orbital and the single-site models of a dilute alloy.

[13]For a discussion of this point, see W. A. Harrison, *Solid State Theory* (McGraw-Hill, New York, 1970), p. 185.

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Spin-Wave Theory of Two-Magnon Raman Scattering in a Two-Dimensional Antiferromagnet*

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The graphical spin-wave approach to two-magnon Raman scattering which was developed in an earlier paper is applied to a study of the two-dimensional Heisenberg antiferromagnet K_2NiF_4. In the present treatment the one-magnon energies are taken to be renormalized in the Hartree-Fock approximation, with the two-magnon Green's function evaluated in the "ladder" approximation. It is found that the Hartree-Fock renormalization gives very good agreement with the experimental results for the temperature shift of the Raman peak up to the measured Néel temperature $T_N = 97.1$ °K, and yields satisfactory agreement above T_N up to the maximum temperatures for which Raman data are available. As in the three-dimensional case, a satisfactory explanation for the observed thermal broadening of the spectra remains to be given. Comparisons between the results for K_2NiF_4 and its three-dimensional analog $KNiF_3$ are made. In particular it is found that the renormalization of the zone-edge magnons for $T \simeq T_N$ is much less marked in the two-dimensional case. As has been discussed by other authors, this indicates a rather different temperature dependence of the "coherence length" for the two-dimensional system.

I. INTRODUCTION

In an earlier paper[1] (hereafter referred to as I), we presented a spin-wave approach to two-magnon Raman scattering and applied it to a study of simple three-dimensional antiferromagnetic systems (e.g., $KNiF_3$). The theory was based on the Dyson-Maleev boson representation of the spin operators, and proceeded through the application of the finite-temperature graphical perturbation theory. In this paper we present results of calculations based on the theory developed in I, but applied to the interesting case of a simple two-dimensional antiferromagnet. Probably the most widely studied example of such a system is the compound K_2NiF_4, which we shall consider in particular in this paper. At various points throughout the paper, we shall, for convenience, use the abbreviations [2] or [3] to stand for two- or three-dimensional systems.

Perhaps the most interesting theoretical question associated with the [2] systems involves the question of long-range ordering. Mermin and Wagner[2] have applied the Bogoliubov inequality to provide a rigorous proof that there can be no long-range order in a [2] system described by the Heisenberg exchange Hamiltonian for $T > 0$ in the absence of anisotropy. This instability is also suggested from simple spin-wave theory using a standard argument which we recall briefly here. Consider an isotropic [2] ferromagnet with a spin-wave branch having energy $\Omega_{\vec{k}} \propto k^2$ for $\vec{k} \to 0$. Calculation of the magnetization involves an integral of the form

$$\int k\,dk \frac{1}{e^{\Omega_{\vec{k}}/k_BT} - 1} ,$$

which diverges logarithmically in the neighborhood of $\vec{k} \to 0$. Next consider the case of an isotropic [2] antiferromagnet with a spin-wave branch having $\Omega_{\vec{k}} \propto k$ as $\vec{k} \to 0$. Here calculation of the sublattice magnetization involves an integral

$$\int \frac{k\,dk}{\Omega_k} \frac{1}{e^{\Omega_{\vec{k}}/k_BT} - 1} ,$$

where the extra factor of Ω_k in the denominator arises from the Bogoliubov transformation which diagonalizes the spin-wave Hamiltonian. Again one obtains a logarithmic divergence from the region $\vec{k} \to 0$. These considerations show that the question of long-range order in [2] depends sensitively on the behavior of the long-wavelength magnons. Also, since a small amount of anisotropy is sufficient to remove the singularity at $\vec{k} \to 0$, the ordering problem (at least in the simple spin-wave approximation[3]) involves a careful treatment of anisotropy effects.

In contrast to the discussion above, we now consider the problem of two-magnon Raman scattering in a [2] antiferromagnet. Here the cross section is determined almost exclusively by the behavior of the short-wavelength (zone-edge) magnons, which should be very insensitive to small effects due to anisotropy. Also one expects the zone-edge magnons to reflect the properties of short-range order in the system, and this can persist well above the

long-range-ordering temperature T_N.

In the lowest-order approximation discussed in I, the one-magnon energies are renormalized by summing the Hartree-Fock self-energy graphs. This yields a result equivalent to that which was first obtained by Bloch[4] from a variational calculation, and which may also be obtained using the method of Keffer and Loudon.[5,6] The renormalized energies in this approximation are given by

$$\Omega_{\vec{k}}(T) = \Omega_{\vec{k}}\alpha(T) ,$$

where

$$\alpha(T) = 1 - \frac{1}{JzS^2}\frac{1}{N}\sum_{\vec{q}}\frac{\Omega_{\vec{q}}}{e^{\Omega_{\vec{q}}\alpha(T)/k_BT}-1} .$$

It should be noted that the integral above is perfectly regular in the region $\vec{q}\to 0$, and will therefore be insensitive to small anisotropy effects. In this paper we show that the above Hartree-Fock theory, applied to the [2] system K_2NiF_4, yields a rather good description of the shift of the Raman peak to lower energies with increasing temperature. As in the [3] case, however, a satisfactory explanation for the observed thermal broadening of the spectrum remains to be given. Nevertheless, the present theory permits some interesting quantitative comparisons to be made between the [2] and [3] systems. In the [3] case of $KNiF_3$, the theory predicts a downward shift of the Raman peak of the order 40–50% for temperatures $T \simeq T_N$. In the [2] case of K_2NiF_4, the theory yields a shift of only 5% near T_N. Both of these results are consistent with the experimental data.

Another interesting point to be made is the following. In the [3] case, $\alpha(T)$ fails to have a solution at a temperature which is close to various theoretical determinations of T_N. In this case, the Hartree-Fock theory permits one to study the Raman shift only up to temperatures $T \simeq T_N$. On the other hand, in the [2] case there can be no obvious connection between the temperature T_α where $\alpha(T)$ fails to have a solution, and the ordering temperature T_N. This is clear because a solution for $\alpha(T)$ exists in the absence of anisotropy, whereas a calculation of the sublattice magnetization diverges in this case. It turns out, in fact, for our particular example of K_2NiF_4, that $\alpha(T)$ fails to have a solution at a temperature which is approximately twice the measured Néel temperature. Thus we are able to compute Raman shifts up to temperatures of the order $2T_N$ for this [2] system.

It is perhaps worth remarking here that the question of actually determining T_N theoretically for [2] systems is a subtle one. Stanley and Kaplan[7] have employed high-temperature expansion methods to infer the possibility of a phase transition (in the absence of anisotropy) to a state lacking long-range order, but in which the spin correlations may have a much longer range below T_c than above. Their formula

$$k_B T_c = \tfrac{1}{10} J(z-1)[2S(S+1)-1] ,$$

applied to the case of K_2NiF_4 [$S=1$, $z=4$, $(J/k_B) = 112\,^\circ\mathrm{K}$], predicts a T_c quite close to the measured Néel temperature, $T_N = 97.1\,^\circ\mathrm{K}$. Lines[8] has recently considered the problem (for the ferromagnetic case) using a new Green's-function decoupling scheme. For the isotropic system he finds two critical temperatures, $T_c^{(2)}$, a Stanley-Kaplan temperature corresponding to a transition to a state with zero magnetization and infinite susceptibility, while $T_c^{(1)} = T_N = 0\,^\circ\mathrm{K}$ is the long-range-ordering temperature. However, for small but nonvanishing anisotropy, Lines finds that $T_c^{(1)}$ and $T_c^{(2)}$ will coincide. It is also pertinent to remark here that Lines's wave-vector-dependent decoupling scheme, applied to the two-magnon scattering problem, should lead to results similar to those we present in this paper. That is, for small $\vec{k}$ magnons, Lines uses the usual Tyablikov[9] or random-phase-approximation (RPA) decoupling which makes the magnon energies renormalize with the magnetization (or sublattice magnetization in the antiferromagnetic case). However, for the large $\vec{k}$ magnons, which are important in two-magnon Raman scattering, he chooses the Keffer-Loudon renormalization, and this is equivalent to our Hartree-Fock renormalization scheme.

In the remainder of the paper we take over, with minor modifications, the theory developed in I, and we apply it to the [2] antiferromagnet K_2NiF_4. In addition to Raman scattering studies,[10,11] this compound has been the subject of recent neutron-scattering,[12,13] and nuclear-magnetic-resonance[14] measurements. The actual modifications of the theory necessary to discuss the [2] problem are slight. First, because we wish to investigate possible effects due to anisotropy, we generalize the Hamiltonian considered previously to include a small dipolar anisotropy contribution. Second, the Oguchi corrections[15] (which arise from normal ordering the two-body-interaction terms) are not so negligible in the [2] case as they were in the [3] case, and these are included in the present analysis. With the above modifications, the theory presented in I may essentially be taken over directly, and the reader is referred to the earlier paper for further details.

II. HARTREE-FOCK THEORY

The basic Hamiltonian we work with is written

$$H = J\sum_{j,\delta}\vec{S}_{j,a}\cdot\vec{S}_{j+\delta,b} - g\mu_B H_A\left(\sum_j S^z_{j,a} - \sum_{j+\delta} S^z_{j+\delta,b}\right) . \tag{1}$$

The first term in Eq. (1) is the usual Heisenberg ex-

change term, with nearest-neighbor interactions assumed only within the plane of a quadratic layer structure. In the subsequent analysis we shall be concerned with the K_2NiF_4 quadratic layer structure, where $z=4$ is the number of nearest-neighbor b sites of a spin at site j, a. The anisotropy contribution as given by the second term in Eq. (1) is expressed in a form assumed to arise from an effective field H_A, which we assume to be proportional to $\langle S_a^z \rangle$, the sublattice magnetization. In reduced units, we define an anisotropy parameter Δ, given by

$$\Delta = \frac{g\mu_B H_A}{SJz} = \frac{\langle S_a^z \rangle}{\langle S_a^z \rangle_0} \Delta_0 , \tag{2}$$

where the zero subscripts refer to zero temperature. The above form[16] for the anisotropy should be valid for dipolar anisotropy, but not for anisotropy arising from single-ion contributions. For the case of K_2NiF_4 we can justifiably assume the condition $\Delta_0 \ll 1$.

Following the same approximate analysis discussed in I, but including the effects of Δ and the Oguchi corrections (from normal ordering the two-body-interaction terms), we obtain the Hamiltonian

$$\tilde{\mathcal{H}} \simeq E_0 + \mathcal{H}_0 + V . \tag{3}$$

The first term in Eq. (3) is a constant contributing to the ground-state energy and given by

$$E_0 = -NJzS\left[(S+1) + \Delta(2S+1) + \frac{C^- C^+}{4S}\right] , \tag{4}$$

with

$$C^{\pm} = \frac{1}{N}\sum_{\vec{k}} \left(1 \pm \frac{(1+\Delta) - \gamma(\vec{k})^2}{[(1+\Delta)^2 - \gamma(\vec{k})^2]^{1/2}}\right) \tag{5}$$

and

$$\gamma(\vec{k}) = \tfrac{1}{2}(\cos k_x a + \cos k_y a) , \tag{6}$$

and all sums go over a [2] Brillouin zone containing N unit cells. Next, $\mathcal{H}_0$ contains the one-magnon spin-wave contributions and is given by

$$\mathcal{H}_0 = \sum_{\vec{k}} \Omega_{\vec{k}}(0)(\alpha_{\vec{k}}^\dagger \alpha_{\vec{k}} + \beta_{\vec{k}}^\dagger \beta_{\vec{k}} + 1) , \tag{7}$$

where

$$\Omega_{\vec{k}}(0) = \Omega_{\vec{k}}\left(1 + \frac{C^-}{2S}\right) - \frac{(SJz)^2}{\Omega_{\vec{k}}} \Delta(1+\Delta)\frac{C^-}{2S} \tag{8}$$

are the spin-wave energies at zero temperature. In Eq. (8), $\Omega_{\vec{k}}$ is the spin-wave energy which obtains if the Oguchi corrections ($C^{\pm}$) are ignored, i.e.,

$$\Omega_{\vec{k}} = SJz\,[(1+\Delta)^2 - \gamma(\vec{k})^2]^{1/2} . \tag{9}$$

Finally, V is given by Eq. (18) of I except that the u's and v's arising from the Bogoliubov transformation are now determined from

$$u_{\vec{k}}^2 + v_{\vec{k}}^2 = (1+\Delta)\left(\frac{SJz}{\Omega_{\vec{k}}}\right) , \tag{10}$$

$$u_{\vec{k}} v_{\vec{k}} = -\frac{1}{2}\gamma(\vec{k})\left(\frac{SJz}{\Omega_{\vec{k}}}\right) . \tag{11}$$

Turning now to the Hartree-Fock self-energy renormalization, we obtain, in the present theory, the following results for the one-magnon energies:

$$\Omega_{\vec{k}}(T) = \alpha(T)\Omega_{\vec{k}} + \frac{(SJz)^2}{\Omega_{\vec{k}}} \Delta(1+\Delta)[1-\alpha(T)] , \tag{12}$$

where $\alpha(T)$ satisfies the implicit equation

$$\alpha(T) = \alpha(0) - \frac{1}{S^2 Jz}\frac{1}{N}\sum_{\vec{q}} \left(\Omega_{\vec{q}} - \frac{(SJz)^2}{\Omega_{\vec{q}}} \Delta(1+\Delta)\right) N(\Omega_{\vec{q}}(T)) . \tag{13}$$

In the equation above

$$\alpha(0) = 1 + C^-/2S , \tag{14}$$

and $N(\Omega)$ denotes the Bose function. If the anisotropy parameter Δ is set equal to zero, the above equations become identical to those derived by Bloch.[4] Also it appears that Eqs. (12)–(14) are identical to those which have been discussed by Keffer,[17] with the identification of symbols $\alpha(T) = 1 - l(T)$. Thus the Hartree-Fock theory is equivalent to the heuristic method of Keffer and Loudon.[5]

As pointed out in the Introduction, the integral which appears in Eq. (13) is perfectly well behaved near $\vec{q} \to 0$ (with or without a finite Δ), and so the renormalized energies will be relatively insensitive to the effects of a finite anisotropy. In contrast to this, the sublattice magnetization, in the Hartree-Fock approximation, is given by

$$\langle S_a^z \rangle = \langle S_a^z \rangle_0 - (1+\Delta)\frac{1}{N}\sum_{\vec{k}} \left(\frac{SJz}{\Omega_{\vec{k}}}\right) N(\Omega_{\vec{k}}(T)) , \tag{15}$$

with

$$\langle S_a^z \rangle_0 = S(1 - C''/2S) \tag{16}$$

and

$$C'' = \frac{1}{N}\sum_{\vec{k}} \left((1+\Delta)\frac{SJz}{\Omega_{\vec{k}}} - 1\right) . \tag{17}$$

It should be noted that the integral occurring in Eq. (15) will diverge logarithmically as $\Delta \to 0$. In the present theory, with the anisotropy inserted in the form given by Eq. (2), one cannot therefore compute an actual Néel temperature, i.e., a temperature $T_N > 0$ such that $\langle S_a^z \rangle_{T_N} = 0$. We can, however, obtain a maximum temperature T_M, above which Eq. (15) fails to have a solution. This temperature lies midway between the measured Néel temperature T_N and the maximum temperature T_α where Eq. (13) for $\alpha(T)$ has a solution. Probably the only conclusion one can reach from this is that the Hartree-Fock theory does not adequately treat

the renormalization of long-wavelength magnons, and that some $\vec{k}$-dependent renormalization scheme is necessary to treat this problem. Nevertheless, as we shall see in the discussion which follows, the Hartree-Fock theory appears to give quite reasonable results for the renormalization of the zone-edge magnons involved in the two-magnon Raman process.

III. RAMAN CROSS SECTION

For the present case of the [2] quadratic-layer structure, we take the following transition operator for the two-magnon Raman scattering process:

$$M=\sum_{j,\delta} B\left(\tfrac{1}{2}\vec{\epsilon}_i\cdot\vec{\epsilon}_f-\frac{1}{\delta^2}(\vec{\delta}\cdot\vec{\epsilon}_i)(\vec{\delta}\cdot\vec{\epsilon}_f)\right)\vec{S}_{j,a}\cdot\vec{S}_{j+\delta,b}\ . \tag{18}$$

In this equation $\vec{\epsilon}_i$ and $\vec{\epsilon}_f$ are the unit polarization vectors for the incident and scattered electric fields, respectively. The vectors $\vec{\delta}$ lying in the plane connect nearest-neighbor spins on opposite sublattices. In this case, only the planar components of $\vec{\epsilon}_i$ and $\vec{\epsilon}_f$ contribute to the scattering process. Additional polarization factors, including out-of-plane components, commute with the spin Hamiltonian and cause no inelastic scattering.

In I we obtained an approximate Raman cross-section formula in terms of renormalized one-magnon propagators and a vertex function which satisfies a general Bethe-Salpeter equation. In the lowest order of approximation we take the one-magnon energies to be renormalized in the Hartree-Fock approximation as discussed in Sec. II, and we take the vertex function to satisfy the "ladder" approximation Bethe-Salpeter equation. The result of this calculation, applied to the [2] antiferromagnet K_2NiF_4, is summarized in the equations which follow. The cross section is proportional to

$$\bar{G}(\hbar\omega)=\frac{\mathrm{Im}G(\xi_1=\hbar\omega+i0^+)}{\pi[1-e^{-\hbar\omega/k_BT}]}\ , \tag{19}$$

where $\omega=\omega_i-\omega_f$ is the frequency shift of the scattered light. Also in Eq. (19),

$$G(\xi_1)=G^{(+)}(\xi_1)+G^{(+)}(-\xi_1)\ , \tag{20}$$

where we find

$$G^{(+)}(\xi_1)=g(\vec{\epsilon}_i,\vec{\epsilon}_f)NS^2B^2\times\left(\frac{L^{(2)}+\frac{1}{2}J(L^{(1)}L^{(1)}-L^{(2)}L^{(0)})}{1-\frac{1}{2}J(L^{(0)}+L^{(2)})-\frac{1}{4}J^2(L^{(1)}L^{(1)}-L^{(2)}L^{(0)})}\right)\ . \tag{21}$$

The factor $g(\vec{\epsilon}_i,\vec{\epsilon}_f)$ contains the polarization dependence for the scattering cross section, and is given in the [2] case by the expression

$$g(\vec{\epsilon}_i,\vec{\epsilon}_f)=(\epsilon_i^x\epsilon_f^x)^2+(\epsilon_i^y\epsilon_f^y)^2-2\epsilon_i^x\epsilon_f^x\epsilon_i^y\epsilon_f^y\ . \tag{22}$$

Also in Eq. (21), the $L^{(m)}$ functions are given by

$$L^{(m)}(\xi_1)=\frac{1}{N}\sum_{\vec{k}}(\cos k_xa-\cos k_ya)^2(u_{\vec{k}}^2+v_{\vec{k}}^2)^m\times\frac{2N(\Omega_{\vec{k}}(T))+1}{2\Omega_{\vec{k}}(T)-\xi_1}\ , \tag{23}$$

where the $\Omega_{\vec{k}}(T)$'s are the Hartree-Fock spin-wave energies, and the sum over $\vec{k}$ is over the [2] Brillouin zone. It is straighforward to check that the integrals (for $m\leq 2$) appearing in Eq. (23) are all well behaved near $\vec{k}\to 0$, and thus the Raman cross section will be insensitive to small anisotropy.

In order to actually compute spectra from the formulas above, we rewrite Eq. (23) in the equivalent form (as $2\Gamma\to 0^+$)

$$L^{(m)}(\hbar\omega+i2\Gamma)=\int_0^{2\Omega_{\max}}dE'\left[(1+\Delta)\left(\frac{2SJz}{E'}\right)\right]^m\frac{2N\{\alpha E'/2+[2(SJz)^2/E']\Delta(1+\Delta)(1-\alpha)\}+1}{\alpha E'+[(2SJz)^2/E']\Delta(1+\Delta)(1-\alpha)-\hbar\omega-i2\Gamma}\times\left\{\frac{2E'}{\pi}\,\mathrm{Im}\frac{1}{N}\sum_{\vec{k}}\frac{(\cos k_xa-\cos k_ya)^2}{E'^2-4\Omega_{\vec{k}}^2-i0^+}\right\}\ , \tag{24}$$

where $\alpha=\alpha(T)$, $\Omega_{\max}=(\Omega_{\vec{k}})_{\max}$, and 2Γ is chosen to be small enough to yield results independent of Γ as $\Gamma\to 0^+$. The last factor (in curly brackets) in Eq. (24) is essentially a density-of-states function which can be expressed in terms of the (tabulated) associated Legendre function. The remaining E' integration was then performed numerically on a computer. In the actual computations we chose $2\Gamma=0.0005$ and employed a Gaussian quadrature routine with a net estimated accuracy of $\frac{1}{2}\%$.

It should be noted, in all the equations where $\Delta\propto\langle S_a^z\rangle$ appears as a parameter, that in principle it is necessary to carry out a self-consistent calculation using Eq. (15) for $\langle S_a^z\rangle$. In practice, however, with $\Delta_0\ll 1$, this is only important for the calculation of $\langle S_a^z\rangle$ itself, because $\alpha(T)$ is very nearly independent of Δ for small values of Δ_0. Thus the Raman cross-section calculations can be carried out by ignoring Δ altogether, and this is what we have done for $T>T_M$. Also, as in the [3] case, we have performed calculations to verify that the cross-section formula is very insensitive to the approximations $L^{(0)}\simeq L^{(1)}\simeq L^{(2)}$. This is again due to the sharp peaking of the density-of-states function near the zone edge. This means that, to a very good approximation, Eq. (21) may

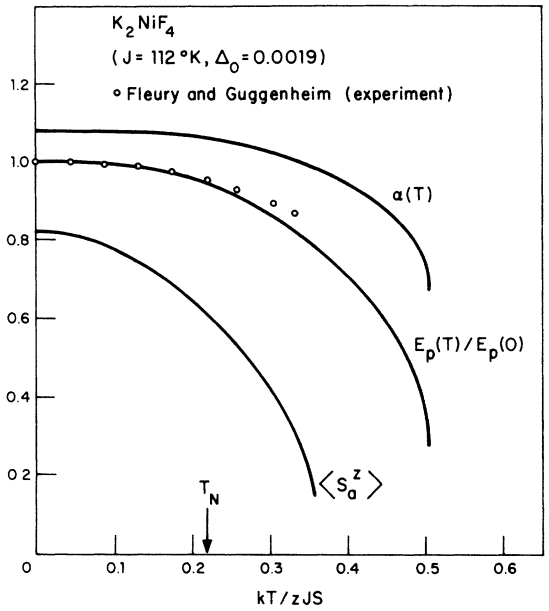

FIG. 1. Hartree-Fock renormalization parameter $\alpha(T)$, the sublattice magnetization $\langle S_a^z \rangle$, and the normalized two-magnon Raman peak position $E_P(T)/E_P(0)$ as functions of temperature in K_2NiF_4.

be approximated as

$$G^{(+)}(\xi_l) = g(\vec{\epsilon}_i, \vec{\epsilon}_f) NS^2 B^2 \frac{L^{(0)}}{1 - JL^{(0)}} . \tag{25}$$

IV. RESULTS AND DISCUSSION

In Fig. 1, along with the function $\alpha(T)$, we present a comparison of our theoretical predictions for the temperature dependence of the two-magnon Raman peak position, with the experimental data for K_2NiF_4 obtained by Fleury and Guggenheim.[10] Also in Fig. 1, we show the results of our calculation for the sublattice magnetization $\langle S_a^z \rangle$. Below T_M these calculations were carried out by solving (iteratively) Eqs. (2), (15), (12), and (13) to obtain a self-consistent result. We have chosen a value of $\Delta_0 = 0.0019$, consistent with experimental determinations[18] of the zero-temperature anisotropy. As discussed previously, $\langle S_a^z \rangle$ fails to have a solution midway between the measured Néel temperature $T_N = 97.1$ °K (close to the Stanley-Kaplan temperature) and the maximum temperature $T_\alpha \simeq 2T_N$ where $\alpha(T)$ has a solution.

The most interesting feature of the results presented in Fig. 1 is the relatively small shift of the Raman peak at the ordering temperature T_N. Both the theory and experiments are consistent with a downward shift of approximately 5% at this temperature. This should be contrasted to the [3] case of $KNiF_3$, results for which are shown in Fig. 2. Here we observe a Raman shift of the order 40–50% for temperatures near T_N. A qualitative interpretation of these observations has been discussed by Fleury and Guggenheim[10] in connection with the theory developed by Lines.[8] Assume there exists some "coherence length" L such that magnons with wavelengths $\lambda < L$ experience much less renormalization than magnons with $\lambda > L$. Then for a given T/T_N, it appears that L is much greater in [2] systems than in [3] systems. Likewise, for essentially the same reasons, it appears that the Hartree-Fock renormalization of the zone-edge magnons is valid for much larger values of T/T_N in the [2] case, although, from Fig. 1 we can claim only fair agreement in the region $T > T_N$.

Finally, in Fig. 3 we present some Raman spectra computed from the formulas of Sec. III. The zero-temperature spectrum is in good agreement with the line shape predicted by the zero-temperature Green's-function theory of Elliott and Thorpe.[11,19] While the higher-temperature spectra correctly shift toward lower energies, they suffer from the same inadequacies discussed in I for the [3] case. Instead of broadening and becoming weaker, the line shapes actually become sharper and the amplitude increases, in contradiction with the experimental results.

The increase in the computed amplitude is due primarily to the increase in the magnon occupation numbers $2N(\Omega_{\vec{k}}(T)) + 1$ and the Stokes factor $(1 - e^{-\hbar\omega/k_B T})^{-1}$ in Eqs. (19) and (23). We stated in I that Sólyom[20] has found an amplitude renormalization factor of α^2 using a different graphical technique. Subsequently Sólyom has informed us that his amplitude factor is actually $\langle S_a^z \rangle$ (although his energies renormalize proportional to α). However, Sólyom obtained the result $\langle S_a^z \rangle$ by neglecting the term $S_{j,a}^z S_{j+b,b}^z$ in the Raman-transition operator. In a future publication we will show that when

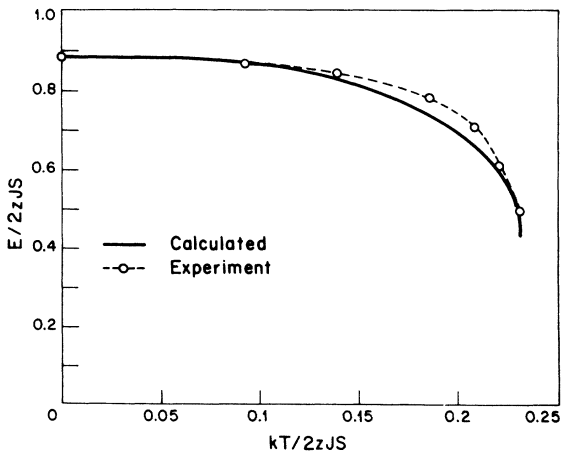

FIG. 2. Two-magnon Raman peak position as a function of temperature in $KNiF_3$. From Refs. 1 and 11.

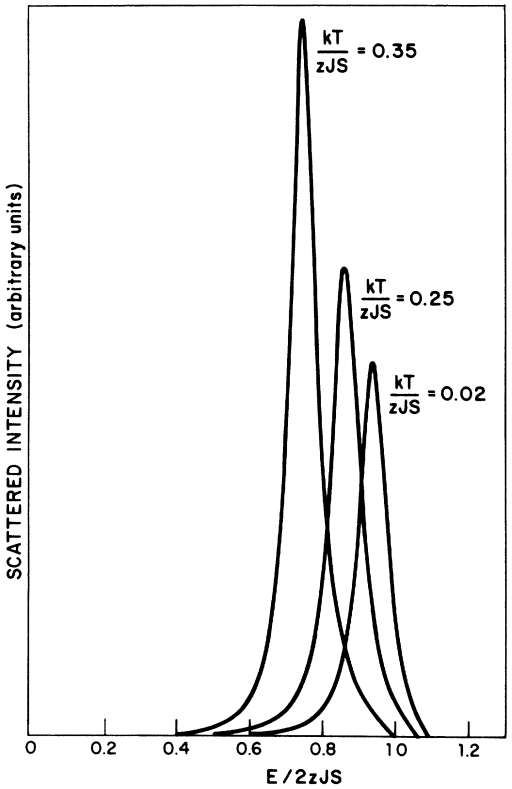

FIG. 3. Computed two-magnon Raman spectra in K_2NiF_4 for different temperatures.

the above term is included, the correct amplitude renormalization factor is indeed α^2. However, by itself, the factor of α^2 is not sufficient to offset the increase in amplitude from the Bose and Stokes factors. The addition of one-magnon damping, discussed below, can contribute significantly to the reduction in amplitude.

One possible source of the observed broadening which was considered in I, and which is often invoked in discussions[21] of line broadening, is the damping of one-magnon states due to higher-order self-energy processes. As shown in I, if one assumes a sufficiently large width Γ for a zone-edge magnon, one can obtain spectra which are in qualitative agreement with the experimental results. However, there are some difficulties with such an explanation. One of these, which can perhaps be dismissed due to the crudeness of calculation, is that we have not been able to compute Γ's which are large enough to explain the experimentally observed broadening. A second, more serious difficulty is that a value of Γ large enough to give the observed broadening produces[22] a high-energy tail extending well above the zero-temperature maximum energy. Such a tail does *not* appear to be observed experimentally. Also, because the simple Hartree-Fock theory appears to give such reasonable results for the shift of the Raman peak, one might argue from the Kramers-Kronig relations (relating the real and imaginary parts of the self-energy) that damping effects should be rather small.

As has been mentioned in I, a satisfactory explanation of the linewidth problem may involve terms in the Heisenberg or Raman Hamiltonians which have been neglected in the present treatment, or, perhaps it may involve the kinematic interaction[23,24] which has been totally ignored thus far. Given enough hard work, the neglected terms in the Hamiltonian should be amenable to calculation. However, if the kinematic interaction is important in this problem, it may be necessary to go to a scheme, such as the drone-fermion[25,26] representation, in which the effect can be included somewhat more conveniently.[27] Application of this scheme for $S > \frac{1}{2}$ is not completely straightforward,[28] however.

In conclusion, while we have shown that the Hartree-Fock renormalization scheme gives satisfactory results for the temperature shift of the two-magnon Raman peak for both [2] and [3] systems, an interesting and challenging problem remains concerning the broadening of the spectra.

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*Work sponsored by the Department of the Air Force.

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[2]N. D. Mermin and H. Wagner, Phys. Rev. Letters 17, 1133 (1966).

[3]In the simple spin-wave model, the ordering temperature varies inversely as the magnitude of the logarithm of the anisotropy. See, however, the discussion which follows concerning the recent theoretical work of Lines. His results appear to indicate that in a more sophisticated treatment, any small (but finite) anisotropy is sufficient to stabilize the long-range order at or below an *anisotropy-independent* temperature.

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Quasiclassical Equation of Motion for the Heisenberg Spin System*

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Previous results of quasiclassical spin dynamics are extended by presenting a new approximation for quantum corrections to the classical equation of motion for the Heisenberg model. The accuracy of the equation with these corrections is determined by comparing computer calculations based on this formalism with exact quantum and classical results for spin pair correlation functions for a six-spin linear chain at infinite temperatures.

I. INTRODUCTION

The dynamical behavior of the Heisenberg spin system, described by the Hamiltonian

$$H = -\sum_{i \neq i'} J_{ii'} \vec{S}_i \cdot \vec{S}_{i'} , \tag{1}$$

has received a great deal of attention. Dynamical quantities of interest are, for example, spin pair correlation functions of the form

$$\langle S_{i\alpha}(0) S_{I\beta}(t) \rangle = \frac{\mathrm{Tr} e^{-\beta H} S_{i\alpha}(0) S_{I\beta}(t)}{\mathrm{Tr} e^{-\beta H}} . \tag{2}$$

The spin pair correlation functions defined in Eq. (2) are directly related to the neutron scattering cross section.[1] Another quantity of interest which is proportional to the neutron cross section is the Fourier transform of the spin pair correlation function:

$$S^{\alpha\beta}(\vec{K}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \sum_{i,I} e^{i\vec{K}\cdot(\vec{r}_I - \vec{r}_i)} \langle S_{i\alpha}(0) S_{I\beta}(t) \rangle . \tag{3}$$

In recent years, the technique of computer simulation has been applied to the classical Heisenberg model, and several interesting results have been obtained.[2-6] It is well known that the classical approximation is valid in the large spin limit.[7] However, for finite spin j, the validity of the classical approximation is not understood very well.

In a previous paper[8] (which will be referred to as I), we have employed the Wigner formalism[9,10] to obtain quantum corrections to the classical spin dynamics. These quantum corrections derived in I can be divided into two types. The first results from approximating the thermal average as an integration over a classical distribution of the form $e^{-\beta H_{\mathrm{cl}}}$. The second is a quantum correction to the classical equation of motion.

These two corrections will be discussed separately in Secs. II and III. In Sec. IV, we present computer calculations to test the accuracy of the quasiclassical equation of motion.

II. THERMAL AVERAGE

In this section, for simplicity, we will consider only the spin-$\frac{1}{2}$ nearest-neighbor exchange model. In I, we obtained spin pair correlation functions in terms of a series in classical spin pair correlation functions. The result to second-order classical correlations is

$$\begin{aligned} \tfrac{1}{2}\langle S_{i\alpha}(0) S_{I\beta}(t) + S_{I\beta}(t) S_{i\alpha}(0) \rangle &= \langle \Omega_{i\alpha}\, \Omega_{I\beta}(t) \rangle_{\mathrm{cl}} \\ &\quad - \tfrac{3}{4}(2\beta - \beta^2 J) \sum_{i'} J_{ii'} \langle \Omega_{i'\alpha}\, \Omega_{I\beta}(t) \rangle_{\mathrm{cl}} \\ &\quad - \tfrac{3}{4}\beta^2 \sum_{i',i''} J_{ii'} J_{i'i''} \langle \Omega_{i''\alpha}\, \Omega_{I\beta}(t) \rangle_{\mathrm{cl}} + \cdots . \end{aligned} \tag{4}$$