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Spin fluctuation theory of specific heat in itinerant electron magnets

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Abstract. The temperature dependence of the specific heat in itinerant electron magnets is discussed by taking into account the effects of both the thermal and the quantum spin fluctuations. We also propose the form of the free energy consistent with the sum rule of the total spin fluctuation amplitude. As a check of the validity of the formalism, the Maxwell relation is studied.

1. Introduction

The effect of exchange enhanced collective magnetic excitations on the specific heat has a long history of theoretical research. It goes back to the paramagnon theories of the exchange enhanced Landau Fermi liquids paying particular attention to their behaviour at low temperature (Doniach and Engelsberg 1966, Brinkmann and Engelsberg 1968, Berk and Schrieffer 1966). On the other hand the temperature dependence of the specific heat has been discussed in the wide temperature range from the ground state to the paramagnetic phase through the magnetic transition temperature (Murata and Doniach 1972, Makoshi and Moriya 1975, Hasegawa 1975). Recently the specific heat has renewed its interest in relation to the low-dimensional itinerant electron systems (Hatatani and Moriya 1995) and the quantum critical phenomena (Hertz 1976, Millis 1993, Zülicke and Millis 1995, Ishigaki and Moriya 1996, Pfleiderer *et al* 1997).

Although the self-consistent renormalization (SCR) spin fluctuation theory has been successful in explaining various magnetic properties of weak itinerant electron magnets (Moriya 1985, Lonzarich and Taillefer 1985), there still remain unresolved problems in its treatment of the specific heat. First the effect of quantum zero-point spin fluctuations is neglected from the beginning by assuming that its temperature dependence is very weak. No convincing arguments have yet been presented. In deriving the temperature dependence of the magnetic susceptibility, for instance, the self-consistent change of the spin fluctuation spectrum is found to play important roles. In the SCR theory the effect, however, has not been reflected on the quantum fluctuation amplitude by assuming its effect is very weak. A series of the present author's studies on the spin fluctuation effects have revealed the significant roles of the quantum spin fluctuation amplitude (Takahashi 1986, 1990, 1992, 1994, 1997a, b, 1998, Takahashi and Sakai 1995, 1998) in deriving the magnetic properties of itinerant magnets. Theoretical consequences of the necessity of including quantum amplitudes have been supported by experimental efforts (Yoshimura *et al* 1988a, b, Shimizu *et al* 1990, Nakabayashi *et al* 1992). Therefore we have to know how the specific heat is influenced by

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the quantum spin fluctuations. As for another problem, the SCR theory predicts a spurious steep decrease, like the dip structure, in the temperature dependence of the specific heat of weak itinerant ferromagnets just above the critical temperature (Makoshi and Moriya 1975). This seems to suggest the presence of a slight inconsistency in the current SCR formalism of the specific heat.

The aim of the present paper is to give an answer to these problems mentioned above. Moreover, in our previous studies on the magnetic properties of itinerant magnetism, no mention has been given of the explicit form of the free energy. On its behalf, we have bypassed our discussions based of the sum rule that the sum of thermal and quantum spin fluctuation amplitudes is almost constant. We propose here a free energy expression that is consistent with the sum rule. Based on the free energy, we will give the consistency check of our formalism based on the Maxwell relation of the thermodynamics.

In the next section, after a brief review of our spin fluctuation theory, a free energy expression is proposed. Based on the free energy, the magnetic entropy is derived. In section 3, the specific heat formula is derived, and the temperature dependence of the specific heat is discussed in the critical and the low temperature regions. As a test of the validity of the present formalism, the Maxwell relation is studied in section 4. The final section is devoted to discussions.

In what follows the magnetization $M_{\boldsymbol{Q}}$ with a wavevector $\boldsymbol{Q}$ is expressed in terms of the dimensionless parameter σ in units of Bohr magnetons μ_B per magnetic atom and the wavevector dependent external field $H_{\boldsymbol{Q}}$ by h in energy units:

$$M_{\boldsymbol{Q}} = N_0 \mu_B \sigma \qquad h = 2\mu_B H_{\boldsymbol{Q}}$$

where N_0 is the number of magnetic atoms in the system. The magnetic susceptibility $\chi(\boldsymbol{Q})$ measured in units of $4\mu_B^2$ is in the present units given by

$$\chi(\boldsymbol{Q})/N_0 = \sigma/2h.$$

2. Free energy expression

Before we introduce a free energy, we briefly summarize the framework of our spin fluctuation theory for later convenience. In our previous investigations, we have assumed that the total spin fluctuation amplitude, defined in the paramagnetic phase, for instance, as the sum of the thermal and the quantum (zero-point) amplitudes by

$$\langle \boldsymbol{S}_i^2 \rangle_{\text{tot}} = \frac{1}{N_0^2} \sum_{\boldsymbol{q}} \int_0^\infty \frac{\mathrm{d}\omega}{\pi} [1 + 2n(\omega)] \operatorname{Im} \chi(\boldsymbol{Q}+\boldsymbol{q}, \omega) = \langle \boldsymbol{S}_i^2 \rangle_Z + \langle \boldsymbol{S}_i^2 \rangle_T \qquad (1)$$

is almost unaffected by the temperature variation and the presence of the external magnetic field h. To generalize our treatment, we assume that the magnetic excitations are enhanced around a wavvector $\boldsymbol{Q}$. The spin fluctuation spectrum is then given by

$$\operatorname{Im} \chi(\boldsymbol{Q}+\boldsymbol{q}, \omega) = \frac{\chi(\boldsymbol{Q}, 0)}{1 + q^2/\kappa^2} \frac{\omega \Gamma_{\boldsymbol{Q}+\boldsymbol{q}}}{\omega^2 + \Gamma_{\boldsymbol{Q}+\boldsymbol{q}}^2} \qquad \Gamma_{\boldsymbol{Q}+\boldsymbol{q}} = \Gamma_0 q^\alpha (\kappa^2 + q^2) \qquad (2)$$

where κ^2 is the squared inverse magnetic correlation length proportional to the inverse magnetic susceptibility. The above expression (2) is justified in the small q, ω region. To be applicable to both the ferro- (F) and the antiferromagnetic (AF) cases, the exponent α is introduced to characterize the wavevector dependence of the damping constant $\Gamma_{\boldsymbol{Q}+\boldsymbol{q}}$ ($\alpha = 1$, 0 for F and AF, respectively). Let us now represent the above spectral form in the parametrized form,

$$\operatorname{Im} \chi(\boldsymbol{Q}+\boldsymbol{q}, \omega)/N_0 = \frac{T_0}{2T_A T} \frac{\xi x^\alpha}{\xi^2 + u^2} \qquad u = x^\alpha (y + x^2)/t$$

by introducing the parameters, y, T_0 and T_A as

$$y = \kappa^2/q_B^2 \qquad T_0 = \Gamma_0 q_B^{2+\alpha}/2\pi \qquad T_A = N_0 q_B^2/[2\chi(\boldsymbol{Q}, 0)\kappa^2]$$

where $\xi = \omega/2\pi T$, $t = T/T_0$, $x = q/q_B$ and q_B is the magnitude of the effective zone-boundary wavevector. In terms of the dynamical magnetic susceptibility in (2), the thermal and the quantum components are, respectively, represented by

$$\begin{aligned} \langle \boldsymbol{S}_i^2 \rangle_T &= \frac{3dT_0}{T_A} A(y, t) \\ A(y, t) &= \int_0^1 \mathrm{d}x x^{d-1+\alpha} [\ln u - 1/2u - \psi(u)] \\ \langle \boldsymbol{S}_i^2 \rangle_Z(y) &= \langle \boldsymbol{S}_i^2 \rangle_Z(0) - \frac{3dT_0}{T_A} c_Z y + \cdots \end{aligned} \tag{3}$$

where $\psi(u)$ is the digamma function. The y-linear dependence of the quantum amplitude is verified for three-dimensional cases for small y value and c_Z is a constant of the order of unity determined by the spin fluctuation spectrum. The y-dependence of the quantum amplitude is modified depending on the dimensionality and the nature of magnetism, F or AF. We have transformed the wavevector summation into the integral form with respect to the reduced dimensionless wavevector $x = q/q_{\mathrm{B}}$:

$$\frac{1}{N_0} \sum_{\boldsymbol{q}} = d \int_0^1 \mathrm{d}x x^{d-1}$$

where d is the dimensionality of the system.

In the presence of the static moment $\sigma = 2|\langle \boldsymbol{S}_{\boldsymbol{Q}} \rangle|/N_0$, the sum rule has to be slightly modified. It is replaced by the condition that the squared static moment and the fluctuation amplitude,

$$\langle \boldsymbol{S}_i^2 \rangle_{\mathrm{tot}} = \frac{\sigma^2}{4} + \langle \delta \boldsymbol{S}_i^2 \rangle_Z + \langle \delta \boldsymbol{S}_i^2 \rangle_T \qquad (\delta \boldsymbol{S}_i = \boldsymbol{S}_i - \langle \boldsymbol{S}_i \rangle) \tag{4}$$

is totally conserved. We also need to account for the anisotropy of the fluctuation amplitude. The effect is included by assuming that reciprocals of the squared magnetic correlation length, κ^2 and κ_z^2, of perpendicular and longitudinal direction are given, respectively, by

$$y = \kappa^2/q_B^2 = \frac{1}{T_A} \frac{h}{\sigma} \qquad y_z = \kappa_z^2/q_B^2 = \frac{1}{T_A} \frac{\partial h}{\partial \sigma}. \tag{5}$$

With the use of the sum rule, the temperature dependence of the magnetic susceptibility, i.e. the t dependence of y, is obtained by solving

$$A(y, t) - c_Z = A(0, t_c) \qquad (t_c = T_c/T_0)$$

in the paramagnetic phase, for instance. The magnetic equation of state is also obtained with the use of (4) (Takahashi 1986).

As the free energy consistent with the sum rule we propose the following expression:

$$F_m(\sigma, t) = \frac{3}{\pi} \sum_{\boldsymbol{q}} \int_0^{\omega_c} \mathrm{d}\omega \left[\frac{\omega}{2} + T \ln(1 - \mathrm{e}^{-\omega/T}) \right] \frac{\Gamma_{\boldsymbol{Q}+\boldsymbol{q}}}{\Gamma_{\boldsymbol{Q}+\boldsymbol{q}}^2 + \omega^2} + \frac{N_0 T_A}{4} y \sigma^2 + \Delta F(y) \tag{6}$$

where ω_c is the upper cut-off frequency. It has explicit t dependence as well as implicit t and σ dependence through that of y. The first term represents the contribution from the collective spin fluctuation modes, that consists of both the effects of thermal and quantum zero-point fluctuations. The second term represents the magnetic Zeeman energy, i.e. the MH term (in the case of F) in the usual notation in the presence of the external magnetic field. The reason to introduce the last term will be explained below. The differences of (6) from the SCR expression are the presence of the quantum fluctuations, the term proportional to $\omega/2$, in the first term and the additional term $\Delta F(y)$.

To clarify the reason to include $\Delta F(y)$, let us see the free energy variation against the parameter y. It is given by

$$\delta(F_m/N_0) = \delta y \frac{3}{\pi} \sum_{q} \int_0^{\omega_c} \mathrm{d}\omega \left[\frac{\omega}{2} + T \ln(1 - \mathrm{e}^{-\omega/T}) \right] \frac{\partial}{\partial \Gamma_{Q+q}} \left(\frac{\Gamma_{Q+q}}{\omega^2 + \Gamma_{Q+q}^2} \right) \Gamma_0 q^\alpha q_B^2 + \delta y \left(\frac{T_A}{4} \sigma^2 + \frac{\partial \Delta F(y)}{\partial y} \right). \tag{7}$$

The above expression is further simplified if we replace the Γ_{Q+q} derivative of the first term by that of ω with the use of the relation

$$\frac{\partial}{\partial \omega} \left(\frac{\omega}{\omega^2 + \Gamma_{Q+q}^2} \right) = \frac{\Gamma_{Q+q}^2 - \omega^2}{(\Gamma_{Q+q}^2 + \omega^2)^2} = -\frac{\partial}{\partial \Gamma_{Q+q}} \left(\frac{\Gamma_{Q+q}}{\omega^2 + \Gamma_{Q+q}^2} \right). \tag{8}$$

After the partial integration, the variation is given by

$$\begin{aligned} \delta(F_m/N_0) &= \delta y \frac{3}{\pi} \sum_{q} \left\{ - \left[\frac{\omega}{2} + T \ln(1 - \mathrm{e}^{-\omega/T}) \right] \frac{\omega \Gamma_{Q+q}}{\Gamma_{Q+q}^2 + \omega^2} \bigg|_0^{\omega_c} \right. \\ &\quad \left. + \int_0^{\omega_c} \mathrm{d}\omega [\tfrac{1}{2} + n(\omega)] \frac{\omega \Gamma_{Q+q}}{\Gamma_{Q+q}^2 + \omega^2} \right\} \frac{1}{y + q^2/q_B^2} + \delta y \left(\frac{T_A}{4} \sigma^2 + \frac{\partial \Delta F(y)}{\partial y} \right) \\ &= \left[T_A \langle \delta \boldsymbol{S}_i^2 \rangle_{\mathrm{tot}} + \frac{T_A}{4} \sigma^2 + \frac{\partial \Delta F(y)}{\partial y} \right] \delta y. \end{aligned} \tag{9}$$

The first term on the top line of (9) is neglected, because the actual frequency dependence of the spectral intensity should decrease faster than the Lorentzian distribution in the high frequency range. We hereafter assume the ω integration is well behaved in the high frequency region and the upper cut-off frequency is infinity. The above result indicates that the extremum condition of the free energy against the variation of y leads to the sum rule of the spin fluctuation amplitude. By expanding $\Delta F(y)$ around $y = 0$, we obtain the following relation by the condition, $\partial F_m/\partial y = 0$:

$$\langle \boldsymbol{S}_i^2 \rangle_{\mathrm{tot}} = \langle \delta \boldsymbol{S}_i^2 \rangle_{\mathrm{tot}} + \frac{1}{4} \sigma^2 = -\frac{1}{T_A} \frac{\Delta F'(0)}{N_0}. \tag{10}$$

Because we are particularly interested in the exchange enhanced nearly magnetic or weakly magnetic cases where y is always very small, the expansion of $\Delta F(y)$ in terms of small y value is justified. From (10) we see that the total spin fluctuation amplitude is related to the first derivative of $\Delta F(y)$ around the origin $y = 0$. In this way we are able to derive the self-consistent equation of y by the stability condition of the free energy. The presence of $\Delta F(y)$, supposed to give rise from a self-energy correction of fluctuations, is crucial. The explicit t and σ dependence of $\Delta F(y)$ is assumed to be very weak.

In the SCR theory, on the other hand, because no explicit correction term ΔF or contribution from quantum spin fluctuations are present, the y derivative of free energy F_{SCR}, in the paramagnetic phase, is proportional to the *thermal* spin fluctuation amplitude given by

$$\frac{1}{N_0} \frac{\partial F_{\mathrm{SCR}}(t, y)}{\partial y} = T_A \langle \delta \boldsymbol{S}_i^2 \rangle_T = 3 d T_0 A(y, t). \tag{11}$$

As an illustration of the application of our free energy, it is easy to see that the thermodynamic relation is obtained as follows:

$$\frac{1}{N_0} \frac{\partial F_m}{\partial \sigma} = \frac{1}{N_0} \left(\frac{\partial F_m}{\partial y} \frac{\partial y}{\partial \sigma} + \frac{\partial F_m}{\partial \sigma} \right) = \frac{T_A}{2} \sigma y = h/2 \tag{12}$$

that is equivalent to the relation $\partial F_m/\partial M = H$ in the F case. The first term proportional to $\partial y/\partial\sigma$ vanishes identically due to the extremum condition.

The magnetic entropy is obtained by the first derivative of the free energy with respect to its explicit temperature dependence, for the implicit temperature dependence through y vanishes from the same reason. The entropy is therefore given by

$$-S_m = \frac{\partial F_m}{\partial T} = \frac{3}{\pi}\sum_q \int_0^\infty \mathrm{d}\omega \left[\ln(1 - \mathrm{e}^{-\omega/T}) - \frac{\omega}{T} n(\omega) \right] \frac{\Gamma_{Q+q}}{\omega^2 + \Gamma_{Q+q}^2}. \qquad (13)$$

In the parametrized form, it is represented by

$$S_m/N_0 = -3d \int_0^1 \mathrm{d}x x^{d-1} [\ln\sqrt{2\pi} - u + (u - 1/2)\ln u - \ln\Gamma(u)]$$
$$+3d \int_0^1 \mathrm{d}x x^{d-1} \left[\ln u - \frac{1}{2u} - \psi(u) \right] \qquad (14)$$

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

3. Specific heat

The specific heat is obtained by differentiating (14) with respect to the temperature T. As the sum of two contributions, it is given by

$$C_m/N_0 t = 3d \left(I_1 - \frac{\partial y}{\partial t} I_2 \right)$$
$$I_1(t, y) = \frac{1}{t} \int_0^1 \mathrm{d}x x^{d-1} u^2 \{-1/u - 1/2u^2 + \psi'(u)\} \qquad (15)$$
$$I_2(t, y) = \frac{1}{t} \int_0^1 \mathrm{d}x x^{d-1+\alpha} u \{-1/u - 1/2u^2 + \psi'(u)\} = \frac{\partial A(y, t)}{\partial t}.$$

Note that the above result is derived by taking explicit account of the effect of quantum spin fluctuations. In contrast with the SCR theory, no additional terms proportional to $\partial^2 y/\partial t^2$ and $(\partial y/\partial t)^2$ are present in (15). They originate from the t dependence of the thermal amplitude in (11) through the implicit t dependence of the parameter y (see the appendix for brief comparison between (15) and the SCR formula). The spurious dip structure in the temperature dependence of the specific heat just above t_c results from the term proportional to the second derivative $\partial^2 y/\partial t^2$ (Makoshi and Moriya 1975). They were therefore sometimes dropped simply to avoid an unfavourable behaviour for the comparison between the theory and experiments (Takeuchi and Masuda 1979). In order to evaluate the temperature dependence of the specific heat at general temperature, we need to obtain those of y and its derivative $\partial y/\partial t$. With these values, the specific heat is numerically evaluated by (15).

Before showing numerical results, we review the temperature dependence of the specific heat in two particular temperature ranges, around the critical temperature and at low temperature. From the numerical estimates, we found that the main contribution comes from the first term I_1. The second term proportional to $\partial y/\partial t$ is always small compared to the first one. Let us first see the behaviours of the integrand of I_1 as a function of x. Depending on the magnitude of u, it behaves as follows:

$$x^{d-1} u^2 [-1/u - 1/2u^2 + \psi'(u)] \sim \begin{cases} x^{d-1}/2 & \text{for } u \ll 1 \\ \dfrac{x^{d-1}}{6u} = \dfrac{t}{6} \dfrac{x^{d-1-\alpha}}{y + x^2} & \text{for } u \gg 1. \end{cases}$$

It follows from the above expression:

(i) The integrand always behaves normally around $x = 0$. Especially at finite temperature t, I_1 is always finite including the case $y = 0$, for u is also finite.

(ii) The asymptotic expansion,

$$u^2[-1/u - 1/2u^2 + \psi'(u)] \sim 1/6u - 1/30u^3 + \cdots$$

to be used in discussing the low temperature behaviour ($1 \ll u$), is not justified around $x = 0$ due to the divergent behaviour of each expansion term. The integrand itself, however, has to be finite as stated above. Therefore if we evaluate the integral with the use of the expansion, we need to introduce the lower cut-off wavevector. If we roughly estimate the lower bound by the condition $u \gtrsim 1$, it is given by $t^{1/(2+\alpha)} \lesssim x$ and $t/y \lesssim x$ depending on the condition $y \ll t^{2/(2+\alpha)}$ and $y \gg t^{2/(2+\alpha)}$, respectively.

(iii) When $d \leqslant 2 + \alpha$, the most dominant term of the asymptotic expansion will show the divergent behaviour as $y \to 0$.

With these properties in mind, the temperature dependence is discussed below in two temperature ranges, i.e. around the critical region, $y \ll t^{2/(2+\alpha)}$, and in the low temperature limit, $y \gg t^{2/(2+\alpha)}$, paying particular attention to the behaviour of I_1.

3.1. Specific heat around the critical region

The critical temperature region is characterized by the condition $y \ll t^{2/(2+\alpha)}$. When t is finite, the integral is always finite even for $y = 0$. Therefore we can assume $y = 0$, to see the t dependence of the critical value of the integral.

If we transform the integration variable from x to $u = x^{2+\alpha}/t$ with the use of the relation

$$x^{d-1}\,\mathrm{d}x = \frac{t^{\nu-1}}{2+\alpha}u^{\nu-2}\,\mathrm{d}u$$

I_1 can be represented by

$$\begin{aligned} I_1(t,0) &= \frac{t^{\nu-2}}{2+\alpha}\int_0^{1/t}\mathrm{d}u\,u^{\nu}\left(-\frac{\partial}{\partial u}\right)\left[\ln u - \frac{1}{2u} - \psi(u)\right] \\ &= \frac{1}{2+\alpha}\{-t^{\nu-2}[u^{\nu}(\ln u - 1/2u - \psi(u))]_0^{1/t} + \nu t^{\nu-2}J(\nu,t)\} \\ &= \frac{1}{2+\alpha}[\nu t^{\nu-2}J(\nu,t) - 1/12] \\ J(\nu,t) &= \int_0^{1/t}\mathrm{d}u\,u^{\nu-1}[\ln u - 1/2u - \psi(u)] \end{aligned} \qquad (16)$$

where $\nu = d/(2+\alpha) + 1$. The upper bound value of the first term of the second line gives a numerical constant $1/12$ from the asymptotic behaviour of the digamma function for large $1/t$ value. The lower bound value at $u = 0$ does not contribute irrespective of the nature of the magnetism (F or AF) or of the space dimensionality d, for ν is always greater than 1, as shown in table 1.

Table 1. The dependence of ν on the dimensionality d for F and AF.

	$d = 3$	$d = 2$	$d = 1$
F	2	5/3	4/3
AF	5/2	2	3/2

Table 2. Numerical values of $C^*_{5/2}$ and C_ν.

$C^*_{5/2}$	$C_{5/3}$	$C_{4/3}$	$C_{3/2}$
$0.080\,064\cdots$	$0.562\,992\cdots$	$1.006\,089\cdots$	$0.653\,093\cdots$

In the small t limit, the value of I_1 is estimated as follows:

$$(2+\alpha)I_1 \simeq \begin{cases} \frac{1}{3} - \frac{5}{2}C^*_{5/2}t^{1/2} & \text{for } \nu = 5/2 \\ \frac{1}{6}\ln(1/t) & \text{for } \nu = 2 \\ \nu C_\nu t^{\nu-2} & \text{for } 1 < \nu < 2. \end{cases}$$

The numerical constants, $C^*_{5/2}$ and C_ν, are defined by

$$C^*_{5/2} = \int_0^\infty \mathrm{d}u u^{3/2}[1/12u^2 - \ln u + 1/2u + \psi(u)] = \frac{\pi\zeta(5/2)\Gamma(5/2)}{(2\pi)^{5/2}\sin(\pi/4)}$$
$$C_\nu = \int_0^\infty \mathrm{d}u u^{\nu-1}[\ln u - 1/2u - \psi(u)] = \frac{\pi\zeta(\nu)\Gamma(\nu)}{(2\pi)^\nu \sin(\nu\pi/2)}$$

where $\zeta(\nu)$ is the zeta function (see table 2 for numerical values of $C^*_{5/2}$ and C_ν).

Before concluding this subsection, let us briefly discuss the contribution of I_2 for $d = 3$. For $u \gg 1$, the integral converges rapidly for finite x because the integrand behaves as follows:

$$\frac{1}{t}x^{2+\alpha}u[-1/u - 1/2u^2 + \psi'(u)] \sim \frac{1}{t}\frac{x^{2+\alpha}}{6u^2}.$$

From the wavevector integral around the origin, there appears no divergent y dependence and its value for $y = 0$ is given by

$$I_2 = \frac{1}{2+\alpha}t^{1/(2+\alpha)}\int_0^{1/t}\mathrm{d}u u^\mu[-1/u - 1/2u^2 + \psi'(u)] \simeq \frac{3+\alpha}{(2+\alpha)^2}C_\mu t^{1/(2+\alpha)} \qquad (t \to 0)$$
$$\mu = \frac{3+\alpha}{2+\alpha}.$$

Because y is proportional to $(t - t_c)^2$ around $t = t_c$, it follows that

$$\frac{\partial y}{\partial t} \propto y^{1/2} \qquad \frac{\partial y}{\partial t}I_2 \propto y^{1/2}I_2.$$

The second term of the first of equations (15) is therefore negligible around t_c and the critical value of C_m is solely determined by the I_1 value. On the other hand, $\partial^2 y/\partial t^2$ is finite around the narrow temperature region around $t = t_c$. If the term proportional to this factor is present, it gives a finite contribution, leading to the dip structure around t_c.

The above result on the critical value of the specific heat is interesting. It is expressed in the following universal form:

$$C_m/N_0T_c = \frac{1}{T_0}I_1(T_c/T_0, 0).$$

Especially for the three-dimensional AF case, since $I_1(t_c, 0)$ is almost given by $3/2$, the limiting value at $t = 0$, the above relation will be used to estimate the parameter T_0 from the value of the critical temperature T_c, if we can extract the spin fluctuation contribution from the observed total specific heat.

3.2. Specific heat in the low temperature region

Let us next deal with the temperature dependence of the specific heat in the $t = 0$ limit when y is finite. In the case of nearly itinerant ferromagnets, we can evaluate the integral I_1 by dividing the range of integration by the value $x_1 = t/y$ into two separate regions: the region (a) $0 \leqslant x \leqslant x_1$, and the region (b) $x_1 \leqslant x \leqslant 1$:

$$I_1 = \frac{1}{t}\left\{\int_0^{x_1} \mathrm{d}x + \int_{x_1}^1 \mathrm{d}x\right\} x^{d-1}u^2[-1/u - 1/2u^2 + \psi'(u)] = I_1^{(a)} + I_1^{(b)}. \tag{17}$$

In the region (a), u is always of the order of 1, i.e. $0 \leqslant u \lesssim 1$, while in the region (b) u rapidly increases its magnitude and the asymptotic expansion of $\psi(u)$ is justified. For the nearly AF case, the asymptotic expansion is justified for whole the x range at low temperature. We can therefore assume $x_1 = 0$. The former contribution $I_1^{(a)}$ is negligible, of the order of t^{d-1}/y^d, for the F case, because of the small phase volume. The most dominant expansion term of the latter is given by the first term, i.e.

$$I_1^{(b)} \sim \frac{1}{6}\int_{x_1}^1 \mathrm{d}x \frac{x^{d-1-\alpha}}{y + x^2}.$$

In the case of $d > \alpha$, since the integrand behaves normally around $x = 0$, the lower bound x_1 can be extended to 0, and the integral is estimated as follows:

$$I_1^{(b)} = \begin{cases} [1 - \sqrt{y}\tan^{-1}(1/\sqrt{y})]/6 & \text{for } d = 3 + \alpha \\ (1/12)\ln(1 + 1/y) & \text{for } d = 2 + \alpha \\ (1/6\sqrt{y})\tan^{-1}(1/\sqrt{y}) & \text{for } d = 1 + \alpha. \end{cases} \tag{18}$$

On the other hand for $d \leqslant \alpha$ ($d = \alpha = 1$, for example), $I_1^{(b)}$ has to be estimated by assuming the finite lower bound t/y. For $d = \alpha$, for instance, it is given by

$$I_1^{(b)} \sim \frac{1}{6}\int_{t/y}^1 \mathrm{d}x \frac{1}{x(y + x^2)} = \frac{1}{12y}\ln(y^3/t^2).$$

The higher order terms are evaluated as follows. In the ferromagnetic case ($\alpha = 1$), each expansion term is evaluated as follows (for $n \geqslant 1$):

$$\frac{1}{t}\int_{t/y}^1 \mathrm{d}x \frac{x^{d-1}}{u^{2n+1}} = t^{2n}\int_{t/y}^1 \mathrm{d}x \frac{1}{x^{2n+2-d}(y + x^2)^{2n+1}} \simeq \frac{1}{t}\left(\frac{t}{y}\right)^{2n+1}\int_{t/y}^1 \mathrm{d}x \frac{1}{x^{2n+2-d}}$$

$$\simeq \begin{cases} \dfrac{1}{2n + 1 - d}(t^{d-1}/y^d) & \text{for } 2n + 1 \neq d \\ (t^{d-1}/y^d)\ln(y/t) & \text{for } 2n + 1 = d. \end{cases} \tag{19}$$

Especially in the case of F for $d = 3$, $I_1^{(b)}$ behaves as follows:

$$I_1^{(b)} = \frac{1}{12}\ln(1 + 1/y) - \frac{t^2}{60y^3}\ln(y^3/t^2) + O(t^2/y^3)$$

as predicted by the paramagnon theory of exchange enhanced Landau Fermi liquids (Doniach and Engelsberg 1966). However, the observation of its behaviour is limited within the extreme low temperature region, $t < y^{3/2}$, as was pointed out by Konno and Moriya (1987). In the nearly AF case each expansion term, $1/u^{2n+1}$, only gives a contribution of the order of $(t/y)^{2n}/y^{1-d/2}$.

The general temperature dependence of the specific heat of exchange enhanced paramagnets can now be stated as follows. When y is very small, as we decrease the temperature t, the specific heat will first show the critical behaviour. As we further decrease the temperature,

Table 3. Characteristic temperature dependence of the specific heat, $3dI_1(\simeq C_m/N_0t)$.

	Ferromagnetism		Antiferromagnetism	
d	$t\sim 0$	$y\sim 0$	$t\sim 0$	$y\sim 0$
3	$\frac{3}{4}\ln(1/y)$	$\frac{1}{2}\ln(1/t)$	$\frac{3}{2}\left(1-\frac{\pi}{2}\sqrt{y}\right)$	$\frac{3}{2}-\frac{45}{4}C^*_{5/2}t^{1/2}$
2	$\frac{\pi}{2}y^{-1/2}$	$\frac{10}{3}C_{5/3}t^{-1/3}$	$\frac{1}{2}\ln(1/y)$	$\frac{1}{2}\ln(1/t)$
1	$\frac{1}{4y}\ln(y^3/t^2)$	$\frac{4}{3}C_{4/3}t^{-2/3}$	$\frac{\pi}{4}y^{-1/2}$	$\frac{9}{4}C_{3/2}t^{-1/2}$

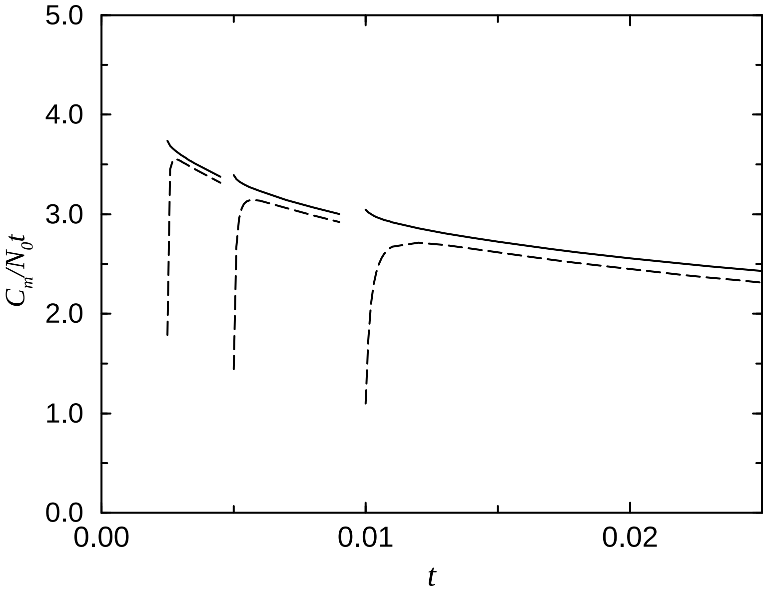

Figure 1. Temperature dependence of the specific heat C_m/N_0t for itinerant weak ferromagnets for $t_c = 0.025$, 0.05 and 0.1 from the left, respectively. Dashed curves represent results of the SCR theory (A3) in the appendix for comparison.

its temperature dependence will change into the low temperature behaviour around the crossover temperature, $y \sim t^{2/(2+\alpha)}$, if no magnetic transition takes place. The temperature dependence of the specific heat in both the temperature regions is summarized in table 3.

We show in figures 1 and 2 numerical results of the temperature dependence of specific heats for F and AF cases, respectively. For comparison, the SCR results are also shown in the same figures. In the case of exchange enhanced paramagnets, the differences are in general small at low temperature.

4. Maxwell relation

To test the validity and the consistency of the formalism, the Maxwell relation is studied here. For the simplicity of the argument, the present discussion is confined in the paramagnetic phase. In our present units, the free energy variation is represented by

$$\mathrm{d}F_m(\sigma, t) = -T_0 S_m\,\mathrm{d}t + N_0 h\,\mathrm{d}\sigma/2.$$

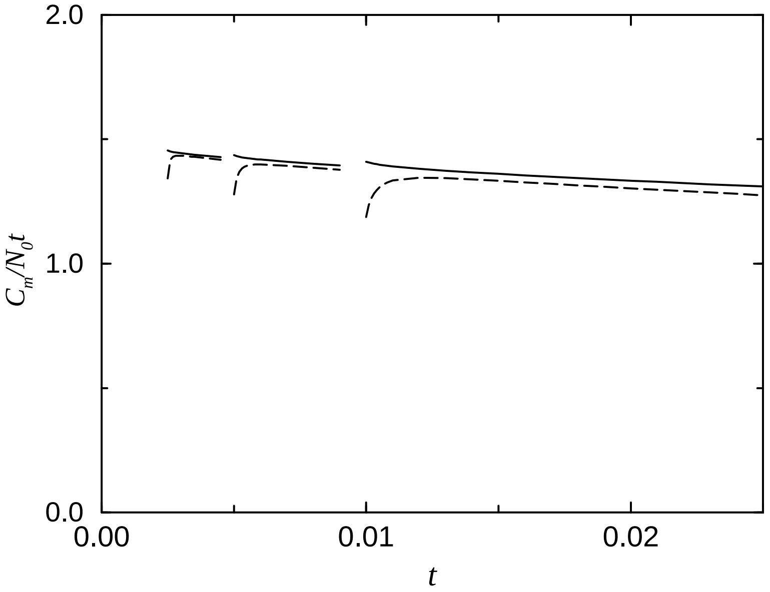

Figure 2. Temperature dependence of the specific heat C_m/N_0t for itinerant weak antiferromagnets for $t_c = 0.01$, 0.05 and 0.1 from the left. Dashed curves represent the SCR results.

The Maxwell relation is stated as follows:

$$-\frac{\partial}{\partial\sigma}\frac{S_m}{N_0} = \frac{T_A}{2T_0}\sigma\frac{\partial y}{\partial t}. \tag{20}$$

To begin with, let us recall our sum rule. In the presence of the static magnetic moment, it is represented by

$$\langle S_i^2\rangle = \frac{\sigma^2}{4} + d\frac{T_0}{T_A}[2B(y,t) + B(y_z,t)]$$
$$B(y,t) = A(y,t) - c_Z y$$

where the function $A(y,t)$ represents the thermal amplitude defined in (3). By differentiating the sum rule by t, the derivative $\partial y/\partial t$ is given by

$$B'(y,t)\frac{\partial y}{\partial t} = C(y,t)$$
$$C(y,t) = \frac{1}{t}\int_0^1 \mathrm{d}x x^{d-1+\alpha}u[1/u + 1/2u^2 - \psi'(u)]. \tag{21}$$

On the other hand, from the derivative of the entropy (14) in σ we obtain

$$\frac{\partial}{\partial\sigma}\frac{S_m}{N_0} = d\left[2C(y,t)\frac{\partial y}{\partial\sigma} + C(y_z,t)\frac{\partial y_z}{\partial\sigma}\right] = dC(y,t)\left(2\frac{\partial y}{\partial\sigma} + \frac{\partial y_z}{\partial\sigma}\right). \tag{22}$$

With the use of the σ derivative of the sum rule, the σ derivatives of y and y_z can be related to $B'(y,t)$ as follows.

$$2B'(y,t)\frac{\partial y}{\partial\sigma} + B'(y_z,t)\frac{\partial y_z}{\partial\sigma} = B'(y,t)\left(2\frac{\partial y}{\partial\sigma} + \frac{\partial y_z}{\partial\sigma}\right) = -\frac{\sigma T_A}{2dT_0}. \tag{23}$$

If we substitute (23) into (22), the σ derivative of the entropy expression is finally transformed into the form

$$\frac{\partial}{\partial\sigma}\frac{S_m}{N_0} = -\frac{\sigma T_A}{2T_0}\frac{C(y,t)}{B'(y,t)}. \tag{24}$$

From the comparison of (21) and (24) the Maxwell relation (20) is now verified.

5. Discussions

In the present paper, we have succeeded in the proper description of the magnetic specific heat of itinerant electron magnets based on the spin fluctuation mechanism. The specific heat formula is derived consistent with our sum rule of the total spin fluctuation amplitude. The result is slightly different from the expression of the SCR theory. Because of the disappearance of the terms proportional to $(\partial y/\partial t)^2$ and $\partial^2 y/\partial t^2$, the spurious dip structure of the SCR theory just above the t_c is absent in our framework. Otherwise, numerical differences between the two expressions are not very significant.

In our derivation we explicitly take account the effect of quantum zero-point spin fluctuations. Although the final result (15) does not seem to contain the effect, its neglect from the beginning is of course not justified. Ishigaki and Moriya (1998) have recently 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 effect has yet been presented.

The significant point of the present derivation is that both the temperature dependence of the magnetic susceptibility and the specific heat are derived from the same form of the free energy expression with the use of the extremum condition. It also plays a significant role in verifying the Maxwell relation between the entropy and the magnetic susceptibility. If there were terms proportional to $(\partial y/\partial t)^2$ and $\partial^2 y/\partial t^2$ in the specific heat, it would be very difficult to prove the relation or it would not be possible. The specific heat formula in the SCR theory is, in this sense, not consistent with its formula for the temperature dependence of the magnetic susceptibility. The explicit form of the free energy proposed in this study will be helpful in our future studies of magnetic properties of itinerant electron systems.

Appendix. Specific heat in SCR theory

For the comparison we show below the derivation of the specific heat in the SCR theory. In the SCR theory, the following form of the free energy is assumed.

$$F_{\mathrm{SCR}} = 3T\sum_q \int_0^\infty \frac{\mathrm{d}\omega}{\pi}\ln(1-\mathrm{e}^{-\omega/T})\frac{\Gamma_q}{\Gamma_q^2+\omega^2}.$$

By differentiating the above expression with respect to the temperature T, the magnetic entropy is given by

$$S_{\mathrm{SCR}} = -\frac{\partial}{\partial T}F_{\mathrm{SCR}} = -3\sum_q \int_0^{\omega_c}\frac{\mathrm{d}\omega}{\pi}\left[\ln(1-\mathrm{e}^{-\omega/T}) - \frac{\omega}{T}n(\omega)\right]\frac{\Gamma_q}{\omega^2+\Gamma_q^2} - \frac{\partial F_{\mathrm{SCR}}}{\partial y}\frac{\partial y}{\partial T}. \tag{A1}$$

The first term represents the explicit derivative with respect to T, the same form as our (14). With the use of (11), the second term is proportional to the thermal amplitude $A(y,t)$. Therefore we obtain the following entropy expression,

$$S_{\mathrm{SCR}}/N_0 = -3d\int_0^1 \mathrm{d}x x^{d-1}[\ln\sqrt{2\pi} - u + (u-1/2)\ln u - \ln\Gamma(u)] \\ +3d\int_0^1 \mathrm{d}x x^{d-1} u\left[\ln u - \frac{1}{2u} - \psi(u)\right] - 3dA(y,t)\frac{\partial y}{\partial t}. \tag{A2}$$

The specific heat is now obtained by differentiating (A2) with respect to the temperature T: i.e. it is given by

$$C_{\mathrm{SCR}}/N_0 t = \frac{3d}{t}\int_0^1 \mathrm{d}x x^{d-1}\left(u - x^{\alpha}\frac{\partial y}{\partial t}\right)u\{-1/u - 1/2u^2 + \psi'(u)\} \\ -\frac{3d}{t}\frac{\partial y}{\partial t}\int_0^1 \mathrm{d}x x^{d}\left(u - x^{\alpha}\frac{\partial y}{\partial t}\right)\{-1/u - 1/2u^2 + \psi'(u)\} - 3d\frac{\partial^2 y}{\partial t^2}A(y,t) \\ = 3d\left\{I_1 - \frac{\partial y}{\partial t}\frac{\partial A(y,t)}{\partial t}\right\} - 3d\frac{\partial y}{\partial t}\left\{\frac{\partial A(y,t)}{\partial t} - \frac{\partial y}{\partial t}\frac{\partial A(y,t)}{\partial y}\right\} - 3d\frac{\partial^2 y}{\partial t^2}A(y,t) \tag{A3}$$

that differs from our expression by the additional last two terms.

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