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Relaxation and Renormalization of Spin Waves in EuO*

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Abstract

The divalent europium chalcogenides include the only known examples of isotropic Heisenberg ferromagnets. As such they offer unique opportunities for comparing the existing highly developed theory of simple Heisenberg ferromagnets with experiments. We have used neutrons to study the dynamic behavior of EuO at temperatures extending up to the Curie temperature T_c and over a fairly wide range of wave vectors. Since the exact form of the scattering cross section is not known, only limited comparisons with the existing theory were possible. Dynamic scaling predictions of spin wave renormalization appear to be qualitatively correct, but the observed line width variations with wave vector and temperature are in only fair agreement with "hydrodynamic" predictions. As was the case with isotropic metallic ferromagnets, the EuO line shapes show no indication of a diffusive mode near T_c .

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1. INTRODUCTION

The divalent europium chalcogenides EuO and EuS are the only known materials which are good representations of simple, isotropic Heisenberg ferromagnets. Since they occupy a unique position in the hierarchy of ferromagnets, their characteristics are of particular interest and have been studied by most if not all of the traditional methods used for magnetic investigations[1-7]. From such studies, a relatively complete picture of the macroscopic thermodynamic properties of these compounds has emerged, but the amount of information available concerning their microscopic properties is very limited indeed. This is because neutron scattering, normally the primary source of such information, is difficult to apply in materials containing europium which is a strong neutron absorber [$\sigma_{n,\gamma}(\text{Eu}) \approx 4000 \text{ bn}$].

In part, the drastic loss of intensity due to europium absorption can be reduced by employing materials prepared with separated Eu^{153} which has the smaller absorption cross section of the two naturally occurring isotopes [$\sigma_{n,\gamma}(\text{Eu}^{153}) \approx 400 \text{ bn}$]. A further improvement in intensity can be achieved by working with thin slab samples (0.5mm thick) so as to use the available beam most efficiently. Practically, this means that all measurements must be made with powder samples since single crystal plates of the required size are beyond the present technology. Experimental flexibility is restricted to some extent by the use of powders, as will be explained, but otherwise it is not a serious limitation.

With this technique, we have found it possible to investigate both the static and dynamic magnetic characteristics of europium compounds. Earlier studies we have made of the static critical behavior of EuO and EuS near the Curie temperature T_c and of their spin-wave excitations are discussed in previous publications [8,9]. In this paper, we wish to describe our observations of the relaxation and renormalization of spin waves in EuO at temperatures approaching T_c . Due to space limitations, we will consider here only those of our measurements made below T_c . Data taken at and above T_c will be reviewed at a later time.

Our measurements show that both the renormalization of spin-wave energies in EuO and the temperature dependence of the line widths are in reasonable accord with theory; agreement between the observed wave vector dependence of the line widths and theory is perhaps less satisfactory. All of these comparisons are, however, complicated by difficulties of interpretation which will be discussed. Near T_c , the excitations have a spectral form similar to that observed in iron [10] and nickel [11]; in particular, there is no evidence of a central component corresponding to a longitudinal diffusive mode.

2. GENERAL CONSIDERATIONS

2.1. Small Angle Measurements with Powders

EuO was chosen for spin-wave studies near T_c in preference to EuS for several reasons. First, the relatively large Curie temperature (69.2°K) offered assurance that the spin-wave energies would be large enough to be resolved by conventional triple-axis spectrometry over an extended temperature range. Second, from studies at low temperatures [9] we had already established that spin-wave dispersion in EuO remains

essentially isotropic almost to the Brillouin zone boundary. This unusual isotropy, which also is a property of iron [10] and nickel [11], is, in EuO, due to a combination of factors. First, the $^8S_{7/2}$ ground state of the Eu^{++} ion has a spherically symmetric spin distribution. Further, EuO is unique among the divalent chalcogenides in that both the nearest and next nearest neighbor exchange constants are of the same sign. In addition the europium ions form an fcc lattice with 12 nearest neighbors which is spatially very isotropic. As a result of this combination of characteristics the magnetic environment of an Eu^{++} ion in EuO is isotropic to a remarkable degree.

A brief consideration of neutron scattering in powders will show why the isotropy of spin-wave dispersion in EuO is essential for line width measurements. When neutrons of wave vector $\vec{k}_i$ are scattered to wave vector $\vec{k}_f$, energy and momentum conservation require that

$$\hbar\omega = (\hbar^2/2m)(k_i^2 - k_f^2)$$

$$\vec{Q} = \vec{k}_i - \vec{k}_f = \vec{q} + \vec{\tau}.$$

Only the reciprocal lattice vector at the origin, $\vec{\tau} = (000)$, is well defined in a powder; all other $\vec{\tau}$'s are distributed uniformly over spheres surrounding the origin. If, as in EuO, the scattering cross section depends only on the modulus of $\vec{q}$, the small angle scattering from a powder will be identical to that observed in a single crystal at the same $\vec{q}$, provided the observations are restricted to values of $\vec{k}_i - \vec{k}_f$ well within the first Brillouin zone. More commonly, however, the cross section depends on the direction of $\vec{q}$ as well as its magnitude. In this case, even well within the first Brillouin zone the scattering from a powder will show a finite energy width simply because of the random orientation of individual crystallites. For example, in EuS this "powder broadening" is large enough to almost completely obscure the effects of relaxation processes[9]. Fortunately, in EuO "powder broadening" of the lines is small and broadening due to relaxation processes can clearly be resolved.

Aside from the limitations outlined above, which affect line width measurements on powders, small angle scattering itself also introduces certain experimental complications which should be mentioned. First, the range of energy transfers accessible with a given value of $|\vec{q}|$ is much more restricted. This is not a problem in EuO where the spin-wave energies are small, but it can become important in materials in which the excitation energies are larger. Second, at small scattering angles (in practice less than about 2°) background variation across the scans is sufficient to introduce unwanted distortions in the observed line shapes. Thus the accessible $|\vec{q}|$ range in powders is limited on the one hand by the requirement that $\vec{k}_i - \vec{k}_f$ be well within the first Brillouin zone and on the other by the requirement that the scattering angles be large enough to avoid background distortion of the line shapes.

2.2. Experimental Methods

All of our measurements were made in the constant- q mode with a triple-axis spectrometer using pyrolytic graphite monochromating and analyzing crystals. The incident neutron energy employed was 4.8 meV. Pyrolytic graphite and polycrystalline Be filters were used to insure against higher order contamination of the beam.

Horizontal and vertical divergences of the collimators between reactor and detector were 0.33° - 0.35° - 0.68° - 0.66° and 0.9° - 1.9° - 1.4° - 1.4° respectively. The energy resolution was approximately 0.10 meV.

3. EXPERIMENTAL RESULTS

Figure 1 shows the spin-wave line shapes observed at $|\vec{q}| = 0.20\text{\AA}^{-1}$ in the temperature range $0.15 \leq (T_c - T)/T_c \leq 0.005$. Both renormalization of the spin-wave energies and line broadening are evident.

In Figure 2 we have shown how spin-wave line shapes observed at a fixed temperature $(T_c - T)/T_c = 0.05$ vary with $|\vec{q}|$. Dispersion effects are apparent and there is also significant q -dependent line broadening. Figures 1 and 2 represent only part of the data collected but the behavior is illustrative of all of our results. It should be noted that the statistical uncertainty of the individual data points is not the square root of the plotted intensity. The data have been normalized to a common counting time shorter than that actually used.

Casual inspection of the observed line shapes is sufficient to establish that there are limits on the extent to which the characteristics of the actual excitations can be determined from the data. The spin-wave peaks merge and broaden as T approaches T_c and the spin-wave energies obviously become less and less well defined. Further, although the line widths of the spin waves decrease more rapidly than their energies, we cannot resolve peaks much narrower than our instrumental energy resolution which is 0.10 meV. Thus our line width measurements were limited for instrumental reasons to values of $q \gtrsim 0.12\text{\AA}^{-1}$ and to temperatures greater than $\sim 60^\circ\text{K}$.

The line shapes of Figures 1 and 2 show no evidence of a three-peaked structure such as was observed in both the isotropic Heisenberg antiferromagnet RbMnF_3 [12] and in the anisotropic Heisenberg ferromagnet MnF_2 [13]. As was the case in iron [10] and nickel [11], while transverse propagating modes are clearly visible, nothing identifiable as a longitudinal mode appears even near T_c .

Although our energy resolution was about the same as that used for the iron and nickel measurements, our observations were limited to somewhat larger values of the wave vector q . This is simply because in both iron and nickel the energy of a spin wave with a given value of q is considerably larger than that of an equivalent mode in EuO . Thus energy resolution, not q resolution, represented the limiting factor in our measurements.

4. ANALYSIS OF THE DATA

4.1. Form of the Cross Section

The inelastic magnetic scattering cross section is customarily expressed in terms of the scattering function $S(\vec{q}, \omega)$ representing the Fourier transform of the spin pair correlation function. $S(\vec{q}, \omega)$ is defined as the product of three terms; a Boltzmann population factor, the static response function $\chi(\vec{q})$ describing the magnetic moment response to a hypothetical static applied field varying sinusoidally in space with wave vector $\vec{q}$, and the dynamic spectral shape function $F(\vec{q}, \omega)$ whose Fourier

transform describes the relaxation in time of the magnetic moment distribution when the field is switched off. Below T_c , in the ordered phase, it is necessary to distinguish between longitudinal and transverse components of both $\chi(\vec{q})$ and $F(\vec{q}, \omega)$. These correspond respectively to the applied field parallel and perpendicular to the direction of the spontaneous moments. In a powder sample [14]

$$S(\vec{q}, \omega) = \frac{\hbar\omega/k_B T}{1 - \exp(-\hbar\omega/k_B T)} \frac{k_B T}{(g\mu)^2} \left[\chi^l(\vec{q}) F^l(\vec{q}, \omega) + 2\chi^t(\vec{q}) F^t(\vec{q}, \omega) \right], \quad (1)$$

where the superscripts l and t refer to longitudinal and transverse components respectively. Here, our primary interest is in the spectral shape function $F^t(\vec{q}, \omega)$ which represents, in the ordered phase, transverse propagating modes or spin waves.

At low temperatures, the transverse magnetic excitations are long lived spin waves with well defined energies $\omega_0(\vec{q})$ i.e. $F^t(\vec{q}, \omega)$ is essentially a two-peaked function of the form $\delta[\omega \pm \omega(\vec{q})]$. At higher temperatures spin-wave interactions assume greater importance and the excitations become shorter lived and less well defined in energy. Nevertheless, in the hydrodynamic regime, both microscopic and macroscopic spin-wave theory [15] predict that the spin-wave damping rate is small compared to the real part of the frequency; hence $F^t(\vec{q}, \omega)$, in this regime, retains its characteristic two-peaked structure. Halperin and Hohenberg [15] have suggested for the spectral shape function in the hydrodynamic regime the form

$$F^t(\vec{q}, \omega) = \frac{1}{\pi} \left\{ \frac{\gamma \omega_0^2(\vec{q})}{[\omega^2 - \omega_0^2(\vec{q})]^2 + \gamma^2 \omega_0^2(\vec{q})} \right\}, \quad (2)$$

where γ is a temperature and q -dependent line width. Alternatively Riedel [17] has employed the double Lorentzian form

$$F^t(\vec{q}, \omega) = \frac{1}{2\pi} \left\{ \frac{\Gamma}{[\omega - \omega_0(\vec{q})]^2 + \Gamma^2} + \frac{\Gamma}{[\omega + \omega_0(\vec{q})]^2 + \Gamma^2} \right\} \quad (3)$$

to represent spin waves under the same conditions. A third approach is the often used damped harmonic oscillator expression

$$F^t(q, \omega) = \frac{1}{\pi} \left\{ \frac{\gamma \omega^2}{[\omega^2 - \omega_0^2(\vec{q})]^2 + \gamma^2 \omega^2} \right\}, \quad (4)$$

which leads to a two-peaked function when the line width $\gamma \ll \omega_0$. The two peaks broaden and merge to a single peak as γ approaches and exceeds ω_0 .

4.2. Comparison with the Experimental Observations

All three of the above expressions, when folded with the

instrumental resolution, satisfactorily represent the observed line shapes. The solid lines in Figures 1 and 2 are the best least square fits obtained with the Halperin-Hohenberg form, Eq. (2); expressions (3) and (4) give fits which are statistically equally good. Unfortunately, no selection between alternative forms of $F^L(\vec{q}, \omega)$ is possible on the basis of quality of fit alone. Furthermore, even near T_c there is no noticeable deterioration in the quality of fit in the region of zero energy transfer as would be expected if a narrow, unresolved central peak [a longitudinal diffusive contribution from $F^L(\vec{q}, \omega)$] was also present. $F^L(\vec{q}, \omega)$ alone, with parameters that vary smoothly with temperature and q , fully describes the observed lines.

In the hydrodynamic limit, the line widths will be very small compared to the energies of the excitations and the three forms of the spectral shape function will be essentially indistinguishable. But it is evident that our data are on the borderline of the hydrodynamic region or perhaps beyond. We are therefore faced with the problem of deciding which form of $F^L(\vec{q}, \omega)$ represents the most realistic extrapolation from the hydrodynamic regime to the regime of our observations. We have already established that there are no experimental grounds for selecting one form of $F^L(\vec{q}, \omega)$ in preference to the others. Since we are not aware of any theoretical basis for such a selection, we decided to make separate analyses of the data using all three forms. Figure 3 shows the temperature dependence of the spin-wave stiffness constant $D = \omega_0(q)/q^2$ derived by fitting alternatively expressions (2), (3), and (4) to the measurements. In Figures 4 and 5 the line widths for the data in Figures 1 and 2 are plotted as functions of temperature and wave vector q . To avoid confusion only the values obtained using the Halperin-Hohenberg form are shown. The widths derived using the other forms show systematic differences but the deviations are somewhat less pronounced than those of Figure 3.

5. COMPARISON WITH THEORY

5.1. The Stiffness Constant

Halperin and Hohenberg [15] have applied dynamic scaling arguments below T_c to predict that in the hydrodynamic regime the spin-wave stiffness constant D will vary as the $\nu' - \beta$ power of the reduced temperature difference $(T_c - T)/T_c$. Here ν' and β represent, respectively, the critical exponents describing the temperature dependence of the spin correlation range and the magnetization. If, in the spirit of the scaling hypothesis, we assume $\nu' = \nu$ i.e. assume that the exponents are symmetric about T_c [16], then, using the values in reference [8], we obtain $\nu' - \beta = 0.323 \pm 0.025$. This result is in excellent agreement with the value 0.329 obtained from Figure 3 using the Halperin-Hohenberg values for D . Alternatively, the double Lorentzian plot leads to a slightly larger value, 0.360, for the exponent. The damped harmonic oscillator plot, however, can hardly be considered to obey a power law at all! It is evident that the form chosen for $F^L(\vec{q}, \omega)$ has enough influence on the values derived for D to affect quantitative comparisons with the theory.

Similar tests of dynamic scaling have also been made with iron [10] and nickel [11]. In these materials spin-wave energies are considerably larger and the peaks relatively sharper. Presumably, ambiguities of the type we have encountered here would be less apparent, but the question was not examined in either case when the data were analyzed.

One final comment concerning dipolar interactions. In the dynamic scaling theory, dipolar contributions to the spin-wave energy are specifically excluded. These interactions become of increasing importance as q decreases and if they are neglected, the stiffness constant D will exhibit an apparent systematic increase with decreasing q . This effect can be clearly seen in our data at very small values of q , but for $q \gtrsim 0.18 \text{ \AA}^{-1}$ it is no longer evident. We have calculated for EuO the dipolar contributions to the spin-wave energies and find that they depend markedly on the direction of propagation of the wave (even changing sign). Consequently, in a powder, dipolar effects are considerably reduced because of the averaging over all orientations. A certain amount of "powder broadening" of the spin-wave lines will, however, occur from this cause at the smallest values of q .

5.2. The Line Widths

Both macroscopic [15] and microscopic [18] spin-wave theory indicate that the spin-wave line widths of Heisenberg ferromagnets will vary as q^4 . Also, according to Halperin and Hohenberg [15], dynamic scaling predicts that line widths in the hydrodynamic region will vary as the $-(3\nu'/2)$ power of the reduced temperature difference. Assuming as before that $\nu' = \nu$, this would imply an exponent of -1.04 ± 0.03 .

Figures 4 and 5 indicate that neither of the above power laws appear to be really good representations of the data. The observed variation with q seems to be somewhat less rapid than q^4 and the variation with temperature marginally smaller than $[(T_c - T)/T_c]^{-1.04}$.

This is hardly surprising. Both power laws are "hydrodynamic" predictions, while our line width measurements (judging from the ratio of widths to energies) appear to have been made outside the hydrodynamic regime. It is undoubtedly more realistic to interpret our results as evidence that line width variation outside the hydrodynamic regime is not very different from what is expected in the hydrodynamic limit.

6. DISCUSSION

As we have said, the most striking feature of the observed EuO line shapes is that they are two-peaked and show no evidence of a diffusive mode near T_c . On the basis of the limited evidence now available it would appear that this is characteristic of isotropic ferromagnets, regardless of whether they are metals or insulators.

We noted in section 4 that we do not find any excess intensity at small energy transfers as would be expected if a long-lived and therefore very narrow diffusive mode were present. At the other extreme, it is pertinent to ask whether there is any evidence to support the view that in these systems the diffusive modes are strongly coupled to other collective modes (possibly propagating modes) and therefore are very short lived. In this case the resulting broad diffusive peak would only manifest itself as a temperature dependent background. We saw no evidence of this, although the statistical accuracy of our background determinations was admittedly not such as to rule out the possibility completely.

In conclusion, we believe the data to be consistent with the predictions of the dynamic scaling theory for the temperature dependence of

the spin-wave stiffness constant; but, as we have said, the comparison necessarily assumes $v' = v$ and is, in addition, sensitive to the form of the spectral shape function $F^t(\vec{q}, \omega)$. Also our results indicate that spin-wave relaxation at the borderline of the hydrodynamic region, or possibly outside of it altogether, is very little different from what is expected in the hydrodynamic limit.

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FIGURE CAPTIONS

- Fig. 1.** Temperature dependence of the spin-wave line shapes observed at $|\vec{q}| = 0.20 \text{ \AA}^{-1}$. The solid lines represent fits to the data using the Halperin-Hohenberg form of the spectral function $F^t(\vec{q}, \omega)$. Note that there is no evidence of a central peak.
- Fig. 2.** Wave-vector dependence of the spin-wave line shapes observed at a reduced temperature difference $(T - T_c)/T_c = 0.05$. The solid lines represent fits to the data^c using the Halperin-Hohenberg form of $F^t(\vec{q}, \omega)$. Note that there is no evidence of a central peak.
- Fig. 3.** Log-log plots of the temperature dependence of the spin-wave stiffness constant $D = \omega_0(q)/q^2$. Values of D are shown for all three forms of $F^t(\vec{q}, \omega)$.
- Fig. 4.** Temperature variation of the spin-wave line widths observed at $|\vec{q}| = 0.20 \text{ \AA}^{-1}$ obtained by fitting to the Halperin-Hohenberg form of $F^t(\vec{q}, \omega)$. The solid line represents a power law with exponent -1.04 .
- Fig. 5.** Wave-vector dependence of the spin-wave line widths observed at a reduced temperature difference $(T_c - T)/T_c = 0.05$ obtained by fitting to the Halperin-Hohenberg form of $F^t(\vec{q}, \omega)$. The solid line represents the Harris expression for the line width [18], which is essentially a power law with exponent 4.0.

FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5