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LETTER TO THE EDITOR

Collective excitations in paramagnets: confrontation of theoretical and experimental results for gadolinium

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Abstract. Recent high-resolution data for the spin response function $S(\boldsymbol{k}, \omega)$ of gadolinium at $T \geqslant 1.5\, T_c$, obtained by neutron scattering, is compared with results derived from the coupled-mode theory of a Heisenberg exchange interaction on an HCP lattice. A well-defined collective mode is obtained in a calculation which uses the long-range interaction derived from an analysis of the low temperature spin-wave dispersions. However, the peak position is at a much lower energy than the corresponding structure in the data, even though the estimated T_c agrees very well with the experimental value. Good overall agreement between theoretical and experimental results is found by using a nearest neighbour exchange deduced from T_c and the measured second-frequency moment.

The spin dynamics of paramagnets continues to be a topic of considerable interest. A recent spate of work has been fuelled by experimental data, obtained by neutron beam techniques, and computer simulations. The latter convincingly demonstrate that the time-dependent behaviour of classical spin chains at infinite temperature has a rich structure (Gerling and Landau 1990). The decay of the energy and spin autocorrelation functions is oscillatory for short times and at long times is consistent with classical diffusion, although the approach to the asymptotic behaviour is very slow. Depending on the degree of spin relaxation, the oscillatory behaviour can translate in the time and space Fourier transform of the spin response function $S(\boldsymbol{k}, \omega)$ to a peak centred at a finite frequency, and this is argued to occur. However, the case for collective excitations in a classical spin chain at infinite temperature is, at best, marginal.

Set against this, neutron scattering data for $S(\boldsymbol{k}, \omega)$ in paramagnetic ($T > T_c$) gadolinium show peaks, albeit heavily damped, when displayed as a function of ω for fixed k near the zone boundary (Cable and Nicklow 1989). Experimental data for some other magnets (insulating and metallic materials) hints at this effect but thus far the data for gadolinium is the best evidence (Lovesey *et al* 1990).

Speculations as to the occurrence of collective excitations in paramagnets have been made on the basis of a variety of calculations. For example, one is based on the behaviour of frequency moments of $S(\boldsymbol{k}, \omega)$ using a Heisenberg exchange model. These moments can be evaluated exactly at infinite temperature, and the results for the second and fourth moments are consistent with a peak in $S(\boldsymbol{k}, \omega)$ at finite ω, for a (fixed) large wavevector (Lovesey 1987). Another type of calculation, based on a generalized

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Langevin equation, produces quite similar predictions (Young and Shastry 1982). Very much more sophisticated calculations of $S(\boldsymbol{k}, \omega)$ with a coupled-mode theory have been performed for realistic models of EuO, EuS, Fe and Pd_2 Mn Sn (Cuccoli *et al* 1989 and 1990) and compared with available experimental data. It was concluded from these exercises that the lattice structure and nature of the exchange interactions play important roles in determining $S(\boldsymbol{k}, \omega)$. In all cases the exchange interactions were derived from the analysis of spin-wave dispersions measured in the low-temperature (ferromagnetic) phase, and the static correlations (susceptibility) derived self-consistently. In consequence, all quantities entering the coupled-mode theory were determined from independent results. The work on the interpretation of critical and paramagnetic fluctuations in these four magnets forms a body of evidence to the effect that coupled-mode theory for $S(\boldsymbol{k}, \omega)$ is to be trusted. However, it is to be remembered that the data do not display definite signatures of a collective excitation, and hence coupled-mode theory is not tested on this issue.

Coupled-mode theory, of the type used in the work described, is known to fail miserably for (one-dimensional) spin chains at temperatures less than the exchange interaction because it does not reproduce the long-lived collective excitation (Lovesey and Megann 1986). In view of this, there is considerable value in testing coupled-mode theory against the data for gadolinium.

The calculations reported here for paramagnetic gadolinium are based on the coupled-mode theory described by Cuccoli *et al* (1989). For the task in hand it has been generalized to the HCP lattice. This is non-trivial because the lattice is non-Bravais and the magnetic atoms are not at centres of inversion symmetry. A full account of the attendant complications and their resolutions will appear, together with a description of critical phenomena.

The theoretical model involves the solution of a (2×2 matrix) generalized Langevin equation for the spin relaxation function $F(\boldsymbol{k}, t)$, namely

$$\partial_t F(\boldsymbol{k}, t) = -\int_0^t \mathrm{d}t' K(\boldsymbol{k}, t - t') F(\boldsymbol{k}, t'). \tag{1}$$

The memory function $K(\boldsymbol{k}, t)$ contains the non-linear spin dynamics. Within the coupled-mode approximation, based on a Heisenberg exchange Hamiltonian, it is the joint function of the product of two relaxation functions, i.e. (1) reduces to an integro-differential equation for $F(\boldsymbol{k}, t)$. It can be shown that the static properties of the approximate equation are consistent with the spherical model of static correlation functions. The time Fourier transform, $F(\boldsymbol{k}, \omega)$, of the relaxation function is an even function of ω, by definition, and related to the neutron scattering response function through

$$S(\boldsymbol{k}, \omega) = \omega(1 - \mathrm{e}^{-\omega/T})^{-1} \times \chi(\boldsymbol{k}) F(\boldsymbol{k}, \omega). \tag{2}$$

In this expression, $\chi(\boldsymbol{k})$ is the isothermal susceptibility, T the absolute temperature ($k_B = 1$), and the definition of $F(\boldsymbol{k}, \omega)$ is such that

$$\int_{-\infty}^{\infty} \mathrm{d}\omega\, F(\boldsymbol{k}, \omega) = 1. \tag{3}$$

Later we discuss the value of the normalized f-sum rule

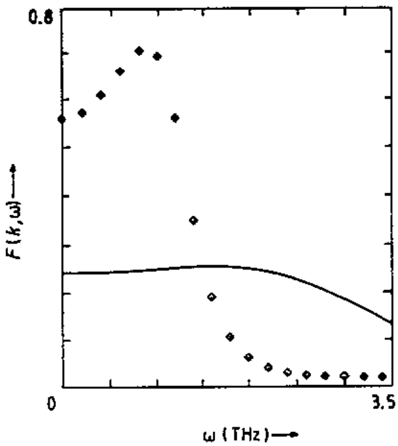

Figure 1. Experimental data for $F(\boldsymbol{k}, \omega)$ at $\boldsymbol{k} = 4\pi(\frac{1}{2}, \frac{1}{2}, 0)/a$ ($\xi_x = \frac{1}{2}$) and $T = 1.5\, T_c$ is displayed together with the corresponding theoretical results obtained using exchange interactions derived from low temperature spin-wave dispersions. Here, and in figure 3, the area enclosed by the curves = 1, i.e. a factor two larger than implied by (3). ———, experiment; ◈, theory.

$$\omega_k^2 = 2 \int_{-\infty}^{\infty} \mathrm{d}\omega \, \{S(\boldsymbol{k}, \omega)/\chi(\boldsymbol{k})\}\omega = \int_{-\infty}^{\infty} \mathrm{d}\omega \, F(\boldsymbol{k}, \omega)\omega^2. \tag{4}$$

The exact value of ω_k^2 for a Heisenberg model with $T \to \infty$ is,

$$\omega_k^2 = \tfrac{8}{3} S(S+1) \, \mathrm{Re} \sum_{l} J^2(\boldsymbol{l})(1 - \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{l}}). \tag{5}$$

Here, S is the magnitude of the effective spin ($2S = 7.63$ for Gd) and $J(\boldsymbol{l})$ is the exchange interaction for spins separated by the lattice vector $\boldsymbol{l}$; the result (5) is appropriate for spins on a non-Bravais lattice.

As a first step toward the interpretation of the experimental data, with the suitably generalized theory, we used the path chosen in previous studies and employed exchange interactions determined from spin-wave dispersion (Lindgård 1978). These produce a tolerable value for the critical temperature and $F(\boldsymbol{k}, \omega)$ contains a significant peak from a collective excitation. However, the position of the peak, and the overall width of $F(\boldsymbol{k}, \omega)$, is much too small by a factor of 3–4. This finding is illustrated in figure 1 and it is consistent with the observation by Cable and Nicklow (1989) that the second frequency moment, evaluated with these exchange interactions, is much smaller than implied by the experimental data.

A plausible explanation for the failure of this scheme for implementing the coupled-mode theory is that the Fermi surface of Gd changes with temperature, so exchange interactions at $T \ll T_c = 293$ K do not keep the same intrinsic values when the temperature is raised to $T > 1.5\, T_c$.

Recognizing that conciliation between theoretical and experimental results hinges on the choice of exchange interactions, we take note of the fact that the experimentally determined second frequency moment is almost independent of temperature. This suggests using the known expression for this moment at infinite temperature, (5), to analyse the data, and thereby determine the exchange, a scheme not unlike the first step using spin-wave dispersions. It is found that a single exchange constant $J = 0.534$ meV provides a tolerable description of both the second moment and critical temperature

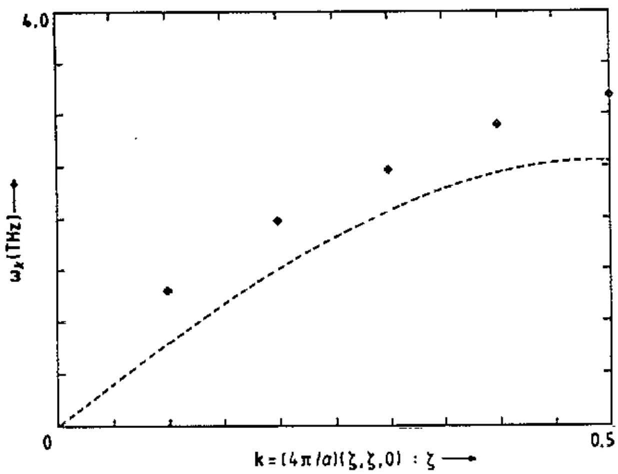

Figure 2. Experimental data for ω_k, defined in (4), at $T = 2T_c$ and $k = 4\pi(\zeta, \zeta, 0)/a$ is shown for $0 \leqslant \zeta \leqslant 0.5$. Theoretical results are derived from (5) using a nearest neighbour exchange $J = 0.534$ meV (1 meV = 0.24 THz), and $S = 7.63/2$.

(300 K), derived from the spherical model. Regarding the critical temperature, the molecular field expression gives the result 455 K and this and the spherical model result (300 K) vary linearly with J. The experimentally determined value of ω_k at $T = 2T_c$ is shown in figure 2 together with the result from (5) using a single exchange interaction ($k = 4\pi(\zeta, \zeta, 0)/a$). While the experimental ($T = 2T_c$) and model ($T = \infty$) results can be made to agree more closely by increasing J this action increases T_c determined from the spherical model. The value $J = 0.534$ meV is a choice which gives reasonable agreement with the chosen input data, with an emphasis on obtaining good agreement with the experimentally determined critical temperatures, 293 K.

Results for $F(k, \omega)$ using this value for the exchange are displayed in figure 3. The agreement between theoretical and experimental results for various temperatures and wavevectors is tolerable. In particular, features attributed to a collective excitation are adequately described. On viewing the data sets it must be kept in mind that all quantities in the model are held fixed, so one sees a one-on-one confrontation of theory and data.

The theoretical results presented in figure 1 demonstrate for the first time that coupled-mode theory applied to a three-dimensional Heisenberg model can display a distinct collective mode peak. However, with respect to gadolinium, its shape and position are at odds with the available experimental data. We interpret this as evidence that exchange interactions change with temperature and the long-range interactions determined from the analysis of low temperature spin-wave dispersions are not appropriate for $T \geqslant T_c$. Here we have chosen the very simple procedure for $T > T_c$ of using a single, nearest neighbour, exchange interaction which is consistent with the observed critical temperature and second frequency moment. Data for the latter quantity show that it is largely independent of temperature ($1.5\,T_c \leqslant T \leqslant 2.9\,T_c$), so a fit to the exact expression available for $T \to \infty$ is a quite reasonable procedure. Looking at figure 2, even closer agreement between results from (5) and experimental data might be achieved, while maintaining the result $T_c = 300$ K, by using a longer-range interaction. If the idea is pursued then the fit should be made to ω_k at a finite temperature, available from the spherical model correlation functions. We have not done this because the results in figure 2 are seen to be consistent with the general tenor of the interpretation displayed in figure 3.

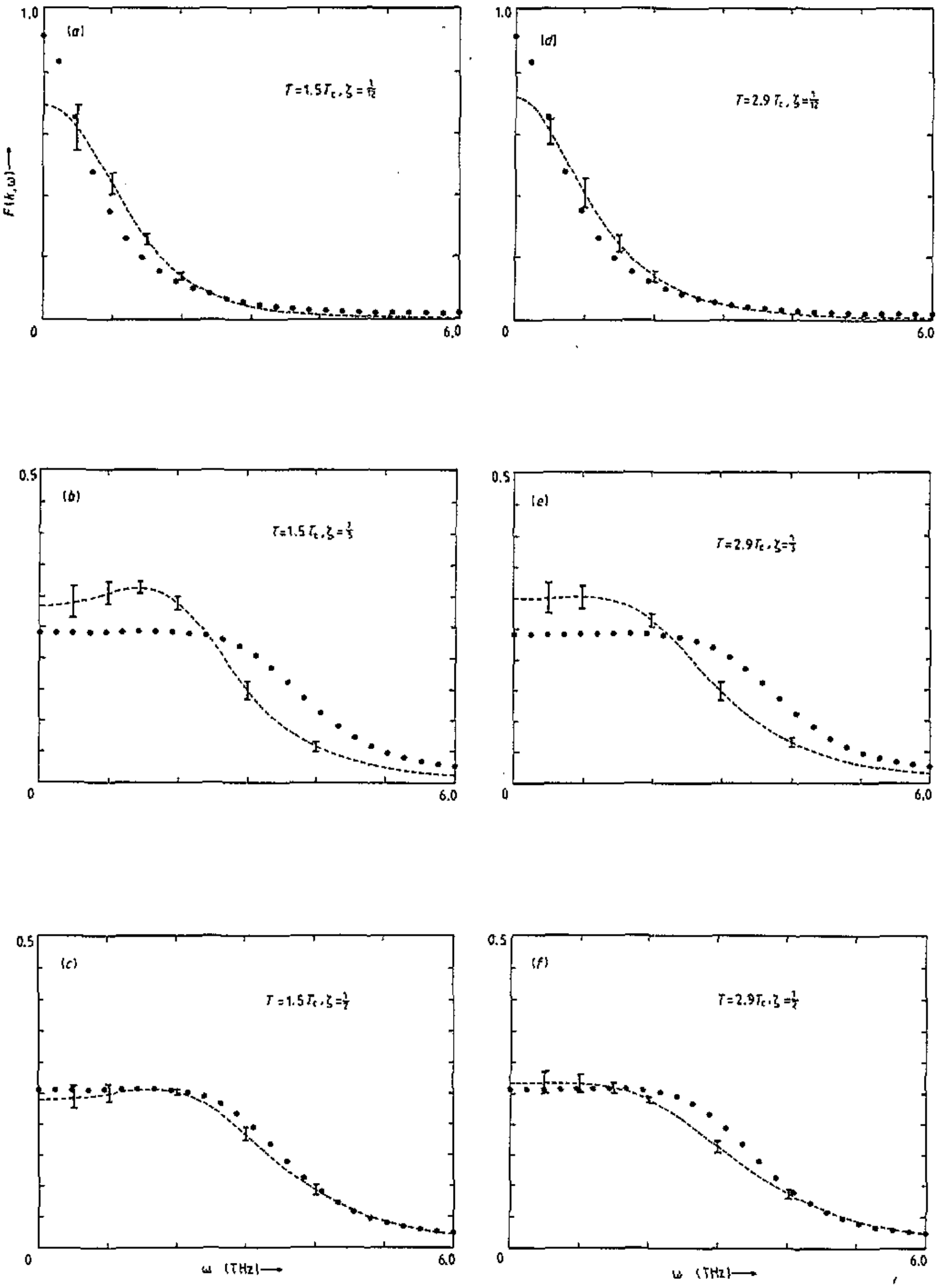

Figure 3. Experimental and theoretical results for $F(\boldsymbol{k}, \omega)$ are displayed for $T = 1.5\ T_c$ and $2.9\ T_c$ with $\boldsymbol{k} = 4\pi(\zeta, \zeta, 0)/a$ and $\zeta = \frac{1}{12}, \frac{1}{3}$ and $\frac{1}{2}$. In calculating $F(\boldsymbol{k}, \omega)$ the input data is the lattice type (HCP), $J = 0.543$ meV, $T_c = 300$ K and $S = 7.63/2$. The experimental results are generated from a damped harmonic oscillator form for $F(\boldsymbol{k}, \omega)$ with parameters provided by Cable and Nicklow (1989). The indicated spread in the data is a conservative estimate provided by Dr Cable (1991, private communication). – – –, experiment; ●●●, theory.

For $F(\boldsymbol{k}, \omega)$ the agreement between theory and experimental results is particularly good for extreme wavevectors, near the zero centre and boundary (along $(1, 1, 0)$). The relatively poor agreement at an intermediate wavevector is found at all temperatures, with the theory consistently underestimating the sharpness of the collective mode peak. However, this finding must be taken in context with the spread in the experimental data, as conservatively indicated in the figures; for the smallest wavevector ($\zeta = 1/12$) there is considerable uncertainty in the data below 0.4 THz, and so much so that a comparison between the theory and experimental data is not really meaningful.

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