Magnetic excitation spectrum of $CoCl_2 \cdot 2H_2O$†

Hans C. Fogedby*
Department of Physics, Harvard University, Cambridge, Massachusetts 02138
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The magnon bound-state spectrum recently observed in the anisotropic magnetic salt $CoCl_2 \cdot 2H_2O$ is discussed in terms of analytic expressions. The transverse energy spectrum including the coupling to an optic phonon and the longitudinal energy spectrum are evaluated to all orders in j^a/H_0, j^a is the transverse anisotropy $[j^a = (j^x - j^y)/2]$ and H_0 is the applied magnetic field, and to second order in the transverse mean exchange j^h $[j^h = (j^x + j^y)/2]$ and expressed in terms of Bessel functions. In the low-field limit the spectrum is analyzed by means of the quasiclassical method. The transverse and longitudinal intensity spectra have the form of infinite continued fractions and can be expressed in terms of Bessel functions. The low-field and zero-field limits are discussed in detail.

I. INTRODUCTION

The recent results of far-infrared transmission measurements on $CoCl_2 \cdot 2H_2O$ (CC2) at helium temperatures and in high magnetic fields by Torrance and Tinkham[1] utilizing a monochromator and by Nicoli and Tinkham[2] employing a laser have strikingly demonstrated the existence of a multimagnon bound-state spectrum in this highly anisotropic magnetic salt.

Utilizing the broad-band far-infrared radiation from a monochromator, Torrance and Tinkham[1] mapped out the magnetic excitation spectrum in fields ranging up to 55 kOe. The limited energy resolution and weak intensity inherent in the monochromator techniques make it difficult, however, to obtain good data on the magnetic intensity spectrum.

The laser combines an intrinsic small linewidth with a high intensity and is therefore ideally suited to perform relative intensity measurements. Employing a cyanide laser, Nicoli and Tinkham[2] measured the magnetic intensity spectrum at fixed energies and in fields ranging up to 90 kOe.

The above authors[1-3] have shown that the magnetic properties of CC2 can be interpreted in terms of Ising-like chains of strongly exchange-coupled Co^{++} ions. Neighboring chains are weakly bonded by the waters of hydration.

The multimagnon bound-state spectrum is accounted for by the following spin-$\frac{1}{2}$ nearest-neighbor exchange Hamiltonian pertaining to an individual chain of N Co^{++} ions:

$$H_{sp} = -2\sum_{i=1}^{N}[j^z S_i^z S_{i+1}^z + (\tfrac{1}{2}j^h)(S_i^+ S_{i+1}^- + \text{H.c.}) + (\tfrac{1}{2}j^a)(S_i^+ S_{i+1}^+ + \text{H.c.})] + H_0\sum_{i=1}^{N} S_i^z. \tag{1.1}$$

The longitudinal exchange constant j^z (the Ising coupling) is of order 13 cm^{-1}, whereas the transverse mean exchange j^h (the Heisenberg coupling) and the transverse anisotropy j^a are of order 2.1 and 1.3 cm^{-1}, respectively. The applied magnetic field in the z direction, H_0, is measured in units of $g^z\mu_B$, where g^z is the longitudinal g factor and μ_B is the Bohr magneton. For CC2 $g^z\mu_B$ is of order 0.35 cm^{-1}/kOe.

The strong longitudinal anisotropy of the system (j^h/j^z and j^a/j^z are both of order $\frac{1}{10}$) makes the Ising model in an applied field a good zeroth-order approximation. The excitation spectrum of the Ising model is conveniently discussed in terms of clusters of adjacent spin deviations with respect to the aligned ferromagnetic ground state. The lowest multiplet corresponds to a single cluster of spin deviations and has the energies $E_n^0 = 2j^z + nH_0$, where n, $n = 1, 2, \ldots, N-1$, is the number of spin reversals in the cluster. The higher-lying multiplets consist of two or more clusters of spin deviations and have the energies $4j^z + nH_0$, $6j^z + nH_0$, etc. In a plot of energy versus field the multiplets are depicted as fans of straight lines converging at the points $2j^z$, $4j^z$, etc., in the limit of zero field (see Refs. 1 and 3).

The spin clusters of adjacent spin deviations are simple kinds of multimagnon bound states, albeit bound states without spatial motion and in the absence of continuum states.

The transverse mean exchange j^h introduces spatial motion and transforms the localized spin clusters of the Ising model into traveling wave packets, the well-known multimagnon bound states and bands of the anisotropic Heisenberg magnet. The zero field degeneracy of the lowest Ising multiplet is partially lifted (to lowest order in j^h/j^z) and the states are transformed into multimagnon bound states. Similarly, the higher-lying Ising multiplets become multimagnon bands.

The transverse anisotropy j^a breaks the rotational invariance about the z axis (the axis of

magnetization) and gives rise to transitions between the multimagnon bound states. Since the parameter characterizing the transitions within a multiplet is j^a/H_0, the effect of even a small transverse anisotropy is greatly enhanced at low field. Experimentally the presence of j^a is of crucial importance. The transverse anisotropy relaxes the selection rule $|\Delta m|=1$ on the transition probabilities and makes it feasible to observe the multimagnon bound-state spectrum in a ferromagnetic resonance experiment.

The above authors[1-3] investigated the magnetic excitation spectrum of (1.1) in detail and solved numerically the secular problem which arises from treating the transverse mean exchange j^h and the transverse anisotropy j^a as perturbations on the Ising model in an applied field.

The exchange forces between the chains due to the waters of hydration are typically an order of magnitude weaker than the exchange forces within a chain. The weak interchain forces are predominantly antiferromagnetic and give rise to various metamagnetic phases.[1-3] Above 45 kOe the system chooses a ferromagnetic arrangement with all the chains aligned in the direction of the field. In the present paper we shall confine our attention to the ferromagnetic phase. The magnetic field in (1.1) is thus measured relative to the ferromagnetic transition field.

A field-independent excitation was observed in CC2. This mode, which only becomes far-infrared active through hybridization with the single magnon mode, was interpreted as an optic phonon by the authors.[1-3] The phonon appears at the energy $E_{\rm ph}$; $E_{\rm ph}$ is of order 29 $\rm cm^{-1}$. In the numerical study of the secular problem the coupling of the phonon to the single magnon mode was accounted for by introducing a phenomenological off-diagonal matrix element A of order 1.3 $\rm cm^{-1}$.

The optic phonon can be included in the Hamiltonian description by adding the term

$$H_{\rm ph}=E_{\rm ph}b^\dagger b+\frac{A}{(N)^{1/2}}\sum_{i=1}^{N}(S_i^+b+\text{H.c.}) \tag{1.2}$$

to (1.1). The first term in (1.2) is the unperturbed phonon energy in the Bose representation; the second term describes the phenomenological coupling to the single magnon mode. The full Hamiltonian describing the magnetic chains interacting with an optic phonon thus assumes the form

$$\begin{aligned}H=&-2\sum_{i=1}^{N}[j^zS_i^zS_{i+1}^z+(\tfrac{1}{2}j^h)(S_i^+S_{i+1}^-+\text{H.c.})\\&+(\tfrac{1}{2}j^a)(S_i^+S_{i+1}^++\text{H.c.})]+H_0\sum_{i=1}^{N}S_i^z\\&+E_{\rm ph}b^\dagger b+\frac{A}{(N)^{1/2}}\sum_{i=1}^{N}(S_i^+b+\text{H.c.}).\end{aligned} \tag{1.3}$$

In a preceding paper[4] this author computed the transverse and longitudinal frequency-dependent magnetic susceptibilities to second order in j^a and j^h for the Hamiltonian (1.1). Since the characteristic dimensionless parameter is j^a/H_0, this calculation was essentially a high-field expansion and therefore did not lend itself to a useful comparison with experimental data. In two other previous papers[5,6] the energy spectrum and intensity spectrum were obtained analytically (i.e., to all orders in j^a/H_0) for the lowest multiplet in the absence of transverse mean exchange (i.e., $j^h=0$) and neglecting coupling to higher multiplets.

The purpose of the present paper is to utilize the analytic solutions obtained in II and III in order to compute the energy spectrum and intensity spectrum pertaining to (1.3) to all orders in j^a/H_0 and to second order in j^h. With the previous authors[1-3] we continue to confine our attention to the lowest multiplet plus optic phonon and neglect transitions to the higher multiplets.

In Sec. II we discuss the spectrum of (1.3) in some detail and compute energy shifts to leading order in j^h. In Sec. III we set up the secular problem for the lowest multiplet plus optic phonon and obtain an analytic solution to all orders in the transverse anisotropy j^a and to second order in j^h.

We obtain the following implicit expressions for the energy spectrum in terms of Bessel functions:

$$2j^a\frac{J_{-(E+H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}{J_{-(E-H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}+\frac{A^2}{E-E_{\rm ph}}+\Delta_1-\Delta=0 \tag{1.4}$$

for the odd manifold including the optic phonon, and

$$2j^a\frac{J_{-(E-2j^z-\Delta)/2H_0}(2j^a/H_0)}{J_{-(E-2H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}+\Delta_2-\Delta=0 \tag{1.5}$$

for the even manifold, where $\Delta_1=-2j^h$, $\Delta_2=-2j^z(j^h/j^z)^2$, and $\Delta=-j^z(j^h/j^z)^2$.

In Sec. IV we formulate the transverse and longitudinal frequency-dependent magnetic susceptibilities $\chi_\perp$ and $\chi_\parallel$ in terms of continued fractions. In Sec. V we sum these continued fractions and obtain analytic expressions for the intensity spectrum.

The transverse susceptibility is given by

$$\chi_\perp(z)=\left[2j^a\left(\frac{J_{-(z+H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}{J_{-(z-H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}\right)+\frac{A^2}{z-E_{\rm ph}}+\Delta_1-\Delta\right]^{-1}. \tag{1.6}$$

The longitudinal susceptibility has the form

$$\chi_{\parallel}(z)=\left[2j^a\left(\frac{J_{-(z-2j^z-\Delta)/2H_0}(2j^a/H_0)}{J_{-(z-2H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}\right)+\Delta_2-\Delta\right]^{-1}+(z\to -z). \tag{1.7}$$

The results reported in the present paper corroborate the numerical results obtained by the previous authors.[1-3] The availability of analytic expressions, however, permits a more complete discussion of the spectrum, in particular of the interesting low-field limit.

II. ANISOTROPIC LINEAR CHAIN

A chain of Co^{++} ions interacting with an optic phonon is described by the Hamiltonian

$$H=-2\sum_{i=1}^{N}[j^z S_i^z S_{i+1}^z+(\tfrac{1}{2}j^h)(S_i^+S_{i+1}^-+\text{H.c.})+(\tfrac{1}{2}j^a)(S_i^+S_{i+1}^++\text{H.c.})]+H_0\sum_{i=1}^{N}S_i^z+E_{\text{ph}}b^\dagger b+\frac{A}{(N)^{1/2}}\sum_{i=1}^{N}(S_i^+b+\text{H.c.}). \tag{2.1}$$

Following the previous authors[1-3] we introduce as basis set the unperturbed states of the Ising model pertaining to the lowest multiplet plus a state characterizing the unperturbed phonon, i.e.,

$$\Psi_n^0=\frac{1}{(N)^{1/2}}\sum_{i=1}^{N}\exp[ik(x_i+\tfrac{1}{2}(n-1)a)]\times S_i^+S_{i+1}^+\cdots S_{i+n-1}^+|0\rangle,\qquad \Phi^0=b^\dagger|0\rangle. \tag{2.2}$$

The Bloch states Ψ_n^0 have the energies $E_n^0=2j^z+nH_0$ and are orthonormal provided $\langle 0|0\rangle=1$, where $|0\rangle$ denotes the aligned ferromagnetic ground state as well as the empty phonon vacuum (i.e., $S_i^-|0\rangle=b|0\rangle=0$). The normalized phonon state Φ^0 has the energy E_{ph} and is orthogonal to Ψ_n^0.

As discussed previously, the effect of j^h does not change the energy spectrum in any substantial way. We shall therefore include j^h to lowest nonvanishing order by means of simple perturbation theory. Since j^h/j^z is of order $\frac{1}{10}$ such an approximation should be adequate in describing the excitation spectrum of CC2.

Following Torrance and Tinkham[3] the energy shift of the Ising state Ψ_1^0 is to lowest order given by $E_1^0=\Psi_1^{0*}H^h\Psi_1^0$, where H^h is the Heisenberg part of (2.1), i.e.,

$$H^h=-\sum_{i=1}^{N}j^h(S_i^+S_{i+1}^-+\text{H.c.}). \tag{2.3}$$

Utilizing the Bloch states (2.2) in evaluating the energy shift ΔE_1^0, we arrive at the result

$$\Delta E_1^0=-2j^h\cos(ka). \tag{2.4}$$

Hence, the dispersion law for the single magnon mode is to lowest order,

$$E_1=E_1^0+\Delta E_1^0=2j^z-2j^h\cos(ka)+H_0, \tag{2.5}$$

where a is the lattice distance (of the order 4 Å in CC2) and the wave number k is restricted to the first Brillouin zone, $-\pi/a<k<\pi/a$. For accidental reasons the higher-order corrections to the energy shift vanish identically, and (2.5) is the exact single magnon dispersion law of the anisotropic Heisenberg chain.

The evaluation of the shifts of $\Psi_n^0(n>1)$ to lowest nonvanishing order in j^h requires the introduction of a more elaborate Ising basis including the higher multiplets. This calculation was performed by Torrance and Tinkham[3] and we shall simply quote their results.

To lowest order the shift of Ψ_2^0 is given by

$$\Delta E_2^0=-2j^z(j^h/j^z)^2\cos^2(\tfrac{1}{2}ka). \tag{2.6}$$

The two-magnon dispersion law is consequently given by

$$E_2=E_2^0+\Delta E_2^0=2j^z-2j^z(j^h/j^z)^2\times\cos^2(\tfrac{1}{2}ka)+2H_0. \tag{2.7}$$

The higher-order corrections to the energy shift again fortuitously vanish, and (2.7) is the exact two-magnon dispersion law of the anisotropic Heisenberg chain.

The shifts of the higher-lying Ising states Ψ_n^0 $(n>2)$ are to second order in j^h given by

$$\Delta E_n^0=-j^z(j^h/j^z)^2\quad(n>2). \tag{2.8}$$

We notice that to leading order $(j^h/j^z)^2$ the energy shifts are the same for $n>2$ and furthermore are k independent.

We conclude that to order $(j^h/j^z)^2$ the Heisenberg part of (2.1) only partially lifts the zero field degeneracy. The states of the multiplet for $n>2$ remain degenerate at zero field whereas the single-magnon state and the two-magnon state are subject to negative shifts with respect to the higher multimagnon states. In our treatment of the secular problem in Sec. III, we include the effects of j^h to second order by shifting the Ising levels by the amounts ΔE_n^0.

In a far-infrared resonance experiment the wavelength of the incoming radiation (~0.03 Å) is much larger than the lattice distance a, i.e., $ka\ll 1$. We shall therefore in the following confine our attention to the zero-wave-number part of the spectrum.

In Fig. 1 we have depicted the energy spectrum of the lowest multiplet plus optic phonon, including the corrections due to j^h. For CC2 the shifts are $\Delta_1=\Delta E_1^0\sim -4\ \text{cm}^{-1}$, $\Delta_2=\Delta E_2^0\sim -0.6\ \text{cm}^{-1}$, and $\Delta=\Delta E_{n>2}^0\sim -0.3\ \text{cm}^{-1}$.

III. SECULAR PROBLEM–ANALYTIC SOLUTION

The secular problem is set up along conventional lines. We consider the eigenvalue equation

$$H\Psi = E\Psi, \tag{3.1}$$

where H is the Hamiltonian (2.1). Confining our attention to the lowest multiplet including the optic phonon we expand the eigenstate Ψ on the basis $\{\Psi_n^0, \Phi^0\}$,

$$\Psi = \sum_{n=1}^{\infty} c_n \Psi_n^0 + a\Phi^0. \tag{3.2}$$

Inserting the expansion (3.2) in (3.1) and evaluating the appropriate matrix elements using the Hamiltonian (2.1) and the basis (2.2) (for $k=0$) we arrive at the following equations for the expansion coefficients c_n and a:

$$E_{\mathrm{ph}}a + Ac_1 = Ea, \tag{3.3a}$$

$$(E_1^0 + \Delta_1)c_1 + Aa - 2j^a c_3 = Ec_1, \tag{3.3b}$$

$$(E_2^0 + \Delta_2)c_2 - 2j^a c_4 = Ec_2, \tag{3.3c}$$

$$(E_n^0 + \Delta)c_n - 2j^a(c_{n+2} + c_{n-2}) = Ec_n. \tag{3.3d}$$

The effect of the transverse mean exchange j^h is taken into account to second order in the eigenvalue equations (3.3a)–(3.3d) by means of the shifts Δ_1, Δ_2, and Δ.

By inspection of the equations (3.3a)–(3.3d) we notice that the states for even values of n, the even manifold, are dynamically decoupled from the states corresponding to odd values of n, the odd manifold. This is due to the fact that the transverse anisotropy j^a induces transitions governed by the selection rule $|\Delta m| = 2$. The optic phonon only interacts with the odd manifold. It is directly coupled to the single-magnon mode by the matrix element A and indirectly coupled to the rest of the odd manifold by j^a.

The boundary condition for the *odd manifold including the phonon* is most conveniently expressed by first introducing the expansion coefficient c_{-1} as a particular linear combination of the phonon amplitude a and the magnon amplitude c_1,

$$c_{-1} = -(A/2j^a)a - [(\Delta_1 - \Delta)/2j^a]c_1. \tag{3.4}$$

From (3.3a) we then obtain the boundary condition

$$2j^a c_{-1} = -A^2/(E - E_{\mathrm{ph}}) + (\Delta_1 - \Delta)c_1. \tag{3.5}$$

Furthermore, with the definition (3.4) of c_{-1} the recursion formula (3.3d) is valid for $n > 0$. The boundary condition (3.5) and the recursion formula (3.3d) together determine the eigenvalue problem. Since $E_n^0 = 2j^z + nH_0$ we recognize (3.3d) as the recursion formula[7]

$$J_{n+1}(z) + J_{n-1}(z) = (2n/z)J_n(z) \tag{3.6}$$

for the Bessel function. As in II we immediately obtain the explicit solution to the recursion formula (3.3d),

$$c_n = J_{-(E - E_n^0 - \Delta)/2H_0}(2j^a/H_0), \tag{3.7}$$

where B is an arbitrary constant. By means of the solution (3.7) and the boundary condition (3.5) we arrive at the following expression for the *energy spectrum of the odd manifold including the phonon*:

$$2j^a \frac{J_{-(E + H_0 - 2j^z - \Delta)/2H_0}(2j^a/H_0)}{J_{-(E - H_0 - 2j^z - \Delta)/2H_0}(2j^a/H_0)} + \frac{A^2}{E - E_{\mathrm{ph}}} + \Delta_1 - \Delta = 0. \tag{3.8}$$

In the case of the *even manifold* we introduce the expansion coefficient c_0,

$$c_0 = [(\Delta - \Delta_2)/2j^a]c_2. \tag{3.9}$$

With the above definition of c_0 the recursion formula (3.3d) is valid for $n > 1$. By means of the solution (3.7) and the boundary condition (3.9) we obtain the following expression for the *energy spectrum of the even manifold*:

$$2j^a \frac{J_{-(E - 2j^z - \Delta)/2H_0}(2j^a/H_0)}{J_{-(E - 2H_0 - 2j^z - \Delta)/2H_0}(2j^a/H_0)} + \Delta_2 - \Delta = 0. \tag{3.10}$$

The expressions (3.8) and (3.10) yield the energy spectrum of the *lowest multiplet including the phonon* in terms of tabulated Bessel functions.[8] We notice that for $A = \Delta_1 = \Delta_2 = \Delta = 0$ the expressions (3.8) and (3.10) are in agreement with the results obtained in II.

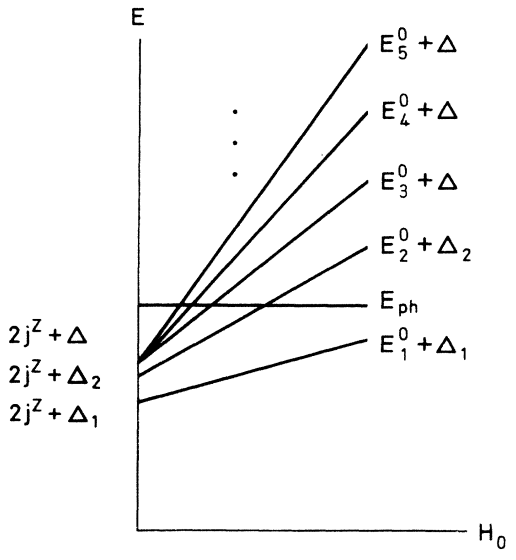

FIG. 1. Energy spectrum of the lowest multiplet including corrections due to j^h (arbitrary units).

A. High-field limit (weak coupling)

In the high-field limit we can, as emphasized in I, II, and III, do perturbation theory in the transverse anisotropy j^a. The *effective coupling* characterizing the transitions within a multiplet is determined by the dimensionless parameter j^a/H_0. A perturbation expansion in powers of j^a thus leads to an expansion in powers of $1/H_0$, i.e., valid at high field.

Inserting the well-known power series expansion[7] of the Bessel function,

$$J_p(z)=\left(\frac{z}{2}\right)^p \sum_{n=0}^{\infty} \frac{(-1)^n}{n!}\left(\frac{z}{2}\right)^{2n} \frac{1}{\Gamma(n+p+1)} \tag{3.11}$$

in (3.8) and (3.10) and utilizing the recursion formula,[7] $\Gamma(z+1)=z\Gamma(z)$, for the Γ function, we are led to the following expressions for the energy spectrum:

$$\sum_{n=0}^{\infty} \frac{(-1)^n}{n!}\left(\frac{j^a}{H_0}\right)^{2n} \times \frac{E-E^0_{2n+1}-\Delta_1-A^2/(E-E_{\rm ph})}{\Gamma((E^0_{2n+3}+\Delta-E)/2H_0)}=0 \tag{3.12}$$

for the *odd manifold including the phonon*, and

$$\sum_{n=0}^{\infty} \frac{(-1)^n}{n!}\left(\frac{j^a}{H_0}\right)^{2n} \times \frac{E-E^0_{2(n+1)}-\Delta_2}{\Gamma((E^0_{2(n+2)}+\Delta-E)/2H_0)}=0 \tag{3.13}$$

for the *even manifold*.

The expressions (3.12) and (3.13) give the solution the eigenvalue problem in terms of power series expansions in j^a/H_0. To finite order in j^a/H_0 the energy spectrum is determined by the roots of two polynomials. The polynomials are equivalent to the finite-order secular determinant arising from the eigenvalue equation (3.1). For purposes of illustration we consider (3.12) to zeroth order in j^a/H_0,

$$[E-E^0_1-\Delta_1-A^2/(E-E_{\rm ph})]/\Gamma((E^0_3+\Delta-E)/2H_0)=0. \tag{3.14}$$

The first factor in (3.14) corresponds to the secular determinant

$$\begin{vmatrix} E-(E^0_1+\Delta) & A \\ A & E-E_{\rm ph} \end{vmatrix} \tag{3.15}$$

describing the interaction and hybridization of the phonon with the single magnon state. The second factor in (3.14) vanishes at the positions of the poles of the Γ function, i.e., for $(E^0_3+\Delta-E)/2H_0 = 0, -1, -2, \ldots$, and thus determines the energies $E^0_n+\Delta$ $(n>2)$ of the unperturbed levels in the multiplet.

The evaluations of (3.12) and (3.13) to higher order in j^a/H_0 rapidly become unmanageable analytically. Torrance and Tinkham[3] diagonalized numerically a 40×40 secular matrix and obtained excellent agreement with the experimental data even in the low-field regime.

B. Low-field limit (strong coupling)

In the low-field limit the unperturbed multimagnon levels approach the degeneracy point $2j^z+\Delta$. The effective coupling j^a/H_0 characterizing the transitions within the multiplet becomes large and the perturbation expansion (3.12) and (3.13) cease to be useful. As the field approaches zero both the order and the argument in the Bessel function solutions (3.8) and (3.10) assume large values and we can as in II and III make use of a well-known double asymptotic expansion[7] of the Bessel function in discussing the low-field limit.

In the present paper, however, we shall choose a more physical approach[9] and relate the low-field properties of the eigenvalue equations (3.3a)–(3.3d) to the one-dimensional motion of a particle of mass $1/16j^a$ in a wedge potential consisting of an infinite potential wall and a constant force potential of slope H_0.

As also discussed in II and III, the recursion formula (3.3d) admits in the *zero-field limit* the continuum solution

$$E=2j^z+\Delta-4j^a\cos\lambda, \quad 0\leq\lambda\leq\pi \tag{3.16a}$$

$$c_n=B\sin(\tfrac{1}{2}\lambda n+\phi), \tag{3.16b}$$

where B is an arbitrary constant (to be determined by normalization requirements) and ϕ an arbitrary phase (to be determined by the boundary condition). In the vicinity of the lower continuum edge, i.e., for $\lambda\ll1$, the expansion coefficient c_n is a slowly varying function of n. Assuming that this behavior also holds for a small but finite field, we replace the difference term $c_{n+2}+c_{n-2}$ in (3.3d) by $2c(n)+4c''(n)$, where we have treated $c_n\cong c(n)$ as a continuous function of n. In the presence of the field the values of n for which c_n is slowly varying depend both on the field and the energy. By inspection of the recursion formula (3.3d) we notice that provided $(E-2j^z-\Delta+4j^a-nH_0)\ll4j^a$ the recursion formula can be written in the form

$$-8j^ac''(n)+nH_0c(n)=(E-2j^z-\Delta+4j^a)c(n) \tag{3.17}$$

for

$$E-2j^z-\Delta+4j^a-nH_0\ll4j^a.$$

The expression (3.17) is the Schrödinger equation for a particle of mass $\frac{1}{16}j^a$ moving in the potential nH_0. The quantum number n plays the role of the

position of the particle. The energy is measured relative to the lower continuum edge $2j^z+\Delta-4j^a$.

The solution of the Schrödinger equation for a particle executing a one-dimensional motion under the influence of a constant force is given in terms of Airy functions[10] which, incidentally, are related to the asymptotic behavior of the Bessel function.[8] For our present purposes, however, it suffices to use the quasiclassical approximation. For the accessible motion to the left of the potential nH_0 the wave function $c(n)$ is given by the expression[10]

$$c(n)=\frac{B}{p(n)^{1/2}}\sin\left(\int_n^{(E-2j^z-\Delta+4j^a)/H_0}p(n')\,dn'+\tfrac{1}{4}\pi\right), \tag{3.18}$$

where $p(n)$ is the canonical momentum

$$p(n)=[(E-2j^z-\Delta+4j^a-nH_0)/8j^a]^{1/2}. \tag{3.19}$$

Evaluating (3.18) using (3.19) we arrive at the expression

$$c(n)=\frac{B}{p(n)^{1/2}}\sin[16j^a p(n)^3/3H_0+\tfrac{1}{4}\pi], \tag{3.20}$$

where B is an arbitrary constant. The quasiclassical approximation holds provided the wavelength $\sim 1/p(n)$ of the particle is slowly varying, i.e., $d[1/p(n)]/dn \ll 1$. Using (3.14) we obtain the condition

$$16j^a p(n)^3/H_0 \gg 1. \tag{3.21}$$

The quasiclassical condition (3.21) together with the assumption (3.17) yield the bounds

$$H_0/16j^a \ll p(n)^3 \ll 1, \tag{3.22}$$

which can only be satisfied provided $H_0/16j^a \ll 1$, i.e., in the low-field limit.

In order to determine the energy spectrum in the low-field limit we invoke the boundary conditions (3.5) and (3.9). For the *even manifold* we obtain

$$\left(\frac{p(2)}{p(0)}\right)^{1/2}\frac{\sin[16j^a p(0)^3/3H_0+\frac{1}{4}\pi]}{\sin[16j^a p(2)^3/3H_0+\frac{1}{4}\pi]}=\frac{\Delta-\Delta_2}{2j^a}, \tag{3.23}$$

whereas for the *odd manifold including the phonon* the boundary condition assumes the form

$$\left(\frac{p(1)}{p(-1)}\right)^{1/2}\frac{\sin[16j^a p(-1)^3/3H_0+\frac{1}{4}\pi]}{\sin[16j^a p(1)^3/3H_0+\frac{1}{4}\pi]}=\left(\Delta-\Delta_1-\frac{A^2}{E-E_{ph}}\right)\Big/2j^a. \tag{3.24}$$

Before we discuss the implications of the rather complicated boundary conditions (3.23) and (3.24) it is elucidating to consider the *even manifold* in the absence of j^h, i.e., for $\Delta=\Delta_2=0$. The boundary condition (3.23) assumes the simple form

$$c(0)=\frac{B}{p(0)^{1/2}}\sin[16j^a p(0)^3/3H_0+\tfrac{1}{4}\pi]=0, \tag{3.25}$$

i.e., the wave function $c(n)$ vanishes at $n=0$. This boundary condition corresponds to the presence of an infinite potential wall at $n=0$ and we conclude that the low-field energy spectrum near the lower continuum edge of the even manifold corresponds to the one-dimensional motion of a particle of mass $1/16j^a$ in the wedge potential

$$U(n)=\begin{cases}\infty & \text{for } n<0\\ nH_0 & \text{for } n>0.\end{cases} \tag{3.26}$$

At a nonvanishing field the motion is bounded by the potential walls and the energy spectrum is discrete. As the field approaches zero the motion becomes unbounded in the right half-plane and the discrete spectrum undergoes a singular transition to a continuous spectrum.

The explicit form of the energy spectrum is obtained from the boundary condition (3.25),

$$16j^a p(0)^3/3H_0+\tfrac{1}{4}\pi=\pi(\nu+1), \tag{3.27}$$

where in the quasiclassical approximation the integer ν is large and positive. The expression (3.27) is readily recognized as the Bohr-Sommerfeld quantization condition for the quasiclassical motion in the potential (3.26),

$$\int_0^{(E-2j^z+4j^a)/H_0}p(n')\,dn'=\pi(\nu+\tfrac{3}{4}). \tag{3.28}$$

Solving (3.27) for E using (3.19) we arrive at the expression

$$E=2j^z-4j^a+2j^a[3\pi H_0(\nu+\tfrac{3}{4})/2j^a]^{2/3} \tag{3.29}$$

which coincides with the energy spectrum derived in Sec. II. In accordance with the bounds (3.22) this expression is only valid for $\nu\gg 1$ and $H_0(\nu+\frac{3}{4})/j^a\ll 1$. The analysis in II shows, however, that (3.29) is valid even in the low quantum-number limit provided we choose the field small enough.

The expression (3.29) exhibits the singular nature of the zero-field transition. The energy spectrum in units of j^a has an algebraic singularity of order $-\frac{2}{3}$ in the coupling parameter j^a/H_0. The energy spectrum is thus not differentiable in j^a/H_0 and could not have been obtained from finite order perturbation theory in j^a/H_0. Since $dE/dH_0 \sim H_0^{-1/3}$ the energy levels approach the lower band edge $2j^z-4j^a$ with infinite slope. By comparison of (3.29) with the zero-field spectrum (3.16a) we infer the correspondence

$$\lambda\cong[3\pi H_0(\nu+\tfrac{3}{4})/2j^a]^{1/3} \tag{3.30}$$

for $\nu\to\infty$, $H_0\to 0$, and $\lambda\ll 1$. Similarly, it can be

shown that the wave function (3.20) assumes the form (3.10b) in the zero-field limit.

For the *odd manifold* in the absence of j^h and in the absence of the phonon coupling, i.e., $\Delta = \Delta_1 = A = 0$, the boundary condition (3.24) takes the form

$$c(-1) = [B/p(-1)^{1/2}]\sin[16j^a p(-1)^3/3H_0 + \tfrac{1}{4}\pi] = 0 \tag{3.31}$$

corresponding to an infinite potential wall at $n = -1$. In a similar fashion to our derivation of (3.29) we obtain the spectrum

$$E = 2j^z - 4j^a + 2j^a\{[3\pi H_0(\nu + \tfrac{3}{4})/2j^a]^{2/3} - H_0/2j^a\} \ . \tag{3.32}$$

We notice, however, that to leading order in H_0/j^a the expressions (3.29) and (3.32) are identical. In the low-field limit the strong effective coupling ($j^a/H_0 \gg 1$) between the levels renders the spectrum insensitive to the boundary condition.

In Fig. 2 we have sketched the wedge potential $U(n)$ and the energy levels in the case of the even manifold.

In order to examine the accumulation of energy levels in the vicinity of the lower band edge it is instructive to evaluate the *density of states* $\rho(E)$ in the low-field limit. The density of states is given by

$$\rho(E) = \sum_\nu \delta(E - E_\nu) \cong \frac{1}{dE/d\nu} \ . \tag{3.33}$$

From (3.29) or (3.32) we obtain

$$\rho(E) = (1/2\pi)(1/H_0)[(E - 2j^z + 4j^a)/2j^a]^{1/2} \ . \tag{3.34}$$

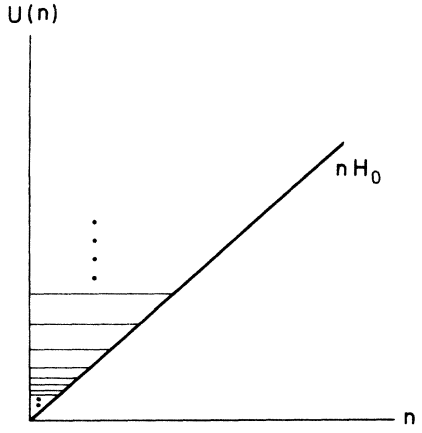

FIG. 2. Wedge potential and the energy levels in the case of the even manifold (arbitrary units).

The density of states has a square-root dependence on the energy at the lower band edge and diverges as the field approaches zero.

In Fig. 3 we have plotted the density of states near the lower band edge.

The presence of the transverse mean exchange j^h and the optic phonon gives rise to energy-dependent phase shifts of the quasiclassical wave functions.

From the boundary condition (3.23) for the *even manifold* we infer

$$16j^a p(0)^3/3H_0 + \tfrac{1}{4}\pi = \phi_{\text{even}} + \pi(\nu + 1) \tag{3.35}$$

or

$$E = 2j^z + \Delta - 4j^a + 2j^a \times [3\pi H_0(\phi_{\text{even}}/\pi + \nu + \tfrac{3}{4})/2j^a]^{2/3} \ , \tag{3.36}$$

where ν is a positive integer. The phase shift ϕ_{even} is to lowest order in $(\Delta - \Delta_2)/2j^a$ [for CC2 $(\Delta - \Delta_2)/2j^a$ is of order $\frac{1}{10}$] and to leading order in H_0/j^a given by

$$\phi_{\text{even}} \simeq [(\Delta - \Delta_2)/j^a](3H_0/16j^a)^{1/3}\pi(\nu + \tfrac{3}{4}) \ . \tag{3.37}$$

The boundary condition (3.24) for the *odd manifold including the phonon* yields the spectrum

$$E = 2j^z + \Delta - 4j^a + 2j^a \times \{[3\pi H_0(\phi_{\text{odd}}/\pi + \nu + \tfrac{3}{4})/2j^a]^{2/3} - H_0/2j^a\} \ , \tag{3.38}$$

where ν is a positive integer. The phase shift ϕ_{odd} is to leading order in H_0/j^a and near the lower band edge given by

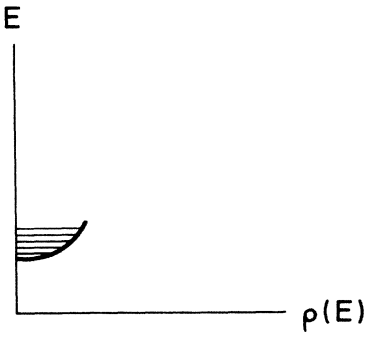

FIG. 3. Density of states near the lower band edge (arbitrary units).

$$\phi_{\text{odd}} \simeq -\frac{[\Delta-\Delta_1-A^2/(2j^z+\Delta-4j^a-E_{\text{ph}})]2\pi(3H_0/16j^a)^{1/3}(\nu+\frac{3}{4})}{\Delta-\Delta_1-A^2/(2j^z+\Delta-4j^a-E_{\text{ph}})-2j^a} \quad . \tag{3.39}$$

The phase shifts ϕ_{even} and ϕ_{odd} due to the altered boundary conditions are vanishingly small in the low-field limit and in the vicinity of the lower band edge. Similarly, the density of states (3.34) is unchanged.

In Sec. V we continue our discussion of the low-field energy spectrum; in particular, the hybridization of the phonon with the odd manifold and the separation of the single magnon and two-magnon states from the continuum states.

IV. MAGNETIC SUSCEPTIBILITIES–CONTINUED FRACTIONS

A convenient and compact theoretical description of the results obtained in a far-infrared transmission experiment on a magnetic system is afforded by introducing the uniform frequency-dependent magnetic susceptibilities.[11]

The magnetic susceptibility is defined as the linear response[11] of the magnetization of the system to a uniform oscillating magnetic field. In order to derive an explicit expression for the susceptibility we add the dipole term

$$H_{\text{dip}} = -\sum_{\alpha=x,y,z} \tfrac{1}{2}(h^\alpha e^{-i\omega t}e^{-\epsilon t}+\text{c.c.})\sum_{i=1}^{N}\gamma^\alpha S_i^\alpha \tag{4.1}$$

to the Hamiltonian (2.1). The causal character of the magnetic response is taken into account as usual by adiabatically switching on the magnetic field. This is accomplished by introducing the factor $e^{-\epsilon t}$ in the definition of the uniform oscillating magnetic field,

$$h^\alpha(t) = \tfrac{1}{2}(h^\alpha e^{-i\omega t}e^{-\epsilon t}+\text{c.c.}) \quad . \tag{4.2}$$

The magnetic field interacts with the total magnetization,

$$M^\alpha = \gamma^\alpha S^\alpha = \sum_{i=1}^{N}\gamma^\alpha S_i^\alpha \tag{4.3}$$

by the usual dipole coupling. γ^α is the effective g factor (in units of the Bohr magneton μ_B), and the index α runs through the Cartesian components x, y, and z. From ordinary time-dependent perturbation theory[10,11] we obtain to linear order in h^α the following expression for the driven part of the total magnetization:

$$\delta M^\alpha(t)_{00} = \gamma^\alpha \delta S^\alpha(t)_{00}$$
$$= \tfrac{1}{2}\gamma^\alpha \sum_{\beta,m}\left(\frac{S_{0m}^\alpha S_{m0}^\beta}{\omega_{m0}-(\omega+i\epsilon)}+\frac{S_{0m}^\beta S_{m0}^\alpha}{\omega_{m0}+(\omega+i\epsilon)}\right)$$
$$\times\gamma^\beta h^\beta e^{-i\omega t}+\text{c.c.} \tag{4.4}$$

In the expression (4.4) S_{mn}^α is the matrix element $\Psi_m^*\sum_{i=1}^{N}S_i^\alpha\Psi_n$, where Ψ_n is an eigenstate of the Hamiltonian H with energy E_n ($H\Psi_n = E_n\Psi_n$). The excitation energy $E_m - E_n$ is denoted by ω_{mn}. Since the system is initially in the ground state Ψ_0, we have computed the average value of the magnetization in the ground state. From (4.4) we conclude that the uniform frequency-dependent susceptibility at zero temperature is given by

$$\chi^{\alpha\beta}(\omega+i\epsilon) = \frac{1}{2N}\sum_m \gamma^\alpha\left(\frac{S_{0m}^\alpha S_{m0}^\beta}{\omega_{m0}-(\omega+i\epsilon)}+\frac{S_{0m}^\beta S_{m0}^\alpha}{\omega_{m0}+(\omega+i\epsilon)}\right)\gamma^\beta \quad . \tag{4.5}$$

For future references we give here the corresponding expression at finite temperature.

$$\chi^{\alpha\beta}(\omega+i\epsilon) = \frac{1}{2N}\sum_{mn} p_n\gamma^\alpha\left(\frac{S_{nm}^\alpha S_{mn}^\beta}{\omega_{mn}-(\omega+i\epsilon)}+\frac{S_{nm}^\beta S_{mn}^\alpha}{\omega_{mn}+(\omega+i\epsilon)}\right)\gamma^\beta \quad . \tag{4.6}$$

In (4.6) we have averaged over the initial states Ψ_n with the Boltzmann weighting factor

$$p_n = \exp(-E_n/k_BT)\Big/\sum_n \exp(-E_n/k_BT) \quad ,$$

T is the temperature and k_B is the Boltzmann constant.

As in I, it is convenient to discuss the dynamical properties in terms of two normalized magnetic susceptibilities, $\chi_\perp(z)$ and $\chi_\parallel(z)$, corresponding to transverse and longitudinal polarizations, respectively. Consequently, we introduce

$$\chi_\perp(\omega+i\epsilon) = \frac{1}{2N}\sum_m\left(\frac{|S_{m0}^+|^2}{\omega_{m0}-(\omega+i\epsilon)}+\frac{|S_{m0}^-|^2}{\omega_{m0}+(\omega+i\epsilon)}\right) \tag{4.7}$$

and

$$\chi_\parallel(\omega+i\epsilon) = \frac{1}{2N}\sum_m |S_{m0}^z|^2\left(\frac{1}{\omega_{m0}-(\omega+i\epsilon)}+\frac{1}{\omega_{m0}+(\omega+i\epsilon)}\right) \quad , \tag{4.8}$$

where $S_{m0}^\pm = S_{m0}^x \pm iS_{m0}^y$.

The magnetic susceptibilities $\chi_\perp$ and $\chi_\parallel$ are in general complex corresponding to a phase difference between the driving field and the magnetization. Employing the well-known relationship $1/(\omega+i\epsilon)=P(1/\omega)-i\pi\delta(\omega)$ we obtain

$$\chi_{\perp,\parallel}(\omega+i\epsilon)=\chi'_{\perp,\parallel}(\omega)+i\chi''_{\perp,\parallel}(\omega)\ , \tag{4.9}$$

where the real (reactive) and imaginary (dissipative) parts, $\chi'_{\perp,\parallel}$ and $\chi''_{\perp,\parallel}$ are

$$\chi'_\perp(\omega)=\frac{1}{2N}P\sum_m\left(\frac{|S^+_{m0}|^2}{\omega_{m0}-\omega}+\frac{|S^-_{m0}|^2}{\omega_{m0}+\omega}\right)\ , \tag{4.10}$$

$$\chi''_\perp(\omega)=\frac{\pi}{2N}\sum_m\left[|S^+_{m0}|^2\delta(\omega_{m0}-\omega)-|S^-_{m0}|^2\delta(\omega_{m0}+\omega)\right] \tag{4.11}$$

and

$$\chi'_\parallel(\omega)=\frac{1}{2N}P\sum_m|S^z_{m0}|^2\left(\frac{1}{\omega_{m0}-\omega}+\frac{1}{\omega_{m0}+\omega}\right)\ , \tag{4.12}$$

$$\chi''_\parallel(\omega)=\frac{\pi}{2N}\sum_m|S^z_{m0}|^2\times[\delta(\omega_{m0}-\omega)-\delta(\omega_{m0}+\omega)]\ . \tag{4.13}$$

The phase difference $\Delta\phi_{\perp,\parallel}$ between the magnetization and the driving field is given by

$$\tan\Delta\phi_{\perp,\parallel}=\chi''_{\perp,\parallel}/\chi'_{\perp,\parallel}\ . \tag{4.14}$$

In the zero-frequency limit the response is static and the magnetization is in phase with the driving field. For each resonance $\omega=\omega_{m0}$ that we pass through, the phase difference increases by π. Only at resonance does energy absorption take place, and it is easily shown[11] that the relative intensity spectrum is given by the expression

$$I_{\perp,\parallel}(\omega)=\omega\chi''_{\perp,\parallel}(\omega)\ . \tag{4.15}$$

From the expressions (4.10)–(4.13) we arrive at the following useful spectral representation:

$$\chi_{\perp,\parallel}(\omega+i\epsilon)=\int\frac{d\omega'}{\pi}\left(\frac{\chi''_{\perp,\parallel}(\omega')}{\omega'(\omega+i\epsilon)}\right)\ . \tag{4.16}$$

In particular, we deduce the well-known Kramers-Kronig relations for the reactive and dissipative parts of the magnetic susceptibilities

$$\chi'_{\perp,\parallel}(\omega)=P\int\frac{d\omega'}{\pi}\left(\frac{\chi''_{\perp,\parallel}(\omega')}{\omega'-\omega}\right) \tag{4.17a}$$

and

$$\chi''_{\perp,\parallel}(\omega)=-P\int\frac{d\omega'}{\pi}\left(\frac{\chi'_{\perp,\parallel}(\omega')}{\omega'-\omega}\right)\ . \tag{4.17b}$$

These dispersion relations are associated with the causal character of the response and express general properties of the magnetic susceptibility. In order to deduce the structures of $\chi_\perp$ and $\chi_\parallel$ for the particular Hamiltonian in question it is convenient to express $\chi_\perp$ and $\chi_\parallel$ as

$$\chi_\perp(z)=\frac{1}{2N}[\Psi_0^*S^-(H-E_0-z)^{-1}\times S^+\Psi_0+\Psi_0^*S^+(H-E_0+z)^{-1}S^-\Psi_0] \tag{4.18}$$

and

$$\chi_\parallel(z)=\frac{1}{2N}\Psi_0^*S^z[(H-E_0-z)^{-1}+(H-E_0+z)^{-1}]S^z\Psi_0\ , \tag{4.19}$$

where we have introduced the complex variable z. In arriving at (4.18) and (4.19) we have removed the set of intermediate states Ψ_n in (4.7) and (4.8) and instead introduced the resolvents $(H-E_0\pm z)^{-1}$. Confining our attention to the lowest multiplet including the optic phonon we insert as intermediate states in (4.18) and (4.19) the unperturbed Ising states Ψ_n^0 and the unperturbed phonon state Φ^0. As in Sec. II, we include the effects of the transverse mean exchange j^h by shifting the Ising levels correspondingly. By inspection of the energy spectrum we notice that $\Psi_1^{0*}S^+\Psi_0$ is of order unity whereas $\Psi_n^{0*}S^+\Psi_0$ $(n\neq1)$ and $\Psi_n^{0*}S^-\Psi_0$ are of order j^a/E_2^0 and $\Phi^{0*}S^+\Psi_0$ is of order $A/E_{\rm ph}$. We thus obtain to leading order the following expression for the transverse susceptibility:

$$\chi_\perp(z)=\frac{1}{2N}|\Psi_1^{0*}S^+\Psi_0|^2\Psi_1^{0*}(H-E_0-z)^{-1}\Psi_1^0\ . \tag{4.20}$$

Similar arguments lead to the following expression for the longitudinal susceptibility:

$$\chi_\parallel(z)=\frac{1}{2N}|\Psi_2^{0*}S^z\Psi_0|^2\Psi_2^{0*}[(H-E_0-z)^{-1}+(H-E_0+z)^{-1}]\Psi_2^0\ . \tag{4.21}$$

Notice, however, that $|\Psi_2^{0*}S^z\Psi_0|^2$ is of order $(j^a/E_2)^2$. Expressing the resolvent $(H-E_0\pm z)^{-1}$ as a quotient of the cofactor $\mathrm{cof}(H-E_0\pm z)$ (defined such that $A^{-1}\,\mathrm{cof}A=\det A$) and the determinant $\det(H-E_0\pm z)$ we arrive at the expressions

$$\chi_\perp(z)=\frac{1}{2N}|\Psi_1^{0*}S^+\Psi_0|^2\Psi_1^{0*}\left(\frac{\mathrm{cof}(H-E_0-z)}{\det(H-E_0-z)}\right)\Psi_1^0 \tag{4.22}$$

and

$$\chi_{\parallel}(z)=\frac{1}{2N}|\Psi_2^{0*}S^z\Psi_0|^2\Psi_2^{0*}\left(\frac{\mathrm{cof}(H-E_0-z)}{\det(H-E_0-z)}\right)\times\Psi_2^0+(z\to -z)\ . \quad (4.23)$$

In the basis $\{\Psi_n^0, \Phi^0\}$ the operator $H-E_0\pm z$ assumes the form

$$\begin{Bmatrix} E_{\mathrm{ph}}\pm z & A & 0 & 0 & 0 & \cdots & \cdots \\ A & E_1^0+\Delta_1\pm z & 0 & -2j^a & 0 & \cdots & \cdots \\ 0 & 0 & E_2^0+\Delta_2\pm z & 0 & -2j^a & \cdots & \cdots \\ 0 & -2j^a & 0 & E_3^0+\Delta\pm z & 0 & \cdots & \cdots \\ 0 & 0 & -2j^a & 0 & E_4^0+\Delta\pm z & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{Bmatrix}. \quad (4.24)$$

It is now an easy task to express $\chi_\perp$ and $\chi_\parallel$ as continued fractions. The matrix element $\Psi_1^{0*}\mathrm{cof}(H-E_0\pm z)\Psi_1^0$ is obtained by deleting the second column and the second row in (4.24) and computing the determinant of the remaining matrix. The matrix element $\Psi_2^{0*}\,\mathrm{cof}(H-E_0\mp z)\Psi_2^0$ is found in a similar way by removing the third column and row and evaluating the determinant of the resulting matrix. As seen by induction the quotients $\Psi_1^{0*}\,\mathrm{cof}/\det\Psi_1^0$ and $\Psi_2^{0*}\,\mathrm{cof}/\det\Psi_2^0$ are expressible in terms of infinite continued fractions. We thus arrive at the following expressions for the transverse and longitudinal susceptibilities:

$$\chi_\perp(z)=\frac{1}{2N}|\Psi_1^{0*}S^+\Psi_0|^2\Big/\left(E_1^0+\Delta_1-z-\frac{A^2}{E_{\mathrm{ph}}-z}-\frac{(2j^a)^2}{E_3^0+\Delta-z-(2j^a)^2/(E_5^0+\Delta-z-\ddots)}\right) \quad (4.25)$$

and

$$\chi_\parallel(z)=\frac{1}{2N}|\Psi_2^{0*}S^z\Psi_0|^2\Big/\left(E_2^0+\Delta_2-z-\frac{(2j^a)^2}{E_4^0+\Delta-z-(2j^a)^2/(E_6^0+\Delta-z-\ddots)}\right)+(z\to -z)\ . \quad (4.26)$$

The continued fractions (4.25) and (4.26) clearly exhibit the admixture effects caused by the transverse anisotropy j^a within the Ising-Heisenberg multiplet.

V. ENERGY AND INTENSITY SPECTRA – ANALYTIC SOLUTION

In Sec. IV we derived expressions for $\chi_\perp$ and $\chi_\parallel$ in terms of continued fractions. Since our aim is to discuss the relative intensity spectrum we introduce the two reduced susceptibilities:

$$\tilde{\chi}_\perp(z)=-1\Big/\left(z-E_1^0-\Delta_1-\frac{A^2}{z-E_{\mathrm{ph}}}-\frac{(2j^a)^2}{z-E_3^0-\Delta-(2j^a)^2/(z-E_5^0-\Delta-\ddots)}\right) \quad (5.1)$$

and

$$\tilde{\chi}_\parallel(z)=-1\Big/\left(z-E_2^0-\Delta_2-\frac{(2j^a)^2}{z-E_4^0-\Delta-(2j^a)^2/(z-E_6^0-\Delta-\ddots)}\right)+(z\to -z)\ , \quad (5.2)$$

i.e.,

$$\chi_\perp=\frac{1}{2N}|\Psi_0^*S^-\Psi_1^0|^2\tilde{\chi}_\perp$$

and

$$\chi_\parallel=\frac{1}{2N}|\Psi_0^*S^z\Psi_2^0|^2\tilde{\chi}_\parallel\ . \quad (5.3)$$

Employing the results of III we obtain immediately closed analytic expressions for $\tilde{\chi}_\perp$ and $\tilde{\chi}_\parallel$:

$$\tilde{\chi}_\perp(z)=\frac{\chi_\perp^0(z)}{1+[A^2/(z-E_{\mathrm{ph}})+\Delta_1-\Delta]\chi_\perp^0(z)} \quad (5.4)$$

and

$$\tilde{\chi}_\parallel(z)=\frac{\chi_\parallel^{0+}(z)}{1+(\Delta_2-\Delta)\chi_\parallel^{0+}(z)}+(z\to -z)\ , \quad (5.5)$$

where

$$\chi_\perp^0(z)=\frac{1}{2j^a}\left(\frac{J_{-(z-H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}{J_{-(z+H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}\right) \quad (5.6)$$

and

$$\chi_\parallel^{0+}(z)=\frac{1}{2j^a}\left(\frac{J_{-(z-2H_0-2j^z-\Delta)/2H_0}(2j^a/H_0)}{J_{-(z-2j^z-\Delta)/2H_0}(2j^a/H_0)}\right). \quad (5.7)$$

Apart from the shift Δ, $\chi_{\perp}^{0}$ is the transverse susceptibility and $\chi_{\parallel}^{0+}$ is the positive frequency part of the longitudinal susceptibility in the absence of the transverse mean exchange j^{h} and the optic-phonon coupling A.

$$\tilde{\chi}_{\perp}(z)=\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}\left(\frac{j^a}{H_0}\right)^{2n}\frac{1}{\Gamma(n+1-(z-H_0-2j^z-\Delta)/2H_0)}\Big/\sum_{p=0}^{\infty}\frac{(-1)^p}{p!}\left(\frac{j^a}{H_0}\right)^{2p}$$
$$\times\frac{(2p+1)H_0+2j^z+\Delta_1+A^2/(z-E_{\mathrm{ph}})-z}{\Gamma(p+1-(z-H_0-2j^z-\Delta)/2H_0)} \tag{5.8}$$

and

$$\tilde{\chi}_{\parallel}^{+}(z)=\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}\left(\frac{j^a}{H_0}\right)^{2n}\frac{1}{\Gamma(n+1-(z-2H_0-2j^z-\Delta)/2H_0)}\Big/\sum_{p=0}^{\infty}\frac{(-1)^p}{p!}\left(\frac{j^a}{H_0}\right)^{2p}$$
$$\times\frac{2(p+1)H_0+2j^z+\Delta_2-z}{\Gamma(p+1-(z-2H_0-2j^z-\Delta)/2H_0)}\ , \tag{5.9}$$

where $\tilde{\chi}_{\parallel}^{+}$ is the positive frequency part of the longitudinal susceptibility $\tilde{\chi}_{\parallel}$.

To finite order in j^a/H_0 the intensity spectrum is determined by the quotients of two polynomials. The polynomials are equivalent to the finite-order secular determinant and cofactor discussed in Sec. IV. Beyond second order in j^a/H_0 the expressions (5.8) and (5.9) become unmanageable analytically. Nicoli and Tinkham,[2] however, computed numerically the appropriate eigenvectors pertaining to a 40×40 secular matrix and obtained excellent agreement with the experimental data on the intensity spectrum.

B. Low-field limit (strong coupling)

In order to discuss the low-field limit it is useful to express (5.4) and (5.5) in the form

$$\tilde{\chi}_{\perp}(z)=\sum_{\nu}\frac{I_{\perp,\nu}}{\omega_{\perp,\nu}-z} \tag{5.10}$$

and

$$\tilde{\chi}_{\parallel}^{+}(z)=\sum_{\nu}\frac{I_{\parallel,\nu}}{\omega_{\parallel,\nu}-z}\ , \tag{5.11}$$

where we have introduced the discrete resonance contributions to the susceptibilities. From III we have

$$\chi_{\perp}^{0}(z)=\sum_{\nu}\frac{I_{\perp,\nu}^{0}}{\omega_{\nu}^{0}-z} \tag{5.12}$$

and

$$\chi_{\parallel}^{0+}(z)=\sum_{\nu}\frac{I_{\parallel,\nu}^{0}}{\omega_{\nu}^{0}-z}\ , \tag{5.13}$$

where to leading order

A. High-field limit (weak coupling)

As in our discussion of the high-field limit of the energy spectrum in Sec. III we make use of the power-series expansion[7] (3.11) of the Bessel function.

$$\omega_{\nu}^{0}=2j^z+\Delta-4j^a+2j^a\times[3\pi H_0(\nu+\tfrac{3}{4})/2j^a]^{2/3}+O(H_0/j^a) \tag{5.14}$$

and

$$I_{\perp,\nu}^{0}=I_{\parallel,\nu}^{0}=H_0/j^a+O((H_0/j^a)^{4/3})\ . \tag{5.15}$$

For the *transverse susceptibility* we obtain in the low-field limit and near the lower band edge

$$\omega_{\perp,\nu}=\omega_{\nu}^{0}+\left(\frac{A^2}{2j^z-4j^a+\Delta-E_{\mathrm{ph}}}+\Delta_1-\Delta\right)I_{\perp,\nu}^{0}\ . \tag{5.16}$$

In accordance with our discussion in Sec. III the spectrum near the lower band edge is to leading order in H_0/j^a unaffected by the presence of the optic phonon and the transverse mean exchange. Similarly, the intensities near the lower band are to leading order given by

$$I_{\perp,\nu}=I_{\perp,\nu}^{0}+O((H_0/j^a)^{4/3})\ . \tag{5.17}$$

The *hybridization of the optic phonon with the* μth *multimagnon level* is easily discussed in terms of the transverse susceptibility (5.10). For energies and fields near the hybridization point the susceptibility assumes the form

$$\tilde{\chi}_{\perp}(z)\simeq\frac{I_{\perp,\mu}^{0}}{\omega_{\mu}^{0}-z}\Big/\left(1+\frac{A^2}{z-E_{\mathrm{ph}}}\,\frac{I_{\perp,\mu}^{0}}{\omega_{\mu}^{0}-z}\right)\ . \tag{5.18}$$

In terms of the two independent resonance contributions to $\tilde{\chi}_{\perp}$ we get

$$\tilde{\chi}_{\perp}(z)\simeq\frac{\tilde{I}_{\mu}}{\tilde{\omega}_{\mu}-z}+\frac{\tilde{I}_{\mathrm{ph}}}{\tilde{E}_{\mathrm{ph}}-z}\ . \tag{5.19}$$

The perturbed phonon and magnon energies are

$$\tilde{E}_{\rm ph} = \tfrac{1}{2}\{E_{\rm ph} + \omega_\mu^0 + [(E_{\rm ph} - \omega_\mu^0)^2 + 4A^2 I_{\perp,\mu}^0]^{1/2}\} \tag{5.20}$$

and

$$\tilde{\omega}_\mu = \tfrac{1}{2}\{E_{\rm ph} + \omega_\mu^0 - [(E_{\rm ph} - \omega_\mu^0)^2 + 4A^2 I_{\perp,\mu}^0]^{1/2}\} \ . \tag{5.21}$$

The phonon and magnon intensities are

$$\tilde{I}_{\rm ph} = \frac{E_{\rm ph} - \omega_\mu^0 + [(E_{\rm ph} - \omega_\mu^0)^2 + 4A^2 I_{\perp,\mu}^0]^{1/2}}{[(E_{\rm ph} - \omega_\mu^0)^2 + 4A^2 I_{\perp,\mu}^0]^{1/2}} I_{\perp,\mu}^0 \tag{5.22}$$

and

$$\tilde{I}_\mu = \frac{\omega_\mu^0 - E_{\rm ph} + [(E_{\rm ph} - \omega_\mu^0)^2 + 4A^2 I_{\perp,\mu}^0]^{1/2}}{[(E_{\rm ph} - \omega_\mu^0)^2 + 4A^2 I_{\perp,\mu}^0]^{1/2}} I_{\perp,\mu}^0 . \tag{5.23}$$

In a similar manner we get for the *longitudinal susceptibility* in the low-field limit near the lower band edge

$$\omega_{\parallel,\nu} = \omega_\nu^0 + (\Delta_1 - \Delta) I_{\parallel,\nu}^0 \ . \tag{5.24}$$

The intensities are to leading order given by

$$I_{\parallel,\nu} = I_{\parallel,\nu}^0 + O((H_0/j^a)^{4/3}) \ . \tag{5.25}$$

In Fig. 4 we have sketched the low-field energy spectrum.

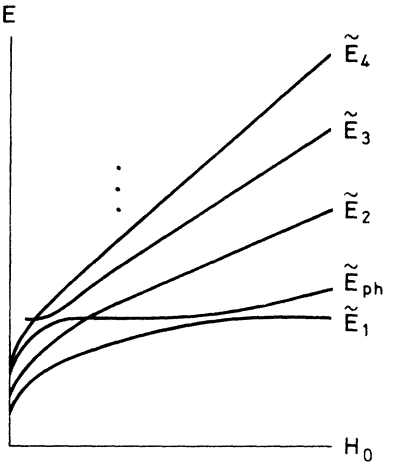

FIG. 4. Low-field energy spectrum (arbitrary units).

C. Zero-field limit

In the zero-field limit the simple branch cut structure of $\chi_\perp^0$ and $\chi_\parallel^{0+}$ (corresponding to the zero-field energy band) allows a more complete discussion of $\chi_\perp$ and $\chi_\parallel^+$ than in the low field case.

At zero field we conclude from III that $\chi_\perp^0$ and $\chi_\parallel^{0+}$ are identical. Denoting the common susceptibility by χ^0 we have

$$\chi^0(z) = \frac{-2}{z - 2j^z - \Delta + [(z - 2j^z - \Delta)^2 - (4j^a)^2]^{1/2}} \ . \tag{5.26}$$

$\chi^0(z)$ has the spectral representation

$$\chi^0(z) = \int \frac{d\omega}{\pi} \left(\frac{\chi^{0\prime\prime}(\omega)}{\omega - z} \right) , \tag{5.27}$$

where the imaginary part (the dissipative part) $\chi^{0\prime\prime}$ is

$$\chi^{0\prime\prime}(\omega) = \begin{cases} \dfrac{1}{2(2j^a)^2}[(4j^a + 2j^z + \Delta - \omega) \\ \qquad \times (\omega - 2j^z - \Delta + 4j^a)]^{1/2} \quad \text{for } -4j^a < \omega - 2j^z - \Delta < 4j^a \\ 0 \quad \text{otherwise.} \end{cases} \tag{5.28}$$

The real part (the reactive part) $\chi^{0\prime}$ is

$$\chi^{0\prime}(\omega) = \begin{cases} -\dfrac{1}{2(2j^a)^2}(\omega - 2j^z - \Delta) \\ \qquad \text{for } -4j^a < \omega - 2j^z - \Delta < 4j^a \\ -\dfrac{1}{2(2j^a)^2}\{\omega - 2j^z - \Delta \\ \qquad - [(\omega - 2j^z - \Delta)^2 - (4j^a)^2]^{1/2}\} \quad \text{otherwise.} \end{cases} \tag{5.29}$$

By means of (5.5) and (5.26) we can express the *longitudinal susceptibility* in the form

$$\tilde{\chi}_\parallel^+(z) = \frac{\tilde{I}_{\parallel,2}}{\omega_{\parallel,2} - z} + \int \frac{d\omega}{\pi} \left(\frac{\tilde{\chi}^{+\prime\prime}(\omega)}{\omega - z} \right) \ . \tag{5.30}$$

The two-magnon bound state is split off from the band and has the energy

$$\omega_{\parallel,2} = 2j^z + \Delta_2 - (2j^a)^2/(\Delta - \Delta_2) \ , \tag{5.31}$$

and the intensity

$$\tilde{I}_{\parallel,2} = 1 - \left(\frac{(2j^a)^2}{\Delta - \Delta_2} \right) \int \frac{d\omega}{\pi} \left(\frac{\chi^{0\prime\prime}(\omega)}{\omega - 2j^z - \Delta_2 + (2j^a)^2/(\Delta - \Delta_2)} \right) . \tag{5.32}$$

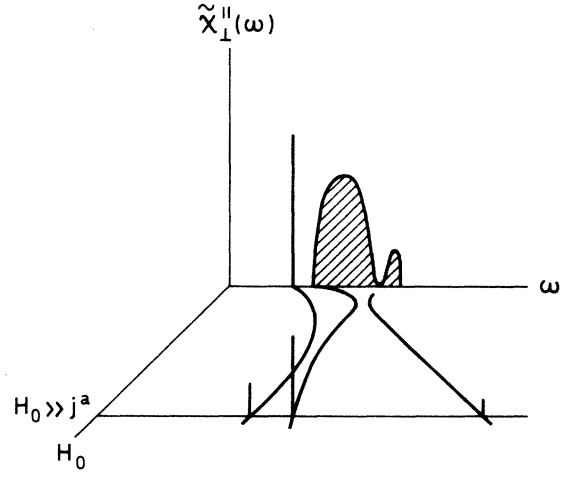

FIG. 5. Transverse susceptibility as function of energy and field (arbitrary units).

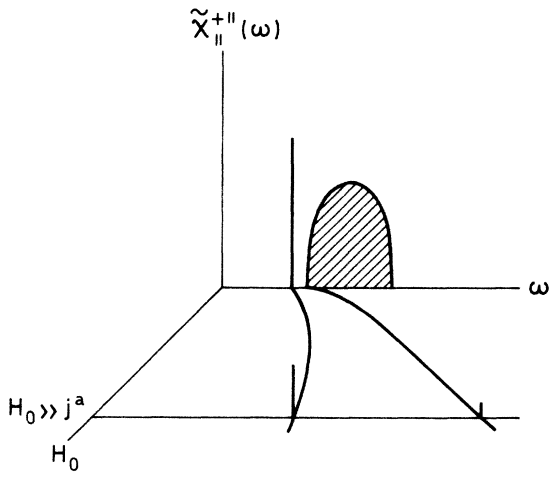

FIG. 6. Longitudinal susceptibility as function of energy and field (arbitrary units).

The modified intensity spectrum corresponding to the energy band has the form

$$\tilde{\chi}^{+\prime\prime}(\omega) = \frac{(2j^a)^2}{\Delta - \Delta_2} \left(\frac{\chi^{0\prime\prime}(\omega)}{\omega - 2j^z - \Delta_2 + (2j^a)^2/(\Delta - \Delta_2)} \right) . \tag{5.33}$$

The *transverse susceptibility* has a more complicated structure due to the hybridization with the optic phonon. Neglecting for simplicity the split-off single-magnon state we express $\tilde{\chi}_{\perp}(z)$ in the form

$$\tilde{\chi}_{\perp}(z) = \int \frac{d\omega}{\pi} \left(\frac{\tilde{\chi}_{\perp}^{\prime\prime}(\omega)}{\omega - z} \right) , \tag{5.34}$$

where the intensity spectrum is given by

$$\tilde{\chi}_{\perp}^{\prime\prime}(\omega) = \frac{(\omega - E_{\rm ph})^2 \chi^{0\prime\prime}(\omega)}{[1 - (A/2j^a)^2][(\omega - \omega_0)^2 + (\tfrac{1}{2}\Gamma)^2]} . \tag{5.35}$$

Since the phonon falls within the band it modifies the intensity spectrum drastically. The intensity spectrum vanishes quadratically in the vicinity of the unperturbed phonon energy. The continuum states are pushed away by the phonon and result in a modification of the intensity spectrum as shown by the Lorentzian factor in (5.35). The Lorentzian is situated at

$$\omega_0 = \frac{E_{\rm ph}[1 - \frac{1}{2}(A/2j^a)^2] - \frac{1}{2}(A/2j^a)^2(2j^z + \Delta)}{1 - (A/2j^a)^2} \tag{5.36}$$

and has the linewidth

$$(\tfrac{1}{2}\Gamma)^2 = \frac{(A/2j^a)^2}{[1 - (A/2j^a)^2]^2} \{ [1 - (A/2j^a)^2] A^2 - \tfrac{1}{4}(A/2j^a)^2 (E - 2j^z - \Delta)^2 \} . \tag{5.37}$$

Notice that the center of the Lorentzian coincides with the phonon energy when the phonon falls in the middle of the band.

The single magnon state is split off from the band and gives rise to a separate resonance contribution. Neglecting the presence of the phonon the position and intensity of the single magnon state are given by (5.31) and (5.32) with Δ_2 replaced by Δ_1.

In Figs. 5 and 6 we have sketched the relative intensity spectra pertaining to the transverse and longitudinal susceptibilities, respectively, as functions of energy and field.

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*Present address: Institut Max von Laue–Paul Langevin, 38042 Grenoble Cedex, France.

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