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Interacting Magnons in the Linear Chain*

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The magnon bound-state spectrum recently observed in the anisotropic magnetic salt $CoCl_2 \cdot 2H_2O$ is investigated by means of response functions. A perturbation scheme is set up for the response functions and the transverse and longitudinal susceptibilities are evaluated to second order in the transverse anisotropy. The effects of the Heisenberg part of the exchange interaction are included to second order by solving a two-magnon and a three-magnon scattering problem. In the zero-field limit we evaluate under certain simplifying assumptions the transverse susceptibility to all orders in the transverse anisotropy.

I. INTRODUCTION

The recent observations by Torrance and Tinkham[1,2] of a magnon bound-state spectrum in the magnetic crystal $CoCl_2 \cdot 2H_2O$ have stimulated renewed theoretical interest in the dynamical properties of the linear anisotropic magnetic chain. The authors[1-3] showed that their helium temperature measurements of the absorption spectra could be interpreted in terms of the spin-$\frac{1}{2}$ Hamiltonian

$$H = -2\sum_{i=1}^{N}\left[-\tfrac{1}{2}g^{z}\mu_B H_0 S^{z} + j^{z} S_i^{z} S_{i+1}^{z} + \tfrac{1}{2}j^{\perp}(S_i^{+}S_{i+1}^{-} + \text{H. c.}) + \tfrac{1}{2}j^{a}(S_i^{+}S_{i+1}^{+} + \text{H. c.})\right], \quad (1.1)$$

which describes a single cluster of exchange-coupled Co^{++} ions. We have included a Zeeman term arising from an applied magnetic field in the z direction. The spin operators satisfy the usual commutation relations

$$[S_i^{\pm}, S_k^{z}] = \mp S_i^{\pm}\delta_{ik} \quad (1.2)$$

and

$$[S_i^{-}, S_k^{+}] = -2S_i^{z}\delta_{ik}, \quad (1.3)$$

where we have chosen units such that $\hbar = 1$. Furthermore, in the spin-$\frac{1}{2}$ case we obtain the length condition

$$S_i^{+}S_i^{-} - S_i^{z} = \tfrac{1}{2} \quad (1.4)$$

and the minimum equations

$$S_i^{+}S_i^{+} = S_i^{-}S_i^{-} = 0 \quad \text{and} \quad S_i^{z}S_i^{z} = \tfrac{1}{4}. \quad (1.5)$$

The longitudinal and transverse anisotropy of the exchange Hamiltonian (1.1) is characterized by the nearest-neighbor exchange constants j^{z}, $j^{\perp}$, and j^{a} [where $j^{\perp} = \frac{1}{2}(j^{x} + j^{y})$ and $j^{a} = \frac{1}{2}(j^{x} - j^{y})$], and by the spectroscopic splitting factors g^{z}, g^{x}, and g^{y}. The dimensionless anisotropy parameters $\sigma = j^{\perp}/j^{z}$ and $\alpha = j^{a}/j^{z}$ assume the values 0.2 and 0.08, respectively, in the case of $CoCl_2 \cdot 2H_2O$.

Owing to the strong longitudinal anisotropy and to the fact that j^{z} is of order 17 °K for $CoCl_2 \cdot 2H_2O$ the measurements by Torrance and Tinkham probe only the zero temperature properties of the system. Any temperature dependence of the excitation spectrum will be exponentially small, i.e., of order $e^{-17/T}$. Consequently, we shall in the present paper confine our attention to zero temperature.

The strong longitudinal anisotropy of the exchange Hamiltonian (1.1) suggests treating the linear chain as a one-dimensional Ising model in the lowest approximation. Such an approach was in fact carried out by Torrance and Tinkham,[1,3] who constructed Bloch functions for the localized Ising spin deviations and computed numerically the effects on the excitation spectrum due to $j^{\perp}$ and j^{a} by means of ordinary perturbation theory.

The excitation spectrum of the Ising model (i.e., $j^{\perp}=j^{a}=0$) is conveniently discussed in terms of clusters of adjacent spin reversals with respect to the aligned ferromagnetic ground state. The lowest-lying states correspond to a single cluster of one or more adjacent spin deviations and have the excitation energies $\omega_0^{(n)}=2j^{z}+n\gamma H_0$, where n is the number of spin reversals and $\gamma=\mu_B g^{z}$. In a plot of energy vs field the states are depicted as a fan of straight lines converging at the point $2j^{z}$ in the limit of zero field. The higher-lying states consist of two or more clusters of adjacent spin deviations and are displayed as fans of straight lines converging at the points $4j^{z}$, $6j^{z}$, etc.

The spin clusters of adjacent spin reversals can in a certain sense be thought of as the simplest kind of bound states encountered in magnetic systems, albeit bound states without spatial motion and in the absence of a band of continuum states.

The transverse mean exchange $j^{\perp}$ gives rise to a spatial dispersion and transforms the localized spin clusters into the well-known multimagnon bound states and bands of the anisotropic Heisenberg magnet. The dispersion law for the single-magnon mode is the familiar result

$$\omega^{(1)}(k)=\omega_0^{(1)}-2j^{z}\sigma\cos ka \quad \text{for } -\pi/a\leq k\leq\pi/a\ ,$$

where a is the nearest-neighbor distance and k ranges over the first Brillouin zone. The two-magnon bound state has the energy

$$\omega^{(2)}(k)=\omega_0^{(2)}-2j^{z}\sigma^2\cos^2\tfrac{1}{2}ka\ ,$$

a result which was first obtained by Bethe[4] in the isotropic case (i.e., $\sigma=1$). Later, Orbach[5] obtained the two-magnon dispersion law in the case of general anisotropy. Recently Wortis[6] and Hanus[7] treated the two-magnon bound-state problem in two and three dimensions. The dispersion laws for the higher multimagnon bound states are not known explicitly for general values of σ. Torrance and Tinkham,[1,3] however, found the dispersion laws $\omega^{(n)}=\omega_0^{(n)}-2j^{z}\frac{1}{2}\sigma^2$ for $\sigma\ll 1$.

Whereas the Heisenberg part $j^{\perp}$ of the exchange interaction does not change the qualitative character of the excitation spectrum, the transverse anisotropy exercises a strong influence on the energy levels. The last term in the Hamiltonian (1.1) breaks the rotational invariance about the axis of magnetization and gives rise to transitions between the multimagnon bound states. The odd multimagnon bound states are admixed into the single-magnon mode. Similarly, the aligned ground state is broken up, owing to the admixture of even multimagnon bound states. In a plot of energy vs field the converging bound states repel one another and assume a downward curvature in the limit of low field.

From an experimental point of view the presence of even a weak transverse anisotropy is of crucial importance. The transverse anisotropy relaxes the selection rules on the transition probabilities and makes it feasible to observe the multimagnon bound-state spectrum in a ferromagnetic resonance experiment.

The admixture of the odd magnon bound states into the single-magnon mode will manifest itself in the transverse frequency-dependent magnetic susceptibility, which will show a set of resonance lines at the positions of the bound states. In a similar fashion, the mixing of the even bound states into the aligned ferromagnetic ground state will be displayed by the longitudinal susceptibility, which will exhibit a set of absorption lines corresponding to the even bound states.

The aim of the present paper is to extend the theory developed by Torrance and Tinkham[1,3] to include other aspects of the dynamical behavior of the linear chain.

In Sec. II, we introduce the magnetic response function which affords a compact description of the absorption spectrum obtained in a resonance experiment. In the absence of transverse anisotropy, the longitudinal response vanishes identically whereas the transverse response has a pole term corresponding to the excitation of a single-magnon mode, i.e., $\chi_{\parallel}=0$ and $\chi_{\perp}\sim(z-\omega^{(1)})^{-1}$. This result is in accordance with the familiar selection rules governing magnetic dipole transitions. The admixture of the higher magnon bound states and bands caused by the transverse anisotropy gives rise to a complicated analytic structure of the transverse and longitudinal responses, i.e.,

$$\chi_{\perp}\sim\cfrac{1}{z-\omega^{(1)}-\cfrac{s^{(3)}}{z-\omega^{(3)}-\cfrac{s^{(5)}}{z-\omega^{(5)}-\cdots}}} \qquad (1.6)$$

and

$$\chi_{\parallel}\sim\cfrac{s^{(2)}}{z^2-\omega^{(2)2}-\cfrac{s^{(4)}}{z^2-\omega^{(4)2}-\cfrac{s^{(6)}}{z^2-\omega^{(6)2}-\cdots}}}\ . \qquad (1.7)$$

We have here represented $\chi_{\perp}$ and $\chi_{\parallel}$ as continued fractions, disregarding for clarity the cut structure which arises from the magnon bands. $\omega^{(i)}$ are the energies of the even and odd multimagnon bound states of the anisotropic Heisenberg chain. The strengths $s^{(i)}$ of the successive admixture of higher bound states are all of order $(j^{a})^2$.

In Sec. III, we set up an approximation scheme for the transverse and longitudinal response functions and expand $\chi_\perp^{-1}$ and $\chi_\parallel$ to second order in j^a. From the continued fractions (1.6) and (1.7) we conclude that an expansion to second order is equivalent to terminating the continued fractions after a single admixture of an even magnon bound state into the ground state in the case of $\chi_\parallel$, and of an odd magnon bound state into the single-magnon mode in the case of $\chi_\perp$, i.e.,

$$\chi_\parallel \sim \frac{s^{(2)}}{z^2 - \omega^{(2)^2}} \tag{1.8}$$

and

$$\chi_\perp \sim \frac{1}{z - \omega^{(1)} - s^{(3)}/(z - \omega^{(3)})} . \tag{1.9}$$

Furthermore, we notice that terminating the continued fractions in the above fashion will in general underestimate the position of the highest bound state involved by a shift of leading order $(j^a)^2/\gamma H_0$. The intensities of the bound states, however, are correctly given to the order in j^a considered. We also notice that since the distance between two unperturbed energy levels is of order γH_0 (disregarding for the moment the effects due to $j^\perp$), the effective dimensionless expansion parameter is $j^a/\gamma H_0$. A perturbation expansion in powers of j^a thus becomes in effect a high-field expansion in powers of $1/\gamma H_0$.

For the longitudinal response in the absence of $j^\perp$ we obtain to second order in $j^a/\gamma H_0$

$$\chi_\parallel(pz) = -2\omega_0^{(2)}\left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \cos^2\frac{pa}{2}\left(\frac{1}{z^2 - \omega_0^{(2)^2}}\right) . \tag{1.10}$$

The corresponding absorptive response is given by

$$\chi_\parallel''(p\omega) = \pi\left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \cos^2\frac{pa}{2} \times[\delta(\omega - \omega_0^{(2)}) - \delta(\omega + \omega_0^{(2)})] . \tag{1.11}$$

To second order in $j^a/\gamma H_0$ we derive the transverse response

$$\chi_\perp(pz) = \left[-\frac{1}{2} + \frac{1}{2}\left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2\right]\Bigg/\left(z - \omega_0^{(1)} - \frac{2(j^a)^2}{\omega_0^{(1)} - j^z} + \frac{4[j^a\omega_0^{(1)}/(\omega_0^{(1)} - j^z)]^2\cos^2 pa}{z + \omega_0^{(1)}} - \frac{4[j^a j^z/(\omega_0^{(1)} - j^z)]^2\cos^2 pa}{z - \omega_0^{(3)}}\right) . \tag{1.12}$$

The absorptive response is

$$\chi_\perp''(p\omega) = \pi I_0^{(1)}(p)\,\delta(\omega - \tilde\omega_0^{(1)}(p)) + \pi I_0^{(3)}(p)\delta(\omega - \tilde\omega_0^{(3)}(p)) + \pi I_0^{(gs)}(p)\,\delta(\omega - \omega_0^{(gs)}(p)) , \tag{1.13}$$

where the respective intensities are given by

$$I_0^{(1)}(p) = \frac{1}{2} - \frac{1}{2}\left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \sin^2 pa - \left(\frac{j^a}{\gamma H_0}\right)^2 \times 2\left(\frac{j^z}{\omega_0^{(1)} - j^z}\right)^2\cos^2 pa ,$$

$$I_0^{(3)}(p) = \left(\frac{j^a}{\gamma H_0}\right)^2 2\left(\frac{j^z}{\omega_0^{(1)} - j^z}\right)^2 \cos^2 pa , \tag{1.14}$$

$$I_0^{(gs)}(p) = -\frac{1}{2}\left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \cos^2 pa .$$

Torrance and Tinkham diagonalized numerically a 40×40 matrix in order to evaluate the effects of $j^\perp$ and j^a on the Ising excitation spectrum. Furthermore, they only considered the lowest series of single Ising clusters as their unperturbed states.

The present calculation goes beyond the results due to Torrance and Tinkham in providing a systematic expansion of the parallel and perpendicular magnetic susceptibility to second order in $j^a/\gamma H_0$. We thereby obtain both the positions and the intensities of the lowest three resonance lines.

The inclusion of the transverse mean exchange $j^\perp$ gives rise to a spatial dispersion, and the evaluation of the longitudinal and transverse responses requires an explicit solution of the two-magnon and three-magnon scattering problems. In Sec. IV, we discuss the structure of the solutions to the two-body and three-body problems. In Sec. V, we evaluate and discuss the effects of the Heisenberg interaction $j^\perp$ on the transverse and longitudinal responses. The expressions for $\chi_\parallel$ and $\chi_\perp$ in the presence of $j^\perp$ are rather lengthy and we shall not give them here.

As mentioned above, the approximation scheme for the response functions in powers of j^a constitutes essentially a high-field expansion. In Sec. VI, we discuss the zero-field limit. Under certain simplifying assumptions it is feasible to evaluate the infinite continued fraction for the response function $\chi_\perp$. In the limit of zero field the perturbed energy levels form a band with an extension determined by j^a. The intensity spectrum is symmetrically shaped around the degeneracy point in contrast to the high-field result, where the intensities fall off rapidly for the higher resonance lines.

At zero field and in the absence of $j^\perp$ and for $p=0$ we obtain the transverse response to all orders in j^a:

$$\chi_\perp(z) = \frac{-1}{(z - 2j^z) + [(z - 2j^z)^2 - (4j^a)^2]^{1/2}} . \tag{1.15}$$

The absorptive response is given by

$$\chi_\perp''(\omega) = \frac{\pi[(4j^a + 2j^z - \omega)(\omega - 2j^z + 4j^a)]^{1/2}}{8j^{a^2}} . \tag{1.16}$$

II. MAGNETIC RESPONSE FUNCTION

We shall discuss the dynamical properties of the linear chain in terms of the so-called magnetic response function[8] or susceptibility χ which expresses the linear response of the magnetization to a varying external magnetic field. The change induced in the average magnetization at the site i is given by the expression

$$\delta\langle m_i^\alpha(t)\rangle = -i\int_{-\infty}^{t} dt' \times\left\langle\left[m_i^\alpha(t),\ -\sum_\beta\sum_{k=1}^{N} m_k^\beta(t')\,h_k^\beta(t')\right]\right\rangle , \tag{2.1}$$

where m_i^α is the α component of the magnetization at the site i, $m_i^\alpha = g^\alpha\mu_B S_i^\alpha$; g^α is the corresponding spectroscopic splitting factor, and h_k^β is the magnetic field component which couples to the magnetization at the site k by the usual dipole coupling.

The absorptive part $\tilde{\chi}_{ik}^{\alpha\beta\prime\prime}(tt')$ of the magnetic response function is defined as the average value of the commutator $[m_i^\alpha(t),\ m_k^\beta(t')]$ in an equilibrium ensemble:

$$\tilde{\chi}_{ik}^{\alpha\beta\prime\prime}(tt') = \tfrac{1}{2}\langle[m_i^\alpha(t),\ m_k^\beta(t')]\rangle . \tag{2.2}$$

The invariance of the equilibrium system under time translations and the invariance under translations through a nearest-neighbor distance a allow us to introduce the Fourier transform

$$\chi^{\alpha\beta\prime\prime}(k\omega) = \sum_{i=1}^{N}\int dt\, e^{-ik(x_i-x_k)}\, e^{i\omega(t-t')}\,\tilde{\chi}_{ik}^{\alpha\beta\prime\prime}(tt') . \tag{2.3}$$

Since $\tilde{\chi}_{ik}^{\alpha\beta\prime\prime}(tt')$ is a commutator of Hermitian operators, we deduce the symmetry properties

$$\chi^{\alpha\beta\prime\prime}(k\omega) = -\chi^{\beta\alpha\prime\prime}(-k-\omega) = [\chi^{\beta\alpha\prime\prime}(k\omega)]^* . \tag{2.4}$$

From time-reversal invariance and rotational invariance follow

$$\chi^{\alpha\beta\prime\prime}(k\omega, B) = -\chi^{\alpha\beta\prime\prime}(k-\omega,\ -B) \tag{2.5}$$

and

$$\chi^{\alpha\beta\prime\prime}(k\omega) = \chi^{\alpha\beta\prime\prime}(-k\omega) , \tag{2.6}$$

where $B = H_0 - \sum_{i=1}^{N}\langle m_i^z\rangle$ is the total magnetic field. Furthermore, we conclude that the only nonvanishing components of the matrix $\chi^{\alpha\beta\prime\prime}$ are $\chi^{zz\prime\prime}$, $\chi^{xx\prime\prime}$, $\chi^{yy\prime\prime}$, $\chi^{xy\prime\prime}$, and $\chi^{yx\prime\prime}$.

The causal character of the response is taken into account by introducing the complex response function

$$\tilde{\chi}_{ik}^{\alpha\beta}(tt') = 2i\,\eta(t-t')\,\tilde{\chi}_{ik}^{\alpha\beta\prime\prime}(tt') , \tag{2.7}$$

where $\eta(t)$ is the step function. Owing to the presence of the step function, the Fourier transform of $\tilde{\chi}_{ik}^{\alpha\beta}$ can be analytically continued to complex values of ω in the upper half-plane. The relationship between $\chi^{\alpha\beta}(k\omega)$ and $\chi^{\alpha\beta\prime\prime}(k\omega)$ is displayed by the Kramers-Konig relation

$$\chi^{\alpha\beta}(kz) = \int\frac{d\omega}{\pi}\,\frac{\chi^{\alpha\beta\prime\prime}(k\omega)}{\omega - z} , \tag{2.8}$$

which provides a spectral representation of $\chi^{\alpha\beta}$ in both half-planes.

From Eqs. (2.1), (2.2), and (2.7) we conclude that for a monochromatic plane-wave field $h_i^\alpha(t) = \frac{1}{2}[h^\alpha e^{-i(\omega t - kx_i)} + \text{c. c.}]$ the mean rate of change of energy per site is given by the following expression:

$$\frac{1}{N}\left\langle\frac{dw}{dt}\right\rangle = \frac{1}{2}\sum_{\alpha\beta}\omega\, h^{\alpha *}\chi^{\alpha\beta\prime\prime}(k\omega)h^\beta . \tag{2.9}$$

In the derivation of (2.9) we have made use of the symmetry properties of $\chi^{\alpha\beta\prime\prime}(k\omega)$.

The expression (2.9) shows that the form of the observed absorption spectrum is directly related to the absorptive part of the response function. As a corollary we conclude that for a stable system $h^{\alpha *}\chi^{\alpha\beta\prime\prime}(k\omega)h^\beta > 0$, from which follows that $\chi^{\alpha\beta\prime\prime}(k\omega)$ is a positive definite matrix.

The ensuing discussion is facilitated by introducing the two-spin correlation functions

$$\chi_\perp''(k\omega) = \tfrac{1}{2}\sum_{i=1}^{N}\int dt\, e^{i\omega(t-t')-ik(x_i-x_k)} \times\tfrac{1}{2}\langle[S_i^-(t),\ S_k^+(t')]\rangle \tag{2.10}$$

and

$$\chi_\parallel''(k\omega) = \sum_{i=1}^{N}\int dt\, e^{i\omega(t-t')-ik(x_i-x_k)}\,\tfrac{1}{2}\langle[S_i^z(t),\ S^z(t')]\rangle . \tag{2.11}$$

The response functions $\chi_\perp''$ and $\chi_\parallel''$ express the transverse and longitudinal susceptibilities of the linear chain and are convenient for a description of the dynamical properties.

In the same way as before we introduce the complex response functions $\chi_\perp$ and $\chi_\parallel$ by means of the spectral representations

$$\chi_{\perp,\parallel}(kz) = \int\frac{d\omega}{\pi}\,\frac{\chi_{\perp,\parallel}''(k\omega)}{\omega - z} . \tag{2.12}$$

In anticipation of the perturbation expansions that we shall derive for the transverse and longitudinal responses, we introduce alternative spectral representations for $\chi_\perp$ and $\chi_\parallel$ in terms of the mass operators $\gamma_\perp$ and $\gamma_\parallel$. From the positive definiteness of $\omega\chi_{\perp,\parallel}$ we conclude that $\chi_{\perp,\parallel}^{-1}$ is analytic in both half-planes with a branch cut along the real axis. Inserting the leading terms in the asymptotic expansions of $\chi_\perp$ and $\chi_\parallel$ we can introduce the mass operators or self-energies $\gamma_\perp$ and $\gamma_\parallel$ in the following manner:

$$\chi_\perp(kz) = \frac{\langle S^z\rangle}{z-\gamma_\perp(kz)} = \frac{-\frac{1}{2}+\langle S^+S^-\rangle}{z-\gamma_\perp(kz)} \tag{2.13}$$

and

$$\chi_\parallel(kz) = \frac{-8j^\perp\langle S_i^+S_{i+1}^-\rangle \sin^2\tfrac{1}{2}ka - 4j^a[\langle S_i^+S_{i+1}^+\rangle + \mathrm{c.\,c}]\cos^2\tfrac{1}{2}ka}{z^2-\gamma_\parallel(kz)} . \tag{2.14}$$

The mass operators $\gamma_\perp$ and $\gamma_\parallel$ have the spectral representations

$$\gamma_{\perp,\parallel}(kz) = \int \frac{d\omega}{\pi} \frac{\gamma''_{\perp,\parallel}(k\omega)}{\omega - z} . \tag{2.15}$$

From the high-frequency expansions of $\chi_\perp$ and $\gamma_\parallel$ we derive the sum rules

$$\int \frac{d\omega}{\pi} \chi''_\perp(k\omega) = -\langle S^z\rangle = \tfrac{1}{2} - \langle S^+S^-\rangle , \tag{2.16}$$

$$\int \frac{d\omega}{\pi} \chi''_\parallel(k\omega) = 0 , \tag{2.17}$$

and

$$\int \frac{d\omega}{\pi} \chi''_\parallel(k\omega)\,\omega = 8j^\perp\langle S_i^+S_{i+1}^-\rangle \sin^2\frac{ka}{2} + 4j^a[\langle S_i^+S_{i+1}^+\rangle + \mathrm{H.\,c.}]\cos^2\frac{ka}{2} . \tag{2.18}$$

The analytic structure of the continued fractions (1.6) and (1.7) satisfies the spectral forms (2.13) and (2.14); consequently, we conclude that the intensity spectrum obtained by terminating the continued fractions at a certain order in j^a will automatically exhaust the sum rules to the same order in j^a.

III. PERTURBATION THEORY TO SECOND ORDER IN j^a

In this section we set up perturbation theory for the response functions $\chi_\perp$ and $\chi_\parallel$ to second order in j^a. From the qualitative discussion in Sec. I and from the spectral representations (2.13) and (2.14) follow that in the case of $\chi_\perp$ such an approximation is obtained by expanding $\chi_\perp^{-1}$ to second order in j^a, i.e., by expanding the mass operator $\gamma_\perp$ and thereby including the admixture of the three-magnon bound state and bands into the single-magnon mode. Since $\chi_\parallel$ vanishes in the absence of transverse anisotropy the second order correction is obtained by a direct expansion of $\chi_\parallel$.

Following the well-known procedure[9] we introduce the two time-ordered Green's functions

$$G_\perp(k\omega) = \sum_{i=1}^N \int dt\, e^{i\omega(t-t')-ik(x_i-x_k)} \times \frac{1}{i}\langle (S_i^-(t)S_k^+(t'))_+\rangle \tag{3.1}$$

and

$$G_\parallel(k\omega) = \sum_{i=1}^N \int dt\, e^{i\omega(t-t')-ik(x_i-x_k)} \times \frac{1}{i}\langle (S_i^+(t^+)S_i^-(t)S_k^+(t'^+)S_k^-(t'))_+\rangle . \tag{3.2}$$

The Green's functions $G_\perp$ and $G_\parallel$ are related to the response functions $\chi_\perp$ and $\chi_\parallel$ by the simple algorithms

$$G_\perp(k\omega) = -2\chi_\perp(k\omega + i\epsilon\,\mathrm{sgn}\omega) \tag{3.3}$$

and

$$G_\parallel(k\omega) = -\chi_\parallel(k\omega + i\epsilon\,\mathrm{sgn}\omega) . \tag{3.4}$$

In order to establish a perturbation expansion of $G_\perp$ and $G_\parallel$ in powers of j^a we express them in the interaction representation, i.e.,

$$G_{\perp ik}(tt') = \frac{1}{i} \frac{\langle 0|(US_i^-(t)S_k^+(t'))|0\rangle}{\langle 0|(U)_+|0\rangle} \tag{3.5}$$

and

$$G_{\parallel ik}(tt') = \frac{1}{i} \frac{\langle 0|(US_i^+(t^+)S_i^-(t)S_k^+(t'^+)S_k^-(t'))|0\rangle}{\langle 0|(U)_+|0\rangle} . \tag{3.6}$$

In this representation the spin operators develop in time according to the anisotropic Heisenberg Hamiltonian

$$H^0 = \sum_{i=1}^N [\omega_0^{(1)}S_i^+S_i^- - j^\perp(S_i^+S_{i+1}^- + \mathrm{H.\,c.}) - 2j^z S_i^+S_i^-S_{i+1}^+S_{i+1}^-] . \tag{3.7}$$

In (3.7) we have made use of the length condition (1.4) in order to eliminate S^z from the Hamiltonian (1.1). The state $|0\rangle$ is the aligned ferromagnetic ground state of H^0. All effects of the perturbation H^a,

$$H^a = \sum_{i=1}^N (-j^a)(S_i^+S_{i+1}^+ + \mathrm{H.\,c.}) , \tag{3.8}$$

are contained in the S matrix U:

$$U = \exp[-i\textstyle\int_{-\infty}^{\infty} dt\, H^a(t)] . \tag{3.9}$$

Using the abbreviation $(i,t) = (1)$ and the repeated index summation convention we can write the perturbation expansions of $G_\perp^{-1}$ and $G_\parallel$ in the form

$$G_\perp^{-1}(12) = [G_\perp^{-1}(12)]_{j^a=0} + \left[\frac{\delta G_\perp^{-1}(12)}{\delta j^a(34)}\right]_{j^a=0} j^a(34)$$

$$+\frac{1}{2}\left[\frac{\delta^2 G_\perp^{-1}(12)}{\delta j^a(34)\,\delta j^a(56)}\right]_{j^a=0} j^a(34) j^a(56) \tag{3.10}$$

and

$$G_\parallel(12)=[G_\parallel(12)]_{j^a=0}+\left[\frac{\delta G_\parallel(12)}{\delta j^a(34)}\right]_{j^a=0} j^a(34)$$
$$+\frac{1}{2}\left[\frac{\delta^2 G_\parallel(12)}{\delta j^a(34)\,\delta j^a(56)}\right]_{j^a=0} j^a(34) j^a(56)\,, \tag{3.11}$$

where

$$j^a(12)=j^a_{i_1 i_2}\,\delta(t_1-t_2)$$
$$=\tfrac{1}{2} j^a(\delta_{i_1+1,i_2}+\delta_{i_1-1,i_2})\,\delta(t_1-t_2)\,.$$

Let us first consider the longitudinal response. From symmetry arguments it follows that $[G_\parallel(12)]_{j^a=0}$ and $[\delta G_\parallel(12)/\delta j^a(34)]_{j^a=0}$ vanish identically. By means of the decomposition of the unit operator

$$1=|\,0\rangle\langle 0\,|+\frac{1}{1!}\sum_{i=1}^{N} S_i^+\,|\,0\rangle\langle 0\,|\,S_i^-$$
$$+\frac{1}{2!}\sum_{i=1,k=1}^{N} S_i^+ S_k^+\,|\,0\rangle\langle 0\,|\,S_i^- S_k^- +\cdots \tag{3.12}$$

and by introducing the two-magnon equal-time Green's function

$$G_{kqk'q'}(\omega)=\langle 0\,|\,S_k^- S_q^-(\omega-H^0+i\epsilon)^{-1} S_{k'}^+ S_{q'}^+\,|\,0\rangle\,, \tag{3.13}$$

we obtain the following result for the second-order correction to $G_\parallel$:

$$G_{\parallel pp'}(\omega)=\sum j^a_{kk'}\,G_{kk'pq}(0)\,G_{pqp'q'}(\omega)$$
$$\times G_{p'q'nn'}(0)\,j^a_{nn'}+\sum j^a_{kk'}\,G_{kk'p'q'}(0)$$
$$\times G_{p'q'pq}(-\omega)\,G_{pqnn'}(0)\,j^a_{nn'}\;. \tag{3.14}$$

The Green's function $G_{kk'pq}(0)$, which multiplies $j^a_{kk'}$, is independent of energy and is essentially a field-dependent vertex correction to the coupling strength j^a; the pair of adjacent spin deviations created in the vacuum interact by means of the Ising interaction j^z.

The admixture of the two-magnon bound state and band into the aligned ground state is described by the energy-dependent two-magnon Green's function $G_{pqp'q'}(\omega)$ which has a pole at the position of the two-magnon bound state $\omega^{(2)}$ and a branch cut corresponding to the two-magnon band.

In the absence of the Heisenberg interaction, i.e., $j^\perp=0$, it is an easy task to evaluate the longitudinal response. The Green's function $G_{kqk'q'}(\omega)$ is symmetric in each pair of indices, but vanishes when $k=q$ or $k'=q'$. This is a reflection of the fact that $S_i^+S_i^+=0$ and is a specific feature of the spin system. The last property is taken into account by expressing $G_{kqk'q'}$ in terms of an antisymmetric unit operator

$$\Delta^1_{kqk'q'}=\frac{1}{2!}\,(\delta_{kk'}\delta_{qq'}-\delta_{kq'}\delta_{qk'})\,. \tag{3.15}$$

Introducing the projection operator

$$Q_{kq}=\delta_{kq+1}+\delta_{kq-1} \tag{3.16}$$

with the eigenvalues 0 and 1 corresponding, respectively, to two nonadjacent and two adjacent spin deviations, we get for the Green's function $G_{kqk'q'}(\omega)$

$$G_{kqk'q'}(\omega)=-2\eta_{kq}\Delta^1_{kqk'q'}\eta_{q'k'}$$
$$\times\left(\frac{Q_{kq}}{\omega-\omega_0^{(2)}+i\epsilon}+\frac{1-Q_{kq}}{\omega-2\omega_0^{(1)}+i\epsilon}\right)\,. \tag{3.17}$$

The sign factor η_{kq} which is $+1$ for $k>q$ and -1 for $k<q$ restores the symmetry properties of $G_{kqk'q'}$. We notice incidentally that the representation (3.17) can only be established for a one-dimensional system, where the spin operators can be arranged in a linear sequence. Fourier transforming and making use of the algorithm (3.4) we obtain the longitudinal response

$$\chi_\parallel(pz)=\left(\frac{j^a}{\omega_0^{(1)}-j^z}\right)^2\cos^2\frac{pa}{2}$$
$$\times\left(\frac{1}{\omega_0^{(2)}-z}-\frac{1}{-\omega_0^{(2)}-z}\right)\,. \tag{3.18}$$

The absorptive response is given by

$$\chi''_\parallel(p\omega)=\pi\left(\frac{j^a}{\omega_0^{(1)}-j^z}\right)^2\cos^2\frac{pa}{2}$$
$$\times[\,\delta(\omega-\omega_0^{(2)})-\delta(\omega+\omega_0^{(2)})\,]\,. \tag{3.19}$$

$\chi''_\parallel$ has an observable resonance line at $\omega_0^{(2)}$, i.e., the excitation energy of two adjacent spin deviations. The intensity of the line is proportional to $[j^a/(\omega_0^{(1)}-j^z)]^2\cos^2\tfrac{1}{2}pa$; in accordance with our discussion in Sec. I, this result, however, is only valid in the high-field limit, i.e., $j^a\ll\gamma H_0$. In this limit the two-magnon line carries all the intensity and (3.19) exhausts the sum rule (2.18). As the field decreases the total intensity is shared by all the converging bound states. The fact that (3.19) continues to exhaust the sum rule is another indication that (3.19) is inadequate at low fields. As discussed in Sec. I the position of the line is underestimated by a shift of leading order $j^{a^2}/\gamma H_0$; this is exactly the shift which gives rise to a downward curvature at low field of the two-magnon bound state in a plot of energy vs field.

Turning our attention to the transverse response we notice again that $[\delta G_\perp^{-1}(12)/\delta j^a(34)]_{j^a=0}$ vanishes from symmetry arguments. In order to evaluate the second-order correction to $G_\perp^{-1}$ we make use of the identity

$$\left[\frac{\delta G_\perp^{-1}(12)}{\delta j^a(34)}\right]_{j^a=0} = -\left[G_\perp^{-1}(15)\right]_{j^a=0}\left[\frac{\delta G_\perp(56)}{\delta j^a(34)}\right]_{j^a=0} \times\left[G_\perp^{-1}(62)\right]_{j^a=0}, \quad (3.20)$$

which follows from the definition of the inverse Green's function,

$$G_\perp(13)\, G_\perp^{-1}(32) = \delta(12) = \delta_{i_1 i_2}\,\delta(t_1 - t_2) . \quad (3.21)$$

The inverse Green's function $[G_\perp^{-1}(12)]_{j^a=0}$ is obtained from the Heisenberg equation of motion applied to the Hamiltonian (3.7). The equation of motion can be written in the form

$$[G_\perp^{-1}(12)]_{j^a=0}\, S^-(2) = 4 j^\perp(13)\, S^+(1)\, S^-(1)\, S^-(3) - 4 j^z(13)\, S^-(1)\, S^+(3)\, S^-(3) , \quad (3.22)$$

where we have introduced the explicit expression for $(G_\perp^{-1})_{j^a=0}$

$$[G_\perp^{-1}(12)]_{j^a=0} = \left(i\, \frac{d}{dt_1} - \omega_0^{(1)}\right)\delta(12) + 2 j^\perp(12) \quad (3.23)$$

and the notation

$$j^{\perp,z}(12) = j^{\perp,z}_{i_1 i_2}\, \delta(t_1 - t_2) = \tfrac{1}{2} j^{\perp,z}(\delta_{i_1+1,i_2} + \delta_{i_1-1,i_2})\, \delta(t_1 - t_2) . \quad (3.24)$$

The second-order correction to $G_\perp^{-1}$ is now evaluated by applying $(G_\perp^{-1})_{j^a=0}$ to the expectation value of the six spin operators arising from differentiation with respect to $j^a(12)$. The time ordering of the spin operators, however, gives rise to rather lengthy expressions and we shall, therefore, defer the details to Appendix A.

It is convenient to analyze the structure of the expansion of $G_\perp^{-1}$ by means of the spectral representation (2.13). There are two distinct contributions to the mass operator $\gamma_\perp$. One part, γ_{gs}, arises from the structure of the perturbed ground state and another, γ_{bs}, contains the contribution from the three-magnon bound state and the three-magnon bands. Splitting off the single-magnon energy $\omega^{(1)}(k)$, we can separate the remaining part of the ground-state contribution into a momentum-dependent shift Δ_{gs}, and a pole term $-S_{gs}/[\omega + \omega^{(1)}(k)]$. We thus obtain the spectral representation

$$G_\perp(k\omega) = \frac{1 - 2\langle S^+ S^-\rangle}{\omega - \omega^{(1)}(k) - \Delta_{gs}(k) - S_{gs}(k)/[\omega + \omega^{(1)}(k)] - \gamma_{bs}(k)} . \quad (3.25)$$

The numerator in the spectral form (3.25) is determined as the coefficient multiplying the leading term in the high-frequency expansion of $G_\perp$ and is given by the expectation value of the equal-time commutator of S^+ and S^-. For an ordinary particle system, e.g., a Bose system, this commutator equals 1, and the leading term in the asymptotic expansion is $1/z$. For the spin system, on the other hand, the commutator is $-2S^z = 1 - S^+S^-$, and its expectation value depends on the structure of ground state of the system. In the aligned ground state of H^0, $\langle 0 \mid S^+S^- \mid 0\rangle$ vanishes and the numerator assumes the value 1. In the perturbed ground state, however, the value is reduced from 1 because of virtual pair fluctuations induced by j^a. In site space we obtain the following contribution to the numerator in (3.25) [see Eq. (A3)]:

$$1 - 2\langle S^+ S^-\rangle = 1 - 2\,\mathrm{F.t.}\textstyle\sum j^a_{kk'}\, G_{kk',pq}(0)\, G_{pqnn'}(0)\, j^a_{nn'} . \quad (3.26)$$

The abreviation F.t. stands for the Fourier transform. The correction term $\langle S^+S^-\rangle$ describes the virtual creation and subsequent annihilation of a pair of spin deviations in the perturbed ground state. We notice again the field-dependent vertex correction of j^a due to the Ising interaction j^z, a feature which occurs whenever a pair of spin deviations is created or annihilated by j^a.

The ground-state shift Δ_{gs}, which is given in Eq. (A4), arises from vacuum fluctuations in the perturbed ground state. Δ_{gs} consists of a momentum-independent shift, which can be absorbed by redefining the energy ω, and a momentum-dependent part, which leads to a renormalization of the single-magnon mode. The correction of the magnon dispersion law due to vacuum fluctuations is quite similar to the renormalization encountered in the random-phase approximation (RPA) applied to the Heisenberg Hamiltonian at finite temperatures.[6]

The second part of the ground-state contribution to the mass operator, the pole term $S_{gs}/[\omega + \omega^{(1)}(k)]$, is evaluated in Eq. (A5). In its evaluation we have introduced the one-magnon equal-time Green's function

$$G_{kk'}(\omega) = \langle 0 \mid S_k^-(\omega - H^0 + i\epsilon)^{-1} S_{k'}^+ \mid 0\rangle . \quad (3.27)$$

The perturbed ground state of the system is strongly correlated, because of virtual pair excitations, and is quite different in structure from the aligned unperturbed ground state. The ground-state mode with energy $\omega = -\omega^{(1)}$ and strength S_{gs} of order $(j^a)^2$ corresponds to the possibility of gaining energy by breaking up a pair of spin deviations in the correlated ground state.

The bound-state contribution to the mass operator γ_{bs} is evaluated in Eq. (A6) making use of the three-magnon equal-time Green's function

$$G_{kqmk'q'm'}(\omega)=\langle 0 \mid S_k^- S_q^- S_m^-(\omega - H^0 + i\epsilon)^{-1} S_{k'}^+ S_{q'}^+ S_{m'}^+ \mid 0\rangle . \tag{3.28}$$

The physical interpretation of γ_{bs} is clear. A pair of adjacent spin deviations excited in the vacuum because of j^a is first renormalized. Subsequently, it interacts with the incoming spin deviation created by the photon $h_{p'}^+ e^{-i\omega t}$ via the dipole coupling $h_{p'}^+ S_{p'}^+ e^{-i\omega t}$. The scattering of the three spin deviations is described by the three-magnon Green's function $G_{pqrp'q'r'}(\omega)$, which has a pole corresponding to the three-magnon bound state and a branch cut arising from the three-magnon bands.

Because of the simplicity of the Ising chain, the transverse response can be easily evaluated in the limit $j^\perp = 0$. Similar to our derivation of the two-magnon Green's function (3.17), we introduce the antisymmetric unit operator

$$\Delta^1_{kqmk'q'm'} = (1/3!)\,[\,\delta_{kk'}\delta_{qq'}\delta_{mm'} - \delta_{kk'}\delta_{qm'}\delta_{mq'} + (\text{cyclic})\,(kqm,\, k'q'm')\,] \tag{3.29}$$

and the projection operator

$$Q_{kqm} = (\delta_{kq+1} + \delta_{kq-1} + \delta_{qm+1} + {}_{qm-1} + \delta_{mk+1} + \delta_{mk-1}) , \tag{3.30}$$

with the eigenvalues 0, 1, and 2 corresponding, respectively, to three nonadjacent, two adjacent, and three adjacent spin deviations. The Green's function $G_{kqmk'q'm'}$ can now be written in the form

$$G_{kqmk'q'm'}(\omega) = -6\eta_{kq}\eta_{qm}\eta_{mk}\Delta_{kqmk'q'm'}\eta_{m'q'}\eta_{q'k'}\eta_{k'm'} \times\left(\frac{\frac{1}{2}(1-Q_{kqm})(2-Q_{kqm})}{\omega - 3\omega_0^{(1)} + i\epsilon} + \frac{Q_{kqm}(2-Q_{kqm})}{\omega - 2\omega_0^{(1)} - \omega_0^{(2)} + i\epsilon} + \frac{\frac{1}{2}Q_{kqm}(Q_{kqm}-1)}{\omega - \omega_0^{(3)} + i\epsilon}\right) . \tag{3.31}$$

The one-magnon Green's function (3.27) which we shall also need in the evaluation of $G_\perp$ is given by

$$G_{kk'}(\omega) = \frac{\delta_{kk'}}{\omega - \omega_0^{(1)}} \tag{3.32}$$

in the limit $j^\perp = 0$.

By means of the explicit expressions (3.17), (3.31), and (3.32) we can now evaluate the various contributions to $G_\perp(p\omega)$. We obtain

$$1 - 2\langle S^+ S^-\rangle = 1 - \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 , \tag{3.33}$$

$$\Delta_{gs}(p) = \frac{2(j^a)^2}{\omega_0^{(1)} - j^z} , \tag{3.34}$$

$$S_{gs}(p) = -4\left(\frac{j^a\omega_0^{(1)}}{\omega_0^{(1)} - j^z}\right)^2 \cos^2 pa , \tag{3.35}$$

and

$$\gamma_{bs}(p\omega) = \frac{S_{bs}(p)}{\omega - \omega_0^{(3)} + i\epsilon} = \frac{4[j^a j^z/(\omega_0^{(1)} - j^z)]^2 \cos^2 pa}{\omega - \omega_0^{(3)} + i\epsilon} . \tag{3.36}$$

Employing the algorithm (3.3) we find the transverse response

$$\chi_\perp(pz) = \frac{-\frac{1}{2} + \langle S^+ S^-\rangle}{z - \omega_0^{(1)} - \Delta_{gs}(p) - S_{gs}(p)/(z + \omega_0^{(1)}) - S_{bs}(p)/(z - \omega_0^{(3)})} . \tag{3.37}$$

In accordance with our discussion in Sec. I, we conclude that the expression (3.37) is only valid for $j^a \ll \gamma H_0$. To second order in $j^a/\gamma H_0$ we obtain the absorptive response

$$\chi_\perp''(p\omega) = \pi I_0^{(1)}(p)\,\delta(\omega - \tilde\omega_0^{(1)}(p)) + \pi I_0^{(3)}(p)\,\delta(\omega - \tilde\omega^{(3)}(p)) + \pi I_0^{(gs)}(p)\,\delta(\omega - \tilde\omega_0^{(gs)}(p)) . \tag{3.38}$$

where the positions of the single-magnon mode, the three-magnon bound-state mode, and the ground-state mode are given, in respective order, by the expressions

$$\tilde\omega^{(1)}(p) = \omega_0^{(1)} + \Delta_{gs}(p) + \frac{S_{gs}(p)}{2\omega_0^{(1)}} - \frac{S_{bs}(p)}{2\gamma H_0} , \tag{3.39}$$

$$\tilde\omega^{(3)}(p) = \omega_0^{(3)} + \frac{S_{bs}(p)}{2\gamma H_0} , \tag{3.40}$$

and

$$\tilde\omega^{(gs)}(p) = -\omega_0^{(1)} - S_{gs}(p)/2\omega_0^{(1)} . \tag{3.41}$$

The intensities of the three modes are given by the following expressions:

$$I_0^{(1)}(p) = \frac{1}{2} - \langle S^+ S^-\rangle - \frac{1}{2}\,\frac{S_{gs}(p)}{(2\omega_0^{(1)})^2} - \frac{1}{2}\,\frac{S_{bs}(p)}{(\gamma H_0)^2} , \tag{3.42}$$

$$I_0^{(3)}(p)=\frac{1}{2}\,\frac{S_{\mathrm{bs}}(p)}{(\gamma H_0)^2}\ , \tag{3.43}$$

and

$$I_0^{(\mathrm{gs})}(p)=\frac{1}{2}\,\frac{S_{\mathrm{gs}}(p)}{(2\omega_0^{(1)})^2}\ . \tag{3.44}$$

$\chi_\perp''$ has two observable resonance lines at $\tilde{\omega}_0^{(1)}$ and $\tilde{\omega}_0^{(3)}$, i.e., the single-magnon mode and the three-magnon bound state. The ground-state mode at energy $\tilde{\omega}_0^{(\mathrm{gs})}$ is not accessible to experiments since it occurs at negative energy. It indirectly contributes, however, to the intensities and positions of the observable resonance lines.

In the high-field limit the positions of the three lines approach the unperturbed Ising values. As the field decreases, the single-magnon mode attains a negative shift causing a downward deflection in a plot of energy vs field. The three-magnon bound state, however, gets a positive shift of the same magnitude and will bend upwards as the field is lowered. As discussed in Sec. I, the position of this line, however, is underestimated by a contribution of leading order $(j^a)^2/\gamma H_0$ arising from the admixture of the higher bound states. This shift will cause the line to assume a downward curvature as the field decreases.

In the high-field limit, i.e., $\gamma H_0 \gg j^a$, the single-magnon mode carries all the intensity and exhausts the sum rule (2.16). The intensities of the two other modes both vanish in the high-field limit. As the field is lowered the intensity of the three-magnon mode increases and at the same time the ground-state mode gets a negative increasing intensity. We notice that the expressions (3.42), (3.43), and (3.44) for the intensities are correctly given to second order in $j^a/\gamma H_0$ and exhaust the sum rule (2.16) to that order.

IV. TWO- AND THREE-MAGNON SCATTERING PROBLEM

In order to estimate the effects of the Heisenberg interaction $j^\perp$ we need a more detailed evaluation of the Green's functions $G_{kqk'q'}$ and $G_{kqmk'q'm'}$.

Separating the Hamiltonian (3.7) in kinetic and potential energy parts, i.e., $H^0=H_0+V$, where

$$H_0=\sum_{i=1}^{N}\left[\omega_0^{(1)}S_i^+S_i^- - j^\perp(S_i^+S_{i+1}^- + \mathrm{H.c.})\right] \tag{4.1}$$

and

$$V=\sum_{i=1}^{N}(-2j^z)S_i^+S_i^-S_{i+1}^+S_{i+1}^-\ , \tag{4.2}$$

and employing the identity (3.12), it is easily seen that $G_{kqk'q'}$ and $G_{kqmk'q'm'}$ satisfy the integral equations

$$G_{kqk'q'}(\omega)=G^0_{kqk'q'}(\omega)+\sum_{k''q''}K_{kqk''q''}(\omega)\,G_{k''q''k'q'}(\omega) \tag{4.3}$$

and

$$G_{kqmk'q'm'}(\omega)=G^0_{kqmk'q'm'}(\omega)+\sum_{k''q''m''}K_{kqmk''q''m''}(\omega)\,G_{k''q''m''k'q'm'}(\omega)\ . \tag{4.4}$$

The Green's functions $G^0_{kqk'q'}$ and $G^0_{kqmk'q'm'}$ are defined as follows:

$$G^0_{kqk'q'}(\omega)=\langle 0\,|\,S_k^-S_q^-(\omega-H_0+i\epsilon)^{-1}S_{k'}^+S_{q'}^+\,|\,0\rangle\ , \tag{4.5}$$

$$G^0_{kqmk'q'm'}(\omega)=\langle 0\,|\,S_k^-S_q^-S_m^-(\omega-H_0+i\epsilon)^{-1}S_{k'}^+S_{q'}^+S_{m'}^+\,|\,0\rangle\ , \tag{4.6}$$

and they describe the propagation of two and three magnons, respectively, in the absence of the two-body potential V. The integral kernels $K_{kqk'q'}$ and $K_{kqmk'q'm'}$ are defined in the following manner:

$$K_{kqk'q'}(\omega)=\frac{1}{2!}\,\langle 0\,|\,S_k^-S_q^-(\omega-H_0+i\epsilon)^{-1}\,VS_{k'}^+S_{q'}^+\,|\,0\rangle \tag{4.7}$$

and

$$K_{kqmk'q'm'}(\omega)=\frac{1}{3!}\,\langle 0\,|\,S_k^-S_q^-S_m^-(\omega-H_0+i\epsilon)^{-1}\,VS_{k'}^+S_{q'}^+S_{m'}^+\,|\,0\rangle\ . \tag{4.8}$$

In order to make the integral equations more manageable it is convenient to introduce the antisymmetric Green's functions $F_{kqk'q'}$ and $F_{kqmk'q'm'}$ defined in the following manner:

$$F_{kqk'q'}(\omega)=\eta_{kq}\,G_{kqk'q'}(\omega)\,\eta_{q'k'} \tag{4.9}$$

and

$$F_{kqmk'q'm'}(\omega)=\eta_{kq}\,\eta_{qm}\,\eta_{mk}\,G_{kqmk'q'm'}(\omega)\,\eta_{m'q'}\eta_{q'k'}\,\eta_{k'm'}\ . \tag{4.10}$$

By Fourier transforming we obtain for $F_{kqk'q'}$ and $F_{kqmk'q'm'}$ the two linear integral equations

$$F(kqk'q'\omega)=F^0(kqk'q'\omega)+\sum_{k''q''}K(kqk''q''\omega)\,F(k''q''k'q'\omega) \tag{4.11}$$

and

$$F(kqmk'q'm'\omega)=F^0(kqmk'q'm'\omega)+\sum_{k''q''m''}K(kqmk''q''m''\omega)\,F(k''q''m''k'q'm'\omega)\ , \tag{4.12}$$

where

$$F^0(kqk'q'\omega)=\frac{-2!\,\Delta_1(kqk'q')}{\omega-\omega^{(1)}(k)-\omega^{(1)}(q)+i\epsilon} \tag{4.13}$$

and

$$F^0(kqmk'q'm'\omega)=\frac{-3!\,\Delta_1(kqmk'q'm')}{\omega-\omega^{(1)}(k)-\omega^{(1)}(q)-\omega^{(1)}(m)+i\epsilon}\ . \tag{4.14}$$

We have introduced the antisymmetric unit operators in two- and three-particle momentum space

$$\Delta_1(kqk'q') = (1/2!)(\delta_{kk'}\delta_{qq'} - \delta_{kq'}\delta_{qk'}) \quad (4.15)$$

and

$$\Delta_1(kqmk'q'm') = (1/3!)[\delta_{kk'}\delta_{qq'} - \delta_{kq'}\delta_{qk'})\delta_{mm'} + (\text{cyclic})(k'q'm')] . \quad (4.16)$$

The antisymmetric kernels are given by the expressions

$$K(kqk'q'\omega) = 2j^{*} \sum_{k''q''} F^0(kqk''q''\omega)\,\Delta_2(k''q''k'q') \quad (4.17)$$

and

$$K(kqmk'q'm'\omega) = \tfrac{2}{9}\, j^{*} \sum_{k''q''m''} F^0(kqmk''q''m''\omega) \times \Delta_2(k''q''m''k'q'm') . \quad (4.18)$$

The nearest-neighbor interaction between two and three magnons is represented by the projection operators

$$\Delta_2(kqk'q') = \frac{1}{N}\, \sin\!\left(\frac{k-q}{2}\right)a \sin\!\left(\frac{k'-q'}{2}\right)a\, \delta_{k+q,k'+q'} \quad (4.19)$$

and

$$\Delta_2(kqmk'q'm') = \frac{1}{N}\, \sin\!\left(\frac{k-q}{2}\right)a \sin\!\left(\frac{k'-q'}{2}\right) \times a\, \delta_{mm'}\delta_{k+q,k'+q'} + (\text{cyclic})(kqm, k'q'm') . \quad (4.20)$$

It is easily verified that $\Delta_2(kqk'q')$ and $\Delta_2(kqmk'q'm')$ are the Fourier transforms of $\frac{1}{2}Q_{kq}\Delta_{kqk'q'}$ and $\frac{3}{2}Q_{kqm}\Delta_{kqmk'q'm'}$, respectively, where Q_{kq} and Q_{kqm} are the projection operators (3.16) and (3.30).

The two-magnon scattering problem can be solved easily. Since the kernel (4.17) is separable in momentum space we are lead to the linear algebraic equation

$$F(kqk'q'\omega) = F^0(kqk'q'\omega) + \sum F^0(kqk''q''\omega) \times \sin[\tfrac{1}{2}(k''-q'')]a\, \alpha(k+q, k'q'\omega) , \quad (4.21)$$

where

$$\alpha(k+q, k'q'\omega) = 2j^{*}(1/N) \sum \delta_{k+q,k''+q''} \times \sin[\tfrac{1}{2}(k''-q'')]a F(k''q''k'q'\omega) . \quad (4.22)$$

Solving for α we obtain an exact solution of the two-magnon scattering problem:

$$F(kqk'q'\omega) = F^0(kqk'q'\omega) + 2j^{*} \frac{\sum F^0(kqk''q''\omega)\,\Delta_2(k''q''\tilde{k}''\tilde{q}'')\,F^0(\tilde{k}''\tilde{q}''k'q'\omega)}{D(k+q\omega)} , \quad (4.23)$$

where

$$D(k+q\omega) = 1 - 2j^{*}\sum F^0(kqk'q'\omega)\,\Delta_2(k'q'kq) . \quad (4.24)$$

In Appendix B we derive a spectral representation for D^{-1},

$$D^{-1}(p\omega) = 1 + \int \frac{A(p\Omega)}{\Omega - \omega - i\epsilon}\, \frac{d\Omega}{\pi} , \quad (4.25)$$

where

$$A(p\Omega) = j^{*}(1 - \sigma^2\cos^2 \tfrac{1}{2}pa)2\pi\delta(\Omega - \omega^{(2)}(p)) + \frac{\frac{1}{2}\{[\omega_+^{(2)}(p) - \Omega][\Omega - \omega_-^{(2)}(p)]\}^{1/2}}{\Omega - \omega^{(2)}(p)} . \quad (4.26)$$

Here $\omega_\pm^{(2)}(p) = 2\omega_0^{(1)} \pm 4j^{\perp}\cos\frac{1}{2}pa$ give the upper and lower edges of the two-magnon band, respectively. We conclude from (4.26) that D^{-1} has a magnon bound-state pole at $\omega^{(2)}(p)$ with strength $2j^{*}(1-\sigma^2 \times\cos^2\frac{1}{2}pa)$ and a branch cut extending from $\omega_-^{(2)}$ to $\omega_+^{(2)}$, corresponding to the two-magnon band.

In contrast to the two-magnon scattering problem, the scattering of three magnons cannot be solved exactly. Furthermore, the integral equation (4.12) is not of the Fredholm type and cannot be treated by applying standard techniques. In addition to describing the simultaneous scattering of three magnons, (4.12) also includes processes in which only two magnons scatter; the third particle assumes the role of a spectator and does not participate in the scattering act. This is reflected in the kernel (4.18) which contains a δ function $\delta_{mm'}$ expressing the separate conservation of momentum of the third particle. The presence of the δ function renders the kernel unbounded, and, therefore, the integral equation cannot be solved by the Fredholm method.

The mathematical aspects of the three-body scattering problem were first treated in a satisfactory manner by Faddeev.[10] We shall employ a formulation due to Weinberg[11] which is more suited for our purposes.

The difficulties associated with the unbounded kernel are circumvented by separating the Green's function F into two parts. The first part, $F^0 + T$, included the inhomogeneous term F^0 and describes all scattering processes in which fewer than three particles participate. The remaining part of F, the connected part C, describes the simultaneous scattering of three particles. That is,

$$F = F^0 + T + C \ . \tag{4.27}$$

A little algebra shows that $C = IF$, where the new kernel I is defined in the following way:

$$I = K - T(F^0)^{-1}(1 - K) \ . \tag{4.28}$$

We obtain the new modified integral equation

$$F = F^0 + T + IF \ . \tag{4.29}$$

The two-particle scattering amplitude T is constructed by means of the solution to the two-body problem obtained previously. We get

$$T(kqmk'q'm'\omega) = 8j^z \frac{\Delta_2(kqk'q')\,\delta_{mm'}}{[\omega - \omega^{(1)}(k) - \omega^{(1)}(q) - \omega^{(1)}(m)]\, D(k+qm\omega)\, [\omega - \omega^{(1)}(k') - \omega^{(1)}(q') - \omega^{(1)}(m')]} \ , \tag{4.30}$$

where

$$D(k+qm\omega) = 1 + 4j^z \sum \frac{\delta_{k+q,k'+q'}\, \Delta_2(k'q'k'q')}{\omega - \omega^{(1)}(k') - \omega^{(1)}(q') - \omega^{(1)}(m)} \ , \tag{4.31}$$

We have introduced the spectator particle by displacing the energy variable ω by the amount $\omega^{(1)}(m)$.

Introducing $A = F^0 + T$ we can write the formal Fredholm solution of (4.29) in the form

$$F = A + NA/D \ , \tag{4.32}$$

where N and D are the usual Fredholm numerator and denominator. The inhomogeneous part A contains the contributions from the three-magnon bands (i.e., the band of three single magnons and the band of a two-magnon bound state plus a single magnon). The three-magnon bound state resides in the Fredholm denominator D. From general analyticity it follows that D^{-1} has the spectral representation

$$D^{-1}(p\omega) = 1 + \int \frac{A(p\Omega)}{\Omega - \omega - i\epsilon} \frac{d\Omega}{\pi} \ . \tag{4.33}$$

In analogy to (4.26) we expect A to have the form

$$A(p\Omega) = S_{\text{bs}}(p)\, \pi\delta(\Omega - \omega^{(3)}(p)) + \Delta_{\text{band}}(p\Omega) \ , \tag{4.34}$$

where $\omega^{(3)}$ is the position of the three-magnon bound state and S_{bs} its strength. Δ_{band} is the contribution to A arising from the three-magnon bands.

V. PERTURBATION THEORY TO SECOND ORDER IN $j^\perp$

As discussed in Sec. I, the isotropic part of the exchange interaction, the Heisenberg part, does not have any pronounced effect on the nature of the excitation spectrum. The Heisenberg interaction $j^\perp$, however, introduces a spatial dispersion and the evaluation of $G_{kqk'q'}$ and $G_{kqmk'q'm'}$ requires the explicit solutions of a two- and three-magnon scattering problem, respectively.

The longitudinal response can be expressed entirely in terms of the two-magnon Green's function $G_{kqk'q'}$. Having obtained an exact solution in Sec. IV, it is an easy task to evaluate the longitudinal response to second order in $j^\perp$. In Appendix C we compute $\chi_\parallel$ in detail. We obtain the following result:

$$\chi_\parallel(pz) = \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \cos^2\frac{pa}{2} \left[1 + \left(2\frac{j^z}{\omega_0^{(1)}}\, \frac{2j^z - \omega_0^{(1)}}{\omega_0^{(1)} - j^z} + \frac{4j^z - \omega_0^{(1)}}{\omega_0^{(1)}} \cos^2\frac{pa}{2}\right)\sigma^2\right]\left(\frac{1}{\omega^{(2)}(p) - z} - \frac{1}{-\omega^{(2)}(p) - z}\right). \tag{5.1}$$

The absorptive response is given by

$$\chi_\parallel''(p\omega) = \pi\left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \cos^2\frac{pa}{2}\left[1 + \left(2\frac{j^z}{\omega_0^{(1)}}\, \frac{2j^z - \omega_0^{(1)}}{\omega_0^{(1)} - j^z} + \frac{4j^z - \omega_0^{(1)}}{\omega_0^{(1)}}\cos^2\frac{pa}{2}\right)\sigma^2\right]\left[\delta(\omega - \omega^{(2)}(p)) - \delta(\omega + \omega^{(2)}(p))\right] . \tag{5.2}$$

The effect of the Heisenberg interaction is to move the resonance line to the position of the two-magnon bound-state energy $\omega^{(2)}(p)$. To second order in σ we obtain the correction factor

$$1 + \left(2\frac{j^z}{\omega_0^{(1)}}\, \frac{2j^z - \omega_0^{(1)}}{\omega_0^{(1)} - j^z} + \frac{4j^z - \omega_0^{(1)}}{\omega_0^{(1)}}\cos^2 \tfrac{1}{2}pa\right)\sigma^2$$

to the intensity of the line.

In order to compute the transverse response we need an explicit solution of the three-magnon scattering problem defined in Sec. IV. We defer the treatment which is of rather technical nature to Appendix C and state the results for the various contributions to $\chi_\perp$ here.

$\chi_\perp$ has again the form

$$\chi_\perp(pz) = \frac{-\frac{1}{2} + \langle S^+ S^- \rangle}{z - \omega^{(1)}(p) - \Delta_{gs}(p) - S_{gs}(p)/[z + \omega^{(1)}(p)] - S_{bs}(p)/[z - \omega^{(3)}(p)]} , \tag{5.3}$$

where to second order in σ we obtain

$$1 - 2\langle S^+ S^- \rangle = 1 - \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 - \frac{j^z}{\omega_0^{(1)}} \frac{3\omega_0^{(1)} - j^z}{\omega_0^{(1)} - j^z} \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \sigma^2 , \tag{5.4}$$

$$\Delta_{gs}(p) = \frac{2 j^{a2}}{\omega_0^{(1)} - j^z} - 2 j^z \frac{j^z}{\omega_0^{(1)}} \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \sigma \cos pa + 2 \frac{j^z}{\omega_0^{(1)} - j^z} (2\omega_0^{(1)} - j^z) \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \sigma^2 , \tag{5.5}$$

$$S_{gs}(p) = -4\left(\frac{j^a \omega_0^{(1)}}{\omega_0^{(1)} - j^z}\right)^2 \cos^2 pa + 8 j^z \omega_0^{(1)} \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \sigma \cos pa - 4 j^{z2} \left(1 + 2 \frac{j^z}{\omega_0^{(1)} - j^z} \cos^2 pa\right) \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \sigma^2, \tag{5.6}$$

$$S_{bs}(p) = 4 \frac{j^a j^z}{\omega_0^{(1)} - j^z} \cos^2 pa - \left[8 \frac{j^{z2}}{\omega_0^{(1)}} (\omega_0^{(1)} - j^z) \left(\frac{j^a}{\omega_0^{(1)} - j^z}\right)^2 \cos pa\right] \sigma + \left[4 j^{a2} j^{z2} \left(\frac{1}{\omega^{(1)2}} - \frac{\omega_0^{(1)2} - 3 j^{z2}}{(\omega_0^{(1)} - j^z)^3 \omega_0^{(1)}} \cos^2 pa\right)\right] \sigma^2 , \tag{5.7}$$

and

$$\omega^{(2)}(p) = \omega_0^{(3)} - j^z \sigma^2 . \tag{5.8}$$

We notice that the dispersion law for the three-magnon bound state is independent of momentum up to second order in σ; this conclusion was also reached by Torrance and Tinkham.[1,3] The shifts of the single-magnon line and the three-magnon bound state lift the degeneracy in the limit of zero field; this fact, however, has no bearing on our previous discussion of $\chi_\perp$ since (5.3) is only valid for $\gamma H_0 \gg j^a$.

VI. ZERO-FIELD LIMIT

In Secs. I–V we developed an approximation scheme for the response functions $\chi_\perp$ and $\chi_\parallel$ to second order in $j^a/\gamma H_0$. Since the validity of the scheme depends on $j^a/\gamma H_0$ being small it is essentially a high-field expansion of $\chi_\perp$ and $\chi_\parallel$. In order to extend the theory to lower values of the field, it is necessary to include the admixture of more than a single bound state, i.e., to terminate the continued fraction at a higher order in $j^a/\gamma H_0$. Such a scheme, however, rapidly becomes unmanageable from an analytical point of view, but can be performed numerically[1,3] under certain simplifying assumptions.

Torrance and Tinkham[1,3] included the admixture of 40 bound states, disregarding the effects of the higher bands, and evaluated the energy eigenvalues by solving numerically the corresponding 40×40 secular determinant. In the zero-field limit, however, simplifications occur and we can under certain conditions find a closed expression for the transverse response $\chi_\perp$ to all orders in j^a.

First of all, we consider for simplification the dispersionless case, i.e., $j^\perp = 0$. Disregarding the ground-state shift Δ_{gs}, the ground-state mode $S_{gs}/(z + \omega_0^{(1)})$, the vertex correction of j^a, and the reduction of $\langle S^z \rangle$ due to correlations in the perturbed ground state, the second-order result (3.37) obtained for $\chi_\perp$ in Sec. III assumes the form

$$\chi_\perp(z) = \frac{-\frac{1}{2}}{z - \omega_0^{(1)} - 4 j^{a2}/(z - \omega_0^{(3)})} . \tag{6.1}$$

Despite the drastic assumptions made, the expression (6.1) evidently contains the basic physics in the problem. Finding the normal modes in (6.1) is tantamount to solving the secular determinantal equation

$$\begin{vmatrix} z - \omega_0^{(1)} & 0 \\ 0 & z - \omega_0^{(3)} \end{vmatrix} = 0 , \tag{6.2}$$

which is just the determinant arising from ordinary time-independent perturbation theory applied to a single admixture. We can now establish the connection between the approach of Torrance and Tinkham[1,3] and the present scheme. These authors showed that the matrix element of j^a between two bound states is equal to $-2j^a$ for $p = 0$. The infinite determinant for the admixture of all the bound states consequently has the form

$$D(E)=\begin{vmatrix} E-\omega_0^{(1)} & -2j^a & \cdots & 0 \\ -2j^a & E-\omega_0^{(3)} & -2j^a & \cdots \\ \cdots & -2j^a & E-\omega_0^{(5)} & -2j^a \\ 0 & \cdots & \cdots & \cdots \end{vmatrix}. \tag{6.3}$$

It is easily seen that the continued fraction

$$\chi_\perp(z)=\cfrac{-\frac{1}{2}}{z-\omega_0^{(1)}-\cfrac{4j^{a^2}}{z-\omega_0^{(3)}-\cfrac{4j^{a^2}}{z-\omega_0^{(5)}-\cdots}}} \tag{6.4}$$

can be expressed in terms of $D(z)$ in the following manner:

$$\chi(z)=-\frac{1}{2}\,\frac{D(z-2\gamma H_0)}{D(z)}\quad. \tag{6.5}$$

The expression (6.5) establishes the connection between the two schemes. In particular we notice that the intensity of the nth line is given by

$$I_0^{(n)}=\frac{1}{2}\,\frac{D(\tilde{\omega}_0^{(n)}-2\gamma H_0)}{D_E'(\tilde{\omega}_0^{(n)})}\quad, \tag{6.6}$$

where $\tilde{\omega}_0^{(n)}$ is the nth root in the determinantal equation $D(E)=0$.

In the zero-field limit, the unperturbed levels are degenerate and $\omega^{(n)}=2j^s$ for all n. In that case we derive the following equation:

$$\chi_\perp(z)=\frac{-\frac{1}{2}}{z-2j^s+8j^{a^2}\chi_\perp(z)}\quad. \tag{6.7}$$

The equation (6.7) can readily be solved; we obtain

$$\chi_\perp(z)=\frac{-1}{(z-2j^s)+[z-2j^s)^2-(4j^a)^2]^{1/2}}\quad. \tag{6.8}$$

The absorptive response is given by

$$\chi_\perp''(\omega)=\frac{\pi[(4j^a+2j^s-\omega)(\omega-2j^s+4j^a)]^{1/2}}{8j^{a^2}}\quad. \tag{6.9}$$

$\chi_\perp(z)$ has a branch cut extending from $2j^s-4j^a$ to $2j^s+4j^a$, i.e., symmetrically around the degeneracy point of the unperturbed levels. The absorptive response has the shape of a semiellipse with height $\pi/2j^a$ and width $8j^a$. As j^a approaches zero, the ellipse degenerates into a δ function corresponding to the excitation of a single-magnon mode. It is interesting to notice that, whereas in the high-field limit the strengths of the higher resonance lines fall off rapidly in powers of $j^a/\gamma H_0$, the intensity gets more evenly shared among the lines as the field decreases. In the zero-field limit the converging lines form a band symmetrically shaped around the degeneracy point $2j^s$.

VII. CONCLUSION

In this paper we have evaluated the transverse and longitudinal dynamical susceptibilities of the anisotropic magnetic chain in an applied field to second order in the transverse anisotropy j^a and to second order in the transverse mean exchange $j^\perp$. Furthermore we have, under certain simplifying assumptions, computed the transverse susceptibility in the absence of $j^\perp$ to all orders in j^a in the limit of zero external field. Because of the nature of the problem, the true expansion parameter is $j^a/\gamma H_0$, where H_0 is the external field. Consequently, the expressions for the susceptibilities can only be considered as valid for high fields, i.e., $\gamma H_0 \gg j^a$.

This fact makes a comparison with the experiments on $CoCl_2 \cdot 2H_2O$ of Torrance and Tinkham[1,2] less interesting since we are only able to predict the initial bending of the three lowest resonance lines in a plot of energy vs field. Preliminary measurements of Nicoli[12] using a far-infrared cyanide laser probe into the interesting regime $\gamma H_0 < j^a$ and indicate that the intensities of the higher resonance lines fall off slowly in accordance with the limiting behavior of the intensity spectrum in the zero-field limit as discussed in Sec. VI.

The perturbation expansions for the transverse and longitudinal susceptibilities only work to second order in j^a. We have been unable to establish a systematic perturbation scheme for the evaluation of the susceptibilities to orders in j^a beyond second order. From the expressions for the susceptibilities in Sec. I it is clear that such a scheme will involve the evaluation of "the self-energy of the self-energy," etc. Such an attempt is, however, beyond the scope of the present paper.

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APPENDIX A

By means of the definition (3.5) of $G_\perp(12)$ and the identity (3.20) we can write the expansion (3.10) in the form

$$G_\perp^{-1}(12)=[G_\perp^{-1}(12)]_{j^a=0}[1-j^a(34)\langle 0\,|\,(S^-(3)\,S^-(4)\,S^+(5)\,S^+(6))_+\,|\,0\rangle j^a(56)]$$

$$-i[G_\perp^{-1}(17)]_{j^a=0}\,j^a(34)\langle 0\,|\,(S^-(3)\,S^-(4)\,S^-(7)\,S^+(8)\,S^+(5)\,S^+(6))_+\,|\,0\rangle j^a(56)\,[G_\perp^{-1}(82)]_{j^a=0}\,. \tag{A1}$$

Using the equation of motion (3.22) and its adjoint we obtain the following lengthy expression for $G_\perp^{-1}$:

$$\begin{aligned}
G_\perp^{-1}(12)= {} & [1-2j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^+(1^+)\,S^-(1)\,S^+(\bar5)\,S^+(\bar6))_+\,|0\rangle j^a(\bar5\bar6)\,]\,[G_\perp^{-1}(12)]_{j^a=0}\\
& +[-4ij^a(\bar3\bar4)\langle 0\,|(S^-(\bar3)\,S^-(\bar4)\,S^+(1)\,S^+(\bar6))_+\,|\,0\rangle j^a(1\bar6)+4j^\perp(1\bar1)\,j^a(\bar3\bar4)\\
& \times\langle 0\,|(S^-(\bar3)\,S^-(\bar4)\,S^-(\bar1)\,S^+(1)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\\
& -4j^z(1\bar1)\,j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^+(\bar1^+)\,S^-(\bar1)\,[1-2S^+(1^+)\,S^-(1)]\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)]\;\delta(12)\\
& +[4j^\perp(12)\,j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^+(1^+)\,S^-(1)\,[1-2S^+(2^+)\,S^-(2)]\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\\
& -4j^z(12)\,j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^-(1)\,S^+(2)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\,]\,[-4j^a(\bar32)\,[G_\perp(\bar3\bar6)\,]_{j^a=0}\,j^a(1\bar6)\\
& +8j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^+(\bar2)\,S^+(2^+)\,S^-(2)\,S^+(\bar6))_+\,|\,0\rangle j^a(1\bar6)\,j^\perp(\bar22)\\
& -8j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^+(\bar2^+)\,S^-(\bar2)\,S^+(2)\,S^+(\bar6))_+\,|\,0\rangle j^a(1\bar6)\,j^z(\bar22)\\
& +8j^\perp(1\bar1)\,j^a(\bar32)\langle 0\,|\,(S^-(\bar3)\,S^+(1^+)\,S^-(1)\,S^-(\bar1)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\\
& -8j^z(1\bar1)\,j^a(\bar32)\langle 0\,|(S^-(\bar3)\,S^-(1^+)\,S^+(\bar1^+)\,S^-(\bar1)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\,]\\
& \times[-16ij^\perp(1\bar1)\,j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^+(1^+)\,S^-(1)\,S^-(\bar1)\,S^+(\bar2)\,S^+(2^+)\,S^-(2)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\,j^\perp(\bar22)\\
& +16ij^\perp(1\bar1)\,j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^+(1^+)\,S^-(1)\,S^-(\bar1)\,S^+(\bar2^+)\,S^-(\bar2)\,S^+(2)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\,j^z(\bar22)\\
& +16ij^z(1\bar1)\,j^a(\bar3\bar4)\langle 0\,|(S^-(\bar3)\,S^-(\bar4)\,S^-(1)\,S^+(\bar1^+)\,S^-(\bar1)\,S^+(\bar2)\,S^+(2^+)\,S^-(2)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\,j^\perp(\bar22)\\
& -16ij^z(1\bar1)\,j^a(\bar3\bar4)\langle 0\,|\,(S^-(\bar3)\,S^-(\bar4)\,S^-(1)\,S^+(\bar1^+)\,S^-(\bar1)\,S^+(\bar2^+)\,S^-(\bar2)\,S^+(2)\,S^+(\bar5)\,S^+(\bar6))_+\,|\,0\rangle j^a(\bar5\bar6)\,j^\perp(\bar22)]\,.
\end{aligned}\tag{A2}$$

Inserting intermediate states by means of the decomposition (3.12), performing the time integrations, and Fourier transforming, we can express (A2) in terms of the equal-time Green's functions (3.13), (3.27), and (3.28). We get for the various terms in the spectral representation (3.25)

$$1-2\langle S^+S^-\rangle=-2\,\mathrm{F.t.}\sum j^a_{kk'}G_{kk'ps}(0)\,G_{psnn'}(0)\,j^a_{nn'}\,, \tag{A3}$$

$$\begin{aligned}
\Delta_{gs}(k)=\mathrm{F.t.}\{[& -4\sum j^a_{kk'}\,G_{kk'ps}(0)\,j^a_{ps}-4\sum j^z_{pq}j^a_{kk'}\,G_{kk'qs}(0)\,G_{qsnn'}(0)\,j^a_{nn'}\\
& +8\sum j^z_{pq}j^a_{kk'}\,G_{kk'pq}(0)\,G_{pqnn'}(0)\,j^a_{nn'}+4\sum j^\perp_{pq}j^a_{kk'}\,G_{kk'ps}(0)\,G_{qsnn'}(0)\,j^a_{nn'}\,]\,\delta_{pp'}\\
& +[-4\sum j^a_{kk'}G_{kk'p's}(0)\,G_{psnn'}(0)\,j^a_{nn'}j^z_{pp'}+4\sum j^a_{kk'}\,G_{kk'ps}(0)\,G_{psnn'}(0)\,j^a_{nn'}j^\perp_{pp'}\\
& -8\sum j^a_{kk'}\,G_{kk'pp'}(0)\,G_{pp'nn'}(0)\,j^a_{nn'}j^\perp_{pp'}\,]\}\,,
\end{aligned}\tag{A4}$$

$$\begin{aligned}
\frac{S_{gs}(k)}{\omega+\omega^{(1)}(k)}=\mathrm{F.t.}[& 4\sum j^a_{ps}\,G_{s's}(-\omega)\,j^a_{s'p'}-8\sum j^a_{kk'}\,G_{kk'sp'}(0)\,G_{sn}(-\omega)\,j^a_{pn}j^z_{sp'}\\
& -8\sum j^z_{ps}j^a_{np'}\,G_{ns}(-\omega)\,G_{pskk'}(0)\,j^a_{kk'}+16\sum j^z_{ps}j^a_{kk'}\,G_{kk's'p'}(0)\,G_{s's}(-\omega)\,G_{psq'q}(0)\,j^a_{qq'}j^z_{s'p'}\\
& +8\sum j^a_{kk'}\,G_{kk'sp'}(0)\,G_{p'n}(-\omega)\,j^a_{pn}j^\perp_{sp'}+8\sum j^\perp_{ps}j^a_{np'}\,G_{np}(-\omega)\,G_{pskk'}(0)\,j^a_{kk'}\\
& +16\sum j^\perp_{ps}j^a_{kk'}\,G_{kk's'p'}(0)\,G_{p'p}(-\omega)\,G_{psqq'}(0)\,j^a_{qq'}j^\perp_{s'p'}-16\sum j^\perp_{ps}j^a_{kk'}\,G_{kk's'p'}(0)\,G_{s'p}(-\omega)\,G_{psqq'}(0)\,j^a_{qq'}j^z_{s'p'}\\
& -16\sum j^z_{ps}j^a_{kk'}\,G_{kk's'p'}(0)\,G_{p's}(-\omega)G_{psqq'}(0)\,j^a_{qq'}J_{s'p'}]\,,
\end{aligned}\tag{A5}$$

$$\gamma_{bs}(k\omega)=T_F\,[16\sum j^z_{ps}j^a_{kk'}\,G_{kk'sr}(0)\,G_{psrp's'r'}(\omega)\,G_{s'r'qq'}(0)\,j^a_{qq'}j^z_{s'p'}$$

$$-16\sum j^z_{ps} j^a_{kk'} G_{kk'sr}(0)\, G_{psrp's'r'}(\omega)\, G_{p'r'qq'}(0)\, j^a_{qq'} j^{\perp}_{s'p'} - 16\sum j^{\perp}_{ps} j^a_{kk'} G_{kk'pr}(0)\, G_{psrp's'r'}(\omega)\, G_{s'r'qq'}(0)\, j^a_{qq'} j^z_{s'p'}$$

$$+16\sum j^{\perp}_{ps} j^a_{kk'} G_{kk'pr}(0)\, G_{psrp's'r'}(\omega)\, G_{p'r'qq'}(0)\, j^a_{qq'} j^{\perp}_{s'p'}] \; . \qquad \text{(A6)}$$

APPENDIX B

From general analyticity it follows that D^{-1} has the spectral representation

$$D^{-1}(p\omega) = 1 + \int \frac{A(p\Omega)}{\Omega - \omega - i\epsilon} \frac{d\Omega}{\pi} \; . \qquad \text{(B1)}$$

Inserting the explicit expressions (4.13) and (4.19) for F^0 and Δ_2 in (4.24) and going to the continuum limit, we obtain

$$D(p\omega) = 1 + 4j^z \int_{-\pi}^{\pi} \frac{dk}{2\pi} \times \frac{\sin^2 k}{\omega - 2\omega_0^{(1)} + 4j^z\sigma\cos\frac{1}{2}p\cos k + i\epsilon} \; . \qquad \text{(B2)}$$

Introducing a new integration variable $u = \cos k$, we get

$$D(p\omega) = 1 + 4j^z \int_1^1 \frac{du}{\pi} \times \frac{(1-u^2)^{1/2}}{\omega - 2\omega_0^{(1)} + 4j^z\sigma u\cos\frac{1}{2}p + i\epsilon} \; . \qquad \text{(B3)}$$

In order to evaluate (B3) we consider the function $F(z)$ given by the integral

$$F(z) = \int_{-1}^{1} \frac{du}{\pi} \frac{(1-u^2)^{1/2}}{u - z} \; . \qquad \text{(B4)}$$

$F(z)$ has a branch cut from -1 to $+1$ with the discontinuity

$$F(\omega + i\epsilon) - F(\omega - i\epsilon) = (1-\omega^2)^{1/2} \qquad \text{(B5)}$$

across the cut and is easily recognized as the function

$$F(z) = (z^2 - 1)^{1/2} - z \; . \qquad \text{(B6)}$$

Alternatively, (B4) can be evaluated by contour integration along the unit circle by means of the substitutions $t = e^{ik}$ and $u = \cos k$. By substitution and choosing the right branch we find

$$D(pz) = 1 + \frac{1}{\sigma\cos\frac{1}{2}pa} \left\{ \frac{z - 2\omega_0^{(1)}}{4j^z\sigma\cos\frac{1}{2}pa} - \left[\left(\frac{z - 2\omega_0^{(1)}}{4j^z\sigma\cos\frac{1}{2}pa}\right)^2 - 1\right]^{1/2} \right\} \; . \qquad \text{(B7)}$$

As is easily verified, D^{-1} has a pole at $\omega_0^{(2)} - 2j^z\sigma^2 \times\cos\frac{1}{2}pa$ and a branch cut extending from $2\omega_0^{(1)} - 4j^z\sigma\cos\frac{1}{2}pa$ to $2\omega_0^{(1)} + 4j^z\sigma\cos\frac{1}{2}pa$. By means of Cauchy's theorem we find the spectral function

$$A(p\Omega) = j^z(1 - \sigma^2\cos^2\tfrac{1}{2}pa)\, 2\pi\delta(\Omega - \omega^{(2)}(p))$$

$$+ \frac{\frac{1}{2}\{[\omega_+^{(2)}(p) - \Omega][\Omega - \omega_-^{(2)}(p)]\}^{1/2}}{\Omega - \omega^{(2)}(p)} \; . \qquad \text{(B8)}$$

APPENDIX C

In order to make use of the solution (4.23) of the two-magnon scattering problem in the evaluation of $\chi_{\parallel}$ we express (3.14) in terms of the Green's function $F_{kqk'q'}$; we obtain for the Fourier transform

$$G_{\parallel}(p\omega) = -\frac{1}{N}\sum j^a\,\Gamma(q)\, F(p+q, -q, p+q', -q', \omega)\,\Gamma(q')j^a - \frac{1}{N}\sum j^a\,\Gamma(q)\, F(-p+q, -q, -p+q', -q', -\omega)\,\Gamma(q')j^a, \qquad \text{(C1)}$$

where we have introduced the vertex function

$$\Gamma(k) = (1/2i)\sum e^{-i(x_p - x_r)k} F_{prkk'}(0)\, (\delta_{kk'-1} - \delta_{kk'+1}) \; . \qquad \text{(C2)}$$

By means of the solution (4.23) it is an easy task to compute the strength of the two-magnon bound-state mode to second order in σ. The bound-state part of F is given by

$$F_{\text{bs}}(kqk'q'\omega) = \frac{\sum 2j^z F^0(kqk''q''\omega)\, \Delta_2(k''q''\tilde{k}''\tilde{q}'')\, F^0(\tilde{k}''\tilde{q}''k'q'\omega)}{D_{\text{bs}}(k+q\,\omega)} \; , \qquad \text{(C3)}$$

where

$$D_{\text{bs}}^{-1}(p\omega) = \frac{-2j^z(1 - \sigma^2\cos^2\frac{1}{2}pa)}{\omega - \omega^{(2)}(p) + i\epsilon} \; . \qquad \text{(C4)}$$

The vertex function $\Gamma(k)$ is given as

$$\Gamma(k) = \frac{\sin ka}{D(00)\,\omega^{(1)}(k)} \; . \qquad \text{(C5)}$$

To second order in σ we obtain

$$\Gamma(k) = \Gamma_0(k) + \Gamma_1(k)\sigma + \Gamma_2(k)\,\tfrac{1}{2}\sigma^2 \; , \qquad \text{(C6)}$$

where

$$\Gamma_0(k) = \frac{\sin ka}{\omega_0^{(1)} - j^z} \; ,$$

$$\Gamma_1(k) = \frac{1}{\omega_0^{(1)} - j^z} \frac{j^z}{\omega_0^{(1)}} \sin 2ka \; , \qquad \text{(C7)}$$

$$\Gamma_2(k) = \frac{2}{\omega_0^{(1)} - j^z}\left(\frac{j^z}{\omega_0^{(1)}}\right)^2 \left(4\cos^2 k\sigma + \frac{j^z}{\omega_0^{(1)} - j^z}\right)\sin ka \; .$$

In a similar fashion we expand $F^0(kqk'q'\omega)$ to second order in σ:

$$F^0 = F_0^0 + F_1^0\sigma + F_2^0\,\tfrac{1}{2}\sigma^2 \; , \qquad \text{(C8)}$$

where

$$F_0^0 = \frac{\Delta_1}{j^z}, \qquad F_1^0 = \frac{\Delta_1}{j^z}(\cos ka + \cos qa),$$
$$F_2^0 = \frac{2\Delta_1}{j^z}\left[(\cos ka + \cos qa)^2 - \cos^2 \tfrac{1}{2}pa\right]. \tag{C9}$$

Inserting Γ, F^0, and $D_{\rm bs}^{-1}$ and collecting terms up to second order in σ we obtain the expression (5.1) for $\chi_{\parallel}(pz)$.

The numerator $1 - 2\langle S^+S^-\rangle$, the ground-state shift $\Delta_{\rm gs}$, and the strength $S_{\rm gs}$ in the spectral form (3.25) can all be expressed in terms of the vertex function $\Gamma(k)$ and thus easily be found to second order in σ. Using (C6) and (C7) we obtain (5.4), (5.5), and (5.6).

In order to evaluate the bound-state part of the mass operator $\gamma_{\rm bs}$, we need an explicit solution of the three-magnon scattering problem to second order in σ. From the formal solution (4.32) of the three-magnon problem we obtain

$$F_{\rm bs} = NA/D, \tag{C10}$$

where, using a well-known identity,

$$D = \det(1 - I) = \exp \operatorname{Tr} \ln(1 - I). \tag{C11}$$

To second order in σ we get

$$F_{\rm bs} = \frac{(NA)_0 + (NA)_1\sigma + (NA)_2 \frac{1}{2}\sigma^2}{D_0 + D_1\sigma + D_2 \frac{1}{2}\sigma^2}. \tag{C12}$$

By means of (C11) we can find D_0, D_1, and D_2 in terms of traces over products of I_0, I_1, and I_2, where I_0, I_1, and I_2 are the coefficients in the expansion of the kernel I to second order in σ. In order to evaluate N_0, N_1, and N_2 we notice that N/D satisfies the integral equation

$$N/D = I + I(N/D). \tag{C13}$$

Expanding both sides of (C13) to second order in σ and solving in succession the resulting separable integral equations for $(N/D)_0$, $(N/D)_1$, and $(N/D)_2$, we can find N_0, N_1, and N_2 and thereby obtain an explicit expression for the bound-state part $F_{\rm bs}$ to second order in σ.

Such procedure is straightforward and leads to a simple but very lengthy calculation. There is, however, a much simpler method which enables us to make contact with ordinary perturbation theory.

Expanding the solution (4.32) for a fixed value of ω in powers of σ we get

$$F_0 = A_0 + \frac{(NA)_0}{D_0}, \tag{C14}$$

$$F_1 = A_1 + \frac{(NA)_1}{D_0} - \frac{(NA)_0}{D_0^2} D_1, \tag{C15}$$

$$F_2 = A_2 + \frac{(NA)_2}{D_0} - \frac{2(NA)_1 D_1 + (NA)_0 D_2}{D_0^2} + \frac{2(NA)_0 D_1^2}{D_0^3}. \tag{C16}$$

From the definitions (3.28) and (4.10) of F we conclude that

$$F_1 = \tfrac{1}{3} j^z F_0 \pi F_0 \tag{C17}$$

and

$$F_2 = \tfrac{2}{3} j^z F_0 \pi F_0 \pi F_0, \tag{C18}$$

where

$$\pi(kqmk'q'm') = \Delta_1(kqmk'q'm')\left[\cos k + \cos q + \cos m\right]. \tag{C19}$$

Using (3.31) and (4.10) and Fourier transforming we get

$$F_0 = -6\left(\frac{p_1}{\omega - 3\omega_0^{(1)} + i\epsilon} + \frac{p_2}{\omega - 2\omega_0^{(1)} - \omega_0^{(2)} + i\epsilon} + \frac{p_3}{\omega - \omega_0^{(3)} + i\epsilon}\right), \tag{C20}$$

where

$$p_1 = \Delta_1 - \tfrac{2}{3}\Delta_2 + \tfrac{2}{3}\Delta_3, \quad p_2 = \tfrac{2}{3}\Delta_2 - \tfrac{4}{3}\Delta_3, \quad p_3 = \tfrac{2}{3}\Delta_3,$$

and

$$\Delta_3 = \tfrac{1}{3}\Delta_2^2 - \tfrac{1}{2}\Delta_2.$$

p_1, p_2, and p_3 are the projection operators for the wave functions of three free magnons (three nonadjacent spin deviations), two bound magnons plus a free one (two adjacent spin deviations and one nonadjacent), and three bound magnons (three adjacent spin deviations), respectively.

Since π is proportional to the Heisenberg part of the Hamiltonian, (C17) and (C18) are readily seen to be the first- and second-order matrix elements of the Heisenberg part taken between the various three-magnon Ising states.

Expanding (C12) around the unperturbed pole $\omega_0^{(3)}$ we get

$$F_{\rm bs} = \frac{[(NA)_0 + (NA)_1\sigma + (NA)_2 \frac{1}{2}\sigma^2]_{\omega=\omega_0^{(3)}} Z_{\rm bs}}{\omega - \omega_0^{(3)} - \Delta_{\rm bs}}, \tag{C21}$$

where

$$\Delta_{\rm bs} = -\left(\frac{D_0 + D_1\sigma + D_2\frac{1}{2}\sigma^2}{D_0' + D_1'\sigma + D_2'\frac{1}{2}\sigma^2}\right)_{\omega=\omega_0^{(3)}} \tag{C22}$$

and

$$Z_{\rm bs}^{-1} = (D_0' + D_1'\sigma + D_2'\tfrac{1}{2}\sigma^2)_{\omega=\omega_0^{(3)}}, \tag{C23}$$

By means of the Eqs. (C14)–(C20), and the identities $p_2\pi p_3 = \pi p_3$, $p_1\pi p_3 = 0$, and $p_3\pi p_3 = 0$, it is easy to identify the various terms in $F_{\rm bs}$. We get

$$F_{bs}=\frac{-6p_3-6(\pi p_3+p_3\pi)\sigma+[18p_3-6(p_3\pi\pi p_2+p_2\pi\pi p_3)-6(p_3\pi\pi+\pi\pi p_3)-12\pi p_3\pi]\frac{1}{2}\sigma^2}{\omega-\omega_0^{(3)}-\Delta_{bs}}\ , \tag{C24}$$

where

$$\Delta_{bs}=-j^z\sigma^2\ . \tag{C25}$$

Inserting (C24) in (A6) and collecting terms to second order in σ, we obtain (5.7). The band contribution to γ_{bs} arising from the three-magnon continuum is in general nonvanishing. A closer inspection, however, shows that it vanishes to second order in σ.

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Anomalous Behavior of the Low-Temperature Magnetic Specific Heat of $NiCl_2 \cdot 2H_2O$

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The specific heat of $NiCl_2 \cdot 2H_2O$ has been measured for $1.2\ K < T < 24.5\ K$. Two peaks are observed, at $T_{1c}=6.309$ K and at $T_{2c}=7.258$ K. The former can be described by a power law $C_{mag}\propto|T-T_{1c}|^{-0.3}$. The T_{2c} peak is asymmetric and displays power-law behavior with smaller exponents. Below ~4 K, $C_p \propto T$, but the line does not extrapolate to zero at $T=0$. The conventional explanations for some previously observed double transitions in other substances do not seem to be applicable to $NiCl_2 \cdot 2H_2O$, thus raising intriguing questions as to the origin of the double C_p peaks. Two possible explanations are sketched.

In order to study the competition between single-ion and exchange effects in Ni^{++} compounds, we have measured the specific heat of $NiCl_2 \cdot 2H_2O$. The structure is monoclinic and belongs to the space group $I2/m$.[1] There are four formula units per unit cell. Two chlorines form an edge of the $(NiCl_4 \cdot 2H_2O)^{--}$ octahedron and link adjacent Ni^{++} ions along the b axis. Although the compounds $MCl_2 \cdot 2H_2O$ (M = Fe, Co, Mn) exhibit very similar features, $NiCl_2 \cdot 2H_2O$ is not isomorphic with these.[1]

Our measurements were performed on a 25.38-g polycrystalline specimen. Small yellow needles of the dihydrate were obtained by slow evaporation at ~75 C from an aqueous solution of reagent grade $NiCl_2 \cdot 6H_2O$. Great care was taken to prevent sample decomposition during encapsulation. Chemical analysis of the specimen showed it to be the dihydrate.[2] Powder and single-crystal x-ray studies on our samples were consistent with the reported[1] structure. Details of the vacuum calorimeter, the phase-sensitive thermometry circuit, and the experimental procedure are discussed elsewhere.[3]

Figure 1 shows our C_p data for $1.2\ K < T < 24.5$ K. There are two peaks, at $T_{1c}=(6.309\pm0.001)$ K and at $T_{2c}=(7.258\pm0.003)$ K, the latter being considerably smaller in magnitude. The quoted uncertainties indicate the maximum scatter from different runs. Below ~4 K, $C_p \propto T$, but the line does not extrapolate to zero at $T=0$. Although double peaks have been observed in other salts,[4] including those