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S W Lovesey

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Stephen W. Lovesey

Rutherford Appleton Laboratory,
Oxfordshire OX11 OQX, U.K.

Abstract

Results from a theoretical analysis of the Heisenberg model of a ferromagnet, including dipolar interactions, are successfully used to interpret recent measurements performed on EuS, just below the critical temperature, that probe the dynamics of long wavelength spin fluctuations parallel (longitudinal) and perpendicular (transverse) to the spontaneous magnetization. Attention is given in the paper to longitudinal fluctuations following this and other related experimental investigations on the topic. With a view to future experiments, properties of the spin dynamics in the critical region are discussed within the framework of the coupled mode approximation.

While realistic models of magnetic materials include dipolar forces, which are responsible among other things for demagnetization effects, many experiments that probe magnetic properties on an atomic scale are relatively insensitive to them. In the case of experiments designed to investigate the dynamics of spin fluctuations, dipolar forces are only influentual at very large wavelengths and small frequencies. To put this comment on a more quantitative basis, let us use a standard measure of the strength of the dipolar forces in a magnetic material, the dipolar wave vector q_d. If c is the exchange stiffness, determined by the strength and range of the Heisenberg exchange interactions, q_d is defined by,

$$\frac{2\pi}{v_o}(g\mu_B)^2 = c q_d^2 , \qquad (1)$$

in which v_o is the unit cell volume and g is the gyromagnetic factor; values of q_d for some materials of current interest are provided in Table 1. It is found that dipolar forces influence the dynamics of spin fluctuations with a wave vector $k \leq q_d$, while for $k >> q_d$ the Heisenberg exchange, which generates Bloch spin waves, is the dominant contribution to the Hamiltonian energy. Clearly, dipolar forces will be important in determining the nature of critical phenomena when the inverse correlation length κ ($\kappa = 0$ at the critical temperature T_c) is comparable to q_d.

Theoretical estimates of long wavelength spin wave frequencies (magnetostatic modes) have been provided by Holstein and Primakoff (1940) and are well understood. Perhaps the first estimate of the influence of dipolar forces on relaxation processes in the critical region was given by Huber (1971) who predicted their extreme importance in determining properties of EuO. He also exposed the special features of the dipolar induced relaxation process that stem from breaking conservation of the total spin (magnetization). Extensive work based on the coupled mode approximation now provides a fairly complete description which is consistent with available experimental findings of the wave vector and frequency dependence of spin fluctuations (Frey and Schwabl 1988, Frey, Schwabl and Thoma 1989).

Another aspect of the spin dynamics of a ferromagnet which has recently received attention is the nature of the spectrum of spin fluctuations parallel to the magnetization, often called longitudinal fluctuations. Measurements made on nickel, in which dipolar interactions are relatively weak (c.f. Table 1), show that the longitudinal spectrum is quasielastic, and very similar to fluctuations observed in the paramagnetic phase (Böni et al. 1991a). The investigators have drawn attention to the unexpectedly large width of the quasielastic spectrum, which is part of the reason why in earlier measurements the longitudinal spectrum was not successfully distinguished from the relatively intense spectrum of transverse fluctuations. However, at present there is no strong experimental evidence for the expected divergence of the integrated intensity (susceptibility) on approaching a magnetic Bragg reflection; see, for example, equ (6), and Cuccoli et al. (1993). The divergence in question is a manifestation of Goldstone bosons, and its prediction dates back to Holstein and Primakoff (1940).

Recent theoretical and experimental work on the influence of dipolar forces on dynamic critical phenomena has focused on properties in the ordered magnetic phase ($T<T_c$). Toperverg and Yashenkin (1992) have used a perturbative treatment of dipolar forces to calculate transverse and longitudinal relaxation rates required for the interpretation of ferromagnetic resonance signals observed in the temperature range up to the critical region. Here, we report findings for the wave vector and frequency dependent response functions observed for EuS in preparatory experiments by Böni (1993). One reason for investigating this material is that the relatively large q_d leads one to expect ready observation of dipolar induced effects. The calculation employs the coupled mode approximation, which should be reliable at high temperatures and in the critical region. There is complete accord between the experimental and theoretical findings at the level permitted by the preliminary nature of the measurements.

We shall choose to report and discuss our findings that directly relate to the experimental observations for EuS, then outline the main features of the calculations which lead to the results, and conclude with predictions for the extreme critical region. For the conditions of the experiment ($T = 0.87T_c$, $k = 0.19Å^{-1}$) it is sufficient to evaluate the dynamic self-energy (memory function) at the first level of approximation while in the critical region it is essential to make a self-consistent, or nonperturbative, calculation. The contribution to the self-energy we first consider is purely dipolar in origin, and naïvely of order λ^2 where the energy parameter $\lambda = cq^2_d$ (= 0.025 meV for EuS). When $k \leq q_d$ the pure exchange and exchange-dipolar induced contributions to damping are relatively small corrections, and they always vanish in the limit $k \rightarrow 0$.

By utilizing polarization analysis Böni (1993) has separated various contributions to the neutron scattering cross section. In view of the dipolar induced spatial anisotropy the direction of **k** relative to the magnetization is an important variable. Spin variables with components parallel and perpendicular to the spontaneous magnetization are referred to as longitudinal and transverse variables, respectively. The scattering geometry did not give access to events in which **k** has a component parallel to the magnetization. The structure of the cross-section observed in the experiments is reviewed by Lovesey and Trohidou (1991), together with a description of the transverse spectrum predicted by linear spin wave theory.

When **k** is perpendicular to the magnetization the full dipolar anisotropy contributes a gap in the spin wave dispersion at the Brillouin zone centre. The appropriate expression is found to be ($\mathbf{k} \perp z$),

$$\varepsilon_k = h + Dk^2 + \lambda < S > \qquad (2)$$

where h is the Zeeman energy, modified by sample shape dependent demagnetizing effects, $<S>$ is the thermally averaged spin moment (in the z - direction) and the spin wave stiffness $D = 2c <S>$. Böni (1993) observed well defined spin wave excitations in the transverse spectrum, consistent with our theory, and report a value $\lambda<S> = 0.041$ meV. This value, taken together with a knowledge of the exchange interactions, yields (for $T = 0.87\ T_c$) the values $D = 1.20$ meV $Å^2$ and $(<S>/S) = 0.47$, where $S = 7/2$. These and various other quantities required in the interpretation of the experiment are gathered in Table 1.

It is well established that the approximation for the temperature dependence of the spin wave stiffness given in the previous paragraph, in which D is proportional to the magnetization, does not give good agreement with experimental data. However, data are in tolerable agreement with a theory, based on the Dyson Hamiltonian, that predicts a renormalization of D with temperature coming from the change with temperature of the (spin wave) energy (Cuccoli et al. 1993). Hence, we are not surprized that the value for D given in Table 1, and used in subsequent numerical estimates, differs from the value deduced from preliminary experimental data (Böni et al., private communication). The discrepancy between the experimental finding and $D = 2c \langle S \rangle = 1.20$ meV Å^2 is completely consistent with results obtained by Cuccoli et al. (1993) from the more sophisticated theory. But, in the present context of a theory for damping internal consistency is respected with use of the quoted value for D. It is also worth remarking that, the value we have deduced for the magnetization is in accord with previous experimental work (Passell et al. 1976) and the theoretical investigation by Cuccoli et al. (1993).

The corresponding expression for the damping of the transverse spin wave is found to be ($\mathbf{k} \perp z$),

$$\Gamma_{\perp} = (1/15\pi)\left(\frac{\lambda T v_o}{4D}\right)^2 \left(\frac{\varepsilon_k}{c\langle S\rangle\omega}\right). \qquad (3)$$

We find $(\Gamma_{\perp}/\varepsilon_k) = 5\%$ at $\omega = \varepsilon_k$. The reported experimental observation that the spin wave line width in EuS is resolution limited is consistent with this estimate, which is the only source of damping in the limit $k \rightarrow 0$.

Turning next to the longitudinal fluctuations, we find that there is no collective mode and the damping ($\mathbf{k} \perp z$),

$$\Gamma(k) = \left(\frac{64\sqrt{2}}{15\pi}\right)\left(\frac{kD^{1/2}}{\omega^{3/2}}\right)(\lambda \langle S \rangle)^2 . \qquad (4)$$

Evaulated for ω at the gap energy we find $(\Gamma/\lambda\langle S\rangle) = 2.0$. These theoretical findings are in accord with the observations that, the longitudinal fluctuations are quasielastic, and their linewidth corresponds roughly to the energy of the transverse spin wave. Note that (3) and (4) are lower bounds in the sense that they are correct in the limit $k \rightarrow 0$. A small k, used in the experiments, will engage small damping contributions generated by pure exchange, and exchange - dipolar processes, and we have more to say on this issue later in the paper.

Non-linear spin wave events and dipolar forces have been shown to generate two other striking features of the neutron scattering cross-section. First, the spectrum of transverse fluctuations contains a quasi-elastic component whose intensity depends on the orientation of the wave vector relative to the reciprocal lattice vector that defines the selected ferromagnetic Bragg peak. Secondly, the intensity of the longitudinal quasi-elastic fluctuations does not decrease smoothly with increasing wave vector, as predicted by linear spin wave theory (Trohidou and Lovesey 1993), but displays some structure for k in the neighbourhood of q_d.

It is interesting to observe that (3) and (4) are consistent with a dynamic scaling exponent $z = 3 - \beta/\nu \sim 5/2$ $(\beta = \frac{1}{2}\nu\,(1 + \eta) \simeq \frac{1}{2}\,\nu)$. To this end, use dimensionless variables $(\omega/\lambda\langle S\rangle)$ and (κ/q_d). On the other hand, it is known that when the dipolar interaction dominates the Heisenberg exchange the marginal dimension = 3, and critical fluctuations are very weak (Als - Nielsen and Birgeneau 1977, Zinn - Justin 1990). In this case, critical exponents agree with the Landau, or molecular field, theory of a continuous phase transition applied to a non-conserved order parameter. Evidently the estimates (3) and (4) are appropriate for the isotropic phase, and the dipolar (anisotropic) phase is reached in the limit $T \rightarrow T_c$. To shed more light on this question, and the range of validity of the estimates (3) and (4), we examine the first correction to the Landau value of the disconinuity in the specific heat. For us to be correct in the use of the thermodynamic (perturbation) approach this must be a small correction, which is indeed the case when,

$$\tau >> G\left(\frac{1}{f}\ln\{f+\sqrt{(1+f^2)}\}\right)^2$$

where $f = (q_d/\kappa)$, the reduced temperature $\tau = (T_c - T)/T_c$ and the Ginzburg parameter,

$$G = \{v_o / 16\pi r_1^3 \Delta C)\}^2$$

in which ΔC is the discontinuity in the specific heat according to the Landau (molecular field) theory, and r_1 is a measure of the range of the exchange interactions. For EuS (EuO) we find G = 0.024 (0.005) and the condition is well satisfied. As $T \rightarrow T_c$, $f \rightarrow \infty$ and eventually the Ginzburg condition is not satisfied (in making this analysis $\kappa^2 \propto \tau$ in keeping with the Landau theory). In the limit $T \rightarrow T_c$, the full non-linear coupled-mode theory is essential to obtain a cross-over from the isotropic to dipolar (anisotropic) behaviour.

By way of contrast to the result (4) for the damping of longitudinal fluctuations in the extreme limit $k \rightarrow 0$, we give the results for the damping generated by the pure exchange mechanism and the first corrections due to dipolar forces. The result valid for small k and leading-order in h is,

$$\Gamma(\mathbf{k}) = \frac{v_o T k}{4\pi D \chi(\mathbf{k})}\{1 - \frac{\lambda \langle S \rangle D k^2}{3\omega^2}(7 + 3\cos^2\theta_{\mathbf{k}})\}, \qquad (5)$$

where $\theta_{\mathbf{k}}$ is the angle between **k** and the direction of the spontaneous magnetization (z-axis), and the appropriate form of the longitudinal susceptibility has been obtained by Lovesey and Trohidou (1991), namely,

$$\chi(\mathbf{k}) = (T v_o / 16 D^2 k). \qquad (6)$$

It is interesting to find that dipolar forces, treated as a weak perturbation, on the one hand decrease the longitudinal susceptibility by approximately a factor of two yet in $\Gamma(\mathbf{k})$ the effect of this change is partially negated by the explicit correction of order λ. The expression (6) for the susceptibility has been used in the calculations that lead to (3) and (4).

The results presented and discussed in the foregoing paragraphs have been obtained from a coupled mode analysis, generated by the generalized Langevin equation, in which five variables are treated on an equal footing. Four variables relate to transverse fluctuations $(S_k^{\pm}, S_{\mp k}^{\pm})$ and the fifth is S_k^z; a feature of the dipolar interactions is that off-diagonal correlations are finite, e.g. $\langle S_k^+ S_{-k}^+ \rangle \neq 0$. The equations of motion for the construction of the self-energies are,

$$i\dot{S}_k^+ = hS_k^+ + (2/N)\sum_p \{\mathcal{J}(p) - \mathcal{J}(k+p)\} S_p^z S_{k+p}^+ + (\lambda/N)\sum_q \{q_+ (q_- S_q^+ + q_+ S_{-q}^-) S_{q-k}^z\}, \tag{7}$$

and

$$\dot{S}_k^z = (i/N)\sum_p \{\mathcal{J}(k+p) - \mathcal{J}(p)\} S_p^+ S_{k+p}^-$$

$$+ (i\lambda/2N)\sum_q \{q_+ q_- (S_{-q}^+ S_{k-q}^- - S_q^- S_{q-k}^+) + q_+^2 S_q^- S_{k-q}^- - q_-^2 S_{-q}^+ S_{q-k}^+\}. \tag{8}$$

In these equations, $\mathcal{J}(\mathbf{q})$ is the spatial Fourier transform of the Heisenberg exchange interaction and $q_{\pm} = \hat{q}_x \pm i\hat{q}_y$ where $\hat{\mathbf{q}}$ is a unit vector (the spontaneous magnetization defines the z-axis). A review of the use of the Langevin equation formalism to construct coupled mode equations is given by Lovesey (1986), together with an introduction to dynamic critical phenomena, while Frey and Schwabl (1988) use the formalism to discuss the ordered isotropic ($q_d = 0$) Heisenberg magnet.

The longitudinal relaxation function $F(\mathbf{k}, t)$ satisfies,

$$\dot{F}(\mathbf{k},t) = -\int_0^t dt'\, F(\mathbf{k}, t-t')\, K(\mathbf{k},t') \tag{9}$$

and in the limit $k \to 0$, where dipolar terms in the equations of motion dominate, the coupled mode approximation for the memory function $K(\mathbf{k},t)$ is

$$K(\mathbf{k},t) = \left(\frac{4\lambda^2 T}{\chi(\mathbf{k})N}\right)\sum_{\mathbf{q}}(q_+q_-)^2\,\chi_\perp^2(\mathbf{q})\,\mathrm{Re}.F_\perp^2(\mathbf{q},t). \qquad (10)$$

Here, $\chi_\perp(\mathbf{q})$ and $F_\perp(\mathbf{q},t)$ are the transverse susceptibility and relaxation function, respectively. The result (4) is obtained from (10) evaluated with a linearized approximation for $F_\perp$ in which $F_\perp(\mathbf{k},t) = \exp(-it\varepsilon_k)$, and $\chi_\perp(\mathbf{k}) = \langle S\rangle/\varepsilon_k$. We will not write out the pure exchange and dipolar-exchange contributions to $K(\mathbf{k},t)$ used to obtain (5). In the latter both contributions are required to obtain the term proportional to λ.

The corresponding equation for $F_\perp(\mathbf{k},t)$ contains an oscillatory component at the natural frequency ε_k, while the memory function is,

$$K_\perp(\mathbf{k},t) = 2\lambda^2 T(\lambda\sin^2\theta_k + 2ck^2)\frac{1}{N}\sum_{\mathbf{q}}(q_+q_-)^2\chi(\mathbf{q})\chi_\perp(\mathbf{q})F(\mathbf{q},t)\mathrm{Re}.F_\perp(\mathbf{q},t). \quad (11)$$

Equation (11) evaluated with linearized relaxation functions leads directly to the estimate (3) for the damping of the transverse collective modes.

By way of orientation to a discussion of relaxation in the extreme critical region we give the result for a simple isotropic ferromagnet ($h = q_d = 0$). In the limit $(\kappa/k) \rightarrow 0$ the self consistent solution of the coupled mode equations is,

$$\Gamma(k) = \Gamma_\perp(k) = e(2/3)^{1/2}k^{5/2}\,;\ \{\Gamma_\perp/\Gamma\} = 1 - \frac{40}{11}(\kappa/k)^2,$$

where the nonuniversal energy constant,

$$e = (2T_c v_o c/3\pi^2)^{1/2}.$$

The critical exponent $z = 5/2$ has been verified in a number of experiments; see Böni et al. (1991a) and references therein.

The memory functions (10) and (11) for relaxation via purely dipolar processes have been analysed in the extreme critical region $T \to T_c$ using the expressions,

$$\chi_{\perp}(\mathbf{q}) = \{2c(q^2 + q_d^2 \sin^2\theta_q)\}^{-1}$$

and

$$\chi(\mathbf{q}) = \{2c(q^2 + q_d^2 \cos^2\theta_q)\}^{-1}.$$

As expected, we find there are no anomolous contributions to the memory functions generated by critical fluctuations. In the anisotropic, dipolar region accessed when $k << q_d$ ($T \to T_c$) the longitudinal and transverse damping functions are independent of the magnitude of the wave vector k, are proportional to $q_d^{5/2}$, and $\Gamma(\Gamma_{\perp})$ varies with $\hat{\mathbf{k}}$ as $\cos^2\theta_k(\sin^2\theta_k)$. The precise magnitude of these functions are determined by the self consistent solutions of (10) and (11). These results, together with a full account of all contributions for $T \leq T_c$ to the lineshapes and damping functions, will be reported in a separate paper. Attention has been given to the interpretation of neutron scattering and muon relaxation experiments. The latter experiments have the potential to provide information not readily obtained from neutron scattering (Lovesey et al. 1992, Yaouanc et al.1993).

Neutron scattering experiments that employ polarization analysis usually have the sample in a static magnetic field. Since a field suppresses critical fluctuations it is natural to question whether a field influences the experimental results we have mentioned. To this end, we have calculated the damping functions for a simple ferromagnet ($q_d = 0$) subject to a magnetic field. The relevant wave vector $q_o = (h/D)^{1/2}$ increases as $T \to T_c$ because D is proportional to the magnetization which ultimately has a field limited value. For the extreme case $(q_o/\kappa)>>1$, the self consistent solutions of the coupled mode equations are,

$$\Gamma(k) = \Gamma_{\perp}(k) = e(\pi/8)^{1/2} q_o^{1/2} k^2 . \tag{12}$$

and writing $x = (\kappa/q_o) < 1$ an expansion in x reveals at leading order,

$$\{\Gamma_{\perp}(k)/\Gamma(k)\} = 1 + (2x)^2.$$

For Ni at T = 0.99 T_c and H = 1.1kG one has (q_o/κ) = 0.38, while for EuS with H = 1.64 kG at the same relative temperature (q_o/κ) = 8.2. The large difference in these values of (q_o/κ) mainly reflects the fact that the stiffness parameter c is much larger for Ni than EuS, c.f. respective critical temperatures. Hence, a modest magnetic field is probably not influential in Ni, whereas it is a significant factor in determining the critical dynamics of EuS.

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Table 1

Material Parameters

Material	q_d (Å^{-1})
Ni	0.01
EuO	0.15
EuS	0.26
$LiTbF_4$	1.31

Experiment on EuS, Böni et al. (1992)

c	=	0.37 meV Å^2	
T	=	$0.87T_c$ = 1.24 meV	
k	=	0.19 Å^{-1}	
v_o	=	52.7 Å^3	
D	=	1.20 meV Å^2	
(<S>/S)	=	0.47	
r_1	=	1.40Å	
ΔC	=	2.42	(1)
H	=	1.64 kG	

(1) Stanley (1971)

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