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# Spin-fluctuation theory for weak itinerant-electron ferromagnets: revisited

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**Abstract.** An explicit *self-consistent* calculation of the zero-point (ZP) and thermally excited (TE) contributions to spin fluctuations in weak itinerant-electron (WI) ferromagnets in the presence and absence of an external magnetic field, based on the version of spin-fluctuation theory that makes use of the Ginzburg–Landau formalism, is presented. These calculations get rid of certain major deficiencies of the conventional spin-fluctuation theories by bringing out clearly the roles of ZP and TE excitations. The results so obtained demonstrate that zero-point spin fluctuations have a *major* share in renormalizing the Landau coefficients of the Stoner theory, are relatively *insensitive* to magnetic field, and make an appreciable contribution to the temperature dependence of magnetization in WI ferromagnets. By contrast, thermally excited collective electron–hole pair excitations almost *entirely* account for the dependences of magnetization on temperature and field, and get *strongly suppressed* by magnetic field. The present theoretical approach, in addition, for the first time, yields an *analytical* expression for the suppression of thermally excited spin fluctuations by field for temperatures just outside the critical region but below the Curie point.

## 1. Introduction

Transition metal-based intermetallic compounds such as MnSi, $Ni_3Al$, NiPt, $Sc_3In$, and $ZrZn_2$ exhibit [1, 2]:

(i) a *low* saturation moment per transition metal (TM) atom at 0 K ($<0.4\ \mu_B$/TM atom),
(ii) a *low* magnetic order–disorder phase transition temperature ($T_C < 45$ K),
(iii) a *large* high-field magnetic susceptibility at 0 K,
(iv) a very *large* coefficient of the term *linear* in temperature in the specific heat at low temperatures,
(v) *negative* magnetoresistance,
(vi) $T^{5/3}$-power-law behaviour of the resistivity over a wide temperature range around $T_C$,
(vii) temperature variations going as $T^2$ and $T^{4/3}$ of the spontaneous magnetization *squared* in the intermediate temperature range and at temperatures just outside the critical region but below $T_C$, respectively, and
(viii) Curie–Weiss behaviour of the magnetic susceptibility in the paramagnetic state.

All of these properties are taken to *characterize* the phenomenon called *weak itinerant-electron* ferromagnetism.

The earliest theoretical attempt, due to Stoner [3], to understand some of these attributes dates back to 1939. The Stoner model [3], like its improvised versions [4], could, at best, provide an adequate explanation for the properties (i), (iii), and (iv) mentioned above, but

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*failed* to account for the remaining ones. The failure of Stoner–Wohlfarth theory [3, 4] to correctly predict the behaviour observed at *finite* temperatures was subsequently traced back to its underlying assumptions, that:

(a) the thermally excited electrons and holes move independently in a common mean (exchange) field, and
(b) the magnetic moments diminish in magnitude with increasing temperature and finally disappear at $T_C$ through spin-flip excitations to the Stoner continuum.

While the assumption (a) leads to a susceptibility that does not obey the Curie–Weiss law in the paramagnetic state, the latter assumption advocates a process for the disappearance of the moment at $T = T_C$ that costs too much energy and hence leads to unphysically large values for the Curie temperature. Following this realization, numerous theoretical attempts [1, 2, 5–29] have been made to take into account the *collective* nature of *correlated* electron–hole *pair* excitations (i.e., exchange-enhanced spin-density fluctuations). As a consequence, substantial progress has been made in understanding qualitatively (or even *quantitatively*) most (*some*) of the characteristic properties of weak itinerant-electron ferromagnets. However, certain inconsistencies and inadequacies still remain. For instance, on the one hand, the spin-fluctuation theories [1, 6, 7, 10, 13, 14, 20, 21, 24, 29–31] that completely dispense with the zero-point contribution to spin fluctuations provide an accurate numerical estimate [1, 20, 21, 24, 29] for $T_C$ and a quantitative explanation for the Curie–Weiss (CW) behaviour [1, 6, 7, 13, 14, 20, 21, 29] of the susceptibility and magnetovolume (MV) effects [26, 30, 31] in itinerant-electron ferromagnets. On the other hand, the CW behaviour and MV effects find alternative explanations [22, 32] in terms of a version of spin-fluctuation theory which assumes that

(I) the zero-point spin-fluctuation contribution *depends* [22, 32] on *temperature* through the temperature dependence of the static susceptibility and *dominates* [22, 32] over the thermal spin-fluctuation contribution for temperatures $T \lesssim T_C$, and
(II) the *sum* of zero-point and thermal spin-fluctuation amplitudes remains *constant* [22] over a fairly wide temperature range that spans both ferromagnetic and paramagnetic regimes.

This approach, however, grossly *overestimates* [33] (by nearly a factor of two) the Curie temperature. Thus, the role of zero-point spin fluctuations is far from being completely understood. Furthermore, failure to yield an expression which *quantifies* the suppression of local spin-density fluctuations by the external magnetic field, $H_{ext}$, figures among the major deficiencies of the theoretical models proposed hitherto. This limitation has its roots in the fact that spin fluctuations do not explicitly depend on $H_{ext}$ but, by virtue of their dependence on the magnetization, $M$, indirectly couple to $H_{ext}$ via $M$. Within the framework of the self-consistent renormalization spin-fluctuation theory [6], this difficulty has been partially overcome [1, 34] by using the electron gas model to calculate a field- and temperature-dependent static susceptibility that is *consistent* with the magnetic equation of state. Considering that the electron gas model forms an *oversimplified* description of the band structure of *real* weak itinerant-electron magnetic systems and that a *number* of adjustable parameters [1, 34] have been used to achieve a quantitative agreement with the experiment, such an approach cannot be regarded as satisfactory.

In this paper, we clearly bring out the role of zero-point (ZP) spin-density fluctuations as well as the influence of $H_{ext}$ on thermally excited (TE) spin-density fluctuations through an explicit *self-consistent* calculation of ZP and TE contributions to spin fluctuations in the absence and presence of $H_{ext}$ based on the version of the spin-fluctuation model [20] which makes use of the Ginzburg–Landau formalism. Besides unambiguously demonstrating that the main function of the ZP spin fluctuations is to *renormalize* the Landau coefficients of the

Stoner mean-field theory and, to a significant extent, contribute to the temperature dependence of magnetization, the present calculations yield an *analytical* expression for the suppression of TE spin fluctuations by $H_{ext}$ for temperatures just outside the critical region but below $T_C$ that is vindicated by the recent experiments [35] on the *crystalline* weak itinerant-electron ferromagnet $Ni_3Al$. Moreover, the theoretical approach adopted by us gives a satisfactory account of many hitherto unexplained properties of *amorphous* weak itinerant ferromagnets as well.

## 2. Theoretical formalism

The inherently *small* magnitude of the order parameter (local magnetization) $\vec{M} + \vec{m}(\vec{r})$ with mean $\vec{M}$ and fluctuation amplitude $\vec{m}(\vec{r})$, even at the *lowest* temperature, in weak itinerant-electron ferromagnets substantially enlarges the temperature domain over which the Ginzburg–Landau expansion of the local free energy in powers of the order parameter is valid. As a consequence, the following forms for the total free energy and magnetic equation of state hold [20] over the temperature range that spans both the ferromagnetic and paramagnetic regimes:

$$F(M) = F(0) + \frac{1}{2}[a(T) + b(3\langle m_\parallel^2\rangle + 2\langle m_\perp^2\rangle)]M^2 + \frac{b}{4}M^4 \tag{1}$$

$$F(0) = F_0 + \frac{1}{2}a(T)(\langle m_\parallel^2\rangle + 2\langle m_\perp^2\rangle) + \frac{b}{4}(\langle m_\parallel^2\rangle^2 + 4\langle m_\perp^2\rangle^2 + 4\langle m_\parallel^2\rangle\langle m_\perp^2\rangle) \tag{2}$$

and

$$\frac{1}{M}\frac{\partial F}{\partial M} = \frac{H}{M(T,H)} = a(T) + b[(3\langle m_\parallel^2\rangle + 2\langle m_\perp^2\rangle) + M^2(T,H)] \tag{3}$$

where $F_0$ is the contribution to the free energy which is *independent* of the order parameter, and $a$ and $b$, the Landau coefficients in the Stoner theory [3, 4], are given by

$$a(T) = -[2\chi(0,0)]^{-1}[1 - (T/T_C^S)^2 - \mathcal{B}ST^4] \tag{4}$$

$$b = [2\chi(0,0)M^2(0,0)]^{-1} \tag{5}$$

and

$$\chi(0,0) = N\mu_B^2 N(E_F)(T_F/T_C^S)^2 = N\mu_B^2 N(E_F)\mathcal{S} = (\chi_p/2)\mathcal{S} \tag{6}$$

$$M^2(0,0) = (N\mu_B\mu_0)^2 = (\mathcal{S}\gamma)^{-1} \tag{7}$$

$$\mathcal{S} = [IN(E_F) - 1]^{-1}$$

$$\gamma = (1/2)\{N\mu_B N(E_F)\}^{-2}\{[N'(E_F)/N(E_F)]^2 - [N''(E_F)/3N(E_F)]\}. \tag{8}$$

In the above expressions, $\chi(0,0)$ and $\mu_0$ are the zero-field differential susceptibility and moment per alloy atom at 0 K, respectively, $H$ stands for the external magnetic field ($H_{ext}$) corrected for demagnetization and other anisotropy fields present, $\mathcal{S}$ is the Stoner enhancement factor, $I$ (the Stoner parameter) is a measure of the exchange splitting of the bands, $N$ is the number of atoms per unit volume, $N(E_F)$ is the density of single-particle states (DOS) at the Fermi level $E_F$ and $N'(E_F)$ ($N''(E_F)$) is its first (second) energy derivative, $T_C^S$ is the Stoner Curie temperature, the coefficient $\mathcal{B}$ of the $T^4$-term in equation (4) involves derivatives of the DOS at $E_F$ up to fourth order and its explicit form is given in reference [36], $\chi_p$ is the Pauli susceptibility, and the thermal variances of the local magnetization parallel ($\parallel$), $\langle m_\parallel^2\rangle$, and perpendicular ($\perp$), $\langle m_\perp^2\rangle$, to the average magnetization, $\vec{M}$, are related to the imaginary part of the dynamical wave-vector-dependent susceptibility, Im $\chi_\nu(\vec{q},\omega)$, where $\nu$ ($=\parallel, \perp$) is the polarization index, through the well-known fluctuation-dissipation relation [1, 14, 20–22]:

$$\langle m_\nu^2\rangle = 4\hbar\int\frac{\mathrm{d}^3\vec{q}}{(2\pi)^3}\int\frac{\mathrm{d}\omega}{2\pi}\left(n(\omega) + \frac{1}{2}\right)\mathrm{Im}\,\chi_\nu(\vec{q},\omega) \tag{9}$$

with

$$n(\omega) = [\exp(\hbar\omega/k_B T) - 1]^{-1} \tag{10}$$

$$\text{Im}\,\chi_\nu(\vec{q},\omega) = \omega\chi_\nu(\vec{q})\frac{\Gamma_\nu(\vec{q})}{\omega^2 + \Gamma_\nu^2(\vec{q})} \tag{11}$$

$$\chi_\nu(\vec{q}) = \chi_\nu(\vec{q},\omega=0) = \chi_\nu(0)\frac{\kappa_\nu^2}{\kappa_\nu^2 + q^2} \tag{12}$$

$$\Gamma_\nu(\vec{q}) = \gamma_\nu q\,\chi_\nu^{-1}(\vec{q}) = \Gamma_\nu(0)q(\kappa_\nu^2 + q^2) \tag{13}$$

$$\chi_\nu(0) = \chi_\nu(\vec{q}=0) = (c_\nu\kappa_\nu^2)^{-1} \tag{14}$$

$$\Gamma_\nu(0) = \Gamma_\nu(\vec{q}=0) = c_\nu\gamma_\nu \tag{15}$$

where $n(\omega)$ is the Bose function, $\Gamma_\nu(\vec{q})$ is the relaxation frequency of a spontaneous spin fluctuation of wave vector $\vec{q}$ and polarization $\nu$, $\chi_\nu(0)$ is the field- and temperature-dependent susceptibility (i.e., $\chi_\nu(0) \equiv \chi_\nu(T,H)$), $c_\nu$ is the coefficient of the gradient term in the Ginzburg–Landau expansion, and the quantity $\gamma_\nu$ depends [20] on the shape of the DOS curve near $E_F$. Equations (11)–(15), obtained by using the self-consistent random-phase approximation [6] (RPA) or by making use of a single-band model and assuming $\langle m_\nu^2\rangle$, $\vec{q}$, and $\omega$ to be *small* [20], correctly describe the results of neutron scattering experiments on archetypal weak itinerant-electron ferromagnets. According to equation (9), spin fluctuations are made up of two *components*, the zero-point (ZP) spin fluctuations, $\langle m_\nu^2\rangle^{ZP}$, and thermally excited (TE) spin fluctuations, $\langle m_\nu^2\rangle^{TE}$, represented in equation (9) by the factors 1/2 and $n(\omega)$, respectively.

### *2.1. Zero-point spin fluctuations*

Zero-point spin fluctuations can be further split into two parts: quantum fluctuations at $T = 0$ K, $\langle m_\nu^2\rangle_0^{ZP}$, and the temperature-induced changes in the zero-point spin (quantum) fluctuations at finite temperatures $T \neq 0$, $\langle m_\nu^2\rangle_T^{ZP}$. Specifically,

$$\langle m_\nu^2\rangle^{ZP} = \langle m_\nu^2\rangle_0^{ZP} + \langle m_\nu^2\rangle_T^{ZP} = \frac{4\hbar\gamma_\nu}{(2\pi)^3}\int_0^{q_c^{ZP}} q^3\,\mathrm{d}q\int_0^{w_c(q)}\frac{\omega\,\mathrm{d}\omega}{\omega^2 + \Gamma_\nu^2(\vec{q})} \tag{16}$$

where $\omega_c(q)$ is the *wave-vector-dependent* upper bound of the relaxation frequency spectrum and $q_c^{ZP}$ is the *temperature- and field-dependent* cut-off wave vector with the property that it approaches a very small but *finite* value $q_0$ as $T \to 0$ K and can possess as large a value as the average radius, $q_B$, of the Brillouin zone at high temperatures (i.e., at $T \gg T_C$). The small magnitude of $q_0$ implies that quantum fluctuations at $T = 0$ K occupy a very small region, centred at $q = 0$, of the Brillouin zone. Deferring the description of the actual functional form of $q_c^{ZP}(T,H) = q_0(0,H) + q_c(T,H)$ to a later section and making use of the reduced variable $x = q/q_c^{ZP}$, equation (16) can be put into the form

$$\langle m_\nu^2\rangle^{ZP} = \frac{2\hbar\gamma_\nu}{(2\pi)^3}(q_c^{ZP})^4\left[\int_0^1 x^3\ln\left[\left(\frac{\omega_c(q)}{A_\nu x}\right)^2 + (x^2+y^2)^2\right]\mathrm{d}x - 2\int_0^1 x^3\ln(x^2+y^2)\,\mathrm{d}x\right] \tag{17}$$

with $y = \kappa_\nu/q_c^{ZP}$ and $A_\nu = \Gamma_\nu(0)(q_c^{ZP})^3$.

Now that the minimum time that ballistic electrons at the Fermi surface take to travel one wavelength $(2\pi/q)$ is $2\pi/qv_F$, the upper bound on the relaxation frequency $\omega_c(q)$ should equal $qv_F$, where $v_F = 3n/2\hbar k_F N(E_F)$ is the Fermi velocity and $N(E_F)$ is the density of states at the Fermi level, $E_F$, per spin per atom. Use of the equality $\omega_c(q) = qv_F$ leads to the

exact result

$$\langle m_\nu^2\rangle^{ZP} = \frac{\hbar\gamma_\nu}{2(2\pi)^3}(q_c^{ZP})^4\left[a_\nu^2\ln\left(\frac{1+u^2}{1+v^2}\right) + a_\nu u(1-a_\nu v)\ln\left(1+\frac{1}{u^2}\right) + a_\nu^2 v^2\ln\left(1+\frac{1}{v^2}\right) - \frac{4a_\nu}{c_\nu(q_c^{ZP})^2\chi_\nu(0)}\tan^{-1}\left(\frac{1/a_\nu}{1+uv}\right)\right] \tag{18}$$

with

$$a_\nu = \frac{v_F}{c_\nu\gamma_\nu(q_c^{ZP})^2} \tag{19}$$

$$u = \frac{1+y^2}{a_\nu} = \frac{\gamma_\nu}{v_F\chi(q_c^{ZP})} \tag{20}$$

$$v = \frac{y^2}{a_\nu} = \frac{\gamma_\nu}{v_F\chi_\nu(0)} \tag{21}$$

which is valid over the entire temperature range extending from $T = 0$ K to temperatures well above $T_C$. In arriving at equation (18) use has been made of the identity

$$\tan^{-1}\alpha - \tan^{-1}\beta = \tan^{-1}[(\alpha-\beta)/(1+\alpha\beta)]$$

and the well-known results

$$\int \ln(x^2+a^2)\,\mathrm{d}x = x\ln(x^2+a^2) - 2x + 2a\tan^{-1}(x/a)$$

$$\int x\ln(x^2+a^2)\,\mathrm{d}x = (1/2)[(x^2+a^2)\ln(x^2+a^2) - x^2)]$$

$$\int x^3\ln(x^2+a^2)\,\mathrm{d}x = (1/4)[(x^4-a^4)\ln(x^2+a^2) + a^2x^2 - (x^4/2)].$$

Note that $a_\nu$ is very *large* because $v_F$ is extremely large while $y$ is *small* compared to unity at temperatures not very far from $T_C$ and in external magnetic fields of moderate strength because $\chi_\nu^{-1}(0)$ is small (note that $\chi_\nu^{-1}(0) = 0$, and hence $y = 0$, at $T = T_C$ in the absence of an external magnetic field). Under these conditions, $v < u < 1$ and logarithmic functions can be expanded in powers of $y$ to yield the result

$$\langle m_\nu^2\rangle^{ZP} \cong \left[\frac{q_c^{ZP}(T,H)}{q_c^{ZP}(T=T_C,H=0)}\right]^4\langle m_\nu^2\rangle_{y=0}^{ZP} - \xi^{ZP}\chi_\nu^{-1}(0) \tag{22}$$

with

$$\langle m_\nu^2\rangle_{y=0}^{ZP} = [q_c^{ZP}(T=T_C,H=0)]^4 f \tag{23}$$

$$f = \frac{\hbar\gamma_\nu}{2(2\pi)^3}\mathcal{F}$$

$$\mathcal{F} = [a_\nu^2\ln(1+a_\nu^{-2}) + \ln(1+a_\nu^2)] \tag{24}$$

$$\xi^{ZP} = [q_c^{ZP}(T,H)]^2 g \tag{25}$$

$$g = \frac{2\hbar\gamma_\nu}{(2\pi)^3 c_\nu}\mathcal{G}$$

$$\mathcal{G} = [a_\nu\tan^{-1}(1/a_\nu)]. \tag{26}$$

In a later section, the expressions (22)–(26) will be used to bring out clearly the role of zero-point spin fluctuations in affecting magnetic properties of weak itinerant-electron ferromagnets. The next subsection is devoted to the calculation of the contributions to $M(T, H)$ arising from thermally excited spin fluctuations in different temperature ranges.

### *2.2. Thermally excited spin fluctuations*

*2.2.1. Low temperatures.* At low temperatures, the main contribution to $\langle m_v^2\rangle^{TE}$ arises from long-wavelength ($q \lesssim q_{SW}$) low-frequency *undamped* (*propagating*) transverse modes, i.e., spin waves (SW). Such a contribution is obtained from equation (9) by dropping the factor 1/2 and inserting the following expression [20] for Im $\chi_v(\vec{q}, \omega)$ in this equation, and then evaluating the integrals:

$$\text{Im}\,\chi_\perp(\vec{q}, \omega) = \frac{\pi}{2}\omega\chi_\perp(\vec{q})[\delta(\omega - \omega(\vec{q})) + \delta(\omega + \omega(\vec{q}))] \tag{27}$$

with the spin-wave propagation frequency $\omega(\vec{q})$ given by [20]

$$\begin{aligned}\hbar\omega(\vec{q}) &= g\mu_B M(T, H)\chi_\perp^{-1}(\vec{q}) = g\mu_B M(T, H)(\chi_\perp^{-1} + c_\perp q^2 + \cdots)\\ &= g\mu_B H + D_\perp^{SW}(T, H)q^2 + \cdots\end{aligned} \tag{28}$$

where $\chi_\perp^{-1} = H/M(T, H)$, the *effective* field $H$ is the external magnetic field, $H_{ext}$, corrected for the demagnetizing field, $H_{dem}$, and other anisotropy fields, $H_A$, i.e.,

$$H = H_{ext} - H_{dem} + H_A = H_{ext} - 4\pi N M(T, H_{ext}) + H_A$$

and $N$ is the demagnetizing factor, $g$ is the Landé splitting factor, and

$$D_\perp^{SW}(T, H) = g\mu_B M(T, H)c_\perp$$

is the spin-wave stiffness. Use of equations (9), (27), and (28) culminates in the result

$$\langle m_\perp^2\rangle_{SW}^{TE} = \zeta(3/2)g\mu_B M(T, 0)\left[\frac{k_B T}{4\pi D_\perp^{SW}(T, 0)}\right]^{3/2}. \tag{29}$$

At low temperatures, $T \ll T_C^S$ and the term $\mathcal{B}ST^4$ in equation (4) is negligibly small, so $a(T) \simeq -[2\chi(0, 0)]^{-1}$. With this value of $a(T)$ and the expansion coefficient $b$ given by equation (5), the magnetic equation of state, equation (3), when $H = 0$, can be approximated by

$$\frac{M(T, 0)}{M(0, 0)} \simeq 1 - \frac{\langle m_\perp^2\rangle}{M^2(0, 0)} - \frac{3}{2}\frac{\langle m_\parallel^2\rangle}{M^2(0, 0)}. \tag{30}$$

Now that the thermal demagnetization of the spontaneous magnetization in crystalline (homogeneous) ferromagnets at low temperatures is mainly due to spin-wave excitations (propagating transverse spin-density fluctuations), the second term in equation (30) is considerably larger in magnitude than the third, and hence the latter term can be dispensed with. Equation (29), when combined with equation (30), yields the well-known Bloch $T^{3/2}$-power law

$$\frac{M(T, 0)}{M(0, 0)} = 1 - \frac{g\mu_B}{M(0, 0)}\zeta(3/2)\left[\frac{k_B T}{4\pi D_\perp^{SW}(T, 0)}\right]^{3/2} \tag{31}$$

where $\zeta(3/2)$ is the Riemann zeta function and $D_\perp^{SW}(T, 0) = g\mu_B c_\perp M(T, 0)$. However, in the presence of an external magnetic field, an energy gap of magnitude $g\mu_B H$ appears in the spin-wave spectrum, as is evident from equation (28), and equations (9), (10), (27), (28) and (3) lead to the expression

$$\frac{M(T, H)}{M(0, H)} = 1 - \frac{g\mu_B}{M(0, H)}Z(3/2, t_H)\left[\frac{k_B T}{4\pi D_\perp^{SW}(T, H)}\right]^{3/2} \tag{32}$$

where the Bose–Einstein integral function

$$Z(3/2, t_H) = \zeta(3/2)F(3/2, t_H) = \sum_{n=1}^{\infty} n^{-3/2}\mathrm{e}^{-nt_H}$$

with $t_H = k_B T_g / k_B T = g\mu_B H / k_B T$ allows for the energy gap in the spin-wave spectrum introduced by $H_{ext}$.

In the case of crystalline or amorphous ferromagnets with competing interactions and/or Invar characteristics, the contribution to $M(T, 0)$ due to longitudinal spin fluctuations, i.e., the third term in equation (30), cannot be ignored even at low temperatures for the following reason. Owing to the constraints imposed by the competing interactions, the orientation of a given magnetic moment in such systems is, in general, not parallel to the direction of bulk magnetization. As a consequence, the displacements of the longitudinal component of magnetization from the local equilibrium value are of the same order of magnitude as the transverse displacements which give rise to spin waves. Thus, as will be apparent from the following treatment, the *diffusive* (overdamped) modes ('*diffusons*'), hydrodynamic in origin, associated with the longitudinal component of the magnetization contribute to the $T^{3/2}$-decrease of the magnetization as *significantly* as the propagating (undamped) transverse (modes) spin fluctuations (spin waves) do. The contribution to $\langle m_v^2 \rangle^{TE}$ on account of diffusons is given by the modified versions of equations (9) and (11), i.e., by the expression

$$\langle m_\parallel^2 \rangle_{D_i}^{TE} = 4\hbar \int_0^\infty \frac{\mathrm{d}^3\vec{q}}{(2\pi)^3} \int_0^\infty \frac{\mathrm{d}\omega}{2\pi} \frac{1}{\mathrm{e}^{\hbar\omega/k_B T} - 1} \omega \chi_\parallel(\vec{q}) \frac{\Gamma_\parallel(\vec{q})}{\omega^2 + \Gamma_\parallel^2(\vec{q})} \tag{33}$$

where [37] $\Gamma_\parallel(\vec{q}) = D_i q^2 = \mathcal{A}\chi_\parallel^{-1}(\vec{q})$, $\mathcal{A}$ is a constant, and $D_i$ is the diffusion coefficient. Use of the standard result

$$\int_0^\infty u^2[\ln u^2 - (1/2u^2) - \psi(u^2)]\,\mathrm{d}u = (1/4(2\pi)^{1/2})\Gamma(3/2)\zeta(3/2)/\cos(\pi/4)$$

where $u = (\hbar D_i / 2\pi k_B T)^{1/2} q$ and $\psi(z)$ is Euler's psi function, permits exact evaluation of the integrals in equation (33) such that

$$\langle m_\parallel^2 \rangle_{D_i}^{TE} = \frac{\mathcal{A}}{2\sqrt{\hbar}} \zeta(3/2) \left( \frac{k_B T}{2\pi D_i} \right)^{3/2}. \tag{34}$$

The spontaneous magnetization is given by the following expression when equations (29) and (34) are used to represent the contributions $\langle m_\perp^2 \rangle$ and $\langle m_\parallel^2 \rangle$ in equation (30):

$$\frac{M(T,0)}{M(0,0)} = 1 - \frac{g\mu_B}{M(0,0)} \zeta\left(\frac{3}{2}\right) \left[ \frac{k_B T}{4\pi D_\perp^{SW}(T,0)} \right]^{3/2} - \frac{3\mathcal{A}}{4\sqrt{\hbar}} [M(0,0)]^{-2} \zeta\left(\frac{3}{2}\right) \left[ \frac{k_B T}{2\pi D_i(T,0)} \right]^{3/2}. \tag{35}$$

Equation (35) demonstrates that the thermal demagnetization of the spontaneous magnetization is *faster* in spin systems in which, besides magnons, diffusons contribute [38] to the $T^{3/2}$-dependence of $M(T, 0)$. However, unlike magnons, diffusons show up as a broad central (elastic) peak [37] in the inelastic neutron scattering (INS) intensity versus neutron energy isotherms taken at constant values of $q$. As a consequence, the value of $D$ deduced from the magnetization data, $D_M$, should be *substantially lower* than that ($D_N$) measured in the INS experiments on such systems. Such a large discrepancy between the values of $D_M$ and $D_N$, i.e., $D_N \gg D_M$, has indeed been found [39–42] in a number of Invar systems. Since $\mathcal{A}$ (and hence $D_i$) has a very weak or even no dependence on the external magnetic field, the contribution to $M(T, H)$ due to diffusons, unlike magnons, is nearly *field independent*.

*2.2.2. Intermediate temperatures.* In the intermediate range of temperatures, the spin-wave contribution is completely overshadowed by the one arising from spin fluctuations (SF). In

view of equations (9)–(11), the latter contribution to $\langle m_\nu^2\rangle^{TE}$ is given by

$$\langle m_\nu^2\rangle_{SF}^{TE} = \frac{8\hbar\gamma_\nu}{(2\pi)^3}\int q^3\,\mathrm{d}q\int_0^{\omega_c(q)}\frac{\omega\,\mathrm{d}\omega}{(\mathrm{e}^{\hbar\omega/k_BT}-1)(\omega^2+\Gamma_\nu^2(\vec{q}))}. \tag{36}$$

Equation (36) is not amenable to analytical solution unless the so-called *classical* approximation is made. This approximation implies that each mode $m_\nu(\vec{q})$ for $\vec{q} < \vec{q}_c$ is thermally excited such that the Bose function $n(\omega)$, equation (10), can be approximated by $k_BT/\hbar\omega$ for those values of $\omega$ for which Im $\chi_\nu(\vec{q},\omega)$ makes an appreciable contribution to the integral over $\omega$ in equation (9). Moreover, $\omega_c(q) = v_Fq$ is extremely large, in general, and hence the upper limit for $\omega$-integration can be replaced by $\infty$ without sacrificing accuracy. With these considerations, equation (36) reduces to

$$\langle m_\nu^2\rangle_{SF}^{TE} = \frac{8\gamma_\nu}{(2\pi)^3}(k_BT)\int_0^{q_c^{TE}} q^3\,\mathrm{d}q\int_0^\infty\frac{\mathrm{d}\omega}{\omega^2+\Gamma_\nu^2(\vec{q})} = \frac{k_BT}{2\pi^2}\int_0^{q_c^{TE}}\frac{q^2\,\mathrm{d}q}{\chi_\nu^{-1}(\vec{q})}. \tag{37}$$

By making use of equations (12) and (14), the integral over $q$ in equation (37) can be evaluated, with the result

$$\langle m_\nu^2\rangle_{SF}^{TE} = \frac{k_BT}{2\pi^2c_\nu}[q_c^{TE} - \kappa_\nu\tan^{-1}(q_c^{TE}/\kappa_\nu)]. \tag{38}$$

At intermediate temperatures, $\kappa_\nu$ is usually large, so $\tan^{-1}z$ with $z < 1$ can be expanded in powers of $z$ to estimate $\langle m_\nu^2\rangle_{SF}^{TE}$, i.e.,

$$\langle m_\nu^2\rangle_{SF}^{TE} \simeq \frac{k_BT}{6\pi^2}\chi_\nu(0)(q_c^{TE})^3 \tag{39}$$

if only the first two leading terms in the expansion are retained. Recalling that $\chi_\nu(0) \equiv \chi_\nu(T,H)$ and $q_c^{TE} \equiv q_c(T,H)$, the variations of $\langle m_\nu^2\rangle_{SF}^{TE}$ with temperature (besides the factor $k_BT$) and field are basically governed by the functional dependences of $\chi_\nu(0)$ and $q_c$ on $T$ and $H$.

*2.2.3. Temperatures outside the critical region but not very far from $T_C$.* In the absence of an external magnetic field and for temperatures close to the Curie point $T_C$ ($T < T_C$) but still away from criticality, where $\chi_\nu^{-1}(0) \cong 0$, equations (9)–(11) yield

$$\langle m_\nu^2\rangle_{SF}^{TE} = \frac{4\hbar\gamma_\nu}{(2\pi)^4}\int_0^\infty q\left[\int_0^\infty\frac{\omega\,\mathrm{d}\omega}{(\mathrm{e}^{\hbar\omega/k_BT}-1)(\omega^2+\Gamma_\nu^2(\vec{q}))}\right]\mathrm{d}^3\vec{q}$$

if the upper limit of integration over $q$ is taken to be infinity rather than $q_B$. This replacement introduces negligible error in most cases. While the result

$$\int_0^\infty t\,\mathrm{d}t/[(t^2+u^2)(\mathrm{e}^{2\pi t}-1)] = (1/2)[\ln u - (1/2u) - \psi(u)]$$

facilitates evaluation of the integral over $\omega$, change of the integration variable from $q$ to $z = (\hbar c_\nu\gamma_\nu/2\pi k_BT)^{1/3}q$ puts the integral over $q$ in the standard form, so

$$\begin{aligned}\langle m_\nu^2\rangle_{SF}^{TE} &= \frac{4\hbar\gamma_\nu}{(2\pi)^3}\left(\frac{2\pi k_BT}{\hbar c_\nu\gamma_\nu}\right)^{4/3}\int_0^\infty\mathrm{d}z\,z^3\left[\ln z^3 - \frac{1}{2z^3} - \psi(z^3)\right]\\ &= \frac{1}{3^{3/2}\pi^2}\Gamma(4/3)\zeta(4/3)(\hbar\gamma_\nu)^{-1/3}\left(\frac{k_B}{c_\nu}\right)^{4/3}T^{4/3}.\end{aligned} \tag{40}$$

By contrast, in the presence of $H_{ext}$, $\chi_\nu^{-1}(0)$ remains *finite* for the range of temperatures in question, and hence equations (9)–(11) can be analytically solved only when the *classical approximation* is used and the cut-off wave vector $q_c$ is allowed to vary with temperature and

field. Such an exercise leads to equation (37), as shown in the preceding subsection. Before proceeding with the solution of the integral appearing in equation (37), we rewrite equation (12) in the form

$$g\mu_B M(T, H)\chi_\nu^{-1}(\vec{q}) = g\mu_B M(T, H)\chi_\nu^{-1}(0) + (g\mu_B M(T, H)c_\nu)q^2$$

and in analogy with the spin-wave dispersion relation, $\hbar\omega(\vec{q}) = g\mu_B H + D_\perp^{SW} q^2$, equation (28), define the spin-fluctuation stiffness as $D_\nu^{SF} = g\mu_B M(T, H)c_\nu$. Furthermore, we assume that for fields of moderate strength

$$\chi_\parallel^{-1}(T, H)\ (\equiv(\chi_\parallel^{-1}(0)) = \partial H/\partial M \cong \chi_\perp^{-1}(T, H)\ (\equiv(\chi_\perp^{-1}(0)) = H/M$$

i.e., longitudinal and transverse spin fluctuations are treated on the same footing in the temperature and field ranges under consideration, with the result that the so-called spin-fluctuation dispersion relation takes the form

$$g\mu_B M(T, H)\chi_\nu^{-1}(\vec{q}) = g\mu_B H + D_\nu^{SF} q^2.$$

In this approximation, equation (37) can be cast into the form

$$\begin{aligned}\langle m_\nu^2\rangle_{SF}^{TE} &= \frac{k_B T}{2\pi^2} g\mu_B M(T, H)\int_0^{q_c} \frac{q^2\,\mathrm{d}q}{g\mu_B H + D_\nu^{SF} q^2} \\ &= \frac{k_B T}{2\pi^2 c_\nu}\left[q_c - (g\mu_B H/D_\nu^{SF})^{1/2}\tan^{-1}(q_c/(g\mu_B H/D_\nu^{SF})^{1/2})\right]. \end{aligned} \tag{41}$$

Equation (41) is an alternative form of equation (38) obtained by replacing $\kappa_\nu$ in equation (38) by $\kappa_\nu = (c_\nu\chi_\nu(0))^{-1/2} = (g\mu_B H/D_\nu^{SF})^{1/2}$. In the range of temperatures close to $T_C$ but outside the critical region as well as for weak and intermediate fields, the argument $x$ of the function $\tan^{-1} x$ in equation (41) has a *sizable magnitude*, so $\tan^{-1} x \cong \pi/2 - (1/x) + \cdots$ and equation (41) reduces to

$$\langle m_\nu^2\rangle_{SF}^{TE} = \left(\frac{k_B T}{2\pi^2}\right)\left(\frac{q_c}{c_\nu}\right)\left[1 - \frac{\pi}{2q_c}\left(\frac{g\mu_B}{D_\nu^{SF}}\right)^{1/2} H^{1/2} + \frac{1}{q_c^2}\left(\frac{g\mu_B}{D_\nu^{SF}}\right)H\right]. \tag{42}$$

In addition to the factors $k_B T$, $H^{1/2}$, and $H$, $q_c(T, H)$ contributes significantly to the temperature and field variations of $\langle m_\nu^2\rangle_{SF}^{TE}$.

### *2.3. Thermal variation of magnetization in the absence and presence of $H_{ext}$*

From the expressions derived for the zero-point and thermal components of spin fluctuations in the subsections 2.1 and 2.2, it is evident that prior knowledge about the actual functional form of $q_c(T, H)$ is needed for arriving at the variations of these components (and hence of the magnetization) with temperature and field.

*2.3.1. Dependence of the cut-off wave vector on temperature and field.* The temperature and field dependences of the cut-off wave-vector $q_c$ are given by the condition [20]

$$\hbar\Gamma_\nu(q_c) \cong k_B T. \tag{43}$$

With the aid of equation (13), the above equality can be put into the form

$$q_c^3 + \kappa_\nu^2 q_c - (k_B T/\hbar\Gamma_\nu(0)) = 0. \tag{44}$$

This cubic equation has only one *real* root, namely

$$q_c(T, H) = q_c(T, H = 0)\left[1 - Z + \frac{Z^3}{3}\right] \tag{45}$$

with

$$q_c(T, H=0) = \left(\frac{k_B T}{\hbar \Gamma_\nu(0)}\right)^{1/3} = \left(\frac{k_B T}{\hbar c_\nu \gamma_\nu}\right)^{1/3} \tag{46}$$

$$Z = \frac{1}{3c_\nu}\left(\frac{\hbar \gamma_\nu c_\nu}{k_B T}\right)^{2/3} \chi_\nu^{-1}(0). \tag{47}$$

Apart from bringing out clearly the functional dependence of $q_c$ on temperature, equations (45) to (47) assert that the variation of $q_c$ with field is basically governed by the field dependence of $\chi_\nu^{-1}(0)$. Moreover, at a given temperature, $\chi_\nu^{-1}(0)$ increases as $H$ increases and hence, according to equations (45)–(47), the cut-off wave vector progressively *diminishes* compared to its value at $H = 0$ as $H$ is increased. Alternatively, the effect of the field is to strongly *suppress* particularly those spin-fluctuation modes that have $q$ close to $q_c(T, H)$. Since $Z < 1$ over a fairly wide temperature range below $T_C$, the third term in equation (45) is not considered in subsequent calculations.

*2.3.2. Temperature evolution, and suppression by magnetic field, of collective electron–hole pair excitations.* The explicit functional form of $q_c(T, H)$ permits one to arrive at the following expressions for the contribution to $M(T, H)$ due to spin-density fluctuations in different temperature ranges. At intermediate temperatures,

$$\begin{aligned}
\langle m_\nu^2 \rangle &= \langle m_\nu^2 \rangle^{ZP} + \langle m_\nu^2 \rangle^{TE} \\
&= \left[ f\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{4/3} - g\left[ q_0^2 + 2q_0\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{1/3} + \left\{ 1 + \frac{4}{3c_\nu}\left(\frac{f}{g}\right)\right\}\right.\right. \\
&\quad \left.\left. \times \left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{2/3}\right] \chi_\nu^{-1}(0) \right]^{ZP} + \left[ \frac{1}{6\pi^2}\left(\frac{k_B^2 \chi_\nu(0)}{\hbar \gamma_\nu c_\nu}\right) T^2 \right. \\
&\quad \left. - \frac{(\hbar \gamma_\nu)^{-1/3}}{6\pi^2}\left(\frac{k_B}{c_\nu}\right)^{4/3} T^{4/3} + \frac{\hbar \gamma_\nu}{18\pi^2}[c_\nu \chi_\nu(0)]^{-1}\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{2/3} \right]^{TE} \\
&= \left[ \frac{1}{6\pi^2}\left(\frac{k_B^2 \chi_\nu(0)}{\hbar \gamma_\nu c_\nu}\right) T^2 + \frac{1}{\pi^2}\left(\frac{3\mathcal{F} - 8\pi}{48\pi}\right)(\hbar \gamma_\nu)^{-1/3}\left(\frac{k_B}{c_\nu}\right)^{4/3} T^{4/3} \right] \\
&\quad - g\left[ q_0^2 + 2q_0\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{1/3} + \left\{ 1 + \frac{3\mathcal{F} - 2\pi}{9\mathcal{G}} \right\}\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{2/3}\right] \chi_\nu^{-1}(0).
\end{aligned} \tag{48}$$

By contrast, for temperatures close to, and below, $T_C$ but outside the critical region,

$$\begin{aligned}
\langle m_\nu^2 \rangle &= \left[ f\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{4/3} - g\left[ q_0^2 + 2q_0\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{1/3} + \left\{ 1 + \frac{4}{3c_\nu}\left(\frac{f}{g}\right)\right\}\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{2/3}\right] \right. \\
&\quad \left. \times \chi_\nu^{-1}(0) \right]^{ZP} + \left[ \frac{(\hbar \gamma_\nu)^{-1/3}}{2\pi^2}\left(\frac{k_B}{c_\nu}\right)^{4/3} T^{4/3}\left[ 1 - \frac{\pi}{2}\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{-1/3} \right.\right. \\
&\quad \left.\left. \times \left(\frac{g\mu_B}{D_\nu^{SF}}\right)^{1/2}\sqrt{H}\right] + \frac{2}{3c_\nu}\left(\frac{\hbar \gamma_\nu}{2\pi^2}\right)\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{2/3} \chi_\nu^{-1}(0) \right]^{TE} \\
&= \left[ \frac{\mathcal{F} + 8\pi}{16\pi^3}(\hbar \gamma_\nu)^{-1/3}\left(\frac{k_B}{c_\nu}\right)^{4/3} T^{4/3}\left[ 1 - \left(\frac{4\pi^2}{\mathcal{F} + 8\pi}\right)\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{-1/3} \right.\right. \\
&\quad \left.\left. \times \left(\frac{g\mu_B}{D_\nu^{SF}}\right)^{1/2}\sqrt{H}\right]\right] - g\left[ q_0^2 + 2q_0\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{1/3} \right. \\
&\quad \left. + \left\{ 1 + \frac{\mathcal{F} - 4\pi}{3\mathcal{G}} \right\}\left(\frac{k_B T}{\hbar \gamma_\nu c_\nu}\right)^{2/3}\right] \chi_\nu^{-1}(0).
\end{aligned} \tag{49}$$

Expressions (48) and (49) are obtained by inserting equations (45)–(47) into equations (39) and (42), respectively, and dropping out terms that involve powers of $\chi_\nu^{-1}(0)$ higher than *one*. Note that the parameters $f$, $\mathcal{F}$, $g$, and $\mathcal{G}$ have already been defined in equations (24) and (26), and $\gamma_\nu c_\nu$ in equation (15). It is evident from the above expressions for $\langle m_\nu^2 \rangle$ that:

(i) both zero-point (ZP) and thermally excited (TE) components *significantly* contribute to the temperature dependence of $\langle m_\nu^2 \rangle$ such that the latter contribution *dominates* over the former over the entire temperature range,
(ii) ZP and TE components make *competing claims* to the $T^{4/3}$-term in the expression for $\langle m_\nu^2 \rangle$ at intermediate temperatures whereas their contributions to the same term at $H = 0$ *supplement* each other for $T \lesssim T_C$,
(iii) the field dependence of $\chi_\nu(0)$ is solely responsible for the variation of $\langle m_\nu^2 \rangle$ with $H$, and
(iv) in zero field, equation (49), but for a slight difference in the numerical factor, has exactly the *same* form as equation (40).

The observation (ii) stated above is a direct consequence of the following fact. The temperature-induced alterations in the zero-point (quantum) spin fluctuations in both zero and finite fields give a contribution to $\langle m_\nu^2 \rangle$ that grows with $T$ as $T^{4/3}$ over the entire temperature range. By contrast, the functional dependence of the contribution to $\langle m_\nu^2 \rangle$ arising from the thermally excited spin-density fluctuations on temperature, $\langle m_\nu^2 \rangle^{TE}(T)$, varies from one temperature range to the other and is *sensitive* to field as well. To elucidate this point further, in *zero* field, $\langle m_\nu^2 \rangle^{TE}$ varies with temperature as $T^{4/3}$ for temperatures close to $T_C$ whereas, in *finite* fields, it *decreases* with increasing temperature as $T^{4/3}$ at all temperatures below $T_C$. Regardless of the temperature range, change in the sign of the coefficient of the $T^{4/3}$-term when the field is 'switched on' reflects the *suppression* of thermally excited spin fluctuations by field. That field suppresses spin fluctuations is obvious from the $\sqrt{H}$-dependence of the coefficient of the $T^{4/3}$-term in equation (49) but such is not the case for the coefficient of the $T^{4/3}$-term in equation (48) because it is essentially *field independent*. At this stage, it should be recalled that:

(a) the $-T^{4/3}$-term in $\langle m_\nu^2 \rangle^{TE}$ at intermediate temperatures has its origin in the field dependence of $\chi_\nu^{-1}(0)$ and hence of $q_c$ (for details see the remarks made in the text that follows equation (47)), and
(b) the function $\mathcal{F}$ in equations (48) and (49) depends on $T$ and $H$ through $q_c(T, H)$ but such dependences are extremely weak and hence can be ignored in practice.

*2.3.3. Magnetic equation of state.* In view of equations (1)–(5), the Stoner magnetic equation of state, modified to account for long-wavelength and low-energy spin fluctuations of small-fluctuation amplitude, assumes the form

$$\left[\frac{M(T,H)}{M(0,0)}\right]^2 = 1 - \left(\frac{T}{T_C^S}\right)^2 - \frac{3\langle m_\parallel^2 \rangle + 2\langle m_\perp^2 \rangle}{M^2(0,0)} + 2\chi(0,0)\frac{H}{M(T,H)}. \qquad (50)$$

Note that in arriving at the above form of the magnetic equation of state the $T^4$-term in equation (4) has been dropped since this term is usually extremely small. This term can, however, become significant when the Stoner enhancement factor $\mathcal{S}$ is extremely large, e.g., for systems on the verge of ferromagnetism in which $IN(E_F) \to 1$ and hence $\mathcal{S} \to \infty$. The dependences of magnetization on temperature and field can be calculated in a *self-consistent* fashion by inserting the expressions for $\langle m_\parallel^2 \rangle$ and $\langle m_\perp^2 \rangle$ valid in different temperature ranges into equation (50). This procedure leads to

$$\left[\frac{M(T,H)}{M(0,0)}\right]^2 = 1 - \left(\frac{T}{T^*}\right)^2 - \left(\frac{T}{T_1}\right)^{4/3} + \frac{3\Phi(T)\chi_\parallel^{-1}(0)}{M^2(0,0)} + 2\chi(T)\frac{H}{M(T,H)} \qquad (51)$$

where

$$\left(\frac{1}{T^*}\right)^2 = \left(\frac{1}{T_C^S}\right)^2 + \left(\frac{1}{T_0}\right)^2 \tag{52}$$

$$T_0^{-2} = \frac{1}{6\pi^2}\left(\frac{k_B^2}{\overline{h}\gamma_\nu c_\nu}\right)\left(\frac{3\chi_\parallel(0) + 2\chi_\perp(0)}{M^2(0,0)}\right) \tag{53}$$

$$T_1^{-4/3} = \frac{5}{2\pi^2}\left(\frac{3\mathcal{F} - 8\pi}{24\pi}\right)(\overline{h}\gamma_\nu)^{-1/3}[M(0,0)]^{-2}\left(\frac{k_B}{c_\nu}\right)^{4/3} \tag{54}$$

$$\Phi(T) = g\left[q_0^2 + 2q_0\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{1/3} + \left\{1 + \frac{3\mathcal{F} - 2\pi}{9\mathcal{G}}\right\}\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{2/3}\right] \tag{55}$$

$$\chi(T) = \chi(0,0)[1 + 2b\Phi(T)] \tag{56}$$

at *intermediate* temperatures, and

$$\left[\frac{M(T,H)}{M(0,0)}\right]^2 = 1 - \left(\frac{T}{T_C^S}\right)^2 - A(H)T^{4/3} + 2\chi(T)\frac{H}{M(T,H)} \tag{57}$$

with

$$A(H) = A(H=0)[1 - \eta\sqrt{H}] \tag{58}$$

$$A(H=0) = (T_C^{SF})^{-4/3} = \frac{5}{2\pi^2}\left(\frac{\mathcal{F} + 8\pi}{8\pi}\right)(\overline{h}\gamma_\nu)^{-1/3}[M(0,0)]^{-2}\left(\frac{k_B}{c_\nu}\right)^{4/3} \tag{59}$$

$$\eta = \left(\frac{4\pi^2}{\mathcal{F} + 8\pi}\right)\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{-1/3}\left(\frac{g\mu_B}{D_\nu^{SF}}\right)^{1/2} \tag{60}$$

$$\chi(T) = \chi(0,0)[1 + 5b\Psi(T)] \tag{61}$$

$$\Psi(T) = g\left[q_0^2 + 2q_0\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{1/3} + \left(1 + \frac{\mathcal{F} - 4\pi}{3\mathcal{G}}\right)\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{2/3}\right] \tag{62}$$

at temperatures just below $T_C$ but outside the critical region ($T \lesssim T_C$), where $\chi_\parallel^{-1}(0)$ and $\chi_\perp^{-1}(0)$ have been assumed to be equal in magnitude. Moreover, the very weak dependence of the functions $\Phi$ and $\Psi$ on field, originating from the extremely slow variations of the quantities $\mathcal{F}$ and $\mathcal{G}$ with $H$, is completely ignored. Since $a_\nu$ is usually very large, such that $\mathcal{F} \simeq 1 + 2\ln a_\nu$ and $\mathcal{G} \simeq 1$, the quantities that involve the functions $\mathcal{F}$ and $\mathcal{G}$ can be estimated from the following expressions without any appreciable loss of accuracy:

$$T_1^{-4/3} \cong \frac{5}{2\pi^2}\left(\frac{3 + 6\ln a_\nu - 8\pi}{24\pi}\right)(\overline{h}\gamma_\nu)^{-1/3}[M(0,0)]^{-2}\left(\frac{k_B}{c_\nu}\right)^{4/3} \tag{63}$$

$$\Phi(T) \cong \frac{1}{4\pi^3}\left(\frac{\overline{h}\gamma_\nu}{c_\nu}\right)\left[q_0^2 + 2q_0\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{1/3} + \frac{4}{3}\left(1 + \frac{\ln a_\nu}{2} - \frac{\pi}{6}\right)\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{2/3}\right] \tag{64}$$

$$A(H=0) = (T_C^{SF})^{-4/3} \cong \frac{5(1 + 2\ln a_\nu + 8\pi)}{16\pi^3}(\overline{h}\gamma_\nu)^{-1/3}[M(0,0)]^{-2}\left(\frac{k_B}{c_\nu}\right)^{4/3} \tag{65}$$

$$\eta \cong \frac{4\pi^2}{1 + 2\ln a_\nu + 8\pi}\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{-1/3}\left(\frac{g\mu_B}{D_\nu^{SF}}\right)^{1/2} \tag{66}$$

$$\Psi(T) \cong \frac{1}{4\pi^3}\left(\frac{\overline{h}\gamma_\nu}{c_\nu}\right)\left[q_0^2 + 2q_0\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{1/3} + \frac{4}{3}\left(1 + \frac{\ln a_\nu}{2} - \pi\right)\left(\frac{k_B T}{\overline{h}\gamma_\nu c_\nu}\right)^{2/3}\right]. \tag{67}$$

In order to facilitate a comparison with equation (3), the magnetic equations of state represented by equations (51) and (57) are recast as

$$\frac{H}{M(T,H)} = a^R(T) + b^R(T)[\langle m^2(T,H)\rangle + M^2(T,H)] \tag{68}$$

with

$$a^R(T) = -\frac{1}{2}\chi^{-1}(T)\left[1-\left(\frac{T}{T_C^S}\right)^2\right] = \frac{a(T)}{1+2b\Phi(T)} \tag{69}$$

$$b^R(T) = \frac{1}{2}\chi^{-1}(T)[M(0,0)]^{-2} = \frac{b}{1+2b\Phi(T)} = \frac{\gamma}{\chi_p + 2\gamma\Phi(T)} \tag{70}$$

$$\langle m^2(T,H)\rangle = M^2(0,0)\left\{\left(\frac{T}{T_0}\right)^2 + \left(\frac{T}{T_1}\right)^{4/3}\right\} - 3\Phi(T)\chi_\parallel^{-1}(0) \tag{71}$$

at intermediate temperatures and

$$a^R(T) = -\frac{1}{2}\chi^{-1}(T)\left[1-\left(\frac{T}{T_C^S}\right)^2\right] = \frac{a(T)}{1+5b\Psi(T)} \tag{72}$$

$$b^R(T) = \frac{1}{2}\chi^{-1}(T)[M(0,0]^{-2} = \frac{b}{1+5b\Psi(T)} = \frac{\gamma}{\chi_p + 5\gamma\Psi(T)} \tag{73}$$

$$\langle m^2(T,H)\rangle = M^2(0,0)A(H)T^{4/3} \tag{74}$$

at $T \lesssim T_C$.

## 3. Discussion and concluding remarks

A close scrutiny of equations (3)–(5) and (68)–(74) reveals that the Landau form of the magnetic equation of state is *retained* even in the presence of zero-point and thermally excited spin-density fluctuations, but the Landau coefficients $a$ and $b$ get *renormalized* in accordance with the relation (69) (relation (72)) and relation (70) (relation (73)) for temperatures in the intermediate range (in the proximity of $T_C$ yet away from criticality). While zero-point spin fluctuations have a *major* share in renormalizing the Landau coefficients of the Stoner theory and significantly contribute to the temperature dependence of the magnetization over a wide range of temperatures, thermally excited electron–hole pair excitations are mainly responsible for the variations of the magnetization with field and temperature over the entire temperature range. Furthermore, zero-point (quantum) spin fluctuations leave the Stoner value of the spontaneous magnetization at 0 K, $M(0,0)$, *unaltered* but *change* the value of the zero-field differential susceptibility at 0 K from that in the Stoner model, i.e., $\chi(0,0)$, to $\chi(0,0)[1 + (5/4\pi^3)(\overline{h}\gamma_v/c_v)bq_0^2]$. Consequently, the Stoner enhancement factor $\mathcal{S}$ and the quantity $\gamma$, equation (8), assume *renormalized* values:

$$\mathcal{S}^R(T=0) = \mathcal{S}[1 + 5b(\overline{h}\gamma_v/4\pi^3 c_v)q_0^2]$$
$$\gamma^R(T=0) = \gamma/[1 + 5b\Psi(T=0)] = \gamma/[1 + 5b(\overline{h}\gamma_v/4\pi^3 c_v)q_0^2]$$

respectively. Another consequence of the *invariance* of $M(0,0)$ and *renormalization* of the coefficients $a$ and $b$ is that equations (30)–(32) and (35) retain their validity at low temperatures despite the fact that they do not account for the zero-point and thermally excited *non-propagating* spin fluctuations.

When $H = 0$, the magnetic equation of state, equation (57) or (68), yields the temperature dependence of the spontaneous magnetization for $T \lesssim T_C$ as

$$M(T,0) = M(0,0)\left[1 - \left(\frac{T}{T_C^S}\right)^2 - A(H=0)T^{4/3}\right]^{1/2}$$
$$= M(0,0)\left[1 - \left(\frac{T}{T_C^S}\right)^2 - \left(\frac{T}{T_C^{SF}}\right)^{4/3}\right]^{1/2}. \tag{75}$$

Use of the condition $M(T,0) = 0$ at $T = T_C$ in the above expression results in [20]

$$1 - \left(\frac{T_C}{T_C^S}\right)^2 - \left(\frac{T_C}{T_C^{SF}}\right)^{4/3} = 0. \tag{76}$$

This relation permits a reasonably accurate determination of the Curie temperature, $T_C$. From equation (76), it follows that $T_C = T_C^S$ if $T_C^S \ll T_C^{SF}$ and $T_C = T_C^{SF}$ if $T_C^{SF} \ll T_C^S$. These two limits represent the extreme situations wherein Stoner single-particle excitations and exchange-enhanced spin-density fluctuations are respectively predominant.

With a view to ascertaining whether zero-point (ZP) spin fluctuations or thermally excited (TE) spin fluctuations *alone* or both of them together determine the Curie temperature of a weak itinerant-electron (WI) ferromagnet, the values of $T_C$ for the archetypal WI ferromagnets $Ni_3Al$, MnSi, and $ZrZn_2$, calculated in the limit $T_C^{SF} \ll T_C^S$ using the *modified* versions of equation (59) or (65) given below, are compared with the experimentally observed [1, 2, 20, 33–36, 43] ones in table 1. The values of $T_C$ displayed in the columns II–VII of the table are respectively the observed ones and those deduced from the expressions

$$T_C = T_C^{TE} = \left(\frac{2\pi^2}{5}\right)^{3/4}[M(0,0)]^{3/2}(\bar{h}\gamma_v)^{1/4}\left(\frac{c_v}{k_B}\right) \tag{77}$$

$$T_C = T_C^{TE} = \left(\frac{2\pi^2}{5\alpha_3}\right)^{3/4}[M(0,0)]^{3/2}(\bar{h}\gamma_v)^{1/4}\left(\frac{c_v}{k_B}\right) \tag{78}$$

$$T_C = T_C^{ZP} = \left(\frac{16\pi^3}{5[1+2\ln a_v]}\right)^{3/4}[M(0,0)]^{3/2}(\bar{h}\gamma_v)^{1/4}\left(\frac{c_v}{k_B}\right) \tag{79}$$

$$T_C = T_C^{ZP+TE} = \left(\frac{16\pi^3}{5[1+2\ln a_v+8\pi]}\right)^{3/4}[M(0,0)]^{3/2}(\bar{h}\gamma_v)^{1/4}\left(\frac{c_v}{k_B}\right) \tag{80}$$

$$T_C = T_C^{ZP+TE} = \left(\frac{16\pi^3}{5[1+2\ln a_v+8\pi\alpha_3]}\right)^{3/4}[M(0,0)]^{3/2}(\bar{h}\gamma_v)^{1/4}\left(\frac{c_v}{k_B}\right) \tag{81}$$

where $\alpha_3 = (2/3^{3/2})\Gamma(4/3)\zeta(4/3) \simeq 1.238$, when the reported [1, 2, 20, 34, 36] values of the parameters appearing in the above expressions are used. Equations (77)–(81), in ascending order of the numerals, describe the following cases:

**Table 1.** Comparison between theoretical and experimental values of the Curie temperature for archetypal weak itinerant-electron ferromagnets.

| Material | $T_C^{obs}$ (K) | $T_C^{TE}$ (K) Equation (77) | $T_C^{TE}$ (K) Equation (78) | $T_C^{ZP}$ (K) Equation (79) | $T_C^{ZP+TE}$ (K) Equation (80) | $T_C^{ZP+TE}$ (K) Equation (81) |
|---|---|---|---|---|---|---|
| $Ni_3Al$ | 41.0(5) | 46.0(5) | 39.5(5) | 90.0(20) | 35.0(10) | 31.0(10) |
| MnSi | 29.5(5) | 36.0(5) | 31.0(5) | 58.0(20) | 26.0(10) | 24.0(10) |
| $ZrZn_2$ | 28.0(5) | 38.5(5) | 33.0(5) | 69.0(20) | 29.0(10) | 26.0(10) |

(i) $T_C$ is determined *solely* by the thermally excited (TE) spin fluctuations, $\langle m_\nu^2 \rangle_{SF}^{TE}$, and the relevant expression for $\langle m_\nu^2 \rangle_{SF}^{TE}$ is obtained using the classical approximation, i.e., equation (42) with $H = 0$;

(ii) the same as (i) but with $\langle m_\nu^2 \rangle_{SF}^{TE}$ calculated without taking recourse to the classical approximation, i.e., using equation (40);

(iii) zero-point (ZP) spin fluctuations *alone* decide the value of $T_C$;

(iv) ZP and TE spin fluctuations *jointly* determine $T_C$ in accordance with equation (65), which is based on the version of equation (42) that describes $\langle m_\nu^2 \rangle_{SF}^{TE}$ in the classical approximation when $H = 0$;

(v) the same as (iv) but the values of $T_C$ are refined by using a more accurate expression, equation (40), for $\langle m_\nu^2 \rangle_{SF}^{TE}$ in equation (65) than equation (42) with $H = 0$.

A cursory glance at the entries in table 1 suffices to reveal the following:

(a) If ZP spin fluctuations alone were responsible for the thermal demagnetization of the spontaneous magnetization, they would overestimate $T_C$ by more than a factor of *two* (cf. columns II and V of table 1).

(b) By contrast, thermally excited spin fluctuations almost entirely account for the observed values of $T_C$ in the case of $Ni_3Al$ and MnSi (cf. columns II and IV of table 1).

(c) While each component (ZP or TE) of spin fluctuations, on its own, fails to reproduce the observed value of $T_C$ for $ZrZn_2$, the combined effect of these components is to essentially eliminate the discrepancy between the calculated and observed $T_C$-values (cf. columns II and VI or VII of table 1).

Therefore, from such a comparison between theory and experiment, we conclude that $T_C$ for $Ni_3Al$ and MnSi is determined primarily by thermally excited spin fluctuations rather than by zero-point spin fluctuations, whereas zero-point spin fluctuations play an important role, though not as significant as that played by thermally excited spin fluctuations, in determining $T_C$ for $ZrZn_2$. Moreover, since $T_C^S$ is at least an order of magnitude larger [20] than $T_C^{SF}$ or even $T_C^{ZP}$ (table 1) for all three materials, a substantial contribution to the thermal demagnetization of $M(T, 0)$ arising from Stoner single-particle excitations is essentially ruled out.

The above remarks concerning the relative importance of spin fluctuations and Stoner single-particle excitations are consistent with the conclusions drawn earlier [20] about the extent to which these excitations influence the temperature dependence of magnetization but are in direct conflict with the assertions made previously by Mohn and Wohlfarth [24] particularly for $ZrZn_2$.

Finally, the functional forms for the dependences of the spin fluctuations (and hence of the magnetization) on temperature and field yielded by the present calculations in the limit in which spin fluctuations *dominate* over single-particle excitations are compared with those predicted by earlier theoretical treatments [1, 2, 5, 6, 10, 20] of spin fluctuations in weak itinerant-electron (WI) ferromagnets. As already mentioned in the introduction, previous versions of the spin-fluctuation (SF) theory (henceforth referred to as the conventional SF theory) do not make any specific prediction regarding the field dependence of spin fluctuations in WI ferromagnets. In the intermediate range of temperatures, the SF theory due to Lonzarich and Taillefer [20] (LT) demonstrates that thermally excited longitudinal ($\parallel$) and transverse ($\perp$) spin fluctuations, when treated on an equal footing, result in a variation of the spontaneous magnetization, $M(T, 0)$, with temperature of the form

$$M(T, 0) = M(0, 0)[1 - (T/T_C)^2]^{1/2}.$$

In the same temperature range, our calculations assert that the magnetization has the functional form

$$M(T, H) = M(0, H)[1 - (T/T_0)^2 - (T/T_1)^{4/3}]^{1/2}$$

i.e., the version of equation (51) in which only the *leading* terms are retained, *irrespective* of whether the field $H$ (recall that $H$ stands for the external magnetic field *corrected* for the demagnetizing and other anisotropy fields present) is *finite* or *zero*. While the $T^2$-term has the same origin as in the LT theory and its coefficient $T_0^{-2}$ *depends* on $H$ through the field dependence of $\chi_\nu(0)$, equation (53), the *extra* $T^{4/3}$-term is a net outcome of the competing claims made by the thermally excited and zero-point components of spin fluctuations (the former contribution *decreases* with $T$ as $T^{4/3}$ whereas the latter one *dominates* over the former and *increases* with $T$ as $T^{4/3}$), equation (54), and its coefficient $T_1^{-4/3}$ is essentially *field independent*. An attempt to assess the relative importance of the $T^2$- and $T^{4/3}$-terms in the truncated version of equation (51) given above at intermediate temperatures using the reported [1, 2, 20, 34, 36] values of the quantities appearing in equations (53) and (54) revealed that the $T^{4/3}$-term, though much smaller in magnitude than the $T^2$-term, is not so small as to justify its total neglect. For temperatures close to $T_C$ but still away from criticality ($T \lesssim T_C$), thermally excited spin fluctuations, according to the conventional SF theory, give rise to a temperature dependence of the spontaneous magnetization of the type

$$M(T, 0) = M(0, 0)[1 - (T/T_C)^{4/3}]^{1/2}.$$

By contrast, on the basis of the calculations presented in this paper, we claim that for $T \lesssim T_C$, the magnetization is given by

$$M(T, H) = M(0, H)[1 - A(H)T^{4/3}]^{1/2}$$

(the truncated form of equation (57) in which the second and fourth terms are dropped because of their negligible magnitude). In this expression, valid for both $H = 0$ and $H \neq 0$, the $T^{4/3}$-term for $H = 0$ has its origin not only in thermally excited (TE) spin fluctuations, as in the conventional SF theory, but also in zero-point (ZP) spin fluctuations, and the contributions to the coefficient $A(H)$ of this term arising from the TE and ZP components are *additive*, equations (58) and (59); for $H \neq 0$, the $T^{4/3}$-term originates from TE spin fluctuations alone. Furthermore, our calculations, besides asserting that, at all temperatures, field *suppresses* thermally excited spin fluctuations but has little, or even no, effect on zero-point spin fluctuations, for the first time, *quantify* the suppression of TE spin fluctuations with magnetic field in the form of equation (58) for $T \lesssim T_C$.

The results of extensive magnetization measurements performed recently [35] on a polycrystalline sample of $Ni_3Al$ and previously [44, 45] on a number of amorphous magnetic systems exhibiting weak itinerant-electron ferromagnetism corroborate all of the predictions of the present theoretical calculations.

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