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Spin density fluctuations in a Heisenberg ferromagnet

A Cuccoli†, S W Lovesey‡, G Pedrolli† and V Tognetti†

† Dipartimento di Fisica dell'Università di Firenze, Largo E Fermi 2, I-50125 Firenze, Italy

‡ Rutherford Appleton Laboratory, Oxfordshire OX11 0QX, UK

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Abstract. The longitudinal susceptibility, a measure of magnetization fluctuations, is studied within the framework of the Dyson Hamiltonian. Results for models appropriate to EuO and EuS show that, near T_c, the fluctuations included in the Hartree–Fock approximation more than double the susceptibility. It is shown that inclusion of the fluctuations does not change the exponent in the power law divergence of the susceptibility with respect to a magnetic field. Results for the magnetization and spin-wave renormalization, including the influence of the dipolar energy, are given for EuO and EuS, and compared favourably with the available experimental data.

1. Introduction

Several groups of researchers have recently noted a somewhat remarkable shortage of precise experimental information on the spectrum of spin density fluctuations in simple magnets that are adequately described by a Heisenberg spin Hamiltonian. Moreover, the data produced by these groups are not in striking agreement with predictions based on linear spin-wave theory; predictions that have been known for several decades. Couched in terms of the wavevector-dependent susceptibility $\chi(\boldsymbol{k})$, the prediction for zero magnetic field is $\chi(\boldsymbol{k}) \propto (1/k)$ near the centre of the Brillouin zone; alternatively, for a finite magnetic field H, $\chi(0) \propto (H)^{-1/2}$.

Neither of these predictions are in tolerable agreement with available data, much of which has been obtained with the neutron scattering technique. The shortage of precise experimental data is largely due to the fact that this technique is not ideally suited to the measurement of the spectrum of spin density fluctuations. Unfortunately, it is the only technique currently available for the measurement of $\chi(\boldsymbol{k})$, which in neutron scattering parlance is referred to as the longitudinal spectrum. In contrast, the transverse spectrum, exhausted by single spin-wave events, is well-researched and understood. Indeed, measurements of the transverse spectrum are the source of data on spin-wave dispersions in magnetic materials. The neutron scattering technique is not ideal for the measurement of $\chi(\boldsymbol{k})$ because it is not observable in isolation from the relatively intense transverse spectrum, unless polarization analysis is employed, although this has the drawback of a significant intensity penalty.

The absence of experimental verification of the divergence of $\chi(\boldsymbol{k})$ at the zone centre, due to a Goldstone mode in the isotropic spin Hamiltonian, prompted investigation of the influence of dipolar forces on the susceptibility. At face value, dipolar forces are a prime candidate for changing the structure of $\chi(\boldsymbol{k})$ because they change the spin symmetry of the Hamiltonian (the total spin is no longer a constant of motion). However, the conclusion is that these forces do not change the structure of $\chi(\boldsymbol{k})$, as far as $\boldsymbol{k}$ is larger than the characteristic wavevectors of the magnetostatic modes. The effect of dipolar forces, to a first approximation, is simply to reduce by about 50% the value of $\chi(\boldsymbol{k})$ from that obtained

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for the isotropic spin system. Detailed calculations have been reported [1,2] for realistic models of EuO and EuS, both of which are simple face-centred cubic ferromagnets with ordering temperatures T_c of 69.5 K and 16.5 K, respectively.

While the investigation of dipolar forces removes an important query in confronting theoretical predictions with experimental data, there remains concern about use of a linear spin-wave approximation, given that experiments are performed close to T_c where the susceptibility is enhanced by fluctuations that are precursors to critical effects. Here, we report the first findings on the influence of non-linear spin wave interactions on the spin density susceptibility, and explicit results for realistic models of EuO and EuS, including accounts of dipolar forces. It is shown that $\chi(0) \propto H^{-1/2}$, i.e. the interactions do not change the exponent in the divergence with respect to the magnetic field. For weak fields, the enhancement of χ by non-linear spin fluctuations is as much as 50% for $T \simeq 0.9T_c$. Inclusion of dipolar forces decreases χ, as already mentioned, but the relative enhancement by non-linear fluctuations remains almost the same.

Let us denote the spin-wave dispersion in a simple ferromagnet ($T < T_c$) by ω_k. The frequency- and wavevector-dependent susceptibility for spin density fluctuations, obtained from linear spin-wave theory, is

$$\chi_0(\boldsymbol{k}, \omega) = (1/N) \sum_{q} \frac{n_q - n_{q+k}}{\omega + \omega_{q+k} - \omega_q} \tag{1}$$

in which

$$n_k = [\exp(\omega_k/T) - 1]^{-1} \tag{2}$$

is the Bose factor ($\hbar = k_B = 1$).

The result for $\chi(\boldsymbol{k}) \equiv \chi(\boldsymbol{k}, 0)$ obtained from (1) is to be contrasted with the corresponding susceptibility for transverse spin fluctuations, namely $2S/\omega_k$, where S is the magnitude of the spin ($S = 7/2$ for EuO and EuS). From (1) we obtain, using the approximation

$$n_k \simeq T/\omega_k \qquad \omega_k \simeq h + Dk^2 \qquad h = g\mu_B H \tag{3}$$

the result

$$\chi_0(0) = T v_0 / 8\pi D^{3/2} h^{1/2} \tag{4}$$

where v_0 is the volume of the unit cell. The estimate (4) is good for temperatures close to T_c, and it displays the anticipated inverse square-root singularity with respect to the magnetic field. It will be shown that, for EuO and EuS and $T \simeq T_c$, (4) significantly underestimates the non-interacting susceptibility.

Our treatment of non-linear spin-wave interactions is based on two spin-wave events described by Dyson's boson Hamiltonian. Evidence to support this treatment comes from several sources. First, it provides an *exact* description of the spin-wave bound state. Second, results obtained for the renormalized spin-wave energies are in excellent agreement with experimental data. In the same vein, critical temperatures obtained from the theory agree well with observed values.

The structure of the equation for the spin density propagator (Green function) has the form of that for a localized single-defect problem of the Lifshitz type, and it can be solved by the established method [3]. For the problem in hand, the defect potential is generated by the dynamic spin-wave interaction. Further details and an explicit solution for a (one-dimensional) chain of spin can be found in [4–6].

Here, we limit attention to the expression for $\chi(\boldsymbol{k} = \boldsymbol{0})$, and reserve for later publication the full expression for $\chi(\boldsymbol{k})$.

2. Renormalized spin-wave theory

The theory of two-spin-wave interactions described by the reduced Hamiltonian of Dyson and its treatment within the Hartree–Fock approximation has recently been reviewed [7]. In view of this, the presentation given here is minimal, and is designed to really do no more than define notation and provide the basic concepts. Our Hamiltonian $\hat{\mathcal{H}}$ is the sum of an isotropic exchange interaction between spins $\boldsymbol{S}_n$, located on a Bravais lattice defined by vectors $\boldsymbol{n}$, and a Zeeman energy created by a magnetic field in the z direction of the coordinate system:

$$\hat{\mathcal{H}} = -\sum_{m,m'} J(\boldsymbol{m}-\boldsymbol{m}')\boldsymbol{S}_m \cdot \boldsymbol{S}_{m'} - h\sum_m S^z_m. \tag{5}$$

Here $J(\boldsymbol{m}-\boldsymbol{m}')$ is the exchange parameter, and is defined such that $J(\boldsymbol{0}) = 0$.

The reduced Hamiltonian introduced by Dyson is the sum of non-interacting spin-waves, with dispersion:

$$\omega_k = 2S(J_o - J_k) + h \tag{6}$$

in which S is the magnitude of the spin and

$$J_k = \sum_m J(\boldsymbol{m})\cos(\boldsymbol{k}\cdot\boldsymbol{m}). \tag{7}$$

The Dyson Hamiltonian is the lowest-order approximation to the dynamic interaction, and correctly describes two-spin-wave processes, e.g. the two-spin-wave bound state. When corrections to the Hartree–Fock approximation for the dynamic interaction are set aside, the dispersion relation is modified by a simple temperature-dependent factor. The renormalized dispersion is

$$\epsilon_k = \omega_k - \frac{2}{N}\sum_q n_q(J_o + J_{k-q} - J_k - J_q) \tag{8}$$

where now

$$n_q = [\exp(\epsilon_q/T) - 1]^{-1}. \tag{9}$$

Results (8) and (9) together give a transcendental equation for the dispersion and occupation number. The corresponding magnetization is determined by

$$\frac{\langle S^z\rangle}{S} = 1 - \frac{1}{NS}\sum_q n_q. \tag{10}$$

Results for the special case of an exchange interaction restricted to one shell of neighbouring magnetic ions, for which $J_q = rJ\gamma_q$ and r is the number of ions in the shell, are discussed in the appendix. A realistic model for EuO and EuS contains two shells of neighbours, and implementation of the theory is necessarily slightly more complicated than for one shell of neighbours. Results for the spin-wave renormalization and magnetization are reported in the next section, together with an estimate of the influence of dipolar forces.

It is well known that dipolar forces have a significant effect on the structure of spin-wave theory of a Heisenberg ferromagnet [7, 8]. However, at elevated temperatures, where many spin waves are excited, their influence is adequately described by the effect of a magnetic field proportional to the magnetization (in this approximation, off-diagonal terms in the Hamiltonian that have a considerable impact on low-temperature properties are neglected). The theory including dipolar forces can thus be mapped into the one described in the previous paragraphs with the magnetic field replaced by the sum of the applied field and the anisotropy energy

$$h_a = \frac{\xi \langle S^z \rangle}{S} \tag{11}$$

in which the constant ξ is proportional to the saturation magnetization; values of ξ for EuO and EuS are provided in table 1.

Table 1. Some useful quantities for EuO and EuS: a is the lattice constant, T_c the experimental critical temperature, J_1 and J_2 the NN and NNN exchange-interaction constants, ξ the parameter, defined in the text, which rules the dipolar effects. Values of ξ are obtained from $\xi = \frac{2}{3}(2\pi g\mu_B M_0)$, where M_0 is the observed saturation magnetization.

	a (Å)	T_c (K)	J_1 (K)	J_2 (K)	ξ (K)
EuO	5.12	69.5	0.61	0.12	1.08
EuS	5.95	16.5	0.24	−0.12	0.67

With the inclusion of the dipolar energy as described above, the magnetization and dispersion are determined by the coupled equations (8) and (10) together with (9) and (11). An investigation of the theory, using a numerical method, is reported in section 3. Fortunately, some key aspects of the results can be understood from a simple analysis reported in the appendix. The equations admit physically acceptable solutions up to a maximum temperature that is quite close to the observed critical temperature. At this maximum temperature, the magnetization and renormalization of the spin-wave dispersion are finite, and the values obtained in a numerical analysis are very close to the estimates reported in the appendix. On the question of the influence of the dipolar force, analytic and numerical works are in good agreement, and the induced changes are found to reduce the discrepancy between theory and experimental data. At this juncture we mention that Passell and co-workers [9] also report numerical calculations of the magnetization and spin-wave renormalization including dipolar energy. However, in their treatment of the dipolar energy the magnetization is inserted (experimental values) whereas here the entire set of physical variables is treated in a self-consistent manner.

3. Magnetization and renormalized dispersion

The theory outlined in section 2 has been applied to a realistic model of the magnetic salts EuO and EuS; various parameters are gathered in table 1. In the absence of dipolar anisotropy ($\xi = 0$), the maximum temperature at which there is a solution of the transcendental equation is 62.5 K for EuO and 16.2 K for EuS. Including the dipolar anisotropy in the approximate form described in the previous paragraph increases this temperature to 63.5 K and 16.8 K respectively. It is gratifying to find that these changes are in good agreement with the estimates derived in the appendix, as are the dipolar-induced changes to the magnetization and spin-wave renormalization.

Figures 1 and 2 contain results for $\langle S^z \rangle / S$ and ϵ_q for EuO and EuS as functions of the temperature, together with available experimental data. The applied field is zero in all cases. The dipolar anisotropy is included in the form described in section 2, and for EuO it goes toward reducing discrepancies between theoretical and experimental data, while for EuS no substantial improvement is obtained.

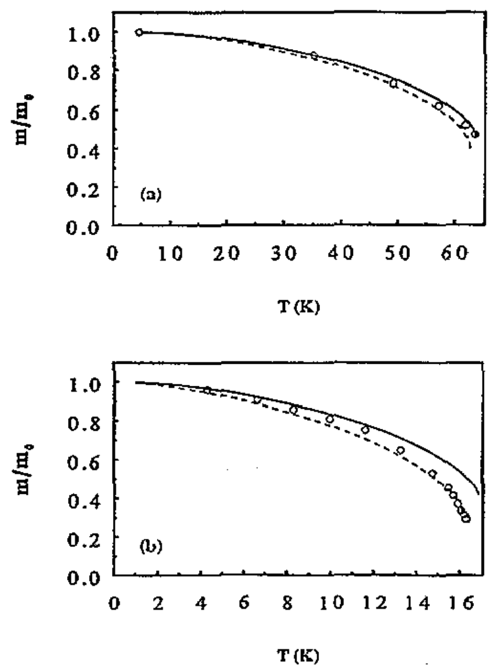

Figure 1. Reduced magnetization against temperature for EuO (*a*), and EuS (*b*). The broken curve is the result for the pure exchange Heisenberg ferromagnet, while the full curve also takes into account the dipolar interaction as described in section 2. The open circles are the experimental results [9].

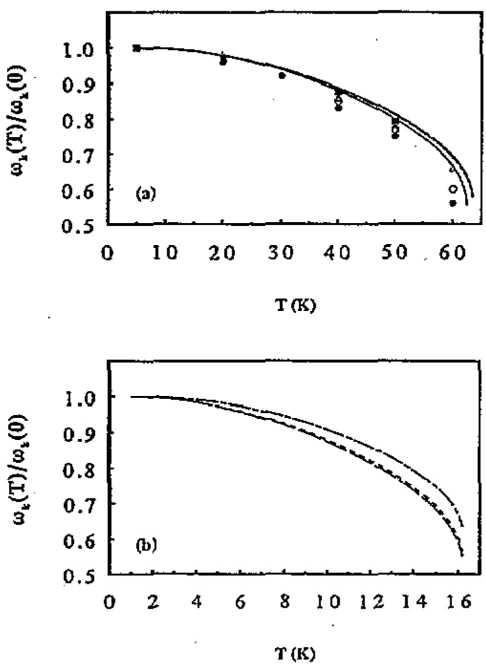

Figure 2. The ratio between the renormalized and bare spin-wave frequency at the zone boundary for EuO (*a*) and EuS (*b*) is reported as a function of temperature. In (*a*) the full curve is the result for the pure exchange Heisenberg ferromagnet, and the long-dash broken, broken and chain curve the results with the dipolar interaction in the (001), (011) and (111) direction, respectively. In (*b*) all the curves are obtained without any dipolar interaction and refer to the (001) (long-dash broken curve), (011) (broken curve), and (111) (chain curve) direction, respectively. The symbols are experimental results for some values of k [9]; full circles: $k = 0.2\,\text{Å}^{-1}$, open circles: $k = 0.6\,\text{Å}^{-1}$, open triangles: $k = 0.8\,\text{Å}^{-1}$, open squares: $k = 1.0\,\text{Å}^{-1}$.

The calculation given in the appendix for a nearest-neighbour exchange model predicts that the magnetization and spin-wave renormalization should be 0.33 and 0.50, respectively, for $\xi = 0$ and at the maximum temperature. The good agreement between the full numerical analysis and these estimates vindicates the assumption that, for high temperatures, it is adequate to use $n_q \simeq T/\epsilon_q$.

4. Longitudinal susceptibility: theory

The longitudinal susceptibility in the classical limit is proportional to the mean-square fluctuation in the magnetization, i.e.

$$\chi(\boldsymbol{k}) \propto \langle (S_{\boldsymbol{k}}^{z})^2 \rangle \qquad \boldsymbol{k} \neq 0 \tag{12}$$

where $S_{\boldsymbol{k}}^{z}$ is the spatial Fourier transform of the magnetization. For a quantum mechanical calculation, a convenient way to proceed is to obtain the Zubarev Green function for the magnetization density operator. The Green function evaluated at zero frequency is proportional to $\chi(\boldsymbol{k})$; see, for example, [10]. Calculated for linear spin-waves with dispersion ω_q the Green function, denoted by $K_0(\boldsymbol{k})$, is simply related to the wavevector- and frequency-dependent susceptibility (1), namely

$$\chi_0(\boldsymbol{k}) = \chi_0(\boldsymbol{k}, 0) = -K_0(\boldsymbol{k}). \tag{13}$$

Taking the limit $k \to 0$ in $\chi_0(\boldsymbol{k}, 0)$ leads to

$$\chi_0(0) = \frac{1}{NT} \sum_{\boldsymbol{q}} n_{\boldsymbol{q}}(1 + n_{\boldsymbol{q}}). \tag{14}$$

In the classical limit $n_q \gg 1$, and then $\chi_0(0)$ is seen to be the mean-square fluctuation in the magnetization. On using the approximation $n_q \simeq T/\omega_q$ in (14), replacing the sum by an integral, in the standard manner, and using the low-k expansion for $\omega_{\boldsymbol{k}}$ we arrive at the result (4). For the simplified nearest-neighbours model discussed in the appendix, the behaviour of the macroscopic static susceptibility for vanishing field is easily recovered without introducing the continuum approximation for the spin wave dispersion. From (A14), when the effects of renormalization are neglected, we get

$$\chi_0(0) = -\frac{T}{e_0^2} I'(\beta) \tag{15}$$

where $I'(\beta)$ is the derivative of the extended Watson integral. Using the expansion (A5), together with $D = 2JSa^2$ and $v_0 = a^3/4$, we see that (15) is identical to (4).

The Green function for the reduced Hamiltonian of Dyson cannot be calculated without approximation. In the present work, the approximation used is well tried and tested; it yields the exact spectrum of the two-spin-wave bound state, and is entirely consistent with the renormalized spin-wave theory described in section 2. The full form of the Green function is quite complicated, and it is readily obtained from the main result provided by Balucani and co-workers [5] in their discussion of fluctuations in the Heisenberg chain. In view of this, we limit ourselves to providing the results for the susceptibility without details of the intermediate working. One new aspect of our result is the generalization to two shells of neighbours labelled by $i = 1, 2$. The Fourier transform of the exchange interaction is

$$J_q = \frac{1}{2} \sum_i \theta_i \gamma_q^{(i)} \tag{16}$$

where $\gamma_q^{(i)}$ are standard geometric factors with the normalization $\gamma_0^{(i)} = 1$, and $\theta_i = 2r_i J_i$.

One effect of the spin-wave interactions is to renormalize the dispersion as described in section 2. Hence, in the remaining discussion n_q is the function (9) in which the dispersion

ϵ_q depends on temperature. The function $K(k)$ used in the following is simply the right-hand side of (1) with ω set to 0 and ω_q replaced by ϵ_q.

The susceptibility is expressed in terms of three functions, in addition to K, that have a structure similar to K. The three additional functions of interest are:

$$\begin{aligned} A^{(i)} &= -\frac{1}{NT}\sum_q n_q(1+n_q)(1-\gamma_q^{(i)}) \\ B^{(i)} &= -\frac{1}{NT}\sum_q n_q(1+n_q)(1-\gamma_q^{(i)})^2 \\ C &= -\frac{1}{NT}\sum_q n_q(1+n_q)(1-\gamma_q^{(1)})(1-\gamma_q^{(2)}) \end{aligned} \tag{17}$$

in terms of which

$$\begin{aligned} \chi(0) = -K(0) + \{\theta_1(A^{(1)})^2 + \theta_2(A^{(2)})^2 + \theta_1\theta_2[(A^{(1)})^2B^{(2)} + (A^{(2)})^2B^{(1)} \\ - 2A^{(1)}A^{(2)}C]\}[1+\theta_1B^{(1)} + \theta_2B^{(2)} + \theta_1\theta_2(B^{(1)}B^{(2)} - C^2)]^{-1}. \end{aligned} \tag{18}$$

Several features of this expression merit some comment. First, in the limit of zero magnetic field $\chi(0) \propto h^{-1/2}$. This behaviour arises, for temperatures less than the maximum temperature, because the three functions listed in (17) are not singular for $h \to 0$ owing to the additional geometric factors in the integrands ($(1-\gamma_q) \propto q^2$ for $q \to 0$). Note that, if we found a result for χ that was not singular for $h \to 0$, the result would be unacceptable, since the singularity is a consequence of a spin symmetry of the Hamiltonian, which must be respected by approximations to observable response functions. Hence, the significant finding is that the exponent of h in $\chi(0)$ is the same as in the non-interacting spin-wave approximation. Second, we find the result $\chi \geqslant \chi_0$. This result is in accord with physical intuition because the corrections to χ_0 in (18) are created by the spin fluctuations, which eventually drive the continuous phase transition as $T \to T_c$. The influence of the spin fluctuations on the susceptibility evaluated in the limit $h \to 0$ is obtained from the relation, derived from (18)

$$\lim_{h\to 0}\frac{\chi}{\chi_0} = \lim_{h\to 0}\frac{K}{K_0}. \tag{19}$$

If we evaluate $K(0)$ in the same fashion as $K_0(0)$, this is evaluated to obtain the estimate (4)

$$\lim_{h\to 0}\frac{\chi}{\chi_0} = \left(\frac{D(0)}{D(T)}\right)^{3/2} \tag{20}$$

where the temperature-dependent spin-wave stiffness $D(T)$ is derived from ϵ_q, namely

$$\epsilon_q = D(T)q^2 \qquad \text{for} \quad h=0,\ aq \ll 1. \tag{21}$$

At low temperatures, $D(0)/D(T)$ increases from unity at $T=0$ with a term proportional to $T^{5/2}$. Figure 3 shows the temperature dependence of $[D(0)/D(T)]^{3/2}$ for EuO and EuS over the entire temperature range for which Hartree–Fock theory is valid.

Finally, we remark on the structure of $\chi(0)$ evaluated with the analysis discussed in the appendix, in which there is just a single shell of ions. With a more or less obvious notation, for a single shell of ions the result (18) reduces to

$$\chi(0) = -K(0) + \frac{\theta A^2}{1+\theta B}. \tag{22}$$

With the approximation $n_q(1+n_q) \simeq (T/\epsilon_q)^2$ we have for the case of zero magnetic field

$$A = -TI/e^2 \qquad B = -T/e^2 \tag{23}$$

where I is the Watson integral and e is the renormalized exchange integral, i.e.

$$\epsilon_q = e(1-\gamma_q) \qquad h = 0. \tag{24}$$

Perhaps the most interesting aspect of these expressions is that, at the maximum temperature for which the Hartree–Fock scheme admits a physical solution of the transcendental equations for the spin-wave renormalization, the analysis in the appendix leads to the result

$$1+\theta B = 1 - \frac{\theta T}{e^2} = 0. \tag{25}$$

Hence, within the approximate analysis based on $n_q \simeq T/\epsilon_q$, which is shown in section 3 to provide a quantitative description of (exact) numerical results, the maximum temperature for a physical solution is also the temperature at which the additional term in the susceptibility, which arises from spin-wave interactions, diverges too. It is particularly satisfying that the divergence with respect to the field h is the same as that of $K(0)$, i.e. at the (zero-field) maximum temperature $T = e_0 S/4$ we obtain

$$1+\theta B \propto h^{1/2} \tag{26}$$

in the limit $h \to 0$, while the function A in the numerator is finite.

5. Longitudinal susceptibility: predictions for EuO and EuS

The result (18) for the susceptibility has been evaluated for the same realistic model of EuO and EuS as we used in section 3 to investigate the magnetization and spin-wave renormalization. We report for these two compounds our predictions with respect to the dependence of the susceptibility on temperature, magnetic field and dipolar anisotropy.

Results for $[D(0)/D(T)]^{3/2}$ in figure 3 give a measure of the influence of spin fluctuations on the susceptibility as a function of temperature. In the temperature range covered in the figures for both materials the susceptibility increases by a factor of about two, and beyond the maximum temperature contained in the figure the susceptibility continues to increase.

In figure 4 we show for EuO and EuS the field dependence of χ and χ_0. The log–log plots illustrate the power law behaviour with respect to h, which is common to χ and χ_0. At the chosen relatively high temperature we see a significant enhancement of the susceptibility by the spin fluctuation effects contained in χ, while the enhancement is found to be negligible at lower temperatures ($T \lesssim 0.7T_c$), as might be expected. We draw attention to the mild field dependence of $(\chi - \chi_0)$.

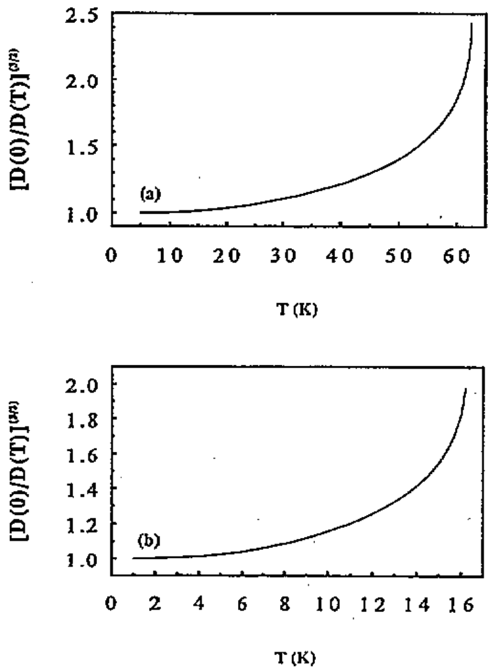

Figure 3. The ratio $[D(0)/D(T)]^{3/2} = K(T)/K(0)$ against temperature for EuO (*a*) and EuS (*b*).

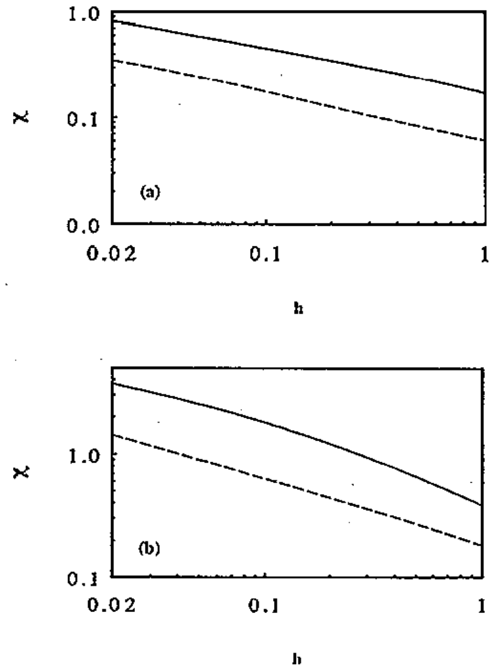

Figure 4. Macroscopic static susceptibility against applied field for EuO at $T = 60\,\mathrm{K}$ (*a*) and EuS at $T = 16\,\mathrm{K}$ (*b*). The broken and full curves refer to the non-interacting and interacting case, respectively, and the Zeeman energy $h = g\mu_B H$ is in units of K.

6. Conclusions

The work reported has essentially two threads. While the main aim is to estimate the influence of spin fluctuations on the longitudinal susceptibility, this naturally entails a study of magnetization and spin-wave renormalization. Results from the latter part of the study are in accord with previous theoretical findings and available experimental data. A new feature of our theoretical work is that the dipole energy contribution is handled in a fully consistent manner, whereas previously recourse was made to experimental data.

Our results for the susceptibility show the significant underestimate of spin fluctuations by the standard spin-wave expression. For a temperature $\simeq 0.96T_c$ the fluctuations more than double the size of the susceptibility, and on approaching the critical temperature from below the susceptibility displays a power law divergence. This, together with other aspects of the study, are clearly revealed in an analysis of a simple model calculation appropriate at high temperatures.

Our results for the longitudinal susceptibility of EuO and EuS can be directly tested with neutron scattering experiments. Less direct evidence is contained in the analysis of muon spin relaxation experiments, where the relaxation rate is proportional to a weighted integral of the susceptibility $\chi(k)$ divided by the damping rate for magnetization fluctuations.

Acknowledgments

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Appendix

While a numerical method must be used to obtain a complete solution of the transcendental equations for the magnetization and renormalization factor, some useful insight to the nature of the solutions can be obtained by elementary analysis, based on a nearest-neighbour model and high-temperature expansion of the Bose factor.

First of all we recall that at very low temperatures the magnetization can be expanded in a power series in T/J, and for a simple magnet the leading-order contribution from the dynamic interaction is found to be proportional to $(T/J)^4$. Furthermore, a quantum spin reduction is present due to the dipolar anisotropy. However, due to the high value of the spin, this reduction turns out to be negligible in the case of interest, being less than 0.2% and 0.6% for EuO and EuS, respectively.

Here, attention is focused on the high-temperature properties of the theory. The equations to be analyzed are

$$m = \frac{\langle S^z \rangle}{S} = 1 - \frac{1}{NS} \sum_k n_k \tag{A1}$$

and

$$C = \frac{1}{N} \sum_k (1 - \gamma_k) n_k \tag{A2}$$

in which the spin-wave dispersion

$$\epsilon_k = e_0(1 - C/S)(1 - \gamma_k) + \xi m + h \tag{A3}$$

and $e_0 = 2rJS$, the exchange interactions being limited to the first shell of neighbours. For high temperatures, $T \gg J$, it is reasonable to approximate the Bose distribution factor by $n_k = T/\epsilon_k$. In this instance, the coupled equations can be expressed in terms of the extended Watson integral

$$I(\beta) = \frac{1}{N} \sum_k \frac{1}{1 + \beta - \gamma_k}. \tag{A4}$$

The series expansion for a FCC lattice is

$$I(\beta) = I(0) - \frac{3\sqrt{3}}{2\pi} \beta^{1/2} + \ldots \qquad \beta < 1 \tag{A5}$$

where $I(0) \simeq 1.3447$. Equations (A1) and (A2) reduce in the high-temperature limit to

$$m = 1 - \frac{T}{eS} I(\beta) \tag{A6}$$

and

$$C = \frac{T}{e}[1 - \beta I(\beta)]. \tag{A7}$$

Here, $\beta = (\xi m + h)/e$ with $e = e_0(1 - C/S)$.

To begin with, let us set aside the contribution to $\epsilon_k(T)$ from the dipolar forces and the applied magnetic field h. With $\xi = h = 0$, $x = e/e_0$ satisfies the equation

$$x(1-x) = (T/e_0 S) \tag{A8}$$

which admits real solutions for $\tau \equiv T/e_0 S \leqslant 1/4$. At the maximum temperature we find $e/e_0 = 1/2$ and

$$m = 1 - \tfrac{1}{2}I(0) = 0.33 \qquad \text{FCC}. \tag{A9}$$

These values are in tolerable agreement with the values obtained from the full numerical solutions reported in the main text; this finding gives confidence in our approximate treatment of the transcendental equations.

Turning next to the influence of dipolar forces on the magnetization and renormalization, still in the absence of any applied magnetic field, we will exploit the fact that the strength of the dipolar forces is weak compared to the exchange forces, i.e. $\xi \ll e_0$. Consider, for example, EuO. The value $J = 0.74\,\mathrm{K}$ gives a critical temperature for the spherical model that agrees with the measured value, and $\xi/e_0 = 0.016$. Working to leading order in ξ/e_0, we find that the coupled equations admit real solutions for the magnetization up to a temperature

$$T_0 \simeq \left(\frac{e_0 S}{4}\right)\left(1 + 2\frac{I(0)\xi m}{e_0}\right) \tag{A10}$$

at which the renormalization parameter is

$$e \simeq \left(\frac{e_0}{2}\right)\left(1 - \frac{I(0)\xi m}{e_0}\right) \tag{A11}$$

and the corresponding value of the magnetization is

$$m = 1 - \frac{1}{2}\left(I(0) - 0.8270\sqrt{\frac{[2 - I(0)]\xi}{e_0}}\right). \tag{A12}$$

The dipolar force is seen to increase the critical temperature and magnetization, and decrease the renormalization function. Evaluated for the simplified model of EuO, with $\xi/e_0 = 0.016$, the dipolar forces increase the critical temperature by 1.4% and the corresponding magnetization by 12.8%.

For the particular case of an applied magnetic field the results (A10) and (A11) apply, with m replaced by unity and $\xi = h$, where h is the Zeeman energy. The corresponding magnetization at the maximum temperature and $h/e_0 \ll 1$ is

$$m = 1 - \tfrac{1}{2}\left[I(0) - 1.1695(h/e_0)^{1/2}\right]. \tag{A13}$$

The same simplified method of analysis can be applied to the static susceptibility; from (22) and the definition of A and B we get

$$\frac{e_0\chi(0)}{S} = f\left\{-I'(\beta) + f\left[I^2(\beta) + I'(\beta)\right]\right\}\left[1 - f\left(1 - \frac{\mathrm{d}}{\mathrm{d}\beta}\left[\beta^2 I(\beta)\right]\right)\right]^{-1} \qquad \text{(A14)}$$

in which $f = \tau/x^2$ and the reduced temperature, τ, and spin-wave renormalization, x, are related by

$$x(1-x) = \tau[1 - \beta I(\beta)]. \qquad \text{(A15)}$$

Some features of this results are mentioned in the main text. Here we note that the susceptibility increases with temperature (see figure 3) and diverges at the maximum temperature.

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