Application of Moments to Light Scattering in Antiferromagnets*

W. J. Brya, Peter M. Richards,† and R. R. Bartkowski
Sandia Laboratories, Albuquerque, New Mexico 87115
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Measurements on two-spin fluctuation light scattering in MnF_2 have been obtained over the temperature range 2–300°K; the results are in good agreement with a theoretical analysis based on frequency moments and intensity of the scattering spectra. We find that the first moment shows critical-type behavior in the vicinity of the Néel temperature T_N and that the measured high-temperature second moment allows a determination of the antiferromagnetic exchange constant in the paramagnetic phase.

We report on experiments and the theory for magnetic light scattering in MnF_2 involving two-spin fluctuations. Our results are novel in that the analysis is based on frequency moments and the integrated intensity of the scattering spectra. Earlier two-magnon studies[1-7] have not taken this point of view and therefore, we feel, have missed certain features of interest. In particular, we find that the first moment $\langle\omega\rangle$ changes rapidly near the Néel temperature T_N and gives an indication of critical behavior in agreement with simple theory. Previous authors have concentrated on the two-magnon peak position which shows much less variation in the critical region.

Measurements of the zeroth (i.e., integrated scattering intensity), first, and second moments of the spectra for MnF_2 have been obtained over the temperature range 2–300°K; these results are compared with theoretical calculations over specific temperature intervals. Agreement between experiment and theory is quite good. We also demonstrate that measurement of the high-temperature second moment shows promise as a convenient method for determination of the antiferromagnetic exchange constant in the paramagnetic phase.

The magnetic scattering spectra of an oriented single crystal of MnF_2 were obtained with a conventional Raman scattering apparatus used in the usual 90° scattering configuration. Temperature control was achieved with a stabilized variable-temperature Dewar; however, we estimate the temperature in the scattering volume to be accurate only to ±3°K. The great majority of the spectra were obtained in the $X(YX)Y$ scattering geometry; the less intense spectra observed in the $X(YZ)Y$ configuration showed essentially the same behavior. At low temperatures the spectrum is characterized by the usual two-magnon line at ~100 cm^{-1} as previously reported.[2] As the temperature is raised, the scattering *increases* in total intensity and the peak shifts to lower frequencies until, at temperatures well above $T_N = 67.7$°K, the spectrum is characterized by a symmetric near-Gaussian line centered at zero frequency shift. In the region of T_N there are marked changes in intensity near zero frequency, whereas the scattering peak shows little change in position. This indicates that the first moment is changing much more rapidly than the two-magnon frequency in the critical region.

The temperature dependences of several aspects of the scattering are given in Fig. 1. The inset shows the variation of the integrated scattering intensity (both Stokes and anti-Stokes components) relative to its 2°K value. Here, the measured intensities have carefully been adjusted to compensate for varying experimental conditions at the different temperatures by normalizing the integrated magnetic scattering intensity to the predicted temperature dependence of several of the Stokes phonon lines which we have also measured. One notes that the high-temperature intensity exceeds the 2°K value by a factor of 4.45; this increased intensity at high temperatures is contrary to previously reported behavior[5] in NiF_2.

Also in Fig. 1 we show the temperature dependences of the first moment of the integrated scattering intensity and the frequency shift of the Stokes scattering peak. The smooth variation in the scattering peak frequency with changing temperature, even through T_N, is similar to behavior previously reported for NiF_2 and other antiferromagnets.[8] The first moment, however, shows a much more pronounced decrease with increasing temperature near T_N.

A high-resolution (1 cm^{-1}) study of the 300°K spectrum has also been conducted in order to determine the high-temperature second moment of the line; a discussion of our results follows.

The basic Hamiltonian for two-spin fluctuation

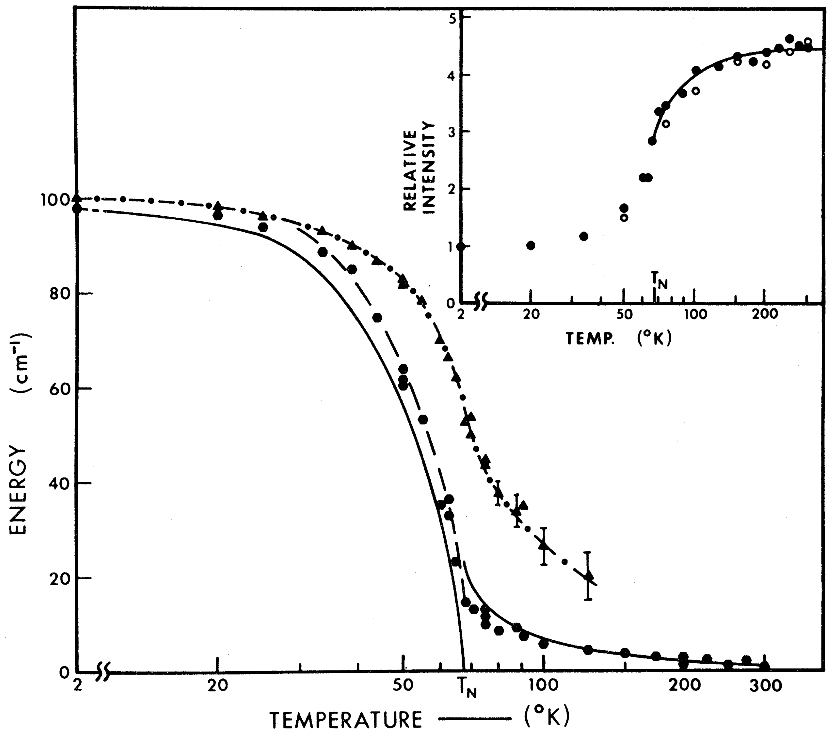

FIG. 1. Temperature dependence of several aspects of magnetic scattering spectra in MnF_2. Triangles and dot-dashed curves: observed frequency shift of the Stokes scattering peak. Hexagons: observed first moment $\langle\omega\rangle$ of the total magnetic scattering. Solid curves: above T_N, theoretical $\langle\omega\rangle$ based on static two-spin correlation functions; below T_N, molecular-field term $\langle\omega\rangle_{mf}$. Dashed curve: theoretical $\langle\omega\rangle$ for $T \leq T_N$ combining $\langle\omega\rangle_{mf}$ and experimental results for $T \geq T_N$, as discussed in text. The inset shows the observed integrated scattering intensity relative to its 2°K value for $X(YX)Y$ scattering geometry (solid circles) and $X(YZ)Y$ geometry (open circles). Solid curve: theoretical relative intensity for $T \geq T_N$.

light scattering in MnF_2 takes the form[2-4]

$$\mathcal{H}' = \sum_{ij} G_{ij}{}^{\alpha\beta}[\vec{S}_i \cdot \vec{S}_j + (\gamma - 1)S_{iz}S_{jz}]E_{1\alpha}E_{2\beta}, \qquad (1)$$

where $E_{1\alpha}$ and $E_{2\beta}$ are electric vectors of the incident and scattered waves polarized in directions α and β, respectively, and $G_{ij}{}^{\alpha\beta}$ is an appropriate next-nearest-neighbor, excited-state–ground-state exchange interaction. The quantity γ represents an anisotropy factor (for $\gamma \neq 1$) which is allowed by the tetragonal symmetry. It has been common practice in previous treatments to consider only the transverse components $S_{i+}S_{j-} + S_{i-}S_{j+}$ in $\mathcal{H}'$ since these give rise to the dominant two-magnon excitation at low temperatures. For a detailed study over the whole temperature range, as presented here, it is, however, essential to retain the longitudinal components $S_{iz}S_{jz}$ as well.

The inelastic scattering intensity at frequency shift ω is proportional to

$$I(\omega) = \int_{-\infty}^{\infty} dt\, \langle \mathcal{H}'(t)\mathcal{H}'\rangle e^{-i\omega t}, \qquad (2)$$

where the time dependence of $\mathcal{H}'$ is governed by the usual isotropic ground-state Heisenberg interaction with antiferromagnetic coupling J between next-nearest neighbors. The nth moment $\langle\omega^n\rangle$ of Eq. (2) is evaluated in terms of commutators[9] and involves static correlations whose complexity increases rapidly with n; the area under the curve of intensity versus frequency is proportional to $A = \langle\mathcal{H}'\mathcal{H}'\rangle$. The moments and area formulas include contributions from both Stokes and anti-Stokes components.

The area can be calculated exactly at infinite temperature and at zero temperature if a Nêel ground state is used. As a result we find

$$A_\infty/A_0 = \tfrac{1}{3}(S+1)^2[\tfrac{1}{3}(2+\gamma^2)] \qquad (3)$$

for the ratio of infinite-to-zero temperature areas. The dependence on S comes from the fact that fluctuations in $\langle(S_{i\alpha})^2\rangle$ are proportional to $S(S+1)$ at $T=\infty$, but only to S at $T=0$ [i.e., $(S_{ix})^2 + (S_{iy})^2 = S(S+1) - (S_{iz})^2 \simeq S$]. For the isotropic ($\gamma = 1$) case, the total integrated intensity is predicted to increase for $S > \frac{1}{2}$, and the ratio is 4.08 for MnF_2 ($S = \frac{5}{2}$). Figure 1 (inset) shows the ratio

A_∞/A_0 to be 4.45 which is in reasonable agreement with the above prediction. A small anisotropy $\gamma-1=0.12$ is one possible means of removing the observed discrepancy. Spin-wave corrections to the Nêel state are in the wrong direction since they decrease A_∞/A_0 by 5–10%.

The basic method of calculating A and $\langle\omega\rangle$ for $T \geq T_N$ is to decouple the four-spin averages into products of two-spin × two-spin correlation functions. (A useful identity reduces $\langle\omega\rangle$ to terms involving only four-spin operators.) Once expectation values have been so reduced to products of two-spin correlation functions, we make use of the recent study of Ritchie and Fisher[10] of two-spin correlations in the Heisenberg ferromagnet[11] at and above the critical temperature.

The solid curves in Fig. 1 for $T \geq T_N$ are results of these calculations. The area is normalized to its high-temperature value, but the first moment contains no adjustable parameters (we take $J=-1.76$°K from neutron-scattering data[12]). Agreement is very good in all aspects except for $\langle\omega\rangle$ at T_N where the observed value of 15 cm^{-1} is significantly less than the theoretical 23 cm^{-1}.

The two-spin correlations are known with less certainty below T_N. We have therefore used the following scheme to estimate $\langle\omega\rangle$ in the region $T<T_N$:

$$\langle\omega\rangle=\langle\omega\rangle_{\rm mf}+f(|T-T_N|),$$

where $\langle\omega\rangle_{\rm mf}$ is the molecular-field ($\langle S_{iz}S_{jz}\rangle=\langle S_{iz}\rangle^2$) approximation to $\langle\omega\rangle$ and $f(|T-T_N|)$ is the short-range fluctuation contribution which is assumed to be the same for a given temperature difference above and below T_N. The solid curve in Fig. 1 for $T<T_N$ is $\langle\omega\rangle_{\rm mf}$ obtained from published values[13,14] for the sublattice magnetization in MnF_2. The dashed curve for $\langle\omega\rangle$ results when $\langle\omega\rangle_{\rm mf}$ is added to the fluctuation term $f(|T-T_N|)$, which is obtained directly from the *experimental* $\langle\omega\rangle$ for $T>T_N$. Our assumption of the form for $f(|T-T_N|)$ is consistent with scaling hypotheses,[15] and the resultant $\langle\omega\rangle$ is seen to give good agreement with the data in the critical region.

The second moment $\langle\omega^2\rangle$ has been calculated at infinite temperature with the result

$$\langle\omega^2\rangle_\infty=\tfrac{16}{3}J^2ZS(S+1)\times\left[1-\frac{2}{5Z}\frac{(\gamma-1)^2}{\gamma^2+2}\left(1+\frac{9}{8S(S+1)}\right)\right], \quad (4)$$

where Z is the number of neighbors coupled by J. Note that for MnF_2 ($Z=8$) the anisotropy ($\gamma\neq 1$) correction is negligible. The effect of nearest-neighbor exchange[12] is also negligible, contributing less than 2% to $\langle\omega^2\rangle_\infty$. We thus predict $\langle\omega^2\rangle_\infty^{1/2}=23.7$ cm^{-1} based on the neutron-scattering result $J=-1.23$ cm^{-1}; this compares very favorably with the observed room-temperature value $\langle\omega^2\rangle^{1/2}=24.3$ cm^{-1} which yields $J=-1.26$ cm^{-1}.

A detailed derivation of the equations for A and $\langle\omega\rangle$ is reserved for a separate publication, together with a discussion of the observed high-temperature line shape and its connection with spin diffusion.

In summary, we have shown the applicability of the moments formalism to light scattering in magnetic systems. From the theoretical standpoint, moments are convenient since their calculation involves static spin-correlation functions which often can be computed with high reliability. Our expression for the high-temperature second moment, for example, is exact. By contrast, theories which attempt to calculate the overall line shape have required several approximations such as decoupling of time-dependent Green's functions.

The experimental determination of moments of light scattering appears to have distinct advantages over, say, spin resonance (ESR) techniques. ESR lines in similar exchange-coupled systems have broad Lorentzian wings so that measurements of their second moment are extemely difficult. This problem does not arise in light scattering where the intensity falls off sufficiently fast that moments can be measured readily. The second moment provides a direct and perhaps most convenient measure of the exchange constant J at high temperature; this should prove to be a useful technique for determining the temperature variation of J in the paramagnetic phase.

Finally, we emphasize that the rapid variation of the first moment has been observed near T_N so that, perhaps contrary to previous analyses, it appears that critical-type behavior of short-range correlations is manifested in certain aspects of two-spin fluctuation light scattering.

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†Permanent address: Department of Physics, The University of Kansas, Lawrence, Kans. 66044.

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Variable Masses in Fission and Heavy-Ion Collisions*

Peter Lichtner and Dieter Drechsel
Institut für Kernphysik der Universität Mainz, Mainz, Germany

and

Joachim Maruhn and Walter Greiner
Institut für Theoretische Physik der Universität Frankfurt, Frankfurt am Main, Germany

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With the use of the cranking formula, the coordinate-dependent mass parameters of the kinetic-energy operator in fission processes and heavy-ion collisions are calculated in the two-center oscillator model. It is shown that the reduced mass and also the classical moment of inertia are obtained for large separations of the fragments. For small separations, however, the mass parameter for the motion of the centers of mass of the fragments is larger than the reduced mass by an order of magnitude.

The double-center shell model developed during the last few years[1-3] has been successful in describing fission phenomena as well as heavy-ion scattering. However, up to now the variation of the inertial parameters in this model has not been studied. In fact, the discussion of fission in previous work has been confined to the mapping of potential energy surfaces, and the treatment of heavy-ion scattering has been carried out under the assumption of a constant reduced mass.

In this paper we show that the inertial parameters change very rapidly, particularly when the two fragments or ions have a large overlap. Thus the fission lifetimes and the cross sections for heavy-ion scattering at high energies will be strongly affected.

The effect of a variable mass has been studied by Hofmann and Dietrich[4] in a one-dimensional model using several phenomenological forms for the mass variation. Similarly Updegraff and Onley[5] have included this effect in a three-dimensional case in their description of photofission in the dynamic collective model.

Griffin[6] has stressed the importance of the Landau-Zener[7] effect of level crossing on the mass parameters and estimated that the masses should be higher than the reduced mass by at least an order of magnitude. A similar conclusion has been reached by Sobiczewski *et al.*[8] in the case of β vibrations and in recent unpublished work.[9] The advantage of the double-center shell model used in the present calculations is its ability to describe the complete fission process to the stage of two separated fragments and, furthermore, its applicability to heavy-ion scattering. In this note we restrict ourselves to the symmetric double-center oscillator which is described