambda(\sigma,t) &= 2\big\{[A'(y_{z0},t)-c_z]\delta y_z-[A_t'(0,t)-c_z]y(\sigma,t)\big\}-5c_z y_{10}(\sigma^2-\sigma_0^2) \\ &= 3[A'(y_{z0},t)-c_z]\delta y_z. \end{aligned} \tag{5.31}$$

On substitution of (4.1) and (5.31) in (5.30), the field-induced variation of the entropy correction is given as follows:

$$\begin{aligned} \frac{\delta\Delta S_{\rm m}(\sigma,t)}{N_0} &= -3\delta y_z\left\{2\frac{\partial A_t(0,t)}{\partial t}+[A'(y_{z0},t)-c_z]\frac{\partial y_{z0}}{\partial t}+5c_z y_{10}\frac{{\rm d}\sigma_0^2}{{\rm d}t}\right\} \\ &\quad +3y\left[2\frac{\partial A_t(0,t)}{\partial t}+5c_z y_{10}\frac{{\rm d}\sigma_0^2}{{\rm d}t}\right]. \end{aligned} \tag{5.32}$$

If we substitute the above expression in (5.30), $\delta S_{\rm m}$ is finally given by the following simple expression:

$$\frac{\delta S_{\rm m}(\sigma,t)}{N_0} = 15A(0,t_{\rm c})y(\sigma,t)\frac{{\rm d}u(t)}{{\rm d}t}, \qquad u(t)=\sigma_0^2(t)/\sigma_{\rm s}^2. \tag{5.33}$$

In the above derivation the δy_z-linear term vanishes identically because of the sum rule (4.2). The entropy change is proportional to the inverse of the transverse magnetic susceptibility y and its coefficient is determined by the slope of the t-dependence of the spontaneous magnetic moment $\sigma_0^2(t)$.

In spite of its simplicity, it is quite easy to confirm that (5.33) satisfies the thermodynamic Maxwell relation. Note first that the partial t-derivative of the magnetic isotherm,

$$y(\sigma,t)=y_1(t)[\sigma^2-\sigma_0^2(t)], \tag{5.34}$$

gives

$$\frac{\partial y(\sigma,t)}{\partial t}=y_1'(t)[\sigma^2-\sigma_0^2(t)]-y_1(t)\frac{{\rm d}\sigma_0^2(t)}{{\rm d}t}\simeq -y_1(t)\sigma_{\rm s}^2\frac{{\rm d}u(t)}{{\rm d}t}. \tag{5.35}$$

On the other hand, from (5.33) the partial σ-derivative of the entropy is given by

$$\frac{1}{N_0}\frac{\partial S_{\rm m}}{\partial\sigma}=30A(0,t_{\rm c})y_1(t)\sigma\frac{{\rm d}u(t)}{{\rm d}t}=30c_z y_{10}\sigma_{\rm s}^2 y_1(t)\sigma\frac{{\rm d}u(t)}{{\rm d}t}.$$

Comparison of the above two expressions verifies the Maxwell relation (5.13).

It is now apparent that the presence of the free energy correction and the use of its stability condition has a key significance in our derivation of the thermodynamically consistent and reasonable result on the field dependence of the entropy. This also justifies our discussion on the temperature dependence of the specific heat in section 4, since the second-order t-derivative terms there result from the same entropy correction $\Delta S_{\rm m}(\sigma,t)$. The presence of the term proportional to the first-order t-derivative of $\sigma_0^2(t)$ in (5.32) also provides the second-order t-derivative term in the field-induced change of the specific heat as shown below.

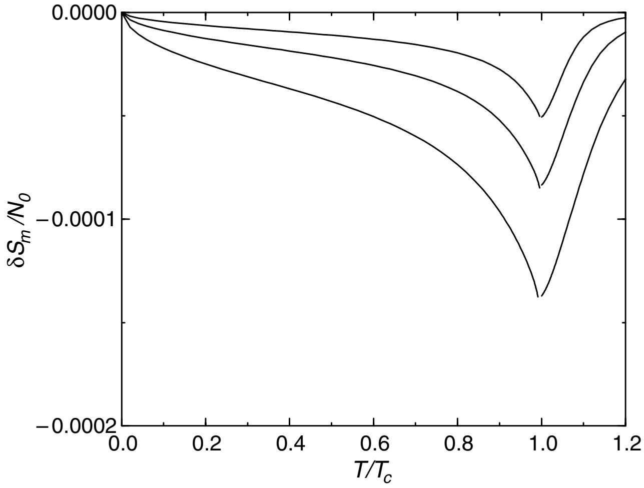

Figure 8. Temperature dependence of the entropy change $\delta S_{\mathrm{m}}/N_0$ for $t_{\mathrm{c}} = 0.01$ under a weak applied magnetic field, $h = 0.05 \times 10^{-5}$, 0.1×10^{-5}, and 0.2×10^{-5}, from the top.

The field-induced specific heat change is simply derived from the t-derivative of the entropy change (5.33) under a fixed external magnetic field. It is given as the sum of two contributions

$$\delta C_{\mathrm{m}}(\sigma, t) = \delta C_{\mathrm{m1}}(\sigma, t) + \delta C_{\mathrm{m2}}(\sigma, t) \tag{5.36}$$

$$\frac{\delta C_{\mathrm{m1}}(\sigma, t)}{N_0 t} = 15A(0, t_{\mathrm{c}})y(\sigma, t)\,\frac{\mathrm{d}^2 u(t)}{\mathrm{d}t^2} \tag{5.37}$$

$$\frac{\delta C_{\mathrm{m2}}(\sigma, t)}{N_0 t} = 15A(0, t_{\mathrm{c}})\,\frac{\mathrm{d}u(t)}{\mathrm{d}t}\,\left.\frac{\partial y(\sigma, t)}{\partial t}\right|_h \tag{5.38}$$

that come from the first-order and the second-order t-derivatives of the spontaneous moment $\sigma_0^2(t)$, respectively. In the low-field limit where (5.34) and (5.35) are justified, (5.37) and (5.38) are also written by

$$\begin{aligned}
\frac{\delta C_{\mathrm{m1}}(\sigma, t)}{N_0 t} &= 15A(0, t_{\mathrm{c}})y_1(t)\,\frac{\mathrm{d}^2 u(t)}{\mathrm{d}t^2}[\sigma^2 - \sigma_0^2(t)] \\
\frac{\delta C_{\mathrm{m2}}(\sigma, t)}{N_0 t} &= 15A(0, t_{\mathrm{c}})\,\frac{\mathrm{d}u(t)}{\mathrm{d}t}\,\frac{y}{y_z}\,\left.\frac{\partial y(\sigma, t)}{\partial t}\right|_\sigma \\
&= -\frac{15}{2}A(0, t_{\mathrm{c}})y_1(t)\sigma_{\mathrm{s}}^2\left(\frac{\mathrm{d}u(t)}{\mathrm{d}t}\right)^2[\sigma^2/\sigma_0^2(t) - 1].
\end{aligned} \tag{5.39}$$

If we notice the low-field limit of the magnetic isotherm,

$$y = y_{10}(\sigma^2 - \sigma_{\mathrm{s}}^2) = \frac{h}{T_A\sigma} \simeq \frac{h}{T_A\sigma_{\mathrm{s}}}$$

it follows that the field-induced change δC_{m} is generally suppressed and is proportional to the applied field h below t_{c}. The h-linear suppression of δC_{m} was actually observed on Sc_3In by Ikeda and Gschneidner (1983).

For the numerical analysis, we need the first-order and the second-order t-derivatives of $u(t)$, the inverse transverse magnetic susceptibility $y(\sigma, t)$ under the presence of the external field, and its t-derivative. We show in figures 8 and 9 the temperature dependence of the

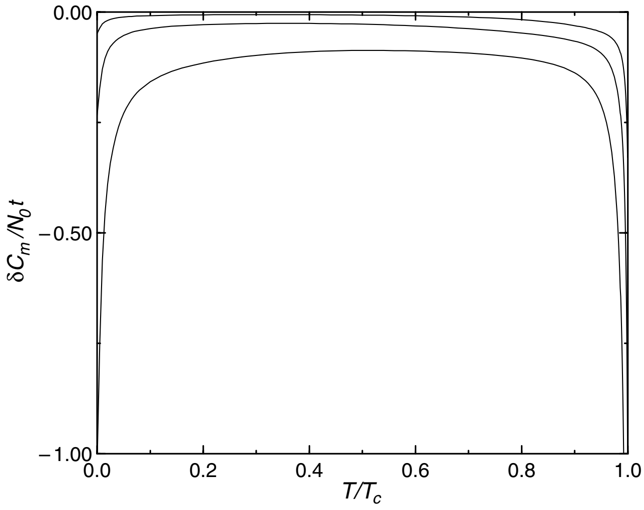

Figure 9. Field-induced change of $\delta C_{\mathrm{m}}/N_0 t$ for $t_{\mathrm{c}} = 0.01$ under applied magnetic fields $h = 0.2 \times 10^{-5}$, 1.0×10^{-5}, and 5.0×10^{-5}, from the top.

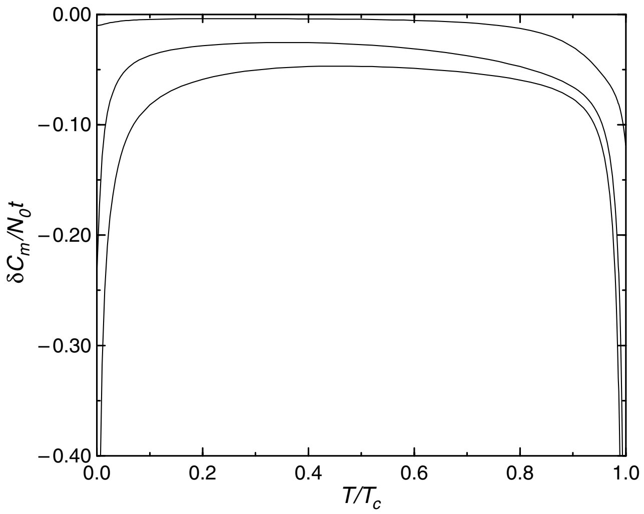

Figure 10. The t-dependence of $\delta C_{\mathrm{m}}/N_0 t$ for $t_{\mathrm{c}} = 0.005$, 0.01, and 0.05 under the applied magnetic field $h = 1.0 \times 10^{-5}$, from the bottom.

entropy and the specific heat, respectively, for various applied fields. The field-induced entropy decreases monotonically with increasing the temperature. Therefore the induced change of the specific heat $\delta C_{\mathrm{m}}/t$ is always negative below t_{c}, including the critical limit. It shows the discontinuous change at $t = t_{\mathrm{c}}$. In figure 10, the temperature dependence of the specific heat under an applied magnetic field is shown for cases with different t_{c}. In figure 11 $\delta C_{\mathrm{m}}/N_0 t$ is plotted against the magnitude of the external magnetic field h at temperature $t/t_{\mathrm{c}} = 0.1$, 0.5, and 0.9. It shows the good linearity at low temperature. At higher temperature, though the deviation from the linearity becomes significant in the weak field region, it shows the monotonic decrease with increasing h, in contrast with experiments on Sc_3In for $T/T_{\mathrm{c}} \sim 0.4$ (Ikeda and Gschneidner 1983), where clear saturation is observed.

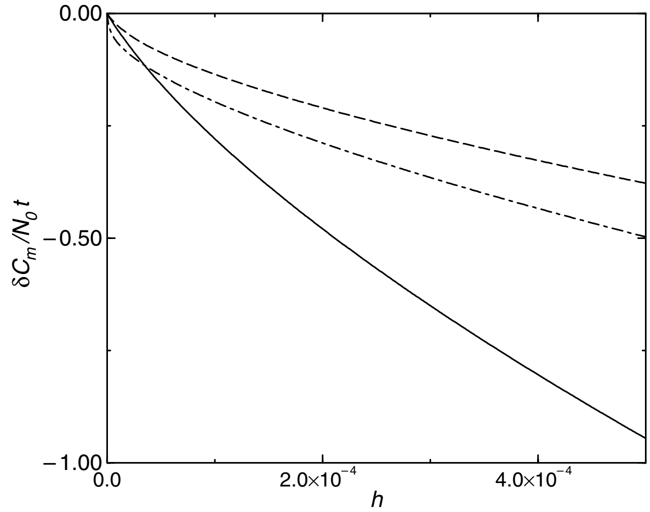

Figure 11. The specific heat suppression as a function of the applied field h for $T/T_c = 0.1$, 0.5, and 0.9 (full, broken, and chain curves, respectively) for $t_c = 0.01$.

In what follows we show the temperature and field dependence of the entropy and the specific heat, paying particular attention to the ranges at low temperature and around t_c.

5.3.1. In the low temperature limit. According to Takahashi (2001) the t-dependence of $y(\sigma, t)$ and $u(t)$ at low temperature is given by

$$\begin{aligned} y(\sigma, t) &= y_1(t)[\sigma^2 - \sigma_0^2(t)] \simeq y_{10}(\sigma^2 - \sigma_s^2) \\ y_z(\sigma, t) &= y_{z0} + 3y_1(t)[\sigma^2 - \sigma_0^2(t)] \simeq 2y_{10}\sigma_s^2 + 3y_{10}(\sigma^2 - \sigma_s^2) \\ u(t) &= 1 - \frac{a_T t^2}{(60c_z y_{10})^2\sigma_s^4} + \cdots, \qquad a_T = 10c_z(r^2 + 5r + 4) \end{aligned} \tag{5.40}$$

where the constant ratio $r = y_{z0}/x_c^2$ has already been introduced in (4.13). From (5.33) the field dependence of the entropy is given by

$$\begin{aligned} \frac{\delta S_m(\sigma, t)}{N_0} &= -15A(0, t_c)\left(\frac{\sigma^2}{\sigma_s^2} - 1\right)\frac{2a_T t}{(60c_z y_{10})^2\sigma_s^2} \\ &= -\frac{a_T t}{120c_z}\left(\frac{\sigma^2}{\sigma_s^2} - 1\right). \end{aligned} \tag{5.41}$$

Therefore the entropy is suppressed proportional to $(\sigma^2 - \sigma_s^2)$ by the applied magnetic field. Its slope is proportional to the temperature.

Because the second term C_{m2} in (5.36) is neglected in the low temperature limit, the field-induced suppression of the T-linear coefficient γ_m of the specific heat is given by

$$\begin{aligned} \frac{\delta\gamma_m(\sigma)}{N_0} &= \frac{\delta C_m}{N_0 T} = \frac{15A(0, t_c)}{T_0} y_{10}(\sigma^2 - \sigma_s^2)\frac{d^2 u(t)}{dt^2} \\ &= \frac{15A^2(0, t_c)}{c_z T_0}\left(\frac{\sigma^2}{\sigma_s^2} - 1\right) = \frac{\bar{F}_{10}}{32}(\sigma^2 - \sigma_s^2)\frac{d^2\sigma_0^2(t)}{dT^2} \end{aligned} \tag{5.42}$$

where $\bar{F}_{10} = 2T_A^2/15c_z T_0$ represents the fourth expansion coefficient of the free energy in powers of σ (Takahashi 2001). It is suppressed because of the negative sign of $d^2u(t)/dt^2$

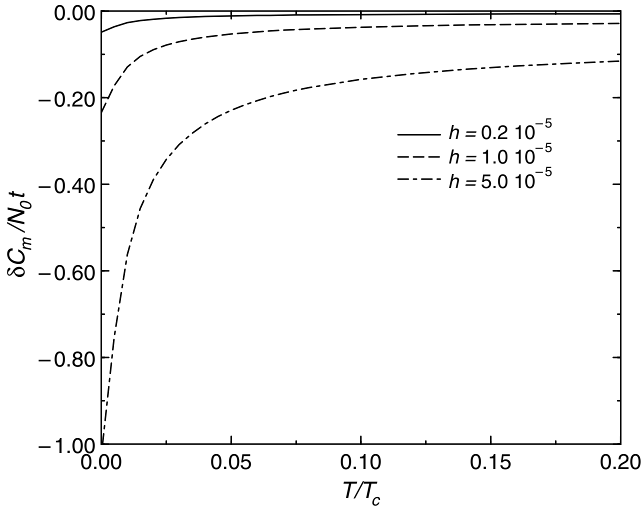

Figure 12. Field-induced change $\delta C_\mathrm{m}/N_0 t$ in the low temperature region for $t_\mathrm{c} = 0.01$ under the applied magnetic fields $h = 0.2 \times 10^{-5}$, 1.0×10^{-5}, and 5.0×10^{-5}.

in (5.40). As we have already shown above, the T-linear coefficient γ_m is suppressed proportional to h. Its negative initial slope is simply determined by the second-order T-derivative of the squared spontaneous moment $\sigma_0^2(t)$,

$$\frac{1}{N_0}\frac{\partial \gamma_m}{\partial h} = \frac{\bar{F}_{10}}{32 T_A y_{10} \sigma_\mathrm{s}} \frac{\partial \sigma_0^2(t)}{\partial T^2} = \frac{1}{4\sigma_\mathrm{s}} \frac{\mathrm{d}^2 \sigma_0^2(t)}{\mathrm{d}^2 T^2}$$
$$= -\frac{a_T}{2T_A^2 \sigma_\mathrm{s}^3}. \tag{5.43}$$

This is the result derived as the natural consequence of the Maxwell relation. In figure 12, the field-induced change of the specific heat is plotted against the reduced temperature T/T_c in the low temperature region. It is remarkable that its magnitude does not decrease monotonically with lowering the temperature. The reason comes from the steep decrease of the entropy there caused by the second-order t-derivative terms.

5.3.2. Field dependence around the critical temperature. In order to discuss the field-induced change of the entropy and the specific heat in the critical limit, we need the t-derivatives of the spontaneous magnetic moment $u(t)$. They are given as follows (see appendix A for the second derivative coefficient u_2):

$$u(t) = a_\mathrm{c}[1 - (t/t_\mathrm{c})^{4/3}], \qquad \frac{\mathrm{d}u(t)}{\mathrm{d}t} = -u_1 = -\frac{4a_\mathrm{c}}{3t_\mathrm{c}}$$
$$\frac{\mathrm{d}^2 u(t)}{\mathrm{d}t^2} \simeq -u_2, \qquad (u_2 > 0). \tag{5.44}$$

If we note the critical magnetization process, $y(\sigma, t_\mathrm{c}) = y_\mathrm{c}\sigma^4$, the following entropy change is derived from (5.33).

$$\frac{\delta S_\mathrm{m}}{N_0} = 15\left(-\frac{4a_\mathrm{c}}{3t_\mathrm{c}}\right) A(0, t_\mathrm{c}) y(\sigma, t_\mathrm{c}) = -\frac{28 A(0, t_\mathrm{c})}{t_\mathrm{c}} y_\mathrm{c}\sigma^4 \tag{5.45}$$

where $a_c = 7/5$ is used (see (4.10)). We can confirm that the entropy change δS_m is continuous at $t = t_c$, from the comparison of the above (5.45) with the paramagnetic result (5.22). This undertakes the validity of our choice of r and ξ in sections 4.1 and 4.2.

By substituting (5.44) into (5.36)–(5.38), the critical field dependence of the specific heat is given by

$$\left.\frac{\delta C_m(h,t)}{N_0 t}\right|_{t=t_c} = -15A(0,t_c)\left[u_2 y(\sigma,t_c) + u_1 \left.\frac{\partial y}{\partial t}\right|_h\right].$$

From the continuity, y and $\partial y/\partial t|_h$ are positive under the applied external field. The field-induced specific heat change is, therefore, negative in the critical limit below t_c. The discontinuous change takes place for $\delta C_m/N_0 t$ at $t = t_c$.

6. Discussion

In the present paper, we have succeeded in giving a comprehensive description of the magnetic entropy and the specific heat of itinerant electron magnets. Thermodynamically consistent formulae are derived with the use of the stability conditions of the free energy. As the result, the entropy satisfies the Maxwell relation of thermodynamics. We have found that these thermal properties are closely related to the global behaviour of the magnetic isotherm in the plane of the temperature and the external magnetic field axes. In this context, the present study owes very much to our previous study (Takahashi 2001).

Various new features predicted in this study are summarized as follows.

(i) In the absence of an applied magnetic field, the specific heat shows a slight but sharp critical peak at $t = t_c$.
(ii) A new additional enhancement of the linear specific heat coefficient is present in the low temperature limit.
(iii) The field-induced change of the entropy is always positive above t_c, while it is negative below t_c.
(iv) For a proper description of the entropy and the specific heat in the critical region, the critical behaviour of the magnetic isotherm has to be taken into account.
(v) At low temperature the field-induced change $\delta C_m/t$ decreases with lowering the temperature.
(vi) The field-induced suppression of the T-linear coefficient $\delta\gamma_m$ of the specific heat is determined by the slope of the T^2-dependence of the spontaneous magnetization at low temperature.

In so far as we are mainly concerned with paramagnetic properties, the thermodynamic consistency is not so severe. However, it becomes very crucial near the critical region. The inadequate inclusion of the term proportional to $\mathrm{d}^2 y(t)/\mathrm{d}t^2$ in the SCR theory gives rise to the spurious behaviour of the specific heat at the critical point, for instance. If there were terms proportional to $(\partial y/\partial t)^2$ and $\partial^2 y/\partial t^2$ in the specific heat, it would also be very difficult to verify the Maxwell relation.

It seems that our final results do not contain any explicit effect of the quantum zero-point spin fluctuations. Note however that we could derive the specific heat formula in consistence with the conserved total spin amplitude. As the stability condition, quantum zero-point spin fluctuations do play significant roles. Their neglect from the beginning is of course not justified logically. Ishigaki and Moriya (1998) have also treated the effect of zero-point spin fluctuations by simply including the effect in the conventional framework of the SCR theory. No treatment of the specific heat including its external field effect has yet been presented. The explicit form of the free energy proposed in this study will be helpful in our future studies on magnetic properties of itinerant electron magnets.

Acknowledgments

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Appendix A. Temperature dependence of $\sigma_0^2(t)$ and $y_{z0}(t)$

According to Takahashi (2001) the temperature dependence of $u(t) = \sigma_0^2(t)/\sigma_s^2$ and $v(t) = y_{z0}(t)/y_{z0}(0)$ is given by solving the following simultaneous equations:

$$\begin{aligned} u &= v\left[1 - \frac{3}{5c_z}A'(y_{z0}, t) - \frac{2}{5c_z}A'_c(0, t)\right] \\ u &= \frac{2v+3}{5} - \frac{1}{5A(0, t_c)}[A(y_{z0}, t) + 2A_t(0, t)] \end{aligned} \tag{A.1}$$

where $y_{z0}(t) = 2A(0, t_c)v(t)/c_z$ and $u(0) = v(0) = 1$. In the low temperature limit, thermal amplitudes show the following t^2-dependence:

$$A(y_z, t) \simeq \frac{t^2}{24y_{z0}(0)}, \qquad A'(y_z, t) \simeq -\frac{t^2}{24y_{z0}^2(0)}, \qquad (y_{z0}(0) = 2y_{10}\sigma_s^2)$$

$$A_c(0, t) \simeq \frac{rt^2}{24y_{z0}(0)}, \qquad A'_c(0, t) \simeq -\frac{r^2t^2}{24y_{z0}^2(0)}.$$

If we define $\delta u = u - 1$ and $\delta v = v - 1$, then the t^2-linear terms of (A.1) can be expressed in the form

$$\delta u = \delta v + \frac{t^2(3+2r^2)}{120y_{z0}^2(0)}, \qquad \delta u = \frac{2\delta v}{5} - \frac{t^2(1+2r)}{120A(0, t_c)y_{z0}(0)}.$$

As the solutions, the second-order derivatives of u and v at $t = 0$ are given by

$$\frac{\mathrm{d}^2u}{\mathrm{d}t^2} = -\frac{c_z(4+5r+r^2)}{180A^2(0, t_c)}, \qquad \frac{\mathrm{d}^2v}{\mathrm{d}t^2} = -\frac{c_z(5+4r+2r^2)}{144A^2(0, t_c)}.$$

Around $t = t_c$, solutions of (A.1) are obtained by assuming the following expansions:

$$u(t) = -u_1(t - t_c) - \frac{u_2}{2}(t - t_c)^2 + \cdots, \tag{A.2}$$

$$v(t) = \frac{v_2}{2}(t - t_c)^2 + \frac{v_3}{6}(t - t_c)^3 + \cdots \tag{A.3}$$

with positive coefficients, u_i and v_{i+1} ($i = 1, 2$). The coefficient u_1 has already been given in (5.44). From the critical behaviour of the thermal amplitude, the first line of (A.1) is well approximated by

$$\begin{aligned} u &\simeq v + \frac{v}{5c_z}\left[\frac{3\pi t}{8\sqrt{y_{z0}}} + \frac{2\xi\pi t}{8\sqrt{y_{z0}}}\right] = v + \frac{\pi t(3+2\xi)}{40c_z\sqrt{2A(0, t_c)/c_z}}\sqrt{v} \\ &= \frac{v_2}{2}(t - t_c)^2 + \frac{\pi t(3+2\xi)\sqrt{v_2}}{80\sqrt{c_zA(0, t_c)}}(t_c - t)\left[1 + \frac{v_3}{6v_2}(t - t_c) + \cdots\right]. \end{aligned} \tag{A.4}$$

The comparison of the $(t - t_c)$-linear coefficient of (A.4) with (A.2) leads to the following estimate:

$$u_1 = \frac{\pi t_c(3+2\xi)\sqrt{v_2}}{80\sqrt{c_zA(0, t_c)}}, \qquad \text{or} \qquad v_2 = \left(\frac{80u_1}{7\pi t_c}\right)^2 c_zA(0, t_c) = c_zA(0, t_c)\left(\frac{64}{3\pi t_c^2}\right)^2.$$

On substitution of the result for u_1 again in (A.4), we obtain

$$u = \frac{v_2}{2}(t - t_c)^2 + u_1 \frac{t}{t_c}(t_c - t)\left[1 + \frac{v_3}{6v_2}(t - t_c) + \cdots\right].$$

From the comparison of the above $(t - t_c)^2$ terms with (A.3), the following relation is derived:

$$v_2 - \left(\frac{2}{t_c} + \frac{v_3}{3v_2}\right)u_1 = -u_2. \tag{A.5}$$

On the other hand, the right-hand side of (A.2) can be expanded as follows:

$$\begin{aligned} u &\simeq \frac{2v}{5} + \frac{3}{5}[1 - A(0,t)/A(0,t_c)] + \frac{1}{5A(0,t_c)}\frac{\pi t}{4}\sqrt{y_{z0}} \\ &\simeq \frac{2}{5}\frac{v_2}{2}(t - t_c)^2 + \frac{3}{5}\left[1 - \left(\frac{t}{t_c}\right)^{4/3}\right] + \frac{1}{5A(0,t_c)}[A(y_{z0},t) - A(0,t)] \\ &= \frac{1}{5}v_2(t - t_c)^2 - \frac{3}{7}u_1(t - t_c) - \frac{3}{5}\frac{4}{9t_c^2}(t - t_c)^2 \\ &\quad - \frac{4t}{7t_c}u_1(t_c - t)\left[1 + \frac{v_3}{6v_2}(t - t_c) + \cdots\right]. \end{aligned}$$

The comparison of $(t - t_c)^2$-linear coefficients in this case gives

$$\frac{4u_1}{7t_c} - \frac{1}{2}u_2 = \frac{1}{5}v_2 - \frac{12}{45t_c^2} - \frac{2v_3}{21v_2}u_1. \tag{A.6}$$

By eliminating u_1v_3/v_2 terms from (A.5) and (A.6) we finally obtain for u_2 the result

$$u_2 = \frac{14}{35}v_2 + \frac{56}{45t_c^2}.$$

Appendix B. The Maxwell relation of the thermodynamics

In this appendix we show that the Maxwell relation of the thermodynamics is satisfied for our entropy (3.1). Against the change of the temperature and the magnetization the free energy change $\mathrm{d}F(M,T)$ can be given by

$$\mathrm{d}F(M,T) = -S(M,T)\,\mathrm{d}T + H(M,T)\,\mathrm{d}M.$$

In our notation, it is written by

$$\mathrm{d}F = -S_m\,\mathrm{d}T + \frac{1}{2}N_0 h\,\mathrm{d}\sigma = -T_0 S_m\,\mathrm{d}t + \frac{N_0}{2}h\,\mathrm{d}\sigma. \tag{B.1}$$

The Maxwell relation is equivalent to saying that (B.1) represents the total derivative. It implies the following relation:

$$\frac{\partial^2 F}{\partial\sigma\,\partial T} = -\frac{\partial S_m}{\partial\sigma} = \frac{N_0}{2T_0}\frac{\partial h}{\partial t} = \frac{\partial^2 F}{\partial T\,\partial\sigma}.$$

The t-derivative under the fixed σ condition can be replaced by $\partial y/\partial t$ as follows:

$$-\frac{\partial}{\partial\sigma}\left(\frac{S_m(\sigma,t)}{N_0}\right) = \frac{T_A}{2T_0}\sigma\left.\frac{\partial y(\sigma,t)}{\partial t}\right|_\sigma. \tag{B.2}$$

The validity of (B.2) will be shown below for our entropy.

From the t-derivatives of the minimum condition of the free energy (2.7) under the fixed σ condition, we can get the following relations:

$$2B_t'(y)\frac{\partial y}{\partial t}+B'(y_z)\frac{\partial y_z}{\partial t}+\frac{T_A}{dT_0}\frac{\partial\eta_1}{\partial t}=-2\frac{\partial A_t(y,t)}{\partial t}-\frac{\partial A(y_z,t)}{\partial t},$$
$$\frac{2T_0}{T_A}\left[B'(y_z)\frac{\partial y_z}{\partial t}-B_t'(y)\frac{\partial y}{\partial t}\right]-\frac{\partial\lambda}{\partial t}-\frac{1}{3}\frac{\partial\eta_1}{\partial t}=-\frac{2T_0}{T_A}\left[\frac{\partial A(y_z,t)}{\partial t}-\frac{\partial A_t(y,t)}{\partial t}\right] \tag{B.3}$$

where $B_t'(y,t)$ and $B'(y_z,t)$ stand for the y- and y_z-derivatives of $B_t(y,t)$, $B(y_z,t)$, respectively, given by

$$B'(y,t)=\frac{1}{t}\int_0^1 \mathrm{d}x\,x^4[1/u+1/2u^2-\psi'(u)]-c_z$$
$$B_t'(y,t)=\frac{1}{t}\int_{x_c}^1 \mathrm{d}x\,x^4[1/u+1/2u^2-\psi'(u)]-c_z+A_{\mathrm{sw}}'(\sigma,t)$$
$$A_{\mathrm{sw}}'(\sigma,t)=-\frac{1}{2t}\left(\frac{T_A\sigma}{T_0}\right)^2\int_0^{x_c}\mathrm{d}x\,x^2\frac{\beta\omega_q\mathrm{e}^{\beta\omega_q}}{(\mathrm{e}^{\beta\omega_q}-1)^2}.$$

The y-derivative of the thermal spin-wave amplitude is also shown above as $A_{\mathrm{sw}}'(\sigma,t)$. The partial t-derivative of the transverse thermal and spin-wave amplitudes, on the other hand, are given by

$$\frac{\partial A_t(y,t)}{\partial t}=-\frac{1}{t}\int_{x_c}^1\mathrm{d}x\,x^3u\left[1/u+1/2u^2-\psi'(u)\right]+\frac{\partial A_{\mathrm{sw}}(\sigma,t)}{\partial t}$$
$$\frac{\partial A_{\mathrm{sw}}(\sigma,t)}{\partial t}=\frac{T_A\sigma}{2T_0t}\int_0^{x_c}\mathrm{d}x\,\frac{x^2\beta\omega_q\mathrm{e}^{\beta\omega_q}}{(\mathrm{e}^{\beta\omega_q}-1)^2}.$$

The following relations are also derived from the σ-derivatives:

$$2B_t'(y,t)\frac{\partial y}{\partial\sigma}+B'(y_z,t)\frac{\partial y_z}{\partial\sigma}+\frac{T_A\sigma}{2T_0d}=-2D_{\mathrm{sw}},$$
$$d\frac{2T_0}{3T_A}\left[B'(y_z,t)\frac{\partial y_z}{\partial\sigma}-B_t'(y,t)\frac{\partial y}{\partial\sigma}\right]-\frac{\sigma}{6}-\frac{\partial\lambda}{\partial\sigma}=d\frac{2T_0}{3T_A}D_{\mathrm{sw}}$$
$$D_{\mathrm{sw}}=\frac{\partial A_{\mathrm{sw}}}{\partial\sigma}+\frac{T_A}{2T_0d}\frac{\partial\eta_1}{\partial\sigma}=\frac{\partial A_{\mathrm{sw}}}{\partial\sigma}-\frac{A_{\mathrm{sw}}}{\sigma}=-\frac{t}{\sigma}\frac{\partial A_{\mathrm{sw}}}{\partial t}$$

where we have used (2.12) between η_1' and A_{sw}, and the relation in the last line,

$$\frac{\partial A_{\mathrm{sw}}}{\partial\sigma}=\frac{A_{\mathrm{sw}}}{\sigma}-\frac{t}{\sigma}\frac{\partial A_{\mathrm{sw}}}{\partial t}.$$

Both the above t- and σ-derivatives of y and y_z can be expressed in terms of those of y and Δy_z in the matrix forms

$$M\begin{pmatrix}\frac{\partial y}{\partial t}\\ \frac{\partial\Delta y_z}{\partial t}\end{pmatrix}=\begin{pmatrix}-v_1\\ v_2\end{pmatrix}$$
$$M\begin{pmatrix}\frac{\partial y}{\partial\sigma}\\ \frac{\partial\Delta y_z}{\partial\sigma}\end{pmatrix}=\left(\frac{T_A\sigma}{6T_0}-\frac{2t}{\sigma}\frac{\partial A_{\mathrm{sw}}}{\partial t}\right)\begin{pmatrix}-1\\ 1/2\end{pmatrix}+\frac{3T_A}{6T_0}\frac{\partial\lambda}{\partial\sigma}\begin{pmatrix}0\\ 1\end{pmatrix} \tag{B.4}$$

where v_1 and v_2 are defined by

$$v_1=-2\frac{\partial A_t(y,t)}{\partial t}-\frac{\partial A(y_z,t)}{\partial t}-\frac{T_A}{3T_0}\frac{\partial\eta_1}{\partial t}$$
$$v_2=\frac{\partial A_t(y,t)}{\partial t}-\frac{\partial A(y_z,t)}{\partial t}+\frac{T_A}{6T_0}\left[3\frac{\partial\lambda}{\partial t}+\frac{\partial\eta_1}{\partial t}\right].$$

The 2×2 matrix M and its inverse are introduced by

$$M = \begin{pmatrix} 2B_t'(y,t) + B'(y_z,t) & B'(y_z,t) \\ -B_t'(y,t) + B'(y_z,t) & B'(y_z,t) \end{pmatrix},$$

$$M^{-1} = \frac{1}{\det M} \begin{pmatrix} B'(y_z,t) & -B'(y_z,t) \\ B_t'(y,t) - B'(y_z,t) & 2B_t'(y,t) + B'(y_z,t) \end{pmatrix} \tag{B.5}$$

$$= \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix}.$$

For the right-hand side of (B.2), the following expression of $\partial y/\partial t$ is derived from the above results:

$$\frac{\partial y}{\partial t} = -m_{11}\left[2\,\frac{\partial A_t(y,t)}{\partial t} + \frac{\partial A(y_z,t)}{\partial t}\right] + m_{12}\left[\frac{\partial A_t(y,t)}{\partial t} - \frac{\partial A(y_z,t)}{\partial t}\right]$$
$$+ \frac{T_A}{6T_0}\left[3m_{12}\,\frac{\partial \lambda}{\partial t} + 2(-m_{11} + m_{12}/2)\,\frac{\partial \eta_1}{\partial t}\right]$$
$$= -m_{11}v_1 + m_{12}v_2. \tag{B.6}$$

On the other hand, σ-derivative of the entropy is given by

$$\frac{1}{N_0}\,\frac{\partial S_{\mathrm{m}}}{\partial \sigma} = -3\left[2\frac{\partial A_t(y,t)}{\partial t}\,\frac{\partial y}{\partial \sigma} + \frac{\partial A(y_z,t)}{\partial t}\,\frac{\partial y_z}{\partial \sigma}\right] + \frac{T_A}{T_0}s_{\mathrm{sw}} + \frac{1}{N_0}\,\frac{\partial \Delta S_{\mathrm{m}}}{\partial \sigma}. \tag{B.7}$$

Among terms of the above result, the last two terms can be rewritten in terms of the derivatives of y and Δy_z as shown below. The third spin-wave term can be expressed as follows:

$$s_{\mathrm{sw}} = -\frac{3T_0}{T_A\sigma}\int_0^{x_{\mathrm{c}}} \mathrm{d}x\, x^2 \frac{\mathrm{e}^{\beta\omega_q}(\beta\omega_q)^2}{(\mathrm{e}^{\beta\omega_q}-1)^2} - \frac{\partial}{\partial \sigma}\left(y\,\frac{\partial \eta_1}{\partial t} + \frac{\partial \eta_0}{\partial t}\right)$$
$$= -\frac{\partial y}{\partial \sigma}\,\frac{\partial \eta_1}{\partial t} + \frac{\partial y}{\partial t}\,\frac{\partial \eta_1}{\partial \sigma} - \frac{\partial}{\partial t}\left(y\,\frac{\partial \eta_1}{\partial \sigma} + \frac{\partial \eta_0}{\partial \sigma}\right) - \frac{3T_0}{T_A\sigma}\int_0^{x_{\mathrm{c}}} \mathrm{d}x\, x^2 \frac{\mathrm{e}^{\beta\omega_q}(\beta\omega_q)^2}{(\mathrm{e}^{\beta\omega_q}-1)^2}$$
$$= -\frac{\partial y}{\partial \sigma}\,\frac{\partial \eta_1}{\partial t} - \frac{6T_0 t}{T_A\sigma}\,\frac{\partial A_{\mathrm{sw}}}{\partial t}\,\frac{\partial y}{\partial t}.$$

We have used the following relation derived from (2.11):

$$\frac{\partial}{\partial t}\left(y\,\frac{\partial \eta_1}{\partial \sigma} + \frac{\partial \eta_0}{\partial \sigma}\right) + \frac{T_0 d}{T_A\sigma}\int_0^{x_{\mathrm{c}}} \mathrm{d}x\, \frac{x^2\mathrm{e}^{\beta\omega_q}(\beta\omega_q)^2}{(\mathrm{e}^{\beta\omega_q}-1)^2} = \frac{\partial \eta_1}{\partial \sigma}\,\frac{\partial y}{\partial t} + \frac{6T_0 t}{T_A\sigma}\,\frac{\partial A_{\mathrm{sw}}}{\partial t}\,\frac{\partial y}{\partial t}.$$

From the definition (2.10) for ΔF_1 and (3.2), the σ-derivative of the last entropy correction is given by

$$\frac{\partial \Delta S_{\mathrm{m}}}{\partial \sigma} = N_0\frac{T_A}{T_0}\left(\frac{\partial \lambda}{\partial t}\,\frac{\partial \Delta y_z}{\partial \sigma} - \frac{\partial \lambda}{\partial \sigma}\,\frac{\partial \Delta y_z}{\partial t}\right).$$

On substitution of these results the left-hand side of (B.2) can be expressed in terms of partial derivatives of y and Δy_z as follows:

$$\frac{1}{N_0}\,\frac{\partial S_{\mathrm{m}}}{\partial \sigma} = -3\left[2\,\frac{\partial A_t(y,t)}{\partial t} + \frac{\partial A(y_z,t)}{\partial t}\right]\frac{\partial y}{\partial \sigma} - 3\,\frac{\partial A(y_z,t)}{\partial t}\,\frac{\partial \Delta y_z}{\partial \sigma}$$
$$- \frac{T_A}{T_0}\,\frac{\partial y}{\partial \sigma}\,\frac{\partial \eta_1}{\partial t} - \frac{6t}{\sigma}\,\frac{\partial A_{\mathrm{sw}}}{\partial t}\,\frac{\partial y}{\partial t} + \frac{T_A}{T_0}\left(\frac{\partial \lambda}{\partial t}\,\frac{\partial \Delta y_z}{\partial \sigma} - \frac{\partial \lambda}{\partial \sigma}\,\frac{\partial \Delta y_z}{\partial t}\right)$$
$$= -3v_1\,\frac{\partial y}{\partial \sigma} + (v_1 - 2v_2)\,\frac{\partial \Delta y_z}{\partial \sigma}$$
$$- \frac{6t}{\sigma}\,\frac{\partial A(y_z,t)}{\partial t}\,\frac{\partial y}{\partial t} - \frac{T_A}{T_0}\,\frac{\partial \lambda}{\partial t}\,\frac{\partial \Delta y_z}{\partial t}. \tag{B.8}$$

It is also given in the form of the simple matrix expression

$$\frac{1}{N_0}\frac{\partial S_{\rm m}}{\partial\sigma} = \left(-\frac{6t}{\sigma}\frac{\partial A_{\rm sw}}{\partial t}, \quad -\frac{T_A}{T_0}\frac{\partial\lambda}{\partial\sigma}\right) M^{-1}\begin{pmatrix}-v_1\\ v_2\end{pmatrix} - \left(v_1, \quad \frac{v_1-2v_2}{3}\right) M^{-1}\begin{pmatrix}-\frac{T_A\sigma}{2T_0}+\frac{6t}{\sigma}\frac{\partial A_{\rm sw}}{\partial t}\\ \frac{T_A\sigma}{4T_0}-\frac{3t}{\sigma}\frac{\partial A_{\rm sw}}{\partial t}+\frac{3T_A}{2T_0}\frac{\partial\lambda}{\partial\sigma}\end{pmatrix}.$$

Let us next introduce two parameters V_1 and V_2 by

$$\frac{1}{N_0}\frac{\partial S_{\rm m}}{\partial\sigma} = v_1 V_1 + v_2 V_2, \tag{B.9}$$

then they are evaluated explicitly as follows:

$$\begin{aligned}
V_1 &= \frac{T_A\sigma}{2T_0}(1, \quad 1/3)\, M^{-1}\begin{pmatrix}1\\ -1/2\end{pmatrix}\\
&\quad + \frac{3t}{\sigma}\frac{\partial A_{\rm sw}}{\partial t}\left[(-2, \quad 0)\, M^{-1}\begin{pmatrix}-1\\ 0\end{pmatrix} - (1, \quad 1/3)\, M^{-1}\begin{pmatrix}2\\ -1\end{pmatrix}\right]\\
&\quad + \frac{T_A}{T_0}\frac{\partial\lambda}{\partial\sigma}\left[(0, \quad -1)\, M^{-1}\begin{pmatrix}-1\\ 0\end{pmatrix} - (1, \quad 1/3)\, M^{-1}\begin{pmatrix}0\\ 3/2\end{pmatrix}\right]\\
&= \frac{T_A\sigma}{2T_0}\left(m_{11}+\frac{m_{12}}{3}-\frac{m_{12}}{2}+\frac{m_{22}}{6}\right)+\frac{3t}{\sigma}\frac{\partial A_{\rm sw}}{\partial t}\left(m_{12}-\frac{2m_{21}}{3}+\frac{m_{22}}{3}\right)\\
&\quad + \frac{T_A}{T_0}\frac{\partial\lambda}{\partial\sigma}\left(-\frac{3m_{12}}{2}+m_{21}+\frac{m_{22}}{2}\right) = \frac{T_A\sigma}{2T_0}m_{11}\\
V_2 &= -\frac{T_A\sigma}{2T_0}(0, \quad -2/3)\, M^{-1}\begin{pmatrix}-1\\ 1/2\end{pmatrix}\\
&\quad + \frac{3t}{\sigma}\frac{\partial A_{\rm sw}}{\partial t}\left[(-2, \quad 0)\, M^{-1}\begin{pmatrix}0\\ 1\end{pmatrix} - (0, \quad -2/3)\, M^{-1}\begin{pmatrix}2\\ -1\end{pmatrix}\right]\\
&\quad + \frac{T_A}{T_0}\frac{\partial\lambda}{\partial\sigma}\left[-(0, \quad 1)\, M^{-1}\begin{pmatrix}0\\ 1\end{pmatrix} - (0, \quad -2/3)\, M^{-1}\begin{pmatrix}0\\ 3/2\end{pmatrix}\right]\\
&= -\frac{T_A\sigma}{2T_0}m_{12}.
\end{aligned}$$

Note the following identity for the inverse matrix elements:

$$3m_{12}/2 - m_{21} + m_{22}/2 = 0.$$

If we finally compare the following result with (B.6),

$$\frac{1}{N_0}\frac{\partial S_{\rm m}}{\partial\sigma} = \frac{T_A\sigma}{2T_0}(v_1 m_{11} - v_2 m_{12})$$

the Maxwell relation is verified.

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