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Excitation spectra for spin-3/2 systems with high-degree pair interactions[*]

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We studied the excitation spectra of a spin-3/2 system with both dipolar and quadrupolar pair interactions. To study the thermal variations of the spectra we calculated the spectral-weight functions by using finite-temperature two-time Green's functions. These functions were evaluated by using the equation-of-motion method and by applying the random-phase decoupling approximation. At low temperatures when the system is ferromagnetically ordered we find for small wave vectors k that the excitation spectra have a quadratic dependence on k. If the dipolar coupling is small compared to the quadrupolar, the system loses its magnetic ordering at a temperature T_d but retains quadrupolar order up to a higher temperature T_q. As one increases the temperature from absolute zero, the excitation spectra for small wave vectors gradually change and become linearly dependent on k when T_d is reached. In the phase with only quadrupolar order, the excitation spectra remain linear in k. The amplitudes of the excitations are shown to satisfy sum rules and we discuss how these excitations may be observed by inelastic neutron scattering experiments.

I. INTRODUCTION

Rare-earth ions retain, in large part, their orbital angular momentum when they form solids. For this reason it has been found that high-degree pair interactions between these ions are comparable to or even stronger than the dipolar coupling. The generic Hamiltonian for spin systems with dipolar and quadrupolar pair interactions is

$$\mathcal{H} = -\Gamma^{(1)} \sum_{\langle ij\rangle} \mathcal{O}_i^1 \cdot \mathcal{O}_j^1 - \Gamma^{(2)} \sum_{\langle ij\rangle} \mathcal{O}_i^2 \cdot \mathcal{O}_j^2 , \qquad (1)$$

where the operators $\mathcal{O}_i^l$ are in the spherical tensor form given by Judd,[1] and the summation is over pairs of nearest neighbors.

In a previous paper[2] we studied the influence of quadrupolar pair interactions on the elementary excitation of a spin-1 ferromagnet $\Gamma^{(1)} > 0$. For $|\Gamma^{(2)}| \leq |\Gamma^{(1)}|$ spin-1 systems have magnetic order. However, for $|\Gamma^{(2)}| > |\Gamma^{(1)}|$ these systems are anomalous in that they do not possess a magnetic phase transition. Systems with $s \geq \frac{3}{2}$ do have magnetic (dipolar) phase transitions no matter how large $\Gamma^{(2)}$ is, as long as $\Gamma^{(1)}$ is nonzero.[3] Of course the transition temperature T_d is proportional to $\Gamma^{(1)}$ and approaches $T = 0$ °K as $\Gamma^{(1)}$ goes to zero. As one lowers the temperature of a system with quadrupolar coupling $\Gamma^{(2)} \gg \Gamma^{(1)} > 0$ the system first undergoes a transition to a phase with only quadrupolar order at a temperature T_q. As the temperature is further lowered the system undergoes a second transition at T_d to a phase with both dipolar and quadrupolar order. If the dipolar and quadrupolar coupling are comparable the transition temperatures T_d and T_q approach one another. For the ratio $\alpha \equiv \Gamma^{(1)}/\Gamma^{(2)}$ greater than some number which depends on the magnitude of the spin s, there is only one phase transition.

The elementary excitations of spin-$\frac{3}{2}$ systems have been studied with only quadrupolar coupling.[4,5] In this paper we consider spin-$\frac{3}{2}$ systems which have both dipolar and quadrupolar pair interactions. We will be particularly interested in studying the variation with temperature of the excitation spectra for systems which have two successive phase transitions. Also, we will be primarily interested in the ratios α much less than 1. The excitation spectra of systems with only quadrupolar pair interactions have dispersion relations which are linear for small values of the wave vector k.[4,5] On the contrary, for ferromagnets the spin-wave spectrum is quadratic for small k, i.e., proportional to k^2. We will show how for our system, Eq. (1), with spin-$\frac{3}{2}$ these disparate excitation spectra change from a quadratic k dependence in the magnetic phase when $T < T_d$ to a linear form in the quadrupolar phase $T_d \leq T < T_q$.

Because these changes in the dispersion curves take place as we vary the temperature we will use finite-temperature two-time Green's functions to determine the spectral weight function for the system. To determine these Green's functions we use the equation-of-motion method, and apply the random-phase decoupling scheme of Haley and Erdős.[6] Our results apply, at least qualitatively, to all systems with spin $s \geq \frac{3}{2}$ which have only dipolar and quadrupolar coupling. When one considers still higher-degree pair interactions there may be additional phase transitions,[7] and we expect new features to the excitation spectra for systems with such interactions. Although the pair interactions between ions in solids are anisotropic, we have considered only isotropic interactions in this paper. In this way we can separate the effects on the excitation spectra due to high-degree pair interactions from those due to anisotropy of the pair interactions.

In Sec. II we calculate the spectral weight function for the excitations of a spin-$\frac{3}{2}$ system with dipolar and quadrupolar pair interactions. We evaluate the dynamical susceptibility for several ratios α of the dipolar to quadrupolar coupling in Sec. III. From the imaginary part of this function we obtain the spectral-weight function. We follow the variation of the excitation spectra as the temperature goes from $T = 0$ °K through the dipole transition T_d and up to the quadrupole transition temperature T_q. In Sec. IV we discuss our results and point out those qualitative features which change when one considers systems with spin greater than $\frac{3}{2}$ and when one uses a decoupling approximation better than the random phase.

II. CALCULATION

To find the excitation spectra of the spin system described by the Hamiltonian Fq. (1) we will calculate the spectral-weight function $A(k, \omega)$. This is related to the dynamical susceptibility as follows:

$$A(k, \omega) = -(1/\pi)\,\mathrm{Im}\chi(k, \omega) , \tag{2}$$

where

$$\chi(k, \omega) = -i \int_{-\infty}^{\infty} e^{i\omega t}\, dt \, \langle [B^\dagger(k, t), B(k, 0)] \rangle \Theta(t) , \tag{3}$$

where

$$B(k, t) = e^{i\mathcal{H}t/\hbar} \sum_j e^{i\vec{k}\cdot\vec{r}_j} B(j)\, e^{-i\mathcal{H}t/\hbar} ,$$

$\Theta(t)$ is the unit step function, and the angular brackets denote a thermal average of the commutator. The wave vector k is a vector quantity but we will for the most part omit the arrow above k. In order to determine the excitation spectra the operator $B(j)$ must have transition matrix elements that connect two states of the system. We will be interested in three types of operators which are categorized according to the change of the z component of the angular momentum $\vec{\mathcal{S}}$. If we assume the ground state of our system is given as $\langle \mathcal{S}_z \rangle_g = -Ns$, where $|\,\rangle_g \equiv |-s, -s, -s, \ldots\rangle$, then the excitations are characterized by the number of spin deviations $m \equiv \langle \mathcal{S}_z \rangle + Ns$. Excitations with one spin deviation $m = 1$ are created by the operator

$$B_1(i) = \mathcal{S}^+(i): \tag{4}$$

These are called spin-wave excitations. Those with two spin deviations $m = 2$ are called quadrupolar like excitations and are excited by the operator

$$B_2(i) = [\mathcal{S}^+(i)]^2 , \tag{5}$$

while octopolar like excitations $m = 3$ are excited by the operator

$$B_3(i) = [\mathcal{S}^+(i)]^3 . \tag{6}$$

The susceptibility can be determined from the following two-time temperature-dependent retarded Green's function:

$$\begin{aligned} G_{\mu_1\nu_1\mu_2\nu_2}(k, t) &\equiv -i\,\Theta(t) \langle [X_{\nu_1\mu_1}(k, t), X_{\mu_2\nu_2}(k, 0)] \rangle \\ &= -i\,\Theta(t) \langle [X^\dagger_{\mu_1\nu_1}(k, t), X_{\mu_2\nu_2}(k, 0)] \rangle , \end{aligned} \tag{7}$$

where the operators $X_{\mu\nu}$ are defined by their matrix elements[7a]

$$\langle \alpha | X_{\mu\nu} | \beta \rangle = \delta_{\mu\alpha}\delta_{\nu\beta} . \tag{8}$$

From this definition it immediately follows that

$$X^\dagger_{\mu\nu} = X_{\nu\mu}$$

and

$$[X_{\mu_1\mu_2}, X_{\mu_3\mu_4}] = \delta_{\mu_2\mu_3} X_{\mu_1\mu_4} - \delta_{\mu_1\mu_4} X_{\mu_3\mu_2} . \tag{9}$$

The relation of these operators to $\mathcal{O}^l_m$ is given by Judd.[1] If we label the eigenstates of our Hamiltonian Eq. (1) by the eigenvalues of the operator $\mathcal{S}_z$ then it follows that the difference between the states is just the number of spin deviations, i.e.,

$$\mu - \nu \equiv m . \tag{10}$$

We will show how we determined the susceptibility for $m = 1$ excitations by determining the appropriate Green's functions. The determination of the excitation spectra for $m = 2$ and 3 follows the same procedure and we present these results at the end of Sec. III.

The $m = 1$ excitations can be measured by inelastic neutron scattering experiments. The cross section for this scattering is given in terms of a spectral weight function[8] $A(k, \omega)$. For $m = 1$ excitations of a system with the ground state $|-s, -s, -s, \ldots\rangle$ this function is the imaginary part of the susceptibility Eq. (3) with $B \equiv g\mu_B \mathcal{S}^+$, i.e.,

$$\chi_1(k, \omega) = -i(g\mu_B)^2 \times \int_{-\infty}^{\infty} e^{i\omega t}\, dt \, \langle [\mathcal{S}^-(k, t), \mathcal{S}^+(k, 0)] \rangle \Theta(t) . \tag{3a}$$

As we have introduced the factor $(g\mu_B)^2$, the spectral weight function is related to this definition of the susceptibility as follows:

$$A_1(k, \omega) = -\frac{1}{\pi (g\mu_B)^2}\, \mathrm{Im}\chi_1(k, \omega) . \tag{2a}$$

For spin $\frac{3}{2}$ the indices μ and ν take on the values $\pm\frac{3}{2}$ and $\pm\frac{1}{2}$, and the spin-raising operator is related to the standard basis operators X in the following way:

$$\mathcal{S}^{+} = \sqrt{3}X_{-1/2,-3/2} + 2X_{1/2,-1/2} + \sqrt{3}\,X_{3/2,1/2}$$

or (11)

$$\mathcal{S}^{+} = \sum_{\alpha} h_{\alpha} X_{\alpha} ,$$

where we use the following contracted index notation: $\alpha \equiv (\mu, \nu) = 1, 2, 3$; $1 \equiv (-\frac{1}{2}, -\frac{3}{2})$, $2 \equiv (\frac{1}{2}, -\frac{1}{2})$, $3 \equiv (\frac{3}{2}, \frac{1}{2})$; $h_1 = h_3 = \sqrt{3}$; and $h_2 = 2$. With this relation and a similar one for the adjoint operator $\mathcal{S}^{-}$, we find

$$[\mathcal{S}^{-}(k, t), \mathcal{S}^{+}(k, 0)] = \sum_{\alpha\beta} h_{\alpha} h_{\beta} [X_{\alpha}^{\dagger}(k, t), X_{\beta}(k, 0)] . \tag{12}$$

Thus, we can write the dynamical susceptibility Eq. (3a) in terms of the Fourier transform of the Green's functions defined by Eq. (7) for $\mu - \nu = 1$,

$$\chi_1(k, \omega) = (g\mu_B)^2 \sum_{\alpha\beta} h_{\alpha} h_{\beta} G_{\alpha\beta}(k, \omega) , \tag{13}$$

where

$$G_{\alpha\beta}(k, \omega) \equiv \int_{-\infty}^{\infty} e^{i\omega t}\, dt\, G_{\mu_1\nu_1\,\mu_2\nu_2}(k, t) . \tag{14}$$

The Green's functions in the susceptibility are determined by finding the solutions to their equations of motion. The method we used to find the solutions follows that of Haley and Erdös.[6] The equation of motion satisfied by the Green's function Eq. (7) is

$$i\frac{d}{dt} G_{\mu_1\nu_1\mu_2\nu_2}(k, t) = \langle [X_{\nu_1\mu_1}(k, 0), X_{\mu_2\nu_2}(k, 0)] \rangle\, \delta(t) - i\langle [[X_{\nu_1\mu_1}(k, t), \mathcal{H}], \times X_{\mu_2\nu_2}(k, 0)] \rangle\, \Theta(t) , \tag{15}$$

where $\delta(t)$ is a Dirac δ function and the Hamiltonian for our system is given by Eq. (1). By using Eq. (1), the commutator relation Eq. (9), and the relation[1] between the operators $\mathcal{O}_m^l$ and $X_{\nu\mu}$, we find that the Green's functions obey the following equation:

$$\begin{aligned} i\frac{d}{dt} G_{\mu_1\nu_1\mu_2\nu_2}(k, t) = {} & (\langle X_{\nu_1\nu_1} \rangle - \langle X_{\mu_1\mu_1} \rangle)\, \delta(t)\, \delta_{\nu_1\nu_2}\, \delta_{\mu_1\mu_2} \\ & + i \sum_{\mu'\nu'\nu''} \sum_{k'} a_{\mu_1\nu''\mu'\nu'}(k') \langle [X_{\nu'\mu'}(k', t) X_{\nu_1\nu''}(k - k', t), X_{\mu_2\nu_2}(k, 0)] \rangle\, \Theta(t) \\ & - i \sum_{\mu'\nu'\mu''} \sum_{k'} a_{\mu''\nu_1\mu'\nu'}(k') \langle [X_{\nu'\mu'}(k', t) X_{\mu''\mu_1}(k - k', t), X_{\mu_2\nu_2}(k, 0)] \rangle\, \Theta(t) , \end{aligned} \tag{16}$$

where

$$a_{\mu''\nu''\mu'\nu'}(k') \equiv (-)^{\nu'+\nu''-2s} \sum_{l=1,2} [l]\, \Gamma^{(l)}(k') \begin{pmatrix} s & l & s \\ -\nu'' & m' & \mu'' \end{pmatrix} \begin{pmatrix} s & l & s \\ -\nu' & m' & \mu' \end{pmatrix} \tag{17}$$

and we have assumed $\Gamma^{(l)} > 0$ [see Eq. (1)] so that the system orders uniformly, i.e.,

$$\langle X_{\nu\nu}(k) \rangle = \langle X_{\nu\nu}(0) \rangle\, \delta_{k0} \equiv \langle X_{\nu\nu} \rangle . \tag{18}$$

The expression in large parentheses is a $3j$ symbol where $m' = \nu' - \mu' = \nu'' - \mu''$ and $[l] \equiv 2l + 1$. The Fourier transform of the nearest-neighbor pair coupling [see Eq. (1)] is given as

$$\Gamma^{(l)}(k') = z\Gamma^{(l)} \gamma_{k'} ,$$

where

$$\gamma_{k'} \equiv (1/z) \sum_{\delta} e^{i\vec{k}'\cdot\vec{\delta}} \tag{19}$$

and the sum is over the z nearest neighbors. By relating the operators $X_{\nu\nu}$ and $X_{\mu\mu}$ to the $\mathcal{O}_m^l$ (see Judd[1]), their thermal averages can be expressed in terms of the dipolar, quadrupolar, and octopolar order parameters,

$$\mathcal{O}_l \equiv \langle \mathcal{O}_o^l \rangle \tag{20}$$

for $l = 1, 2, 3$.

From Eq. (16) we see that the equation for a Green's function involves those of a higher order. To find them one has to solve yet another equation which contains Green's functions of even higher order. This leads to a hierarchy of equations which are not readily soluble. Therefore, to find the solution of $G_{\mu_1\nu_1\mu_2\nu_2}$ we have used a decoupling scheme which is based on the random-phase approximation; that is to say, we make the following approximation[6,9] in Eq. (16):

$$\begin{aligned} & \langle [X_{\nu'\mu'}(k', t) X_{\mu''\nu''}(k - k', t), X_{\mu\nu}(k, 0)] \rangle \\ & \quad \cong \langle X_{\nu'\mu'}(k', t) \rangle \langle [X_{\mu''\nu''}(k - k', t), X_{\mu\nu}(k, 0)] \rangle \\ & \quad + \langle X_{\mu''\nu''}(k - k', t) \rangle \langle [X_{\nu'\mu'}(k', t), X_{\mu\nu}(k, 0)] \rangle \\ & \quad + \langle [X_{\nu'\nu''}(k, t), X_{\mu\nu}(k, 0)] \rangle\, \delta_{\mu'\mu''} \\ & \quad - \langle [X_{\mu''\mu'}(k, t), X_{\mu\nu}(k, \ldots, 0)] \rangle\, \delta_{\nu'\nu''} . \end{aligned} \tag{21}$$

By assuming uniaxial ordering, the thermal expectation value of the operator $X_{\mu\nu}$ is

$$\langle X_{\mu'\nu'}(k',t)\rangle = \langle X_{\nu'\nu'}(k')\rangle\,\delta_{\nu'\mu'}\ . \tag{22}$$

For a system that orders in a translationally invariant mode, this reduces to

$$\langle X_{\mu'\nu'}(k',t)\rangle = \langle X_{\nu'\nu'}\rangle\,\delta_{\nu'\mu'}\,\delta_{k'0}\ . \tag{23}$$

By using this expectation value and the decoupling approximation Eq. (21) we find that the equation of motion for the Green's function Eq. (16) reduces to

$$i\frac{d}{dt}G_{\mu_1\nu_1\mu_2\nu_2}(k,t) = -(\langle X_{\mu_1\mu_1}\rangle - \langle X_{\nu_1\nu_1}\rangle)\,\delta(t)\delta_{\mu_1\mu_2}\,\delta_{\nu_1\nu_2} + \overline{a}_{\mu_1\nu_1}(0)\,G_{\mu_1\nu_1\mu_2\nu_2}(k,t)$$
$$+(\langle X_{\mu_1\mu_1}\rangle - \langle X_{\nu_1\nu_1}\rangle)\sum_{\mu',\nu'} a_{\mu_1\nu_1\mu'\nu'}(k)\,G_{\mu'\nu'\mu_2\nu_2}(k,t)\ , \tag{24}$$

where

$$\overline{a}_{\mu_1\nu_1}(0) \equiv \sum_{\nu'}\langle X_{\nu'\nu'}\rangle\left[a_{\nu_1\nu_1\nu'\nu'}(0) - a_{\mu_1\mu_1\nu'\nu'}(0)\right]$$
$$= \sum_{l=1,2}[l]^{1/2}\mathcal{O}_l\,\Gamma^{(l)}(0)\left[(-)^{\nu_1-s}\begin{pmatrix} s & l & s\\ -\nu_1 & 0 & \nu_1\end{pmatrix} - (-)^{\mu_1-s}\begin{pmatrix} s & l & s\\ -\mu_1 & 0 & \mu_1\end{pmatrix}\right]\ .$$

Finally, by taking the Fourier time transform of these equations and by using the definition Eq. (14) of the time-transformed Green's function, we find

$$\sum_{\gamma}\left[M_{\alpha\gamma}(k) - \omega\delta_{\alpha\gamma}\right]G_{\gamma\beta}(k,\omega) = e_\alpha\,\delta_{\alpha\beta}\ , \tag{25}$$

where

$$M_{\alpha\gamma}(k) \equiv e_\alpha\,a_{\alpha\gamma}(k) + \overline{a}_\alpha(0)\,\delta_{\alpha\gamma} = M_{\mu_1\nu_1\mu'\nu'}(k)$$
$$= e_{\mu_1\nu_1}\,a_{\mu_1\nu_1\mu'\nu'}(k) + \overline{a}_{\mu_1\nu_1}(0)\,\delta_{\mu_1\mu'}\,\delta_{\nu_1\nu'}\ , \tag{26}$$

$$e_\alpha \equiv e_{\mu_1\nu_1} = \langle X_{\mu_1\mu_1}\rangle - \langle X_{\nu_1\nu_1}\rangle\ . \tag{27}$$

Equation (25) is identical in *form* to the one obtained by Haley and Erdös.[10] The only difference is that the matrix $M_{\alpha\gamma}$ here refers to our Hamiltonian Eq. (1), which contains quadrupolar as well as dipolar pair interactions.

The matrix equation (25) represents a system of $(2s+1-m)^2$ coupled equations for the excitations $m=\mu-\nu$. For spin $\frac{3}{2}$ and for the $m=1$ excitations, Eq. (25) represents nine equations for the nine-unknown Green's function $G_{\alpha\beta}(k,\omega)$. The solution to the matrix equation is found by multiplying both sides of Eq. (25) by the inverse of the matrix $(M-\omega)$; we find

$$G_{\alpha\beta}(k,\omega) = (M-\omega)^{-1}_{\alpha\beta}\,e_\beta = e_\beta\,c_{\beta\alpha}(k,\omega)/D(k,\omega)\ , \tag{28}$$

where D is the determinant of the matrix $(M-\omega)$ and $c_{\beta\alpha}$ is a cofactor of the determinant. When we calculate the matrix elements $M_{\alpha\gamma}$ and e_β for $(\alpha,\beta) = (-\frac{1}{2},-\frac{3}{2})$, $(\frac{1}{2},-\frac{1}{2})$, $(\frac{3}{2},\frac{1}{2})$ we obtain the Green's functions that give us the dynamical susceptibility for $m=1$ excitations, Eq. (13).

We have evaluated the matrix elements $M_{\alpha\gamma}$ for spin-$\frac{3}{2}$, $m=1$ excitations and find

$$\begin{aligned}
M_{11} &= -\tfrac{1}{2}[a(1-\gamma_k)\,e_1 + c\,e_2 + b\,e_3]\ ,\\
M_{22} &= -\tfrac{1}{2}[\tfrac{1}{2}(a+b)(e_1+e_3) + c(1-\gamma_k)\,e_2]\ ,\\
M_{33} &= -\tfrac{1}{2}[b\,e_1 + c\,e_2 + a(1-\gamma_k)\,e_3]\ ,\\
M_{21} &= M_{23} = \tfrac{1}{5}\sqrt{3}\,z\Gamma^{(1)}e_2\,\gamma_k\ ,\\
M_{12} &= \tfrac{1}{5}\sqrt{3}\,z\Gamma^{(1)}\,e_1\,\gamma_k\ ,\\
M_{32} &= \tfrac{1}{5}\sqrt{3}\,z\,\Gamma^{(1)}e_3\,\gamma_k\ ,\\
M_{31} &= \tfrac{1}{2}\,b\,e_3\,\gamma_k\ ,\\
M_{13} &= \tfrac{1}{2}\,b\,e_1\,\gamma_k\ ,
\end{aligned} \tag{29}$$

where the indices 1, 2, 3 refer to the transitions $(-\frac{1}{2},-\frac{3}{2})$, $(\frac{1}{2},-\frac{1}{2})$, $(\frac{3}{2},\frac{1}{2})$, and $\alpha = \Gamma^{(1)}/\Gamma^{(2)}$ (this α should not be confused with the index α). The γ_k is defined in Eq. (19), and the remaining constants are defined as follows:

$$\begin{aligned}
a &\equiv (1+\tfrac{3}{5}\alpha)\,z\,\Gamma^{(2)}\ ,\\
b &\equiv (-1+\tfrac{3}{5}\alpha)z\,\Gamma^{(2)}\ ,
\end{aligned} \tag{30}$$

and

$$c \equiv \tfrac{4}{5}\,\alpha z\,\Gamma^{(2)}\ .$$

The coefficients e_α defined by Eq. (27) are

$$\begin{aligned}
e_1 &= \tfrac{1}{5}\sqrt{5}(\mathcal{O}_1 + \sqrt{5}\mathcal{O}_2 + 2\mathcal{O}_3)\ ,\\
e_2 &= \tfrac{1}{5}\sqrt{5}(\mathcal{O}_1 - 3\mathcal{O}_3)\ ,
\end{aligned} \tag{31}$$

and

$$e_3 = \tfrac{1}{5}\sqrt{5}(\mathcal{O}_1 - \sqrt{5}\mathcal{O}_2 + 2\mathcal{O}_3)\ .$$

The $\mathcal{O}_l$ are the parameters Eq. (20) that describe the ordered phase when $T<T_q$ or $T<T_d$. They can be determined self-consistently by taking appropriate combinations of the Green's functions Eq. (28).[6] However, in Sec. III we resort to the simpler procedure of using order parameters calculated in the mean-field approximation. For

spin $\frac{3}{2}$ the dipolar and quadrupolar order parameters have already been calculated by Sivardière and Blume.[3]

The general solution to the susceptibility $\chi_1(k, \omega)$, Eq. (13), is rather complicated, as it involves a cubic equation in the frequency ω. However, for special cases of interest to us this equation can be factored and the solutions are simpler.

III. EXCITATION SPECTRA

A. $m=1$

The excitation spectra for a spin-$\frac{3}{2}$ system with only quadrupolar interactions, $\Gamma^{(1)}=0$, have been determined previously.[4,5] Let us now determine the effects of dipolar pair interactions $\alpha \neq 0$ on the spectra. In general, there are no simple relations between the order parameters $\mathcal{O}_1$, $\mathcal{O}_2$, and $\mathcal{O}_3$, and the expression for the susceptibility of Eq. (13) is formidable. However, in the limit of weak dipolar interactions where $\alpha \ll 1$, i.e., $\Gamma^{(1)} \ll \Gamma^{(2)}$, and for low temperatures $T < T_d \ll T_q$, we find in the mean-field approximation that[11]

$$\mathcal{O}_1 \simeq 3\mathcal{O}_3 , \qquad (32)$$

where $\mathcal{O}_1 = \frac{1}{5}\sqrt{5}\langle S_z\rangle = \frac{1}{5}\sqrt{5}M$ is given as the solution the following self-consistent equation[12]:

$$M = \tfrac{3}{2}\tanh(\tfrac{3}{2}\beta\Gamma^{(1)}M) \qquad (33)$$

and $\beta \equiv 1/kT$. As pointed out by Sivardière and Blume,[12] the quadrupole moment is constant at these low temperature, $T < T_d \ll T_q$:

$$\mathcal{O}_2 \simeq \tfrac{1}{2} . \qquad (34)$$

Therefore, for $\alpha \ll 1$ and $T < T_d$ we find from Eqs. (31), (32), and (34) that

$$\begin{aligned} e_1 + e_3 &\simeq \tfrac{2}{3}\sqrt{5}\,\mathcal{O}_1 , \\ e_1 - e_3 &\simeq 2\mathcal{O}_2 , \end{aligned} \qquad (35)$$

and

$$e_2 \simeq 0 .$$

In Fig. 1 we show the variation with temperature of the order parameters for spin-$\frac{3}{2}$ as determined by Sivardière and Blume[3] in the mean-field approximation.

As the coefficient e_2 is approximately zero, it follows immediately from Eq. (28) that

$$G_{\alpha 2}(k, \omega) \sim 0 \qquad (36)$$

and that the matrix elements M_{21} and M_{23} are zero. The expression for the susceptibility is considerably simpler because of these cancellations. By inserting Eqs. (29), (30), and the values of e_α, Eq. (35), in Eq. (28) we have evaluated the Green's functions G_{11}, G_{33}, G_{13}, and G_{31}. By using them in Eq. (13) we find that the dynamical susceptibility for $m=1$ excitations for $T < T_d$ is given as

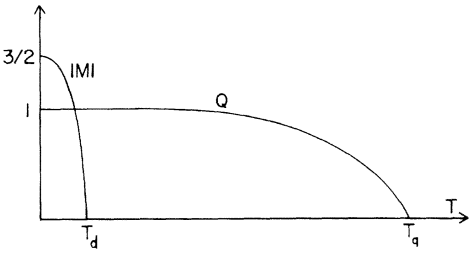

FIG. 1. Thermal variation of the order parameters $M \equiv \sqrt{5}\,\mathcal{O}_1$ and $Q \equiv 2\mathcal{O}_2$ for spin $\frac{3}{2}$ and for $\Gamma^{(2)} \gg \Gamma^{(1)} > 0$ in the mean-field approximation (see Sivardière and Blume, Ref. 3). Note that T_d is much less than T_q.

$$\chi_1(k, \omega) = (g\mu_B)^2 \left(\frac{A_+(k)}{\omega - \omega_+} + \frac{A_-(k)}{\omega - \omega_-}\right) , \qquad (37)$$

where

$$A_\pm(k) = -\sqrt{5}\,\mathcal{O}_1 \pm 3[\tfrac{5}{9}(\mathcal{O}_1)^2(1 - \tfrac{3}{5}\alpha)\gamma_k + 2(\mathcal{O}_2)^2(1-\gamma_k)]/\Delta ,$$

$$\omega_\pm(k) = \tfrac{1}{2}z\Gamma^{(2)}\{\tfrac{1}{3}\sqrt{5}\,\mathcal{O}_1[(1+\tfrac{3}{5}\alpha)\gamma_k - \tfrac{6}{5}\alpha] \pm \Delta\} ,$$

and

$$\begin{aligned} \Delta = [&\tfrac{5}{9}(\mathcal{O}_1)^2(1-\tfrac{3}{5}\alpha)^2\gamma_k^2 \\ &+ 4(\mathcal{O}_2)^2(1-\gamma_k)(1-\tfrac{3}{5}\alpha\gamma_k)]^{1/2} . \end{aligned} \qquad (38)$$

The spectral weight function $A_1(k, \omega)$, Eq. (2a), is found by taking the imaginary part of the susceptibility Eq. (37). Since we have used retarded Green's functions the complex frequency is $\omega + i\epsilon$ with $\epsilon = 0^+$. By using the identity

$$\lim_{\epsilon \to 0} \frac{1}{x + i\epsilon} = P\frac{1}{x} - i\pi\,\mathrm{sgn}(\epsilon)\,\delta(x) \qquad (39)$$

for $x = \omega - \omega_\pm$ we find that the spectral weight function $A_1(k, \omega)$ is

$$A_1(k, \omega) = A_+(k)\,\delta(\omega - \omega_+) + A_-(k)\,\delta(\omega - \omega_-) . \qquad (40)$$

We evaluate $A_\pm(k)$ and $\omega_\pm$ by using the mean-field values for the $\mathcal{O}_l$; we find that A_+, $\omega_+ \geq 0$ while A_-, $\omega_- \leq 0$ for all temperatures. Therefore the first term in Eq. (40) represents energy $\hbar\omega_+$ *absorbed* by the spin system from inelastically scattered neutrons with a probability proportional to[13] $[A_+(k)/(1 - e^{-\beta\omega_+})]\,\delta(\omega - \omega_+)$. The second term corresponds to energy $\hbar|\omega_-|$ *emitted* by the system or absorbed by the inelastically scattered neutrons with a probability proportional to $[|A_-(k)|/(e^{-\beta\omega_-} - 1)]\,\delta(\omega - \omega_-)$.

The sum of the two amplitudes satisfies the following sum rule:

$$\int_{-\infty}^{\infty} A_1(k, \omega)\,d\omega = -\frac{1}{(g\mu_B)^2}\left(\frac{1}{\pi}\int_{-\infty}^{\infty} \mathrm{Im}\,\chi_1(k, \omega)\,d\omega\right)$$

$$= \langle [S_k^-(0),\ S_k^+(0)] \rangle$$

or

$$\int_{-\infty}^{\infty} A_1(k,\ \omega)\, d\omega = -2\langle S_z\rangle = -2\sqrt{5}\mathcal{O}_1\ , \tag{41}$$

where we have used the definitions of A_1 and χ_1, Eqs. (2a) and (3a), and the commutator $[S_i^+,\ S_j^-] = 2S_{zi}\delta_{ij}$. From the expressions for $A_\pm(k)$ [see Eq. (37)] we readily ascertain that Eq. (41) is satisfied by our amplitudes

$$A_+(k) + A_-(k) = -2\sqrt{5}\mathcal{O}_1\ . \tag{42}$$

Since we have chosen our ground state to be $|-\frac{3}{2}, -\frac{3}{2}, \ldots\rangle$, $\mathcal{O}_1$ is negative and the sum of the amplitudes is positive.

To gain further insight into the response of our system to $m = 1$ excitations let us trace the variation of the spectra with temperature. At $T = 0$ °K

$$\mathcal{O}_1 = -3/2\sqrt{5} \quad \text{and} \quad \mathcal{O}_2 = \tfrac{1}{2}\ ,$$

so that from Eq. (37)

$$A_+ = 3\ , \quad A_- = 0\ ,$$

$$\omega_+ = \tfrac{1}{2}z\Gamma^{(2)}(1 + \tfrac{3}{5}\alpha)(1 - \gamma_k)\ ,$$

and

$$\omega_- = -\tfrac{1}{2}z\Gamma^{(2)}(1 - \tfrac{3}{5}\alpha)\ . \tag{43}$$

Since we derived Eq. (37) on the assumption that $\alpha \ll 1$, we note that in fact ω_- is negative. The $m = 1$ excitation spectra at $T = 0$ °K consist of only one mode $\omega_+(k)$ which goes as k^2 in the limit of long wavelengths. This corresponds to a spin-wave excitation from the ferromagnetically aligned ground state $|-\frac{3}{2}, -\frac{3}{2}, \ldots\rangle$. As the temperature is raised, $0 < T < T_d$, the excited states of the Hamiltonian Eq. (1) become thermally populated. Then the system can *emit* $m = 1$ excitations as well as absorb them, and we find A_- is *nonzero*.

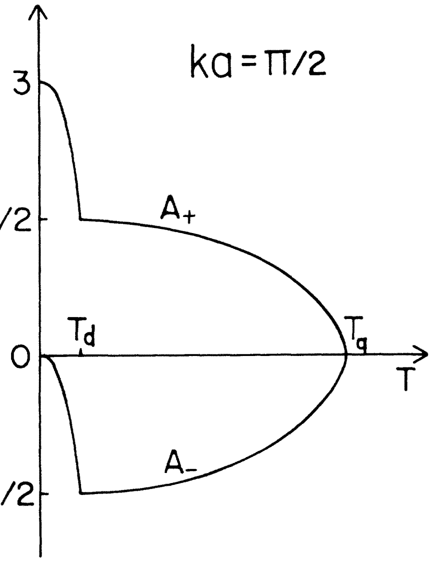

FIG. 2. Thermal variation of the spectral weights for $m = 1$ excitations $A_+(k)$ and $A_-(k)$ at $k = \pi/2a$.

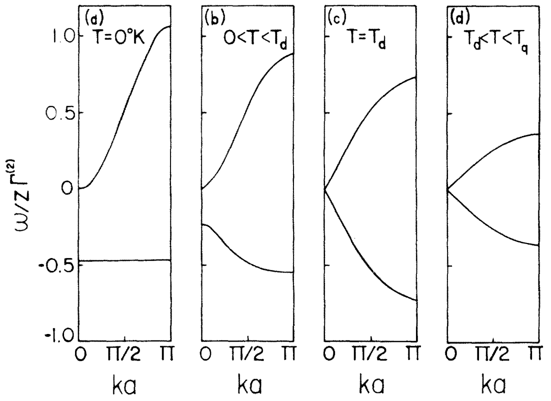

FIG. 3. Spectra of $m = 1$ excitations for $\alpha \equiv \Gamma^{(1)}/\Gamma^{(2)} = 0.1$ for several temperatures: (a) $T = 0$ °K, (b) a temperature T in the range $0 < T < T_d$ such that $\mathcal{O}_1(T)/\mathcal{O}_1(0) = \frac{1}{2}$, (c) $T = T_d$, and (d) a temperature T in the range $T_d < T < T_q$ such that $\mathcal{O}_2(T)/\mathcal{O}_2(0) = \frac{1}{2}$.

As we approach the second-order dipolar transition at T_d, $\mathcal{O}_1$ approaches zero and we find the amplitudes and frequencies of the spectral-weight function Eq. (40) become equal in magnitude and remain opposite in sign,

$$A_\pm = \pm\frac{3}{2}\left(\frac{1-\gamma_k}{1-\frac{3}{5}\alpha\gamma_k}\right)^{1/2}$$

and

$$\omega_\pm = \pm\tfrac{1}{2}z\Gamma^{(2)}[(1-\gamma_k)(1-\tfrac{3}{5}\alpha\gamma_k)]^{1/2}\ . \tag{44}$$

We note that at T_d the dispersion curve is linear in k in the long-wavelength limit for $\alpha < \frac{5}{3}$. In Fig. 2 we show the variation of the amplitudes $A_\pm$ with temperature for $k = \pi/2a$. We note that A_+ decreases while A_- increases (in a negative sense), until $T = T_d$, when they become equal in magnitude. Since $\mathcal{O}_1 = 0$ for $T = T_d$ it follows from the sum rule Eq. (41) that the amplitudes $A_\pm$ must be equal and opposite if they are nonzero. We have also shown in Fig. 3 the excitation spectra $\omega_\pm(k)$ at $T = 0$ °K, $T = T_d$, and an intermediate temperature such that $\mathcal{O}_1(T) = \frac{1}{2}\mathcal{O}_1(T = 0\ °\text{K})$. We take particular note of the change of the cosinelike dispersion curve at $T = 0$ °K into a sinelike curve as we approach the dipolar phase transition at T_d.

In the temperature range $T_d \le T \le T_q$ we have only quadrupolar ordering and it follows that $\mathcal{O}_1 = \mathcal{O}_3 = 0$. Therefore, we find from Eqs. (31) that

$$e_1 + e_3 = 0\ , \quad e_1 - e_3 = 2\sqrt{5}\mathcal{O}_2\ ,$$

and (45)

$$e_2 = 0\ .$$

By inserting these values in Eq. (29) we find M_{22}

$=M_{21}=M_{23}=0$. As before, we find from Eq. (28) that $G_{\alpha 2}=0$. The remaining Green's functions are found by placing the appropriate matrix elements in Eq. (28) and we find the dynamical susceptibility for $m=1$ excitations for $T_d<T<T_q$ is

$$\chi_1(k,\omega)=\frac{2A\omega_o}{\omega^2-\omega_o^2}=A\left(\frac{1}{\omega-\omega_o}-\frac{1}{\omega+\omega_o}\right), \tag{46}$$

where

$$A=3\mathcal{O}_2\left(\frac{1-\gamma_k}{1-\frac{3}{5}\alpha\gamma_k}\right)^{1/2}$$

and

$$\omega_o=\mathcal{O}_2 z\Gamma^{(2)}[(1-\gamma_k)(1-\tfrac{3}{5}\alpha\gamma_k)]^{1/2}. \tag{47}$$

At $T=T_d^+$, $\mathcal{O}_2\cong\frac{1}{2}$ and these expressions agree with those found in Eq. (44) for $T=T_d^-$; i.e., the spectra do not have any discontinuities at T_d. The variation of $\mathcal{O}_2$ with temperature for spin $\frac{3}{2}$ has been given by Sivardière and Blume[3] and we see from Fig. 1 that as T approaches T_q the quadrupolar order parameter goes to zero continuously. Therefore, both the amplitude A and frequency ω_o go to zero as we approach T_q. The variation of the amplitude A is shown in Fig. 2. The excitation spectrum $\omega_0(k)$ is of the form shown in Fig. 3(d) for $T_d<T<T_q$. Because the bandwidth of the curve is proportional to $\mathcal{O}_2$ [see Eq. (47)] this goes to zero as we approach the second-order quadrupolar phase transition at T_q.

As dipolar pair interactions increase and become comparable in size to the quadrupolar interactions, the two phase transitions approach one another,[3] and finally there is but one transition to an ordered phase for which all three order parameters are nonzero. When α is not small the approximations for the order parameters, Eqs. (32), (33), and (34), are no longer valid. In particular, e_2 and thus M_{21}, M_{23} will be nonzero so that the excitation $X_{1/2,-1/2}$ mixes with the other two, and the dynamical susceptibility Eq. (13) involves a cubic equation in the frequency ω. The partial-fraction decomposition of this susceptibility into simple poles is difficult and we will not present the cumbersome expressions.

Finally, in the limit of only dipolar interactions, i.e., $\Gamma^{(2)}=0$, all three order parameters are nonzero. The three $m=1$ excitations mix, and the decomposition of the dynamical susceptibility in partial fractions, i.e., simple poles, leads to a very cumbersome expression. The *low-lying* $m=1$ excitations for general spin s have been previously studied by Tahir-Kheli, ter Haar, and Callen.[14]

B. $m=2$

In the same way we determined the $m=1$ excitation spectra for our system, Eq. (1), we have also found these spectra for $m=2$ excitations. The susceptibility we calculate from Eqs. (3) and (5) is given as

$$\chi_2(k,\omega)=-i\int_{-\infty}^{\infty}e^{i\omega t}\,dt\langle[S^{-2}(k,t),S^{+2}(k,0)]\rangle\Theta(t),$$

where

$$S^{+2}(k)=\sum_i e^{i\vec{k}\cdot\vec{r}_i}[S^+(i)]^2,$$

$$[S^+(i)]^2=2\sqrt{3}[X_{1/2,-3/2}(i)+X_{3/2,-1/2}(i)]$$

$$=2\sqrt{3}\sum_\alpha X_\alpha(i), \tag{48}$$

and now $\alpha\equiv(\mu,\nu)=1,2$; $1\equiv(\frac{1}{2},-\frac{3}{2})$; $2\equiv(\frac{3}{2},-\frac{1}{2})$. In terms of the Green's functions Eq. (7) this susceptibility is given as

$$\chi_2(k,\omega)=12\sum_{\alpha\beta}G_{\alpha\beta}(k,\omega), \tag{49}$$

where $G_{\alpha\beta}$ is given by Eq. (14) and α, β are given above. By using Eqs. (26)–(28) and by decomposing our result into partial fractions, we find the susceptibility for $m=2$ excitations is

$$\chi_2(k,\omega)=\left(\frac{A_+(k)}{\omega-\omega_+}+\frac{A_-(k)}{\omega-\omega_-}\right), \tag{50}$$

where

$$A_\pm=-6e\left(1\pm\frac{\frac{1}{4}e\gamma_k+\tilde{e}\mathcal{O}_2}{[(\frac{1}{4}e\gamma_k)^2+(\mathcal{O}_2)^2(1-\gamma_k)]^{1/2}}\right), \tag{51}$$

$$\omega_\pm=z\Gamma^{(2)}\{\tfrac{1}{4}e\gamma_k-2\alpha\mathcal{O}_1'\pm[(\tfrac{1}{4}e\gamma_k)^2+(\mathcal{O}_2)^2(1-\gamma_k)]^{1/2}\},$$

$$e\equiv 4\mathcal{O}_1'-2\mathcal{O}_3', \quad \tilde{e}\equiv -2\mathcal{O}_2/e, \tag{52}$$

and

$$\mathcal{O}_l'\equiv\tfrac{1}{5}\sqrt{5}\,\mathcal{O}_l.$$

The spectral weight function for the $m=2$ excitations follows from its definition Eq. (2) in terms of the above susceptibility,

$$A_2(k,\omega)=A_+\delta(\omega-\omega_+)+A_-\delta(\omega-\omega_-). \tag{53}$$

We readily find that the sum rule for this function, i.e.,

$$\int_{-\infty}^{\infty}A_2(k,\omega)\,d\omega=12\sum_{\alpha\beta}\langle[X_\alpha^\dagger,X_\beta]\rangle=-12e, \tag{54}$$

is satisfied by the amplitudes given by Eq. (51).

In the limit of no quadrupole interactions $\Gamma^{(2)}=0$, we find the $m=2$ excitation spectrum is dispersionless,

$$\omega_\pm=-2\mathcal{O}_1' z\Gamma^{(1)}$$

and

$$A_\pm(k)=-6e. \tag{55}$$

The lack of dispersion in this limit is due to our decoupling approximation. Where we include the contributions of two $m=1$ excitations at different sites to the susceptibility Eq. (49), we find that the energies Eq. (55) are complex and depend on the wave vector k. If we set the dipolar coupling equal to zero, we find

$$\omega_\pm = \pm \mathcal{O}_2 z \Gamma^{(2)} (1-\gamma_k)^{1/2} \text{.} \tag{56}$$

This agrees with the results obtained previously.[4,5] We note for spin-$\frac{3}{2}$ pure quadrupolar systems that the $m=2$, $k=0$ mode is a Goldstone mode, i.e., the spectrum is gapless. The amplitudes when $\Gamma^{(1)}=0$ are

$$A_\pm(k) = \pm 12 \mathcal{O}_2/(1-\gamma_k)^{1/2} , \tag{57}$$

and we note as k tends to zero that this amplitude becomes infinite. This behavior is to be expected for a Goldstone mode.

In the case when both dipolar and quadrupolar pair interactions are present, we find the $m=2$ excitation spectra have gaps as long as there is dipolar order, i.e., $\mathcal{O}_1 \neq 0$. At $T=0$ °K, the expectation values e and $\tilde{e}$ [see Eq. (52)] in the ground state $|-\frac{3}{2}, -\frac{3}{2}, \ldots\rangle$ are $e=-1$ and $\tilde{e}=1$, so that Eqs. (50) and (51) become

$$A_\pm = \begin{cases} 12 \\ 0 \end{cases}$$

and (58)

$$\omega_\pm = \begin{cases} \frac{1}{2} z \Gamma^{(2)} (1-\gamma_k + \frac{6}{5}\alpha) \\ -\frac{1}{2} z \Gamma^{(2)} (1-\frac{6}{5}\alpha) \text{ .} \end{cases}$$

We note that one of the spectra goes as k^2 in the long-wavelength limit and that they are *not* gapless. The excitation corresponding to A_-, $h\omega_-$ is not observed at zero temperature, as it corresponds to a $m=2$ deexcitation from an excited state.

For finite temperatures e goes from -1 to zero as one approaches the dipole phase transition at $T=T_d$, and the amplitude $A_-(k)$ increases in magnitude. If α is small then $T_d < T_q$ and one finds for $T_d \leq T < T_q$ that the amplitudes are the same as for $\Gamma^{(1)}=0$, i.e., they are equal and opposite. However, the magnitude of $A(k)$ depends on the wavelength of the excitation and may be very large, i.e., as $k \to 0$, $A_\pm(k) \to \infty$. The excitation spectrum in the region $T \geq T_d$ is given by Eq. (56) and we note that it is linear in k in the long-wavelength limit. Therefore, for $m=2$ as for $m=1$ excitations, we find that the quadratic dispersion relation at zero temperature gradually goes over to a linear one for small k, as one approaches the dipole phase-transition temperature.

C. $m=3$

The spectrum for $m=3$ excitations from the ground state $|-\frac{3}{2}, -\frac{3}{2}, \ldots\rangle$ is found from the susceptibility [see Eqs. (3) and (6)]

$$\chi_3(k, \omega) = -i \int_{-\infty}^{\infty} e^{i\omega t}\, dt \langle [S^{-3}(k,t), S^{+3}(k,0)] \rangle \Theta(t) , \tag{59}$$

where

$$S^{+3} = 6X_{3/2,-3/2} \text{ .}$$

By evaluating the appropriate Green's function we find

$$\chi_3(k, \omega) = -\frac{36(3\mathcal{O}_1' + \mathcal{O}_3')}{\omega + 3\mathcal{O}_1' z \Gamma^{(1)}} \text{ .} \tag{60}$$

The spectral weight function is readily found to be

$$A_2(k, \omega) = A_o \delta(\omega - \omega_o) , \tag{61}$$

where

$$A_o = -36(3\mathcal{O}_1' + \mathcal{O}_3') \quad \text{and} \quad \omega_o = -3\mathcal{O}_1' z \Gamma^{(1)} \text{ .}$$

Since $\mathcal{O}_1'$ and $\mathcal{O}_3'$ are negative for our ground state, both the amplitude A_o and frequency ω_o are positive. We verify that this amplitude satisfies the sum rule

$$\begin{aligned} \int_{-\infty}^{\infty} A_3(k, \omega)\, d\omega &= 36 \langle [X^\dagger_{3/2,-3/2}, X_{3/2,-3/2}] \rangle \\ &= -36(3\mathcal{O}_1' + \mathcal{O}_3') \text{ .} \end{aligned} \tag{62}$$

As this excitation corresponds to the transition $X_{3/2,-3/2}$ we find that its amplitude and frequency go to zero as T approaches T_d. If α is small so that $T_d < T_q$, then A_3 is zero in the quadrupolar phase, i.e., for $T_d \leq T \leq T_q$, as well as for $T \geq T_q$.

To arrive at these results we have used a decoupling approximation which neglects the possibility that one can obtain $m=3$ excitations from three $m=1$ excitations at different sites or from a $m=1$ excitation at one site combining with a $m=2$ excitation at another site. For this reason, we find that the $m=3$ excitation spectrum is flat. If one considers these two- and three-step processes in calculating the susceptibility Eq. (60) one finds the spectrum is k dependent and that the energies are complex, i.e., the $m=3$ excitations have finite lifetimes.

IV. DISCUSSION OF RESULTS

We have used a random-phase approximation to decouple the equations of motion of the Green's functions. By using this approximation we neglected multiple excitation processes which are responsible for the finite lifetime of these excitations and in some cases for their dispersion. When one includes these processes one finds that the δ functions in the spectral-weight functions are broadened. In the case of $m=2$ and 3 excitations, one finds these functions spread over a whole continuum of energies,[2] i.e., two and three spin-wave bands. The random-phase approximation is most drastic for these excitations as it neglects multi-excitation processes.

In spite of these shortcomings inherent in the random-phase approximation, we have found a complete qualitative description of the variation of the excitation spectra with temperature from $T = 0$ °K through the dipole ordering temperature T_d to the quadrupole ordering temperature T_q. The excitations given by $A_+(k)$ and ω_+ are predominantly $X_{\mu,-3/2}$ excitations from the ground state, with $\mu = m - \frac{3}{2}$; these travel through the crystal with wave vector k and energy $\hbar\omega_+(k)$. The excitations given by $A_-(k)$ and ω_- are from excited states of the system μ to other states ν with $\mu - \nu = m$. When the dipolar pair interaction is small compared to the quadrupolar pair interactions, the single-ion picture of the level scheme is such that the levels $|\frac{3}{2}\rangle$ and $|-\frac{1}{2}\rangle$ lie below the level $|\frac{1}{2}\rangle$, with $|-\frac{3}{2}\rangle$ lying lowest of all.[3] Therefore, in these cases the excitations given by $A_-(k)$ and $\omega_-(k)$ correspond to energy deexcitations, i.e., $\omega_-(k)$ is negative, even though m is still positive. These excitations are observed as inelastic scattering events in which neutrons gain energy $\hbar|\omega_-|$ rather than lose it.

The $m = 1$ excitations can be measured on real systems by inelastic neutron scattering experiments. The scattering cross sections are related to the transition probability of flipping a neutron's spin[8] and this in turn is related to the spectral-weight function as follows[13]:

$$P_{\alpha\beta}(k, \omega) \sim \frac{A_1^{\beta\alpha}(k, \omega)}{1 - e^{-\omega/kT}} \tag{63}$$

where $\alpha\beta = \pm$ is spin up or down of the neutron and $A_1^{-+}(k, \omega)$ corresponds to the spectral-weight function we have calculated, Eq. (40). Since we assumed our ground state was $|-s, -s, \ldots\rangle$, one has to scatter neutrons with spin up (+) off the system to produce an $m = +1$ excitation at $T = 0$ °K. From Eq. (63) we note that at $T = 0$ °K, the transition for the $m = 1$ excitation corresponding to $\omega_-(k) < 0$ is not seen *even if* $A_-(k, \omega) \neq 0$, because the term $e^{-\omega_-/kT}$ in the denominator is infinite. At finite temperatures, neutrons with down spin as well as up spin are inelastically scattered from *thermally* excited states.

The $m = 2$ excitations can also be measured from inelastic neutron scattering experiments. However, it is the spin-*independent* part of the cross section that one should look at. Through their mass-interaction term neutrons create phonons which, through the orbit-lattice interaction, create $m = 2$ quadrupolar excitations.[15] Thus, one should repeat our calculations with the orbit-lattice coupling included to determine the $m = 2$ susceptibility which yields the proper spectral-weight function for the spin-*independent* neutron scattering cross section. The $m = 3$ excitations are still harder to observe.

In real systems the pair interactions are not isotropic. However, from previous work[2,5] it is not difficult to determine how the spectra we have found are changed when one considers the effects of anisotropy. Finally, we have presented results for a system with spin $\frac{3}{2}$ with only dipolar and quadrupolar pair interactions. The presence of still higher-degree pair interactions can alter our results. For example, if the octopolar pair interaction is sufficiently large, one finds that the system undergoes a transition from a paramagnetic phase to an ordered phase in which there is no dipolar or quadrupolar order but only octopolar order.[7]

Also, we have specifically considered the ratio of dipolar to quadrupolar interactions so that both phase transitions are second order. However, at first-order phase transitions the excitation spectra and the spectral weights change discontinuously. This is readily seen as these quantities depend on the order parameters $\mathcal{O}_l$ which are discontinuous at first-order phase transitions. For spin $\frac{3}{2}$ it is possible that the dipolar phase transition is first order.[3] For spin greater than $\frac{3}{2}$, both dipolar and quadrupolar phase transitions can be first-order.[3]

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Note added in proof. For large-momentum transfer the magnetic dipole-dipole interaction between neutrons and magnetic ions is capable of producing excitations with $m > 1$.[16] Therefore we do not have to go through the two-step process of first creating phonons and then using the orbit-lattice interaction to create $m > 1$ excitations. We are indebted to Professor G. T. Trammell for pointing this out to us.

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(a) See Ref. 1 p. 109, Eq. (5-30) and ensuing relations.

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[9] The last two terms in Eq. (21) arise when we write

$$X_{\nu'\mu'}(k')\,X_{\mu''\nu''}(k-k')$$
$$=\sum_i\sum_j e^{i\vec{k}'\cdot\vec{r}_i+i(\vec{k}-\vec{k}')\cdot\vec{r}_j}X_{\nu'\mu'}(i)\,X_{\mu''\nu''}(j)$$

and consider the case $i=j$. Then we find by using the commutator relation Eq. (9) that

$$X_{\nu'\mu'}(k')X_{\mu''\nu''}(k-k')\approx X_{\nu'\nu''}(k)\,\delta_{\mu'\mu''}-X_{\mu''\mu'}(k)\,\delta_{\nu'\nu''}\,.$$

As these terms do not depend on k', they do not contribute to the equation of motion for the Green's function Eq. (16). That is to say, when we place these terms in Eq. (16) and sum over k' we find they vanish because

$$\sum_{k'}\Gamma^{(l)}(k')\propto\Gamma^{(l)}(r_i-r_j=0)=0.$$

[10] See Ref. 6, Eq. (5.12).

[11] This result follows from the definition of the octopole operator for $s=\frac{3}{2}$, $\mathcal{O}_0^3=(1/3\sqrt{5})\,\{5s_z^3-[3s(s+1)-1]s_z\}$ and from the mean-field results of Sivardière and Blume (see Ref. 3). Equation (32) immediately follows when (i) we neglect the terms proportional to $e^{-2\beta KQ}$ in the expression for $\mathcal{O}_3$ and (ii) we recognize that $\mathcal{O}_1=(1/\sqrt{5})M$, where M is given by Sivardière and Blume; see Ref. 3, Eq. (18) and discussion on p. 1128.

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