Intensity Spectrum of the Anisotropic Magnetic Chain

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The transverse and longitudinal intensity spectrum pertaining to the lowest (one-cluster) multiplet of the linear anisotropic spin-ferromagnetic chain is obtained analytically in the absence of transverse mean exchange ($j^x + j^y = 0$) and neglecting coupling to higher multiplets. The magnetic susceptibilities, which have the form of continued fractions, are expressed in terms of Bessel functions. At high fields the susceptibilities have the form of quotients of power-series expansion in j^a/H_0, where j^a is the transverse anisotropy parameter ($j^a = (j^x - j^y)/2$) and H_0 is the magnetic field. At low field the total intensity is evenly shared among the energy levels. At zero field the intensity spectra are bounded and continuous and assume the shape of a semiellipse about the degeneracy point. A magnetic intensity spectrum of that character has recently been observed by Nicoli and Tinkham in the magnetic salt $CoCl_2 \cdot 2H_2O$.

In a previous publication,[1] referred to as II, an analytical expression was obtained for the energy spectrum of the lowest (one-cluster) multiplet of the linear anisotropic spin-$\frac{1}{2}$ ferromagnetic chain in the absence of transverse mean exchange ($j^x + j^y = 0$) and neglecting coupling to other states.

In the present article, an analytical expression is derived for the relative intensity spectrum pertaining to the lowest (one-cluster) multiplet in the absence of transverse mean exchange ($j^x + j^y = 0$) and disregarding coupling to higher multiplets. An intensity spectrum of this character has recently been observed in the magnetic salt $CoCl_2 \cdot 2H_2O$ by Nicoli and Tinkham,[2] and it is therefore of considerable interest to obtain an analytical expression for the relative intensity spectrum.

As discussed in a previous publication,[3] which shall be referred to as I, the intensity spectrum falls into two parts. The part corresponding to transverse polarization is given by the absorptive component of the transverse dynamical susceptibility,

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

Similarly, the part of the intensity spectrum corresponding to longitudinal polarization is described by the absorptive component of the longitudinal dynamical susceptibility,

$$\chi''_\parallel(k, \omega) = \sum_{i=1}^{N} \int dt \exp\{i\omega(t-t') - ik(x_i - x_k)\} \times \langle [S_i^z(t), S_k^z(t')] \rangle . \tag{2}$$

In the exact expressions (1) and (2) the square brackets denote an expectation value in the true ground state of the system. The spin operators referring to the site indices i and k develop in time according to the spin-$\frac{1}{2}$ nearest-neighbor exchange Hamiltonian

$$\mathcal{H}' = -2 \sum_{i=1}^{N} [j^z S_i^z S_{i+1}^z + \tfrac{1}{2} j^a (S_i^+ S_{i+1}^+ + \text{H.c.})] + \gamma H_0 \sum_{i=1}^{N} S_i^z . \tag{3}$$

j^z and j^a are the longitudinal and transverse exchange constants, respectively. γH_0 is the effective g factor in units of the Bohr magneton μ_B times an external field applied along the z direction.

As in I, it is convenient to introduce the complex susceptibilities $\chi_\perp(k, z)$ and $\chi_\parallel(k, z)$ defined by the spectral representations

$$\chi_{\perp,\parallel}(k, z) = \int \frac{d\omega}{\pi} \frac{\chi''_{\perp,\parallel}(k, \omega)}{\omega - z} . \tag{4}$$

As is well known, the poles and corresponding residues of $\chi_\perp(k, z)$ and $\chi_\parallel(k, z)$ yield the energy spectrum and relative intensity spectrum corresponding to transverse and longitudinal polarizations, respectively.

Performing the time integrations in (1) and (2) by means of the Heisenberg equations of motion for the spin operators, we can express $\chi_\perp(k, z)$ in the resolvent forms

$$\chi_\perp(k, z) = \frac{1}{2} \sum_{i=1}^{N} e^{-ik(x_i - x_k)} [\langle S_i^-(\mathcal{H}' - E_0 - z)^{-1} S_k^+ \rangle - \langle S_k^+(E_0 - \mathcal{H}' - z)^{-1} S_i^- \rangle] , \tag{5}$$

and

$$\chi_\parallel(k, z) = \sum_{i=1}^{N} e^{-ik(x_i - x_k)} [\langle S_i^z(\mathcal{H}' - E_0 - z)^{-1} S_k^z \rangle - \langle S_k^z(E_0 - \mathcal{H}' - z)^{-1} S_i^z \rangle] . \tag{6}$$

In order to investigate the intensity spectrum of the lowest (one-cluster) multiplet, we insert into expressions (5) and (6) a set of intermediate Bloch states referring to the lowest multiplet of the

Ising model (i.e., $j^a = 0$),

$$\psi_n^0(k) = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{ik[x_i + (n-1)a/2]} S_i^+ S_{i+1}^+ \cdots S_{i+n-1}^+ |0\rangle . \quad (7)$$

The Bloch states $\psi_n^0(k)$, corresponding to a single cluster of n adjacent spin deviations with respect to the aligned ferromagnetic ground state $|0\rangle$, have the energies $E_n^0 = 2j^z + n\gamma H_0$ ($n = 0, 1, 2, \ldots, N$) and form an orthonormal set, assuming $\langle 0|0\rangle = 1$. a is the interatomic instance and N is the total number of spins; the angular wave number k runs over the first Brillouin zone $-\pi/a < k \leq \pi/a$. Confining our attention to the admixture effects within the lowest multiplet, i.e., neglecting coupling to higher multiplets, we get

$$\chi_\perp(k, z) = \psi_1^0(k)^* (\mathcal{H}' - z)^{-1} \psi_1^0(k) \quad (8)$$

and

$$\chi_\parallel(k, z) = \psi_2^0(k)^* [(\mathcal{H}' - z)^{-1} + (\mathcal{H}' + z)^{-1}] \psi_2^0(k) , \quad (9)$$

where we have absorbed the ground-state energy E_0 in $\mathcal{H}'$ and included z-independent constants in the definitions of $\chi_\perp$ and $\chi_\parallel$. Expressing the resolvent $(\mathcal{H}' - z)^{-1}$ as the quotient of the cofactor cof$(\mathcal{H}' - z)$ and the determinant det$(\mathcal{H}' - z)$ in the Bloch basis (7), we can show that $\chi_\perp$ and $\chi_\parallel$ can be expressed in terms of continued fractions in the following manner:

$$\chi_\perp(k, z) = \cfrac{-1}{z - E_1^0 - \cfrac{(2j^a \cos ka)^2}{z - E_3^0 - \cfrac{(2j^a \cos ka)^2}{z - E_5^0 - \cdots}}} \quad (10)$$

and

$$\chi_\parallel(k, z) = \cfrac{-1}{z - E_2^0 - \cfrac{(2j^a \cos ka)^2}{z - E_4^0 - \cfrac{(2j^a \cos ka)^2}{z - E_6^0 - \cdots}}} + (z \to -z) . \quad (11)$$

By inspection we notice that the poles in (10) and (11) coincide with the roots in the secular determinant arising from the eigenvalue equation $\mathcal{H}'\psi = E\psi$ for the lowest multiplet,

$$\begin{vmatrix} E - E_1^0 & 0 & -2j^a \cos ka & 0 \\ 0 & E - E_2^0 & 0 & -2j^a \cos ka \\ -2j^a \cos ka & 0 & E - E_3^0 & 0 \\ 0 & -2j^a \cos ka & 0 & E - E_4^0 \end{vmatrix} = 0 . \quad (12)$$

The diagonal elements originate from the unperturbed Ising model (i.e., $j^a = 0$); the off-diagonal elements arise from evaluating the j^a term in $\mathcal{H}'$ in the Bloch basis (7). We also notice that the dynamical decoupling of the states that correspond to an odd number of spin deviations (the "odd" states) from the states that correspond to an even number of spin deviations (the "even" states) is reflected in the two components of the susceptibility. $\chi_\perp$ yields the intensity spectrum of the odd states, and $\chi_\parallel$ gives the intensity spectrum of the even states.

It is perhaps interesting to observe that since the unperturbed levels are equidistant the eigenvalue problem corresponds to a harmonic oscillator with a constant coupling between adjacent levels. In the well-known Bose representation of a quantum oscillator, the Hamiltonian for the odd levels assumes the form

$$\mathcal{H}_{\text{osc}}^{\text{odd}} = 2j^z + \gamma H_0 + 2\gamma H_0 b^\dagger b - (2j^a \cos ka)[(1 + b^\dagger b)^{-1/2} b + \text{H.c.}] . \quad (13)$$

The structure of (13) clearly exhibits the strong nonlinearity of the problem.

Returning to the continued fraction (10), it is easy to show that $\chi_\perp$ satisfies the nonlinear difference equation

$$(2j^a \cos ka)^2 \chi_\perp(k, z - 2\gamma H_0) \chi_\perp(k, z) + (z - E_1^0) \chi_\perp(k, z) + 1 = 0. \quad (14)$$

By means of the substitution

$$\chi_\perp(k, z) = \frac{1}{2j^a \cos ka} \frac{\Omega(k, z)}{\Omega(k, z + 2\gamma H_0)} , \quad (15)$$

a linear difference equation is obtained for $\Omega(k, z)$:

$$\Omega(k, z + 2\gamma H_0) + \Omega(k, z - 2\gamma H_0) = \frac{z - E_1^0}{2j^a \cos ka} \Omega(k, z) . \quad (16)$$

The substitution (15) essentially amounts to expressing $\chi_\perp(k, z)$ as a quotient of two secular determinants, as was done in I. The linear difference equation (16) is then obtained simply by expanding the determinants in minors.

As in our analysis of the energy spectrum in II, a solution is immediately obtained by observing that (16) is a recursion formula for the solutions to the Bessel equation.[4] The Bessel function has the appropriate high-field behavior; we obtain

$$\Omega(k, z) = J_{(E_1^0 - z)/2\gamma H_0}\left(\frac{2j^a \cos ka}{\gamma H_0}\right) . \tag{17}$$

Hence

$$\chi_\perp(k, z) = \frac{1}{2j^a \cos ka} \times \frac{J_{(E_1^0 - z)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)}{J_{(E_1^0 - 2\gamma H_0 - z)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)} . \tag{18}$$

By a similar analysis we obtain

$$\chi_\parallel(k, z) = \frac{1}{2j^a \cos ka}\, \frac{J_{(E_2^0 - z)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)}{J_{(E_2^0 - 2\gamma H_0 - z)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)} , + \frac{1}{2j^a \cos ka}\, \frac{J_{(E_2^0 + z)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)}{J_{(E_2^0 + 2\gamma H_0 + z)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)} . \tag{19}$$

The expressions (18) and (19) provide explicit forms for $\chi_\perp$ and $\chi_\parallel$ in terms of tabulated function.[5] As in II the energy spectrum is obtained by determining the roots of the Bessel functions in the denominators of $\chi_\perp$ and $\chi_\parallel$, i.e., the poles of $\chi_\perp$ and $\chi_\parallel$. The intensity spectrum is given by the residues of the corresponding poles in (18) and (19). Since $\chi_\perp$ and $\chi_\parallel$ have a similar structure, we confine our detailed discussion to $\chi_\perp$.

At zero field, the unperturbed Ising levels are degenerate and the continued fraction (10) for $\chi_\perp$ can be expressed in a closed form. Solving (14), which for $H_0 = 0$ becomes a simple algebraic equation, we obtain (as in I)

$$\chi_\perp(k, z) = \frac{-2}{(z - 2j^z) + [(z - 2j^z)^2 - (4j^a \cos ka)^2]^{1/2}} . \tag{20}$$

$\chi_\perp(k, z)$ has a branch cut along the real axis extending from $2j^z - 4j^a \cos ka$ to $2j^z + 4j^a \cos ka$. This branch cut corresponds to the bounded continuous energy spectrum obtained in II in the zero-field limit. The relative intensity spectrum is given by $\chi_\perp''(k, \omega) = \mathrm{Im}\chi_\perp(k, \omega + i\epsilon)$. From (20) we obtain

$$\chi_\perp''(k, \omega) = \frac{[(4j^a \cos ka + 2j^z - \omega)(\omega - 2j^z + 4j^a \cos ka)]^{1/2}}{(4j^a \cos ka)^2} . \tag{21}$$

The relative intensity spectrum in the zero-field limit has the shape of a semiellipse centered about the degeneracy point $2j^z$.

In order to discuss the intensity spectrum at finite field, we emphasize that the dimensionless parameter characterizing the problem is the coupling strength divided by the energy difference between adjacent unperturbed levels. It is therefore useful to introduce the dimensionless parameter $\eta = (2j^a \cos ka)/\gamma H_0$.

For large values of the field, i.e., $\eta \ll 1$, an asymptotic expansion in $1/\gamma H_0$ is obtained by doing perturbation theory in j^a, as discussed in I and II. The procedure amounts to terminating the continued fraction (10) after admixture of a finite number of higher levels. In terms of the secular determinant (12), the above procedure is equivalent to diagonalizing a finite determinant, as was done by Torrance and Tinkham[6] numerically in their analysis of the energy spectrum and by Nicoli and Tinkham[2] in their numerical investigation of the intensity spectrum. An expansion of $\chi_\perp$ in powers of η is readily obtained by means of the well-known series expansion of the Bessel function,[4]

$$J_\lambda(z) = \left(\frac{z}{2}\right)^\lambda \sum_{n=0}^{\infty} \frac{(-1)^n}{n!}\left(\frac{z}{2}\right)^{2n} \frac{1}{\Gamma(n + \lambda + 1)} . \tag{22}$$

We get

$$\chi_\perp(k, z) = \frac{1}{2\gamma H_0}\left[\sum_{n=0}^{\infty} \frac{(-1)^n}{n!}\left(\frac{\eta}{2}\right)^{2n} \frac{1}{\Gamma(n + (E_1^0 - z)/2\gamma H_0 + 1)}\right] \Big/ \left[\sum_{p=0}^{\infty} \frac{(-1)^p}{p!}\left(\frac{\eta}{2}\right)^{2p} \frac{1}{\Gamma(p + (E_1^0 - z)/2\gamma H_0)}\right] , \tag{23}$$

which expresses $\chi_\perp(k, z)$ as the quotient of two power-series expansions on the dimensionless parameter η. For completeness we note that the expansions in (23) are convergent for all values of η and uniformly convergent for all values of z.

Employing the fundamental recursion formula $\Gamma(z+1) = z\Gamma(z)$ for the Γ function,[4] we get for $\eta = 0$, $\chi_\perp(k, z) = 1/(E_1^0 - z)$, thus corroborating the continued fraction (10) for $\eta = 0$.

As in our discussion of the energy spectrum in

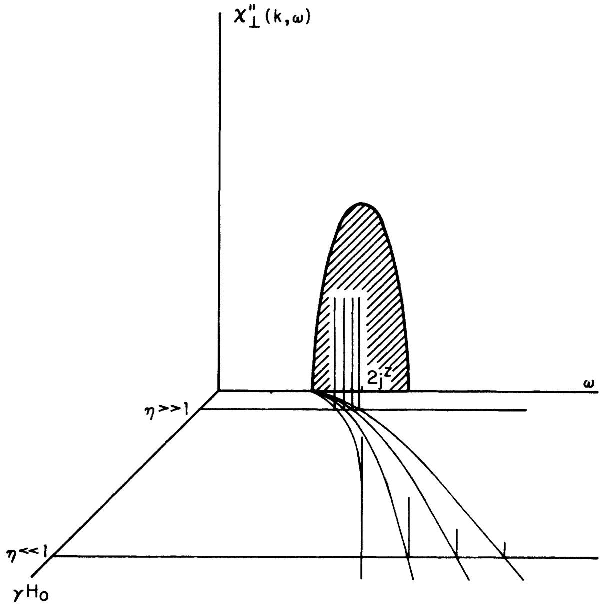

FIG. 1. Relative-intensity spectrum as a function of energy and field (arbitrary units).

II, the interesting regime is the low-field limit, where the discrete infinite spectrum collapses to a bounded continuous spectrum. Since the spectrum for nonvanishing field is discrete, $\chi_\perp(k, z)$ can be written in the form

$$\chi_\perp(k, z) = \sum_{\nu=0}^{\infty} \frac{I_\nu}{\omega_\nu - z} , \tag{24}$$

i.e.,

$$\chi_\perp''(k, \omega) = \pi \sum_{\nu=0}^{\infty} I_\nu \delta(\omega - \omega_\nu) , \tag{25}$$

where I_ν is the relative intensity of the energy level ω_ν. An expression for I_ν is obtained by expanding $\chi_\perp(k, z)$ near the pole ω_ν. From (18) we get

$$I_\nu = -\frac{1}{2j^a \cos ka} \times \frac{J_{(E_1^0-\omega_\nu)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)}{[(d/d\omega) J_{(E_1^0-2\gamma H_0-\omega)/2\gamma H_0}(2j^a \cos ka/\gamma H_0)]_{\omega=\omega_\nu}} . \tag{26}$$

In order to discuss the low-field limit we make use of the following double asymptotic expansion of the Bessel function[4]:

$$J_{\mu\cos\alpha}(\mu) = (2/\pi\mu \sin\alpha) \times \{\cos[\mu(\sin\alpha - \alpha\cos\alpha) - \tfrac{1}{4}\pi] + O(\mu^{-1/5})\} , \tag{27}$$

where $0 < \alpha < \frac{1}{2}\pi$ and $\mu > 0$.

As in II, the energy spectrum is determined by the condition

$$J_{(E_1^0-2\gamma H_0-\omega_\nu)/2\gamma H_0}(2j^a \cos ka/\gamma H_0) = 0 . \tag{28}$$

By means of (27), we get

$$\eta(\sin\theta_\nu - \theta_\nu \cos\theta_\nu) = \pi(\nu + \tfrac{3}{4}), \quad \nu = 0, 1, 2, \ldots , \tag{29}$$

where we have introduced the notation

$$\omega_\nu = 2j^z - 4j^a \cos ka\, (\cos\theta_\nu + \tfrac{1}{2}\eta^{-1}) . \tag{30}$$

By expanding (29) and (30) at low field we obtain to leading order

$$\omega_\nu = 2j^z - 4j^a \cos ka \{1 - \tfrac{1}{2}[3\pi(\nu + \tfrac{3}{4})]^{2/3}\eta^{-2/3} + O(\eta^{-1})\} . \tag{31}$$

As described in II, the energy levels approach the lower edge of the zero-field band with infinite slope as a function of field.

In order to examine the intensity spectrum given by (20) at low field, we introduce the notation

$$\omega = 2j^z - 4j^a \cos ka(\cos\theta + \tfrac{1}{2}\eta^{-1}) \tag{32}$$

and

$$\omega_\nu = 2j^z - 4j^a \cos ka(\cos\theta_\nu' - \tfrac{1}{2}\eta^{-1}) . \tag{33}$$

By means of (27) we get

$$I_\nu = \left(\frac{\sin\theta_\nu}{\sin\theta_\nu'}\right)^{1/2} (-1)^\nu \frac{2}{\eta} \frac{1}{\theta_\nu} \times \cos[\eta(\sin\theta_\nu' - \theta_\nu' \cos\theta_\nu') - \tfrac{1}{4}\pi] , \tag{34}$$

where

$$\cos\theta_\nu' = \eta^{-1} + \cos\theta_\nu . \tag{35}$$

Examining (34) at low field, (i.e., about the lower edge, since ν is fixed) we obtain to leading order

$$I_\nu = 2\eta^{-1} + O(\eta^{-4/3}) . \tag{36}$$

It is interesting to note that the leading term in the low-field expansion is *independent* of the quantum number ν. The total intensity is shared evenly among the energy levels in the low-field limit. However, since the levels converge towards the lower-band edge in the limit of zero field, the continuous intensity spectrum at zero field assumes the shape of a semiellipse.

In Fig. 1 we have sketched the relative intensity spectrum as a function of energy and field.

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[1]H. C. Fogedby and H. H. Jensen, Phys. Rev. B **6**, 3444 (1972).
[2]D. Nicoli and M. Tinkham (unpublished).
[3]H. C. Fogedby, Phys. Rev. B **5**, 1941 (1972).
[4]R. Courant and D. Hilbert, *Methods of Mathematical*

Physics (Interscience, New York, 1966).
[5]*Tables of Function with Formulae and Curves*, edited by E. Jahnke and F. Emde (Dover, New York, 1945).
[6]J. B. Torrance, Jr. and M. Tinkham, Phys. Rev. **187**, 587 (1969).