

## Approximate calculation of the excitation spectrum of the Heisenberg linear chain

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An approximate calculation of the excitation spectrum of the Heisenberg linear chain is performed by the Green's-function technique. Apart from the usual transverse Green's function, a longitudinal Green's function is also found useful. The decoupling procedure is not based on the two-sublattice picture, but utilizes the knowledge of known exact results about the ground state of the linear chain and the insight obtained from a model with explicitly known ground state. The equations are solved in a self-consistent manner, and the spectrum, except near the edge of the Brillouin zone, compares well the exact des Cloizeaux-Pearson result. We also provide an expression for the intensity for neutron-scattering studies; this can be checked against measurements on  $\text{CuCl}_2 \cdot 2\text{N}(\text{C}_5\text{D}_5)$  (CPC).

### I. INTRODUCTION

Recent work<sup>1</sup> on the dynamics of a spin- $\frac{1}{2}$  one-dimensional Heisenberg antiferromagnet, dichlorobis(pyridine)copper(II) (CPC), by Endoh, Shirane, Birgeneau, Richards, and Holt has fully confirmed the des Cloizeaux-Pearson excitation spectrum,<sup>2</sup> and has asked for "renewed theoretical effort on the  $S = \frac{1}{2}$  system in one dimension." Subsequently, Hohenberg and Brinkman<sup>3</sup> have shown that the spectral weight of the spin correlation functions measured by neutron-scattering studies is concentrated on these excited states. The des Cloizeaux-Pearson eigenvalues are essentially exact, but the scattering intensity is not exactly known, because the corresponding wave functions are very complicated. The antiferromagnetic spin-wave theory provides a reasonable description of the scattering intensity but is widely off on the spectrum. The purpose of the present work is to develop an approximation scheme that does better than the classical spin-wave theory for the eigenvalue spectrum as well as for the scattering intensity.

The Hamiltonian for the spin- $\frac{1}{2}$  linear chain with isotropic exchange is

$$H = 2J \sum_{i=1}^N \tilde{S}_i \cdot \tilde{S}_{i+1} \quad (1)$$

( $J > 0$ ,  $N$  even,  $N+1 \equiv 1$ ,  $|\tilde{S}_i| = \frac{1}{2}$ ). The ground state<sup>4</sup> has total spin  $S = 0$  and has energy  $-0.88629NJ$ . The first excited states are spin  $S = 1$  states, and the eigenvalues are given by<sup>2</sup>

$$\hbar\omega = \pi J |\sin q|. \quad (2)$$

$q$  is the wave vector measured with respect to that of the ground state (the wave vector of the ground state is 0 if  $N$  is a multiple of 4, and  $\pi$  if  $N$  is of the form  $4j+2$ , where  $j$  is an integer). We take

the lattice spacing to be unity. Note the characteristic double periodicity of spectrum (2).

The classical spin-wave theory<sup>5</sup> is based on the two-sublattice picture and uses the Holstein-Primakoff transformation for each sublattice. The underlying ground state is supposed to be the Néel state: up spins on one sublattice and down spins on the other. The excitation spectrum is

$$\hbar\omega = 2J |\sin k|. \quad (3)$$

A general critique of this approach is found in the work of Marshall.<sup>6</sup> The two-sublattice picture is based on Néel's insight into the physics of the problem, but is not justified from Hamiltonian (1). It is known that this picture is essentially the molecular-field idea, the staggered field for the antiferromagnets.<sup>7,8</sup> The Néel states are not eigenstates of the Hamiltonian, and are quite different from the ground state, which has all the  $\binom{N}{N/2}$  states with total  $S_z = 0$  contributing to it. That the excitation spectrum (3) differs from the exact one of Eq. (2) by a factor  $2/\pi$  has been regarded as mystifying,<sup>9</sup> but the double periodicity of (3) is artificially produced, and the degeneracy of these states is not correct.<sup>2</sup> Actually, spectrum (3) becomes correct in the limit<sup>10</sup>  $S \rightarrow \infty$ , while the  $S = \frac{1}{2}$  system is the extreme quantum limit.

To set up a simple approximation scheme, we try to avoid the pitfalls of the spin-wave theory for Hamiltonian (1). Fortunately, many exact results are known about (1); these are collected in Sec. II. We also know an antiferromagnetic model, Eq. (4) below, with known ground state,<sup>11</sup> from which we can derive useful insight and confirm many ideas of the approximation scheme. Both these models, Eqs. (1) and (4), do not show long-range order in the ground state, and are useful in the context of CPC. In Sec. III we write the equations for Green's functions for this problem. The main innovation here is the use of a

longitudinal Green's function in addition to the more familiar transverse ones. As usual we get a hierarchy of equations. Using the simplest decoupling possible, we terminate the hierarchy. Section IV solves the set of equations for the linear chain self-consistently, and compares the results with the exact ones and experiments. Section V discusses some features of the solution, and the generalizations that could perhaps be made from the approximation scheme.

## II. EXACT RESULTS IN ONE-DIMENSIONAL MODELS

For the linear chain of Eq. (1) many exact results are known. The relevant points for our work are as follows. The exact ground state is unique and nondegenerate and has total spin  $S=0$ . For the Heisenberg model, this result is proved by Peierls,<sup>6</sup> Marshall,<sup>6</sup> and Lieb and Mattis<sup>12</sup>; but for the linear chain it could also be inferred from the work of Hulthén.<sup>4</sup> It follows that the average value  $\langle 0|S_i^z|0\rangle$  is zero for every site  $i$ . The lowest excitation spectrum was computed by des Cloizeaux and Pearson,<sup>2</sup> as stated earlier. The correlation functions in the ground state  $\langle S_i^z S_j^z \rangle$  has not been calculated by the Bethe-Hulthén method, although the nearest-neighbor correlation function  $\langle S_i^z S_{i+1}^z \rangle$  is known from the work of Orbach<sup>13</sup> and Walker.<sup>14</sup> A detailed knowledge of these functions  $\langle S_i^z S_j^z \rangle$  can be obtained from the short-chain results of Bonner and Fisher,<sup>8</sup> who have also estimated the correlation function for the infinite linear chain (Table I) from short-chain calculations. The ground state has no long-range order. The correlation functions alternate in sign, and fall off with the separation between the spins. The alteration in sign is reminiscent of Néel's arrangement of spins. In other words, although the ground state has a very complicated structure, the correlation functions have the behavior expected from the Néel picture.

All these points can be demonstrated explicitly from the antiferromagnetic model with known ground state, discussed earlier by one of us.<sup>11</sup> The Hamiltonian here is a linear chain with the

next-nearest-neighbor (antiferromagnetic) interaction half that of the nearest neighbors:

$$H = 2J \sum_{i=1}^N \tilde{S}_i \cdot \tilde{S}_{i+1} + J \sum_{i=1}^N \tilde{S}_i \cdot \tilde{S}_{i+2} \quad (4)$$

( $J > 0$ ,  $N+1 \equiv 1$ ,  $N+2 \equiv 2$ ). The ground-state energy is  $E_0 = -\frac{3}{4}NJ$ , and the ground-state wave functions can be written

$$\Psi^+ = [1, 2][3, 4] \cdots [N-1, N] \\ + [2, 3][4, 5] \cdots [N, 1], \quad (5)$$

$$\Psi^- = [1, 2][3, 4] \cdots [N-1, N] \\ - [2, 3][4, 5] \cdots [N, 1], \quad (6)$$

with

$$[l, m] = \alpha(l)\beta(m) - \beta(l)\alpha(m). \quad (7)$$

The first one,  $\Psi^+$ , belongs to the  $k=0$  representation of the translation group. If  $T$  corresponds to the unit translation, we have

$$T\Psi^+ = \Psi^+ = e^{i0}\Psi^+. \quad (8)$$

Similarly,  $\Psi^-$  belongs to the  $k=\pi$  representation

$$T\Psi^- = -\Psi^- = e^{i\pi}\Psi^-. \quad (9)$$

We note that the ground state has spin 0 and  $\langle 0|S_i^z|0\rangle = 0$ ,  $\forall i$ . The order properties in these ground states can be calculated explicitly for the limit  $N \rightarrow \infty$  and they show only short-range order, either in the longitudinal  $\langle S_i^z S_j^z \rangle$  or the transverse  $\langle S_i^- S_j^+ \rangle$  correlation function. If the evanescent order is studied for short chains, one gets a remarkable result (Table II, cf. Table I), that the correlation functions  $\langle \sigma_i^z \sigma_{i+n}^z \rangle$  not only alternate in sign, but settle down to a constant absolute value, very much like what the Néel picture suggests. This absolute value represents long-range order, which diminishes with increasing number of spins in the chain, and disappears ultimately. Since the ground-state functions are so different from the Néel states, this behavior of the correlation functions is surprising, and reinforces the view that

TABLE I. Correlation functions of the linear chain as calculated by Bonner and Fisher (Ref. 8). (The last column is read from the figure of Ref. 8.  $\langle S_i^z S_j^z \rangle$  can be exactly calculated, since the ground-state energy can be expressed in terms of this correlation function.)

No. of spins	4	6	8	10	$\infty$
$4\langle S_1^z S_2^z \rangle$	-0.666 67	-0.622 84	-0.608 52	-0.602 06	-0.590 86
$4\langle S_1^z S_3^z \rangle$	0.333 33	0.277 35	0.261 04	0.254 07	0.25
$4\langle S_1^z S_4^z \rangle$		-0.309 02	-0.251 94	-0.231 17	-0.19
$4\langle S_1^z S_5^z \rangle$			0.198 83	0.173 07	0.15
$4\langle S_1^z S_6^z \rangle$				-0.187 81	

TABLE II. Spin correlation functions of the Hamiltonian (4). “+” indicates the state  $\Psi^+$ , Eq. (5), and “-” the state  $\Psi^-$ , Eq. (6).

No. of spins State	6		8		10		12		$\infty$	
	+	-	+	-	+	-	+	-	+	-
$\langle \sigma_1^z \sigma_2^z \rangle$	-0.333	-0.600	-0.556	-0.429	-0.467	-0.529	-0.516	-0.484	-0.5	-0.5
$\langle \sigma_1^z \sigma_3^z \rangle$	-0.333	0.200	0.111	-0.143	-0.067	0.059	0.030	-0.032	0	0
$\langle \sigma_1^z \sigma_4^z \rangle$	0.333	-0.200	-0.111	0.143	0.067	-0.059	-0.030	0.032	0	0
$\langle \sigma_1^z \sigma_5^z \rangle$			0.111	-0.143	-0.067	0.059	0.030	-0.032	0	0
$\langle \sigma_1^z \sigma_6^z \rangle$					0.067	-0.059	-0.030	0.032	0	0
$\langle \sigma_1^z \sigma_7^z \rangle$							0.030	-0.032	0	0

the Néel states really give a good clue to them.

Note that in this model (4) it is possible to construct a one-parameter family of ground states that breaks the translational symmetry of the problem. It might be possible to interpret the linearity of the  $\omega - k$  relationship (as  $\omega \rightarrow 0$ ) in terms of the so-called Goldstone bosons.<sup>15</sup> It ought to be pointed out that the discussion of the Goldstone theorem for antiferromagnetic models is extremely poor and incomplete. Anderson<sup>5</sup> first discussed the broken symmetry of antiferromagnets; since he invoked anisotropy fields in the course of his argument that discussion applied strictly to the strongly anisotropic or the Ising limit. Later Anderson<sup>16</sup> has given a physical discussion of the broken symmetry of the Heisenberg Hamiltonian. A rigorous mathematical discussion, such as exists for ferromagnets,<sup>15</sup> has never been given. The linear chain (1) also shows an asymptotic degeneracy (in the  $N \rightarrow \infty$  limit) in the ground state, and the symmetry properties of the Hamiltonian have been the subject of controversy in the literature.<sup>17</sup>

### III. GREEN'S-FUNCTION EQUATIONS AND DECOUPLING

We shall consider only the zero temperature theory. We define the transverse Green's function<sup>18</sup> as usual.

$$G^{-+}(i, j; t) = -i\Theta(t)\langle 0 | [S_i^-(t), S_j^+(0)] | 0 \rangle. \quad (10)$$

$\Theta(t)$  is 1 if  $t > 0$  and zero otherwise. Zubarev<sup>19</sup> has introduced a convenient notation

$$\langle\langle A; B \rangle\rangle = -i\Theta(t)\langle 0 | [A(t), B(0)] | 0 \rangle; \quad (11)$$

so

$$G^{-+}(i, j; t) = \langle\langle S_i^-; S_j^+ \rangle\rangle. \quad (12)$$

We also need the longitudinal Green's function

$$G^{zz}(i, j; t) = -i\Theta(t)\langle 0 | [S_i^z(t), S_j^z(0)] | 0 \rangle = \langle\langle S_i^z; S_j^z \rangle\rangle. \quad (13)$$

As far as we know, this has not been used earlier. The ferromagnetic ground state is an eigenfunction of every  $S_i^z$  operator, and the equation of motion for  $G^{zz}$  is trivial. If one starts from the two-sublattice picture of the antiferromagnet, the Néel states are again eigenstates of the site spin operators  $S_i^z$ , so that there also this Green's function  $G^{zz}$  is trivial. For the actual  $S=0$  ground state, however, the operator  $S_i^z$  will produce spin-1 excitations, and the longitudinal Green's function  $G^{zz}$  becomes relevant and it is important to treat its equation of motion on the same footing as that of  $G^{-+}$ . Now

$$i \frac{\partial}{\partial t} G^{-+}(i, j; t) = \delta(t)\langle 0 | [S_i^-, S_j^+] | 0 \rangle + \langle\langle [S_i^-, H]; S_j^+ \rangle\rangle. \quad (14)$$

The inhomogeneous term can be written

$$\langle 0 | [S_i^-, S_j^+] | 0 \rangle = -2\delta_{ij}\langle 0 | S_i^z | 0 \rangle. \quad (15)$$

If one uses the two-sublattice picture one replaces  $\langle 0 | S_i^z | 0 \rangle$  by  $S$  for one sublattice and  $-S$  for the other. On the other hand, for the exact ground state, this expectation value is strictly zero. Thus the equation of motion with the first-order time derivative has no inhomogeneous term

$$i \frac{\partial}{\partial t} G^{-+}(i, j; t) = \langle\langle [S_i^-, H]; S_j^+ \rangle\rangle. \quad (16)$$

To get an inhomogeneous term on the right, we consider the second time derivative

$$-\frac{\partial^2}{\partial t^2} G^{-+}(i, j; t) = \delta(t)\langle 0 | [[S_i^-, H], S_j^+] | 0 \rangle + \langle\langle [[S_i^-, H], H]; S_j^+ \rangle\rangle. \quad (17)$$

That the second commutator plays a role in determining the excitation energy was recognized earlier,<sup>11</sup> and is now given a natural explanation in this setup.

For the longitudinal case,

$$i \frac{\partial}{\partial t} G^{zz}(i, j; t) = \delta(t) \langle 0 | [S_i^z, S_j^z] | 0 \rangle + \langle \langle [S_i^z, H]; S_j^z \rangle \rangle. \quad (18)$$

The inhomogeneous term now disappears identically and we go on to the second time derivative:

$$-\frac{\partial^2}{\partial t^2} G^{zz}(i, j; t) = \delta(t) \langle 0 | [[S_i^z, H], S_j^z] | 0 \rangle + \langle \langle [[S_i^z, H], H]; S_j^z \rangle \rangle. \quad (19)$$

The inhomogeneous terms can be simplified by using the notation for the correlation functions

$$K^{\alpha\beta}(i, j) = \langle 0 | S_i^\alpha S_j^\beta | 0 \rangle. \quad (20)$$

Translational invariance would imply that

$$K^{\alpha\beta}(i, j) = K^{\alpha\beta}(\vec{r}_i - \vec{r}_j). \quad (21)$$

We have

$$\begin{aligned} & \langle 0 | [[S_i^-, H], S_j^+] | 0 \rangle \\ &= 2J_{ij} [K^{-+}(i, j) + 2K^{zz}(i, j)] \\ & \quad - 2\delta_{ij} \sum_l J_{il} [K^{-+}(i, l) + 2K^{zz}(i, l)] \end{aligned} \quad (22)$$

and

$$\begin{aligned} & \langle 0 | [[S_i^z, H], S_j^z] | 0 \rangle \\ &= 2J_{ij} K^{-+}(i, j) - 2\delta_{ij} \sum_l J_{il} K^{-+}(i, l). \end{aligned} \quad (23)$$

The full equations of motion can be written

$$\begin{aligned} -\frac{\partial^2}{\partial t^2} G^{-+}(i, j; t) &= \delta(t) \langle 0 | [[S_i^-, H], S_j^+] | 0 \rangle + 4 \sum_{\substack{l, k \\ l \neq k}} J_{il} J_{lk} \langle \langle S_i^- S_k^z S_l^z - S_i^z S_k^z S_l^-; S_j^+ \rangle \rangle + \sum_l J_{il}^2 \langle \langle S_i^-; S_j^+ \rangle \rangle \\ & \quad + 2 \sum_{\substack{l, k \\ k \neq l}} J_{il} J_{lk} \langle \langle S_i^- S_l^+ S_k^- - S_i^- S_l^- S_k^+; S_j^+ \rangle \rangle - \sum_l J_{il}^2 \langle \langle S_i^-; S_j^+ \rangle \rangle - 2 \sum_{\substack{l, k \\ l \neq k}} J_{il} J_{lk} \langle \langle S_i^+ S_k^- S_l^- - S_i^+ S_k^+ S_l^-; S_j^+ \rangle \rangle \\ & \quad + \sum_l J_{il}^2 \langle \langle S_i^-; S_j^+ \rangle \rangle - 4 \sum_{\substack{l, k \\ k \neq l}} J_{il} J_{lk} \langle \langle S_i^z S_k^z S_l^- - S_i^z S_k^z S_l^+; S_j^+ \rangle \rangle - \sum_l J_{il}^2 \langle \langle S_i^-; S_j^+ \rangle \rangle \end{aligned} \quad (24)$$

and

$$\begin{aligned} -\frac{\partial^2}{\partial t^2} G^{zz}(i, j; t) &= \delta(t) \langle 0 | [[S_i^z, H], S_j^z] | 0 \rangle - 2 \sum_{\substack{l, k \\ l \neq k}} J_{il} J_{lk} \langle \langle S_i^+ S_k^z S_l^- - S_i^z S_k^+ S_l^-; S_j^z \rangle \rangle \\ & \quad + 2 \sum_{\substack{l, k \\ k \neq l}} J_{il} J_{lk} \langle \langle S_i^+ S_l^- S_k^z - S_i^+ S_l^z S_k^-; S_j^z \rangle \rangle - 2 \sum_{\substack{l, k \\ l \neq k}} J_{il} J_{lk} \langle \langle S_i^- S_k^z S_l^+ - S_i^- S_k^- S_l^+; S_j^z \rangle \rangle \\ & \quad + 2 \sum_{\substack{l, k \\ k \neq l}} J_{il} J_{lk} \langle \langle S_i^- S_l^+ S_k^z - S_i^- S_l^z S_k^+; S_j^z \rangle \rangle + 2 \sum_l J_{il}^2 \langle \langle S_i^z; S_j^z \rangle \rangle - 2 \sum_l J_{il}^2 \langle \langle S_i^z; S_j^z \rangle \rangle. \end{aligned} \quad (25)$$

These equations start the hierarchy of the Green's function equations. We shall do a very simple decoupling of the hierarchy with the help of the correlation functions  $K^{\alpha\beta}$ , for example,

$$\langle \langle S_i^- S_k^z S_l^z; S_j^+ \rangle \rangle \approx K^{zz}(k, l) \langle \langle S_i^-; S_j^+ \rangle \rangle. \quad (26)$$

When two indices are equal, we use the identities for spin- $\frac{1}{2}$  operators.

$$(S_i^z)^2 = \frac{1}{4}, \quad S_i^- S_i^z = \frac{1}{2} S_i^-, \quad S_i^+ S_i^z = -\frac{1}{2} S_i^+, \quad S_i^+ S_i^- = S_i^z + \frac{1}{2}, \quad S_i^- S_i^+ = -S_i^z + \frac{1}{2}. \quad (27)$$

After decoupling we obtain the equations

$$\begin{aligned}
-\frac{\partial^2}{\partial t^2} G^{-+}(i, j; t) &= 2J_{ij}[K^{-+}(i, j) + 2K^{zz}(i, j)] - 2\delta_{ij} \sum_l J_{il}[K^{-+}(i, l) + 2K^{zz}(i, l)] \\
&+ 2 \sum_{l, k} J_{il} J_{lk} \{G^{-+}(i, j; t)[K^{-+}(l, k) + 2K^{zz}(k, l)] - G^{-+}(k, j; t)[K^{-+}(l, i) + 2K^{zz}(i, l)]\} \\
&+ 2 \sum_{l, k} J_{il} J_{lk} \{G^{-+}(k, j; t)[K^{-+}(i, l) + 2K^{zz}(i, l)] - G^{-+}(l, j; t)[K^{-+}(k, i) + 2K^{zz}(i, k)]\} \\
&- 2 \sum_l J_{il}^2 [G^{-+}(i, j; t) - G^{-+}(l, j; t)][K^{-+}(i, l) + 2K^{zz}(i, l)] \\
&+ 2 \sum_l J_{il}^2 G^{-+}(i, j; t)[1 - K^{-+}(l, l) - 2K^{zz}(l, l)] \\
&- 2 \sum_l J_{il}^2 G^{-+}(l, j; t)[1 - K^{-+}(i, i) - 2K^{zz}(i, i)]
\end{aligned} \tag{28}$$

and

$$\begin{aligned}
-\frac{\partial^2}{\partial t^2} G^{zz}(i, j; t) &= 2J_{ij}K^{-+}(i, j) - 2\delta_{ij} \sum_l J_{il}K^{-+}(i, l) \\
&- 4 \sum_{l, k} J_{il} J_{lk} [G^{zz}(k, j; t)K^{-+}(i, l) - G^{zz}(i, j; t)K^{-+}(l, k)] \\
&+ 4 \sum_{l, k} J_{il} J_{lk} [G^{zz}(k, j; t)K^{-+}(l, i) - G^{zz}(l, j; t)K^{-+}(k, i)] \\
&+ 4 \sum_l J_{il}^2 [K^{-+}(i, l)G^{zz}(l, j; t) - K^{-+}(l, i)G^{zz}(i, j; t)] \\
&+ 2 \sum_l J_{il}^2 G^{zz}(i, j; t)[1 - 2K^{-+}(l, l)] - 2 \sum_l J_{il}^2 G^{zz}(l, j; t)[1 - 2K^{-+}(i, i)].
\end{aligned} \tag{29}$$

Note that the equations of motion (24) and (26) as well as the decoupling procedure are completely general, and do not depend on the one-dimensional nature of the system. We now take the Fourier transform

$$G(i, j; t) = \int d\omega e^{-i\omega t} G(i, j; \omega) = \int d\omega e^{-i\omega t} \frac{1}{N} \sum_{\vec{k}} G(\vec{k}, \omega) \exp[i\vec{k} \cdot (\vec{r}_i - \vec{r}_j)]. \tag{30}$$

Then we get

$$G^{-+}(\vec{k}, \omega) = \frac{A_T(\vec{k})}{2\pi(\omega^2 - E_T^2)}, \quad G^{zz}(\vec{k}, \omega) = \frac{A_L(\vec{k})}{2\pi(\omega^2 - E_L^2)}, \tag{31}$$

with

$$A_T(\vec{k}) = \frac{2}{N} \sum_{\vec{q}} [J(\vec{k} - \vec{q}) - J(\vec{q})][K^{-+}(\vec{q}) + 2K^{zz}(\vec{q})], \tag{32}$$

$$\begin{aligned}
E_T^2(\vec{k}) &= \frac{2}{N} \sum_{\vec{q}} [J(\vec{k}) - J(\vec{q})][J(\vec{k} - \vec{q}) - J(\vec{q})][K^{-+}(\vec{q}) + 2K^{zz}(\vec{q})] \\
&+ \frac{2}{N^2} \sum_{\vec{q}, \vec{q}'} J(\vec{q}') [J(\vec{k} - \vec{q} - \vec{q}') - J(-\vec{q} - \vec{q}')] [K^{-+}(\vec{q}) + 2K^{zz}(\vec{q})] \\
&+ \left(1 - \frac{1}{N} \sum_{\vec{q}} K^{-+}(\vec{q}) - \frac{2}{N} \sum_{\vec{q}} K^{zz}(\vec{q})\right) \frac{2}{N} \sum_{\vec{q}} J(\vec{q}) [J(-\vec{q}) - J(\vec{k} - \vec{q})]
\end{aligned} \tag{33}$$

and

$$A_L(\vec{k}) = \frac{2}{N} \sum_{\vec{q}} [J(\vec{k} - \vec{q}) - J(\vec{q})] K^{-+}(\vec{q}), \quad (34)$$

$$E_L^2(\vec{k}) = \frac{4}{N} \sum_{\vec{q}} [J(\vec{k}) - J(\vec{q})][J(\vec{k} - \vec{q}) - J(\vec{q})] K^{-+}(\vec{q}) + \frac{4}{N^2} \sum_{\vec{q}, \vec{q}'} J(\vec{q}') [J(\vec{k} - \vec{q} - \vec{q}') - J(-\vec{q} - \vec{q}')] K^{-+}(\vec{q}) \\ + \frac{2}{N} \left( 1 - \frac{2}{N} \sum_{\vec{q}} K^{-+}(\vec{q}) \right) \sum_{\vec{q}} J(\vec{q}) [J(-\vec{q}) - J(\vec{k} - \vec{q})]. \quad (35)$$

Here

$$J(\vec{q}) = \frac{1}{N} \sum_{i,j} J_{ij} \exp[-i\vec{q} \cdot (\vec{r}_i - \vec{r}_j)], \quad (36)$$

$$K(\vec{q}) = \frac{1}{N} \sum_{i,j} K(i,j) \exp[-i\vec{q} \cdot (\vec{r}_i - \vec{r}_j)]. \quad (37)$$

We have  $J(\vec{q}) = J(-\vec{q})$ ,  $K(\vec{q}) = K(-\vec{q})$  from the symmetry  $J_{ij} = J_{ji}$ ,  $K(i,j) = K(j,i)$ . The energy eigenvalues  $E_T$  and  $E_L$  are expressed in terms of the correlation functions  $K^{-+}$  and  $K^{zz}$ .

But the correlation functions can be expressed in terms of the Green's functions. For two operators  $A$  and  $B$ , we get<sup>19</sup>

$$-\frac{1}{2} \langle 0 | AB | 0 \rangle = \int_0^\infty d\omega \operatorname{Im} G_{AB}(\omega). \quad (38)$$

Hence we can write

$$K^{-+}(i,j) = \langle 0 | S_i^- S_j^+ | 0 \rangle = -2 \int_0^\infty d\omega \operatorname{Im} G^{-+}(i,j;\omega), \quad (39)$$

$$K^{zz}(i,j) = \langle 0 | S_i^z S_j^z | 0 \rangle = -2 \int_0^\infty d\omega \operatorname{Im} G^{zz}(i,j;\omega). \quad (40)$$

From the expression of  $G^{-+}$  and  $G^{zz}$ , we get

$$K^{-+}(\vec{k}) = \frac{1}{2} A_T / E_T, \quad K^{zz}(\vec{k}) = \frac{1}{2} A_L / E_L. \quad (41)$$

Therefore, the equations for the correlation functions are

$$K^{-+}(\vec{k}) = \frac{1}{2E_T} \frac{2}{N} \sum_{\vec{q}} [J(\vec{k} - \vec{q}) - J(\vec{q})] \\ \times [K^{-+}(\vec{q}) + 2K^{zz}(\vec{q})], \quad (42)$$

$$K^{zz}(\vec{k}) = \frac{1}{2E_L} \frac{2}{N} \sum_{\vec{q}} [J(\vec{k} - \vec{q}) - J(\vec{q})] K^{-+}(\vec{q}). \quad (43)$$

We thus obtain four equations (33), (35), (42), and (43) for determining four quantities  $E_T$ ,  $E_L$ ,  $K^{-+}$ , and  $K^{zz}$ .

For the isotropic Hamiltonian, with a rotationally

invariant ground state, we have a further simplification

$$K^{-+}(\vec{q}) = 2K^{zz}(\vec{q}). \quad (44)$$

It follows from (33) and (35) that

$$E_T^2 = E_L^2. \quad (45)$$

Thus we have a triply degenerate excitation spectrum,  $E_T$  being doubly degenerate. We then have two quantities to determine from two equations. Dropping suffixes, we call these  $K$  and  $E$ .

#### IV. SOLUTION FOR THE LINEAR CHAIN

The equations to be solved for the linear chain are

$$K(k) = \frac{1}{2E(k)} \frac{4}{N} \sum_q [J(k-q) - J(q)] K(q) \quad (46)$$

and

$$E^2(k) = \frac{4}{N} \sum_q [J(k) - J(q)] [J(k-q) - J(q)] K(q) \\ + \frac{4}{N^2} \sum_{q,q'} J(q' - q) [J(k - q') - J(q')] K(q) \\ + \left( 1 - \frac{2}{N} \sum_q K(q) \right) \frac{2}{N} \sum_q J(q) [J(q) - J(k - q)]. \quad (47)$$

With only nearest-neighbor interaction, we get

$$J(q) = 2J \cos q. \quad (48)$$

Recalling that  $K(q)$  is an even function of  $q$ , we get the equation for  $K(k)$  as

$$K(k) = \frac{1}{\pi E} \int_{-\pi}^{\pi} 2J [\cos(k-q) - \cos q] K(q) dq. \quad (49)$$

Hence

$$K(k) = 4J(1 - \cos k) I_1 / E, \quad (50)$$

with

$$I_1 = -\frac{1}{2\pi} \int_{-\pi}^{\pi} \cos q K(q) dq. \quad (51)$$

Define

$$I_0 = \frac{1}{N} \sum_q K(q) = \frac{1}{2\pi} \int_{-\pi}^{\pi} K(q) dq. \quad (52)$$

The expression for the excitation energy is from (47), (48), and (50)–(52):

$$E^2(k) = 16J^2(1 - \cos k) \times \left(\frac{1}{4} - \frac{1}{2}I_0 + \frac{1}{2}I_1 + I_2 + I_1 \cos k\right), \quad (53)$$

with

$$I_2 = \frac{1}{2\pi} \int_{-\pi}^{\pi} \cos^2 q K(q) dq. \quad (54)$$

Hence, we get

$$K(k) = \frac{(1 - \cos k)^{1/2} I_1}{\left(\frac{1}{4} - \frac{1}{2}I_0 + \frac{1}{2}I_1 + I_2 + I_1 \cos k\right)^{1/2}}, \quad (55)$$

and the three integrals  $I_0$ ,  $I_1$ , and  $I_2$  can be expressed in terms of the parameter

$$\alpha = (1/2I_1)\left(\frac{1}{4} - \frac{1}{2}I_0 - \frac{1}{2}I_1 + I_2\right). \quad (56)$$

From (52), (51), (54), (55), and (56), we obtain

$$I_0 = (2/\pi)I_1^{1/2} \ln\left\{[1 + (\alpha + 1)^{1/2}]/\alpha^{1/2}\right\}, \quad (57)$$

$$I_1 = -(2/\pi)I_1^{1/2}\left\{(\alpha + 1)^{1/2} - (\alpha + 1) \ln\left\{[1 + (\alpha + 1)^{1/2}]/\alpha^{1/2}\right\}\right\}, \quad (58)$$

$$I_2 = (2/\pi)I_1^{1/2}\left\{-\alpha(\alpha + 1)^{1/2} + \left(\frac{3}{2}\alpha + 1\right) \ln\left\{[1 + (\alpha + 1)^{1/2}]/\alpha^{1/2}\right\}\right\}. \quad (59)$$

Combining with the definition of  $\alpha$ , Eq. (56), we get the equation of self-consistency for  $\alpha$  as

$$f(\alpha) = (\alpha + 1)\left\{\alpha(\alpha + 1)^{1/2} \ln\left\{[1 + (\alpha + 1)^{1/2}]/\alpha^{1/2}\right\} - (\alpha + 1)\right\} \times \left\{(\alpha + 1)^{1/2} \ln\left\{[1 + (\alpha + 1)^{1/2}]/\alpha^{1/2}\right\} - 1\right\} - \frac{1}{8}\pi^2 \equiv 0. \quad (60)$$

The solution of this equation — there is only one — gives (Fig. 1)

$$\alpha = 0.0798. \quad (61)$$

The integrals at this value of  $\alpha$  are

$$I_0 = 0.8765, \quad I_1 = 0.4856, \quad I_2 = 0.5089. \quad (62)$$

Hence, we have the excitation spectrum:

$$E(k) = 4J(1 - \cos k)^{1/2}(0.5634 + 0.4856 \cos k)^{1/2}. \quad (68)$$

Figure 2 shows the excitation spectrum which compares well with the des Cloizeaux–Pearson spec-

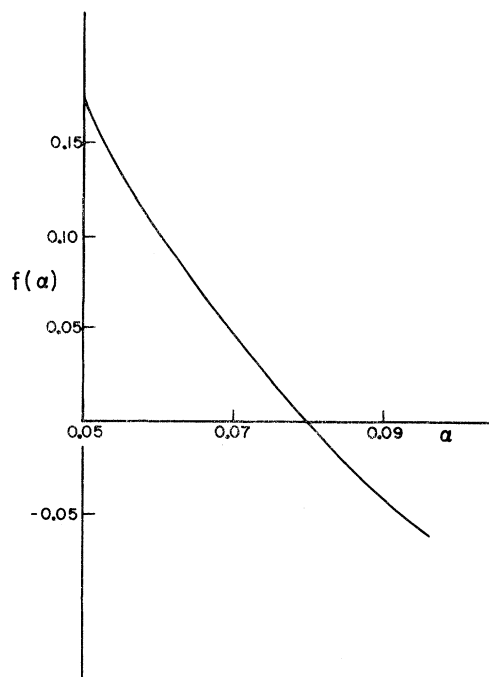


FIG. 1. Solution of Eq. (60) for  $\alpha$ .

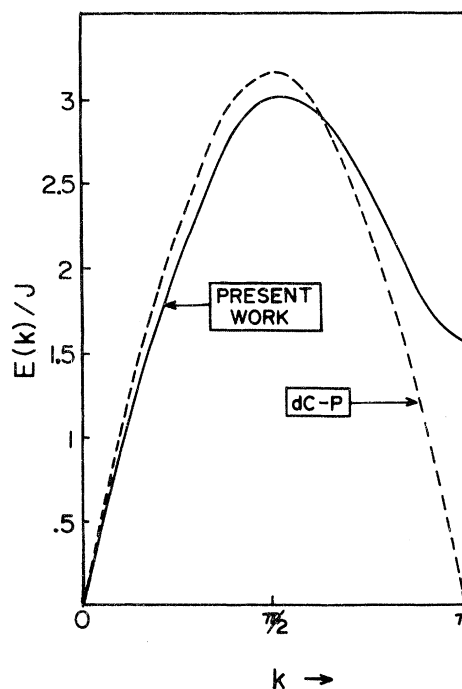


FIG. 2. Excitation spectrum compared with the exact des Cloizeaux–Pearson (dC–P) spectrum.

trum except near  $k = \pi$ . As  $k \rightarrow 0$ ,

$$E(k) \approx 2.9J|k|. \quad (64)$$

The Fourier transform of the spin correlation function becomes

$$K(k) = 0.4856(1 - \cos k)^{1/2} / (0.5634 + 0.4856 \cos k)^{1/2}. \quad (65)$$

This quantity is zero at  $k=0$  (which, in fact, is an exact result), and has a broad maximum at  $k=\pi$ . The spin correlation function  $K(i, j)$  therefore does not have a true long-range order. Its values can be calculated at several points (Fig. 3) and it shows the expected oscillating behavior.

The ground-state energy  $E_0$  can be expressed in terms of the correlation function

$$\begin{aligned} E_0 &= \sum_{i,j} J_{ij} [K^{zz}(i, j) + K^{-+}(i, j)] \\ &= 3NJK(|r_i - r_j| = 1) \\ &= -3NJI_1 = -1.4568NJ. \end{aligned} \quad (66)$$

As happens very often with truncation procedures of Green's functions, the ground-state energy is lower than the true energy. Thus, truncation does not have the good features of a variation calculation.

Of considerable interest to the neutron-scattering studies is expression (65) of  $K(k)$ . Endoh *et al.*<sup>1</sup> quote and use the classical result

$$K(q) = (1 - \cos q) / \sin q. \quad (67)$$

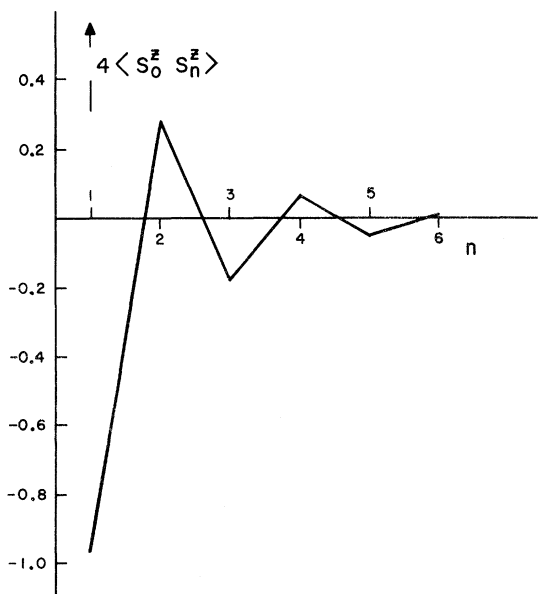


FIG. 3. Spin correlation function  $K(i, j)$ .

Normalized to the intensity at  $k = \frac{1}{2}\pi$ , the intensity of scattering is

$$I(k)/I(\frac{1}{2}\pi) = (1.16)^{1/2}(1 - \cos k)^{1/2} / (1.16 + \cos k)^{1/2}. \quad (68)$$

Figure 4 shows a plot of the above expression as well as that of (67). Our curve lies lower than the classical curve for large  $k$ , as do the experimental points in Ref. 1, but a direct comparison with experimental points requires form factor corrections. Very close to  $k \approx \pi$ , our approximation obviously will not work.

## V. CONCLUSION

The self-consistent solution of our equation gives a fair description of the properties of the Heisenberg linear chain. Of course, there are some obvious defects. The quantity  $I_0$  ought to be 0.5 exactly, but comes out to be higher 0.8765. One can get a constrained solution, insisting on  $I_0$  to be 0.5. This gives  $\alpha = 0.18$ ,  $E(k) \sim 2.3J|k|$  as  $k \rightarrow 0$ , and  $E_0 = -0.765NJ$ . This is still a fair description, so that the violation of self-consistency [Eq. (60)] is not serious.

We can also make contact with Anderson's spin-wave spectrum.<sup>5</sup> If we consider  $K^{-+} = 0$ ,  $E_L = 0$ , but let the transform of the longitudinal spin correlation function be sharply peaked

$$K^{zz}(q) = \frac{1}{4}N\delta_{q,\pi}, \quad (69)$$

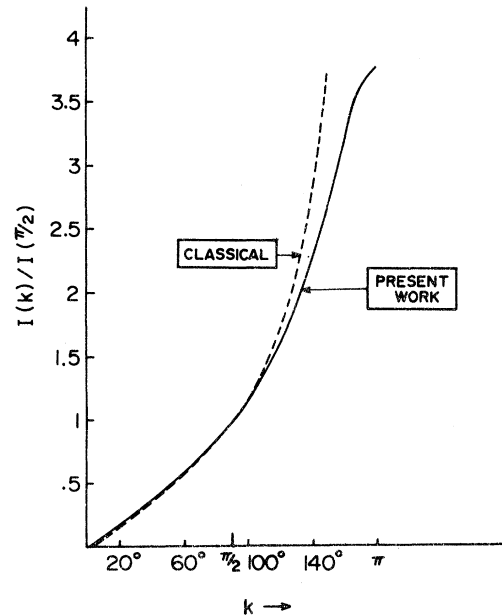


FIG. 4. Normalized scattering intensity of Eq. (68).



we get  $\hbar^2\omega^2 = 4J^2 \sin^2 k$ , provided we ignore the last two terms in Eq. (47) for  $E_T$ . This shows why Anderson did not get the degeneracy correctly. He had ignored the longitudinal part of the spectrum.<sup>20</sup>

We have noted that the decoupling scheme does not specifically depend on the one dimensionality. This, however, is no guarantee that a self-consistent or even a constrained solution can be found in the three-dimensional case. That some new features might be expected can be seen as follows. Our  $K(k)$  has only a broad maximum but no singularity. It follows that there is no long-range order. In order that  $K(i, j)$  be finite, oscillating in sign,

as the separation  $|\vec{r}_i - \vec{r}_j|$  becomes large, we must have a singularity in  $K(\vec{q})$  or  $K(\vec{q})$  must be a generalized function<sup>21</sup> [cf. Eq. (69)]. Unfortunately, the simple decoupling procedure used in this paper does not harbor any hope of generating a singularity. In three dimensions some long-range order is expected; or, if Anderson's argument<sup>16</sup> is followed, one must incorporate the broken symmetry in the correlation functions. So the decoupling scheme would require reexamination and possibly modification before we can develop a theory of the Heisenberg antiferromagnet in three dimensions.

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