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Magnetovolume effect of itinerant electron ferromagnets

Yoshinori Takahashi and Hiroki Nakano

Graduate School of Science, University of Hyogo, Kouto 3-2-1, Kamigori, Ako-gun, 678-1297, Japan

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Abstract
The magnetovolume effect of itinerant electron magnets is discussed by the explicit examination of the volume dependence of the spin fluctuation free energy. Magnetic Grüneisen parameters are introduced instead of magnetovolume coupling constants, that enable us to describe all the magnetovolume properties in terms of these parameters. We have particularly found the presence of a new thermal expansion, showing T^2-like temperature dependence. It explains the appreciable electronic volume thermal expansions observed experimentally. We also show that the magnetovolume coupling constants for spontaneous and forced magnetostrictions, of different magnitudes, are temperature dependent. Analysis of the pressure effect on the Curie temperature T_c and the spontaneous magnetic moment M shows that the linear relation between them, $dT_c/dp \propto dM/dp$, is generally violated.

1. Introduction

There has already been a long history of intensive experimental and theoretical investigations on the pressure effect of magnetic materials. This is partly because the effect is closely related to the well known invar effect, that already has a wide area of technical applications. Due to the recent development of high pressure techniques one can now easily perform the various measurements under high pressure and get reliable experimental data. These bring us a surge of substantial interest in the pressure effects on various magnetic properties.

It seems, however, that the theoretical basis of the pressure effects has not yet been established very well. Until around 1980, magnetovolume effects of itinerant electron magnets were mainly analysed by Stoner–Edwards–Wohlfarth (SEW) theory (Wohlfarth 1969, 1977, Franse 1977, 1979). They are described by the free energy expanded in powers of the uniform magnetization M,

$$F(M, T, \omega) = F(0, T, \omega) + \frac{\Omega}{2\kappa}\omega^2 + \frac{1}{2}a(T, \omega)M^2 + \frac{1}{4}b(T, \omega)M^4 + \cdots, \quad (1)$$

where Ω, $\omega = \delta\Omega/\Omega$, and κ represent a reference volume, a volume strain, and the compressibility, respectively. The volume expansion is found by minimizing the free energy.

For instance, from the strain variation of the free energy (1) the induced volume expansion is expressed in the form

$$\omega = \frac{\kappa C}{\Omega} M_0^2(T) + \frac{\kappa C}{\Omega}[M^2 - M_0^2(T)], \qquad C = -\frac{1}{2}\frac{\partial a(T,\omega)}{\partial \omega}, \tag{2}$$

where the first and second terms represent spontaneous and forced magnetostrictions, respectively, and $M_0(T)$ and C are the spontaneous magnetization and the magnetovolume coupling constant. As far as the volume effect is concerned, the theory is consistent because all the magnetic properties including volume effects are derived by the same free energy. The theory, however, was incapable of explaining the Curie–Weiss law behaviour of the magnetic susceptibility observed even for very weak itinerant electron ferromagnets. The effects of thermal spin fluctuations are therefore invoked to resolve the difficulty. According to the self-consistent renormalization (SCR) spin fluctuation theory (Moriya 1985), the Curie–Weiss law dependence is now realized as resulting from the effect of non-linear mode–mode couplings among spin fluctuation modes. Particular attention of theoretical studies is focused on the effect of the second coefficient a in (1). The effects on higher order coefficients, b for example, have not been so crucial.

Despite its pronounced success, spin fluctuation theories have some drawbacks. Rotationally invariant treatments in spin space do always predict a first order phase transition and hence the temperature dependence of the spontaneous magnetic moment shows a discontinuous change at the critical temperature. The difficulty has already been solved by Takahashi (1986, 2001) by taking explicit account of the effect of zero-point spin fluctuations. For this purpose he introduced a new idea of spin amplitude conservation (Nakano and Takahashi 2004). He has also succeeded in deriving the first order differential equation that determines the magnetic isotherm of the system. This implies that the M dependence of the free energy can be determined as its solution. There is thus no need to assume an expansion like (1) from the start. Many qualitative and quantitative new fascinating results are derived in subsequent studies by Takahashi (1990, 1992, 1994, 1997a, 1997b, 1998, 1999, 2001), Takahashi and Sakai (1995, 1998), Takahashi and Nakano (2004), and confirmed by experiments (Yoshimura *et al* 1987, 1988, Shimizu *et al* 1990, Nakabayashi *et al* 1992, Fujita *et al* 1995, Koyama *et al* 2000a, 2003). To avoid the fictitious first order transition, one has to accept that the magnetization curve is affected as a whole by the effect of spin fluctuations. The fourth order coefficient b is of course no exception.

The beginning of spin fluctuation theories of magnetovolume effects goes back to Moriya and Usami (1980). According to their arguments, (2) has to be modified by

$$\omega = \frac{\kappa C}{\Omega}[M_0^2(T) + \xi^2(T)], \qquad \xi^2(T) = \sum_{\mathbf{q}} \langle \delta\mathbf{M}_q \cdot \delta\mathbf{M}_{-q} \rangle, \tag{3}$$

because of the presence of the thermal spin fluctuation amplitude squared $\xi^2(T)$. They also introduced a parameter η by

$$\eta(T_c) = \xi^2(T_c)/M_0^2(0), \tag{4}$$

as the ratio of the mean squared local spin amplitude at T_c to the same value at $T = 0$ K. The volume expansion in the ground state is then given by

$$\omega(0) - \omega(T_c) = \frac{\kappa C}{\Omega} M_0^2(0)(1-\eta) = \frac{2}{5}\frac{\kappa C}{\Omega} M_0^2(0),$$

relative to the volume reference at T_c, in contrast to $\kappa C M_0^2(0)/\Omega$ by the SEW theory. As an approach to extend the Moriya and Usami (MU) theory and to deal with spin fluctuations of finite amplitude, the temperature dependence of the spontaneous magnetostriction was calculated by Hasegawa (1981). Within the static single-site approximation, the tight-binding Hubbard model was treated based on the functional integral method, assuming $\Omega^{-5/3}$ volume dependence of the d-band width (Heine 1967). Similar calculations were also reported by Kakahashi (1981) with the use of the Liberman and Pettifor virial theorem (Pettifor 1978).

If we take into account the effect of zero-point spin fluctuations, it seems to be difficult to explain the magnetovolume effect. Based on an extended MU free energy that includes zero-point fluctuations, Takahashi (1990), however, claimed that the wavevector dependence of the magnetovolume coupling constant will produce the effect, even if we assume the spin amplitude constraint. He has found that the same reduction of the magnetostriction as given by the MU theory rather comes from the difference in spontaneous and forced magnetovolume coupling constants. Later the influence of the anharmonicity of zero-point spin fluctuations on the magnetovolume effect was also treated by Solontsov and Wagner (1995).

In spite of these theoretical efforts, there still remain several difficulties involved in magnetovolume properties. Some of them are listed below.

- The magnetovolume coupling constant includes a slight T^2-linear dependence. Kortekaas and Franse (1976) reported that the forced magnetovolume coupling constants of Ni_3Al and Ni–Pt alloys obey the T^2 dependence at low temperatures as predicted by Wohlfarth (1969) and Mills (1971). It is not so clear whether we should associate this behaviour with the T dependence coming from the Fermi distribution function.
- The aim of the spin fluctuation theories has been particularly focused on the thermal expansion. The forced magnetostriction is not discussed so well. Though Takahashi (1990) argued that the forced striction is proportional to M^4 at the critical point instead of M^2, the effect of spin fluctuations on the forced striction has to be elaborated to the same extent as the spontaneous volume striction.
- As comments on the MU theory, Wohlfarth (1980) insisted on the applicability of their theory in view of the presence of the T^2-linear term in the volume thermal expansion at low temperatures. No convincing explanation for this term has yet been presented. If it is analysed as the electronic Grüneisen parameters, anomalously large values are obtained (Wohlfarth 1980). The origin of the large T^2-dependent term has to be clarified.
- In the analysis of experiments, Grüneisen parameters are sometimes introduced. Underlying these is a belief in the close relationship between the specific heat and the thermal expansion coefficient. Not so much theoretical attention has been paid to this issue to date except for the review by Kakahashi (1989), who showed the presence of the term proportional to the specific heat in the insulator limit of the degenerate Hubbard model.
- The pressure dependence of T_c and $M_0(0)$ is usually analysed based on either of the following relations:

$$\frac{\mathrm{d}\ln T_c}{\mathrm{d}p} = \frac{\mathrm{d}\ln M_0(0)}{\mathrm{d}p}, \qquad \frac{\mathrm{d}\ln T_c}{\mathrm{d}p} = \frac{3}{2}\frac{\mathrm{d}\ln M_0(0)}{\mathrm{d}p},$$

  as predicted by the SEW theory and the SCR theory. The observed dependence is not so consistently well fitted with either of the above relations. The relation between pressure effects on T_c and the spontaneous magnetic moment has not yet been studied very well.

It is important to note that the MU theory is not fully self-consistent as far as the magnetovolume effect is concerned. Their magnetostriction is *not* derived from their free energy expression. The purpose of this paper is to discuss magnetovolume effects from a different viewpoint, paying particular attention to the effect of spin fluctuations, and to explain the above mentioned unresolved properties. In this paper we deal with the explicit volume dependence of the free energy due to the collective magnetic excitation modes. From the logical consistency of the theory, such an approach is highly necessary. It will also allow us to define magnetic Grüneisen parameters.

In the next section, from the explicit volume dependence of the spin fluctuation free energy, general formulae are derived for spontaneous and forced magnetostrictions. Magnetic Grüneisen parameters are also introduced as the volume derivatives of characteristic parameters of the free energy. Succeeding sections 3 and 4 are devoted to discussions on the effects in the ordered and paramagnetic phases, respectively. In section 5, the critical forced magnetostriction and the pressure effect on T_c are discussed. Through the present paper, the uniform magnetization M is expressed in terms of the dimensionless parameter σ in units of Bohr magnetons μ_B per magnetic atom and the uniform external magnetic 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 component of the dynamical magnetic susceptibility $\chi(q, \nu)$ measured in units of $4\mu_B^2$ is in the present units given by

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

2. Volume dependence of the free energy

Thermal expansion of the crystal due to the lattice vibration results from the anharmonicity of the elastic energy. It is equivalent to the assumption of the volume-dependent phonon frequencies. In the Debye model of lattice vibrations, it is known that the *volume* thermal expansion coefficient $\beta_{ph}(T)$ is proportional to the lattice specific heat per unit volume as given by

$$\beta_{ph}(T) = \gamma_{ph}\kappa c_v(T). \tag{5}$$

Their ratio γ_{ph} is the phonon Grüneisen parameter defined from the strain (i.e., $\omega = \delta\Omega/\Omega$) derivative of the Debye temperature Θ_D by

$$\gamma_{ph} = -\frac{\partial \ln \Theta_D}{\partial \omega}.$$

The parameter Θ_D represents a characteristic scale of the phonon free energy. By finding out the explicit deviation of the phonon free energy due to the strain through the ω-derivative of Θ_D, we can obtain the relation (5).

What we do in this section is to follow the same argument as above to obtain the magnetovolume effects that originate from the strain (i.e. volume) dependence of the magnetic free energy consisting of collective magnetic excitation energies of the crystal. Such an approach will also allow us to introduce *magnetic* Grüneisen parameters most naturally by the strain derivatives of characteristic parameters of the magnetic free energy. Let us now start from the following free energy expression:

$$F_m = \frac{1}{\pi}\sum_{\mathbf{q}}\int_0^{\nu_c} d\nu \left[\frac{\nu}{2} + T\ln(1 - e^{-\nu/T})\right]\left\{2\frac{\Gamma_q}{\nu^2 + \Gamma_q^2} + \frac{\Gamma_q^z}{\nu^2 + (\Gamma_q^z)^2}\right\}$$
$$+ N_0 T_A \left\{y\sigma^2/4 - (y + \Delta y_z/3)\left\langle \mathbf{S}_i^2\right\rangle_{\rm tot}\right\} + \Delta F_1(\sigma, t) \tag{6}$$

where ν_c is the upper bound of the frequency integral. The free energy (6) was proposed by us in our previous study of the magnetic entropy and the specific heat (Takahashi and Nakano 2004). The extended MU free energy, on the other hand, was employed by the previous study (Takahashi 1990). The first term comes from the collective spin fluctuation modes with transverse and longitudinal polarization to the induced moment in the z-direction. The parameters, Γ_q and Γ_q^z, denote the wavevector dependent damping constants. Throughout the paper, the magnetic moment and the applied magnetic field are assumed to be along the z-axis. The subscript α (superscript for Γ) for the parallel modes is, therefore, denoted by y_z for instance, while those for transverse modes are suppressed. The second term represents the sum of the Zeeman energy and the correction necessary for the spin-amplitude conservation. The last term ΔF_1 is an additional correction introduced for the description of the ordered state.

The above free energy assumes the presence of exchange-enhanced spin fluctuation modes. The wavevector dependent frequency spectrum for each α-mode is given in terms of the imaginary part of the dynamical magnetic susceptibility of the double-Lorentzian form,

$$\begin{aligned} \mathrm{Im}\,\chi_\alpha(q,\nu) &= \chi_\alpha(q,0)\frac{\nu\Gamma_q^\alpha}{\nu^2+\Gamma_q^{\alpha 2}}, \\ \chi_\alpha(q,0) &= \frac{\chi_\alpha(0,0)}{1+q^2/K_\alpha^2}, \qquad \Gamma_q^\alpha = \Gamma_0 q(K_\alpha^2+q^2), \end{aligned} \tag{7}$$

where K_α^2 is the squared inverse correlation length proportional to $\chi_\alpha^{-1}(0,0)$. For later convenience two energy (temperature) scales T_0 and T_A are introduced by

$$T_0 = \Gamma_0 q_{\mathrm{B}}^3/2\pi, \qquad T_A = N_0 q_{\mathrm{B}}^2/2\chi_\alpha(0,0)K_\alpha^2. \tag{8}$$

The zone-boundary wavevector is denoted by q_{B}. These are measures of distributions of the wavevector dependent damping Γ_q^α and the static inverse susceptibility $\chi(q,0)^{-1}$ in the wavevector space. They correspond to the Debye temperature of lattice vibrations or the exchange constant of the Heisenberg magnets. The wavevector dependence of $\chi_\alpha(q,0)$ and Γ_q^α is then written by

$$\chi_\alpha(q,0) = \frac{N_0}{2T_A}\frac{1}{y_\alpha+x^2}, \qquad \Gamma_q^\alpha = 2\pi T_0 x(y_\alpha+x^2), \tag{9}$$

where x is a reduced wavevector defined as a ratio q/q_{B}. The parameter, y_α, stands for the reduced inverse magnetic susceptibility, i.e. the second order derivatives of the free energy with respect to the parallel and perpendicular moment $\delta M_\parallel$ and $M_\perp$, defined by

$$T_A y_z = \frac{N_0}{2\chi_\parallel(0,0)} = \frac{\partial h}{\partial\sigma} \propto \frac{\partial^2 F}{\partial\delta M_\parallel^2}, \qquad T_A y = \frac{N_0}{2\chi_\perp(0,0)} = \frac{h}{\sigma} \propto \frac{\partial^2 F}{\partial M_\perp^2}. \tag{10}$$

In the presence of static induced magnetic moment, the spin fluctuation amplitudes become anisotropic. The effect is included in terms of reduced anisotropic inverse susceptibilities, y and y_z.

The free energy (6) is a function of its arguments, σ, T, y, and y_z. The temperature is, hereafter, measured in reduced units, $t = T/T_0$. We assume that the equilibrium values of these variables are always determined by the following stability conditions of the free energy.

$$\begin{aligned} \frac{\partial F_m}{\partial y} &= N_0 T_A\left\{\sum_\alpha \left\langle \delta S_{\alpha i}^2\right\rangle_T (y_\alpha,t) + \sum_\alpha \left\langle \delta S_{\alpha i}^2\right\rangle_Z (y_\alpha) + \frac{\sigma^2}{4} - \left\langle \mathbf{S}_i^2\right\rangle_{\mathrm{tot}}\right\} = 0, \\ \frac{\partial F_m}{\partial\Delta y_z} &= N_0 T_A\left\{\left\langle \delta S_{zi}^2\right\rangle_T (y_z,t) + \frac{1}{3}\left[\sum_\alpha \left\langle \delta S_{\alpha i}^2\right\rangle_Z (y_\alpha) - \left\langle \mathbf{S}_i^2\right\rangle_{\mathrm{tot}}\right] - \lambda(\sigma,t)\right\} = 0, \\ \frac{\partial F_m}{\partial\sigma} &= \frac{1}{2}N_0 T_A y\sigma = \frac{1}{2}N_0 h. \end{aligned} \tag{11}$$

The parameter λ in the second line is related to the derivative of the correction ΔF_1 with respect to σ. The amplitudes of local spin fluctuations in the right-hand side of (11) are evaluated by the wavevector and frequency integrals of the imaginary part of the dynamical magnetic susceptibility. Their thermal and zero-point components are, respectively, given by

$$\begin{aligned}
\langle\delta S_{\alpha i}^2\rangle_T(y_\alpha,t) &= \frac{2}{N_0^2}\sum_q\int_0^\infty\frac{\mathrm{d}\nu}{\pi}n(\nu)\,\mathrm{Im}\,\chi_\alpha(q,\nu) = \frac{3T_0}{T_A}A(y_\alpha,t),\\
A(y_\alpha,t) &= \int_0^1\mathrm{d}x\,x^3[\ln u-1/2u-\psi(u)], \qquad u=x(y_\alpha+x^2)/t \qquad (12)\\
\langle\delta S_{\alpha i}^2\rangle_Z(y_\alpha) &= \frac{1}{N_0^2}\sum_q\int_0^\infty\frac{\mathrm{d}\nu}{\pi}\,\mathrm{Im}\,\chi_\alpha(q,\nu) = \frac{1}{3}\langle\mathbf{S}_i^2\rangle_Z(0,0)-\frac{3T_0}{T_A}c_z y_\alpha+\cdots,
\end{aligned}$$

where c_z is a numerical constant of order unity. The digamma function denoted by $\psi(u)$ is defined by the integral

$$\int_0^\infty \mathrm{d}\nu\frac{\nu}{\mathrm{e}^{\beta\nu}-1}\frac{1}{\nu^2+\Gamma^2} = \frac{1}{2}[\log u-1/2u-\psi(u)], \qquad \Gamma=2\pi Tu. \quad (13)$$

The first line of (11) shows the spin amplitude conservation. We have already shown that many magnetic properties can be deduced with the use of this condition, such as magnetic isotherms, the temperature dependence of the magnetic susceptibility in the paramagnetic phase, and the spontaneous moments in the ordered state (Takahashi 1986, 2001). The last line is nothing but the thermodynamic relation, $H=\partial F_m/\partial M$, in our reduced units. In the following discussions, we assume that all the above conditions in (11) are always satisfied adiabatically for each value of the volume strain of the crystal.

2.1. *Explicit free energy deviation due to the volume strain*

In order to obtain magnetovolume effects, we need the explicit volume dependence of the free energy. The direct volume dependence was discussed by Edwards and Macdonald (1983) based on the free energy of Moriya and Kawabata (1973a, 1973b) that lacks the rotational invariance in spin space. They assumed a single energy band for conduction electrons with parabolic energy dispersion and the volume dependence is included by the band width proportional to $\Omega^{-3/5}$ (Heine 1967). From the calculated thermal expansion, they showed $\eta(T_\mathrm{c})>1$, i.e. no volume expansion below T_c, in contradiction to the frequently observed invar effects. Although we employ the same idea, our free energy is different and the way to introduce the volume dependence is different as well.

Under the isothermal condition, let us now define a free energy variation in the presence of the volume strain by

$$\delta' F_m(y,y_z,\sigma,t,\omega) \equiv \frac{\partial F_m(y,y_z,\sigma,t,\omega)}{\partial\omega}\delta\omega. \quad (14)$$

It is evaluated, for instance, by assuming that spectral widths in (9) show deviations given by

$$\delta'\Gamma_q = 2\pi\delta T_0 x(y+x^2) = \frac{\delta T_0}{T_0}\Gamma_q, \qquad \delta'\Gamma_q^z = \frac{\delta T_0}{T_0}\Gamma_q^z.$$

Be careful that the implicit dependence through the changes, δy and δy_z, is neglected because of the stability conditions (11). In this way the free energy variation is written as follows.

$$\begin{aligned}
\delta' F_m &= \delta' F_s + \delta' F_h,\\
\delta' F_s &= \frac{\delta T_0}{T_0}\sum_{\mathbf{q}}\int_0^{\nu_c}\frac{\mathrm{d}\nu}{\pi}\left[\frac{\nu}{2}+T\ln(1-\mathrm{e}^{-\nu/T})\right]\\
&\quad\times\left\{2\Gamma_q\frac{\partial}{\partial\Gamma_q}\left(\frac{\Gamma_q}{\nu^2+\Gamma_q^2}\right)+\Gamma_q^z\frac{\partial}{\partial\Gamma_q^z}\left(\frac{\Gamma_q^z}{\nu^2+(\Gamma_q^z)^2}\right)\right\}+\cdots. \qquad (15)
\end{aligned}$$

The first and second terms in the first line are defined as components that produce spontaneous and forced strictions depending on the absence and the presence of the external magnetic field, respectively. The first term $\delta' F_s$ is discussed in further detail just below. The second term $\delta' F_h$ is treated later in section 2.5.

2.2. *Spontaneous magnetostriction*

The spontaneous magnetostriction emerges from the first term of the free energy variation $\delta' F_s$ in (15). It is further divided into the two contributions

$$\delta' F_s = \delta' F_0 + \delta' F_t,$$

$$\delta' F_t = \frac{\delta T_0}{T_0} \sum_{\mathbf{q}} \int_0^{\nu_c} \frac{d\nu}{\pi} n(\nu) \left\{ 2 \frac{\nu \Gamma_q}{\nu^2 + \Gamma_q^2} + \frac{\nu \Gamma_q^z}{\nu^2 + (\Gamma_q^z)^2} \right\} + \delta' \Delta F_t, \tag{16}$$

where we have used the following relation:

$$\frac{\partial}{\partial \Gamma_q} \left(\frac{\Gamma_q}{\nu^2 + \Gamma_q^2} \right) = -\frac{\partial}{\partial \nu} \left(\frac{\nu}{\nu^2 + \Gamma_q^2} \right).$$

It is better to call the second term $\delta' F_t$ in (16) a *thermal* magnetostriction for the presence of the T-dependent factor of Bose excitations. On the other hand, the first term will give a similar behaviour as predicted by the MU theory.

In the weak field limit, $y(\sigma, t)$ is expanded in the form

$$y(\sigma, t) = y_0(t) + y_1(t)[\sigma^2 - \sigma_0^2(t)] + \cdots$$

where $\sigma_0(t)$ and $y_1(t)$ are the spontaneous moment and the reduced fourth order expansion coefficient of the free energy, respectively. The free energy variation $\delta' F_0(y_0, \Delta y_{z0}, \omega)$ is a function of its small arguments, $y_0(t)$ and $\Delta y_{z0}(t) = 2y_1(t)\sigma_0^2(t)$, i.e. values of $y(\sigma, t)$ and $\Delta y_z(\sigma, t) = y_z(\sigma, t) - y(\sigma, t)$ at $\sigma = \sigma_0(t)$. Since this term does not contain thermal spin fluctuations, no anomalous critical behaviour is expected around its origins, $y_0 \sim 0$, $\Delta y_{z0} \sim 0$. It is hence expanded as follows.

$$\delta' F_0(y_0, \Delta y_{z0}, 0) = \delta' F_0(0, 0, 0) + \frac{\partial \delta' F_0}{\partial y_0} y_0 + \frac{\partial \delta' F_0}{\partial \Delta y_{z0}} \Delta y_{z0} + \cdots. \tag{17}$$

Exchange of the order of the y-differentiation with the variation δ' due to the strain allows us to evaluate the above coefficients explicitly, as given by

$$\frac{\partial \delta' F_0}{\partial y} = \delta' \left(\left. \frac{\partial F_0}{\partial y} \right|_{y_0=0} \right) = -N_0 \delta \left\{ T_A \Delta \left\langle \mathbf{S}_i^2 \right\rangle \right\} + \frac{1}{4} N_0 \sigma_0^2(t) \delta T_A,$$

$$\frac{\partial \delta' F_0}{\partial \Delta y_z} = \delta' \left(\left. \frac{\partial F_0}{\partial \Delta y_z} \right|_{\Delta y_z=0} \right) = \frac{1}{3} N_0 \delta \left\{ T_A \Delta \left\langle \mathbf{S}_i^2 \right\rangle \right\} + \frac{1}{12} N_0 \sigma_0^2(t) \delta T_A,$$

where we have made use of the stability conditions in (11). The squared moment σ^2 has to be treated as an independent variable. After the manipulation, it was replaced by its thermal equilibrium value $\sigma_0^2(t)$. Thermal amplitudes $A(y, t)$ and $A(y_z, t)$ do not appear in the above expressions because they are all incorporated in $\delta' F_t$. The difference of the spin amplitudes is therefore introduced by $\Delta \left\langle \mathbf{S}_i^2 \right\rangle = \left\langle \mathbf{S}_i^2 \right\rangle_{\mathrm{tot}} - \left\langle \mathbf{S}_i^2 \right\rangle_Z (0)$. Its value is related to the spontaneous moment squared or the inverse magnetic susceptibility in the ground state (Takahashi 2001) by

$$\Delta \left\langle \mathbf{S}_i^2 \right\rangle = \begin{cases} 3\sigma_s^2/20, & \text{for ferromagnets,} \\ -3y_0(0)/20y_{10}, & \text{for exchange-enhanced paramagnets,} \end{cases}$$

where $\sigma_s = \sigma_0(0)$ and $y_{10} = y_1(0) = T_A/60c_z T_0$ (see Takahashi 2001, for instance).

The free energy variation due to the volume strain is now written by

$$\begin{aligned} \delta' F_0(y_0, \Delta y_{z0}, 0) &= \delta' F_0(0,0,0) - N_0 C_0(t)\omega(3y_0 + \Delta y_{z0}) + \cdots, \\ C_0(t)\omega &= \tfrac{1}{3}\left\{\delta\left[T_A \Delta \left\langle \mathbf{S}_i^2 \right\rangle\right] - \tfrac{1}{4}\sigma_0^2(t)\delta T_A\right\}. \end{aligned} \tag{18}$$

From the minimum condition of the free energy with respect to ω, the spontaneous magnetostriction is expressed in the form

$$\omega_0(t) = \rho\kappa C_s(t) \begin{cases} \dfrac{\Delta y_{z0}(t)}{2y_1(t)}, & \text{for the ferromagnetic phase,} \\ \dfrac{y_0(t)}{y_1(t)}, & \text{for the paramagnetic phase,} \end{cases} \tag{19}$$

where $\rho = N_0/\Omega$ and $C_s(t) = 2y_1(t)C_0(t), 3y_1(t)C_0(t)$, for ferro- and paramagnetic phases.

In the presence of the spontaneous magnetic moment below T_c, $\omega_0(t)$ in (19) is given by $\rho\kappa C_s(t)\sigma_0^2(t)$, since $y_0(t) = 0$ and $\Delta y_{z0}(t) = 2y_1(t)\sigma_0^2(t)$ are satisfied. The parameter $C_s(t)$ therefore has the meaning of the magnetovolume coupling constant. Equation (19) also results in the positive volume striction proportional to the inverse magnetic susceptibility above T_c. Its expression is in agreement with the result by Takahashi (1990) except for the temperature dependence of the magnetovolume coupling constant. Though it behaves similarly with the result of the MU theory, they are not exactly the same, since the right-hand side of (19) is not proportional to the spin fluctuation amplitudes, $\xi^2(T)$, but rather to the inverse of the magnetic susceptibility, $\Delta y_{z0}(t)$ or $y_0(t)$.

2.3. *Effect of thermal spin fluctuations*

Let us next discuss the thermal magnetostriction caused by thermal spin fluctuations. The frequency integral of $\delta' F_t$ in (16) is performed with the use of the digamma function $\psi(x)$ defined in (13). The wavevector summation of this term is then written by

$$\begin{aligned} &\sum_{\mathbf{q}} \int_0^\infty \frac{d\nu}{\pi} \frac{\nu}{e^{\beta\nu} - 1} \frac{\Gamma}{\nu^2 + \Gamma^2} = 3N_0 T_0 t \int_0^1 dx\, x^2 \Phi(u), \\ &\Phi(u) = u[\log u - 1/2u - \psi(u)], \end{aligned}$$

where x is the reduced wavevector, q/q_B. The free energy variation $\delta' F_t$ is thus given by

$$\begin{aligned} &\delta' F_t = 3N_0 T_0 \frac{\delta T_0}{T_0} t \left\{ 2\int_{x_c}^1 dx x^2 \Phi(u) + \int_0^1 dx x^2 \Phi(u_z) \right\}, \\ &u = x(y_0(t) + x^2)/t, \qquad u_z = x(y_{z0}(t) + x^2)/t. \end{aligned}$$

As the consequence, the strain derivative of $\delta' F_t$ gives the thermal expansion,

$$\omega_t(t) = 3\rho\kappa T_0 \gamma_0 t \left\{ 2\int_{x_c}^1 dx\, x^2 \Phi(u) + \int_0^1 dx\, x^2 \Phi(u_z) \right\}, \tag{20}$$

where $\gamma_0 = -d \ln T_0/d\omega$. Note that the wavevector integral of the first term for transverse modes is limited by the lower bound x_c due to the spin-wave modes around the origin. The presence of this term $\omega_t(t)$ has long been disregarded in previous theoretical studies. If we start from the free energy expansion (1) following the SEW theory, such a term never appears. It results from the explicit volume dependence of the free energy (6). This will partly explain the reason for the ignorance of this term.

In the low temperature limit, the following asymptotic expansion of $\Phi(u)$ in (20) is verified:

$$\Phi(u) \simeq \frac{1}{12u}, \qquad \text{as } u \to \infty$$

leading to the thermal expansion given by

$$\omega_t(t) \simeq \frac{1}{8}\rho\kappa T_0\gamma_0 \begin{cases} t^2[2\ln x_c^{-2} + \ln(1+y_{z0}^{-1})], & \text{for } 0 < y_{z0} \ll 1 \\ 2t^2\ln(1/t), & \text{for } y_{z0} = 0 \end{cases} \tag{21}$$

as far as both $t \ll 1$ and $y_0, y_{z0} \ll 1$ are satisfied. For weak itinerant electron ferromagnets, because y_{z0} and x_c^2 are both proportional to σ_s^2, it is written in the form

$$\omega_t(t) \simeq \tfrac{3}{4}\rho\kappa T_0\gamma_0 t^2 \ln \sigma_s^{-1}. \tag{22}$$

In the case of exchange-enhanced paramagnets, it is written as

$$\omega_t(t) \simeq \tfrac{3}{8}\rho\kappa T_0\gamma_0 t^2 \ln y_0(0)^{-1}, \tag{23}$$

where $y_0(0)$ stands for the reduced reciprocal magnetic susceptibility at $T = 0$ K. From these results we easily find that the thermal magnetostriction $\omega_t(t)$ cannot be expanded in terms of its parameters, y_0 and y_{z0}, because their derivatives cannot be defined around their origins. In contrast, $\omega_0(t)$ is assumed to be expanded.

As for the correction of the thermal magnetostriction, the part of the free energy correction of ΔF_1 resulting from thermal spin fluctuations satisfies the relation (Takahashi and Nakano 2004)

$$\frac{\partial \Delta F_{1t}}{\partial \sigma} = -2N_0T_0[A(y_z,t) - A_t(y,t)]\frac{\partial \Delta y_z}{\partial \sigma}.$$

Its strain variation is given by

$$\delta'\Delta F_{1t} \simeq 2N_0T_0\Delta y_{z0}\gamma_0\delta\omega\left\{A(y_{z0},t) - A_t(0,t) - t\left[\frac{\partial A(y_{z0},t)}{\partial t} - \frac{\partial A_t(0,t)}{\partial t}\right]\right\},$$

after the succeeding σ-integration. Long-wave modes of the transverse thermal amplitude $A_t(y,t)$ have to be described by spin-wave excitations in the ordered phase (Takahashi 2001). The correction to the $\omega_t(t)$ is therefore written in the form

$$\Delta\omega_t(t) = 2\rho\kappa T_0\Delta y_{z0}\gamma_0\left\{t\left[\frac{\partial A(y_{z0},t)}{\partial t} - \frac{\partial A_t(0,t)}{\partial t}\right] - A(y_{z0},t) + A_t(0,t)\right\}. \tag{24}$$

It is well known that the T-linear coefficient of the specific heat is strongly enhanced in proportion to $\ln\sigma_s^{-1}$ for systems in the vicinity of the magnetic instability point. It is derived by the T^2-dependence of the free energy given in our notation by

$$F(T) = F(0) - \frac{3N_0T^2}{4T_0}\ln\frac{1}{\sigma_s} + \cdots. \tag{25}$$

It is also easy to find its strain variation,

$$\delta' F(T) = -\frac{3N_0}{4}T_0\gamma_0 t^2\omega\ln\frac{1}{\sigma_s},$$

consistent with the thermal expansion, (22). Both the thermal expansion (22) and the enhanced T-linear specific heat coefficient result from the same free energy expression. The linear relation between them like (5) is therefore satisfied. The presence of $\omega_t(t)$ is obvious from this argument as far as γ_0 is appreciable. Later we show the coefficient γ_0 cannot be neglected based on the analysis of experiments.

At the critical temperature $T = T_c$, the thermal magnetostriction is given by

$$\omega_t(t_c) = 3\rho\kappa T_0\gamma_0 t_c^2\int_0^{1/t_c} dv\,\Phi(v) \simeq \tfrac{1}{4}\rho\kappa T_0\gamma_0 t_c^2 \ln(1/t_c), \qquad (t_c \ll 1) \tag{26}$$

where $v = x^3/t$. In the paramagnetic state we obtain

$$\omega_t(t) \simeq \tfrac{3}{8}\rho\kappa T_0\gamma_0 t^2 \ln y_0^{-1}(t),$$

as far as the condition $y_0(t) \ll 1$ is satisfied. The thermal magnetostriction derived above gives rise to the T^2-like temperature dependence because its coefficient shows only very weak logarithmic dependence. After subtraction of the spontaneous magnetostriction $\omega_0(t) \propto \sigma_0^2(t)$ from the observed volume change of the crystal, the thermal expansion is usually analysed according to the following sum of the T-dependence at low temperatures:

$$\Delta L/L = AT^2 + BT^4,$$

where the first and second terms are supposed to originate from the electronic and the phonon contributions. Wohlfarth (1980) argued that the presence of the significant T^2 dependence in the thermal expansion would demonstrate that a part of the first term is attributable to the SEW mechanism. Our present result suggests that the T^2-linear electronic volume expansion will rather result from the thermal magnetostriction $\omega_t(t)$ due to thermal spin fluctuations.

2.4. Magnetic Grüneisen parameters

The free energy variation in (6) in the presence of the volume strain can be described by the following three parameters.

$$\frac{\mathrm{d}\Delta\left\langle \mathbf{S}_i^2\right\rangle}{\mathrm{d}\omega} = \gamma_m \left|\Delta\left\langle \mathbf{S}_i^2\right\rangle_0\right|, \qquad \gamma_A = -\frac{\mathrm{d}\ln T_A}{\mathrm{d}\omega}, \qquad \gamma_0 = -\frac{\mathrm{d}\ln T_0}{\mathrm{d}\omega}. \tag{27}$$

The parameter γ_0 is already introduced in (20). The temperature scales, T_0 and T_A, defined in (8) represent the spectral widths of the spin fluctuation spectra in frequency and wavevector spaces. These two parameters γ_0 and γ_A correspond to the Grüneisen parameter γ_{ph} for lattice vibrations. They are also in accordance with the usual definition by the negative of the logarithmic derivatives of the characteristic energies of the system with respect to the strain (Fawcett 1989). The parameter γ_m is defined as the slope of the spin amplitude squared at some reference volume Ω_0.

$$\Delta\left\langle \mathbf{S}_i^2\right\rangle = \Delta\left\langle \mathbf{S}_i^2\right\rangle_0 (1 + \gamma_m\omega) = \Delta\left\langle \mathbf{S}_i^2\right\rangle_0 (1 - \kappa\gamma_m p).$$

It implies that $\Delta\left\langle \mathbf{S}_i^2\right\rangle$ is not strictly constant, but will show a slight temperature dependence through that of ω. The difference $\Delta\left\langle \mathbf{S}_i^2\right\rangle_0$ stands for its value at $\Omega = \Omega_0$. Note that the $\Delta\left\langle \mathbf{S}_i^2\right\rangle$ will vanish at the magnetic instability point as a function of the strain. The reason for the above definition is to make it possible that the value $\Delta\left\langle \mathbf{S}_i^2\right\rangle$ will change its sign at some critical strain $\omega = -1/\gamma_m$. It is also expressed in the form

$$\frac{\mathrm{d}\Delta\left\langle \mathbf{S}_i^2\right\rangle}{\mathrm{d}\omega} = \frac{3}{20}\sigma_{s0}^2\gamma_m, \qquad \frac{\mathrm{d}\Delta\left\langle \mathbf{S}_i^2\right\rangle}{\mathrm{d}\omega} = \frac{3y_0(0)}{20y_{10}}\gamma_m, \tag{28}$$

for ferromagnets and exchange-enhanced paramagnets, respectively.

With these parameters, the coupling constant $C_0(t)$ for ferromagnets is represented by

$$\begin{aligned} C_0(t) &= \tfrac{1}{3}\left[-\gamma_A T_A \Delta\left\langle \mathbf{S}^2\right\rangle + \gamma_m T_A \Delta\left\langle \mathbf{S}^2\right\rangle_0 + \tfrac{1}{4} T_A \gamma_A \sigma_0^2(t)\right] \\ &= \frac{C_{h0}}{5y_{10}}\left[1 - \frac{\gamma_A}{\gamma_m}\left(\frac{\sigma_s^2}{\sigma_{s0}^2} - \frac{5\sigma_0^2(t)}{3\sigma_{s0}^2}\right)\right]. \end{aligned} \tag{29}$$

where C_{h0} has a meaning of the magnetovolume coupling constant for forced strictions defined by

$$C_{h0} = \tfrac{1}{4} T_A y_{10} \sigma_{s0}^2 \gamma_m. \tag{30}$$

From ratios of the above Grüneisen parameters, let us define additional parameters by

$$g_0 = \frac{\gamma_0}{\gamma_m}, \qquad g_A = \frac{\gamma_A}{\gamma_m}.$$

In view of the analogy with the thermal expansion due to lattice vibrations, it is reasonable to call γ_m, γ_0, and γ_A magnetic Grüneisen parameters for itinerant electron magnets. Among them the parameter γ_m is specific to itinerant magnets. It is determined by the volume dependence of $\Delta\langle\mathbf{S}_i^2\rangle = \langle\mathbf{S}_i^2\rangle_{\rm tot} - \langle\mathbf{S}_i^2\rangle_Z(0)$. Its value is also not influenced by the ω-dependence of the total spin amplitude but by the zero-point amplitude. Kambe *et al* (1997) introduced a Grüneisen parameter to the SCR model in their analysis of the heavy fermion $Ce_{1-x}La_xRu_2Si_2$ compounds. Their definition is, however, different from ours.

To conclude, the spontaneous magnetostriction consists of the sum of two contributions,

$$\omega_s(t) = \omega_0(t) + \omega_t(t). \tag{31}$$

The first term corresponds to the conventional expression of the spontaneous magnetostriction (2), for instance by the SEW theory. The magnetovolume coupling constant $C_s(t)$ is only defined for $\omega_0(t)$. The second term is the thermal magnetostriction derived in this study for the first time.

2.5. Forced magnetostriction and Maxwell relation

Forced magnetostriction is caused from the second term $\delta' F_h$ of the free energy variation in (15). We can derive the general formula for the forced magnetostriction by making use of the Maxwell relation of the thermodynamics. In our reduced units, the total differential of the free energy as a function of the magnetization σ and the pressure p is given by

$$\mathrm{d}F(\sigma, p) = \Omega\omega\,\mathrm{d}p + \tfrac{1}{2}N_0 h\,\mathrm{d}\sigma. \tag{32}$$

This is equivalent to the following Maxwell relation:

$$\Omega\frac{\partial\omega}{\partial\sigma} = \frac{1}{2}N_0\frac{\partial h}{\partial p} = \frac{N_0\sigma}{2}\frac{\partial(T_A y)}{\partial\omega}\frac{\partial\omega}{\partial p} = -\frac{N_0\kappa\sigma}{2}\frac{\partial(T_A y)}{\partial\omega}.$$

That is, the forced magnetostriction is evaluated by solving the differential equation,

$$\frac{\partial\omega}{\partial\sigma} = -\frac{1}{2}\rho\kappa\sigma\frac{\partial(T_A y)}{\partial\omega}. \tag{33}$$

According to the expression of the volume strain in (2), let us define the forced magnetovolume coupling constant $C_h(\sigma, t)$ by

$$\frac{\partial\omega}{\partial\sigma} = 2\rho\kappa C_h(\sigma, t)\sigma. \tag{34}$$

The comparison of (33) and (34) leads to the general expression of the coupling constant $C_h(\sigma, t)$,

$$C_h(\sigma, t) = -\frac{1}{4}\frac{\partial(T_A y)}{\partial\omega} = \frac{T_A}{4}\left[\gamma_A y(\sigma, t) - \frac{\partial y(\sigma, t)}{\partial\omega}\right]. \tag{35}$$

We can thus obtain the forced magnetostriction by the integration of (34) with respect to σ. We will later show how to evaluate the strain derivative, $\partial y(\sigma, t)/\partial\omega$.

2.6. Magnetovolume effect in the ground state

In preceding sections, we have obtained general expressions for spontaneous and forced magnetostrictions. Before going into detailed discussions, it is better to explain how to apply them on a very simple example and to find various pressure effects. Therefore let us study the magnetovolume effect in the ground state. Inverses of magnetic susceptibilities of transverse and longitudinal components are then given, respectively, by

$$\begin{aligned} &y(\sigma, 0) = y_{10}(\sigma^2 - \sigma_s^2), \qquad y_z(\sigma, 0) = y(\sigma, 0) + 2y_{10}\sigma^2, \\ &\Delta y_z(\sigma, 0) = y_z(\sigma, 0) - y(\sigma, 0) = 2y_{10}\sigma^2 = 2y_{10}\sigma_s^2 + 2y(\sigma, 0). \end{aligned} \tag{36}$$

To begin with, from (19) and (29) the spontaneous magnetostriction is given by

$$\omega_0(0) = \rho\kappa C_{s0}\sigma_s^2, \qquad C_{s0} = \tfrac{1}{10} T_A y_{10}\sigma_{s0}^2 \left(\gamma_m + \tfrac{2}{3}\gamma_A\right). \tag{37}$$

The forced magnetostriction, on the other hand, is related to the second term $\delta' F_h$ in (15). Exchange of the order of the σ-differentiation and the variation δ' gives

$$\frac{\partial \delta' F_h}{\partial \sigma} = \delta'[T_A y(\sigma, 0)]\sigma. \tag{38}$$

From (36) the above right-hand side is evaluated as follows.

$$\begin{aligned}\delta'(T_A y) &= \delta'(T_A y_{10})(\sigma^2 - \sigma_s^2) - T_A y_{10}\sigma_{s0}^2\gamma_m\omega \\ &\simeq -T_A y_{10}\sigma_{s0}^2\gamma_m\omega \qquad (\sigma \simeq \sigma_s).\end{aligned} \tag{39}$$

After integration of (38) starting from the initial condition $\delta' F_h = 0$ for $\sigma = \sigma_s$, the variation of the free energy is given by

$$\delta' F_h = -\tfrac{1}{4} N_0 T_A y_{10}\sigma_{s0}^2\gamma_m(\sigma^2 - \sigma_s^2)\omega.$$

This leads to the forced magnetostriction,

$$\omega_h(\sigma, 0) = \rho\kappa C_{h0}(\sigma^2 - \sigma_s^2), \tag{40}$$

where C_{h0} defined in (30) is the forced magnetovolume coupling constant in the ground state. From the comparison of (37) and (40) the ratio of two magnetovolume coupling constants in the ground state is given by

$$\frac{C_{s0}}{C_{h0}} = \frac{2}{5}\left(1 + \frac{2\gamma_A}{3\gamma_m}\right) = \frac{2}{5}\left(1 + \frac{2}{3}g_A\right), \tag{41}$$

in agreement with (Takahashi 1990) when g_A is negligible.

3. Magnetovolume effect in the ordered state

Magnetovolume properties observed in the ordered phase of ferromagnets are studied in this section. The discussions are based on the magnetic isotherm, given in the form of the field dependence of reciprocal magnetic susceptibilities,

$$\begin{aligned}&y(\sigma, t) = y_1(t)[\sigma^2 - \sigma_0^2(t)], \qquad y_z(\sigma, t) = y_1(t)[3\sigma^2 - \sigma_0^2(t)], \\ &\Delta y_z = 2y_1(t)\sigma^2 = 2y_1(t)\sigma_0^2(t) + 2y(\sigma, t),\end{aligned} \tag{42}$$

in the weak field limit. The inverse magnetic susceptibility $y_0(t) = y(0, t)$ therefore always vanishes below T_c, for $\sigma = \sigma_0(t)$ in the absence of the external magnetic field, while $y_{z0}(t)$ is finite. Equation (42) is nothing but the magnetic equation of states in reduced units derived from the free energy (1), i.e.

$$H = \frac{\partial F}{\partial M} = a(T)M + b(T)M^3 + \cdots.$$

The coefficient $y_1(t)$ is the reduced fourth order coefficient $b(T)$ of the free energy expansion. The temperature dependence of the spontaneous and the forced magnetostrictions is discussed in detail in subsequent sections.

3.1. Spontaneous magnetostriction

To begin with, after substitution of (29) into (19), the minimum condition of the free energy with respect to ω gives

$$\begin{aligned}\omega_0(t) &= \rho\kappa C_{h0}\frac{2y_1(t)}{5y_{10}}\left\{1+\frac{g_A}{3\sigma_{s0}^2}\left(5\sigma_0^2(t)-3\sigma_s^2\right)\right\}\sigma_0^2(t)\\ &= \tfrac{2}{5}\omega_{s0}V_0(t)\left[1+g_A\left(\tfrac{5}{3}U_0(t)-1\right)\right], \qquad (\text{for } \sigma_s\simeq\sigma_{s0}) \qquad (43)\\ U_0(t) &= \frac{\sigma_0^2(t)}{\sigma_s^2}, \qquad V_0(t)=\frac{y_{z0}(t)}{2y_{10}\sigma_s^2}=\frac{y_1(t)}{y_{10}}U_0(t),\end{aligned}$$

where we have defined $\omega_{s0}=\rho\kappa C_{h0}\sigma_s^2$ as a unit of the magnetostriction. The ground state value $\omega_0(0)$ agrees with (37) in section 2.6. New parameters $U_0(t)$ and $V_0(t)$ denote the reduced spontaneous moment squared and the inverse of the longitudinal magnetic susceptibility, respectively. Both are normalized to unity at $t=0$. The ratios of the thermal magnetostrictions $\omega_t(t)$ and $\Delta\omega_t(t)$ to ω_{s0} are given by

$$\begin{aligned}\frac{\omega_t(t)}{\omega_{s0}} &= \frac{g_0 t}{5c_z(y_{10}\sigma_s^2)^2}\left\{2\int_{x_c}^1 \mathrm{d}x\, x^2u[\log u-1/2u-\psi(u)]\right.\\ &\quad\left.+\int_0^1 \mathrm{d}x\, x^2u_z[\log u_z-1/2u_z-\psi(u_z)]\right\}+\frac{\Delta\omega_t(t)}{\omega_{s0}}, \qquad (44)\\ \frac{\Delta\omega_t(t)}{\omega_{s0}} &= \frac{2g_0y_{z0}}{15c_z(y_{10}\sigma_s^2)^2}\left\{t\left[\frac{\partial A(y_{z0},t)}{\partial t}-\frac{\partial A_t(0,t)}{\partial t}\right]-A(y_{z0},t)+A_t(0,t)\right\},\end{aligned}$$

where x_c is the lower cut-off wavevector due to spin-waves. The volume thermal expansion coefficient $\beta(t)$ is defined by the temperature derivative of the thermal expansion, and is divided into the following components:

$$\begin{aligned}\beta(t) &= \frac{\mathrm{d}\omega_s(t)}{\mathrm{d}T}=\beta_0(t)+\beta_t(t)+\Delta\beta_t(t)\\ &= \frac{1}{T_0}\frac{\mathrm{d}\omega_s(t)}{\mathrm{d}t}=\frac{\omega_{s0}}{T_0}[\bar\beta_0(t)+\bar\beta_t(t)+\Delta\bar\beta_t(t)]. \qquad (45)\end{aligned}$$

The t-dependence of reduced components is expressed by

$$\begin{aligned}\bar\beta_0(t) &= \frac{2}{5}\left\{(1-g_A)V'(t)+\frac{5g_A}{3}[V'(t)U_0(t)+V_0(t)U'(t)]\right\},\\ \bar\beta_t(t) &= \frac{g_0}{5c_z(y_{10}\sigma_s^2)^2}\left\{-2\int_{x_c}^1\mathrm{d}x\,x^2u^2\left(\frac{1}{u}+\frac{1}{2u^2}-\psi'(u)\right)\right.\\ &\quad -\int_0^1\mathrm{d}x\,x^2u_z^2\left(\frac{1}{u_z}+\frac{1}{2u_z^2}-\psi'(u_z)\right)\\ &\quad +\frac{\mathrm{d}V_0(t)}{\mathrm{d}t}\left[-\frac{tx_c}{V_0(t)}x_c^2u_c\left(\ln u_c-\frac{1}{2u_c}-\psi(u_c)\right)\right.\\ &\quad \left.\left.+\ 2y_{10}\sigma_s^2\left(A(y_{z0},t)-t\frac{\partial A(y_{z0},t)}{\partial t}\right)\right]\right\},\\ \Delta\bar\beta_t(t) &= \frac{4g_0}{15c_zy_{10}\sigma_s^2}\left\{V'(t)\left[t\left(\frac{\partial A(y_{z0},t)}{\partial t}-\frac{\partial A_t(0,t)}{\partial t}+y_{z0}\frac{\partial A'(y_{z0},t)}{\partial t}\right)\right.\right.\\ &\quad \left.-A(y_{z0},t)+A_t(0,t)-y_{z0}A'(y_{z0},t)\right]\\ &\quad \left.+tV_0(t)\left[\frac{\partial^2A(y_{z0},t)}{\partial t^2}-\frac{\partial^2A_t(0,t)}{\partial t^2}\right]\right\},\end{aligned}$$

where $u_c=x_c^3/t$ and $A'(y,t)$ denotes the derivative with respect to y.

As the coefficient of $\sigma_0^2(t)$ in the first line of (43), we can find the spontaneous magnetovolume coupling constant $C_s(t)$,

$$C_s(t) = \frac{2C_{h0}}{5}\frac{y_1(t)}{y_{10}}\left\{1 + \frac{g_A}{3\sigma_{s0}^2}\left(5\sigma_0^2(t) - 3\sigma_s^2\right)\right\}$$
$$= \frac{2C_{h0}}{5}\frac{V_0(t)}{U_0(t)}\left\{1 + g_A\left[\frac{5}{3}U_0(t) - 1\right]\right\}, \qquad (\text{for } \sigma_s \simeq \sigma_{s0}) \tag{46}$$

revealing clearly that the coupling constant $C_s(t)$ does show temperature dependence. Notice that our coupling constant $C_0(t)$ in (18) is defined by the free energy expansion in small parameters $y_0(t)$ and $\Delta y_{z0}(t) = 2y_1(t)\sigma_0^2(t)$. The t-dependent coefficient $y_1(t)$ in (46) results from the ratio of $\Delta y_{z0}(t)$ to $\sigma_0^2(t)$ when we show $\omega_0(t)$ in the form proportional to $\sigma_0^2(t)$. An extra dependence due to $U_0(t)$ will also appear in $C_0(t)$ if the ratio g_A is finite and non-negligible. The coupling $C_s(t)$ vanishes at the critical point in reflection to the t-dependence of $y_1(t)$. Further details of the temperature dependence at low temperatures and around T_c are given below.

At low temperatures, we have shown (Takahashi 2001) that the temperature dependence of $U_0(t)$ and $V_0(t)$ is given by

$$\begin{aligned}\frac{y_1(t)}{y_{10}} &= \frac{V_0(t)}{U_0(t)} = 1 - \frac{3+2r^2}{480c_z(y_{10}\sigma_s^2)^2}t^2 + \cdots,\\ \frac{\sigma_0^2(t)}{\sigma_s^2} &= U_0(t) = 1 - \frac{4+5r+r^2}{360c_z(y_{10}\sigma_s^2)^2}t^2 + \cdots,\end{aligned} \tag{47}$$

where $r = \pi^2/4$ (Takahashi and Nakano 2004). The substitution of the result into (46) gives the following t^2-dependence of $C_s(t)$.

$$C_s(t) = C_{s0}\left\{1 - \frac{t^2}{120c_z(y_{10}\sigma_s^2)^2}\left[\frac{3+2r^2}{4} + \frac{5g_A}{3+2g_A}\frac{4+5r+r^2}{3}\right] + \cdots\right\}, \tag{48}$$

where C_{s0} is defined in (37). The volume thermal expansion also shows the same t^2-dependence,

$$\begin{aligned}\frac{\omega_s(t)}{\omega_{s0}} = \frac{2}{5}\left(1 + \frac{2}{3}g_A\right)&\left\{1 - \frac{t^2}{120c_z(y_{10}\sigma_s^2)^2}\left(\frac{3+2r^2}{4} + \frac{3+7g_A}{3+2g_A}\frac{4+5r+r^2}{3}\right) + \cdots\right\}\\ &+ \frac{g_0t^2}{20c_z(y_{10}\sigma_s^2)^2}\ln\sigma_s^{-1} + \frac{g_0(1-r^2)t^2}{180c_z(y_{10}\sigma_s^2)^2}.\end{aligned}$$

The first term comes from $\omega_0(t)$, while the last two terms represent the thermal magnetostriction and its correction. Owing to the presence of the extra logarithmic enhancement factor $\ln\sigma_s^{-1}$, the second term finally becomes predominant in the $\sigma_s \to 0$ limit. We show in figure 1 numerical results of $[\beta_t(t) + \Delta\beta(t)]/(3\rho\kappa\gamma_0 T)$ against the reduced temperature T/T_c for several values of t_c. The increasing enhancement of the T-linear coefficient of $\beta(t)$ with decreasing σ_s is evident in the figure. We hope the presence of the logarithmic enhancement is confirmed from the analysis of the observed thermal expansion at low temperatures.

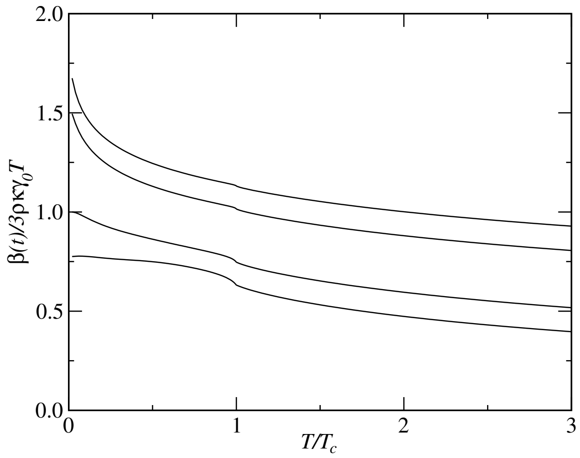

Figure 1. Enhancement of the T-linear coefficient of the thermal expansion coefficient, $[\beta_t(t) + \Delta\beta(t)]/(3\rho\kappa\gamma_0 T)$ for $g_0 = g_A = 0.1$. Solid curves from the top correspond to $t_c = 0.005, 0.01, 0.05, 0.1$.

Around the critical temperature, the temperature dependence of $\sigma_0^2(t)$ and $y_1(t)$ is given by Takahashi (2001)

$$U_0(t) = \frac{\sigma_0^2(t)}{\sigma_s^2} = \frac{7}{5}\left[1 - \left(\frac{t}{t_c}\right)^{4/3}\right],$$
$$\frac{V_0(t)}{U_0(t)} = \frac{y_1(t)}{y_{10}} = \left[\frac{2\sqrt{2}(2+\sqrt{5})}{7}\right]^2 \frac{y_c\sigma_0^2(t)}{y_{10}} = y_{10}\sigma_s^2\left[\frac{20\sqrt{2}c_z}{7\pi t_c}\right]^2 U_0(t), \tag{49}$$
$$y_c = \left[\frac{20c_z y_{10}}{\pi(2+\sqrt{5})t_c}\right]^2.$$

Substitution of these results into (43) leads to the t-dependence of $C_s(t)$ and $\omega_s(t)$,

$$\frac{C_s(t)}{C_{h0}} = \frac{14}{25}(1-g_A)y_{10}\sigma_s^2\left(\frac{40\sqrt{2}c_z}{7\pi t_c}\right)^2\left[1 - \left(\frac{t}{t_c}\right)^{4/3}\right] + \cdots,$$
$$\frac{\omega_s(t)}{\omega_{s0}} = \frac{\omega_t(t)}{\omega_{s0}} + \frac{98}{125}(1-g_A)y_{10}\sigma_s^2\left(\frac{40\sqrt{2}c_z}{7\pi t_c}\right)^2\left[1 - \left(\frac{t}{t_c}\right)^{4/3}\right]^2 + \cdots. \tag{50}$$

As we approach the critical temperature, the spontaneous coupling constant $C_s(t)$ vanishes in proportion to $T - T_c$. Aside from the thermal magnetostriction, $\omega_0(t)$ and its temperature coefficient $\beta_0(t)$ show dependence proportional to $(T - T_c)^2$ and $(T - T_c)$, respectively.

3.2. *Forced magnetostriction*

According to our general expressions, (34) and (35), the forced magnetostriction is evaluated by the following integration with respect to the moment σ:

$$\omega_h(\sigma, t) = \frac{1}{2}\rho\kappa T_A \int_{\sigma_0(t)}^{\sigma} d\sigma'\sigma' \left[\gamma_A y(\sigma', t) - \frac{\partial y(\sigma', t)}{\partial \omega}\right]. \tag{51}$$

The σ-dependence of $y(\sigma, t)$ is determined by the first line of the conditions in (11) written in the form

$$2A_t(y, t) + A(y_z, t) - c_z(2y + y_z) + 5c_z y_{10}\sigma^2 = 3A(0, t_c).$$
$$y_z = y + \sigma\frac{\partial y}{\partial \sigma}. \tag{52}$$

Takahashi (2001) showed how to evaluate $y(\sigma, t)$ by solving (52) as a first order differential equation for $y(\sigma, t)$ with respect to σ. The y dependence of the transverse thermal spin fluctuation amplitude $A_t(y, t)$ around the origin is modified due to the long wave spin-wave excitations in the ordered phase. In the ground state where $y = 0$, $y_z = 2y_{10}\sigma_s^2$, and $\sigma = \sigma_s$ are satisfied, the above equation reduces to the relation between t_c and σ_s,

$$A(0, t_c) = c_z y_{10}\sigma_s^2. \tag{53}$$

The σ-dependence of the strain-derivative $y_\omega \equiv \partial y/\partial\omega$ in the second term of (51) is also evaluated by making use of this equation. The partial ω-derivative of (52) leads to another differential equation for y_ω,

$$\begin{aligned}2[A_t'(y, t) - c_z]y_\omega + [A'(y_z, t) - c_z]\left(y_\omega + \sigma\frac{\partial y_\omega}{\partial\sigma}\right) + 5c_z y_{10}(-\gamma_A + \gamma_0)\sigma^2 \\ = 3c_z y_{10}[\sigma_{s0}^2\gamma_m - \sigma_s^2(\gamma_A - \gamma_0)].\end{aligned} \tag{54}$$

As solutions of simultaneous differential equations, (52) and (54), the σ-dependence of both $y(\sigma, t)$ and $y_\omega(\sigma, t)$ is obtained at the same time. Notice that the σ-dependence of $y(\sigma, t)$ is given by $y(\sigma, t) = y_1(t)[\sigma^2 - \sigma_0^2(t)]$ around $\sigma = \sigma_0(t)$, then the initial condition of $y_\omega(\sigma_0, t)$ is determined from its strain derivative by

$$y_\omega(\sigma_0, t) = -y_1(t)\frac{\partial\sigma_0^2(t)}{\partial\omega} = -y_{10}\sigma_s^2 V_0(t)\left[\gamma_m + \frac{1}{U_0(t)}\frac{\partial U_0(t)}{\partial\omega}\right]. \tag{55}$$

The first order σ^2-derivative of y_ω at $\sigma = \sigma_0(t)$ is also given by

$$\begin{aligned}\frac{\partial y_\omega}{\partial U} &= \sigma_s^2\frac{\partial y_1(t)}{\partial\omega} \\ &= y_{10}\sigma_s^2\frac{V_0(t)}{U_0(t)}\left[-\gamma_A + \gamma_0 + \frac{1}{V_0(t)}\frac{\partial V_0(t)}{\partial\omega} - \frac{1}{U_0(t)}\frac{\partial U_0(t)}{\partial\omega}\right],\end{aligned} \tag{56}$$

where $U = \sigma^2/\sigma_s^2$ is the reduced squared moment. See the appendix on how to evaluate derivatives $\partial U_0(t)/\partial\omega$ and $\partial V_0(t)/\partial\omega$ numerically. Numerical integration of (51) is also performed by solving third-order differential equations consisting of (52), (54), and the following third equation for $\omega_h(\sigma, t)$:

$$\frac{\partial\omega_h(\sigma, t)}{\partial U} = \frac{\omega_{s0}}{y_{10}\sigma_s^2}\left[g_A y(\sigma, t) - \frac{1}{\gamma_m}y_\omega(\sigma, t)\right].$$

We can then obtain the forced magnetostriction $\omega_h(\sigma, t)$ for any arbitrary value of σ. In the weak field limit for $\sigma \simeq \sigma_0(t)$, it is explicitly given by

$$\begin{aligned}\omega_h(\sigma, t) &= \rho\kappa C_h(t)[\sigma^2 - \sigma_0^2(t)] \\ C_h(t) &= -\frac{1}{4}T_A y_\omega = C_{h0}V_0(t)\left[1 + \frac{1}{\gamma_m U_0(t)}\frac{\partial U_0(t)}{\partial\omega}\right],\end{aligned} \tag{57}$$

on substitution of (55) into (51) followed by the σ-integration.

Together with our result of the spontaneous magnetostriction in section 3.1, the magnetostriction in the ordered phase now turns out to be summarized as the following formula:

$$\begin{aligned}&\omega(\sigma, t) = \omega_s(t) + \omega_h(\sigma, t) \\ &\omega_s(t) = \rho\kappa C_s(t)\sigma_0^2(t) + \omega_t(t), \qquad \omega_h(\sigma, t) = \rho\kappa C_h(t)[\sigma^2 - \sigma_0^2(t)].\end{aligned} \tag{58}$$

It corresponds to (2) by the SEW theory or its amendment by MU. Our result particularly differs from them in the following several respects.

(i) The magnetovolume coupling constants, $C_s(t)$ and $C_h(t)$, are different in their magnitudes, i.e. $C_s(t) \neq C_h(t)$. Their temperature dependence is also different.

(ii) The appearance of the new thermal magnetostriction $\omega_t(t)$ is predicted. Although thermal spin fluctuation amplitudes are responsible for this term, its temperature dependence is different from the term proportional to the thermal spin fluctuation amplitude squared, $\xi^2(T)$, predicted by MU. It causes the enhancement of the T-linear coefficient of the thermal expansion coefficient $\beta(t)$ around the magnetic instability point.

All the analysis of experiments to date did not take this term into account. Because it gives the T^2-like dependence to the thermal expansion, it may have been attributed to the electronic origin.

(iii) If we take the presence of the zero-point spin fluctuations into account, we cannot use the definition of the amplitude ratio (4) for $\eta(t_c)$ introduced by MU in our analysis of experiments. Let us therefore introduce an alternative definition by

$$\eta(t) = 1 + \frac{\Delta\omega_s(t)}{\omega_{s0}}, \qquad \Delta\omega_s(t) = \omega_s(\sigma_0(t), t) - \omega_s(\sigma_s, 0).$$

Substitution of (58) then gives

$$\begin{aligned}\eta(T_c) &= 1 + \frac{[-\rho\kappa C_s(0)\sigma_s^2 + \omega_t(t_c)]}{\rho\kappa C_h(0)\sigma_s^2} \\ &= 1 - \frac{C_s(0)}{C_h(0)} + \frac{\omega_t(t_c)}{\omega_{s0}} = \frac{3}{5} - \frac{4}{15}g_A + \frac{\omega_t(t_c)}{\omega_{s0}}.\end{aligned}$$

It agrees with the value 3/5 by MU when both g_A and $\omega_t(t_c)$ are negligible. The SEW theory predicts $\eta = 0$. The finite positive η by the MU theory results from the temperature induced thermal spin fluctuation amplitude $\xi^2(T)$ at $T = T_c$. In our view it is caused by the difference of two magnetovolume coupling constants $C_s(t)$ and $C_h(t)$ as well as the presence of the thermal magnetostriction $\omega_t(t)$. The value of η is, therefore, not necessarily fixed to the unique constant, 3/5.

In figure 2, the temperature dependence of our volume thermal expansion $\omega_s(t)$ is compared with those by the SEW theory and the MU theory. For the purpose of qualitative comparisons, $U_0(t) = \sigma_0^2(t)/\sigma_s^2$ is plotted as $\omega_{\mathrm{SEW}}(t)/\omega_{s0}$. As $\omega_{\mathrm{MU}}(t)/\omega_{s0}$, the values of $U_0(t) + 3(T/T_c)^{4/3}/5$ and $3[1 + V_0(t)]/5$ are plotted for the temperature ranges $T \leqslant T_c$ and $T > T_c$, respectively.

3.2.1. Forced magnetostriction in the low temperature limit and around T_c. In the weak field limit for $\sigma \simeq \sigma_0(t)$, analytical treatments are possible in some restricted temperature ranges. At low temperatures, from the temperature dependence of $U_0(t)$ in (47) we obtain

$$\frac{\partial U_0(t)}{\partial\omega} = \frac{4 + 5r + r^2}{180c_z(y_{10}\sigma_s^2)^2}(\gamma_m - \gamma_A)t^2 + \cdots. \tag{59}$$

Substituting the result into (57) gives the following t^2-linear dependence of the coupling constant:

$$\begin{aligned}\frac{C_h(t)}{C_{h0}} &= V_0(t)\left(1 + \frac{1}{\gamma_m U_0(t)}\frac{\partial U_0(t)}{\partial\omega}\right) \\ &= 1 + \frac{t^2}{120c_z(y_{10}\sigma_s^2)^2}\left[(1 - 2g_A)\frac{4 + 5r + r^2}{3} - \frac{3 + 2r^2}{4}\right] + \cdots,\end{aligned} \tag{60}$$

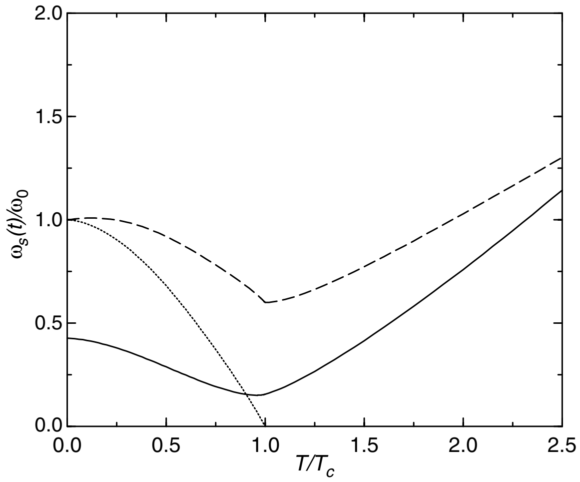

Figure 2. Qualitative comparisons among three theoretical results of spontaneous magnetostrictions. Dotted and broken curves denote those derived by the SEW and the MU theories, respectively. The solid curve represents the ratio $\omega_s(t)/\omega_{s0}$ evaluated by assuming $t_c = 0.1$, $g_0 = g_A = 0.1$, and $T_A/T_0 = 10$.

where r is given by $\pi^2/4$ (Takahashi and Nakano 2004). Comparison of (48) and (60) shows that $C_s(t)$ and $C_h(t)$ have different slopes of their t^2-linear dependence. Around the critical point, the strain derivative of the moment is, from (49), given by

$$\frac{\partial U_0(t)}{\partial \omega} = \frac{28}{15}\left(\frac{t}{t_c}\right)^{4/3}\frac{\mathrm{d}\ln T_c}{\mathrm{d}\omega} \simeq \frac{28}{15}\frac{\mathrm{d}\ln T_c}{\mathrm{d}\omega},$$

leading to the temperature dependence of $C_h(t)$,

$$\frac{C_h(t)}{C_{h0}} = V_0(t)\left(1 + \frac{1}{\gamma_m U_0(t)}\frac{\partial U_0(t)}{\partial \omega}\right) \simeq \frac{V_0(t)}{\gamma_m U_0(t)}\frac{\partial U_0(t)}{\partial \omega}$$
$$= \frac{196}{75}y_{10}\sigma_s^2\frac{1}{\gamma_m}\frac{\mathrm{d}\ln T_c}{\mathrm{d}\omega}\left[\frac{40\sqrt{2}c_z}{7\pi t_c}\right]^2\left[1 - \left(\frac{t}{t_c}\right)^{4/3}\right] + \cdots. \tag{61}$$

In this study the forced magnetovolume coupling constant $C_h(t)$ is defined as the initial slope of $\omega_h(\sigma, t)$ against σ^2. We have already shown that $C_s(t)$ vanishes at $T = T_c$ in (50). Both the magnetovolume coupling constants therefore vanish at T_c in proportion to $T - T_c$. The effect of the volume strain on T_c, i.e. $\mathrm{d}\ln T_c/\mathrm{d}\omega$, will be discussed later.

4. Magnetovolume effect in the paramagnetic state

The purpose of this section is to discuss further details of magnetovolume properties of exchange-enhanced paramagnets and ferromagnets in their paramagnetic phases.

4.1. Exchange-enhanced paramagnets

In order to proceed in parallel with discussions on the ferromagnetic phases, let us define a *paramagnetic* or pseudo-Curie temperature T_c^* from the ratio $t_c^* = T_c^*/T_0$ given as a solution of

$$A(0, t_c^*) = c_z y_0(0),$$

where $y_0(0)$ is the inverse magnetic susceptibility in the ground state. The above relation between t_c^* and $y_0(0)$ corresponds to (53) for ferromagnets. Although no magnetic transition occurs at $T = T_c^*$, the ratio t_c^* characterizes how close the system is to the magnetic instability point. In this meaning, it is better to associate T_c^* with the Curie temperature for ferromagnets. From the correspondence between $y_0(0)$ and $y_{10}\sigma_s^2$ in the ground state, we can also define a pseudo induced magnetic moment squared $\sigma_0^{*2}(t) = y_0(t)/y_1(t)$ for exchange enhanced paramagnets (Takahashi 1994). With these parameters the magnetic isotherm is written in the form

$$y(\sigma, t) = y_0(t) + y_1(t)\sigma^2 + \cdots = y_1(t)[\sigma^2 + \sigma_0^{*2}(t)] + \cdots,$$

that corresponds to (42) for ferromagnets. To pursue the similarity with ferromagnets further, reduced parameters $U_0(t)$ and $V_0(t)$ are also defined by

$$U_0(t) = \frac{y_0(t)}{y_0(0)}\frac{y_{10}}{y_1(t)} = \frac{\sigma_0^{*2}(t)}{\sigma_0^{*2}(0)}, \qquad V_0(t) = \frac{y_0(t)}{y_0(0)}, \qquad \frac{V_0(t)}{U_0(t)} = \frac{y_1(t)}{y_{10}}. \tag{62}$$

As a unit of the size of the magnetostriction, ω_{s0} in (43) is defined by

$$\omega_{s0} = \frac{1}{4}\rho\kappa T_A \frac{y_0^2(0)}{y_{10}}\gamma_m = \frac{1}{4}\rho\kappa T_A y_{10}\gamma_m \sigma_0^{*2}(0) = \rho\kappa C_{h0}\sigma_0^{*2}(0).$$

4.1.1. Spontaneous magnetostriction for paramagnets. Also for paramagnets, the volume thermal expansion, $\omega_s(t) = \omega_0(t) + \omega_t(t)$, has the same origins as those in the ferromagnetic cases. Among them the thermal magnetostriction $\omega_t(t)$ has an isotropic form of (20) with $y_0(t) = y_{z0}(t)$ and $x_c = 0$. Substitution of $C_0(t)$ in (29) for paramagnets into (19) gives the following expression for the first term $\omega_0(t)$:

$$\omega_0(t) = \frac{3}{20}\rho\kappa T_A \frac{y_0(0)}{y_{10}}(\gamma_m + \gamma_A)y_0(t) = \frac{3}{5}\omega_{s0}(1 + g_A)V_0(t), \tag{63}$$

that corresponds to (43) for ferromagnets. Since both the transverse and the longitudinal modes contribute to the volume expansion, the numerical factor $3/5$ appears above instead of $2/5$. In a strict sense, we cannot define a spontaneous magnetostriction in paramagnetic phases. We can, however, attribute the thermal volume expansion $\omega_0(t)$ defined here to the appearance of the pseudo spontaneous moment $\sigma_0^*(t)$. From the definition of $\sigma_0^*(t)$, $\omega_0(t)$ is written in the form

$$\begin{aligned} \omega_0(t) &= \rho\kappa C_{h0}\frac{3y_1(t)}{y_{10}}(1 + g_A)\sigma_0^{*2}(0) = \rho\kappa C_s(t)\sigma_0^{*2}(t), \\ C_s(t) &= \frac{3V_0(t)}{5U_0(t)}(1 + g_A)C_{h0}, \qquad C_{h0} = \frac{1}{4}T_A y_{10}\sigma_0^{*2}(0)\gamma_m, \end{aligned} \tag{64}$$

corresponding to (43) and (46) for ferromagnets. The t-dependence of the thermal magnetostriction $\omega_t(t)$ for paramagnets was already discussed in section 2.3. Numerical results of $\omega_s(t)$ for $g_0 = g_A = 0.1$ and $T_A/T_0 = 10$ are shown in figure 3 with its components $\omega_0(t)$ and $\omega_t(t)$. The temperature dependence of the volume thermal expansion coefficient $\beta(t)$ is given by

$$\begin{aligned} \beta(t) &= \frac{1}{T_0}\frac{\mathrm{d}\omega_s(t)}{\mathrm{d}t} = \frac{\omega_{s0}}{T_0}\bar{\beta}(t) = \frac{\omega_{s0}}{T_0}[\bar{\beta}_0(t) + \bar{\beta}_t(t)], \\ \bar{\beta}_0(t) &= \frac{3}{5}(1 + g_A)\frac{\mathrm{d}V_0(t)}{\mathrm{d}t}, \\ \bar{\beta}_t(t) &= \frac{g_0}{5c_z y_0^2(0)}\left\{-3\int_0^1 \mathrm{d}x\, x^2\Phi(u) + 3y_0(0)\frac{\mathrm{d}V_0(t)}{\mathrm{d}t}\left[A(y_0, t) - t\frac{\partial A(y_0, t)}{\partial t}\right]\right\}. \end{aligned} \tag{65}$$

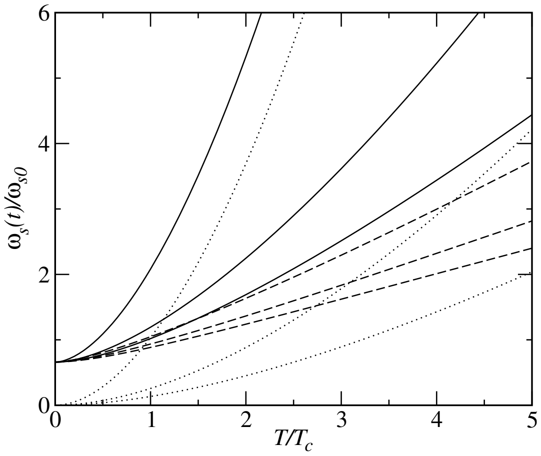

Figure 3. Temperature dependence of the thermal expansion $\omega_s(t)/\omega_{s0}$ for paramagnets for $t_c^* = 0.01, 0.05, 0.1$ from the top (solid curves). The components $\omega_0(t)$ and $\omega_t(t)$ are shown by broken and dotted curves.

Numerical results of its T-linear coefficient $\beta(t)/(3\rho\kappa\gamma_m T)$ are also shown in figure 4 with its components.

At low temperatures, the inverse magnetic susceptibility obeys the following t^2-linear dependence:

$$y_0(t) = y_0(0) + \frac{t^2}{24c_z y_0(0)} + \cdots.$$

Substitution of the above result into (63) leads to the t^2-linear dependence of $\omega_0(t)$. Together with the same dependence of $\omega_t(t)$ in section 2.3, the thermal expansion is given by

$$\frac{\omega_s(t)}{\omega_{s0}} = \frac{3}{5}(1+g_A) + \frac{t^2}{40c_z y_0^2(0)}\left\{g_0 \ln y_0^{-1}(0) + 1 + g_A\right\} + \cdots. \tag{66}$$

The T-linear coefficient of the thermal expansion coefficient $\beta(t)$, i.e. the coefficient of the second t^2-linear term of $\omega_s(t)$, shows the $\ln y_0^{-1}(0)$ enhancement for systems in the vicinity of the magnetic instability point. The linear thermal expansions of Ni_3Al and Ni–Pt alloys have been analysed in their paramagnetic and ferromagnetic phases at low temperatures, according to the following temperature dependence (Franse 1977, Brommer and Franse 1990).

$$\Delta L/L = aT^2 + bT^4.$$

The first and second terms represent the electronic and lattice contributions, respectively. The coefficient a for paramagnetic alloys shows a small upturn near the critical concentration. Upturns are suppressed by high magnetic fields. These observations demonstrate that the above T^2-linear dependence will be of magnetic origin, and hence it is better to attribute it to our thermal magnetostriction $\omega_t(t)$.

4.1.2. Forced magnetostriction for paramagnets. In the weak field limit, the forced magnetovolume coupling constant can be explicitly obtained according to our general formula (35). For this purpose we can make use of the relation (Takahashi 1994)

$$A(y_0(t), t) - c_z y_0(t) = -c_z y_0(0) = -A(0, t_c^*), \tag{67}$$

for the inverse magnetic susceptibility $y_0(t)$, that corresponds to (52) for paramagnets in the absence of the applied field. The temperature dependence of $y_0(t)$ is determined as a solution of (67). The ω-derivative of (67) gives the following equation for $\partial y_0/\partial\omega$.

$$\left[A'(y_0, t) - c_z\right]\left(t\gamma_0\frac{\partial y_0}{\partial t} + \frac{\partial y_0}{\partial \omega}\right) = -t\gamma_0\frac{\partial A(y_0, t)}{\partial t} - c_z\frac{\partial y_0(0)}{\partial \omega}. \tag{68}$$

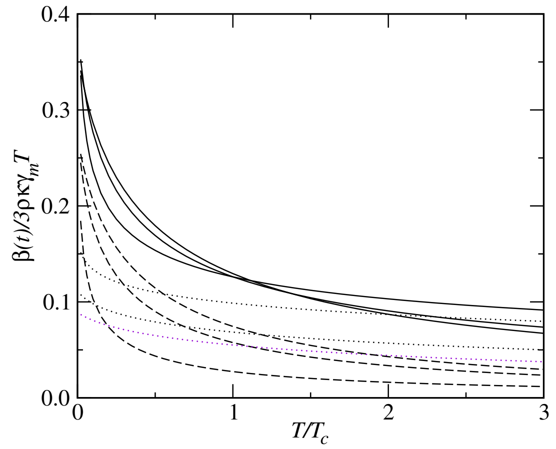

Figure 4. T-linear coefficient of thermal expansion coefficient, $\beta(t)/(3\rho\kappa\gamma_m T)$, for the same paramagnets as figure 3 (solid curves) for $t_c^* = 0.01, 0.05, 0.1$ from the top at high temperatures. Dotted and dashed curves represent components proportional to $\beta_t(t)/t$ and $\beta_0(t)/t$ from the bottom and the top in ascending order of t_c^*, respectively.

The t-derivative terms in the above do not contribute, since $\partial y_0(t)/\partial t$ satisfies the relation

$$\left[A'(y_0, t) - c_z\right] \frac{\partial y_0}{\partial t} = -\frac{\partial A(y_0, t)}{\partial t}, \tag{69}$$

derived from the t-derivative of (67). The strain derivative $\partial y_0(0)/\partial\omega$ in the right-hand side of (68) can be written in terms of Grüneisen parameters. From the definition of $\Delta\left\langle \mathbf{S}_i^2 \right\rangle$ for paramagnets in (28), $y_0(0)$ is given by

$$y_0(0) = -\frac{T_A}{9c_z T_0}\Delta\left\langle \mathbf{S}_i^2 \right\rangle.$$

Substitution of the ω-derivative of both sides,

$$\frac{\partial y_0(0)}{\partial\omega} = [-\gamma_m - \gamma_A + \gamma_0] y_0(0),$$

into (68) therefore leads to

$$\begin{aligned} \frac{\partial y_0(t)}{\partial\omega} &= -\frac{c_z}{A'(y_0, t) - c_z}\frac{\partial y_0(0)}{\partial\omega} \\ &= \frac{y_1(t)}{y_{10}}\frac{\partial y_0(0)}{\partial\omega} = -\frac{y_0(0)}{y_{10}}(\gamma_m + \gamma_A - \gamma_0) y_1(t). \end{aligned} \tag{70}$$

Now according to the general formula (35), the coupling constant $C_h(t)$ is finally written in the form

$$\begin{aligned} \frac{C_h(t)}{C_{h0}} &= \frac{1}{y_0(0)}\left(g_A y_0(t) - \frac{1}{\gamma_m}\frac{\partial y_0(t)}{\partial\omega}\right) = \left\{g_A \frac{y_0(t)}{y_0(0)} + \frac{y_1(t)}{y_{10}}(1 + g_A - g_0)\right\} \\ &= \frac{V_0(t)}{U_0(t)}\left\{1 - g_0 + g_A\left[1 + U_0(t)\right]\right\}. \end{aligned} \tag{71}$$

At low temperatures $C_h(t)$ exhibits the t^2-linear dependence owing to the same temperature dependence of $y_0(t)$ and $y_1(t)$. Numerical results of $C_s(t)$ and $C_h(t)$ for exchange-enhanced paramagnets are shown in figure 5.

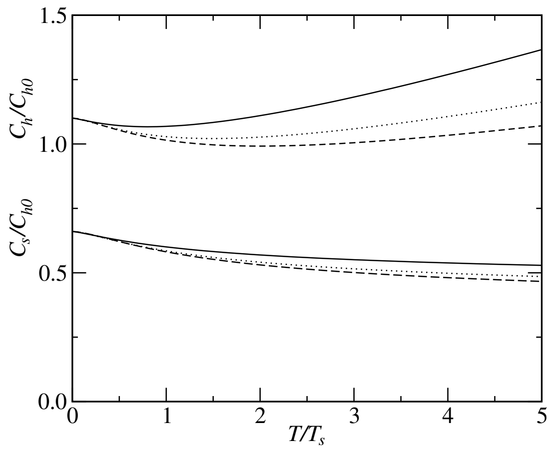

Figure 5. Temperature dependence of magnetovolume coupling constants, $C_s(t)$ and $C_h(t)$, for paramagnets with the same parameters as figure 3. Numerical results for $t_c^* = 0.01$, 0.05, and 0.1 are shown by solid, dotted, and broken curves, respectively.

4.2. Spontaneous magnetostriction in the paramagnetic phase

Though we cannot literally define spontaneous magnetostriction for ferromagnets in their paramagnetic phases with no spontaneous moment, we can associate it with the first term of $\omega_s(t) = \omega_0(t) + \omega_t(t)$ along with the discussion for paramagnets. The second term $\omega_t(t)$ represents the thermal magnetostriction. According to (19), $\omega_0(t)$ is given by

$$\begin{aligned}\omega_0(t) &= \rho\kappa C_s(t)\frac{y_0(t)}{y_1(t)},\\ C_s(t) &= 3y_1(t)C_0(t) = \frac{3y_1(t)}{5y_{10}}\left(1 - g_A\frac{\sigma_s^2}{\sigma_{s0}^2}\right)C_{h0}.\end{aligned}\tag{72}$$

In parallel with the argument for paramagnets, we can define the pseudo magnetic moment squared $\sigma_0^{*2}(t)$ by the ratio $y_0(t)/y_1(t)$. The correspondence between $\sigma_0^*(0)$ and σ_s then leads to the definition of reduced parameters $U_0(t)$ and $V_0(t)$,

$$U_0(t) = \frac{y_0(t)}{y_1(t)\sigma_s^2} = \frac{\sigma_0^{*2}(t)}{\sigma_s^2}, \qquad V_0(t) = \frac{y_0(t)}{y_{10}\sigma_s^2}, \qquad \frac{V_0(t)}{U_0(t)} = \frac{y_1(t)}{y_{10}}.\tag{73}$$

Substituting the above definitions gives analogous expressions to (63) and (64) for the thermal expansion $\omega_s(t)$ and the magnetovolume coupling constant $C_s(t)$,

$$\begin{aligned}&\omega_s(t) = \omega_0(t) + \omega_t(t), \qquad \omega_0(t) = \frac{3}{5}\omega_{s0}V_0(t)\left(1 - g_A\frac{\sigma_s^2}{\sigma_{s0}^2}\right),\\ &\frac{C_s(t)}{C_{h0}} = \frac{3}{5}\frac{V_0(t)}{U_0(t)}\left(1 - g_A\frac{\sigma_s^2}{\sigma_{s0}}\right) = \frac{3}{5}\frac{V_0(t)}{U_0(t)}(1 - g_A). \qquad (\sigma_s \simeq \sigma_{s0}).\end{aligned}\tag{74}$$

The volume thermal expansion coefficient $\beta(t)$ is obtained by the t-derivative of $\omega_s(t)$, as given by

$$\begin{aligned}\beta(t) &= \frac{1}{T_0}\frac{d\omega_s(t)}{dt} = \frac{\omega_{s0}}{T_0}[\bar{\beta}_0(t) + \bar{\beta}_t(t)],\\ \bar{\beta}_0(t) &= \frac{3}{5}(1 - g_A)\frac{dV_0(t)}{dt},\\ \bar{\beta}_t(t) &= \frac{g_0}{5c_z(y_{10}\sigma_s^2)^2}\left\{-3\int_0^1 dx\, x^2\Phi(u) + 3y_{10}\sigma_s^2\frac{dV_0(t)}{dt}\left[A(y_0,t) - t\frac{\partial A(y_0,t)}{\partial t}\right]\right\}.\end{aligned}\tag{75}$$

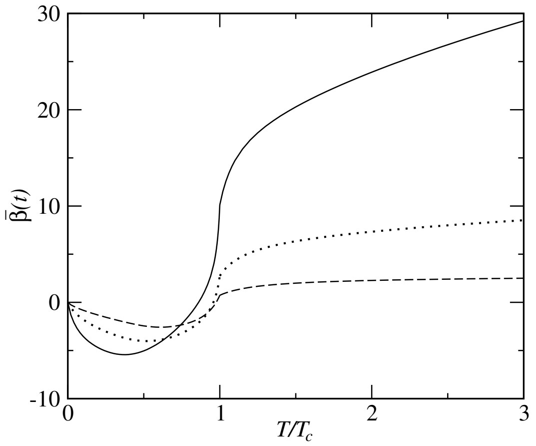

Figure 6. Temperature dependence of the reduced thermal expansion coefficient, $\bar{\beta}(t) = T_0\beta(t)/\omega_{s0}$, for $t_c = 0.05$, 0.1, and 0.2, by solid, dotted, and broken curves, respectively. The same values of parameters g_0, g_A, and the ratio T_A/T_0 as in figure 2 are used.

The analytical expressions of the temperature dependence of these properties are obtained explicitly around the Curie temperature and at higher temperatures where Curie–Weiss law behaviour of the magnetic susceptibility is observed. Around T_c, the parameters $U_0(t)$ and $V_0(t)$ behave as

$$\begin{aligned} U_0(t) &= \tfrac{1}{2}[(t/t_c)^{4/3} - 1], \\ \frac{V_0(t)}{U_0(t)} &= \frac{y_1(t)}{y_{10}} = \left[\frac{\sqrt{2}(2+\sqrt{5})}{5}\right]^2 [(t/t_c)^{4/3} - 1]\frac{y_c\sigma_s^2}{y_{10}} \\ &= \left(\frac{4\sqrt{2}c_z}{\pi t_c}\right)^2 y_{10}\sigma_s^2[(t/t_c)^{4/3} - 1] \end{aligned}$$

where y_c is defined in (49). These are, respectively, proportional to $(t - t_c)$ and $(t - t_c)^2$. Substitution of these results into (74) leads to the t-dependence of $\omega_0(t)$ and $C_s(t)$,

$$\begin{aligned} \frac{\omega_0(t)}{\omega_{s0}} &= \frac{3}{10}(1 - g_A)\left(\frac{4\sqrt{2}c_z}{\pi t_c}\right)^2 y_{10}\sigma_s^2[(t/t_c)^{4/3} - 1]^2, \\ \frac{C_s(t)}{C_{h0}} &= \frac{3}{5}(1 - g_A)\left(\frac{4\sqrt{2}c_z}{\pi t_c}\right)^2 y_{10}\sigma_s^2[(t/t_c)^{4/3} - 1]. \end{aligned} \tag{76}$$

The thermal expansion coefficient $\beta(t)$ is obtained by the t-derivative of the volume thermal expansion. Together with (50) in the ferromagnetic phase, the above result (76) for $\omega_0(t)$ indicates the continuous change of $\beta(t)$ at $T = T_c$. In contrast, both the SEW theory and the MU theory predict the discontinuous change there. The thermal expansion measurements on $ZrZn_2$ by Ogawa and Kasai (1969) and $TiBe_{2-x}Cu_x$ by Creuzet *et al* (1983) seem to support our prediction. Numerical results of $\beta(t)$ and $\omega_s(t)$ in a wide temperature range are shown in figures 6 and 7, respectively.

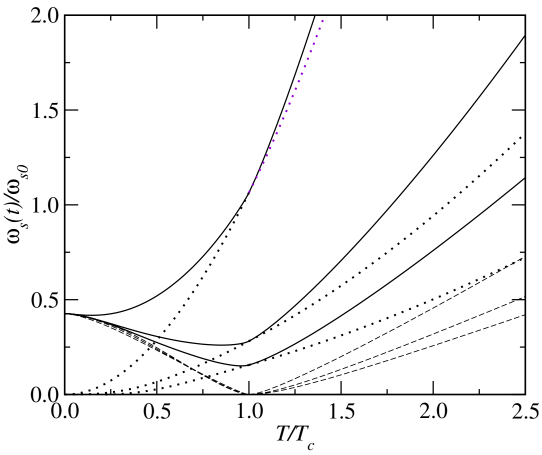

Figure 7. Temperature dependence of the spontaneous magnetostriction $\omega_s(t)/\omega_{s0}$ for $t_c = 0.01$, 0.05, and 0.1 from the top (solid curves). Dotted and broken curves represent ω_t/ω_{s0} (from the top) and ω_0/ω_{s0} (from the bottom for $t < t_c$ and from the top for $t > t_c$) in ascending order of t_c. (This figure is in colour only in the electronic version)

At high temperatures, the Curie–Weiss law of the magnetic susceptibility, and hence $V_0(t)$, are expressed in our reduced units by

$$y_0(t) \simeq \frac{1}{10c_z y_{10}\sigma_{\rm eff}^2}(t-t_c) = \frac{1}{10A(0,t_c)}\frac{\sigma_s^2}{\sigma_{\rm eff}^2}(t-t_c),$$
$$V_0(t) = \frac{y_0(t)}{y_{10}\sigma_s^2} \simeq \frac{c_z}{10A^2(0,t_c)}\frac{\sigma_s^2}{\sigma_{\rm eff}^2}(t-t_c),$$

where $\sigma_{\rm eff}$ is the effective moment per magnetic ion in units of μ_B. The parameter $V_0(t)$ is therefore proportional to $t - t_c$, while the ratio $V_0(t)/U_0(t)$ is almost independent of t. Substitution of these results into (74) gives the following temperature dependence:

$$\begin{aligned}
\frac{\omega_0(t)}{\omega_{s0}} &= \frac{C_s(t)}{C_{h0}}\frac{y_0(t)}{y_1(t)\sigma_s^2} \simeq (1-g_A)\frac{3}{50c_z(y_{10}\sigma_s^2)^2}\frac{\sigma_s^2}{\sigma_{\rm eff}^2}(t-t_c),\\
C_s(t) &\simeq \frac{3(1-g_A)y_1(t)}{5y_{10}}C_{h0},\\
\frac{T_c\beta_0(t)}{\omega_{s0}} &= t_c\bar{\beta}_0(t) = \frac{3}{5}(1-g_A)t_c\frac{dV_0(t)}{dt} = (1-g_A)\frac{3c_z t_c}{5A(0,t_c)}\frac{dy_0(t)}{dt}\\
&\simeq (1-g_A)\frac{3c_z t_c}{50A^2(0,t_c)}\frac{\sigma_s^2}{\sigma_{\rm eff}^2} \simeq (1-g_A)\frac{27c_z}{50C_{4/3}{}^2 t_c^{5/3}}\frac{\sigma_s^2}{\sigma_{\rm eff}^2},\\
C_{4/3} &= 1.006\,089\cdots
\end{aligned} \tag{77}$$

where we have used the relation for the critical thermal amplitude, $A(0, t_c) = c_z y_{10}\sigma_s^2 = C_{4/3}t_c^{4/3}/3$, justified for $t_c \ll 1$. Whereas $\omega_0(t)$ increases with temperature proportional to $T - T_c$, $C_s(t)$ and hence $\beta(t)/\omega_{s0}$ are almost independent of temperature. Owing to the universal relation between ratios of $\sigma_{\rm eff}^2/\sigma_s^2$ and $t_c = T_c/T_0$ (Takahashi 1986, 2001), (77) shows that $T_c\beta_0(t)/\omega_{s0}$ depends solely on the ratio T_c/T_0 (Takahashi 1990) aside from the factor $(1 - g_A)$. The numerical result of its t_c dependence is shown in figure 8. The observed linear thermal expansion coefficients $\alpha_m = \beta(t)/3$ above T_c and the values of ω_{s0} estimated from the forced magnetovolume constant C_{h0} are shown in table 1 for MnSi (Matsunaga *et al* 1982),

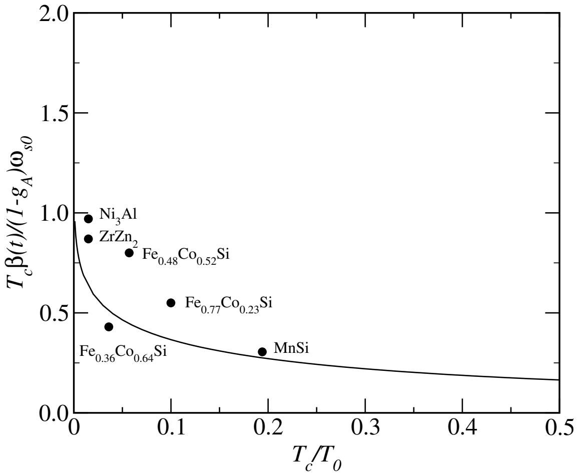

Figure 8. Dependence of the ratio of $T_c\beta_0(t)/(1-g_A)\omega_{s0}$ on $t_c = T_c/T_0$. Numerical results of $\beta_0(t)$ evaluated at $t/t_c = 5$ are shown as a solid curve. Solid circles represent ratios estimated experimentally.

Table 1. Observed thermal expansion coefficient at high temperatures.

	T_c	T_0	T_c/T_0	α_m	ω_{s0}	$T_c\beta/\omega_{s0}$
MnSi	30	155	0.194	30×10^{-7}	6.70×10^{-4}	0.403
$ZrZn_2$	21.3	1390	0.015	12	0.88	0.87
Ni_3Al	41.3	2760	0.015	2.2	0.28	0.97
$Fe_x\,Co_{1-x}\,Si$						
$x = 0.36$	23	640	0.036	2.5	0.4	0.43
$x = 0.48$	48	841	0.057	9.4	1.7	0.80
$x = 0.77$	40	399	0.100	5.5	1.2	0.55

$ZrZn_2$ (Ogawa 1983, Meincke *et al* 1969), Ni_3Al (Suzuki and Masuda 1985), and (Fe,Co)Si (Shimizu *et al* 1989, 1990). Solid circles in figure 8 represent compounds shown in this table (the value corrected by the factor $(1-g_A) = 1.32$ for MnSi by assuming $g_0 = g_A$). Such a t_c-dependence cannot be expected either from the SEW theory or from the MU theory. Both of them assume a larger ratio $C_s/C_{h0} = 1$. Notice that the theoretical curve in figure 8 is determined by the numerical difference between C_s and C_{h0} (i.e. the ratio $C_s(t)/C_{h0} < 1$) and the $\sigma_{\text{eff}}^2/\sigma_s^2$ versus T_c/T_0 relation. The SEW theory always predicts $\beta_0(t) = 0$, since no thermal expansion is present, while in the MU theory no correlation between $\sigma_{\text{eff}}^2/\sigma_s^2$ and T_c/T_0 is expected in general.

4.3. *Forced magnetostriction in the paramagnetic phase*

In the weak field limit, the forced magnetostriction in the paramagnetic phase for ferromagnets can be treated in just the same way as in section 4.1.2. In place of (67) the t-dependence of the magnetic susceptibility is obtained by solving the equation

$$A(y_0(t), t) - c_z y_0(t) = A(0, t_c). \tag{78}$$

In order to get the magnetovolume coupling constant $C_h(t)$, we need the ω-derivative of $y_0(t)$. It is also evaluated with the use of (78). The ω-derivative of (78) gives the equation for $\partial y_0(t)/\partial\omega$,

$$\begin{aligned}\frac{\partial y_0(t)}{\partial\omega} &= \frac{1}{A'(y_0,t)-c_z}\frac{\partial A(0,t_c)}{\partial\omega} = -\frac{y_1(t)}{c_z y_{10}}A(0,t_c)\frac{\partial \ln A(0,t_c)}{\partial\omega}\\ &= -y_1(t)\sigma_{s0}^2\left[\gamma_m - \frac{\sigma_s^2}{\sigma_{s0}^2}(\gamma_A-\gamma_0)\right],\end{aligned} \tag{79}$$

where t-derivative terms do not appear for the same reason as before. The spin amplitude conservation yields the relation between $\Delta\left\langle \mathbf{S}_i^2\right\rangle$ and the thermal spin amplitude $A(0, t_c)$,

$$\Delta\left\langle \mathbf{S}_i^2\right\rangle = \frac{9T_0}{T_A}A(0, t_c) = \frac{3}{20}\sigma_s^2. \tag{80}$$

The ω-derivative of this relation leads to the following expression for $\mathrm{d}\ln A(0, t_c)/\mathrm{d}\omega$ in (79),

$$\frac{\partial \ln A(0, t_c)}{\partial\omega} = \frac{\sigma_{s0}^2}{\sigma_s^2}\gamma_m - \gamma_A + \gamma_0,$$

in terms of Grüneisen parameters. Except for the narrow temperature region around T_c, $y_1(t)$ is almost independent of temperature (Takahashi 2001), implying the almost temperature independent slope of $y_0(t)$ against ω from (79).

The above conclusion is supported by the pressure effect measurements of the magnetic susceptibility on Ni_3Al by Brommer *et al* (1995). Their results are fitted well with the linear relation

$$\frac{\mathrm{d}\ln\chi(T)}{\mathrm{d}\omega} = \frac{1}{\chi(T)}\frac{\mathrm{d}\chi(T)}{\mathrm{d}\omega} \propto \chi(T),$$

being equivalent with the temperature independence of $\chi^{-2}(T)\,\mathrm{d}\chi(T)/\mathrm{d}\omega = \mathrm{d}\chi^{-1}(T)/\mathrm{d}\omega$. Since $y_0(t)$ is the reduced inverse magnetic susceptibility, i.e. $y_0(t) \propto \chi^{-1}(T)$, the observed linear relation is consistent with our prediction (79). We can further check the validity of (79) from the quantitative comparison with experiments. The observed slope in our notation is written in the form

$$\begin{aligned}\frac{N_0}{2\chi(T)}\frac{\mathrm{d}\ln\chi(T)}{\mathrm{d}\ln V} &= -T_A y_0\frac{\partial \ln y_0}{\partial\omega} = -T_A\frac{\partial y_0}{\partial\omega}\\ &= T_A y_1(t)\sigma_s^2\frac{\mathrm{d}\ln A(0, t_c)}{\mathrm{d}\omega} = T_A y_1(t)\sigma_s^2(\gamma_m - \gamma_A + \gamma_0).\end{aligned} \tag{81}$$

The estimated slope of $\ln\chi(T)/\mathrm{d}\ln V$ against $\chi(T)$ in the left-hand side of (81) is given by 2.27×10^3 K. On the other hand, spectral parameters T_0 and T_A are estimated to be 3.6×10^3 K and 3.1×10^4 K for Ni_3Al, respectively, from the microscopic measurements (Bernhoeft *et al* 1983, 1986). These values imply $y_1(t) \sim y_{10} \sim 0.29$. The observed pressure effect measurement $\mathrm{d}\ln\sigma_s^2/\mathrm{d}p = 27.2\times 10^{-3}\ \mathrm{kbar}^{-1}$ and the bulk modulus, $B = \kappa^{-1} = 1.7$ M bar, lead to the Grüneisen parameter,

$$\gamma_m = -B\frac{\mathrm{d}\ln\sigma_s^2}{\mathrm{d}p} \simeq 46.2.$$

On substituting these observed and estimated values, the right-hand side of (81) is given by 2.0×10^3 K for $\sigma_s = 0.07$, respectively, on neglecting parameters γ_0 and γ_A. Fair agreement of these two slopes estimated independently demonstrates the quantitative validity of (79).

The forced magnetovolume coupling constant $C_h(t)$ in the weak field limit is obtained by substituting (79) into (51).

$$\begin{aligned}\frac{C_h(t)}{C_{h0}} &= \frac{1}{y_{10}\sigma_{s0}^2}\left(g_A y_0(t) - \frac{1}{\gamma_m}\frac{\partial y_0(t)}{\partial\omega}\right) = \frac{1}{y_{10}\sigma_{s0}^2}\left\{g_A y_0(t) + \frac{y_1(t)}{c_z y_{10}\gamma_m}\frac{\partial A(0, t_c)}{\partial\omega}\right\}\\ &= \frac{y_1(t)}{y_{10}}\left\{1 - \frac{\sigma_s^2}{\sigma_{s0}^2}\left[\left(1 - \frac{y_0(t)}{y_1(t)\sigma_s^2}\right)g_A - g_0\right]\right\}\\ &= \frac{V_0(t)}{U_0(t)}\{1 - g_A[1 - U_0(t)] + g_0\}, \qquad (\sigma_s \simeq \sigma_{s0}).\end{aligned} \tag{82}$$

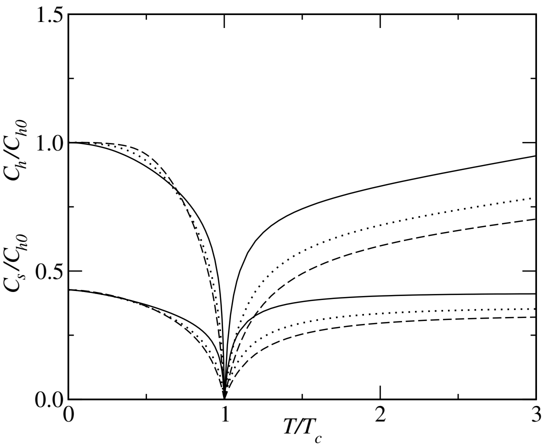

Figure 9. Temperature dependence of $C_s(t)$ and $C_h(t)$ for $t_c = 0.01$ (solid curves), 0.05 (dotted curved), and 0.1 (broken curves).

Around the critical point, it shows the $(t - t_c)$-linear dependence, given by

$$\frac{C_h(t)}{C_{h0}} = (1 - g_A + g_0)\left(\frac{4\sqrt{2}c_z}{\pi t_c}\right)^2 y_{10}\sigma_s^2[(t/t_c)^{4/3} - 1], \tag{83}$$

and vanishes at the critical point. With increasing the temperature it exhibits the tendency of saturation. If the magnitude of g_A is appreciable, an additional $(t - t_c)$-linear increase will be observed due to the presence of $U_0(t)$. Numerical results for $C_s(t)$ and $C_h(t)$ for ferromagnets in a wider temperature range are shown in figure 9.

In the presence of a finite external magnetic field, we have to employ a numerical method to find the forced magnetostriction by solving the following third order simultaneous differential equations for $y(\sigma, t)$, $y_\omega(\sigma, t)$, and $\omega_h(\sigma, t)$,

$$\begin{aligned}
&2A(y, t) + A(y_z, t) - c_z(2y + y_z) + 5c_z y_{10}\sigma^2 = 3A(0, t_c),\\
&2[A'(y, t) - c_z]y_\omega + [A'(y_z, t) - c_z]\left(y_\omega + \sigma\frac{\partial y_\omega}{\partial \sigma}\right) + 5c_z y_{10}(-\gamma_A + \gamma_0)\sigma^2\\
&\qquad = 3c_z y_{10}\sigma_{s0}^2\left[\gamma_m - \frac{\sigma_s^2}{\sigma_{s0}^2}(\gamma_A - \gamma_0)\right],\\
&\frac{\partial \omega_h(\sigma, t)}{\partial U} = \frac{\omega_{s0}}{y_{10}\sigma_s^2}\left[g_A y_0(\sigma, t) - \frac{1}{\gamma_m}y_\omega(\sigma, t)\right],
\end{aligned} \tag{84}$$

where y_z is given by $y_z = y + 2U\partial y/\partial U$. The initial conditions of the equations are determined by the series expansion of solutions in powers of U. Let $y_\omega(\sigma, t)$ be expanded as follows.

$$y_\omega(\sigma, t) = y_\omega(0, t) + y'_\omega(0, t)\sigma_s^2 U + \cdots, \tag{85}$$

where the derivative of $y_\omega(\sigma, t)$ with respect to σ^2 is denoted by y'_ω. Then after substitution, the comparison of the coefficients of zeroth order terms gives the initial condition at $U = 0$,

$$y_\omega(0, t) = -y_{10}\sigma_s^2\gamma_m(1 - g_A + g_0)\frac{V_0(t)}{U_0(t)}.$$

On the other hand, the first order coefficients provide the condition

$$[A'(y_0, t) - c_z]y'_\omega(0, t)\sigma_s^2 + A''(y_0, t)y_1(t)\sigma_s^2 y_\omega(0, t) + A(0, t_c)(-\gamma_A + \gamma_0) = 0.$$

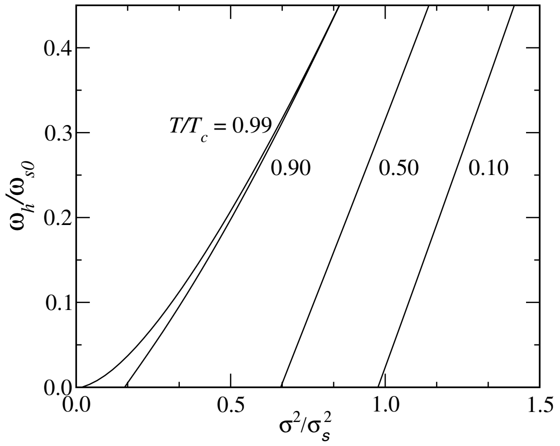

Figure 10. Forced magnetostriction $\omega_h(\sigma, t)/\omega_{s0}$ against σ^2/σ_s^2 for $t_c = 0.05$ at various temperatures below T_c.

The initial conditions of y'_ω and the derivative of $\omega_h(\sigma, t)$ with respect to U are given by

$$y'_\omega(0,t)\sigma_s^2 = \frac{A(0,t_c)}{A'(y_0,t)-c_z}\left[\gamma_A - \gamma_0 - A''(y_0,t)\frac{y_1(t)}{c_z y_{10}} y_\omega(0,t)\right]$$
$$= -A(0,t_c)\frac{V_0(t)}{c_z U_0(t)}\left[\gamma_A - \gamma_0 - \frac{V_0(t)}{c_z U_0(t)} A''(y_0,t) y_\omega(0,t)\right],$$
$$\frac{\partial \omega_h(\sigma,t)}{\partial U} = \frac{\omega_{s0}}{y_{10}\sigma_s^2}\left[g_A y_0(t) - \frac{1}{\gamma_m} y_\omega(0,t)\right].$$

The initial conditions of $y(\sigma, t)$ and $y_z(\sigma, t)$ are simply given by the inverse of the magnetic susceptibility, $y_0(t)$. In figure 10, the forced magnetostriction $\omega_h(\sigma, t)$ is plotted against σ^2 for several reduced temperatures below T_c.

5. Magnetovolume effect at the critical point

Not so much attention has been paid so far to the critical magnetovolume behaviour except for the study on the forced magnetostriction by Takahashi (1990). In this section, the critical forced magnetostriction is discussed again from a slightly different point of view. The pressure dependence of T_c is also discussed in relation to our Grüneisen parameters.

5.1. Critical forced striction

Magnetic isotherms in general exhibit a peculiar critical anomaly around the critical point T_c. In order to obtain the critical forced magnetostriction, let us follow the same procedure as in preceding sections. We have already demonstrated that $y(\sigma, t)$ and $y_z(\sigma, t)$ show critical σ^4-linear dependence (Takahashi 1986, 2001) as solutions of the first line of (84). The first two terms of the left-hand side of its second equation for $y_\omega = \partial y/\partial\omega$ can be written as

$$2[A'(y,t) - c_z]\frac{\partial y}{\partial \omega} + [A'(y_z,t) - c_z]\frac{\partial y_z}{\partial \omega} \simeq -\frac{\pi t_c}{8}\left(\frac{2}{\sqrt{y}}\frac{\partial y}{\partial \omega} + \frac{1}{\sqrt{y_z}}\frac{\partial y_z}{\partial \omega}\right),$$

owing to the critical behaviour of the thermal amplitude,

$$A'(y,t) \simeq -\frac{\pi t}{4\sqrt{y}}.$$

Substituting the result into the second line of (84) leads to

$$-\frac{\pi t_c}{8}\left(\frac{2}{\sqrt{y}}\frac{\partial y}{\partial \omega} + \frac{1}{\sqrt{y_z}}\frac{\partial y_z}{\partial \omega}\right) = 3c_z y_{10}\sigma_s^2\gamma_m(1 - g_A - g_0). \tag{86}$$

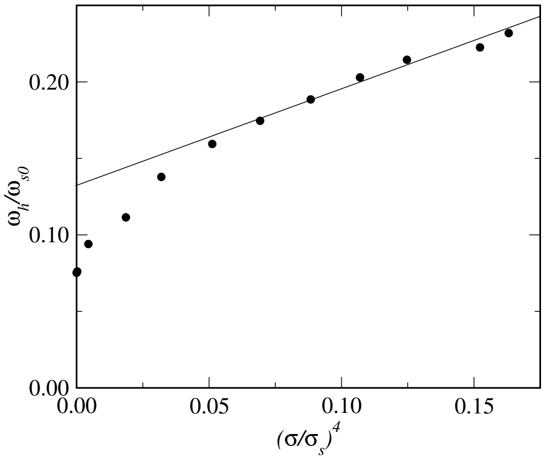

Figure 11. Critical forced magnetostriction of MnSi. Experiments are shown by solid circles, while the thin solid line is a guide for the eyes.

The third term in the same line proportional to σ^2 is of higher order in σ, and hence neglected. The σ-independence of the right-hand side of (86) and the critical σ^4-linear dependence of $y(\sigma, t)$ and $y_z(\sigma, t)$ imply $y_\omega \propto \sigma^2$ and $\partial y_z/\partial\omega = y_\omega + \sigma\partial y_\omega/\partial\sigma = 3y_\omega$. On substituting these σ^2-linear dependences into (86), one can determine their coefficients. The derivative $y_\omega(\sigma, t)$ is thus obtained by

$$\frac{1}{\gamma_m}\frac{\partial y}{\partial\omega} = -\frac{24\sqrt{5}}{3+2\sqrt{5}}(1-g_A-g_0)\frac{\sqrt{y_c}}{\pi t_c}A(0,t_c)\sigma_s^2 U.$$

The critical σ^4-linear dependence of the forced volume striction is finally deduced from our general formula (51),

$$\begin{aligned}\frac{\omega_h(\sigma,t)}{\omega_{s0}} &= \frac{12\sqrt{5}}{3+2\sqrt{5}}(1-g_A-g_0)\frac{\sqrt{y_c}}{\pi t_c y_{10}}A(0,t_c)U^2 \\ &= (1-g_A-g_0)\frac{80\sqrt{5}c_z C_{4/3}}{(16+7\sqrt{5})\pi^2 t_c^{2/3}}\frac{\sigma^4}{\sigma_s^4} \\ &\simeq 0.2881(1-g_A-g_0)t_c^{-2/3}\frac{\sigma^4}{\sigma_s^4}, \qquad (\text{for } c_z = 0.5).\end{aligned} \tag{87}$$

As shown in figure 10, the non-linear behaviour becomes prominent around the weak external field range as we approach the critical temperature. The observed forced volume strains for MnSi (Matsunaga *et al* 1982) at the critical point $T = 29$ K are replotted in figure 11 in our reduced units. The deviation from the σ^2-linear behaviour is fitted well with the relation $\omega_h/\omega_{s0} = 0.627(\sigma/\sigma_s)^4$. If we assume $t_c = 0.3$, the slope of (87) against $(\sigma/\sigma_s)^4$ gives 0.634 apart from the factor $(1 - g_A - g_A)$, in good quantitative agreement with experiments.

5.2. *Pressure dependence of* T_c

According to the SEW theory, the pressure effect on T_c is derived from the strain dependence of the second order coefficient $a(T, \omega)$ in (1). Although it is a slightly cumbersome procedure, let us find the pressure effect on T_c from the same condition as the SEW theory below. The critical temperature T_c is thus determined by the condition $y_0(t_c, \omega) = 0$. From the variation of the condition with respect to the strain, we obtain

$$\left.\frac{\partial y_0(t,\omega)}{\partial t}\right|_{t=t_c}\left(\frac{\delta T_c}{T_0} - \frac{T_c}{T_0^2}\delta T_0\right) + \left.\frac{\partial y_0(t,\omega)}{\partial\omega}\right|_{t=t_c}\delta\omega = 0. \tag{88}$$

With the use of (69) and (79), the above two partial derivatives of $\partial y_0/\partial t$ and $\partial y_0/\partial \omega$ can be rewritten as follows.

$$\begin{aligned} \frac{\partial y_0(t,\omega)}{\partial \omega} &= \frac{1}{[A'(y_0,t)-c_z]}\frac{\partial A(0,t_c)}{\partial \omega} = \frac{c_z}{[A'(y_0,t)-c_z]}\frac{d(y_{10}\sigma_s^2)}{d\omega}, \\ \frac{\partial y_0(t,\omega)}{\partial t} &= -\frac{1}{[A'(y_0,t)-c_z]}\frac{\partial A(0,t)}{\partial t}. \end{aligned} \tag{89}$$

Substituting these results into (88) leads to the relation

$$t_c\frac{\partial A(0,t_c)}{\partial t_c}\left(\frac{d\ln T_c}{d\omega}+\gamma_0\right) = c_z\frac{d(y_{10}\sigma_s^2)}{d\omega}. \tag{90}$$

The left-hand side is given by $dA(0,t_c)/d\omega$ from the $t_c^{4/3}$-linear dependence of $A(0,t_c)$ in (77) in the limit $t_c \ll 1$. The result of (90), i.e. $d[A(0,t_c)-c_z y_{10}\sigma_s^2]/d\omega = 0$, therefore implies that the condition (88) is equivalent to the following relation, satisfied for every value of volume strain adiabatically.

$$\sigma_s^2 = \frac{1}{c_z y_{10}}A(0,t_c) = \frac{C_{4/3}}{3c_z y_{10}}t_c^{4/3} = \frac{20C_{4/3}T_0}{T_A}\left(\frac{T_c}{T_0}\right)^{4/3}. \tag{91}$$

Our introduction of Grüneisen parameters, defined from the volume dependence of σ_s^2, T_0, and T_A in (27), amounts to the following ω (or pressure) dependence of T_c given by

$$\left(\frac{T_c}{T_{c0}}\right)^{4/3} = e^{-(\gamma_A+\gamma_0/3)\omega}(1+\gamma_m\omega), \tag{92}$$

where T_{c0} is the Curie temperature at the reference volume for $\omega = 0$. Its initial slope around $\omega \simeq 0$ is given by

$$T_c^{4/3} = T_{c0}^{4/3}\left\{1+\omega\left(\gamma_m-\gamma_A-\frac{\gamma_0}{3}\right)\right\},$$

or, in the differential form, by

$$\frac{4}{3}\frac{d\ln T_c}{d\omega} = \gamma_m-\gamma_A-\frac{1}{3}\gamma_0. \tag{93}$$

With our definition of γ_m in (27), we have found that the pressure dependence of σ_s^2 and T_c is governed by the different sets of magnetic Grüneisen parameters. The strain derivative of the logarithm of T_c in (92) exhibits divergent behaviour around the critical volume strain, $\omega_c = -1/\gamma_m$.

$$\frac{4}{3}\frac{d\ln T_c}{d\omega} = \frac{\gamma_m}{1+\gamma_m\omega}-\gamma_A-\frac{1}{3}\gamma_0 = \gamma_m\left(\frac{\omega_c}{\omega_c-\omega}-g_A-\frac{1}{3}g_0\right).$$

As the pressure effect, (93) is also written in the form

$$\frac{d\ln T_c}{dp}-\frac{3}{4}\frac{d\ln\sigma_s^2}{dp} = \frac{\kappa}{4}(3\gamma_A+\gamma_0). \tag{94}$$

Around the magnetic instability point $\omega \simeq \omega_c = -1/\gamma_m$, on the other hand, the slope of $T_c^{4/3}$ against ω is in general different from the initial value at $\omega = 0$ and is given by

$$\frac{d}{d\omega}\left(\frac{T_c}{T_{c0}}\right)^{4/3} = e^{g_A+g_0/3}\gamma_m.$$

Currently, it is widely believed that the pressure effects on T_c and σ_s obey the relation

$$\frac{d\ln\sigma_s}{dp} = \frac{2}{3}\frac{d\ln T_c}{dp}, \tag{95}$$

based on the SCR theory. The SEW theory rather insists on

$$\frac{\mathrm{d}\ln\sigma_s}{\mathrm{d}p} = \frac{\mathrm{d}\ln T_\mathrm{c}}{\mathrm{d}p}. \tag{96}$$

In view of our result (94), neither relation is justified generally. Both the above linear relations imply the same sign of slopes, $\mathrm{d}T_\mathrm{c}/\mathrm{d}p$ and $\mathrm{d}\sigma_s/\mathrm{d}p$. The observed variety of their signs also shows the limitation of these relations. According to our (94), on the other hand, various cases are possible depending on magnitudes and signs of our three Grüneisen parameters. For example, we can predict the following general tendencies.

- When $|\gamma_m| \gg |\gamma_0|, |\gamma_A|$ is satisfied, then
$$\frac{\partial T_\mathrm{c}}{\mathrm{d}p} > 0, \quad \frac{\partial\sigma_s}{\mathrm{d}p} > 0, \qquad \text{or } \frac{\partial T_\mathrm{c}}{\mathrm{d}p} < 0, \quad \frac{\partial\sigma_s}{\mathrm{d}p} < 0.$$
- When $\gamma_m \simeq 0$, then $\partial\sigma_s/\mathrm{d}p \simeq 0$ and
$$\frac{\partial T_\mathrm{c}}{\mathrm{d}p} > 0, \qquad \text{or } \frac{\partial T_\mathrm{c}}{\mathrm{d}p} < 0,$$
depending on the sign of γ_{0A}.

Fåk *et al* (2005) have recently suggested the violation of the linear relation, $\sigma_s^2 \propto T_\mathrm{c}^{4/3}$, implied by (91) from the pressure dependence of sublattice magnetization of MnSi by neutron diffraction measurements. This should not, however, be attributed to the violation of (91) but to the presence of the pressure dependence of spectral parameters, T_0 and T_A.

5.3. *Experimental determination of Grüneisen parameters*

We have introduced three magnetic Grüneisen parameters as the ω-dependence of the characteristic scales of our spin fluctuation model. We show below how to determine them experimentally by the pressure-effect measurements. The parameter γ_m is simply determined by the variation of the spontaneous moment σ_s under pressure in the ground state. From the observed pressure effect on T_c, the weighted average of γ_0 and γ_A, i.e. the value of $\gamma_{0A} = (\gamma_0 + 3\gamma_A)/4$, is estimated by (94). We show in table 2 values of γ_m and γ_{0A} for various itinerant electron ferromagnets determined experimentally. Compounds are in ascending order depending on the magnitude of the ratio γ_{0A}/γ_m (except for Y(Co, Al)$_2$ and (Fe,Co)Si). The last row but one of table 2 clearly suggests that the strain dependence of the spectral width is not always negligible. We also find that the ratio γ_{0A}/γ_m is likely to become negative for compounds with larger magnitudes of their ratio. The reason is unknown at present. In order to check the mutual correlation between parameters γ_m, γ_0, and γ_A experimentally, values (in units of 10^{-3} kbar^{-1}) of $\kappa\gamma_{0A}$ for compounds shown in table 2 are plotted against $\kappa\gamma_m$ in figure 12. Judging from the figure, it is reasonable to assume these two parameters as independent.

If another independent pressure effect measurement is available, we can determine γ_0 and γ_A individually. For instance, the parameter y_{10} in (36) is proportional to the ratio T_A/T_0. The fourth order expansion coefficient $b(0)$ of the free energy in (1) is related to y_{10} by $b(0) \propto T_A y_{10}$. The pressure derivative of $b(0)$ is therefore given by

$$\frac{\mathrm{d}\ln b(0)}{\mathrm{d}p} = 2\frac{\mathrm{d}\ln T_A}{\mathrm{d}p} - \frac{\mathrm{d}\ln T_0}{\mathrm{d}p} = \kappa(2\gamma_A - \gamma_0). \tag{97}$$

As solutions of linear simultaneous equations of (94) and (97), Grüneisen parameters γ_0 and γ_A are given by

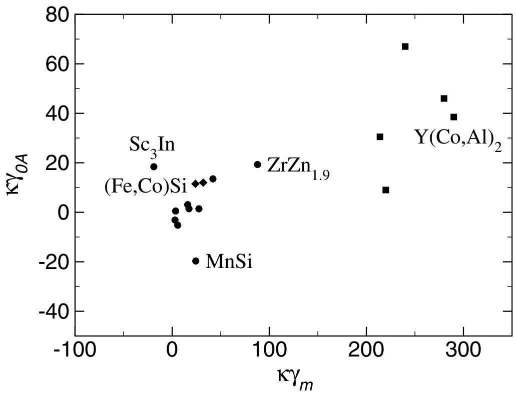

Figure 12. Correlation between Grüneisen parameters.

Table 2. Grüneisen parameters estimated experimentally.

Compounds	$\kappa\gamma_m$ (10^{-3} kbar^{-1})	$-\mathrm{d}\ln T_c/\mathrm{d}p$ (10^{-3} kbar^{-1})	$\kappa\gamma_{0A}$ (10^{-3} kbar^{-1})	γ_{0A}/γ_m	References
$TiFe_{0.5}Co_{0.5}$	27.6	19.3	1.4	0.051	Beille *et al* (1978)
$Ni_{75}Al_{25}$	17.4	11.6	1.45	0.083	Buis *et al* (1981)
$Y(Co_{1-x}Al_x)_2$					
$x = 0.145$	290	179	38.5	0.133	Duc *et al* (1993)
$x = 0.15$	240	113	67	0.279	Armitage *et al* (1990)
$x = 0.16$	214	130	30.5	0.143	Duc *et al* (1993)
$x = 0.185$	280	164	46	0.164	Duc *et al* (1993)
$x = 0.2$	220	156	9	0.041	Duc *et al* (1993)
Co_2ZrAl	3.6	2.2	0.5	0.139	Kanomata *et al* (2005)
$Fe_{67}Ni_{33}$	16.0[a]	8.9[b]	3.1	0.194	[a]Kanomata (2005), [b]Shiga (1993)
$ZrZn_{1.9}$	88	46.7	19.3	0.219	Huber *et al* (1975)
$Ni_{45}Pt_{55}$	42	18	13.5	0.321	Koyama (2005)
$Fe_xCo_{1-x}Si$					
$x = 0.3$	32[c]	12[d]	12	0.375	[c]Beille *et al* (1979), [d]Miura *et al* (2005),
$x = 0.5$	24[e]	6.5[f]	11.5	0.479	[e]Beille *et al* (1979), [f]Miura *et al* (2005)
MnSi	24.4	38	−19.7	−0.807	Koyama *et al* (2000b)
Co_2TiGa	5.8[g]	9.5[h]	−5.2	−0.897	[g]Sasaki (1999), [h]Sasaki *et al* (2001)
$Sc_{75.7}In_{24.3}$	−18.8	−32.5	18.4	−0.979	Grewe *et al* (1989)
Rh_2NiGe	3.0	5.3	−3.1	−1.033	Adachi *et al* (2005)

$$
\begin{aligned}
\gamma_0 &= \frac{1}{\kappa}\frac{\mathrm{d}\ln T_0}{\mathrm{d}p} = \frac{6}{5}\gamma_m + \frac{1}{\kappa}\left(\frac{8}{5}\frac{\mathrm{d}\ln T_c}{\mathrm{d}p} - \frac{3}{5}\frac{\mathrm{d}\ln b(0)}{\mathrm{d}p}\right), \\
\gamma_A &= \frac{1}{\kappa}\frac{\mathrm{d}\ln T_A}{\mathrm{d}p} = \frac{3}{5}\gamma_m + \frac{1}{\kappa}\left(\frac{4}{5}\frac{\mathrm{d}\ln T_c}{\mathrm{d}p} + \frac{1}{5}\frac{\mathrm{d}\ln b(0)}{\mathrm{d}p}\right).
\end{aligned}
\tag{98}
$$

The pressure dependence of $b(0)$ estimated from the slope of the Arrott plot of magnetization measurements will determine them separately. The observed pressure effect on the Arrott plot of magnetization curves on Ni_3Al (Buis *et al* 1976) seems to suggest $\mathrm{d}b(0)/\mathrm{d}p \simeq 0$ for this compound. On the other hand, when $\gamma_0 \simeq \gamma_A$ is satisfied due to the localized character of the spin fluctuation spectrum, the $b(0)$ will show the same pressure dependence of these spectral widths. Though this is the first time we publish our idea of the volume dependence of spectral parameters, T_0 and T_A, we have recognized its importance before. From the Arrott plot analysis of the magnetization curve under pressure, the pressure dependences of these parameters of MnSi have already been estimated by Thessieu *et al* (1998) based on the above relation.

6. Discussion

The magnetovolume effects of itinerant electron ferro- and paramagnets have been discussed in this paper by the inspection of the explicit volume dependence of the free energy consisting of spin fluctuation excitations. We have thus found the presence of the thermal magnetostriction $\omega_t(t)$. The term is necessary in order to guarantee the consistency between the temperature dependence of thermal volume expansions at low temperatures and the enhancement of the T-linear coefficient of the specific heat. Unfortunately, little theoretical concern has been directed to this intimate relationship between them to date. We have also succeeded in deriving various new striking results, that can be summarized as follows.

- Three magnetic Grüneisen parameters, γ_m, γ_0, and γ_0, are introduced as the volume dependence of the characteristic parameters of the free energy. Theoretically, the volume dependence is usually introduced by the $\Omega^{-3/5}$ dependence of the band width. Our semi-phenomenological approach has an advantage when we try to understand various pressure effect measurements in their mutual relationships in accordance with theoretical predictions. The negative volume expansion in our picture is related to the parameter γ_m. It results from the volume dependence of the ratio of the zero-point spin fluctuation amplitude to the total amplitude, even if we assume the total amplitude conservation.
- The spontaneous magnetostriction consists of the sum of two kinds of contributions, given by
$$\omega_s(t) = \omega_0(t) + \omega_t(t), \qquad \omega_0(t) = \rho\kappa C_s(t)\sigma_0^2(t).$$
The second term represents the thermal magnetostriction, newly acquired in the present study. This term gives rise to a T^2-like temperature dependence of the volume thermal expansion, that leads to an enhancement of the T-linear coefficient of the thermal expansion coefficient $\beta(t)$ in the vicinity of the magnetic instability point. Although the presence of the term proportional to the specific heat was proposed by Kakahashi (1989), neither an explicit expression of the logarithmic enhancement of the T-linear coefficient of $\beta(t)$ nor mention of such a behaviour was given. In the weak interaction limit, his result leads to the expression proportional to the thermal spin amplitude squared as given by Moriya and Usami. A part of the anti-invar-like anomalously large volume expansion observed in the paramagnetic phase may be attributed to the term $\omega_t(t)$.
- Magnetovolume coupling constants $C_s(t)$ and $C_h(\sigma, t)$ defined for spontaneous and forced magnetostrictions are different from each other. Both of them show temperature dependence. Similar T^2 dependence has already been predicted by the SEW theory at low temperatures. Our temperature dependence comes from the spin fluctuation mechanism. The forced magnetovolume coupling $C_h(\sigma, t)$ shows an external field dependence as well.
- Pressure effects on σ_s and T_c are controlled by a different set of Grüneisen parameters. The linear relation between $\mathrm{d}\ln T_c/\mathrm{d}p \propto \mathrm{d}\ln\sigma_s/\mathrm{d}p$ does not hold generally.
- We have confirmed the critical forced striction, $\omega \propto \sigma^4$, including its explicit coefficient.
- Because of the multiple Grüneisen parameters, we cannot generally expect a linear relation between the thermal expansion coefficient $\beta(t)$ and the magnetic specific heat in the wide range of temperature.

We would like many existing magnetovolume properties to be examined again according to our new theoretical results presented in this paper to check the validity.

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Appendix. On the strain derivatives of $U_0(t)$ and $V_0(t)$

In order to find initial conditions for the simultaneous differential equations, (52) and (54), we need to find derivatives of $U_0(t)$ and $V_0(t)$ with respect to ω. In this appendix we show briefly how to evaluate them numerically. To begin with, the t-dependence of $U_0(t)$ and $V_0(t)$ defined in (43) is obtained by solving the following non-linear simultaneous equations:

$$\begin{aligned} F_1(U_0, V_0) &= V_0\left[1 - \frac{3}{5c_z}A'(y_{z0}, t) - \frac{2}{5c_z}A'_{\mathrm{c}}(0, t)\right] - U_0 = 0, \\ F_2(U_0, V_0) &= U_0 - \frac{2}{5}V_0 - \frac{3}{5} + \frac{1}{5A(0, t_{\mathrm{c}})}[A(y_{z0}, t) + 2A_t(0, t)] = 0. \end{aligned} \tag{A.1}$$

Their ω-derivatives, $U_\omega = \partial U_0(t)/\partial\omega$ and $V_\omega = \partial V_0(t)/\partial\omega$, are also evaluated with the use of (A.1). The ω-derivative of (A.1) leads to the following coupled equations for U_ω and V_ω:

$$\begin{aligned} &\frac{U_0}{V_0}V_\omega - \frac{V}{5c_z}\left[3\frac{\partial A'(y_{z0}, t)}{\partial\omega} + 2\frac{\partial A'_{\mathrm{c}}(0, t)}{\partial\omega}\right] - U_\omega = 0, \\ &U_\omega - \frac{2}{5}V_\omega + \left(U_0 - \frac{2}{5}V_0 - \frac{3}{5}\right)\left(\frac{\sigma_{s0}^2}{\sigma_s^2}\gamma_m - \gamma_A + \gamma_0\right) \\ &\qquad + \frac{1}{5A(0, t_{\mathrm{c}})}\left[\frac{\partial A(y_{z0}, t)}{\partial\omega} + 2\frac{\partial A_t(0, t)}{\partial\omega}\right] = 0. \end{aligned} \tag{A.2}$$

When $\sigma_s \simeq \sigma_{s0}$ is satisfied, the strain derivatives of the thermal amplitudes, $A(0, t)$ and $A(y_{z0}, t)$ in the above equations, are written by

$$\begin{aligned} \frac{\partial A_t(0, t)}{\partial\omega} &= x_{\mathrm{c}}^2 A'_{\mathrm{c}}(0, t)\left(\gamma_m - \gamma_A + \gamma_0 + \frac{1}{V_0(t)}\frac{\partial V_0(t)}{\partial\omega}\right) + \gamma_0 t\left[\frac{\partial A_{sw}}{\partial t} + \frac{\partial A_{\mathrm{c}}(0, t)}{\partial t}\right], \\ \frac{\partial A(y_{z0}, t)}{\partial\omega} &= y_{z0}A'(y_{z0}, t)\left(\gamma_m - \gamma_A + \gamma_0 + \frac{1}{V_0(t)}\frac{\partial V_0(t)}{\partial\omega}\right) + \gamma_0 t\frac{\partial A(y_{z0}, t)}{\partial t}, \\ \frac{\partial A'_{\mathrm{c}}(0, t)}{\partial\omega} &= x_{\mathrm{c}}^2 A''_{\mathrm{c}}(0, t)\left(\gamma_m - \gamma_A + \gamma_0 + \frac{1}{V_0(t)}\frac{\partial V_0(t)}{\partial\omega}\right) + \gamma_0 t\frac{\partial A'_{\mathrm{c}}(0, t)}{\partial t}, \\ \frac{\partial A'(y_{z0}, t)}{\partial\omega} &= y_{z0}A''(y_{z0}, t)\left(\gamma_m - \gamma_A + \gamma_0 + \frac{1}{V_0(t)}\frac{\partial V_0(t)}{\partial\omega}\right) + \gamma_0 t\frac{\partial A'(y_{z0}, t)}{\partial t}, \end{aligned}$$

where $A'(y, t)$, $A''(y, t), \ldots$, stand for derivatives with respect to the variable y. Substituting the results into (A.2) gives

$$\begin{aligned} &\left\{\frac{U_0(t)}{V_0(t)} - \frac{1}{5c_z}\left[3y_{z0}A''(y_{z0}, t) + 2V_0(t)\frac{\partial A'_{\mathrm{c}}(0, t)}{\partial V}\right]\right\}V_\omega - U_\omega \\ &\qquad = \frac{V_0(t)}{5c_z}(\gamma_m - \gamma_A + \gamma_0)[3y_{z0}A''(y_{z0}, t) + 2x_{\mathrm{c}}^2A''_{\mathrm{c}}(0, t)] \\ &\qquad\quad + \gamma_0\frac{V_0(t)t}{5c_z}\left[2\frac{\partial A'_{\mathrm{c}}(0, t)}{\partial t} + 3\frac{\partial A'(y_{z0}, t)}{\partial t}\right], \\ &\left\{\frac{2}{5} - \frac{1}{5A(0, t_{\mathrm{c}})V_0(t)}[y_{z0}A'(y_{z0}, t) + 2x_{\mathrm{c}}^2A'_{\mathrm{c}}(0, t)]\right\}V_\omega - U_\omega \\ &\qquad = (\gamma_m - \gamma_A + \gamma_0)\left\{U_0(t) - \frac{2}{5}V_0(t) - \frac{3}{5}\right. \\ &\qquad\quad \left. + \frac{1}{5A(0, t_{\mathrm{c}})}[y_{z0}A'(y_{z0}, t) + 2x_{\mathrm{c}}^2A'_{\mathrm{c}}(0, t)]\right\} \\ &\qquad\quad + \gamma_0\frac{t}{5A(0, t_{\mathrm{c}})}\left[2\frac{\partial A_t(0, t)}{\partial t} + \frac{\partial A(y_{z0}, t)}{\partial t}\right]. \end{aligned} \tag{A.3}$$

In a matrix form, (A.3) is simply written as follows.

$$M\begin{pmatrix} U_\omega \\ V_\omega \end{pmatrix} = -\gamma_0 t \begin{pmatrix} \partial F_1/\partial t \\ \partial F_2/\partial t \end{pmatrix} + (\gamma_m - \gamma_A + \gamma_0)\left[\begin{pmatrix} U_0(t) \\ 3/5 - U_0(t) \end{pmatrix} - V_0(t)\begin{pmatrix} \partial F_1/\partial V \\ \partial F_2/\partial V \end{pmatrix}\right]. \tag{A.4}$$

The above 2×2 matrix M is defined by

$$M = \begin{pmatrix} \partial F_1/\partial U & \partial F_1/\partial V \\ \partial F_2/\partial U & \partial F_2/\partial V \end{pmatrix},$$

where $F_1(U_0, V_0)$ and $F_2(U_0, V_0)$ are defined in (A.1). From the property of the matrix algebra, the last term of (A.4) proportional to $V_0(t)$ does not contribute to U_ω and V_ω. The ω derivatives are finally given by

$$\begin{pmatrix} U_\omega \\ V_\omega \end{pmatrix} = -\gamma_0 t M^{-1}\begin{pmatrix} \partial F_1/\partial t \\ \partial F_2/\partial t \end{pmatrix} + (\gamma_m - \gamma_A + \gamma_0)M^{-1}\begin{pmatrix} U_0(t) \\ 3/5 - U_0(t) \end{pmatrix}. \tag{A.5}$$

Once the value of U_ω is evaluated either from (A.3) or (A.5), we can get y_ω from (55), and therefore $C_h(t)$. In this way, all the quantities that we need for the forced magnetostriction are evaluated in terms of thermal spin fluctuation amplitudes and their y- and t-derivatives. These values required for magnetovolume effects are necessary for our numerical calculations of the magnetic entropy and the specific heat.

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