

# Finite-temperature dynamical structure factor of alternating Heisenberg chains

A. J. A. James,<sup>1</sup> F. H. L. Essler,<sup>1</sup> and R. M. Konik<sup>2</sup>

<sup>1</sup>*Rudolf Peierls Centre for Theoretical Physics, University of Oxford, Oxford OX1 3NP, United Kingdom*

<sup>2</sup>*Brookhaven National Laboratory, Upton, New York 11973, USA*

(Received 23 June 2008; revised manuscript received 7 August 2008; published 16 September 2008)

We develop a low-temperature expansion for the finite-temperature dynamical structure factor of the spin half Heisenberg chain with alternating nearest-neighbor exchange in the limit of strong alternation of the exchange constants. We determine both the broadening of the low-lying triplet lines and the contribution of the thermally activated intraband scattering.

DOI: [10.1103/PhysRevB.78.094411](https://doi.org/10.1103/PhysRevB.78.094411)

PACS number(s): 75.10.Jm, 75.10.Pq, 75.40.Gb

## I. INTRODUCTION

Much is now known about the physics of quasi-one-dimensional Heisenberg antiferromagnetic chain materials at zero temperature. This understanding has benefited from powerful analytical techniques (see, e.g., Refs. 1–4 and references therein) as well as highly accurate experiments performed at temperatures much smaller than the relevant exchange constants.<sup>5</sup> As such, the regime in which these materials are understood is dominated by quantum fluctuations. In contrast, far less is known about their finite-temperature behavior, where there is an interplay of quantum and thermal fluctuations.<sup>6–11</sup>

Although the chemistry of these materials can be exceedingly complicated, a wide range of their properties, up to energy scales set by their exchange constants, are well described by simple lattice models of the form

$$H = \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

Here  $J_{ij}$  is the exchange constant between spins at sites  $i$  and  $j$ .

Perhaps the most encompassing probe of the properties of spin chains is inelastic neutron scattering. Such experiments provide detailed information on the chains' excitations.<sup>12</sup> In theoretical terms, these experiments specifically yield the spin dynamical structure factor,

$$S^{\alpha\gamma}(\omega, Q) = -\frac{1}{\pi} \frac{1}{1 - e^{-\beta\omega}} \text{Im}[\chi^{\alpha\gamma}(\omega, Q)],$$

$$\chi^{\alpha\gamma}(\omega, Q) = -\int_0^\beta d\tau e^{i\omega_n\tau} \frac{1}{N} \sum_{l,l'} e^{-iQ(l-l')} \langle S_l^\alpha(\tau) S_{l'}^\gamma \rangle_{\omega_n \rightarrow \omega_n - i\omega}. \quad (2)$$

Here  $\beta = 1/k_B T$  and  $\alpha, \gamma = x, y, z$  and the brackets imply a thermal expectation. We have used the Matsubara formalism with imaginary times  $\tau$  and frequencies  $\omega_n$ . The dynamical susceptibilities  $\chi^{\alpha\gamma}(\omega, Q)$  take their simplest form at zero temperature, where they provide direct information on the energies and lifetimes of the spin excitations. At finite temperature the  $\chi^{\alpha\gamma}(\omega, Q)$  become more complex functions, determined now by a competition between thermal and quantum effects. The value of the exchange coupling  $J$  determines the extent of the temperatures' role. If  $J \gg T$ , there exists an

appreciable range of energies  $\omega$  over which inelastic neutron-scattering experiments effectively probe the zero-temperature form of  $\chi^{\alpha\gamma}(\omega, Q)$ . However, if  $J \sim T$  or one is interested in energies  $\omega$ , on the order of  $T$ , the effects of temperature must be taken into account when calculating  $\chi^{\alpha\gamma}(\omega, Q)$ .

In this paper, we are interested in calculating the dynamical structure factor at low but finite temperature for a class of dimerized spin-1/2 chain materials. These materials are well described by the Hamiltonian

$$H_{\text{dimer}} = \sum_{a=0}^{N/2-1} (J \mathbf{S}_{2a} \cdot \mathbf{S}_{2a+1} + J' \mathbf{S}_{2a+1} \cdot \mathbf{S}_{2a+2}). \quad (3)$$

Here  $J$  is the exchange coupling, through which pairs of neighboring  $S=1/2$  spins form singlet dimers while  $J' = \alpha J$  gives the strength of the interdimer interaction. We will be interested in the case  $\alpha \ll 1$ . The lowest-lying excitations of this model, which we will refer to as magnons, are characterized by a gap  $\Delta$ , which (at zeroth order in  $\alpha$ ) represents the cost of breaking one dimer. With small but finite  $\alpha$ , the magnons disperse according to

$$\epsilon_k = J - \frac{\alpha J}{2} \cos(kd),$$

where  $d$  is the interdimer distance. A particular realization of this material is  $\text{Cu}(\text{NO}_3)_2 \cdot 2.5\text{H}_2\text{O}$ , where  $J = 5.22$  K and  $\alpha = 0.27$ .<sup>13,14</sup> While this material has been studied by inelastic neutron scattering at 300 mK,<sup>13</sup> a temperature far smaller than  $\Delta = 4.4$  K, more recent experiments have been performed<sup>15</sup> at temperatures on the same order as the gap where thermal effects on the form of  $\chi^{\alpha\gamma}(\omega, q)$  cannot be ignored. Previously, much theoretical work has concentrated on the zero-temperature limit, establishing various properties of the spectrum of Eq. (3) including multiparticle continua and bound states<sup>16–23</sup> as well as spectral weights<sup>24,25</sup> and the dynamical structure factor.<sup>22,25,26</sup> However, at finite temperature, the most pertinent features to explore are the broadening of the single-particle modes, due to interaction with the thermally populated background, and the low-frequency response arising from intraband transitions.

By virtue of the spin-rotational invariance of the Hamiltonian (3), all off-diagonal elements of the dynamical susceptibility vanish and the three diagonal elements are the

same, i.e.,  $\chi^{xx}(\omega, Q) = \chi^{yy}(\omega, Q) = \chi^{zz}(\omega, Q)$ . In what follows we will therefore only consider  $\chi^{zz}$ .

Accounting for the effects of temperature in the dynamical susceptibilities makes this problem particularly challenging. To see why, we consider a Lehmann expansion of the spin response function in terms of the eigenstates of the model  $\{|l\rangle\}$ . Defining  $C(\tau, x) = \langle S_j^z(\tau) S_k^z \rangle$ , where  $x = R_j - R_k$ , this expansion takes the form

$$C(\tau, x) = \frac{1}{Z} \sum_{l,m} e^{-\beta E_l} \langle l | S_j^z(\tau) | m \rangle \langle m | S_k^z | l \rangle. \quad (4)$$

The double sum in this representation for  $C(\tau, x)$  arises on one hand from the Boltzmann sum  $\sum_l e^{-\beta E_l}$ , where  $E_l$  is the energy of eigenstate  $|l\rangle$ , and on the other hand from an insertion of a resolution of the identity between the two operators  $S_j^z(\tau)$  and  $S_k^z$ . This expansion renders the task of finding  $C(\tau, x)$  into a matter of computing individual matrix elements  $\langle l | S_j^z(\tau) | m \rangle$ . At zero temperature, this computation is simplified on two counts:<sup>3</sup> (i) the sums over eigenstates in Eq. (4) reduce to a single sum and (ii) the matrix elements needed are of a single type, namely, those connecting the ground with various excited states. At finite temperature, however, we must deal both with the double sum in Eq. (4) and the matrix elements in their full generality.

To make this task tractable, we exploit the fact that the spin chain material is gapped. On a qualitative level the excitations can be divided according to the number  $n$  of magnons they contain. The energy of an excitation with  $n$  magnons is then at least  $n\Delta$ . This notion is imprecise because the number of magnons is not a good quantum number; nonetheless, at small  $\alpha$ , it can serve as a rough guide to the excitations' energies. In turn, provided the temperature does not exceed the gap  $\Delta$ , the contribution to the sum in Eq. (4) of excitations containing large numbers of magnons will be exponentially suppressed by the Boltzmann factor  $e^{-\beta E_l}$ . In such a case, we thus need only to consider excitations in the Boltzmann sum  $\sum_l e^{-\beta E_l}$ , involving only a few magnons. Concomitantly, provided we are interested in determining  $\text{Im } \chi^{zz}(\omega, Q)$  at energies not far in excess of the gap, we can similarly restrict the sum in Eq. (4) ( $\Sigma_m$ ) arising from the resolution of the identity.

The evaluation of  $\chi^{zz}(\omega, Q)$  is, however, more delicate than what the above implies. When evaluating the leading terms in the Fourier transform of the spectral representation (4), one finds divergences when the frequency approaches the magnon dispersion. Such divergences are expected, since the spectral sum still contains the  $T=0$  result, which is a delta function at the position of the single-magnon dispersion. On physical grounds the single-magnon line is expected to broaden at  $T>0$ . Analytically this is achieved by carrying out a resummation on the divergences of the higher-order terms in the expansion. Specifically, the sum in Eq. (4) can be reorganized according to a Dyson's equation,<sup>11</sup> where we write  $\chi^{zz}(\omega, Q)$  in the form

$$\chi^{zz}(\omega, Q) = \frac{D(\omega, Q)}{1 - D(\omega, Q)\Sigma(\omega, Q)}. \quad (5)$$

Here  $D(\omega, q)$  can be thought of as the propagator for noninteracting magnons and  $\Sigma(\omega, q)$  is the magnon self-energy.

The key is to match the perturbative expansion of Eq. (5),

$$\chi^{zz}(\omega, Q) = D(\omega, Q) + D^2(\omega, Q)\Sigma(\omega, Q) + \dots, \quad (6)$$

to the spectral representation of  $\chi^{zz}(\omega, Q)$ , which is given in terms of the Fourier transform of Eq. (4). In this way we obtain a controlled low-temperature expansion of the self-energy  $\Sigma(\omega, q)$  in lieu of  $\chi^{zz}(\omega, Q)$ .

This approach has been used successfully in the study of finite-temperature dynamical correlation functions in gapped one-dimensional quantum antiferromagnets with continuum integrable field-theoretic representations.<sup>11</sup> There the matrix elements  $\langle l | S_j^z(0) | m \rangle$  were computed exactly via analyticity constraints coming from integrability.<sup>3,27</sup> However, the model of the dimerized spin chain [Eq. (3)] is not exactly solvable. But because  $\alpha$  is small, we can compute the necessary matrix elements perturbatively in  $\alpha$ .

The  $T>0$  dynamical susceptibility has been studied previously using exact diagonalization of finite-length chains.<sup>10</sup> We believe our approach provides a useful complement to this work. The numerical approach yields results for all  $\alpha$  and is not restricted to small temperatures. However, the system size that can be studied is quite small. We, on the other hand, must proceed perturbatively in  $\alpha$  and are restricted to low temperatures, but our calculations do not suffer from finite-size effects. Moreover, the nature of low-lying excitations is more apparent and we can identify the specific processes that give rise to the various finite-temperature effects in the structure factor.

With this in mind, a specific feature that we focus upon in our analysis is the presence of temperature-induced neutron-scattering intensity at low frequencies much smaller than the zero-temperature gap. The origin of this intensity is intraband scattering. The analogous phenomenon in Ising-type antiferromagnetic spin chains was first pointed out by Villain<sup>28</sup> and was first observed in the anisotropic spin chain material CsCoBr<sub>3</sub>.<sup>29,30</sup> For Ising antiferromagnets, the relevant excitations are domain walls in the antiferromagnetic order. In contrast, in the dimer model, the relevant excitations correspond to low-lying magnons. In both cases these excitations experience intraband transitions.

An outline of this paper is as follows. In Sec. II, using first-order degenerate perturbation theory, we determine the dimer model's low-lying spectrum and the corresponding matrix elements. In Sec. III we discuss in detail how to use these ingredients to compute the susceptibility  $\chi^{zz}(\omega, Q)$ . In particular, we explain the use of the resummation implied by the Dyson equation. In Sec. IV, we present the actual results for  $\chi^{zz}(\omega, Q)$ .

## II. GROUND STATE AND EXCITED STATES OF WEAKLY COUPLED DIMERS

Our starting point is the Hamiltonian given in Eq. (3) with an even number of sites  $N$  and periodic boundary conditions. For small  $\alpha = J'/J$ , we split the Hamiltonian into a solvable part proportional to  $J$ ,  $H_0$ , and a perturbation  $H'$  (proportional to  $J'$ ),

$$H_{\text{dimer}} = \sum_{a=0}^{N/2-1} (J\mathbf{S}_{2a} \cdot \mathbf{S}_{2a+1} + J'\mathbf{S}_{2a+1} \cdot \mathbf{S}_{2a+2}) = H_0 + H'. \quad (7)$$

We first make some remarks about the  $J'=0$  case, in which the spins decouple into pairs on the bonds  $J$ . The ground state of  $H_0$  is unique and is given by

$$|0\rangle = \prod_{a=0}^{N/2-1} |0\rangle_a, \quad (8)$$

$$|0\rangle_a = \frac{1}{\sqrt{2}}(|\uparrow\rangle_{2a}|\downarrow\rangle_{2a+1} - |\downarrow\rangle_{2a}|\uparrow\rangle_{2a+1}) \quad (9)$$

so that  $|0\rangle_0$  is a singlet between sites 0 and 1. We take the associated eigenvalue  $E_0 = -3NJ/8$  as zero energy.

### A. Excitations

Excitations are formed by breaking singlets to create triplets. The spectrum of  $H_0$  then consists of degenerate levels at energies  $nJ$  relative to the ground state, where  $n < N/2$  is the number of triplets. These excitations are dispersionless hard-core bosons. When the perturbation  $H'$  is applied, the degeneracies are removed, and coherent single-particle excitations with dispersion relation  $\epsilon_p$  are formed. These magnons are not free, but interact with each other through the perturbation  $H'$  in addition to being subject to the hard-core constraint. The first-excited state consists of  $N/2-1$  singlets and one triplet, leading to a total spin  $S_{\text{tot}}=1$ .

We define  $d_a(m)$  as the operator that breaks a dimer between sites  $2a$  and  $2a+1$ , creating a state with  $z$  component of spin  $m$ . For example the explicit form of one of these operators is

$$d_a(0) = d_a^\dagger(0) = 2S_{2a}^z, \quad (10)$$

although it is important to realize that  $d(\pm 1) \neq d^\dagger(\pm 1)$ . A translationally invariant state is formed by taking the Fourier transform

$$|p, m\rangle = \sqrt{\frac{2}{N}} \sum_{a=0}^{N/2-1} e^{2ipa} d_a(m) |0\rangle. \quad (11)$$

Here the factor of 2 in the exponent accounts for the inter-dimer distance. Periodic boundary conditions lead to the quantization condition

$$e^{ipN} = 1,$$

so that

$$p = \frac{2\pi n}{N}, \quad n = 0, 1, 2, \dots, N/2 - 1.$$

Strictly at the point  $\alpha=0$ , these single-particle excitations are  $N/2$ -fold degenerate with a flat dispersion  $\epsilon_p = J$ . A finite value of  $\alpha$  causes magnons to ‘‘hop.’’ To the first order in  $\alpha$ , the dispersion is

$$\epsilon_p = J - \frac{J'}{2} \cos(2p), \quad (12)$$

resulting in a gap

$$\Delta = J - \frac{J'}{2}. \quad (13)$$

Using translational and spin-rotational invariances, we can express two-magnon states (to lowest order in  $\alpha$ ) in the form

$$|p_1, p_2, S, m\rangle = \mathcal{N}_S(p_1, p_2) \sum_{a=1}^{N/2-1} \sum_{b=0}^{a-1} \psi_{ab}^S(p_1, p_2) \Phi_{ab}^{S,m} |0\rangle. \quad (14)$$

Here  $S=0, 1, 2$  and the normalization  $\mathcal{N}$  will, in general, be dependent on the linear and angular momenta. Explicit expressions for the spin part  $\Phi_{ab}^{S,m}$  are given in Appendix A. The wave function is given by

$$\psi_{ab}^S(p_1, p_2) = e^{2i(p_1 a + p_2 b)} + A_{p_1 p_2}^S e^{2i(p_1 b + p_2 a)}. \quad (15)$$

Embodied in the nontrivial relative phase  $A_{p_1 p_2}^S$  is the magnon-magnon interaction. For  $\alpha=0$  the form of  $A$  is unspecified because the magnons cannot hop onto the same site and as such do not interact. To the lowest order in  $\alpha$ , the correct basis in degenerate perturbation theory is given by requiring

$$\mathcal{P}_2 H' |p_1, p_2, S, m\rangle = -\frac{J'}{2} [\cos(2p_1) + \cos(2p_2)] |p_1, p_2, S, m\rangle, \quad (16)$$

where  $\mathcal{P}_2$  is the projection operator onto the two-particle states. When the triplets in the sum given in Eq. (14) are well separated ( $|a-b| > 1$ ), the condition (16) is trivially satisfied, independently of  $A$ . When the triplets are neighboring ( $|a-b|=1$ ), we find

$$A_{p_1 p_2}^0 = -\frac{1 + e^{-2i(p_1 + p_2)} - 2e^{-2ip_2}}{1 + e^{-2i(p_1 + p_2)} - 2e^{-2ip_1}}, \quad (17a)$$

$$A_{p_1 p_2}^1 = -\frac{1 + e^{-2i(p_1 + p_2)} - e^{-2ip_2}}{1 + e^{-2i(p_1 + p_2)} - e^{-2ip_1}}, \quad (17b)$$

$$A_{p_1 p_2}^2 = -\frac{1 + e^{-2i(p_1 + p_2)} + e^{-2ip_2}}{1 + e^{-2i(p_1 + p_2)} + e^{-2ip_1}}. \quad (17c)$$

The magnons therefore experience both an infinite onsite repulsion and a nearest-neighbor momentum and spin-dependent interaction. Periodic boundary conditions and the restriction on the sums lead to the conditions

$$A_{p_1 p_2}^S = e^{ip_1 N} \quad \text{and} \quad A_{p_1 p_2}^S e^{ip_2 N} = 1, \quad (18)$$

with the implication that the quantization of the two-particle momenta depends on the total spin  $S$ . These are in fact the Bethe-ansatz equations for the spin-1/2 XXZ chain, where the sectors  $S=0, 1, 2$  correspond to anisotropies  $\Delta=1, +\frac{1}{2}, -\frac{1}{2}$ , respectively. Solving these equations for finite  $N$  to find  $p_1$  and  $p_2$  is a numerical task, which we outline in Appendix B.

TABLE I. Nonzero matrix elements of the interband type for  $S_j^z$  acting at sites  $j=0, 1$ .

$\langle 0 S_j^z p, m\rangle$	$(-1)^j \sqrt{\frac{1}{2N}} \delta_{m,0}$
$\langle p_1, p_2, 0, 0 S_j^z p, 0\rangle$	$(-1)^j \sqrt{\frac{2}{3N^3}} U_0(p, p_1, p_2)$
$\langle p_1, p_2, 2, 0 S_j^z p, 0\rangle$	$(-1)^{j+1} \sqrt{\frac{4}{3N^3}} U_2(p, p_1, p_2)$
$\langle p_1, p_2, 1, 1 S_j^z p, 1\rangle$	$(-1)^{j+1} \sqrt{\frac{1}{N^3}} U_1(p, p_1, p_2)$
$\langle p_1, p_2, 1, -1 S_j^z p, -1\rangle$	$(-1)^j \sqrt{\frac{1}{N^3}} U_1(p, p_1, p_2)$
$\langle p_1, p_2, 2, 1 S_j^z p, 1\rangle$	$(-1)^{j+1} \sqrt{\frac{1}{N^3}} U_2(p, p_1, p_2)$
$\langle p_1, p_2, 2, -1 S_j^z p, -1\rangle$	$(-1)^{j+1} \sqrt{\frac{1}{N^3}} U_2(p, p_1, p_2)$

We note here, however, that the solutions may be complex, leading to bound states, in agreement with results in the literature.<sup>16–18</sup> For notational convenience, we define a phase shift by

$$\delta_{p_1 p_2}^S = -\frac{i}{2} \ln(A_{p_1 p_2}^S). \quad (19)$$

For real  $p_1, p_2$ , the normalization of a two-particle state is given by

$$\mathcal{N}_S(p_1, p_2) = \left[ \frac{N}{2} \left( \frac{N}{2} - 1 \right) - \frac{N \cos(2\delta_{p_1 p_2}^S) - \cos(2p_1 - 2p_2 - 2\delta_{p_1 p_2}^S)}{1 - \cos(2p_1 - 2p_2)} \right]^{-1/2}. \quad (20)$$

We note that two-magnon states have the symmetry

$$|p_1, p_2, S, m\rangle = e^{-2i\delta_{p_1 p_2}^S} |p_2, p_1, S, m\rangle. \quad (21)$$

To avoid an overcomplete basis we make the restriction  $p_1 > p_2$ .

### B. Matrix elements

For small  $\alpha$  the gap to excitations is of order  $J$  and the states with  $n$  magnons are suppressed by a factor  $\exp(-\beta n J)$  in the thermal trace (2). At low temperatures  $\beta J \gg 1$ , we then make the approximation that the relevant states are those with  $n \leq 2$  magnons. Due to the isotropy in spin space of  $H_{\text{dimer}}$ , we require only the  $\chi^{zz}$  component of the dynamical susceptibility. In addition the Hamiltonian is invariant under translations by two sites. When evaluating Eq. (2), it is then sufficient to consider matrix elements of the form  $\langle l|S_j^z|m\rangle$ , where  $j=0, 1$  and  $l, m$  correspond to states with zero, one, or two magnons. Calculating such elements is simple to zeroth order in perturbation theory and the results are summarized in Tables I and II. Certain matrix elements are not given in the tables because they are identically zero. This can be seen by taking account of the fact that the operator  $S_j^z$  commutes with the total  $z$  component of spin, lead-

 TABLE II. Nonzero matrix elements of the intraband type for  $S_j^z$  acting at sites  $j=0, 1$ .

$\langle p', m S_j^z p, m\rangle$	$\frac{1}{N}(\delta_{m,1} - \delta_{m,-1})$
$\langle p'_1, p'_2, 1, 0 S_j^z p_1, p_2, 0, 0\rangle$	$\sqrt{\frac{8}{3N^4}} V_{10}(p'_1, p'_2, p_1, p_2)$
$\langle p'_1, p'_2, 2, 0 S_j^z p_1, p_2, 1, 0\rangle$	$\sqrt{\frac{4}{3N^4}} V_{21}(p'_1, p'_2, p_1, p_2)$
$\langle p'_1, p'_2, 1, \pm 1 S_j^z p_1, p_2, 1, \pm 1\rangle$	$\mp \sqrt{\frac{1}{N^4}} V_{11}(p'_1, p'_2, p_1, p_2)$
$\langle p'_1, p'_2, 2, \pm 1 S_j^z p_1, p_2, 1, \pm 1\rangle$	$\pm \sqrt{\frac{1}{N^4}} V_{21}(p'_1, p'_2, p_1, p_2)$
$\langle p'_1, p'_2, 2, \pm 1 S_j^z p_1, p_2, 2, \pm 1\rangle$	$\pm \sqrt{\frac{1}{N^4}} V_{22}(p'_1, p'_2, p_1, p_2)$
$\langle p'_1, p'_2, 2, \pm 2 S_j^z p_1, p_2, 2, \pm 2\rangle$	$\mp \sqrt{\frac{4}{N^4}} V_{22}(p'_1, p'_2, p_1, p_2)$

ing to the transition selection rule  $\Delta S^z=0$ . In addition some elements are zero by inspection of the states given in Eqs. (A2a)–(A2i).

We define the functions

$$U_S(p, p_1, p_2) = \frac{N}{2} \mathcal{N}_S(p_1, p_2) e^{-i\delta_{p_1 p_2}^S} \left[ \frac{\sin(p - p_1 + \delta_{p_1 p_2}^S)}{\sin(p - p_1)} + \frac{\sin(p - p_2 - \delta_{p_1 p_2}^S)}{\sin(p - p_2)} \right], \quad (22)$$

and

$$V_{S'S}(p'_1, p'_2, p_1, p_2) = \left( \frac{N}{2} \right)^2 \mathcal{N}_S(p_1, p_2) \mathcal{N}_{S'}(p'_1, p'_2) \exp[i(\delta_{p_1 p_2}^S - \delta_{p'_1 p'_2}^{S'})] \left[ \frac{\sin(p_1 - p'_1 - \delta_{p_1 p_2}^S + \delta_{p'_1 p'_2}^{S'})}{\sin(p_1 - p'_1)} + \frac{\sin(p_2 - p'_2 + \delta_{p_1 p_2}^S - \delta_{p'_1 p'_2}^{S'})}{\sin(p_2 - p'_2)} + \frac{\sin(p_1 - p'_2 - \delta_{p_1 p_2}^S - \delta_{p'_1 p'_2}^{S'})}{\sin(p_1 - p'_2)} + \frac{\sin(p_2 - p'_1 + \delta_{p_1 p_2}^S + \delta_{p'_1 p'_2}^{S'})}{\sin(p_2 - p'_1)} \right], \quad (23)$$

which are useful when calculating matrix elements that involve two-particle states.

### III. SPECTRAL REPRESENTATION AND RESUMMATION

Taking the definition of the susceptibility in the Matsubara formalism (2), it is helpful to expand in terms of operators at even and odd sites,

$$\begin{aligned}\chi^{zz}(\omega, Q) = & - \int_0^\beta d\tau e^{i\omega_n\tau} \frac{1}{N} \sum_{l, l'=0}^{N/2-1} e^{-i2Q(l-l')} [\langle S_{2l}^z(\tau) S_{2l'}^z \rangle \\ & + \langle S_{2l+1}^z(\tau) S_{2l'}^z \rangle e^{-iQ} + e^{iQ} \langle S_{2l}^z(\tau) S_{2l'+1}^z \rangle \\ & + \langle S_{2l+1}^z(\tau) S_{2l'+1}^z \rangle] \Big|_{\omega_n \rightarrow \eta - i\omega}.\end{aligned}\quad (24)$$

Using translational symmetry and grouping terms according to magnon number, the susceptibility can be written as

$$\begin{aligned}\chi^{zz}(\omega, Q) & \equiv \frac{1}{Z} \sum_{r,s=0}^{\infty} C_{rs}, \\ C_{rs} = & - \int_0^\beta d\tau e^{i\omega_n\tau} \frac{1}{N} \sum_{l, l'=0}^{N/2-1} e^{-i2Q(l-l')} \sum_{\gamma_r, \gamma_s} e^{-\beta E_{\gamma_r}} \\ & \times e^{-\tau[E_{\gamma_s} - E_{\gamma_r}]} e^{i2(l-l')[P_{\gamma_s} - P_{\gamma_r}]} M_{\gamma_r, \gamma_s} \Big|_{\omega_n \rightarrow \eta - i\omega}.\end{aligned}\quad (25)$$

Here  $\gamma_s$  is a multi-index enumerating all  $s$ -particle states,  $E_{\gamma_s}$  and  $P_{\gamma_s}$  are the energy and momentum of the excited state  $|\gamma_s\rangle$ ,  $Z$  is the partition function, and

$$\begin{aligned}M_{\gamma_r, \gamma_s} = & |\langle \gamma_r | S_0^z | \gamma_s \rangle|^2 + e^{iQ} \langle \gamma_r | S_0^z | \gamma_s \rangle \langle \gamma_s | S_1^z | \gamma_r \rangle + |\langle \gamma_r | S_1^z | \gamma_s \rangle|^2 \\ & + e^{-iQ} \langle \gamma_r | S_1^z | \gamma_s \rangle \langle \gamma_s | S_0^z | \gamma_r \rangle.\end{aligned}\quad (26)$$

We have also suppressed the energy and momentum labels on  $C_{rs}$  for notational simplicity. Carrying out the Fourier transform, we have

$$C_{rs} = \sum_{\gamma_r, \gamma_s} \frac{N}{4} \delta_{Q+P_{\gamma_r}, P_{\gamma_s}} \frac{e^{-\beta E_{\gamma_r}} - e^{-\beta E_{\gamma_s}}}{\omega + i\eta + E_{\gamma_r} - E_{\gamma_s}} M_{\gamma_r, \gamma_s}.\quad (27)$$

The nonvanishing contribution at  $T=0$  is obtained from  $C_{10} + C_{01}$ ,

$$C_{10} + C_{01} = \frac{1 - \cos(Q)}{4} (1 - e^{-\beta\epsilon_Q}) \left[ \frac{1}{\omega + i\eta - \epsilon_Q} - \frac{1}{\omega + i\eta + \epsilon_Q} \right]\quad (28)$$

$$\equiv (1 - e^{-\beta\epsilon_Q}) D(\omega, Q).\quad (29)$$

$D(\omega, Q)$  is then the bare magnon propagator. The remaining  $C_{rs}$  terms, up to  $C_{12} + C_{21}$ , are obtained in a simple manner using Eq. (27) and the matrix elements in Tables I and II,

$$C_{11} = \frac{1 + \cos(Q)}{N} \sum_p \frac{e^{-\beta\epsilon_p} - e^{-\beta\epsilon_{p+Q}}}{\omega + \epsilon_p - \epsilon_{p+Q} + i\eta},\quad (30)$$

$$\begin{aligned}C_{12} + C_{21} = & \frac{1 - \cos(Q)}{4} \left( \frac{2}{N} \right)^2 \sum_S \sum_{p_1 > p_2} \left( \frac{2S+1}{3} \right) U_S^2(Q + p_1 \\ & + p_2, p_1, p_2) [e^{-\beta\epsilon_{Q+p_1+p_2}} - e^{-\beta(\epsilon_{p_1} + \epsilon_{p_2})}] \\ & \times \left\{ \frac{1}{\omega + \epsilon_{Q+p_1+p_2} - \epsilon_{p_1} - \epsilon_{p_2} + i\eta} - \frac{1}{\omega - \epsilon_{Q+p_1+p_2} + \epsilon_{p_1} + \epsilon_{p_2} + i\eta} \right\}.\end{aligned}\quad (31)$$

We reiterate that the allowed values of  $p_1, p_2$  in the sum above depend on  $S$  and that the sum is only over those momenta that produce unique two-particle states.

We now make some initial remarks about the structure of the  $C_{rs}$  terms. First, the two-site dimer basis leads to a  $Q$ -dependent prefactor that differs between the interband terms  $C_{r,r+1}, C_{r+1,r}$  and intraband terms  $C_{rr}$ . Second, the  $C_{12} + C_{21}$  term will diverge with system size as  $N$ . This divergence is expected and should cancel with terms arising from an expansion of the partition function  $Z$ . Lastly, the  $\omega$  dependence in the denominators is such that interband terms diverge as  $\omega \rightarrow \epsilon_Q$ . This divergence is reflected in the intraband terms, which diverge for  $\omega \rightarrow \pm J' \sin(Q)$ . Higher-order terms have stronger divergences. This standard behavior is a consequence of the essentially nonperturbative nature of the finite  $T$ -magnon lifetime. Perturbation theory is unable to capture, order by order, the decay-enhancing effect of particle-particle scattering processes, hence, lifetimes remain infinite to all orders. Treating the interaction accurately and rendering the lifetimes finite require a summation of perturbation-theory terms to infinite order. We achieve this infinite summation for certain processes as follows. Taking into account interactions between the magnons by a Dyson equation, we conclude that we can write

$$\chi^{zz}(\omega, Q) = \frac{D(\omega, Q)}{1 - D(\omega, Q)\Sigma(\omega, Q)}.\quad (32)$$

Expanding this we obtain

$$\chi^{zz}(\omega, Q) = [D(\omega, Q) + D^2(\omega, Q)\Sigma(\omega, Q) + \dots].\quad (33)$$

On the other hand, from our low-temperature expansion of the spectral representation, we have

$$\begin{aligned}\chi^{zz}(\omega, Q) & = \frac{1}{Z} [C_{01} + C_{10} + C_{11} + C_{12} + C_{21} + \dots] \\ & = \frac{1}{1 + Z_1 + Z_2 + \dots} [(1 - e^{-\beta\epsilon_Q})D(\omega, Q) + C_{11} + C_{12} \\ & \quad + C_{21} + \dots] \\ & = (1 - e^{-\beta\epsilon_Q})D(\omega, Q) + C_{11} + C_{12} + C_{21} \\ & \quad - Z_1(1 - e^{-\beta\epsilon_Q})D(\omega, Q) + \dots \\ & = D(\omega, Q) + [C_{11} + C_{12} + C_{21} - Z_1(1 - e^{-\beta\epsilon_Q}) \\ & \quad \times D(\omega, Q) - e^{-\beta\epsilon_Q}D(\omega, Q)] + \dots.\end{aligned}\quad (34)$$

The contribution to the partition function from the one-particle states  $Z_1 = 3 \sum_p e^{-\beta\epsilon_p}$  must be included to cancel the  $N$  dependence of the  $C_{12} + C_{21}$  contribution. Comparing the two expansions, we make the identification

$$\begin{aligned}\Sigma(\omega, Q) \approx & D^{-2}(\omega, Q) \{ (C_{11} + C_{12} + C_{21}) - [Z_1(1 - e^{-\beta\epsilon_Q}) \\ & + e^{-\beta\epsilon_Q}] D(\omega, Q) \}.\end{aligned}\quad (35)$$

Finally we calculate the quantity of experimental interest as

$$S^{zz}(\omega, Q) = - \lim_{\eta \rightarrow 0} \frac{1}{\pi} \frac{1}{1 - e^{-\beta\omega}} \text{Im} \left[ \frac{D(\omega, Q)}{1 - D(\omega, Q)\Sigma(\omega, Q)} \right].\quad (36)$$

**IV. RESULTS AND DISCUSSION**

We now choose  $J=1$  and  $J'=0.1$  so that  $\alpha=0.1$  is small as required by our expansion. We consider temperatures less than the gap, so that the low-magnon number approximation holds. We then calculate the dynamical structure factor at a given  $\omega$  and  $Q$  numerically. Sums over momenta such as those in Eqs. (30) and (31) are performed for systems of  $N/2$  dimers. Analytically, the standard procedure for evaluating Eq. (2) is to take the thermodynamic limit, perform the resulting momentum integrals, extract the imaginary part of the resulting susceptibility, and finally take the limit  $\eta \rightarrow 0$ . The zero-temperature result for  $S^{zz}(\omega, Q)$  then takes the form of a delta function at  $\omega = \epsilon_Q$ . For the purposes of numerics, it is necessary to stipulate  $\eta$  before performing the sums. This results in a broadened zero-temperature result, a Lorentzian peak of width  $\eta$ . To obtain accurate results at finite  $T$ , this width must be small in comparison with the thermally activated broadening, which scales as  $J'e^{-\beta J}$ . In contrast, to avoid finite-size effects, the sums must be evaluated on a grid in wave vector space that is fine enough to resolve the Lorentzian. The condition for producing accurate numerics is then

$$e^{-\beta J} \gg \frac{2\eta}{J'} > \frac{4\pi}{N}. \tag{37}$$

In principle by increasing the system size  $N$ , very small values of  $\eta$  could be used. However, as explained above this is only necessary at very low temperatures, where the broadening is not sufficiently asymmetric to be interesting. For the range of temperatures we investigate, we find a suitable value of  $\eta$  to be 0.001. When calculating the intraband response, we typically use systems of size  $N/2=600$ . The interband response is less sensitive to finite-size effects and sums, using a smaller system size  $N/2=400$ , are permissible. We find that the effects of using a larger number of dimers are negligible. If the delta function in Eq. (27) is ever to be satisfied, we must restrict the external momentum to  $Q = 4\pi n/N$  with integer  $n$ . We leave the discussion of further issues affecting numerical accuracy, in particular, bound-state solutions of Eq. (18) to Appendix B.

We first consider the behavior of the one-magnon or interband response and examine the lineshape in energy by fixing the external wave vector  $Q$ . Figure 1 shows the dynamical structure factor at  $Q = \pi$  for a range of temperatures. At temperatures less than the gap, the main feature is a peak at  $\omega = \epsilon_Q$ . At temperatures below  $T \sim 0.3J$ , the peak is approximately Lorentzian. The maximum response falls rapidly with increasing temperature and the peak broadens asymmetrically. In particular the peak becomes skewed with a tail extending toward energies  $\omega \sim J$ .

The degree of asymmetry is large compared to that found for a variety of other spin chains using a semiclassical approach.<sup>7,8</sup> Instead it is similar to the asymmetry found by exact diagonalization.<sup>10</sup> The broadening also resembles that found for two integrable spin chains in Ref. 11, which uses the same resummation scheme as this paper. Most importantly the asymmetry has been found to be consistent with recent data on copper nitrate.<sup>15</sup> We also point out that the

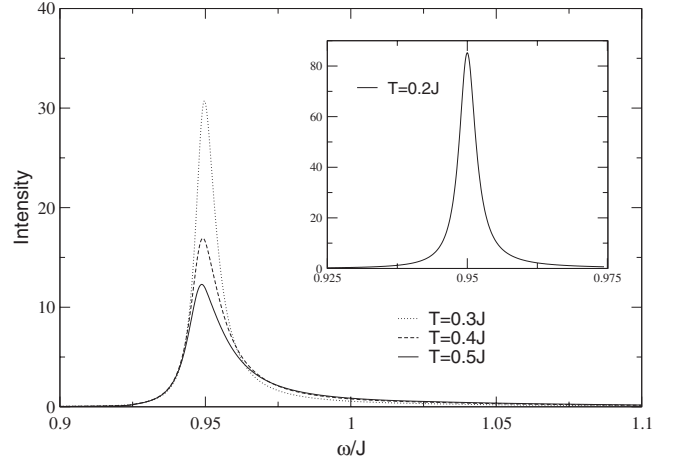


FIG. 1. The one-magnon response at  $Q = \pi$  for  $N/2=400$ . At  $T = 0.2J$  the lineshape is nearly Lorentzian (inset) but is increasingly asymmetric as temperature approaches the energy gap.

asymmetry is primarily a consequence of the two-magnon interband terms  $C_{12} + C_{21}$ . Intraband terms  $C_{rr}$  have vanishing spectral weight in this region and their lack of influence is confirmed by observing that the one-magnon mode is unaffected if we neglect them in the resummation.

The behavior in wave-vector space is dominated by the static structure factor,  $1 - \cos(Q)$ , arising from the two-site dimer basis. As a result, the response is maximal for  $Q = (2n+1)\pi$  and disappears altogether at  $Q = 2n\pi$  as in Figs. 2 and 3 (for integer  $n$ ). Exactly at  $Q = (2n+1)\pi/4$ , the susceptibility  $\chi^{zz}$  is a symmetric function of  $\omega$  about the point  $\omega = J$ ; however, the dynamical structure factor  $S^{zz}$  is not symmetric because spectral weight is shifted to higher energies by the factor  $(1 - e^{-\beta\omega})^{-1}$ .

The direction of asymmetry in the dynamical response at  $\omega \sim \epsilon_Q$  can be understood qualitatively in terms of a joint density of states for transitions between occupied one-magnon and unoccupied two-magnon states. For  $\omega > 0$ , this takes the form

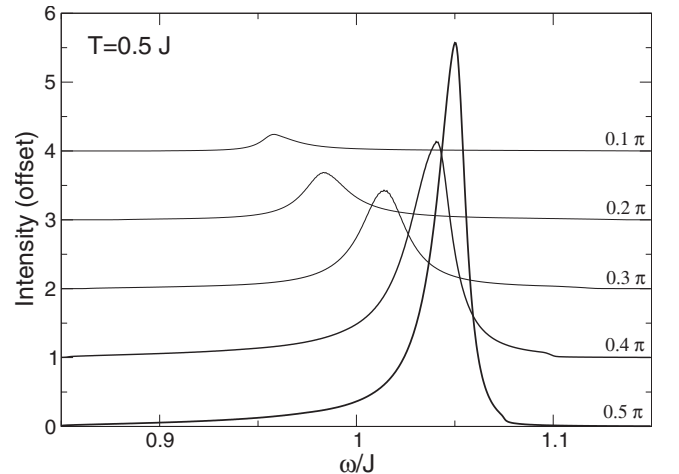


FIG. 2. Wave-vector dependence of the one-magnon or interband response. The peak position is given by  $\omega = \epsilon_Q$ . At  $Q=0$  the response vanishes. Top: Wave vectors between  $Q=2\pi/10$  and  $\pi/2$ . The vertical axis is offset by integer values  $5 - m$  for  $Q = m\pi/10$ .

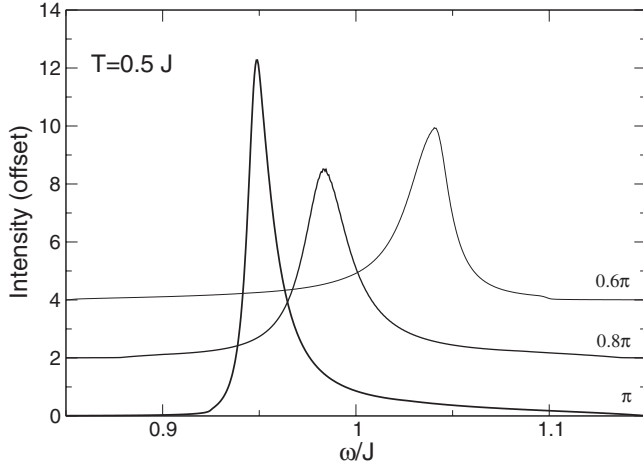


FIG. 3. Wave-vector dependence of the one-magnon or interband response. The peak position is given by  $\omega = \epsilon_Q$ . At  $Q=0$  the response vanishes. Wave vectors between  $Q=3\pi/5$  and  $\pi$ . The vertical axis is offset by integer values  $10-m$  for  $Q=m\pi/10$ .

$$N_{1 \rightarrow 2} = \sum_{p, p_1, p_2} n(p) \bar{n}(p_1, p_2) \delta_{Q+p, p_1+p_2} \delta_{\omega+\epsilon_p, \epsilon_{p_1}+\epsilon_{p_2}} \quad (38)$$

Here  $n(p)$  is the thermal occupation number for a one-magnon state with momentum  $p$  and  $\bar{n}(p_1, p_2)$  is the probability that the two-magnon state characterized by momenta  $p_1$  and  $p_2$  is unoccupied. At low temperatures and weak interdimer interactions, we have approximately

$$n(p) \approx e^{-\beta\epsilon_p}, \quad \bar{n}(p_1, p_2) \approx (1 - e^{-\beta\epsilon_{p_1}})(1 - e^{-\beta\epsilon_{p_2}}). \quad (39)$$

For  $-\pi/4 < Q < \pi/4$  and  $\pi/4 < Q < 3\pi/4$ , this function is skewed toward higher and lower energies, respectively. On the other hand the specific form of the lineshape is dictated by the matrix element  $M_{12}$  and hence by the magnon-magnon interaction. The fact that the lineshape of the dynamical response at  $\omega \approx \epsilon_Q$  is skewed toward low frequencies in some regions of the Brillouin zone and toward high frequencies in others is a consequence of the smallness of the ratio of bandwidth to magnon gap. This should be contrasted to the findings of Ref. 11 for the lineshape in the O(3) nonlinear sigma model, for which the bandwidth is infinite and, concomitantly, the asymmetry was found to always extend toward higher energies.

Next we turn to the intraband response. When magnons are thermally excited, incident neutrons can scatter them within the same band with energy transfers, which are small compared to the gap. Accordingly, at finite temperatures, there is a spin response at energies  $\omega \sim 0$ . To the lowest order, the contribution to the intraband response is given by

$$-\frac{1}{\pi} \frac{1}{1 - e^{-\beta\omega}} \text{Im} C_{11} = \frac{1 + \cos(Q)}{2\pi} \frac{e^{-\beta(J-\omega/2)}}{\sqrt{[J' \sin(Q)]^2 - \omega^2}} \times \cosh \left\{ \frac{\beta}{2} \cot(Q) \sqrt{[J' \sin(Q)]^2 - \omega^2} \right\}, \quad (40)$$

valid for  $|\omega| \leq J' |\sin(Q)|$ . This response is bounded by in-

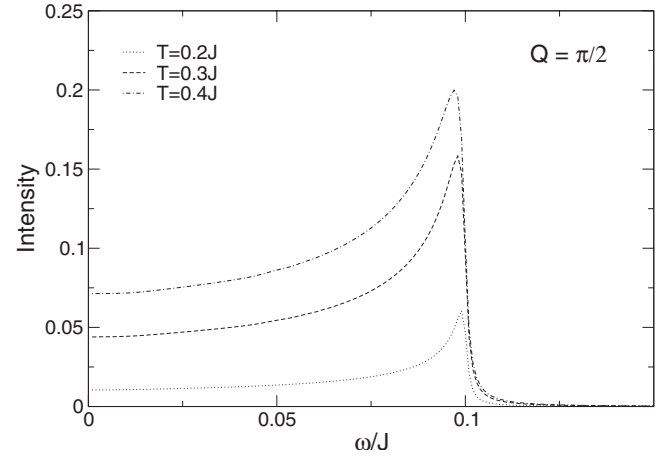


FIG. 4. Development of intraband scattering with temperature at  $Q = \pi/2$  for  $N/2 = 600$  dimers.

verse square-root singularities and has an overall magnitude that grows with temperature as  $e^{-\beta J}$ . We have calculated the next leading contribution  $C_{22}$  using the matrix elements in Table II and found it to exhibit a stronger divergence at  $\omega = \pm J' \sin(Q)$ . This shows that, just as for the interband contributions, a resummation needs to be carried out. This is achieved by including the intraband scattering contributions in the low-temperature expansion of the self-energy in Eq. (32). A complication that arises in doing so is that because of the different prefactors  $1 \pm \cos(Q)$  for intraband and interband scatterings, the result of the resummation is reliable only at very low temperatures for certain wave vectors. For such values of  $Q$ , higher-order terms, such as  $C_{22}$ , should be taken into account. However, a consistent treatment of such terms would require the incorporation of interband contributions involving three-magnon states, which is beyond the scope of this work. By including  $C_{11}$  in the resummation, we remove the square-root singularities and associated threshold. Instead the response has two finite peaks and falls rapidly to zero for  $|\omega| > J' |\sin(Q)|$ . This is physically sensible and is analogous to what is found for the intraband scattering in the spin-1/2 Heisenberg-Ising chain.<sup>31</sup> In Fig. 4 we show the calculated intraband scattering at  $Q = \pi/2$ . The  $Q$ -dependent range in  $\omega$  of this scattering compares well with that calculated previously by exact diagonalization of chains with  $N=16$  sites.<sup>10</sup> In that case, however, the small system size limited the number of available transitions and so the lineshape was not representative of the thermodynamic limit.

In conclusion we have calculated the approximate dynamical structure factor at finite temperature of the alternating Heisenberg chain in the limits of strong alternation and low temperature. The method we use has previously been applied to integrable spin chains, but in this case, the system is nonintegrable. We find that the lineshape of the lowest-lying one-magnon mode is increasingly asymmetric with temperature, a direct consequence of magnon-magnon interactions. We also establish a prediction for the low-temperature lineshape of the intraband scattering.

### ACKNOWLEDGMENTS

We are grateful to Bella Lake and Alan Tennant for their numerous enlightening discussions. This work was supported by the EPSRC-GB under Grant No. GR/R83712/01 (F.H.L.E. and A.J.A.J.), the DOE under Contract No. DE-AC02-98 CH 10886 (R.M.K.), and the ESF network INSTANS.

### APPENDIX A: TWO-MAGNON STATES

The spin part of the two-magnon states is of the form

$$\Phi_{ll'}^{S,m} = \sum_{\{m_1, m_2\}} c_{m_1, m_2}^{S,m} d_l(m_1) d_{l'}(m_2). \quad (\text{A1})$$

The  $c$ 's are Clebsch-Gordan coefficients but the explicit expressions are given below for convenience,

$$\Phi_{ll'}^{0,0} = \frac{1}{\sqrt{3}} [d_l(1) d_{l'}(-1) + d_l(-1) d_{l'}(1) - d_l(0) d_{l'}(0)], \quad (\text{A2a})$$

$$\Phi_{ll'}^{1,0} = \frac{1}{\sqrt{2}} [d_l(1) d_{l'}(-1) - d_l(-1) d_{l'}(1)], \quad (\text{A2b})$$

$$\Phi_{ll'}^{2,0} = \frac{1}{\sqrt{6}} [d_l(1) d_{l'}(-1) + d_l(-1) d_{l'}(1) + 2d_l(0) d_{l'}(0)], \quad (\text{A2c})$$

$$\Phi_{ll'}^{1,1} = \frac{1}{\sqrt{2}} [d_l(1) d_{l'}(0) - d_l(0) d_{l'}(1)], \quad (\text{A2d})$$

$$\Phi_{ll'}^{1,-1} = \frac{1}{\sqrt{2}} [d_l(0) d_{l'}(-1) - d_l(-1) d_{l'}(0)], \quad (\text{A2e})$$

$$\Phi_{ll'}^{2,1} = \frac{1}{\sqrt{2}} [d_l(1) d_{l'}(0) + d_l(0) d_{l'}(1)], \quad (\text{A2f})$$

$$\Phi_{ll'}^{2,-1} = \frac{1}{\sqrt{2}} [d_l(0) d_{l'}(-1) + d_l(-1) d_{l'}(0)], \quad (\text{A2g})$$

$$\Phi_{ll'}^{2,2} = d_l(1) d_{l'}(1), \quad (\text{A2h})$$

$$\Phi_{ll'}^{2,-2} = d_l(-1) d_{l'}(-1). \quad (\text{A2i})$$

### APPENDIX B: QUANTIZATION OF THE TWO-MAGNON MOMENTA

In order to carry out momentum sums over two-particle states on a finite lattice, we require knowledge of the allowed values of the momenta  $p_1$  and  $p_2$  in each sector  $S$ . In practice, this means that we must solve Eqs. (17) and (18) numerically to find the  $(N/2-1)N/4$  pairs  $\{p_1, p_2\}$  allowed by the condi-

tion  $p_1 > p_2$ . This is a problem usually encountered in models solvable by Bethe ansatz.<sup>32</sup>

#### 1. Real solutions

We first consider scattering states of two magnons, for which  $p_1$  and  $p_2$  are both real. In each spin sector  $S$ , Eq. (18) can be written in the form

$$e^{iNp_1} = A_{p_1 p_2}^S, \quad e^{iNp_2} = A_{p_2 p_1}^S. \quad (\text{B1})$$

In order to enumerate all roots of the coupled Eq. (B1), we take the logarithm. We choose a branch cut, such that

$$p_1 = -\frac{i}{N} \ln(-A_{p_1 p_2}^S) + \frac{\pi}{N} (2I_1 + 1),$$

$$p_2 = -\frac{i}{N} \ln(-A_{p_2 p_1}^S) + \frac{\pi}{N} (2I_2 + 1). \quad (\text{B2})$$

Here the integers  $I_{1,2}$  have the range  $0 \leq I_{1,2} < N/2$ . We note that  $I_1 \geq I_2$  implies that  $p_1 > p_2$  and, using the indistinguishability of particles, we can restrict ourselves without loss of generality to the case  $I_1 > I_2$ . Using the parametrization (B2), it is now a relatively straightforward matter to determine real roots corresponding to pairs of integers  $I_1 > I_2$  by standard numerical root-finding algorithms.

There are a number of roots, which require special treatment. Specifically, in the singlet sector, there is a class of real roots  $\{p_1 = 2(I_1 + 1)\pi/N, p_2 = 0\}$  and in the triplet and quintet sectors, there are solutions  $\{p_1 = \pi, p_2 = 0\}$ . For these cases the derivation of Eqs. (20), (22), and (23) needs to be revisited. For the special solutions in the singlet sector, the phase shift is zero and one finds (setting  $p_1 = q$ ),

$$\mathcal{N}^2(q, 0) = \left[ \left( \frac{N}{2} \right)^2 - N \right]^{-1}, \quad (\text{B3})$$

$$U(p, q, 0) = \mathcal{N}(q, 0) \left( \frac{N}{2} \delta_{p,0} - \frac{N}{2} \delta_{p-q,0} - 2 \right). \quad (\text{B4})$$

In the triplet and quintet sectors, the same expressions are found, but the arguments of the two Kronecker deltas above are never satisfied.

The matrix elements for  $C_{22}$  involving the special solutions are similarly affected and need to be replaced by

$$\begin{aligned} V_{S'S}(q', 0, p_1, p_2) &= -\mathcal{N}(q', 0) \mathcal{N}_S(p_1, p_2) \\ &\times \left( \frac{N}{2} \right)^2 e^{i\delta_{p_1 p_2}^S} \left[ 2 \cos(\delta_{p_1 p_2}^S) \right. \\ &+ \frac{\sin(p_1 - q' - \delta_{p_1 p_2}^S)}{\sin(p_1 - q')} \\ &\left. + \frac{\sin(p_2 - q' + \delta_{p_1 p_2}^S)}{\sin(p_2 - q')} \right], \quad (\text{B5}) \end{aligned}$$

$$V_{S'S}(q', 0, q, 0) = \mathcal{N}(q', 0) \mathcal{N}(q, 0) \left( \frac{N}{2} \right)^2 \left[ \frac{N}{2} - 4 \right], \quad (\text{B6})$$



$$V_{SS}(p_1, p_2, p_1, p_2) = 2\mathcal{N}_S^2(p_1, p_2) \left( \frac{N}{2} \right)^2 \left[ \frac{N}{2} - 1 - \frac{\sin(p_1 - p_2 - 2\delta_{p_1 p_2}^S)}{\sin(p_1 - p_2)} \right]. \quad (\text{B7})$$

The numerical root finder does not converge for  $\mathcal{O}(N)$  pairs of integers  $\{I_1^c, I_2^c\}$ . Most of these correspond to the complex solutions of Eq. (B2), which are discussed in Appendix B 2. So far we have restricted our discussion to real roots with distinct integers  $I_1 \neq I_2$ . The reason for this restriction is that  $I_1 = I_2$  corresponds generically to  $p_1 = p_2$ , which does not yield a valid solution of the Schrödinger equation. However, in analogy to what was shown in Ref. 33, there are additional “good” real solutions with repeating integers  $I_1 = I_2$ , which have to be treated with care.

## 2. Complex solutions

In addition to real roots, there exist complex solutions of Eq. (B2). These give rise to wave functions that exhibit an exponential decay with respect to the distance between the two magnons and hence correspond to bound states. As the Eq. (18) is closed under complex conjugation, complex roots must come in pairs,

$$p_1 = x + iy, \quad p_2 = x - iy, x, y \text{ real}. \quad (\text{B8})$$

Adding the two momenta using Eq. (B2) gives

$$p_1 + p_2 = 2x = \pi \frac{2}{N} (I_1^c + I_2^c + 1). \quad (\text{B9})$$

The real component  $x$  is now uniquely defined by the integers  $I_1^c, I_2^c$ . Defining  $\phi_s = S(S+1)/2 - 2$  and substituting for  $x$ , the value of  $y$  can be found by solving

$$e^{ip_1 N} = A_{p_1 p_1}^S$$

or equivalently

$$e^{ixN} e^{-yN} + \frac{1 + e^{-4ix} + \phi_s e^{-2ix} e^{-2y}}{1 + e^{-4ix} + \phi_s e^{-2ix} e^{2y}} = 0 \quad (\text{B10})$$

using a Newton-Raphson method. The resulting values of  $y$  are such that  $A_{p_1 p_1}^S$  or, equivalently,  $e^{ip_1 N}$  is either very small or very large. Consequently, in order to demonstrate that our solutions satisfy both of the Bethe-ansatz equations given in (18) numerically, the value of  $p_1$  must be known to very high precision. Fortunately the matrix elements are less sensitive and for the system sizes we consider, it transpires that to evaluate  $C_{12} + C_{21}$  accurately, 17 significant figures of  $p_1$  are sufficient. The two-particle normalization for complex momenta is given by

$$\mathcal{N}_S(x + iy, x - iy) = e^{yN/2} \left[ e^{ixN} \frac{N}{2} \left( \frac{N}{2} - 1 \right) - \frac{N \cosh(yN) - \cosh(4y - Ny)}{1 - \cosh(4y)} \right]^{-1/2}, \quad (\text{B11})$$

and for the matrix elements, the modification is

$$U_S(p, p_1, p_1^*) = \frac{N}{2} \mathcal{N}_S(x + iy, x - iy) \times \frac{(1 + A_{p_1 p_1}^S) \cos(2p - 2x) - (e^{-2y} + A_{p_1 p_1}^S e^{2y})}{\cos(2p - 2x) - \cosh(2y)}, \quad (\text{B12})$$

which is an even function of  $y$  as required. We find that the bound states affect only the fifth significant digit and higher of  $C_{12} + C_{21}$ , so that they are negligible in comparison to the contributions from other states.

<sup>1</sup>I. Affleck, in *Fields, Strings and Critical Phenomena*, edited by E. Brézin and J. Zinn-Justin (Elsevier, Amsterdam, 1989).  
<sup>2</sup>A. H. Bougourzi, M. Karbach, and G. Müller, Phys. Rev. B **57**, 11429 (1998).  
<sup>3</sup>F. H. L. Essler and R. M. Konik, in *From Fields to Strings: Circumnavigating Theoretical Physics*, edited by M. Shifman, A. Vainshtein, and J. Wheeler (World Scientific, Singapore, 2005).  
<sup>4</sup>J.-S. Caux, R. Hagemans, and J.-M. Maillet, J. Stat. Mech.: Theory Exp. (2005) P09003.  
<sup>5</sup>B. Lake, D. A. Tennant, and S. E. Nagler, Phys. Rev. Lett. **85**, 832 (2000); I. A. Zaliznyak, S.-H. Lee, and S. V. Petrov, *ibid.* **87**, 017202 (2001); M. Kenzelmann, G. Xu, I. A. Zaliznyak, C. Broholm, J. F. DiTusa, G. Aeppli, T. Ito, K. Oka, and H. Takagi, *ibid.* **90**, 087202 (2003); M. B. Stone, D. H. Reich, C. Broholm, K. Lefmann, C. Rischel, C. P. Landee, and M. M. Turnbull, *ibid.* **91**, 037205 (2003); A. Zheludev, Z. Honda, C. L. Broholm, K. Katsumata, S. M. Shapiro, A. Kolezhuk, S. Park, and Y. Qiu,

Phys. Rev. B **68**, 134438 (2003); I. A. Zaliznyak, H. Woo, T. G. Perring, C. L. Broholm, C. D. Frost, and H. Takagi, Phys. Rev. Lett. **93**, 087202 (2004); A. Zheludev, T. Masuda, G. Dhalenne, A. Revcolevschi, C. Frost, and T. Perring, Phys. Rev. B **75**, 054409 (2007); V. O. Garlea, A. Zheludev, L.-P. Regnault, J.-H. Chung, Y. Qiu, M. Boehm, K. Habicht, and M. Meissner, Phys. Rev. Lett. **100**, 037206 (2008).  
<sup>6</sup>S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, 1999).  
<sup>7</sup>S. Sachdev and A. P. Young, Phys. Rev. Lett. **78**, 2220 (1997).  
<sup>8</sup>K. Damle and S. Sachdev, Phys. Rev. B **57**, 8307 (1998).  
<sup>9</sup>R. M. Konik, Phys. Rev. B **68**, 104435 (2003); F. H. L. Essler and A. M. Tsvelik, Phys. Rev. Lett. **90**, 126401 (2003); K. Damle and S. Sachdev, *ibid.* **95**, 187201 (2005); B. Doyon, J. Stat. Mech.: Theory Exp. (2005) P11006; A. Rapp and G. Zarand, Phys. Rev. B **74**, 014433 (2006); S. A. Reyes and A. M. Tsvelik, *ibid.* **73**, 220405(R) (2006); B. Altshuler, R. M. Konik, and A. Tsvelik, Nucl. Phys. B **68**, 104435 (2006); B. Doyon and

- A. Gamsa, J. Stat. Mech.: Theory Exp. (2008) P03012.
- <sup>10</sup>H. J. Mikeska and C. Luckmann, Phys. Rev. B **73**, 184426 (2006).
- <sup>11</sup>F. H. L. Essler and R. M. Konik, arXiv:0711.2524 (unpublished).
- <sup>12</sup>I. Zaliznyak and S. Lee, in *Modern Techniques for Characterizing Magnetic Materials*, edited by Y. Zhu (Springer, Heidelberg, 2005).
- <sup>13</sup>D. A. Tennant, C. Broholm, D. H. Reich, S. E. Nagler, G. E. Granroth, T. Barnes, K. Damle, G. Xu, Y. Chen, and B. C. Sales, Phys. Rev. B **67**, 054414 (2003).
- <sup>14</sup>G. Xu, C. Broholm, D. H. Reich, and M. A. Adams, Phys. Rev. Lett. **84**, 4465 (2000).
- <sup>15</sup>D. A. Tennant *et al.* (unpublished).
- <sup>16</sup>T. Barnes, J. Riera, and D. A. Tennant, Phys. Rev. B **59**, 11384 (1999).
- <sup>17</sup>G. S. Uhrig and H. J. Schulz, Phys. Rev. B **54**, R9624 (1996).
- <sup>18</sup>A. Brooks Harris, Phys. Rev. B **7**, 3166 (1973).
- <sup>19</sup>W. Zheng, C. J. Hamer, and R. R. P. Singh, Phys. Rev. B **74**, 172407 (2006); W. Zheng, J. Oitmaa, and C. J. Hamer, arXiv:cond-mat/0412184 (unpublished).
- <sup>20</sup>G. Bouzerar, A. P. Kampf, and G. I. Japaridze, Phys. Rev. B **58**, 3117 (1998).
- <sup>21</sup>S. Trebst, H. Monien, C. J. Hamer, Z. Weihong, and R. R. P. Singh, Phys. Rev. Lett. **85**, 4373 (2000).
- <sup>22</sup>K. P. Schmidt, C. Knetter, and G. S. Uhrig, Phys. Rev. B **69**, 104417 (2004).
- <sup>23</sup>A. Collins, C. J. Hamer, and Z. Weihong, Phys. Rev. B **74**, 144414 (2006).
- <sup>24</sup>K. P. Schmidt and G. S. Uhrig, Phys. Rev. Lett. **90**, 227204 (2003).
- <sup>25</sup>C. J. Hamer, W. Zheng, and R. R. P. Singh, Phys. Rev. B **68**, 214408 (2003).
- <sup>26</sup>F. H. L. Essler, A. M. Tselik, and G. Delfino, Phys. Rev. B **56**, 11001 (1997).
- <sup>27</sup>F. Smirnov, *Form Factors in Completely Integrable Models of Quantum Field Theory* (World Scientific, Singapore, 1992).
- <sup>28</sup>J. Villain, Physica B (Amsterdam) **79**, 1 (1975).
- <sup>29</sup>S. E. Nagler, W. J. L. Buyers, R. L. Armstrong, and B. Briat, Phys. Rev. Lett. **49**, 590 (1982).
- <sup>30</sup>S. E. Nagler, W. J. L. Buyers, R. L. Armstrong, and B. Briat, J. Magn. Magn. Mater. **31-34**, 1213 (1983).
- <sup>31</sup>A. J. A. James and F. H. L. Essler (unpublished).
- <sup>32</sup>F. H. L. Essler, H. Frahm, F. Göhmann, A. Klümper, and V. E. Korepin, *The One-Dimensional Hubbard Model* (Cambridge University Press, Cambridge, 2005).
- <sup>33</sup>F. H. L. Essler, V. E. Korepin, and K. Schoutens, J. Phys. A **25**, 4115 (1992).