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The spectrum of longitudinal spin fluctuations in a ferromagnet including dipolar and Zeeman energies

K N Trohidou† and S W Lovesey‡

† National Centre for Scientific Research, Institute for Materials Science, 'Democritos', PO Box 60228, Athens, Greece

‡ Rutherford Appleton Laboratory, Didcot, Oxfordshire OX11 0QX, UK

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Abstract. The spectrum of longitudinal spin fluctuations, observed in inelastic neutron scattering experiments, is provided for a Heisenberg model of an ordered ferromagnet, including dipolar forces and a magnetic field. A numerical method is used to obtain results, from an expression derived with linear spin-wave theory, for wavevectors ($\boldsymbol{k}$) throughout the Brillouin zone as a function of frequency (ω) and at various temperatures. Particular attention is given to realistic models of EuO and EuS. Results show that dipolar forces diminish the intensity in the longitudinal response function $S(\boldsymbol{k},\omega)$ by about a factor of 2, in accord with previous results for the isothermal susceptibility. The evolution of $S(\boldsymbol{k},\omega)$ with $\boldsymbol{k}$ displays several pronouced features, and for EuS there is a strong dependence on the direction of $\boldsymbol{k}$ for k near the zone boundary.

1. Introduction

The dearth of experimental data on the spectrum of spin fluctuations along the spontaneous magnetization axis (longitudinal fluctuations) in a ferromagnet is an anomaly when taken in context with the abundant, detailed data available on transverse (spin-wave) excitations in various compounds and metals. By and large, neutron scattering is the preferred experimental method by which to study spin-wave dispersions and lifetimes. The shortage of data on longitudinal fluctuations is caused largely by technical difficulties in extracting this component from the neutron cross section. A simple scattering geometry exists to measure just the transverse spectrum, but the longitudinal spectrum is always measured together with the transverse. In consequence, a differencing method is required to separate the longitudinal and transverse components in data sets, and the quality of data for the longitudinal spectrum is much inferior to that for the directly measured transverse spectrum. Polarization analysis affords in principle a clean separation of the two components, but the apparatus for the method is not yet very efficient.

Even so, recent progress in studies of the longitudinal spin-fluctuation spectrum has been achieved by using polarization analysis. Interestingly, results are not in accord with theory, which dates back to pioneering work on excitations in the Heisenberg ferromagnet; see [1,2] and references therein. If $\chi(\boldsymbol{k})$ denotes the longitudinal wavevector-dependent susceptibility, the prediction is $\chi(\boldsymbol{k}) \to (1/k)$ for $k \to 0$. The singular behaviour results from the presence of Goldstone bosons in an isotropic Heisenberg magnet. Application of a magnetic field generates a gap in the spectrum (the bosons acquire a mass) and $\chi(0) \sim (1/H)^{1/2}$, where H is the applied field. These results can be verified from the longitudinal spin response function $S(\boldsymbol{k},\omega)$ using the standard relation

$$\chi(\boldsymbol{k}) = \int_{-\infty}^{\infty} \mathrm{d}\omega\, [S(\boldsymbol{k},\omega)/\omega] \tag{1.1}$$

and the result, obtained for non-interacting spin waves with dispersion $E_k = Dk^2$,

$$S_0(\boldsymbol{k},\omega) = \left(\frac{Tv_0k^3}{16\pi^2 E_k^2}\right)[1+n(\omega)] \times \ln \left[\!\left[1 + \left\{[1+n(\omega)]\left[\exp\left(\frac{4hE_k + (\omega - E_k)^2}{4E_kT}\right) - 1\right]\right\}^{-1}\right]\!\right] \tag{1.2}$$

where v_0 is the volume of a unit cell, $h = g\mu_{\mathrm{B}}H$ is the Zeeman energy, and (Boltzmann's constant = 1)

$$n(\omega) = [\exp(\omega/T) - 1]^{-1}. \tag{1.3}$$

In the limit $k \to 0$ (ω fixed) $S_0(\boldsymbol{k},\omega)$ vanishes, while for $\omega \to 0$ (k fixed) it achieves a value that increases with decreasing k. The peak in $S_0(k,\omega)$ at the spin-wave energy $\omega = E_k$ is suppressed by the Zeeman energy, and it coalesces with the quasi-elastic response when k is decreased and $E_k \to 0$. The frequency dependence of $S_0(\boldsymbol{k},\omega)$ is illustrated in figure 1. Observe that (1.2) is the density fluctuation response for a Bose fluid with a chemical potential of $-h$.

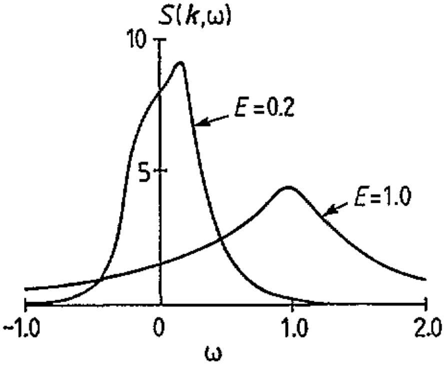

Figure 1. The frequency dependence of $S_0(\boldsymbol{k},\omega)$ defined in (1.2) illustrated for $h = 0.01$, $T = 0.5$ and two values of $E_k = 0.2$ and 1.0; these parameters are in units of meV.

The discrepancy between measured results and theoretical predictions, for response functions of an isotropic Heisenberg magnet, has prompted investigations of the influence on static and dynamic response functions of the usually very weak dipolar interactions. It has been shown that dipolar interactions, present to some extent in all magnets, reduce the magnitude of $\chi(\boldsymbol{k})$ but leave unchanged the divergence at

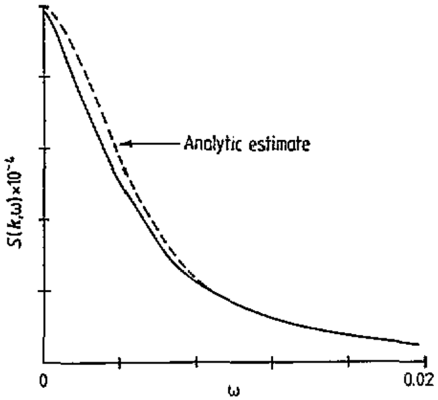

Figure 2. Results from $S(\boldsymbol{k},\omega)$ obtained from the analytic expression (1.2), derived for a spin-wave dispersion Dk^2 and a numerical evaluation of the full form given in (2.3) shown for EuO and a small wavevector, $k = 0.1(\pi/a)$. The other parameters are $H = 0.1$ T and $T = 0.25T_c$.

long wavelengths [1]. The contribution to the longitudinal response function induced by dipolar forces is significantly different from $S_0(\boldsymbol{k},\omega)$, at least at long wavelengths, where it stems from two spin-wave creation events, which contribute even at absolute zero temperature [2]. The dipolar induced response vanishes below

$$\omega_0 = 2(h^2 + 2\epsilon h)^{1/2} \tag{1.4}$$

where ϵ determines the strength of the dipolar forces (see table 1), and it decreases rapidly with increasing ω.

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

	J_1 (K)	J_2 (K)	D (meV Å^2)	M_0 (G)	ϵ (meV)
EuO ($a = 5.14$ Å, $T_c = 69.5$ K)	0.61	0.12	11.65	1910	0.14
EuS ($a = 5.95$ Å, $T_c = 16.5$ K)	0.24	−0.12	2.56	1184	0.09

[a] After [3,4].

In this paper we provide, for the first time, a comprehensive survey of the longitudinal dynamic response function $S(\boldsymbol{k},\omega)$, including the influences of dipolar forces, Zeeman energy and temperature. Results are given for wavevectors out to the zone boundary. The definition of $S(\boldsymbol{k},\omega)$ is reviewed in the next section. While the result used in our calculations is based on linear (non-interacting) spin-wave theory, it might be useful quite close to the critical temperature, T_c, since the lowest-order processes that contribute to $S(\boldsymbol{k},\omega)$ involve two spin-wave events.

The numerical method for calculating $S(\boldsymbol{k},\omega)$, for arbitrary $\boldsymbol{k}$ and ω, is described in section 3. Results for realistic models of EuO and EuS are gathered in section 4, and section 5 contains a discussion of our findings.

2. Spin-wave expression for $S(\boldsymbol{k}, \omega)$

The longitudinal spin response function $S(\boldsymbol{k}, \omega)$ is the spatial and temporal Fourier transforms of the correlation function $\langle S_l^z S_m^z(t)\rangle$, where S_m^z is the spin component at the lattice site defined by $\boldsymbol{m}$ parallel to the spontaneous magnetization (z axis). In linear spin-wave theory, it is adequate to use for S^z the approximate relation [3]

$$S^z = S - S^- S^+/2S \tag{2.1}$$

where S is the magnitude of the spin, and $S^\pm$ are standard spin raising and lowering operators. It follows that $S(\boldsymbol{k}, \omega)$ is the sum of two components, one that vanishes unless $\omega = 0$, and a second non-trivial component derived from

$$(1/2S)^2 \langle S^- S^+ S^-(t) S^+(t)\rangle. \tag{2.2}$$

In linear spin-wave theory, $S^- \sim a^+$, where a and a^+ are Bose operators; hence (2.2) is of the form of a particle density autocorrelation function, cf. (1.2).

The full calculation of $S(\boldsymbol{k}, \omega)$ using linear spin-wave theory for a Heisenberg ferromagnet including dipolar and Zeeman energies is provided by Lovesey and Trohidou [1]. Here we limit ourselves to just recording the final expression, namely

$$\begin{aligned} S(\boldsymbol{k}, \omega) = [1 + n(\omega)] \frac{1}{N} \sum_{\boldsymbol{p},\boldsymbol{q}} \delta_{\boldsymbol{p}-\boldsymbol{q},\boldsymbol{k}} \{ \tfrac{1}{2} E(\boldsymbol{p}, \boldsymbol{q})(1 + n_{\boldsymbol{p}} + n_{\boldsymbol{q}})[\delta(\omega - E_{\boldsymbol{p}} - E_{\boldsymbol{q}}) \\ - \delta(\omega + E_{\boldsymbol{p}} + E_{\boldsymbol{q}})] + F(\boldsymbol{p}, \boldsymbol{q})(n_{\boldsymbol{p}} - n_{\boldsymbol{q}})\delta(\omega + E_{\boldsymbol{p}} - E_{\boldsymbol{q}})\}. \end{aligned} \tag{2.3}$$

In this expression, the wavevectors $\boldsymbol{p}$ and $\boldsymbol{q}$ are restricted to the Brillouin zone, and $n_{\boldsymbol{p}}$ is the Bose distribution function for a spin wave with energy $E_{\boldsymbol{p}}$ (the result $E_{\boldsymbol{p}} = Dp^2$ applies in the limit of long wavelengths for a simple isotropic ferromagnet). The structure factors $E(\boldsymbol{p}, \boldsymbol{q})$ and $F(\boldsymbol{p}, \boldsymbol{q})$ can be shown to satisfy E, $F > 0$ and E vanishes while $F \to 1$ in the absence of dipolar forces; explicit expressions are given in [1]. In the absence of dipolar forces, $S(\boldsymbol{k}, \omega)$ is generated by events that involve creation and annihilation of spin waves, as expected for density fluctuations, i.e. $a_{\boldsymbol{q}}^+(t) a_{\boldsymbol{p}}^-(t)$. Dipolar forces induce off-diagonal fluctuations, of the form $a_{\boldsymbol{q}}(t) a_{\boldsymbol{p}}(t)$, as well as contributing to the spin-wave energy and $F(\boldsymbol{p}, \boldsymbol{q})$. The result (1.2) is obtained from (2.3) on taking $E = 0$, $F = 1$, and using $E_k = h + Dk^2$, which is appropriate for long-wavelength spin fluctuations.

When $k \sim (\epsilon/D)^{1/2} \sim 0$ the contribution to $S(\boldsymbol{k}, \omega)$ induced by dipolar forces is, to a good approximation ($\omega \geqslant 0$),

$$S_{\mathrm{d}}(k \sim 0, \omega) = [1 + n(\omega)] \frac{1}{2N} \sum_{\boldsymbol{p}} E(\boldsymbol{p}, \boldsymbol{p})(1 + 2n_{\boldsymbol{p}})\delta(\omega - 2E_{\boldsymbol{p}}). \tag{2.4}$$

It can be shown that $S_{\mathrm{d}}(k \sim 0, \omega)$ vanishes for $0 < \omega < \omega_0$, where ω_0 is given by (1.4) [2]. In the limit of small magnetic fields, $h \to 0$,

$$S_{\mathrm{d}}(k \sim 0, \omega_0) \sim (T^2/he^{1/2}) \qquad h \ll e.$$

Hence, the dipolar contribution to the longitudinal response is large for small magnetic fields.

3. Numerical method

The longitudinal response function $S(\boldsymbol{k},\omega)$ has been evaluated numerically for a range of parameters. The summation in equation (2.3) was done over a mesh of evenly spaced points in the Brillouin zone. For values of k along symmetry directions the numerical procedure could be made more efficient by restricting the summation to a reduced region of k-space.

For a general k value, however, 6.4×10^7 points in k-space were sufficient to eliminate spurious structure in $S(\boldsymbol{k},\omega)$ due to the finite mesh size. Frequency was sampled in intervals of $\omega \simeq 0.03$ meV. The magnetic field is parallel to the easy axis, which is $(1,1,1)$ for both 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.

To assess the accuracy of our numerical procedure, we compare our numerical results for a very small value of k with the analytically exact ones given by equation (1.2). This comparison is given in figure 2. The full curve shows $S(k,\omega)$ for EuO without the dipolar forces for $k = 0.1(\pi/a)$ Å^{-1}, temperature $T = 0.25T_c$, in the presence of a magnetic field $H = 0.1$ T and the broken curve shows the results for $S(k,\omega)$ for the same values of k, T and H using equation (1.2). The good agreement between the two sets of results shown in figure 2 gives us confidence in our numerical method.

4. Results

4.1. General comments

Values of $S(\boldsymbol{k},\omega)$ defined in equation (2.3) and [1] have been obtained, for realistic models of EuO and EuS, using the numerical method described in section 3. Parameters for these two ferromagnets are gathered in table 1. The dipolar energy parameter ϵ that appears in equation (1.4) is defined to be

$$\epsilon = 2\pi g \mu_B M_0 \tag{4.1}$$

where M_0 is the saturation magnetization. The spin-wave energy $\omega_{\boldsymbol{k}}$ satisfies

$$\omega_{\boldsymbol{k}}^2 = (E_{\boldsymbol{k}} + h)(E_{\boldsymbol{k}} + h + 2\epsilon \sin^2 \theta_{\boldsymbol{k}}) \tag{4.2}$$

in which $\theta_{\boldsymbol{k}}$ is the angle between $\boldsymbol{k}$ and the easy axis, and $E_{\boldsymbol{k}}$ is the exchange energy derived from nearest (J_1) and next-nearest (J_2) shells of magnetic ions, which has the form Dk^2 in the limit of long wavelengths ($ak \ll 1$). Note that, for an FCC lattice, the spin-wave stiffness

$$D = a^2 2S(J_1 + J_2) \tag{4.3}$$

and for europium ions $S = 7/2$.

Regarding the Zeeman energy $h = g\mu_B H$, it is useful to note that, for a field $H = 1$ T and $g = 2.0$, we have $h = 0.116$ meV. At high temperature ($T \geqslant 0.8T_c$) the dipolar contribution to the spin dynamics can be safely mimicked by an effective magnetic field. It has been shown [5] that in this temperature range the dipolar energy in the spin-wave energy is adequately represented by use of the expression

$$\omega_{\boldsymbol{k}} = E_{\boldsymbol{k}} + h + \tfrac{2}{3}\epsilon. \tag{4.4}$$

The last term in this expression, obtained by averaging the dipolar contribution in (4.2) over the directions of $\boldsymbol{k}$, is just an addition to the Zeeman energy, i.e. an effective Zeeman energy equal to $(h + 2\epsilon/3)$.

To set the scene for our findings for $S(\boldsymbol{k},\omega)$ over a wide range of $\boldsymbol{k}$, and including the dipolar energy, we refer to figure 1. Here, we illustrate the frequency dependence of the longitudinal response at long wavelengths of a simple ferromagnet (no dipolar forces). The results are derived from equation (1.2). The intensity of the spin-wave excitation at $\omega \sim E_k$ is determined by the Zeeman energy, and there is no significant structure in the response near $\omega \sim -E_k$. In all other figures we report data only for $\omega \geqslant 0$. Note that the function in (1.2) has a first derivative with respect to ω that is positive definite at $\omega = 0$ for all values of the parameters.

Calculations reported in the balance of this section have been made for a field $H = 0.1$ T, so $h = 0.01$ meV. It is useful to bear in mind values of $(2\epsilon/3)$ for EuO and EuS, namely 0.09 and 0.06 meV, respectively. For both materials, the dipolar energy is therefore large compared to the Zeeman energy. All the calculations that include dipolar forces are made with equation (2.3) and not the simplified treatment, appropriate only at high temperatures, in which they are represented by an effective magnetic field of magnitude $2\epsilon/3$.

The results of calculations of the response function $S(\boldsymbol{k},\omega)$ in the presence of a magnetic field $H = 0.1$ T are shown in figures 3 and 4 for several values of k in the (1,0,0) direction less than or equal to the zone boundary for temperatures $T = 0.4T_c$ and $0.8T_c$. There, $S(\boldsymbol{k},\omega)$ is in units of eV^{-1}, the full curves give the response function without the dipolar forces and the broken curves give $S(\boldsymbol{k},\omega)$ including dipolar forces. The magnitude of the wavevector k is in units (π/a), which is 0.61 and 0.53 $Å^{-1}$ for EuO and EuS, respectively.

4.2. EuO

As we can see from figure 3, for a small value of the wavevector there is a spin-wave peak in $S(\boldsymbol{k},\omega)$ around the value predicted from the spin-wave theory ($\omega = Dk^2 + h = 1.10$ meV for $k = 0.5$ in figure 3(*a*)). The diminution of intensity at $\omega = 0$ fades with increasing k, cf. figures 1 and 3(*b*)–(*d*). The influence of the dipolar forces is to decrease the response function, and to suppress features in the intensity distribution, including the spin-wave peak. This finding is consistent with the notion that, in some respects, dipolar forces have an effect that is akin to an external magnetic field; some of the important differences to be borne in mind are that an external field creates a gap in the spin-wave spectrum, and it does not break the conservation of the total magnetization (in the direction of the field).

Moving to larger values of k, the results displayed in figures 3(*b*)–(*d*) reveal the appearance of a structured quasi-elastic peak, in contrast to a diminution of intensity for smaller wavevectors, and a pronounced peak at finite ω. The latter is not associated with excitation of a single spin wave, at least near the zone boundary, where it arises simply from a cut-off in intensity imposed by the delta functions in (2.3). It is this feature of the spectrum that is most severely affected by dipolar

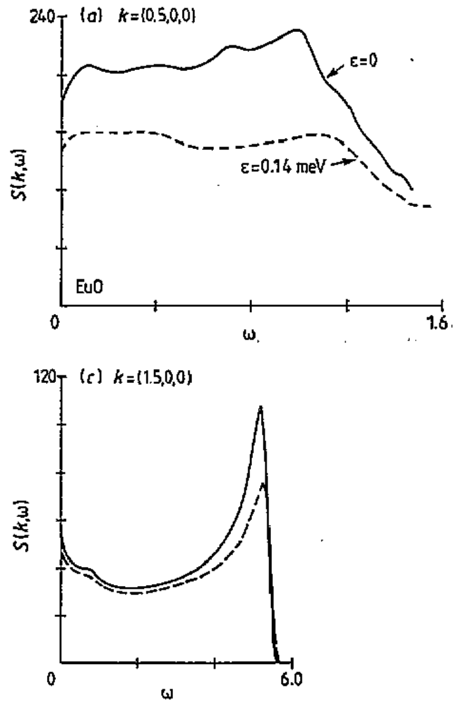

Figure 3. $S(\boldsymbol{k}, \omega)$ for EuO with $\epsilon = 0$ and 0.14 meV displayed for four values of $\boldsymbol{k}$ as a function of ω; $H = 0.1$ T and $T = 0.4T_c$.

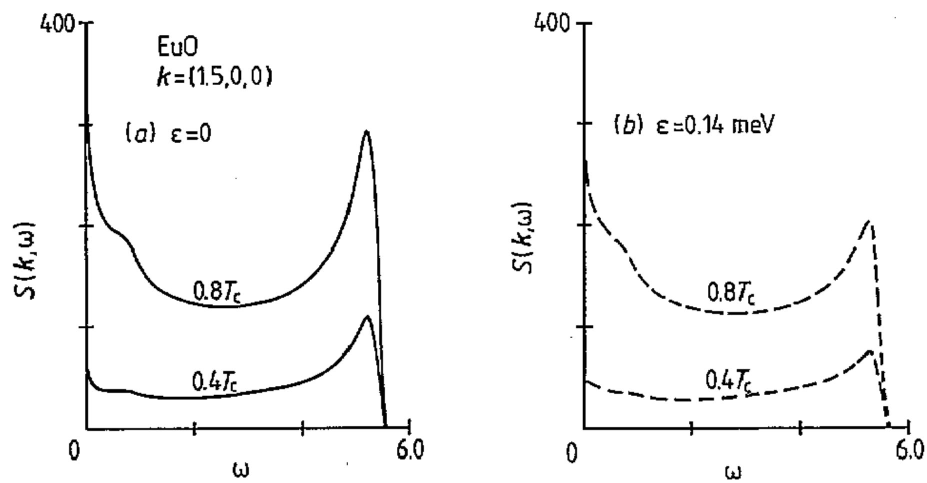

Figure 4. The influence of temperature on $S(\boldsymbol{k}, \omega)$ for EuO shown for $\boldsymbol{k} = (1.5, 0, 0)$ with and without dipolar forces.

forces, and relatively more so than the spin-wave peak in the spectrum for the smallest wavevector.

Figures 4(*a*) and (*b*) show the influence of temperature on $S(\boldsymbol{k}, \omega)$ with and without dipolar forces. As a rough guide, the intensity, being derived from two spin-wave events, might be expected to vary as the square of the temperature, i.e.

a factor of 4 difference between the two plots in each figure. With regard to the high-frequency peak in the spectra, this is an overestimate, and the factor is more like 2.7.

An investigation of the dependence of $S(\boldsymbol{k}, \omega)$ on the direction of $\boldsymbol{k}$ revealed an anticipated behaviour. For small k the spectrum is almost isotropic, whereas for $\boldsymbol{k}$ at the zone boundary the slight variation stems from changes in the cut-offs imposed by the delta functions in (2.3). In this respect, EuS, to which we next turn, is more interesting.

4.3. EuS

Results shown in figure 5 are to be compared with the corresponding data for EuO displayed in figure 3. The main differences between the two sets of results arise from a change in effective energy scales, brought about by the competing nature of the exchange interaction in EuS, e.g. T_c for EuO and EuS is in the ratio 4.2:1, cf. table 1. Thus, we find that the width of $S(\boldsymbol{k}, \omega)$ for EuS is much less than for EuO; in consequence, structure is less pronounced in the relatively compressed spectrum, but the maximum intensities are larger.

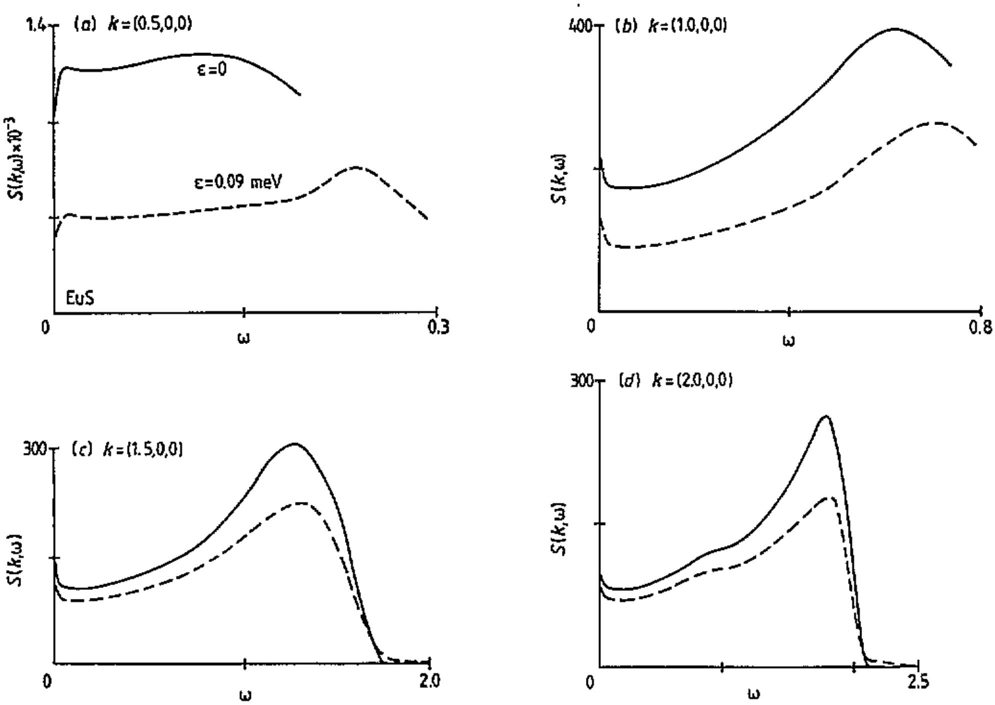

Figure 5. Comparison data to figure 3 shown for EuS; $\epsilon = 0$ and 0.09 meV.

Another effect of the competing interactions in EuS is pronounced spatial anisotropy. This feature is illustrated in figure 6, which shows $S(\boldsymbol{k}, \omega)$ with $k = (2\pi/a)$ and $\boldsymbol{k}$ directed along the three principal directions (relative to the easy axis). Once more, we find that the main effect of dipolar forces is to diminish the intensity of $S(\boldsymbol{k}, \omega)$, rather than to modify the distributions of intensity.

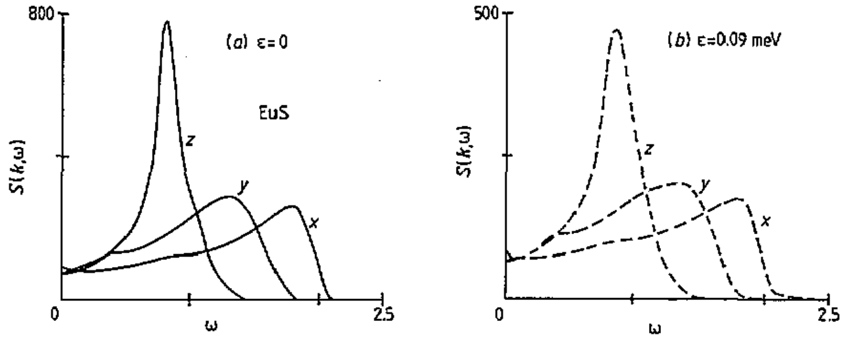

Figure 6. $S(\boldsymbol{k},\omega)$ for EuS with $\epsilon = 0$ and 0.09 meV displayed for three different directions of $\boldsymbol{k}$ with $k = 2\pi/a$; $T = 0.4T_c$ and $H = 0.1$ T.

5. Discussion

Our comprehensive investigation of the influence of dipolar forces on the longitudinal dynamic response function $S(\boldsymbol{k},\omega)$ shows that the main effect, for both EuO and EuS, is a reduction in intensity by roughly a factor of 2. It seems that no new structure in $S(\boldsymbol{k},\omega)$ results from the dipolar forces. These findings are in accord with previous results for the isothermal susceptibility, which is formally the inverse frequency moment of $S(\boldsymbol{k},\omega)$, equation (1.1).

There is a rich structure in $S(\boldsymbol{k},\omega)$ with variation of k. For small k, the quasi-elastic peak is weak and of an intensity approximately equal to the spin-wave contribution (in a finite field). The result for a simple magnet, illustrated in figure 1 and appropriate for a small k, never displays more than one peak. In the opposite extreme, when k is at the zone boundary, the quasi-elastic response is more intense than the high-frequency contribution. For a large k, the latter is not attributable to the excitation of a spin wave, but arises from a cut-off in intensity created by the energy-conserving delta functions in the spin-wave formula for $S(\boldsymbol{k},\omega)$.

Let us consider the effect of non-linear spin-wave events. These represent spin-wave fluctuations that become more important as $T \to T_c$. In consequence, including non-linear events increases $\chi(\boldsymbol{k})$ beyond the values obtained from simple (linear) spin-wave theory, and the increase becomes more pronounced as the critical temperature is approached [6]. Near T_c the dipolar forces can be mimicked by an effective magnetic field, which will, of course, suppress spin fluctuations. In conclusion, for given parameters, results provided here are consistent with a lower bound on $\chi(\boldsymbol{k})$ (the integrated intensity).

Acknowledgment

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