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Static and dynamic susceptibilities of ferromagnets calculated with spin-wave theory including dipolar forces

Stephen W Lovesey† and Kalliopi N Trohidou‡
† Rutherford Appleton Laboratory, Didcot, Oxfordshire, OX11 0QX, UK and Institute of Physics, Uppsala University, S-751 21, Sweden
‡ National Research Centre, Democritos, PO Box 60228, Athens, Greece

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Abstract. Static and dynamic susceptibilities for a Heisenberg ferromagnet with dipolar forces and an external magnetic field are calculated with linear spin-wave theory. Attention is focused on quantities that can be observed in neutron diffraction and spectroscopy experiments. Analytic results for the susceptibilities are reported for a region of modest temperatures and long wavelengths. Extensive numerical results for the static longitudinal susceptibility of models of EuO and EuS, including first- and second-neighbour exchange interactions, are summarized.

1. Introduction

The influence of dipolar forces on the static and dynamic properties of simple ferromagnets has been convincingly demonstrated in measurements of spin fluctuations in the critical region. A striking example is the time dependence of the spin autocorrelation function at the critical temperature T_c, which is predicted to change as a result of dipolar forces from a Gaussian to exponential form (Lovesey and Williams 1986, Aberger and Folk 1988, Frey and Schwabl 1989). This is consistent with data at small wavevectors k obtained by the spin-echo technique for EuO (Mezei 1986). Moreover, the observed k dependence of the damping constant is in excellent agreement with the value derived from a dynamic critical exponent $z = 2.5$. A dipolar-induced cross-over to $z = 2.0$ occurs, in the theory, at a k value that is about an order of magnitude smaller than the value for cross-over in static properties and time dependence, and confirmation remains a challenge to the experimentalist. The influence of dipolar forces is also manifest in a variety of other careful experiments on spin fluctuations in the critical region of insulating and metallic magnets; see, for example, Hohenemser *et al* (1989).

Dipolar forces, present to some extent in all magnetic materials, complicate the basic structure of a model in which the ordered state is ascribed to a simple Heisenberg exchange interaction. In some ways the most important complication is that the total magnetization is not conserved. This means, for one thing, that off-diagonal spin correlation functions $\langle S^{\pm}S^{\pm}\rangle$, which contribute to static and dynamic susceptibilities, acquire a finite value. Another consequence is that, in the critical and paramagnetic regions, relaxational dynamics prevails in the long-wavelength limit, i.e. the dynamic critical

Table 1. Dipolar wavevector[a] ($\mathrm{\AA}^{-1}$).

Material	q_D
Fe	0.045
Ni	0.013
EuO	0.147
EuS	0.24
$LiTbF_4$	1.31

[a] After Mezei (1984) and Kötzler (1986).

exponent $z = 2.0$. Furthermore, spin isotropy is broken, leading to different properties longitudinal and transverse to $\boldsymbol{k}$. The longitudinal susceptibility does not diverge at T_c, but saturates to a value determined by the dipolar (demagnetization) anisotropy. This effect has been observed in a polarized neutron scattering study of the critical fluctuations around Bragg reflections in EuS and EuO crystals (Kötzler *et al* 1984). In the ordered phase dipolar forces create magnetostatic modes which make up the ferromagnetic resonance spectrum, and cause a discontinuity in the spin-wave dispersion. The latter has been observed in a crystal Ho/10% Tb by inelastic neutron scattering (Larsen *et al* 1987).

By and large, the influence of dipolar forces has been revealed most clearly in properties of long-wavelength spin fluctuations. The examples of static and dynamic effects in EuO and EuS cited above appear at wavevectors $k < q_D$, where q_D is the so-called dipolar wavevector. Values of q_D, compiled by Mezei (1984) and Kötzler (1986), are gathered in table 1. Four of the materials are cubic ferromagnets with weak dipolar forces, while $LiTbF_4$ is a tetragonal uniaxial dipolar ferromagnet, extensively studied by Als-Nielsen (1976a, b) using neutron diffraction. The values in the table have been determined from data on volume susceptibilities and inverse correlation lengths in the critical region.

The behaviour of the longitudinal susceptibility $\chi(\boldsymbol{k})$ in the ordered phase is not clearly understood. For a simple ferromagnet, $\chi(0)$ is predicted to diverge as the applied field H vanishes, and the form $H^{-1/2}$ has long been known from spin-wave theory. Calculations using the renormalization group technique predict that $\chi(\boldsymbol{k})$ diverges with vanishing $\boldsymbol{k}$ and H; $\chi(0)$ should behave like $H^{-1/3}$ at all temperatures below T_c. The experimental situation is not clear-cut. Kötzler and Muschke (1986) report an indication of $H^{-1/3}$ behaviour in an analysis of bulk data for EuS.

Neutron scattering measurements of $\chi(\boldsymbol{k})$ for EuO and Ni, at about $0.9T_c$, by Mitchell and collaborators (unpublished and referred to by Toh and Gehring (1990)), which extend a previous investigation (Mitchell *et al* 1984) on a disordered alloy Pd/10% Fe, do not reveal a divergent behaviour. In addition, Böni *et al* (1990) find for Ni at $T = 0.987T_c$ that the longitudinal intensity decreases slightly on increasing the field by a factor 3.

Here we use linear ferromagnetic spin-wave theory to calculate the longitudinal and transverse neutron cross sections, including effects of polarization in the incident and scattered beams. The corresponding static susceptibilities are also provided. With regard to the latter, our results correct work by Toh and Gehring (1990) who use a theory, originally developed by Holstein and Primakoff (1940), that is incorrect; see for example, Lowde (1965), Keffer (1966) and Lovesey (1987).

Spin-wave theory of a ferromagnet, described by a Heisenberg exchange interaction and dipolar forces, is briefly reviewed in section 2. Results for the transverse and longitudinal neutron cross sections and wavevector-dependent susceptibilities are presented in sections 3 and 4, respectively. It is shown that dipolar forces determine $\chi(0)$ in zero field, while for k just beyond the magnetostatic region the dependence $\chi(k) \propto k^{-1}$ holds. Section 5 contains numerical results for the susceptibilities of FCC materials with particular attention to parameter sets appropriate for EuO and EuS. For the latter material dipolar forces have a significant effect on $\chi(\boldsymbol{k})$ even for $k > q_{\mathrm{D}}$. In section 6 there are some analytic results for the longitudinal response function observable in an inelastic neutron scattering experiment; without dipolar forces our result corrects an error in the expression provided by Vaks *et al* (1968). The longitudinal cross section possesses a field-limited peak when the energy transfer matches the spin-wave dispersion, which is not suppressed by dipolar forces although they change the lineshape. Conclusions are gathered in section 7.

2. Spin-wave theory

In this and subsequent sections we follow the development of spin-wave theory by Lovesey (1987) for a Heisenberg ferromagnet with dipolar forces. For the most part, the notation adopted is consistent with Lowde (1965) and Keffer (1966).

Spin operators $\{S_{\boldsymbol{l}}\}$ are assigned to sites defined by vectors $\{\boldsymbol{l}\}$ on a Bravais crystal lattice with N unit cells. Within a linear spin-wave theory,

$$[S_{\boldsymbol{l}}^{+}, S_{\boldsymbol{l}'}^{-}] = 2S\delta_{\boldsymbol{l},\boldsymbol{l}'} \tag{2.1}$$

and

$$S_{\boldsymbol{l}}^{z} = S - (1/2S)S_{\boldsymbol{l}}^{-}S_{\boldsymbol{l}}^{+} \tag{2.2}$$

where S is the spin magnitude. Neutron scattering experiments measure fluctuations in the spatial Fourier components $\{S_{\boldsymbol{q}}^{\pm}\}$ where

$$S_{\boldsymbol{l}}^{\pm} = (1/N)\sum_{\boldsymbol{q}} \exp(\pm \mathrm{i}\boldsymbol{q}\cdot\boldsymbol{l})S_{\boldsymbol{q}}^{\pm}. \tag{2.3}$$

For a simple ferromagnet, these components obey the equation of motion ($\hbar = 1$)

$$\mathrm{i}\partial_t S_{\boldsymbol{q}}^{+} = \varepsilon_{\boldsymbol{q}} S_{\boldsymbol{q}}^{+} \tag{2.4}$$

in which the spin-wave dispersion $\varepsilon_{\boldsymbol{q}}$ is the sum of exchange and external magnetic field energies, namely

$$\varepsilon_{\boldsymbol{q}} = g\mu_{\mathrm{B}}H + 2S[J(0) - J(\boldsymbol{q})]. \tag{2.5}$$

Here, H is the strength of the applied field, g the gyromagnetic factor and $J(\boldsymbol{q})$ is the spatial Fourier transform of the exchange interactions; $J(\boldsymbol{q}) = J(-\boldsymbol{q})$ because the lattice is Bravais, and $J(\boldsymbol{q}) = J(\boldsymbol{q}+\boldsymbol{\tau})$ by definition of the reciprocal attice vectors $\{\boldsymbol{\tau}\}$.

On adding dipolar forces to the Hamiltonian, the equation of motion (2.4) is changed by the addition of a term $B_{\boldsymbol{q}}^{*}S_{-\boldsymbol{q}}^{-}$, and $\varepsilon_{\boldsymbol{q}}$ is replaced by

$$A_{\boldsymbol{q}} = \varepsilon_{\boldsymbol{q}} + |B_{\boldsymbol{q}}| = A_{-\boldsymbol{q}} = A_{\boldsymbol{q}}^{*}. \tag{2.6}$$

The quantity $B_{\boldsymbol{q}} = B_{-\boldsymbol{q}} \neq B_{\boldsymbol{q}}^{*}$ is the Fourier transform of part of the dipolar force field.

Various authors have shown that (see Keffer 1966 and references therein), for all but the extreme value $\boldsymbol{q} = 0$, the appropriate functional form is

$$B_q = 2\pi g\mu_B M_0 \sin^2\theta_q \exp(-2i\varphi_q) \tag{2.7}$$

where θ_q and φ_q define the orientation of $\boldsymbol{q}$ with respect to the preferred (easy) axis, e.g. $q_x = q \sin\theta \cos\varphi$, and M_0 is the saturation magnetization. Also, demagnetizing effects modify the influence of the applied field (Keffer 1966).

Diagonalization of the new equation of motion is readily achieved in terms of Bose operators a and a^+ that satisfy

$$[a_q, a^+_{q'}] = \delta_{q,q'} \tag{2.8}$$

with

$$S^+_q = u_q a_q + v_q a^+_{-q}. \tag{2.9}$$

The coefficients in (2.9) are taken to be

$$u^2_q = (2SN)(A_q + \omega_q)/2\omega_q \tag{2.10}$$

and

$$v_q = -u_q B^*_q/(A_q + \omega_q) \tag{2.11}$$

where the spin-wave dispersion ω_q satisfies

$$\omega^2_q = A^2_q - |B_q|^2. \tag{2.12}$$

Note that, with this choice for the coefficients, which is not unique, v_q vanishes and $u_q = (2SN)$ as $B_q \to 0$, and u_q is purely real whereas v_q is complex. Holstein and Primakoff (1940) and Toh and Gehring (1990) are at fault in the determination of these coefficients, which the latter authors incorrectly find to be purely real.

3. Neutron scattering cross sections and polarization effects

The partial differential cross section for magnetic scattering of a polarized beam of neutrons from an array of spins is compactly expressed in terms of spin autocorrelation functions (Lovesey 1987, section 10.6). If the change in energy and wavevector in scattering are denoted by $\omega = E - E'$ and $\boldsymbol{k}$, respectively, and the atomic form factor $F(\boldsymbol{k})$ is taken to be purely real, the cross section is

$$\frac{d^2\sigma}{d\Omega\, dE'} = r_0^2 \left(\frac{E'}{E}\right)^{1/2} \sum_{l,l'} e^{i\boldsymbol{k}\cdot(\boldsymbol{r}-\boldsymbol{l}')} [\tfrac{1}{2} g F(\boldsymbol{k})]^2 \int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{-i\omega t} [\langle \boldsymbol{S}^{(\perp)}_l \cdot \boldsymbol{S}^{(\perp)}_{l'}(t)\rangle$$
$$+ i\boldsymbol{P} \cdot \langle \boldsymbol{S}^{(\perp)}_l \times \boldsymbol{S}^{(\perp)}_{l'}(t)\rangle]. \tag{3.1}$$

Here, $r_0 = -0.54 \times 10^{-12}$ cm, $\boldsymbol{P}$ is the polarization vector,

$$k^2 \boldsymbol{S}^{(\perp)} = \boldsymbol{k} \times (\boldsymbol{S} \times \boldsymbol{k}) \tag{3.2}$$

and $\boldsymbol{S}(t)$ is the standard Heisenberg operator in which t has the dimension of time.

Within the linear spin-wave approximation, correlation functions $\langle S^x S^z\rangle$ and $\langle S^y S^z\rangle$ vanish, because they involve an odd number of Bose operators. The functions formed with S^x and S^y describe single-spin-wave events, whereas the inelastic part of $\langle S^z S^z\rangle$

involves the creation and annihilation of two spin-waves, as is clearly evident in the subsequent expression. The combinations

$$\langle S^x S^y \pm S^y S^x \rangle \tag{3.3}$$

deserve some comment; this quantity with a positive sign occurs in the first contribution to the cross section (3.1), and with a negative sign in the second, polarization-dependent, contribution. The latter is finite and has a weight that is totally independent of the details of the magnetic Hamiltonian since it is, within the linear spin-wave approximation, simply $[S^x, S^y] = \mathrm{i}S$. This observation also tells us that the polarization created in the scattered beam is independent of parameters in the Hamiltonian. In particular, these contributions are not sensitive to dipolar forces, which are manifest only insofar as they contribute to the spin-wave dispersion. In contrast, the combination (3.3) with a positive sign is proportional to the strength of the dipolar force, and its finite value is a direct consequence of the fact that the total spin, in the direction of the easy axis, is not a conserved quantity.

The single-spin-wave events in (3.1) arise from

$$\sum_{l,l'} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{l}'-\boldsymbol{l})} \int_{-\infty}^{\infty} \frac{\mathrm{d}t}{2\pi} \mathrm{e}^{-\mathrm{i}\omega t} [(1-\hat{k}_x^2)\langle S_l^x S_{l'}^x(t)\rangle + (1-\hat{k}_y^2)\langle S_l^y S_{l'}^y(t)\rangle$$
$$- \hat{k}_x \hat{k}_y \langle S_l^x S_{l'}^y(t) + S_l^y S_{l'}^x(t)\rangle + \mathrm{i}(\hat{\boldsymbol{k}}\cdot\boldsymbol{\eta})(\hat{\boldsymbol{k}}\cdot\boldsymbol{P})\langle S_l^x S_{l'}^y(t) - S_l^y S_{l'}^x(t)\rangle] \tag{3.4}$$

where $\hat{\boldsymbol{k}}$ and $\boldsymbol{\eta}$ are unit vectors in the direction of $\boldsymbol{k}$ and the easy axis, respectively. Evaluating (3.4) with the results recorded in section 2, the spin-wave creation event is found to have a cross section

$$(SN/2)(1+n_q)\delta(\omega-\omega_q)\{(1+\hat{k}_z^2)(A_q/\omega_q) + (1-\hat{k}_z^2)(|B_q|/\omega_q)\cos[2(\varphi-\varphi_q)]$$
$$- 2(\hat{\boldsymbol{k}}\cdot\boldsymbol{\eta})(\hat{\boldsymbol{k}}\cdot\boldsymbol{P})\}. \tag{3.5}$$

Here, n_q is the standard Bose occupation factor, and φ is the azimuthal angle of $\boldsymbol{k}$ in the plane perpendicular to the easy axis. The corresponding annihilation cross section is proportional to

$$n_q \delta(\omega + \omega_q)$$

and the sign of $\boldsymbol{P}$ is reversed. With these expressions it is understood that $\boldsymbol{k} = \boldsymbol{\tau} + \boldsymbol{q}$, where $\boldsymbol{\tau}$ is a reciprocal lattice vector.

Turning now to the contribution to the cross section proportional to $\langle S^z S^z \rangle$, it is convenient to introduce two structure factors defined by

$$\left.\begin{matrix} E(\boldsymbol{p},\boldsymbol{q}) \\ F(\boldsymbol{p},\boldsymbol{q}) \end{matrix}\right\} = [A_p A_q \mp \omega_p \omega_q + |B_p|\,|B_q| \cos 2(\varphi_p - \varphi_q)]/(2\omega_p \omega_q). \tag{3.6}$$

It can be shown that $E, F \geqslant 0$, and E vanishes while $F \to 1$ in the absence of dipolar forces. We find for the inelastic part of

$$\sum_{l,l'} \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{l}'-\boldsymbol{l})} \int_{-\infty}^{\infty} \frac{\mathrm{d}t}{2\pi} \mathrm{e}^{-\mathrm{i}\omega t} (1-\hat{k}_z^2)\langle S_l^z S_{l'}^z(t)\rangle \tag{3.7a}$$

the result (β is the inverse temperature)

$$N(1-\hat{k}_z^2) S(\boldsymbol{k}, \omega)$$

with

$$S(\boldsymbol{k},\omega)=(1-\mathrm{e}^{-\beta\omega})^{-1}\frac{1}{N}\sum_{p,q}\delta_{p-q,k}\{\tfrac{1}{2}E(p,q)(1+n_p+n_q)[\delta(\omega-\omega_p-\omega_q)$$
$$-\delta(\omega+\omega_p+\omega_q)]+F(p,q)(n_p-n_q)\delta(\omega+\omega_p-\omega_q)\}. \qquad (3.7b)$$

Here, the first factor in $S(\boldsymbol{k},\omega)$ is a consequence of the condition of detailed balance. The following features of the cross section are worth noting: (i) it is not explicitly proportional to the magnitude of the spins, in contrast to single-spin-wave events; (ii) processes engaged in scattering involve two-spin-wave creation, annihilation and difference events, and only the latter occur in the absence of dipolar forces; and (iii) there is a zero-point contribution on account of the dipolar forces.

If the scattering wavevector $\boldsymbol{k}$ is arranged parallel to the easy axis, the $\langle S^zS^z\rangle$ and the dipolar-induced contribution to the single-spin-wave cross section are not observed since both appear together with $(1-\hat{k}_z^2)$. Moving $\boldsymbol{k}$ away from the easy axis engages these contributions to the cross section, and reduces the influence of the polarization of the incident beam, which ultimately vanishes when $\boldsymbol{k}$ is perpendicular to $\boldsymbol{\eta}$.

To round off this discussion of neutron scattering from ferromagnetic spin waves requires a record of the expressions for polarization of the scattered beam. Polarization of the incident and scattered beams is aligned with the easy axis, and the components of the polarization vectors are P_z and P_z' respectively. Using results from section 2, and formula (10.125) from Lovesey (1987) for $\boldsymbol{P}'$, we find that P_z' is proportional to

$$-NS(\boldsymbol{k},\omega)P_z\sin^2\theta\cos 2\theta+(SN/2)(1+n_q)\delta(\omega-\omega_q)$$
$$\times\{2\cos^2\theta-(P_z/\omega_q)[A_q(1+\cos^2\theta\cos 2\theta)$$
$$+|B_q|(1+2\cos^2\theta)\sin^2\theta\cos 2(\varphi-\varphi_q)]\}. \qquad (3.8)$$

Here, just the single-spin-wave creation contribution is included, since it dominates the annihilation contribution in the limit of low temperatures; the latter is derived from the creation contribution by replacing

$$(1+n_q)\delta(\omega-\omega_q)$$

by

$$n_q\delta(\omega+\omega_q)$$

and reversing the sign of the created polarization. The other response function in (3.8) is $S(\boldsymbol{k},\omega)$ defined in (3.7b). The polar angles θ, φ define the orientation of $\boldsymbol{k}$ relative to the easy axis, $\boldsymbol{\eta}$. The weight of the created polarization, which exists even when $P_z=0$, is independent of details of the Hamiltonian, for reasons mentioned already, and it vanishes when $\boldsymbol{k}$ and $\boldsymbol{\eta}$ are perpendicular. Contributions to P_z' weighted by $S(\boldsymbol{k},\omega)$ and $|B_q|$ vanish when $\boldsymbol{k}$ and $\boldsymbol{\eta}$ are aligned. For $\theta=\pi/2$ the longitudinal and single-spin-wave contributions have opposite signs, given $A_q>|B_q|$, which is required for stability.

4. Wavevector-dependent susceptibilities

The static wavevector-dependent susceptibilities are conveniently calculated from the standard expression

$$\chi^{\alpha\beta}(\boldsymbol{k})=\frac{1}{N}\int_0^\beta \mathrm{d}\mu\sum_{l,l'}\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{l}'-\boldsymbol{l})}\langle\Delta S_l^\alpha\Delta S_{l'}^\beta(\mathrm{i}\mu)\rangle \qquad (4.1)$$

in which β is the inverse temperature, and ΔS denotes the fluctuation in the spin variable. For the system under discussion, $\Delta S^z=S^z-\langle S^z\rangle$ and $\Delta S^\alpha=S^\alpha$ for $\alpha=x,y$.

The susceptibilities can be measured by neutron diffraction (Als-Nielsen 1976a, b). The formal relation between $\chi(\boldsymbol{k})$ and the cross section is not required at present, because we will use (4.1), but it is worth mentioning that $\chi(\boldsymbol{k})$ is proportional to the cross section multiplied by $[1 - \exp(-\beta\omega)]/\omega$ and integrated over all ω. In consequence, there are strong similarities between terms in the cross section and $\chi(\boldsymbol{k})$, as will be evident in subsequent results.

We choose to define two transverse susceptibilities, derived from the correlation functions

$$\tfrac{1}{2}\langle S^\alpha S^\beta + S^\beta S^\alpha\rangle$$

with $\alpha = \beta = x, y$ and $\alpha = x, \beta = y$, and denote them by $\chi_{\mathrm{d}}^{\perp}$ and $\chi_{0\mathrm{d}}^{\perp}$. The latter is finite because dipolar forces break the conservation of the total spin. Using results recorded in section 2, we find from (4.1) the values

$$\chi_{\mathrm{d}}^{\perp}(\boldsymbol{k}) = (SA_k/\omega_k^2) \tag{4.2}$$

and

$$\chi_{0\mathrm{d}}^{\perp}(\boldsymbol{k}) = -[S|B_k|\sin(2\varphi_k)]/\omega_k^2. \tag{4.3}$$

In the limit $B_k \to 0$, $\chi_{0\mathrm{d}}^{\perp}$ vanishes and $\chi_{\mathrm{d}}^{\perp}$ reverts to the usual expression for the spin-wave value of the transverse susceptibility. Referring to the single-spin-wave cross section (3.5) note that $\chi_{\mathrm{d}}^{\perp}$ and $\chi_{0\mathrm{d}}^{\perp}$ are closely related to the weights of the transverse and longitudinal components, respectively. With regard to the latter component, and $\chi_{0\mathrm{d}}^{\perp}(\boldsymbol{k})$ in (4.3), while these might not be positive quantities the cross section (3.5) is positive, or possibly zero, since it is an observable response function. So the quantity $\chi_{0\mathrm{d}}^{\perp}(\boldsymbol{k})$ is seen as a component of an observable response, and in isolation it does not have genuine physical significance. However, $\chi_{0\mathrm{d}}^{\perp}(\boldsymbol{k})$ to some extent embodies, through its marked dependence on the direction of $\boldsymbol{k}$, the rather special features of the dipolar force field.

The longitudinal susceptibility $\chi(\boldsymbol{k})$ is obtained from (4.1) on taking $\alpha = \beta = z$, and using (2.2) and (2.9)–(2.11). Employing the two structure factors defined in (3.6)

$$\chi(\boldsymbol{k}) = \frac{1}{N}\sum_{p,q} \delta_{p-q,k}\left(\frac{(1 + n_p + n_q)}{(\omega_p + \omega_q)}\, E(\boldsymbol{p}, \boldsymbol{q}) + \frac{(n_p - n_q)}{(\omega_q - \omega_p)}\, F(\boldsymbol{p}, \boldsymbol{q})\right). \tag{4.4}$$

In common with the longitudinal cross section (3.7*b*), there is a zero-point contribution in $\chi(\boldsymbol{k})$ induced by dipolar forces.

To gauge the magnitude of $\chi(\boldsymbol{k})$, and its dependence on the external field in (4.4) set $\boldsymbol{k} = 0$, $n_p = (T/\omega_p)$, and employ

$$2S[J(0) - J(\boldsymbol{p})] = Dp^2 \tag{4.5}$$

where D is the spin-wave stiffness, which is strictly valid in the limit of long wavelengths. In the absence of dipolar forces, the result is

$$\chi^0(0) = (T/8\pi)\,[v_0^{2/3}/D(g\mu_{\mathrm{B}}H)^{1/3}]^{3/2} \tag{4.6}$$

where v_0 is the unit-cell volume. Equation (4.6) shows a divergence proportional to $H^{-1/2}$ as the field tends to zero. Extending the analysis to small k,

$$\chi^0(k) = \chi^0(0)y^{-1/2}\sin^{-1}[y/(1 + y)]^{1/2} \tag{4.7}$$

where $y = (Dk^2/4h)$ and $h = g\mu_{\mathrm{B}}H$. In the limit $H \to 0$, (4.7) gives $\chi^0(k) \propto k^{-1}$.

The influence of dipolar forces on $\chi(k)$ at small k is readily found from an asymptotic analysis that exposes possible divergent behaviour. To this end, in (4.4) set $k = 0$ in the argument of the sum, and replace the sum over q, say, by an integration in which the minimum wavevector is k. Differentiating the result for $\chi(k)$ with respect to k leads to the expression

$$\left(\frac{\mathrm{d}\chi(k)}{\mathrm{d}k}\right) = -\left(\frac{k^2 v_0}{4\pi^2}\right)\int_0^{\pi} \mathrm{d}(\cos\theta)\left[\frac{(1+2n_k)}{2\omega_k}E(k,k) + \left(\frac{1}{T}\right)n_k(1+n_k)F(k,k)\right]. \tag{4.8}$$

Consider first the extreme case $k = 0$. Values of A_0 and B_0, which completely determine the integrand, are given by Keffer (1966). They depend on the shape of the sample, which is a characteristic of excitations in the magnetostatic region. In general, A_0 and B_0 are finite even in the absence of an external magnetic field. From (4.8), it then follows that $\chi(0)$ is a constant.

Turning to the case where k is small, but beyond the region described by magnetostatic modes, the appropriate values of A_k and B_k are given in section 2. On replacing the Bose factors in (4.8) by $(T/\omega_k) \gg 1$, as in the calculation leading to (4.6) and (4.7), the angular integration can be accomplished by elementary methods. The result reveals that, for zero magnetic field, $\chi(k) \propto k^{-1}$ and the coefficient is independent of the strength of the dipolar forces. Hence, dipolar forces determine $\chi(0)$ when $h = 0$, while for small k just beyond the magnetostatic region the dependence of $\chi(k)$ on k is the same as in (4.7), obtained without dipolar forces. Corrections to $\chi(k)$ from the latter decrease its value. When the field is finite $\chi(k)$ is a constant, in the region of k in question.

An estimate of $\chi(k)$ for k at the limit of the magnetostatic region is obtained using arguments that lead to (4.6) and (4.7) together with the results given in section 2. Defining a reduced variable $x = (4\pi M_0/H)$,

$$\chi(k \sim 0) = \tfrac{1}{2}\chi^0(0)\{1 + x^{-1/2}\sin^{-1}[x/(1+x)]^{1/2}\} \tag{4.9}$$

which predicts that dipolar forces cause a significant decrease in the susceptibility.

An estimate of the magnitude of the zero-point contribution to $\chi(0)$, induced by the dipolar forces, can be obtained from (4.4), evaluated at $T = 0$, together with the approximation (4.5). As a function of the dipolar force, the leading-order contribution to $\chi(0)$ arises from an integral of $|B_q|^2/\varepsilon_q^3$. Keeping just this term,

$$\chi(k \sim 0) = \zeta^3 v_0 (D\zeta^2/h^3)^{1/2}/(120\pi) \tag{4.10}$$

and some values of D and the wavevector ζ, defined in (5.1), are given in table 2. For the moment, note that the zero-point contribution is proportional to $H^{-3/2}$.

The range of validity of these approximate results, and the influence of dipolar forces at finite wavevectors, are explored in the next section in which a numerical evaluation of (4.4) is reported.

5. Longitudinal susceptibility: numerical results

The longitudinal susceptibility $\chi(k)$ has been evaluated numerically for a range of parameters. It is a quantity of interest in both its own right, since $\chi(k)$ is a basic

Table 2. Parameters for EuO and EuS[a] (FCC, easy axis (1, 1, 1)).

J_1 (K)	J_2 (K)	D (meV Å^2)	$M_0(T=0)$ (G)	ζ (Å^{-1})
EuO (a = 5.14 Å, T_c = 69.5 K)				
0.61	0.12	11.65	1910	0.11
EuS (a = 5.95 Å, T_c = 16.5 K)				
0.24	−0.12	2.56	1184	0.18

[a] After Keffer (1966) and Passell *et al* (1976).

response function, and as a guide to the total intensity observable in a neutron scattering experiment.

Results for the dipolar wavevector q_D presented in table 1 lead us to expect that dipolar forces are more significant in EuO and EuS than either of the transition-metal magnets ($LiTbF_4$ is not described by the spin-wave theory presented in section 2). A numerical evaluation of (4.4) has been made for an FCC lattice, and exchange interactions appropriate for EuO and EuS out to second-nearest neighbours. Values of the parameters used for EuO and EuS are given in table 2. In the spin-wave region, an appropriate measure of the strength of dipolar forces is a wavevector ζ defined through

$$\zeta^2 = (2\pi M_0 g\mu_B/D) \tag{5.1}$$

where M_0 is the saturation magnetization. For a FCC lattice, and first- and second-nearest-neighbour exchange interactions, J_1 and J_2 respectively, the spin-wave stiffness is

$$D = a^2 2S(J_1 + J_2) \tag{5.2}$$

and for europium ions $S = 7/2$. Note that values of q_D, derived from properties measured near the phase transition, exceed ζ by a factor of about 1.35. The value for $|B_q|$ derived from (2.7) has been compared by Passell *et al* (1976) with the results obtained by computing the full lattice sums in the basic definition (Lovesey 1987), and these authors found that they agree to within two parts in 10^3 over the entire Brillouin zone. In view of this, equation (2.7) with the value of M_0 given in table 2 was used in obtaining results presented here.

For the wavevector sum in expression (4.4) for $\chi(\boldsymbol{k})$ the argument is evaluated at N_0 points on a cubic mesh in the (BCC) reciprocal lattice unit cell. Various N_0 were used in exploratory calculations performed to ascertain the accuracy of the numerical routine. A test with $N_0 = 5 \times 10^5$ points gave 0.5% accuracy for Watson's (FCC) integral, and this N_0 was used for most of the data reported in table 3 and figures 1 and 2. The magnetic field is parallel to the easy axis, which is (1, 1, 1) for EuO and EuS. The wavevector $\boldsymbol{k}$ is specified with respect to this axis, and $(\xi, 0, 0)$, $(0, \xi, 0)$ and $(0, 0, \xi)$ are respectively parallel to $(-1, -1, 2)$, $(1, -1, 0)$ and $(1, 1, 1)$ in real-space cubic axes. The dimension of the Brillouin zone is such that k_y (k_z) passes through the K (L) point at which $\xi = 3/\sqrt{2}$ ($\sqrt{3}$). On the other hand, k_x is not aligned with a high-symmetry axis; it passes through a hexagonal face of the Brillouin zone, at a point $(3\pi/4a)(-1, -1, 2)$ at which $\xi = (3/2)^{3/2}$.

Table 3. Values of $T_c\chi(k)$ ($k = (0.1, 0, 0)(\pi/a)$, $H = 1$ kG or $h = 0.012$ meV).

T/T_c \ ξ (Å^{-1}) =	EuO 0.0	EuO 0.11	EuS 0.0	EuS 0.18
0.01	0.051	0.022	0.029	0.025
0.10	0.804	0.423	0.702	0.368
0.25	2.254	1.286	2.013	1.147
0.50	4.783	2.843	4.273	2.529

The temperature dependence of $\chi(k)$ for a small k is illustrated by the data contained in table 3. It increases more rapidly than a linear dependence, particularly when the dipolar forces are included in the calculation. There is next to no difference in the temperature dependence of $\chi(k)$ for EuO and EuS, although in other respects there are marked differences, cf figures 1 and 2. The value of k used in the calculations reported in table 3, 0.061 and 0.053 Å^{-1} for EuO and EuS, respectively, is relatively small on the scale that can readily be achieved in a neutron diffraction experiment. We have chosen to list $T_c\chi(k)$ because it is a dimensionless quantity, given our definition of static susceptibilities, and it is also of a similar magnitude for the two europium compounds.

Looking more closely at the data in table 3 reveals that the reduction in $\chi(k)$ caused by dipolar forces is the same for EuO and EuS at each temperature except the very lowest one. The reduction factor, which ranges from 0.53 to 0.60 for the three highest temperatures, is roughly in accord with (4.7), about which we will say more later. Hence, within the spin-wave approximation and for modest temperatures, dipolar forces reduce the longitudinal susceptibility by an amount that is largely independent of the magnitude of exchange forces and strength of the dipolar force. However, at a sufficiently low temperature the relative magnitudes of $\chi(k)$ with and without dipolar forces is reversed. For, without dipolar forces $\chi(k)$ vanishes in the limit $T \rightarrow 0$, whereas it is finite with dipolar forces, i.e. there is a zero-point contribution in the latter case, since the exact ground state is not a simple collinear ferromagnetic configuration. This cross-over in the magnitude of $\chi(k)$ is illustrated in the data for EuS at $T = 0.01T_c$, where $\chi(k)$ is more or less the same for the two cases.

To gauge the accuracy of (4.9) for $\chi(k)$ in the absence of dipolar forces, it can be compared with data given in table 3. As an example, for $T = T_c/4$ results from (4.9) differ from the numerical data by 29% and 1% for EuO and EuS, respectively. In fact, (4.9) appears to be tolerable when $y \leqslant 1.0$, and the large differences in accuracy found for the two compounds reflects significantly different values of y, namely 3.758 and 0.616, which is largely attributed to the different values of D for EuO and EuS.

Turning now to (4.7), which contains the influence of dipolar forces on $\chi(0)$, it appears from table 3 to overestimate by a factor that is typically 40%. However, this is based on a comparison of $\chi(0)$ with data of a small finite wavevector, and $\chi(k)$ certainly increases as k tends to zero, cf figures 1 and 2. Going to much smaller values of k turns up a special situation in the numerical evaluation of $\chi(k)$, and since our smallest k is more or less at the limit of what can be readily achieved in an experiment, extremely small k-values have not been tackled.

There is significantly more spatial anisotropy in $\chi(k)$ for EuS than EuO, on account of the competing exchange interactions in the former compound. This feature of $\chi(k)$ is

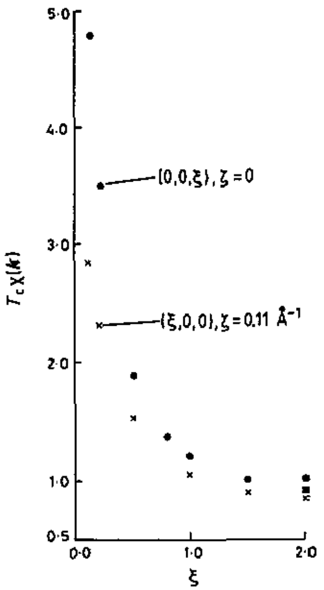

Figure 1. Values of $T_c\chi(\boldsymbol{k})$ for EuO at a temperature $T = T_c/2$ and a magnetic field $H = 1$ kG ($h = 0.012$ meV) parallel to the easy axis are displayed for various values of k_x and k_z. Wavevectors are taken relative to the easy axis, which is $(1, 1, 1)$ in the FCC lattice, and measured in units of π/a, where values of the lattice constant a are provided in table 2, e.g. $k_x = (\pi/a)(\xi, 0, 0)$ with $0 \leq \xi \leq 2$. Results without dipolar forces are denoted by full circles and are for k_z, values being very little different in the other principal directions. Dipolar forces induce more anisotropy but it is significant only at large $\boldsymbol{k}$. The full square (k_z) and cross (k_x) for $\xi = 2.0$ are extreme values at this ξ, and values of $\chi(k_x)$ are provided for smaller ξ.

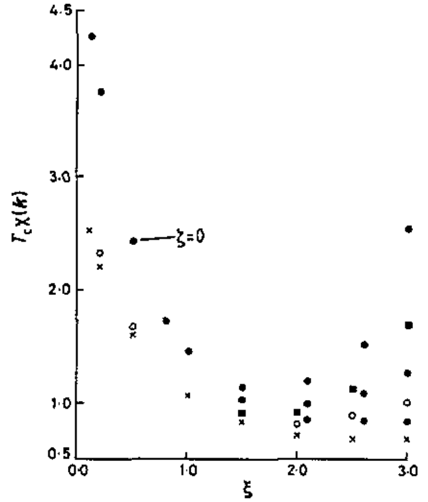

Figure 2. As in figure 1 but with exchange parameters appropriate for EuS, and $0 \leq \xi \leq 3$. Results without dipolar forces are denoted by full circles. For $\xi \leq 1.0$ the anisotropy in $\chi(\boldsymbol{k})$ is too small to display effectively. It is found that $\chi(k_x) < \chi(k_y) < \chi(k_z)$, and the extreme values are shown for $\xi = 1.5$, while all values are displayed at other selected $\xi > 2$. Including dipolar forces reduces $\chi(\boldsymbol{k})$, and values at k_x, k_y and k_z are denoted by crosses, open circles and full squares, respectively. At $\xi = 0.1$ and 1.0 there is minimal difference in the values of $\chi(\boldsymbol{k})$ so one value is shown, while at $\xi = 0.2$, 0.5 and 1.5 the extreme values are given. Beyond $\xi = \sqrt{3}$ crystal symmetry strongly influences the behaviour of $\chi(\boldsymbol{k})$ with respect to the direction of $\boldsymbol{k}$.

illustrated in figures 1 and 2, which display values of $T_c\chi(\boldsymbol{k})$ for $\boldsymbol{k}$ along the principal axes, relative to the easy axis. For EuO without dipolar forces, there is minimal anisotropy out to $k = 2\pi/a$, and with dipolar forces the anisotropy is significant only at large wavevectors, e.g. for $k = 2\pi/a$ there is a 9% difference between $\chi(k_x)$ and $\chi(k_z)$. The corresponding figures for EuS are 28% and 33% with and without dipolar forces, respectively, and the latter reduce $\chi(\boldsymbol{k})$ by about 50% at all $\boldsymbol{k}$ points included in figure 2.

6. Longitudinal dynamic susceptibility

Properties of the longitudinal susceptibility, or response function, $S(\boldsymbol{k}, \omega)$ defined in (3.7b) can be extracted in various limiting cases. First, we consider the case of exchange and magnetic field interactions. Using (4.5), $E_k = Dk^2$ and $h = g\mu_B H$, the result is

$$S(k,\omega)=\frac{Tv_0k^3}{16\pi^2E_k^2}(1-\mathrm{e}^{-\beta\omega})^{-1}\ln\left(\frac{1+(1-\mathrm{e}^{-\beta\omega})}{\exp\{(\beta/4E_k)[4hE_k+(\omega-E_k)^2]\}-1}\right). \tag{6.1}$$

This expression corrects the result of Vaks *et al* (1968). It is consistent with the susceptibility (4.9) calculated via the standard relation

$$\chi^0(k)=2\int_{-\infty}^{\infty}\frac{\mathrm{d}\omega}{\omega}S(k,\omega). \tag{6.2}$$

In the limit $k\to 0$ (ω finite), $S(k,\omega)$ vanishes, while for $\omega\to 0$ (k finite) it achieves a value that increases with decreasing k. The intensity of the peak in $S(k,\omega)$ at $\omega=E_k$ is obviously suppressed by the applied field, but this effect is mildly dependent on the magnitude of H because of the behaviour of the logarithm.

While for finite dipolar forces we have not found an expression equivalent to (6.1), behaviour of the frequency moments reveals that dipolar forces decrease $S(\boldsymbol{k},\omega)$ at large ω, at least for small k. From (3.7*b*), with $n=-1,1,3,\ldots$,

$$\int_{-\infty}^{\infty}\mathrm{d}\omega\,\omega^nS(k,\omega)=\frac{1}{2N}\sum_{p,q}\delta_{p-q,k}[E(p,q)(1+n_p+n_q)(\omega_p+\omega_q)^n$$
$$+F(p,q)(n_p-n_q)(\omega_q-\omega_p)^n] \tag{6.3}$$

Setting $\boldsymbol{k}=0$ in the expression for $n\geqslant 1$ shows that the frequency moments are proportional to E, and hence the square of the dipole force parameter B. The value of (6.3) for $\boldsymbol{k}=0$, $n=1$ is $(3T\zeta^3v_0/32\sqrt{2})$ in the limit $H\to 0$ and to leading order in ζ, defined in (5.1). In conjunction with this result we have the result (4.7) for $\chi(0)$, related to $S(\boldsymbol{k},\omega)$ as in (6.2). A numerical evaluation of $S(\boldsymbol{k},\omega)$, for a wide range of parameters, is the subject of a separate paper.

7. Conclusions

With regard to the influence of dipolar forces on the neutron cross section for single-spin-wave events, Lowde's (1965) longitudinal dipolar-induced contribution has been confirmed, and the polarization-dependent contribution is shown to have a weight that does not depend on the nature of the Hamiltonian, other than that the configuration is ferromagnetic. The same remark holds for the created polarization. However, the final polarization of an initially polarized beam contains a term that is an analogue of Lowde's term in the cross section. The magnitude of this term depends on the directions of the spin-wave momentum and the scattering vector. The so-called longitudinal cross section contains two-spin-wave scattering events. Unlike single-spin-wave scattering, zero-point fluctuations, due to dipolar corrections to the assumed classical ferromagnetic ground state, appear explicitly. An analytic result for the longitudinal response, valid for modest temperatures and small wavevectors, displays a field-limited enhancement at the spin-wave dispersion frequency. This effect is not removed by dipolar forces, although these change the lineshapes at higher frequencies. The predicted decrease in intensity due to dipolar forces is consistent with the decrease found in the longitudinal susceptibility, $\chi(\boldsymbol{k})$.

Numerical results for $\chi(k)$ with a small k for EuO and EuS show that at a given temperature ($T<T_c/2$) dipolar forces reduce it by about the same amount for the two

compounds. Differences in $\chi(\boldsymbol{k})$ for these magnets appear at temperatures low enough for zero-point fluctuations to be significant, and also in the dependence of $\chi(\boldsymbol{k})$ on the wavevector. There is next to no spatial anisotropy in $\chi(\boldsymbol{k})$ for EuO, whereas there is pronounced anisotropy at large k for EuS, generated by the competing exchange interactions. Moreover, dipolar forces influence $\chi(\boldsymbol{k})$ for EuO most keenly at small k, whereas in EuS there is a large reduction at all wavevectors examined. Analytic results for $\chi(0)$ show the expected $H^{-1/2}$ field dependence at modest temperatures, which changes to $H^{-3/2}$ at zero temperature. An expression for $\chi(\boldsymbol{k})$ valid for small k displays a divergence in the long-wavelength limit of the form k^{-1} when $H = 0$.

Our results for the longitudinal and transverse static susceptibilities correct previous work by Toh and Gehring (1990). In essence, their expressions are obtained from our findings by setting to zero the azimuthal angle of the wavevector, with respect to the easy axis, which is explicit in (3.6) and (4.4) and (4.3). In consequence, their results do not contain geometric factors characteristic of the dipolar force field, which contribute to the spatial anisotropy in $\chi(\boldsymbol{k})$ and directly influence the single-spin-wave cross section.

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Note added in proof. Results for the influence of dipolar forces on the longitudinal response function have been reported by Lovesey (1991).

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