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CORRELATION THEORY APPLIED TO THE STATIC AND DYNAMIC PROPERTIES

OF EuO and EuS

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Correlation theory applied to the static and dynamic properties of EuO and EuS

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ABSTRACT

The paramagnetic scattering was recently measured for EuO¹⁾. It was found that spin-wave-like excitations develop for wave vectors approaching the zone boundary. The spectrum was found to be well described by damped harmonic oscillators (also called the two-pole-approximation). This approximation was used previously in the correlation theory²⁾ primarily to calculate static properties. Selfconsistent dynamic and static calculations have here been performed for EuO, which is an ideal Heisenberg magnet with significant second nearest neighbour interaction ($J_2 = J_1/5$). The two-pole approximation describes accurately the correlation range, the static susceptibility and the qualitative behaviour of the dynamic properties (i.e. the wave vector at which peaks appear in the spectrum as a function of temperature). However, in order to also obtain the correct frequency scale it is necessary to use a cut-off of the spectrum at high frequencies, which cannot be seen experimentally, but which significantly influences the frequency moments. It was found that the finite J_2 has significant importance for a comparison between theory and experiment. It is concluded that the calculation for a simple cubic n.n. magnet by Hubbard³⁾ does not describe the EuO data accurately, neither with respect to lineshape nor frequency scale. Significant differences are to be expected between EuO and EuS having opposite sign for J_2 .

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INTRODUCTION

In order to describe accurately static and dynamic properties of a magnetic system a generalization of the random phase approximation (RPA) has been developed²⁾ in which essential correlation effects are included self-consistently. In this correlation theory (CT) an attempt is made to provide a simple scheme to calculate the dynamic response in an approximate, yet realistic way (i.e. yielding renormalization and damping of excitations) in terms of static properties like correlation functions, to be calculated selfconsistently by exact relations using the dynamics. EuO and EuS represent ideal realizations of the 3-d Heisenberg model (apart from a weak dipolar anisotropy, which may in fact also be treated by CT). The structure is fcc, $S = 7/2$ and the nn and nnn exchange constants J_1 and J_2 are known from spin wave measurements^{1,4)}: for EuO $J_1 = 0.625$ K, $J_2 = +J_1/5$ and for EuS $J_1 = 0.253$ K, $J_2 = -J_1/5$; thus the nnn interaction is significant and having opposite sign. Extensive measurements of dynamic^{1,4)} and static⁴⁾ properties for $T \geq T_c$ have been performed in particular for EuO. In the present paper the range of validity of CT is tested versus these experimental results.

CORRELATION THEORY FOR THE HEISENBERG MODEL ($T \geq T_c$)

A brief summary of the approximation used in CT:

By the Mori projection operator formalism the dynamical response $(S_q^z S_{-q}^z)_\omega$ is related to the static susceptibility χ_q and frequency moments $\langle \omega_q^n \rangle$, which are expressed as wave vector sums over exchange and static correlation function e.g. $\chi_q \langle \omega_q^2 \rangle = 4 \sum_k (J_k - J_{k-q}) \langle S_k^z S_{-k}^z \rangle$. For small frequencies $\omega \lesssim 2S(J_0 - J_q)$ the two-pole-approximation is expected to describe the line shape well:

$$(S_q^z S_{-q}^z)_\omega = \frac{\chi_q}{\pi\omega} \langle \omega_q^2 \rangle \text{Im} \left\{ \frac{1}{(\omega - \alpha_q - i\beta_q)(\omega + \alpha_q - i\beta_q)} \right\} \quad (1)$$

This corresponds to assuming a Lorentzian line shape for the random force response function $(\dot{S}_q^z \dot{S}_{-q}^z)_\omega$ for small ω , with the halfwidth $2\beta_q$. $(\dot{S}_q^z \dot{S}_{-q}^z)_\omega$ is approximately evaluated in terms of products of $(S_k^z S_{-k}^z)_\omega$ using the mode-mode-decoupling (MMD) and β_q is selfconsistently obtained at the halfwidth frequency ω_{HW} of $(S_q^z S_{-q}^z)_\omega$. (Approximations amounting to those in the MMD were also made by Hubbard³⁾, who, did not, however, use (1) and did not attempt to selfconsistently calculate the static properties). Eq. (1) yields χ_q and $\langle \omega_q^2 \rangle = \alpha_q^2 + \beta_q^2$ selfconsistently, which suffices for a quite accurate description of the static correlation function, using $\langle S_q^z S_{-q}^z \rangle = \int (S_q^z S_{-q}^z)_\omega / (1 - e^{\beta\omega}) d\omega$. The dynamical properties obtained are qualitatively correct, i.e. a) diffusion for $q \rightarrow 0$ ($\alpha_q + i\beta_q$ for $T > T_c$, b) dynamical scaling $\omega_{HW} \sim q^{5/2}$ at $T = T_c$, and c) development of spin wave like peaks at high q ($\alpha_q > \beta_q$). It is, however, clear that (1) is not valid at high frequencies and must be cut off in order

to give finite higher moments $\langle \omega_q^n \rangle$, $n > 2$. If a cut-off determined by $\langle \omega_q^4 \rangle$ is introduced, the linewidth ω_{HW} increases in particular for q near the zone boundary. (In evaluating $\langle \omega_q^4 \rangle$ the four spin correlation functions involving different sites were decoupled in products of pair correlation functions, analogous to the MMD; those involving only two sites were directly calculated). Writing $S_q = C + \sum_n a_n (J_0 - J_q)^n$, where C and a_n are temperature dependent constants, one finds that $C \rightarrow 0$ for $T \rightarrow T_c$. This means that both $(S_q^z S_q^z)_\omega$ and $(\hat{S}_q^z \hat{S}_q^z)_\omega$ exhibit critical slowing down, and therefore the two-pole approximation is not satisfactory very close to T_c .

APPLICATION OF THE CORRELATION THEORY TO EuO AND EuS

The following figures show a comparison between experiments and the results of CT using no adjustable parameters. Comments are given in the figure texts. The calculated transition temperatures are given in table 1. The contribution from the dipole forces may account for the difference of a few K between $T_c(CT)$ and $T_c(exp)$.

Table 1	$T_c(\text{mean field})$	$T_c(CT)$	$T_c(exp)^{4)}$	$T_c(CT)/T_c(MF)$
EuO	86.6 K	66.6 K	69.1 K	0.768
EuS	25.8 K	17.1 K	16.6 K	0.662

CONCLUSION

It is found that the correlation theory with no adjustable parameters accounts for the measured static and dynamic properties of EuO for $1.02 < T/T_c < \infty$. The effect of the next nearest neighbour interaction, J_2 , is important and the calculated spectrum of EuS is therefore significantly different from that of EuO.

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REFERENCES

1. H.A. Mook. Phys. Rev. Lett. 46, 508 (1981).
2. P.-A. Lindgård and D. Yang. J. Magn. and Magn. Mat. 15-18, 1037 (1980), and P.-A. Lindgård. Proc. Int. Conf. on 4-f Systems. Philadelphia Nov. 1979 (Plenum Press), p 153 (1980).
3. J. Hubbard. J. Phys. C. 4, 53 (1971).
4. J. Als-Nielsen, O.W. Dietrich and L. Passell, I-III, Phys. Rev. B. 14, 4897-4945.
5. N. Menyuk, K. Dwigth, and J.B. Reed. Phys. Rev. B 3, 1689 (1971), see also J.T. Ho Phys. Rev. B 22, 467 (1980).

FIGURE CAPTIONS

Fig. 1. The calculated inverse correlation range times the nearest neighbour distance, $\kappa_1 a_{nn}$, agrees with no adjustable parameters with experiment⁴⁾ for $0.02 < (T - T_c)/T_c$. For EuS $\kappa_1 a_{nn}$ (not shown) is 15% smaller, caused by the larger depression (table 1) of T_c when $J_2 < 0$; the difference is small and in agreement with the experimental trend⁴⁾. The susceptibility fits similarly for $0.02 < (T - T_c)/T$ with the neutron data⁴⁾ (scaled). The static data⁵⁾ (which yield a lower γ -exponent than the neutron data) give a too small susceptibility for all $T > T_c$. $\chi(CT)$ reduces to the mean field $\chi(MF)$ for $(T - T_c)/T \rightarrow 1$.

Fig. 2. Calculated and experimental¹⁾ line shape and line width for EuO. The agreement is good except at high q near T_c , where the cut-off two-pole line shape is not adequate. The calculation by Hubbard³⁾ for a simple cubic structure (here using $J = J_1 + J_2$, which gives the same spin wave energy at the (111) zone boundary) is not relevant for the fcc EuO with $J_2 = J_1/5$. The spin wave energy at $T=0$ is indicated by an $\uparrow$.

Fig. 3. The influence of J_2 is evident when comparing the calculation of ω_{HW} , $\sqrt{\omega_q^2}$ and ω_{peak} for EuO ($J_2 = J_1/5$) and EuS ($J_2 = -2J_1/5$). In EuO a peak (at ω_{peak}) develops at $q > 0.5$ q_{zone} for $T > T_c$, whereas in EuS this only happens closer to T_c and in a limited q -range. As a function of temperature $\sqrt{\omega_q^2}$ rises rapidly near T_c to a value $= 2\sqrt{\omega_q^2}_{T=\infty}$ for $q = q_{zone}$ in EuO.

Fig. 4. For small q and $T > T_c$ the line shape is Lorentzian and the line width $\omega_{HW} = \Lambda(T)q^2$ shows spin diffusional behaviour. $\Lambda(T) \rightarrow 0$ for $T \rightarrow T_c$ due to critical slowing down. At any T one finds that $\omega_{HW} \ll 2\theta_q$.

Fig. 5. Line width at $T = T_c$. Dynamical scaling, $\omega_{HW} \sim q^{5/2}$, is found to hold for $q < 0.1$ q_{zone} . The experimental data by Dietrich *et al*⁴⁾ seem not consistent with those of Mook¹⁾ at higher q -values. This may be due to the Lorentzian line shape used in the analyses in ref. 4.

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