

Alternating Linear Heisenberg Antiferromagnet: The Exciton Limit*

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The one-dimensional alternating antiferromagnet with $\mathcal{H} = 2J \sum_n \vec{S}_{2n} \cdot \vec{S}_{2n-1} + 2J' \sum_n \vec{S}_{2n} \cdot \vec{S}_{2n+1}$ is studied for $J' \ll J$. For $\beta^{-1} \equiv k_B T \ll J$ the susceptibility is expanded in powers of the exciton density as $\chi T \propto A(\beta J', J'/J) e^{-2\beta J} + B(\beta J', J'/J) e^{-4\beta J} + \dots$ and the coefficients A and B are calculated for $J'/J \rightarrow 0$. The calculation of $B(\beta J', 0)$ required the evaluation of the two-exciton scattering matrix. The interactions between excitons which affect the susceptibility are found to be repulsive. As a result, the coefficient B is correctly predicted by the usual assumption that excitons obey localized statistics. A general discussion relating statistics to the on-shell forward-scattering t matrix enables one to understand the difference between the statistical properties of spin waves and excitons. For opposite-spin excitons an attractive bound state is found to exist for all values of total momentum. Perturbation theory in J'/J is used to calculate the single-exciton dispersion relation at zero temperature as $E(k) = (2J + 5J'^3/32J^2) - (J' + J'^2/2J - 5J'^3/32J) \cos k - (J'^2/4J + J'^3/8J^2) \cos^2 k - (J'^3/8J^2) \cos^3 k$.

I. INTRODUCTION

It is well known that almost all three-dimensional models are intractable. Although a number of two-dimensional models have been solved exactly,¹ most of these are related to the Ising model, which has a discrete eigenvalue spectrum. On the other hand, even rather complicated one-dimensional systems have been treated exactly.² Perhaps the most important such model is the one-dimensional Heisenberg antiferromagnet, for which several ground-state properties have been calculated.³ Another example closely related to the present work is the description of the two-spin-wave states in a ferromagnet.⁴ For the ferromagnet, the three-dimensional case is rather complicated, whereas the one-dimensional case can be treated in terms of simple functions. The situation is similar with regard to the two-exciton states and, as we shall see, simple explicit results can be obtained in one dimension.

In the past, theoretical interest in one-dimensional systems was confined to workers in statistical mechanics, since it was hard to find real systems which conformed to such theoretical models. However, recently a number of one-dimensional systems have been studied experimentally,⁵⁻⁷ and this activity has stimulated theorists to consider more complicated models. In view of the recent interest in the tetracyanoquinodimethan salts⁷ we have treated the alternating Heisenberg antiferromagnet in one dimension. This model differs from the usual Heisenberg antiferromagnet in that the exchange integrals alternate periodically between two values J and J' . Thus the Hamiltonian for this system may be written as

$$\mathcal{H} = 2J \sum_n \vec{S}_{2n} \cdot \vec{S}_{2n-1} + 2J' \sum_n \vec{S}_{2n} \cdot \vec{S}_{2n+1}. \quad (1.1)$$

This model has been studied previously by many authors,⁸⁻¹² and the results of noninteracting exciton theory have been fully investigated. Here we shall study the extent to which such results are modified by interactions between excitons.

In the present paper we shall be concerned with this model in the "exciton limit," in which $J \gg J'$. In this limit the ground state consists essentially of pairs of spins each of which forms a state of total spin zero due to the antiferromagnetic exchange interaction J . Localized excitations in which one or more pairs have total spin one are broadened into exciton bands by the J' exchange interactions. The objectives of the present paper are, first, to calculate the single-exciton dispersion relation to as high order in (J'/J) as is convenient and, second, to calculate the second virial coefficient for the susceptibility of the exciton gas. Whereas properties may be calculated correctly to the first order in the exciton density using simple noninteracting-exciton theory, the second-order terms depend on exciton-exciton interactions, and these effects are conveniently described by the second virial coefficient.¹³ The properties of this model in the "regular limit," i.e., when $|J - J'| \ll J$, will be discussed in a future publication.

This paper is organized as follows. In Sec. II we give a general discussion of the structure of perturbation theory and of the type of results one can expect for the model. Here we discuss the construction of an effective Hamiltonian $\mathcal{H}_{\text{eff}}^{(n)}$ for the manifold of states containing n excitons. In general $\mathcal{H}_{\text{eff}}^{(n)}$ is given by an infinite series in the parameter (J'/J) . In Sec. III we study the scattering of two excitons using $\mathcal{H}_{\text{eff}}^{(2)}$ to lowest order in (J'/J) . Here we include in addition to the terms given in Eq. (1.1) also those describing the interaction with an external magnetic field and we introduce anisotropy by replacing J by an anisotropic exchange

integral. It is shown that for the total spin m of the two colliding excitons equal to 2 or 1, the exciton-exciton interactions are repulsive, whereas for $m=0$ they are mainly attractive. In all cases, since we work in one dimension, these interactions give rise to bound states of two excitons. In Sec. IV the results of the preceding section are specialized to the isotropic model, and the zero-field susceptibility is calculated as a power series in the exciton density. The term which is second order in the density shows that the combined effects of dynamic and kinematic interactions do not differ markedly from what one would find using a hybrid picture in which the excitons obey a noninteracting-exciton dispersion relation, but also obey statistics of the localized type rather than of the Fermi or Bose type. Here we derive a general condition for the validity of localized statistics in terms of quasiparticle interactions. In Sec. V we present results for $\mathcal{H}_{\text{eff}}^{(1)}$, the effective Hamiltonian for the single-exciton manifold, including effects up to third order in perturbation theory in (J'/J) . The resulting dispersion relation and bandwidth coincide with the usual noninteracting result⁹ only to first order in (J'/J) . These calculations can be used to calculate the first virial coefficient for the susceptibility for $J'/J < \frac{1}{2}$. Finally, our results and conclusions are summarized in Sec. VI.

In Appendix A we use Bethe's ansatz¹⁴ to obtain the eigenvalue condition for two parallel spin excitons, and show that the treatment of Ref. 8 is in error. Appendix B contains the details of the scattering calculation for antiparallel spins. In Appendix C a technique suggested by Bloomfield¹⁵ for analyzing singular integrals is used to simplify the expressions for the two-exciton scattering matrix. Finally, Appendix D contains the details of the calculation of $\mathcal{H}_{\text{eff}}^{(1)}$ discussed in Sec. V.

II. QUALITATIVE CONSIDERATIONS

The purpose of this section is to give a general qualitative discussion of the properties of the model of Eq. (1.1). For $J'=0$, it is clear that the system consists of independent pairs of spins antiferromagnetically coupled, so that for each pair a total-spin-zero singlet ground state lies at an energy $2J$ below the total-spin-one triplet excited state. The susceptibility χ is then given by

$$\chi T/NC = 3\rho/(1+3\rho), \quad (2.1)$$

where C is the Curie constant: $C = 2g^2\mu_B^2/3k_B$, where $g\mu_B/\hbar$ is the gyromagnetic ratio of the spins, and also in Eq. (2.1) $\rho = e^{-2\beta J}$, where $\beta = (k_B T)^{-1}$. Thus at low temperatures, $\rho \ll 1$ and then ρ is approximately proportional to the density of excitons. For $\rho \ll 1$, i. e., at low exciton density, one may write

$$\chi T/NC = 3\rho - 9\rho^2 + \dots \quad (2.2)$$

To proceed further, let us introduce the simplest possible approximation, namely, we neglect terms in J' which do not conserve the number of excitons. Within this approximation the number of excitons is a good quantum number, and the energy levels of the system can be written

$$E(n; \lambda) = E_0 + 2Jn + J'\varphi(n, \lambda), \quad (2.3)$$

where E_0 is the ground-state energy, λ is a quantum number labeling the particular n -exciton state, and $\varphi(n, \lambda)$ is a dimensionless energy obtained from $\mathcal{H}_{\text{eff}}^{(n)}$. In this case $\mathcal{H}_{\text{eff}}^{(n)}$ consists of the secular terms involving J' . The neglected terms in $\mathcal{H}_{\text{eff}}^{(n)}$ which do not conserve the number of excitons will modify $\mathcal{H}_{\text{eff}}^{(n)}$ by the addition of terms of relative order $(J'/J)^r$, with $r \geq 2$. For $n=1$, the well-known result is that exciton eigenstates are labeled by a wave vector and a spin-projection quantum number, and that in zero field one has^{8,9}

$$E(1; k, m) = E_0 + 2J - J' \cos ka, \quad (2.4)$$

where a is the separation between pairs. Henceforth we shall set $a=1$. Intuitively, one might argue that for $J' \neq 0$ Eq. (2.1) should be generalized to read

$$\frac{\chi T}{NC} = \frac{3\rho}{N} \sum_k \frac{e^{\beta J' \cos k}}{1 + 3\rho e^{\beta J' \cos k}}, \quad (2.5a)$$

so that when $\rho \ll 1$, one can write

$$\chi T/NC = 3\rho A(\beta J') - 9\rho^2 A(2\beta J') + \dots, \quad (2.5b)$$

where

$$A(\beta J') = N^{-1} \sum_k e^{\beta J' \cos k}. \quad (2.6)$$

In fact, Lynden-Bell and McConnell justify writing Eq. (2.5a) by arguing that excitons do obey localized statistics.⁸

By obeying localized statistics, we mean that the thermal occupation number $n_m(k)$ is given by

$$n_m(k) = (e^{\beta E(k)} + 3)^{-1}, \quad (2.7a)$$

$$n_m(k) = \frac{\rho e^{\beta J' \cos k}}{1 + 3\rho e^{\beta J' \cos k}}. \quad (2.7b)$$

This expression is to be contrasted to the corresponding results for bosons and fermions, which are

$$n_m(k)_B = (e^{\beta E(k)} - 1)^{-1}, \quad (2.8a)$$

$$n_m(k)_F = (e^{\beta E(k)} + 1)^{-1}, \quad (2.8b)$$

respectively. If triplet excitons are imagined to obey Bose or Fermi statistics, then the susceptibility is given by

$$\frac{\chi_B T}{NC} = \frac{3\rho}{N} \sum_k \frac{e^{\beta J' \cos k}}{(1 - \rho e^{\beta J' \cos k})^2}, \quad (2.9a)$$

$$\frac{\chi_F T}{NC} = \frac{3\rho}{N} \sum_k \frac{e^{\beta J' \cos k}}{(1 + \rho e^{\beta J' \cos k})^2}, \quad (2.9b)$$

in the respective cases. The corresponding low-density expansions are

$$\chi_B T/NC = 3\rho A(\beta J') + 6\rho^2 A(2\beta J') + \dots, \quad (2.10a)$$

$$\chi_F T/NC = 3\rho A(\beta J') - 6\rho^2 A(2\beta J') - \dots. \quad (2.10b)$$

From Eqs. (2.5b) and (2.10) it is clear that statistics affect the ρ^2 (and higher-order) terms in χ . The type of statistics quasiparticles obey can be thought of as arising from so-called "kinematic" interactions. When J' is nonzero, there are in addition "dynamic" interactions due to the fact that two excitons no longer propagate like two independent quasiparticles.

These considerations motivate our approach to this question, which is to calculate χ in the low-density limit as a power series in the density of excitons:

$$\chi T/NC = 3\rho A(\beta J') + \rho^2 B(\beta J') + \dots. \quad (2.11)$$

The result $B(\beta J') = -9A(2\beta J')$ would then be an indication that localized statistics are appropriate: see Eq. (2.5b). In contrast, the result $B(\beta J') = +$ (or $-$) $6A(2\beta J')$ would indicate that we should use Bose (or Fermi) statistics.

In order to evaluate the second virial coefficient $B(\beta J')$, it is necessary to treat the interactions of two excitons. This we do exactly insofar as the secular terms in J' are concerned. Presumably, inclusion of nonsecular terms would not qualitatively affect the results inasmuch as they lead to contributions to $\mathcal{H}_{\text{eff}}^{(2)}$ of order (J'/J) smaller than the secular terms.

According to conventional wisdom, the best approximation is to use localized statistics to describe triplet excitons. This is clearly correct at high temperature ($\beta J' \ll 1$), when incoherent delocalization becomes unimportant. For low temperatures this conclusion is based on rather superficial evidence. In particular this result is obtained in Ref. 8 by considering the problem of excitons interacting via a hard-core repulsion. However, it is quite obvious that such a calculation, even if it were correct, would not be definitive, because for the spin- $\frac{1}{2}$ Heisenberg ferromagnet spin waves are subject to the same hard-core repulsion when they attempt to visit the same site, and yet, as Dyson¹⁶ has shown for the three-dimensional case, long-wavelength spin waves interact quite weakly. It is this lack of interaction which validates classifying spin waves as bosons. In fact, as shown in Appendix A, the treatment of the hard-core exciton gas given in Ref. 8 is incorrect. Thus, while it is clear that localized statistics must be correct at high temperature, it is an open question as to which statistics are correct at low temperatures when only low-energy long-wavelength excitons are excited.

At low temperatures our result should depend decisively on the interactions between low-energy long-wavelength excitons. In fact, one can discuss this problem in a general way. For repulsive dynamical interactions, as is the case for parallel spin excitons, or for the case of weakly attractive interactions, localized statistics are found to be appropriate. For strongly attractive interactions, bound states of two excitations are formed; there is attraction in momentum space, and no independent quasiparticle statistics can describe the two-excitation manifold. Finally, when, as for spin waves, the attractive dynamical interactions are just strong enough to balance the effects of the hard-core interaction, no interference effects occur in momentum space and Bose statistics are appropriate.

These ideas are confirmed by our explicit numerical calculation of $B(\beta J')$ which shows that $B(\beta J')/A(2\beta J')$ tends to the value -9 in both the high- and low-temperature limits. At intermediate temperatures the ratio is found to be not very different from -9 , so that our conclusion is that excitons approximately obey localized statistics at all temperatures.

The second aim of this paper is to treat the effects which are higher order in (J'/J) . In this case, for simplicity, we study only the single-exciton manifold. This calculation is tedious, but straightforward. By successive canonical transformations we obtain all terms in $\mathcal{H}_{\text{eff}}^{(1)}$ up to relative order $(J'/J)^3$. We can compare our results with the version of linear-exciton theory which includes two-exciton creation and annihilation processes, which gives⁹

$$E(1; k, m) = E_0 + 2J[1 - (J'/J) \cos k]^{1/2}. \quad (2.12)$$

As one might expect, our result agrees with this formula only up to first order in (J'/J) .

Finally, one would like to know how rapidly the expansion in (J'/J) is converging. Linear-exciton theory, viz., Eq. (2.12), indicates convergence for $|J'/J| < 1$, and this property is undoubtedly equivalent to the statement that the exciton spectrum has a gap for $(J'/J) < 1$. For short chains, Brinkman¹⁷ has found that perturbation theory does converge for $|J'/J| < 1$. Thus it is suggested that the ratio of succeeding terms in the (J'/J) expansion is of order $1(J'/J)$, and hence that our results may be useful as long as (J'/J) is smaller than, say, $\frac{1}{2}$.

III. SCATTERING STATES OF TWO EXCITONS

A. Construction of the Boson Hamiltonian

The model we consider is described by the Hamiltonian,

$$\mathcal{H} = 2J \sum_n \vec{S}_{2n} \cdot \vec{S}_{2n-1} + D \sum_n (S_{2n}^z + S_{2n-1}^z)^2$$

$$-h \sum_n (S_{2n}^z + S_{2n-1}^z) + 2J' \sum_n \vec{S}_{2n} \cdot \vec{S}_{2n-1}. \quad (3.1)$$

For the case of $J' \ll J$ which we consider, one may visualize the Hamiltonian as describing a linear system of N unit cells, in each of which there are two spins with $S = \frac{1}{2}$. Spins in the same unit cell are strongly coupled via an antiferromagnetic Heisenberg exchange interaction scaled by an energy J , whereas exchange coupling between different unit cells is scaled by the much smaller energy J' . Dipolar interactions^{8,9} are included insofar as they are described by the second term, where D would then be of order $g^2 \mu_B^2 \nu_0^{-3}$, where ν_0 is the separation between spins in the same unit cell. More generally, D may be viewed as an anisotropic exchange constant. Finally, the third term in Eq. (3.1) is the energy of the spins in an external magnetic field.

To treat the low-density (of excitons) limit, we map the spin Hamiltonian of Eq. (3.1) into a boson Hamiltonian via the following transformation:

$$\sqrt{2} S_{2n}^+ = c_{1,n}^\dagger (1 - N_n) - (1 - N_n) c_{-1,n} + (c_{1,n}^\dagger c_{0,n} + c_{0,n}^\dagger c_{-1,n}), \quad (3.2a)$$

$$\sqrt{2} S_{2n-1}^+ = -c_{1,n}^\dagger (1 - N_n) + (1 - N_n) c_{-1,n} + (c_{1,n}^\dagger c_{0,n} + c_{0,n}^\dagger c_{-1,n}), \quad (3.2b)$$

$$S_{2n}^- = (S_{2n}^+)^{\dagger}, \quad (3.2c)$$

$$S_{2n-1}^- = (S_{2n-1}^+)^{\dagger}, \quad (3.2d)$$

$$2S_{2n}^z = -c_{0,n}^\dagger (1 - N_n) - (1 - N_n) c_{0,n} + (c_{1,n}^\dagger c_{1,n} - c_{-1,n}^\dagger c_{-1,n}), \quad (3.2e)$$

$$2S_{2n-1}^z = c_{0,n}^\dagger (1 - N_n) + (1 - N_n) c_{0,n} + (c_{1,n}^\dagger c_{1,n} - c_{-1,n}^\dagger c_{-1,n}), \quad (3.2f)$$

where $N_n = \sum_m c_{m,n}^\dagger c_{m,n}$. The operator $c_{m,n}^\dagger$ creates an ($S = 1$), ($S^z = m$) exciton in the n th unit cell. Although these substitutions reproduce the matrix elements of the spin operators in the states for

which no site has more than one exciton, they also give finite results when two or more excitons are present on a single site. To eliminate these spurious states, we shall add to the Hamiltonian a repulsive interaction of the form

$$\frac{1}{2} T \sum_{m, m', n} c_{m,n}^\dagger c_{m',n}^\dagger c_{m',n} c_{m,n} \quad (3.3)$$

and the limit $T \rightarrow \infty$ will be taken.

When the expressions of Eq. (3.2) are substituted into Eqs. (3.1) and (3.3), the result can be written as

$$\mathcal{H} = \mathcal{H}_0 + V, \quad (3.4)$$

where \mathcal{H}_0 is

$$\mathcal{H}_0 = \sum_{k,m} (2J + Dm^2 - hm - J' \cos k) c_{m,k}^\dagger c_{m,k}, \quad (3.5a)$$

$$\mathcal{H}_0 \equiv \sum_{k,m} E_m(k) c_{m,k}^\dagger c_{m,k}, \quad (3.5b)$$

where $c_{m,k}^\dagger$ creates an ($S^z = m$) exciton wave:

$$c_{m,k}^\dagger = N^{-1/2} \sum_n e^{ink} c_{m,n}^\dagger. \quad (3.6)$$

There are various terms in V . There are those which conserve the number of excitons, i. e., those which commute with \mathcal{H}_0 . In the usual nomenclature these are called secular terms, and they will be denoted V_{sec} . As mentioned in Sec. II, the nonsecular terms lead to effects which are higher order in (J'/J). The effects of these terms on the single-exciton manifold will be considered in Sec. V. Here we neglect such effects and set

$$\mathcal{H} = \mathcal{H}_0 + V_{\text{sec}}. \quad (3.7)$$

It may be seen that V_{sec} commutes with the total z component of spin. We can therefore classify terms in V_{sec} according to the total z component of spin of the colliding excitons:

$$V_{\text{sec}} = \sum_{m=-2}^{+2} V_{\text{sec}}^{(m)}. \quad (3.8)$$

Within the manifold of two-exciton states we find that

$$V_{\text{sec}}^{(2)} = \frac{1}{2} J' \sum_n (c_{1,n}^\dagger c_{1,n}^\dagger c_{1,n} c_{1,n+1} + c_{1,n}^\dagger c_{1,n+1}^\dagger c_{1,n+1} c_{1,n+1} + c_{1,n+1}^\dagger c_{1,n+1}^\dagger c_{1,n+1} c_{1,n} + c_{1,n+1}^\dagger c_{1,n}^\dagger c_{1,n} c_{1,n} + c_{1,n+1}^\dagger c_{1,n}^\dagger c_{1,n+1} c_{1,n}) + \frac{1}{2} T \sum_n c_{1,n}^\dagger c_{1,n}^\dagger c_{1,n} c_{1,n}, \quad (3.9a)$$

$$V_{\text{sec}}^{(1)} = \frac{1}{2} J' \sum_n (c_{1,n}^\dagger c_{0,n}^\dagger c_{0,n} c_{1,n+1} + c_{1,n}^\dagger c_{0,n+1}^\dagger c_{0,n+1} c_{1,n+1} + c_{1,n}^\dagger c_{0,n}^\dagger c_{0,n} c_{1,n+1} + c_{0,n}^\dagger c_{1,n+1}^\dagger c_{0,n+1} c_{1,n} + c_{1,n+1}^\dagger c_{0,n+1}^\dagger c_{0,n+1} c_{1,n} + c_{1,n+1}^\dagger c_{0,n}^\dagger c_{0,n} c_{1,n} + c_{0,n}^\dagger c_{1,n+1}^\dagger c_{1,n} c_{0,n+1} + c_{0,n+1}^\dagger c_{1,n+1}^\dagger c_{1,n+1} c_{0,n} + c_{0,n+1}^\dagger c_{1,n}^\dagger c_{1,n} c_{0,n}) + T \sum_n c_{1,n}^\dagger c_{0,n}^\dagger c_{1,n} c_{0,n}, \quad (3.9b)$$

$$V_{\text{sec}}^{(0)} = \frac{1}{2} J' \sum_n (c_{1,n}^\dagger c_{-1,n}^\dagger c_{-1,n} c_{1,n+1} + c_{1,n}^\dagger c_{-1,n+1}^\dagger c_{-1,n+1} c_{1,n+1} + c_{1,n+1}^\dagger c_{-1,n+1}^\dagger c_{-1,n+1} c_{1,n} + c_{1,n+1}^\dagger c_{-1,n}^\dagger c_{-1,n} c_{1,n} + c_{-1,n+1}^\dagger c_{1,n+1}^\dagger c_{1,n+1} c_{-1,n} + c_{-1,n+1}^\dagger c_{1,n}^\dagger c_{1,n} c_{-1,n} + c_{-1,n}^\dagger c_{1,n+1}^\dagger c_{1,n+1} c_{-1,n} + c_{-1,n}^\dagger c_{1,n}^\dagger c_{1,n} c_{-1,n})$$

$$\begin{aligned}
& -c_{1,n}^\dagger c_{-1,n+1}^\dagger c_{1,n} c_{-1,n+1} - c_{-1,n}^\dagger c_{1,n+1}^\dagger c_{-1,n} c_{1,n+1} + c_{1,n}^\dagger c_{-1,n+1}^\dagger c_{0,n} c_{0,n+1} + c_{-1,n}^\dagger c_{1,n+1}^\dagger c_{0,n} c_{0,n+1} + c_{0,n}^\dagger c_{0,n}^\dagger c_{0,n} c_{0,n+1} \\
& + c_{0,n}^\dagger c_{0,n+1}^\dagger c_{0,n+1} c_{0,n+1} + c_{0,n+1}^\dagger c_{0,n+1}^\dagger c_{0,n+1} c_{0,n} + c_{0,n+1}^\dagger c_{0,n}^\dagger c_{0,n} c_{0,n} \\
& + c_{0,n+1}^\dagger c_{0,n}^\dagger c_{1,n+1} c_{-1,n} + c_{0,n}^\dagger c_{0,n+1}^\dagger c_{1,n} c_{-1,n+1} + T \sum_n (c_{1,n}^\dagger c_{-1,n}^\dagger c_{1,n} c_{-1,n} + \frac{1}{2} c_{0,n}^\dagger c_{0,n}^\dagger c_{0,n} c_{0,n}) . \quad (3.9c)
\end{aligned}$$

Expressions for $V_{\text{sec}}^{(-1)}$ and $V_{\text{sec}}^{(-2)}$ can be obtained from those given for $V_{\text{sec}}^{(1)}$ and $V_{\text{sec}}^{(2)}$, respectively, by changing the signs of all the spin-projection quantum numbers on the boson operators. In writing Eq. (3.9) we have dropped terms involving six exciton operators, since they have no effect on states with less than three excitons.

The next step in the calculation of the two-exciton states is to write down the equation for the scattering matrix t for each of the channels characterized by a given value of m . These equations are schematically of the form

$$t = V + VGt, \quad (3.10)$$

where V is the interaction and G describes two-particle propagation. Since the dependence on the sign of m is trivial, we will consider only the values $m = 2, 1$, and 0 .

It is useful to have a qualitative picture of exciton-exciton interactions. For instance, consider the case $m = 2$, in Eq. (3.9a). The hard-core repulsion T moves the states with two excitons on the same site to infinite energy in the limit $T \rightarrow \infty$. The first four terms proportional to J' in Eq. (3.9a) then becomes irrelevant, since they describe coupling to the infinite-energy states. The remaining term,

$$\frac{1}{2} J' \sum_n c_{1,n+1}^\dagger c_{1,n}^\dagger c_{1,n+1} c_{1,n}, \quad (3.11)$$

is a repulsive term. Thus, in addition to the hard-core or kinematic repulsion, excitons have a dynamic repulsion when they are on neighboring sites. Since we are working in one dimension, we expect these interactions to lead to repulsive bound states. For $m = 1$ the analogous dynamical terms are

$$\frac{1}{2} J' \sum_n (c_{1,n}^\dagger c_{0,n+1}^\dagger c_{0,n} c_{1,n+1} + c_{0,n}^\dagger c_{1,n+1}^\dagger c_{1,n} c_{0,n+1}) \quad (3.12)$$

and the effect of these is similar to those for $m = 2$. For $m = 0$ the dynamical terms are

$$\begin{aligned}
& \frac{1}{2} J' \sum_n (-c_{1,n}^\dagger c_{-1,n+1}^\dagger c_{1,n} c_{-1,n+1} - c_{-1,n}^\dagger c_{1,n+1}^\dagger c_{-1,n} c_{1,n+1} \\
& + c_{1,n}^\dagger c_{-1,n+1}^\dagger c_{0,n} c_{0,n+1} + c_{-1,n}^\dagger c_{1,n+1}^\dagger c_{0,n} c_{0,n+1} \\
& + c_{0,n}^\dagger c_{0,n+1}^\dagger c_{1,n+1} c_{-1,n} + c_{0,n}^\dagger c_{0,n+1}^\dagger c_{-1,n+1} c_{1,n}) . \quad (3.13)
\end{aligned}$$

In this case there are some attractive static interactions, and we expect these to give rise to attractive bound states.

B. Solution for $m = 2$

In terms of exciton waves we may write

$$\begin{aligned}
V_{\text{sec}}^{(2)} &= \frac{1}{2N} \sum_{K, p, p'} V_K^{(2)}(p, p') \\
&\times c_{1, K/2+p}^\dagger c_{1, K/2-p}^\dagger c_{1, K/2+p} c_{1, K/2-p}, \quad (3.14)
\end{aligned}$$

and comparison with Eq. (3.9a) yields

$$\begin{aligned}
V_K^{(2)}(p, p') &= T + 2J' \cos \frac{1}{2} K (\cos p + \cos p') \\
&+ J' \cos p \cos p'. \quad (3.15)
\end{aligned}$$

The t matrix for $m = 2$ then obeys Eq. (3.10), which in this case reads

$$\begin{aligned}
t_{K,z}^{(2)}(p, p') &= V_K^{(2)}(p, p') + \frac{1}{N} \sum_{p''} V_K^{(2)}(p, p'') t_{K,z}^{(2)}(p'', p') \\
&\times \frac{1}{z - E_1(\frac{1}{2}K + p'') - E_1(\frac{1}{2}K - p'')} . \quad (3.16)
\end{aligned}$$

This equation is represented diagrammatically in Fig. 1, and we shall adhere to the convention that

$$k_1 = \frac{1}{2}K + p; \quad k'_1 = \frac{1}{2}K + p'; \quad (3.17a)$$

$$k_2 = \frac{1}{2}K - p; \quad k'_2 = \frac{1}{2}K - p'. \quad (3.17b)$$

Additional momenta will be introduced by double-primed variables in conformity with Eqs. (3.17). When clarity requires, we shall use the momenta k_1, k_2, k'_1, k'_2 , and the associated spin projections as arguments of the t matrix. Equation (3.16) is an integral equation with a kernel which is the sum of separable terms, and as such, it is solved by an ansatz of the form

$$t_{Kz}(p, p') = A + B \cos p + C \cos p' + D \cos p \cos p'. \quad (3.18)$$

The unknown coefficients A, B, C , and D , which are functions of K and z , are found by substituting Eq. (3.18) into Eq. (3.16). The resulting equations for the coefficients are

$$\begin{aligned}
A &= T + N^{-1} \sum_{p''} (T + 2J' \alpha \cos p'') (A + B \cos p'') \\
&\times [z - E_{11}(p'')]^{-1}, \quad (3.19a)
\end{aligned}$$

$$\begin{aligned}
B &= J' N^{-1} \sum_{p''} (2\alpha + \cos p'') (A + B \cos p'') \\
&\times [z - E_{11}(p'')]^{-1}, \quad (3.19b)
\end{aligned}$$

$$\begin{aligned}
C &= N^{-1} \sum_{p''} (T + 2J' \alpha \cos p'') (C + D \cos p'') \\
&\times [z - E_{11}(p'')]^{-1}, \quad (3.19c)
\end{aligned}$$

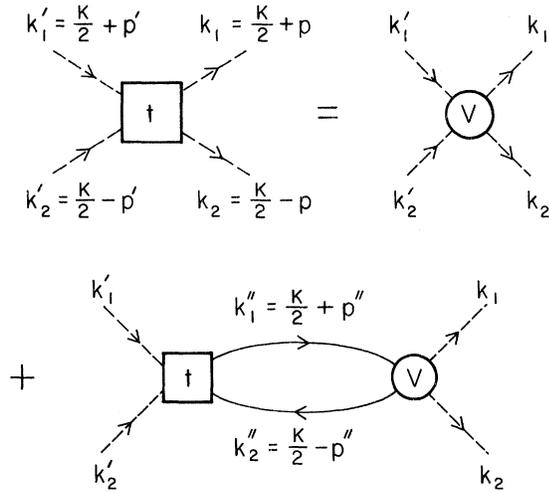


FIG. 1. Diagrammatic representation of Eq. (3.10) for the t matrix. The momenta are labeled in accord with Eq. (3.17).

$$D = J' + J'N^{-1} \sum_{p''} (2\alpha + \cos p'')(C + D \cos p'') \times [z - E_{11}(p'')]^{-1}, \quad (3.19d)$$

where $\alpha = \cos \frac{1}{2}K$ and

$$E_{mnt}(p) = E_m(\frac{1}{2}K + p) + E_{m'}(\frac{1}{2}K - p). \quad (3.20)$$

We use the evaluations

$$2J'N^{-1} \sum_{p''} [z - E_{11}(p'')]^{-1} = (xr)^{-1}, \quad (3.21a)$$

$$2J'N^{-1} \sum_{p''} \cos p'' [z - E_{11}(p'')]^{-1} = (\alpha r)^{-1}(r-1), \quad (3.21b)$$

$$2J'N^{-1} \sum_{p''} \cos^2 p'' [z - E_{11}(p'')]^{-1} = -x(r-1)/\alpha^2 r, \quad (3.21c)$$

where

$$x = (z - 4J - 2D + 2h)/2J', \quad (3.22)$$

$$r = (1 - \alpha^2/x^2)^{1/2}. \quad (3.23)$$

In Eq. (3.23) r is defined to have a branch cut for $|x| < \alpha$ on the real axis and its phase is determined so that $r \rightarrow 1$ for large $|x|$.

Using the evaluations of Eq. (3.21) one may write the solutions of Eq. (3.19) as

$$A = T - 4J'\alpha^2 + 4J'\alpha^2[1 + x(r-1)/2\alpha^2]^{-1} + T^2(2J'x - T)^{-1}, \quad (3.24a)$$

$$B = C = 2J'\alpha[1 + x(r-1)/2\alpha^2]^{-1} + 2\alpha T J'(2J'x - T)^{-1}, \quad (3.24b)$$

$$D = J'[1 + x(r-1)/2\alpha^2]^{-1} + 4J'^2\alpha^2(2J'x - T)^{-1}. \quad (3.24c)$$

Thus the explicit form for t is

$$t(z; k_1, 1; k_2, 1; k'_1, 1; k'_2, 1) = t_{k_2}^{(2)}(p, p') = T - 4J'\alpha^2 + (T + 2J'\alpha\gamma)(T + 2J'\alpha\gamma')(2J'x - T)^{-1}$$

$$+ J'[1 + x(r-1)/2\alpha^2]^{-1}(2\alpha + \gamma)(2\alpha + \gamma'), \quad (3.25)$$

where

$$\gamma = \cos p, \quad \gamma' = \cos p'. \quad (3.26)$$

The poles at $x = T/2J'$ reflect the bound states of two excitons on a single site. These states are eliminated in the limit $T \rightarrow \infty$. Otherwise, the condition for the existence of bound states is seen from Eqs. (3.24) or (3.25) to be

$$r = 1 - 2\alpha^2/x. \quad (3.27)$$

Analysis of this condition shows that there are no bound states for $|\alpha| > \frac{1}{2}$. For $|\alpha| < \frac{1}{2}$ the bound-state energy is determined by

$$x = \frac{1}{4} + \alpha^2 \quad (3.28a)$$

which, according to Eq. (3.22), yields an energy

$$E = 4J + 2D - 2h + 2J'(\frac{1}{4} + \alpha^2). \quad (3.28b)$$

The two-exciton continuum and the location of this bound state are shown in Fig. 2. Note that the bound state of Eq. (3.28) lies above the continuum.

The eigenvalue condition for the scattering states and the bound-state energy, Eqs. (3.28), are also obtained in Appendix A using Bethe's ansatz.¹⁴

C. Solution for $m = 1$

For $m = 1$ we write

$$V_{\text{sec}}^{(1)} = N^{-1} \sum_{K, p, p'} V_K^{(1)}(p, p') \times c_{1, K/2+p}^\dagger c_{0, K/2-p}^\dagger c_{1, K/2+p'} c_{0, K/2-p'}, \quad (3.29)$$

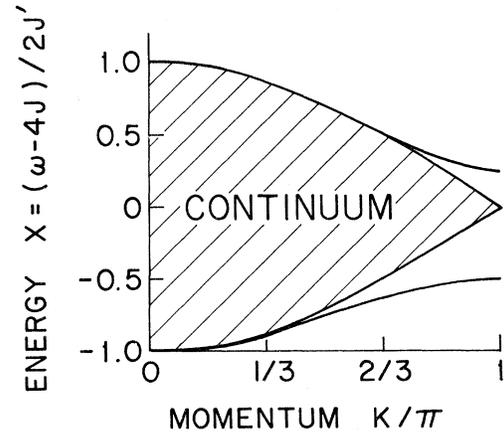


FIG. 2. The two-exciton band for $m = 0$ for $D = 0$. The repulsive bound state above the continuum emerges from the continuum at $\alpha = \frac{1}{2}$ (i.e., $K = \frac{2}{3}\pi$), and its dispersion relation is $E = 4J + 2J'(\frac{1}{4} + \cos^2 \frac{1}{2}K)$. The attractive bound state lies below the continuum for $K \neq 0$ and its dispersion relation is $E = 4J - J'(1 + \cos^2 \frac{1}{2}K)$. The band for $m \neq 0$ does not have the attractive bound state and its energy scale is shifted by replacing ω by $\omega' = \omega + mh - |m|D$. For $m = 0$ and $D \neq 0$, the bound-state condition, Eq. (B9) is more complicated.

and comparison with Eq. (3.9b) yields

$$V_K^{(1)}(p, p') = T + 2J' \cos \frac{1}{2}K(\cos p + \cos p') \\ + J' \cos p \cos p' - J' \sin p \sin p'. \quad (3.30)$$

The t matrix for $m = 1$ satisfies the equation

$$t_{Kz}^{(1)}(p, p') = V_K^{(1)}(p, p') + N^{-1} \sum_{p''} V_K^{(1)}(p, p'') \\ \times t_{Kz}^{(1)}(p'', p') [z - E_1(\frac{1}{2}K + p'') - E_0(\frac{1}{2}K - p'')]^{-1}, \quad (3.31)$$

which is solved by the ansatz

$$t_{Kz}^{(1)}(p, p') = -J' \Gamma_0 \sin p \sin p' + A' + B' \cos p \\ + C' \cos p' + D' \cos p \cos p'. \quad (3.32)$$

Here Γ_0 describes scattering in the odd-parity channel corresponding to total spin one and the other coefficients describe scattering in the even-parity channel corresponding to total spin two. Thus it is not surprising that the coefficients A' , B' , C' , and D' are simply related to the corresponding unprimed coefficients of Eq. (3.18). In particular, the primed coefficients are obtained from the expressions for the unprimed coefficients, Eq. (3.24), by replacing x by x' , where

$$x' = (z - 4J - D + h) / 2J'. \quad (3.33)$$

The difference in the definitions of Eq. (3.22) and (3.33) corresponds merely to a shift of all energies by an amount $D - h$. Otherwise, the even-

parity $m = 1$ states have the same dynamics as the $m = 2$ states. Using Eq. (3.31) it is found that Γ_0 satisfies the equation

$$-J' \Gamma_0 = -J' + J' N^{-1} \sum_{p''} \Gamma_0 \sin^2 p'' [z - E_{01}(p'')]^{-1}, \quad (3.34)$$

whose solution is

$$\Gamma_0 = 2\alpha^2 (2\alpha^2 - x' + x' r')^{-1}, \quad (3.35)$$

where $r' = (1 - \alpha^2/x'^2)^{1/2}$. Comparison with Eq. (3.27) shows that the bound states in the odd-parity $m = 1$ channel occur at the same energies as those in the even-parity $m = 2$ channel.

The complete expression for the $m = 1$ t matrix is thus

$$t(z; k_1, 1; k_2, 0; k'_1, 1; k'_2, 0) = t_{Kz}^{(1)}(p, p') \\ = T - 4J' \alpha^2 + (T + 2J' \alpha \gamma)(T + 2J' \alpha \gamma')(2J' x' - T)^{-1} \\ + J' [1 + x'(r' - 1) / 2\alpha^2]^{-1} \\ \times [(2\alpha + \gamma)(2\alpha + \gamma') - \sin p \sin p']. \quad (3.36)$$

D. Solution for $m = 0$

The case $m = 0$ is the only one where inclusion of the anisotropy leads to any extra complication. In this case the scattering terms are of the form shown in Fig. 3, and we write $V_{\text{sec}}^{(0)}$ as

$$V_{\text{sec}}^{(0)} = \frac{1}{2N} \sum_{K, p, p'} [2V_K^{1,1}(p, p') c_{1, K/2+p}^\dagger c_{-1, K/2-p}^\dagger c_{1, K/2+p} c_{-1, K/2-p} + 2V_K^{0,1}(p, p') c_{0, K/2+p}^\dagger c_{0, K/2-p}^\dagger c_{1, K/2+p} c_{-1, K/2-p} \\ + V_K^{1,0}(p, p') c_{1, K/2+p}^\dagger c_{-1, K/2-p}^\dagger c_{0, K/2+p} c_{0, K/2-p} + V_K^{0,0}(p, p') c_{0, K/2+p}^\dagger c_{0, K/2-p}^\dagger c_{0, K/2+p} c_{0, K/2-p}]. \quad (3.37)$$

Note that the potential coefficients are labeled by additional superscripts indicating the various processes shown in Fig. 3. Comparison with Eq. (3.9c) yields the following expressions for $V_K^{n, n'}(p, p')$:

$$V_K^{1,1}(p, p') = T + 2J' \cos \frac{1}{2}K(\cos p + \cos p') \\ - J' \cos p \cos p' - J' \sin p \sin p', \quad (3.38a)$$

$$V_K^{1,0}(p, p') = 2J' \cos p \cos p', \quad (3.38b)$$

$$V_K^{0,1}(p, p') = J' \cos p \cos p', \quad (3.38c)$$

$$V_K^{0,0}(p, p') = T + 2J' \cos \frac{1}{2}K(\cos p + \cos p'). \quad (3.38d)$$

Likewise, the t matrix is labeled by superscripts, and the scattering equation is now of the form

$$t_{Kz}^{n, n'}(p, p') = V_K^{n, n'}(p, p') \\ + \frac{1}{N} \sum_{p'', n''} V_K^{n, n''}(p, p'') t_{Kz}^{n'', n'}(p'', p) \\ \times [z - E_{n''}(\frac{1}{2}K + p'') - E_{-n''}(\frac{1}{2}K - p'')]^{-1}. \quad (3.39)$$

Again the integral equation for the t matrix has a kernel which is the sum of separable terms. This time it is solved by the ansatz

$$t_{Kz}^{n, n'}(p, p') = -\delta_{n,1} \delta_{n',1} J' \Gamma_{00} \sin p \sin p' + A^{n, n'} \\ + B^{n, n'} \cos p + C^{n, n'} \cos p' + D^{n, n'} \cos p \cos p', \quad (3.40)$$

where $\delta_{n, n'}$ is the Kronecker δ function. The determination of the coefficients in Eq. (3.40) is straightforward, but since the details are complicated, we relegate them to Appendix B.

In the case when D vanishes, the conclusions are as follows. Since $m = 0$ contains total spin 2, 1, and 0, the $m = 0$ channel has the same bound states as the $m = 2$ and $m = 1$ channels. In addition, there are bound states of total spin 0. In accord with the discussion following Eq. (3.13) these are found to be attractive bound states whose dispersion relation is given by (B11):

$$E = 4J - J'(1 + \alpha^2) \quad (3.41)$$

for all values of the total momentum as shown in Fig. 2.

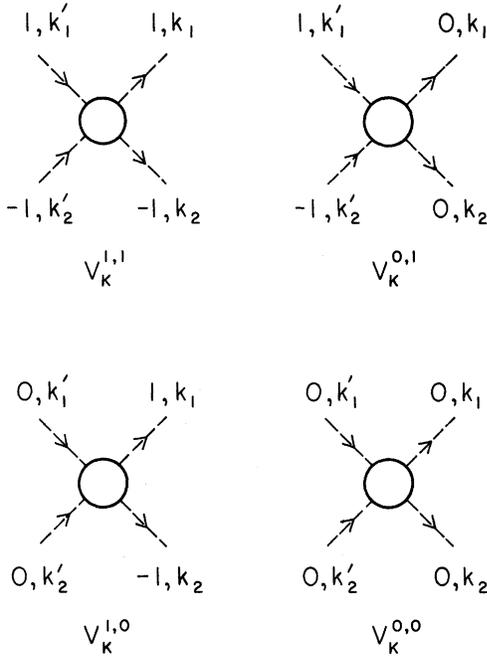


FIG. 3. Diagrammatic representation of $V_{\text{sec}}^{(0)}$ given in Eq. (3.37).

IV. SECOND VIRIAL COEFFICIENT FOR THE SUSCEPTIBILITY

A. Formal Expression for the Susceptibility

In this section we shall develop an expression for the susceptibility which includes exactly all effects up to second order in the density of excitons. What we wish to do is to relate the susceptibility χ to the t matrix, whose explicit evaluation was performed in Sec. III. To do this it suffices to calculate $\langle c_{m,n}^\dagger c_{m,n} \rangle_T$, since the magnetic moment is proportional to $\Sigma_m n \langle c_{m,n}^\dagger c_{m,n} \rangle_T$. However, a calculation of $\langle c_{m,n}^\dagger c_{m,n} \rangle_T$ requires knowing the temperature dependence of the exciton energy. The temperature-dependent energy shift is given by the self-energy Σ which in turn can be calculated if the t matrix is known.

The most important properties of the Green's functions we shall employ are the following:

$$G_m(k, t) = -i \langle c_{m,k}(t) c_{m,k}^\dagger(0) \rangle_T, \quad 0 < it < \beta \quad (4.1a)$$

$$G_m(k, t + i\beta) = G_m(k, t), \quad (4.1b)$$

$$G_m(k, z_\nu) = \int_0^{-i\beta} G_m(k, t) e^{iz_\nu t} dt, \quad (4.1c)$$

$$G_m(k, t) = (-i\beta)^{-1} \sum_\nu G_m(k, z_\nu) e^{-iz_\nu t}, \quad (4.1d)$$

$$G_m(k, z_\nu) = [z_\nu - E_m(k) - \Sigma_m(k, z_\nu)]^{-1}, \quad (4.1e)$$

where $z_\nu = 2\pi i\nu/\beta$ with ν an integer. Further discussion of this formalism can be found in the literature.¹⁸

The first step in the calculation is to relate the self-energy to the t matrix. This relation is shown in Fig. 4 and may be written as

$$\Sigma_m(k_1, z_\nu) = \sum_{m', k_2} (1 + \delta_{m, m'}) \sum_{\nu'=-\infty}^{+\infty} [z_{\nu'} - E_{m'}(k')]^{-1} \times (NB)^{-1} t(z_\nu + z_{\nu'}; k_1, m; k_2, m'; k_1, m; k_2, m'), \quad (4.2)$$

where we have indicated all arguments of the t matrix. Henceforth we shall drop the arguments in the t matrix describing the scattered excitons, it being understood that only matrix elements describing forward scattering will appear in this section. In Eq. (4.2) the factor $\delta_{m, m'}$ comes from the "exchange" term. Using contour integral techniques and dispersion relations [cf. Eqs. (4.7) and (4.8)], one can show that to lowest order in ρ ,

$$\Sigma_m(k_1, z_\nu) = N^{-1} \sum_{m', k_2} (1 + \delta_{m, m'}) n^0(k_2, m') \times t(z_\nu + E_{m'}(k_2); k_1, m; k_2, m'), \quad (4.3)$$

where

$$n^0(k, m) = (e^{\beta E_m(k)} - 1)^{-1}. \quad (4.4)$$

Since $n^0(k, m)$ is of order ρ , we see that $\Sigma_m(k_1, z_\nu)$ is also of order ρ .

We now evaluate $n_m = \langle c_{m,n}^\dagger c_{m,n} \rangle_T$. From Eq. (4.1) we have that

$$n_m = - (N\beta)^{-1} \sum_{k_1, \nu} e^{\epsilon_\nu 0^+} G_m(k_1, z_\nu), \quad (4.5)$$

where 0^+ denotes a positive infinitesimal. Using Eq. (4.3) for the self-energy and expanding in powers of Σ (i. e., in powers of ρ), we obtain

$$n_m = - (N\beta)^{-1} \sum_{k_1, \nu} \{ e^{\epsilon_\nu 0^+} [z_\nu - E_m(k_1)]^{-1} + \Sigma_m(k_1, z_\nu) [z_\nu - E_m(k_1)]^{-2} + \dots \}, \quad (4.6a)$$

$$n_m = n_m^0 - (N^2\beta)^{-1} \sum_{m', \nu} \sum_{k_1, k_2} (1 + \delta_{m, m'}) n^0(k_2, m') \times t(z_\nu + E_{m'}(k_2); k_1, m; k_2, m') [z_\nu - E_m(k_1)]^{-2}, \quad (4.6b)$$

where

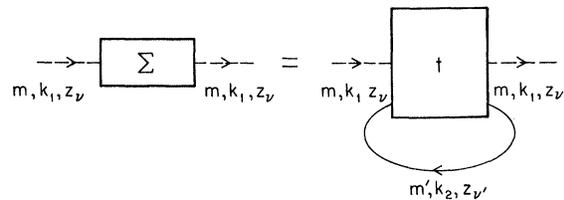


FIG. 4. Diagrammatic representation of Eq. (4.2). Here m' , k_2 , and $z_{\nu'}$ are summed over all values. Since the t matrix includes all "ladder" diagrams, this equation includes all diagrams for Σ with the minimum number (i. e., one) of backward, or hole, lines, and thus gives Σ correct to first order in the thermal density of excitons.

$$n_m^0 = N^{-1} \sum_k n^0(k, m).$$

To perform the sum over ν in Eq. (4.6b) we use the algorithm

$$-\beta^{-1} \sum_{\nu} F(z_{\nu}) = -\frac{1}{2\pi i} \oint_{C_1} \frac{F(z) dz}{e^{\beta z} - 1} = \frac{1}{2\pi i} \oint_{C_2} \frac{F(z) dz}{e^{\beta z} - 1}, \quad (4.7)$$

where C_1 is a counterclockwise contour surrounding only the imaginary axis and C_2 is a counterclockwise contour surrounding only the real axis. In using this method it is convenient to write the

t matrix in the form

$$t(z; k_1, m; k_2, m') = t(\infty; k_1, m; k_2, m') + \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{\rho(\omega; k_1, m; k_2, m')}{z - \omega}, \quad (4.8)$$

where

$$\rho(\omega; k_1, m; k_2, m') = \sum_{\sigma=\pm 1} \sigma t(\omega - i\sigma 0^+; k_1, m; k_2, m'). \quad (4.9)$$

Evaluating the ν sum in this fashion, we find the result

$$n_m = n_m^0 + N^{-2} \sum_{k_1, k_2, m'} (1 + \delta_{m, m'}) n^0(k_2, m') \left[-\beta t(\infty; k_1, m; k_2, m') n^0(k_1, m) [n^0(k_1, m) + 1] + \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{\rho(\omega; k_1, m; k_2, m')}{[\omega - E_m(k_1) - E_{m'}(k_2)]^2} \right. \\ \left. \times \left(\frac{1}{e^{\beta[\omega - E_{m'}(k_2)]} - 1} + \beta[\omega - E_m(k_1) - E_{m'}(k_2)] n^0(k_1, m) [n^0(k_1, m) + 1] - n^0(k_1, m) \right) \right]. \quad (4.10)$$

To evaluate this expression to leading order in ρ we may drop terms involving extra powers of $e^{-\beta E_m(k)}$. Thus we have

$$n_m = n_m^0 + N^{-2} \sum_{k_1, k_2, m'} (1 + \delta_{m, m'}) e^{-\beta[E_m(k_1) + E_{m'}(k_2)]} \left(-\beta t(\infty; k_1, m; k_2, m') + \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \rho(\omega; k_1, m; k_2, m') \Phi(\omega; k_1, m; k_2, m') \right), \quad (4.11)$$

where

$$\Phi(\omega; k_1, m; k_2, m') = \frac{e^{\beta[E_m(k_1) + E_{m'}(k_2) - \omega]} + \beta[\omega - E_m(k_1) - E_{m'}(k_2)] - 1}{[\omega - E_m(k_1) - E_{m'}(k_2)]^2}. \quad (4.12)$$

The zero-field susceptibility is given as

$$\frac{\chi}{N} = g^2 \mu_B^2 \frac{\partial}{\partial h} (n_1 - n_{-1}), \quad (4.13a)$$

$$\frac{\chi}{N} = 2g^2 \mu_B^2 \frac{\partial n_1}{\partial h}, \quad (4.13b)$$

where the derivatives are evaluated at $h=0$. Use of Eq. (4.11) for n_1 yields

$$\frac{(\chi - \chi_0)T}{NC} = \left(\frac{3k_B T}{N^2} \right) \frac{\partial}{\partial h} \sum_{k_1, k_2, m'} (1 + \delta_{1, m'}) \\ \times e^{-\beta[E_1(k_1) + E_{m'}(k_2)]} \left(-\beta t(\infty; k_1, 1; k_2, m') + \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \rho(\omega; k_1, 1; k_2, m') \Phi(\omega; k_1, 1; k_2, m') \right), \quad (4.14)$$

where

$$\frac{\chi_0 T}{NC} = (3k_B T) \frac{\partial n_1^0}{\partial h}, \quad (4.15a)$$

$$\frac{\chi_0 T}{NC} = 3N^{-1} \sum_k n^0(k, 1) [n^0(k, 1) + 1], \quad (4.15b)$$

$$\frac{\chi_0 T}{NC} = 3\rho A(\beta J') + 6\rho^2 A(2\beta J') + \dots, \quad \rho \ll 1. \quad (4.15c)$$

B. Explicit Evaluation of the Susceptibility

From now on we shall only consider the case $D=0$. It can be shown that both $\rho(\omega; 1, k_1, m', k_2)$ and $\Phi(\omega; 1, k_1, m', k_2)$ are functions of $\omega + h(1 + m')$. Consequently, the ω integral in Eq. (4.14) is h -independent, and we find that

$$\frac{(\chi - \chi_0)T}{NC} = 3N^{-2} \sum_{k_1, k_2, m'} (1 + m') (1 + \delta_{1, m'}) \\ \times e^{-\beta[E(k) + E(k')]} \left(-\beta t(\infty; k_1, 1; k_2, m') + \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \rho(\omega; k_1, 1; k_2, m') \Phi(\omega; k_1, k_2) \right), \quad (4.16)$$

where we have dropped those m labels which are irrelevant when $h=0$. It is convenient to write Eq. (4.16) as

$$(\chi - \chi_0)T/NC = \Lambda_{\infty} + \Lambda_P + \Lambda_C, \quad (4.17)$$

corresponding to the contributions from $t(\infty)$, from the poles of t , and from the continuum, respectively.

Using Eqs. (3.25) and (3.36) for the t matrices at infinite energy, we obtain Λ_∞ as

$$\Lambda_\infty = - \left(\frac{3\beta}{N^2} \right) \sum_{k_1, k_2} e^{-\beta [E(k_1) + E(k_2)]} \times \{ 5[T - 4J'\alpha^2 + J'(2\alpha + \gamma)^2] - \sin^2 p \}, \quad (4.18a)$$

$$\Lambda_\infty = -3\beta[(5T + 2J')I_0^2 + 20J'I_1I_0 + 3J'I_1^2], \quad (4.18b)$$

where

$$I_n = N^{-1} \sum_k \cos^n k e^{-\beta E(k)}. \quad (4.19)$$

The term in Eq. (4.18b) involving T will be cancelled by another term.

To evaluate the pole and continuum terms, we need to construct the spectral representation for the t matrix. For this purpose the following dispersion relation is helpful:

$$\left(1 + \frac{x(r-1)}{2\alpha^2} \right)^{-1} = 1 + \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{1}{z-\omega} \sigma(\alpha, \omega), \quad (4.20)$$

where $x = (z - 4J)/2J'$, r is defined in Eq. (3.23), and

$$\begin{aligned} \sigma(\alpha, \omega) = & 4 \left(1 - \frac{2\tilde{\omega}}{J'} + 4\alpha^2 \right)^{-1} \left[\alpha^2 - \left(\frac{\tilde{\omega}}{2J'} \right)^2 \right]^{1/2} \\ & \times \theta \left(\alpha^2 - \left(\frac{\tilde{\omega}}{2J'} \right)^2 \right) \\ & + \pi J' (1 - 4\alpha^2) \theta(1 - 4\alpha^2) \delta(\tilde{\omega} - \frac{1}{2}J' - 2\alpha^2 J'), \end{aligned} \quad (4.21)$$

where $\theta(x) = \frac{1}{2}(x + |x|)/x$, and $\tilde{\omega} = \omega - 4J$. By comparing this relation with Eqs. (4.8), (3.25), and (3.36) one can construct the spectral weight functions for the t matrices. In particular, if we determine the residue of the t matrices at the bound-state pole and also read off the residue of the pole at $T/2J'$ from Eqs. (3.25) and (3.36), we are led to the result

$$\begin{aligned} \Lambda_p = & 15N^{-2} \sum_{k_1, k_2} e^{-\beta [E(k_1) + E(k_2)]} \frac{(T + 2J'\alpha\gamma)^2}{[4J + T - E(k_1) - E(k_2)]^2} \{ e^{\beta [E(k_1) + E(k_2) - 4J - T]} + \beta [4J + T - E(k_1) - E(k_2)] - 1 \} \\ & + 3N^{-2} \sum_{k_1, k_2} e^{-\beta [E(k_1) + E(k_2)]} \frac{1}{2} J'^2 \frac{[5(2\alpha + \gamma)^2 - (1 - \gamma^2)][1 - 4\alpha^2]}{[4J + \frac{1}{2}J' + 2J'\alpha^2 - E(k_1) - E(k_2)]^2} \theta(1 - 4\alpha^2) \\ & \times (\exp\{ \beta [E(k_1) + E(k_2) - 4J - \frac{1}{2}J' - 2J'\alpha^2] \} + \beta [4J + \frac{1}{2}J' + 2J'\alpha^2 - E(k_1) - E(k_2)] - 1). \end{aligned} \quad (4.22)$$

Note that $E(k_1) + E(k_2) = 4J - 2J'\alpha\gamma$, using which we see that the term proportional to βT cancels the similar term in Eq. (4.18b).

With the help of Eq. (4.21) we evaluate the continuum contribution to Λ as

$$\begin{aligned} \Lambda_c = & \frac{3}{\pi^3} \int_{-1}^1 \frac{d\alpha}{(1 - \alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1 - \gamma^2)^{1/2}} \int_{-1}^1 \frac{dy(1 - y^2)^{1/2}}{1 + 4\alpha^2 - 4\alpha y} [5(2\alpha + \gamma)^2 - (1 - \gamma^2)] e^{-\beta [E(k_1) + E(k_2)]} \\ & \times \left(\frac{e^{-2\beta J'\alpha\gamma - 4\beta J} e^{\beta [E(k_1) + E(k_2)]}}{(y + \gamma)^2} + \frac{2\beta J'\alpha}{y + \gamma} - \frac{1}{(y + \gamma)^2} \right). \end{aligned} \quad (4.23)$$

In obtaining this result we transformed the ω integral in Eq. (4.16) using $\omega = 4J + 2J'\alpha\gamma$ and also the replacements

$$\begin{aligned} N^{-2} \sum_{k_1, k_2} &= \frac{1}{\pi^2} \int_0^{2\pi} dK \int_0^{\pi - K/2} dp \\ &= \frac{1}{\pi^2} \int_{-1}^1 \frac{d\alpha}{(1 - \alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1 - \gamma^2)^{1/2}}, \end{aligned} \quad (4.24)$$

which are valid for integrands $I(\alpha, \gamma)$ satisfying $I(\alpha, \gamma) = I(-\alpha, -\gamma)$.

Consider the y integral in Eq. (4.23). The integrand is regular at $y = -\gamma$. However, we wish to treat the three terms in the final bracket sep-

arately, because the last two terms give rise to dispersion-relation integrals which are easily done in closed form:

$$\begin{aligned} \oint \frac{1}{2\pi} \int_{-1}^1 \frac{dy(1 - y^2)^{1/2}}{(y + \gamma)(1 - 4\alpha y + 4\alpha^2)} \\ = \left(\frac{\frac{1}{2}\alpha\gamma + \frac{1}{4} + \theta(1 - 4\alpha^2)(\alpha^2 - \frac{1}{4})}{(\frac{1}{4} + \alpha^2 + \alpha\gamma)(4\alpha)} \right), \end{aligned} \quad (4.25a)$$

$$\begin{aligned} \oint \frac{1}{2\pi} \int_{-1}^1 \frac{dy(1 - y^2)^{1/2}}{(y + \gamma)^2(1 - 4\alpha y + 4\alpha^2)} \\ = - \frac{|\alpha^2 - \frac{1}{4}|}{8(\frac{1}{4} + \alpha^2 + \alpha\gamma)^2}. \end{aligned} \quad (4.25b)$$

In using such singular integrals one must treat all three terms in the final bracket of Eq. (4.23) in the same way. To obtain Eq. (4.25) we replaced the final bracket, denoted $G(y)$, by $\frac{1}{2}[G(y+i0^*) + G(y-i0^*)]$. Furthermore, for the first term in the final bracket, we would like to interchange the order of integration, since the γ integral can be done analytically. However, since the integrand is singular, it is not obvious that one can interchange

the order of integration. As Bloomfield¹⁵ has pointed out, this difficulty can be surmounted by deforming the contour in the y integral, so that the integrand is never singular. After the order of integration is interchanged, the contour is restored to its original shape, and one obtains a prescription analogous to the Plemelj formulas.¹⁹ This analysis is carried out in detail in Appendix C. The result of the analysis is that

$$\Lambda_c = -\frac{15\rho^2}{\pi} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} e^{2\beta J'(\alpha-1)} + \frac{12\rho^2}{\pi^2} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} e^{2\beta J'(\alpha\gamma-1)} \times \left(\frac{\frac{3}{2}(1-\gamma^2)}{D} + \beta J' \left[\frac{1}{2}\alpha\gamma + \frac{1}{4} + \theta(1-4\alpha^2)(\alpha^2 - \frac{1}{4}) \right] [5(2\alpha+\gamma)^2 + \gamma^2 - 1]/D + \left| \alpha^2 - \frac{1}{4} \right| [5(2\alpha+\gamma)^2 + \gamma^2 - 1]/D^2 \right), \tag{4.26}$$

where

$$D = 1 + 4\alpha\gamma + 4\alpha^2. \tag{4.27}$$

Collecting the results of Eqs. (4.15c), (4.18b), (4.22), and (4.26) we obtain

$$\chi T/NC = 3\rho A(\beta J') + \rho^2 B(\beta J') + \theta(\rho^3), \tag{4.28}$$

with

$$B(\beta J') = -9A(2\beta J') - 15A^2(\beta J') - 3\beta J'(2I_0^2 + 10I_1I_0 + 3I_1^2) + \frac{12}{\pi^2} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} e^{2\beta J'\alpha\gamma} \left\{ \frac{1}{2} [5(2\alpha+\gamma)^2 + \gamma^2 - 1] (1-4\alpha^2)\theta(1-4\alpha^2) [e^{-J'D\beta/2} + \frac{1}{2}\beta J'D - 1]/D^2 + \frac{3}{2}(1-\gamma^2)/D + \left[\frac{1}{2}\alpha\gamma + \frac{1}{4} + \theta(1-4\alpha^2)(\alpha^2 - \frac{1}{4}) \right] [5(2\alpha+\gamma)^2 + \gamma^2 - 1] \beta J'/D + \left| \alpha^2 - \frac{1}{4} \right| [5(2\alpha+\gamma)^2 + \gamma^2 - 1]/D^2 \right\}. \tag{4.29}$$

We now obtain expansions for $B(\beta J')$ in the extremes of temperature. At low temperatures, we shall keep all contributions larger than linear in temperature. Within this accuracy one has $I_n \sim A(\beta J')$, and these are of order $(\beta J')^{-1/2}$. Thus Eq. (4.29) yields

$$B(\beta J') \sim -9A(2\beta J'). \tag{4.30}$$

The high-temperature expansion is most readily obtained by noting from Eqs. (4.18) and (4.23) that Λ_∞ and Λ_c vanish for $\beta=0$. Inspection of Eq. (4.22) shows that $\Lambda_p = -15$ for $\beta=0$. Taking account of Eq. (4.15c) we find that

$$B(0) = -9. \tag{4.31}$$

As suggested in Sec. II, we can determine what kind of statistics excitons obey when two-particle collisions are included. To do this we study the quantity

$$R(\beta J') \equiv B(\beta J')/A(2\beta J'). \tag{4.32}$$

The results of Eq. (4.30) and (4.31) show that $R = -9$ in both the low- and high-temperature lim-

its. Numerical evaluation of $R(\beta J')$ shows that it remains close to its limiting value over the entire temperature range. This is seen in Fig. 5, where our numerical results for $R(\beta J')$ are plotted. The conclusion is therefore that over the entire temperature range excitons obey localized statistics.

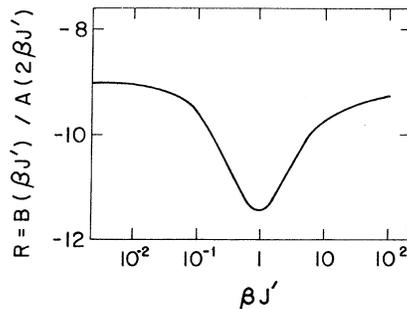


FIG. 5. Normalized second virial coefficient $R(\beta J') = B(\beta J')/A(2\beta J')$ for the susceptibility vs $\beta J'$. The values of R corresponding to localized, Fermi, and Bose statistics are -9 , -6 , and 6 , respectively.

C. Discussion of Kinematic Properties

Having obtained, via a complicated and perhaps opaque calculation, a result which conforms to the simple argument that momentum states cannot be doubly occupied, it is natural to see whether such a result can be related to the interactions between quasiparticles. In other words, we seek to formulate a general criterion for the exclusion in momentum space which our explicit calculations have established.

We shall consider only the low-temperature limit, since it is then that the exclusion in momentum space differs from Dyson's result¹⁶ for a three-dimensional spin- $\frac{1}{2}$ ferromagnet. To investigate the kinematics in momentum space, we study the quantity

$$f_{k, k'} \equiv \langle c_{1, k}^\dagger c_{1, k'}^\dagger c_{1, k'} c_{1, k} \rangle . \quad (4.33)$$

For simplicity, we restrict the discussion to the $m=2$ manifold and will set $h=0$. By a derivation quite similar to that leading to Eq. (4.6) one obtains the expression

$$f_{k, k'} = (1 + \delta_{k, k'}) \left(n_k^0 n_{k'}^0 - \frac{1}{N\beta} \sum_z [z - E(k) - E(k')]^{-2} \times t(z; k, k') \right), \quad (4.34a)$$

$$f_{k, k'} \equiv (1 + \delta_{k, k'}) n_k^0 n_{k'}^0 (1 + N^{-1} g_{k, k'}) , \quad (4.34b)$$

where $t(z; k, k') \equiv t(z; k, 1; k', 1; k, 1; k', 1)$ and the magnetic quantum numbers corresponding to the $m=2$ manifold have been dropped. Note the occurrence of the factor N^{-1} in the second term in Eq. (4.34). On general grounds one can write

$$g_{k, k'} = NA_g(k, k') + B_g(k, k'), \quad (4.35)$$

where A_g and B_g are of order unity, independent of N . True exclusion in momentum space would require that A_g be nonzero, which is not possible without long-range interactions. However, if $B_g(k, k')$ becomes large and negative for $|k - k'| \rightarrow 0$, then $f_{k, k'}$ will display a "Fermi hole" in momentum space.

We therefore study $g_{k, k'}$ for $k' \rightarrow k$. Following the procedure leading to Eq. (4.16) we find that

$$g_{k, k} = -\beta t(2E(k), k, k) + \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{\rho(\omega; k, 1; k, 1)}{[\omega - 2E(k)]^2} \times (e^{\beta[2E(k) - \omega]} - 1) . \quad (4.36)$$

Examination of the two-particle kinematics indicates that for a fixed total momentum K the minimum kinetic energy, denoted ω_{\min} , occurs for $k_1 = k_2 = \frac{1}{2}K$, so that for $K=2k$, $\omega_{\min} = 2E(k) = 4J - 2J'\alpha$. Hence, unless $\rho(\omega)$ has an attractive bound-state contribution, both terms on the right-hand side of Eq. (4.36) are related to the t matrix at the bottom of the two-particle continuum.

The behavior of the t matrix at the bottom of the band can be obtained from a simple argument. We determine t from

$$t = V + VGt \quad (4.37a)$$

or

$$t = (1 - VG)^{-1} V . \quad (4.37b)$$

But VG has a behavior dominated by the divergence in the one-dimensional density of states, $g(\omega')$:

$$VG \sim \int_{\omega_{\min}} (\omega - \omega')^{-1} g(\omega') d\omega' , \quad (4.38a)$$

$$VG \sim \int_{\omega_{\min}} (\omega - \omega')^{-1} (\omega' - \omega_{\min})^{-1/2} d\omega' , \quad (4.38b)$$

$$VG \sim i(\omega - \omega_{\min})^{-1/2} + \text{const} , \text{ as } \omega \rightarrow \omega_{\min}^+ . \quad (4.38c)$$

Thus we expect that

$$t \sim i(\omega - \omega_{\min})^{1/2} . \quad (4.39)$$

In contrast, in three dimensions one does not have a divergent density of states and there the result is²⁰

$$t \sim t_0 + ci(\omega - \omega_{\min})^{1/2} . \quad (4.40)$$

If we accept Eq. (4.39), we see that the first term of Eq. (4.36) vanishes. Dimensional analysis, taking $\rho(\omega) \sim (\omega - \omega_{\min})^{1/2}$ from Eq. (4.39), leads to the estimate

$$g_{k, k} \sim -\beta^{1/2} \quad (4.41)$$

indicating momentum space exclusion.

We now consider a more general interaction in one dimension. However, to avoid becoming immersed in the technical details of a general argument, we give here a simple treatment from which the main physical results are apparent. We consider the case of a lattice gas with nearest-neighbor interactions and a hard-core repulsion, for which

$$V = \frac{1}{2} T \sum_n c_n^\dagger c_n^\dagger c_n c_n + \alpha_+ \sum_n c_n^\dagger c_n^\dagger c_n c_{n+1} + \alpha_- \sum_n c_n^\dagger c_n^\dagger c_n c_{n-1} + \beta_+ \sum_n c_n^\dagger c_{n+1}^\dagger c_{n+1} c_n + \beta_- \sum_n c_n^\dagger c_{n-1}^\dagger c_{n-1} c_n + \xi J' \sum_n c_n^\dagger c_{n+1}^\dagger c_{n+1} c_n . \quad (4.42)$$

We take the unperturbed single-particle spectrum to be

$$E(k) = E_0 - J' \cos k . \quad (4.43)$$

We will ignore the terms in α_+ and β_+ , since they can have no effect in the $T \rightarrow \infty$ limit. In other words, we take the potential to have a matrix element,

$$V^K(p, p') = T + 2\xi J' \cos p \cos p' \quad (4.44)$$

in the notation of Eq. (3.14). For $\xi = \frac{1}{2}$, this model

is the exciton gas of Eq. (3.15), while for $\xi = -1$, the model reproduces the spin- $\frac{1}{2}$ Heisenberg ferromagnet.

We now construct the t matrix which is found, as in Sec. III, by solving Eq. (3.10). To this end we write V , G , and t in the form

$$V(p, p') = \sum_{i,j} V_{ij} \varphi_i(p) \varphi_j(p'), \quad (4.45a)$$

$$G(p, p') = \sum_{i,j} G_{ij} \varphi_i(p) \varphi_j(p'), \quad (4.45b)$$

$$t(p, p') = \sum_{i,j} t_{ij} \varphi_i(p) \varphi_j(p'), \quad (4.45c)$$

where $\varphi_1(p) = 1$ and $\varphi_2(p) = \sqrt{2} \cos p$. Thus the potential is represented by the matrix

$$V = \begin{pmatrix} T & 0 \\ 0 & \xi J' \end{pmatrix}. \quad (4.46)$$

The propagator has a matrix representation obtainable from Eq. (3.21) as

$$G = \frac{1}{2Jr} \begin{bmatrix} \frac{1}{x} & -\sqrt{2} \left(\frac{1-r}{\alpha} \right) \\ -\sqrt{2} \left(\frac{1-r}{\alpha} \right) & \frac{2x(1-r)}{\alpha} \end{bmatrix}, \quad (4.47)$$

where now $x = (\omega - 4J)/2J'$. In the $T \rightarrow \infty$ limit, we find that

$$t = V(1 - GV)^{-1}$$

$$= \begin{bmatrix} -2J'x + \frac{2J'x(1-r)}{D} & \frac{\sqrt{2}\xi x J'(1-r)}{\alpha D} \\ \frac{\sqrt{2}\xi x J'(1-r)}{\alpha D} & \frac{\xi J'}{D} \end{bmatrix}, \quad (4.48)$$

where

$$D = 1 - \frac{\xi x}{\alpha^2} (1-r). \quad (4.49)$$

We wish to study the value of t near threshold, i. e., for $\omega \rightarrow \omega_{\min}$, i. e., for $x \rightarrow -\alpha$. Thus we set $x = -\alpha + \hat{\omega}$ and evaluate $t_0(\hat{\omega}) \equiv t(\omega_{\min} + 2J'\hat{\omega}; \frac{1}{2}K, \frac{1}{2}K)$. Thus we set $p = p' = 0$ and have that

$$t_0(\hat{\omega}) = \sum_{i=1,2} \sum_{j=1,2} t_{ij}(x = -\alpha + \hat{\omega}) \varphi_i(0) \varphi_j(0). \quad (4.50)$$

In the limit $\hat{\omega} \rightarrow 0$, we find that

$$t_0(\hat{\omega}) \sim 2J' i \alpha \hat{\omega}^{1/2}, \quad \xi + \alpha \neq 0 \quad (4.51a)$$

$$t_0(\hat{\omega}) \sim -2J'\hat{\omega}, \quad \xi + \alpha = 0. \quad (4.51b)$$

To complete the discussion we consider possible effects due to bound states, the condition for which is

$$D = 0. \quad (4.52)$$

Analysis of this condition shows that $D = 0$ if $|\xi|$ $|\xi| > \alpha$ and

$$x = \frac{1}{2\xi} (\alpha^2 + \xi^2), \quad (4.53)$$

and in particular for $\xi < -1$, a bound state appears below the continuum even for zero total momentum. Equation (4.53) gives the bound-state energy ω_B as

$$\omega_B = 2E_0 + \frac{1}{\xi} J' (\alpha^2 + \xi^2), \quad (4.54a)$$

$$\omega_B = \omega_{\min} + \frac{1}{\xi} J' (\alpha + \xi)^2. \quad (4.54b)$$

The spectral weight function ρ then has a bound-state contribution

$$\rho(\omega; k, 1; k, 1) \sim A \delta(\omega - \omega_B). \quad (4.55)$$

For the discussion of $g_{k,k}$ as $k \rightarrow 0$, we thus distinguish three regimes: $\xi > -1$, $\xi = -1$, and $\xi < -1$. For all these cases one has $t_0(0) = 0$, so that $t(2E(k), k, k) = 0$. Thus we need consider only the integral in Eq. (4.36). For $\xi > -1$, as is the case for excitons with $m = 2$, we have from Eq. (4.51a) that $\rho(\omega) \sim (\omega - \omega_{\min})^{1/2}$ independent of ξ , and the argument leading to Eq. (4.41) holds. For $\xi = -1$, i. e., for spin waves, we have the result given in Eq. (4.51b), so that $\rho(\omega)/(\omega - \omega_{\min}) \rightarrow 0$ as $\omega \rightarrow \omega_{\min}$, and as a result $g_{k,k}$ does not become large at low temperatures. For $\xi < -1$, the bound-state contribution to $\rho(\omega)$ given in Eq. (4.55) dominates the integral of Eq. (4.36) and $g_{k,k} \sim e^{\beta(\omega_{\min} - \omega_B)}$ becomes large at low temperatures, since $\omega_B < \omega_{\min}$. If also $\omega_B < 0$, then the system is unstable relative to formation of bound pairs of excitations.

V. PERTURBATION THEORY IN J'/J

A. Formalism

The objective of this section is to study the effects of the nonsecular terms on the exciton dispersion relation. The most convenient way to do this is to use perturbation theory in the parameter (J'/J) . For this purpose we construct an effective Hamiltonian for the single-exciton manifold of states which is correct to third order in $(J'/J) \equiv \hat{j}'$. To do this we write the Hamiltonian for $D = \hbar = 0$ in the form

$$\mathcal{H} = \mathcal{H}_0 + V, \quad (5.1)$$

with

$$\mathcal{H}_0 = 2J \sum_n N_n, \quad (5.2)$$

where N_n is the number of excitons on the n th site and

$$V = 2J' \sum_n \vec{S}_{2n} \cdot \vec{S}_{2n+1}. \quad (5.3)$$

A convenient prescription for constructing an effective Hamiltonian for a degenerate manifold is described by Messiah.²¹ There it is shown that the eigenvalues of the degenerate manifold a , corresponding to an unperturbed energy E_a^0 , are determined by the generalized eigenvalue equation which we write in the form

$$\det |W_a + (E_a^0 - \lambda)K_a| = 0, \quad (5.4)$$

where W_a and K_a are given by a perturbation series. Thus the effective Hamiltonian for the space a is

$$\mathcal{H}_{\text{eff}} = E_a^0 \underline{1} + K_a^{-1/2} W_a K_a^{-1/2}. \quad (5.5)$$

If we keep only those terms in the perturbation series for V_a and K_a which are required to construct \mathcal{H}_{eff} to third order in $(J'/J) \equiv \hat{j}'$, we then have

$$K_a = P_a \left(\underline{1} - V \frac{1 - P_a}{E_a^2} V \dots \right) P_a, \quad (5.6a)$$

$$W_a = P_a \left(V + V \frac{1 - P_a}{E_a} V + V \frac{1 - P_a}{E_a} V \frac{1 - P_a}{E_a} V - VP_a V \frac{1 - P_a}{E_a^2} V - V \frac{1 - P_a}{E_a^2} VP_a V + \dots \right) P_a, \quad (5.6b)$$

where P_a is the projection operator for the manifold a and

$$E_a = E_a^0 - H_0. \quad (5.7)$$

Using this prescription, we construct H_{eff} correct to third order in V as

$$\mathcal{H}_{\text{eff}} = P_a \left(V + V \frac{1 - P_a}{E_a} V + V \frac{1 - P_a}{E_a} V \frac{1 - P_a}{E_a} V - \frac{1}{2} VP_a V \frac{1 - P_a}{E_a^2} V - \frac{1}{2} V \frac{1 - P_a}{E_a^2} VP_a V + \dots \right) P_a + E_a^0 \underline{1}. \quad (5.8)$$

We now specialize to the exciton problem and note the simplifying feature that \mathcal{H}_0 , given by Eq. (5.2), produces equally spaced eigenvalues. Hence we classify terms in V according to the number of excitons they create:

$$V \equiv \sum_n V_n, \quad (5.9)$$

where

$$[\mathcal{H}_0, V_n] = 2nJV_n \quad (5.10)$$

and, of course, $P_a VP_a = V_0$ is the secular part of V . The integer n in Eq. (5.9) will be shown to be confined to the range $-2 \leq n \leq 2$. Use of Eqs. (5.9) and (5.10) enables us to write \mathcal{H}_{eff} in the following way:

$$\begin{aligned} \mathcal{H}_{\text{eff}} = & V_0 + \frac{1}{2J} [V_1, V_{-1}] + \frac{1}{4J} [V_2, V_{-2}] \\ & + \frac{1}{8J^2} \{ [V_{-1}, [V_0, V_1]] + \text{H. c.} \} \\ & + \frac{1}{32J^2} \{ [V_{-2}, [V_0, V_2]] + \text{H. c.} \} \\ & + \frac{1}{8J^2} \{ [V_1, [V_1, V_{-2}]] + \text{H. c.} \}. \quad (5.11) \end{aligned}$$

Here H. c. denotes the Hermitian conjugate of the preceding factor.

Explicit expressions for the V_n 's are most easily obtained by writing

$$\vec{\mathcal{S}}_n = \vec{\mathcal{S}}_{2n} + \vec{\mathcal{S}}_{2n-1}, \quad (5.12a)$$

$$\vec{\mathcal{T}}_n = \vec{\mathcal{S}}_{2n} - \vec{\mathcal{S}}_{2n-1}, \quad (5.12b)$$

so that

$$V = \frac{1}{2} J' \sum_n (\vec{\mathcal{S}}_n + \vec{\mathcal{T}}_n) \cdot (\vec{\mathcal{S}}_{n+1} - \vec{\mathcal{T}}_{n+1}). \quad (5.13)$$

We write

$$T_n^\sigma = -d_{0,n}^\dagger - d_{0,n}, \quad (5.14a)$$

$$2^{-1/2} T_n^+ = d_{1,n}^\dagger - d_{-1,n}, \quad (5.14b)$$

$$2^{-1/2} T_n^- = -d_{-1,n}^\dagger + d_{1,n}, \quad (5.14c)$$

so that

$$(V_{-2})^+ = V_2 = -\frac{1}{2} J' \sum_{n,m} (-1)^m d_{m,n}^\dagger d_{-m,n+1}^\dagger, \quad (5.15a)$$

$$(V_{-1})^\dagger = V_1 = -\frac{1}{2} J' \sum_{n,m,\sigma} (-1)^m \sigma d_{m,n}^\dagger s_{n+\sigma}^{-m}, \quad (5.15b)$$

$$V_0 = \frac{1}{2} J' \sum_{nm} (-d_{m,n}^\dagger d_{m,n+1} - d_{m,n+1}^\dagger d_{m,n} + s_n^m s_{n+1}^{-m} (-1)^m), \quad (5.15c)$$

where m ranges over the values $-1, 0$, and 1 ; σ over the values -1 and 1 ; and n indexes the unit cells. The operators appearing in Eq. (5.15) are defined as

$$d_{m,n}^\dagger = c_{m,n}^\dagger (1 - N_n), \quad (5.16a)$$

$$(s_n^{-1})^\dagger = -s_n^1 = c_{1,n}^\dagger c_{0,n} + c_{0,n}^\dagger c_{-1,n}, \quad (5.16b)$$

$$s_n^0 = c_{1,n}^\dagger c_{1,n} - c_{-1,n}^\dagger c_{-1,n}, \quad (5.16c)$$

and they obey the commutation relations

$$[d_{m',n'}, d_{m,n}^\dagger] = \delta_{n,n'} [c_{m,n}^\dagger (1 - N_n) c_{m',n} + c_{m',n}^\dagger c_{m,n} (1 - N_n) - (1 - N_n)^2 \delta_{m,m'}], \quad (5.17a)$$

$$[s_{n'}^{m'}, d_{m,n}^\dagger] = \delta_{n,n'} A_{m,m'} c_{m+m',n}^\dagger (1 - N_n), \quad (5.17b)$$

where

$$A_{1,1} = A_{0,0} = A_{-1,-1} = 0, \quad (5.18a)$$

$$A_{1,0} = A_{1,-1} = A_{0,-1} = -A_{0,1} = -A_{-1,1} = -A_{-1,0} = 1. \quad (5.18b)$$

In the following, we restrict ourselves to the single-exciton manifold. For such states one has

$$s_n^m s_{n+\sigma}^{m'} = 0, \quad (5.19a)$$

$$(1 - N_n)^2 = 1 - N_n, \quad (5.19b)$$

$$s_n^m d_{m',n'} = 0, \quad (5.19c)$$

and so forth.

B. Calculation up to Second Order in \hat{j}'

Within the manifold of one-exciton states, the term which is first order in V , denoted $\mathcal{H}_{\text{eff},1}$, is simply

$$\mathcal{H}_{\text{eff},1} = -\frac{1}{2}J' \sum_{m,n,\sigma} c_{m,n}^\dagger c_{m,n+\sigma}, \quad (5.20a)$$

$$\mathcal{H}_{\text{eff},1} = -J' \sum_{m,k} \cos k c_{m,k}^\dagger c_{m,k}, \quad (5.20b)$$

in agreement with the well-known results.

The terms which are second order in V , denoted $\mathcal{H}_{\text{eff},2}$, are

$$(J/J'^2)\mathcal{H}_{\text{eff},2} = \frac{1}{8} \sum_{m,n,\sigma} \sum_{m',n',\sigma'} \sigma\sigma' [d_{m,n}^\dagger s_{n+\sigma}^{-m}, d_{m',n'} s_{n'+\sigma'}^{m'}] (-1)^{m'} + \frac{1}{16} \sum_{m,n} \sum_{m',n'} (-1)^{m+m'} [d_{m,n}^\dagger d_{-m,n+1}^\dagger, d_{m',n'} d_{-m',n'+1}] \quad (5.21)$$

Expanding the commutators and keeping only nonzero terms, we have

$$(J/J'^2)\mathcal{H}_{\text{eff},2} = \frac{1}{8} \sum_{m,n,\sigma} \sum_{m',\sigma'} (-1)^{m'} \sigma\sigma' \{d_{m,n}^\dagger [s_{n+\sigma}^{-m}, d_{m',n'} s_{n'+\sigma'}^{m'}] s_{n+\sigma}^{-m} + d_{m,n}^\dagger d_{m',n'+\sigma'} [s_{n+\sigma}^{-m}, s_{n'+\sigma'}^{m'}] s_{n+\sigma}^{-m} \\ + [d_{m,n}^\dagger, d_{m',n'}] s_{n+\sigma}^{-m} s_{n'+\sigma'}^{m'} + d_{m',n'} [d_{m,n}^\dagger, s_{n'+\sigma'}^{m'}] s_{n+\sigma}^{-m}\} + \frac{1}{16} \sum_{m,m',n} (-1)^{m+m'} \{d_{m,n}^\dagger [d_{-m,n+1}^\dagger, d_{m',n'+1}] d_{-m',n'+2} \\ + d_{m,n}^\dagger d_{m',n'} [d_{-m,n+1}^\dagger d_{-m',n'+1}] + [d_{m,n}^\dagger, d_{m',n'}] d_{-m',n'+1} d_{-m,n+1}^\dagger + d_{m',n'-1} [d_{m,n}^\dagger, d_{-m',n'+1}] d_{-m',n'+1}\}. \quad (5.22)$$

In Eq. (5.22) since we are considering only the one-exciton manifold, we need take only the projection onto the ground state of the commutators with superscripts 0. Thus we find that

$$(J/J'^2)\mathcal{H}_{\text{eff},2} = \frac{1}{8} \sum_{m,m',\sigma,n} (-1)^{m'} \{-\delta_{m,m'} s_{n+\sigma}^{-m} s_{n+\sigma}^{-m'} + d_{m',n'+\sigma'} [d_{m,n}^\dagger, s_{n'+\sigma'}^{m'}] s_{n+\sigma}^{-m}\} + \frac{1}{16} \sum_{m,m',n} (-1)^{m+m'} \\ \times \{-\delta_{-m,m'} c_{m,n}^\dagger c_{m,n+2} - \delta_{m,m'} c_{m,n}^\dagger c_{m,n} - [\delta_{m,m'}(1-N_n) - c_{m,n}^\dagger c_{m',n}] \delta_{m,m'}(1-N_{n+1}) - \delta_{m,-m'} c_{-m,n+1}^\dagger c_{-m,n-1}\}. \quad (5.23)$$

A short calculation leads to the result

$$(J/J'^2)\mathcal{H}_{\text{eff},2} = -\frac{3}{16}N + \sum_{m,k} c_{m,k}^\dagger c_{m,-k} \\ \times \left(-\frac{1}{8} - \frac{1}{2}\cos k - \frac{1}{8}\cos 2k\right). \quad (5.24)$$

C. Third-Order Result and Discussion

The evaluation of the terms of order J'^3/J^2 in the dispersion relation is performed via a similar, but more tedious, calculation which is given in detail in Appendix D. Here we summarize the calculation by giving the results for the various terms appearing in Eq. (5.11):

$$[V_{-1}, [V_0, V_1]] = \left(\frac{1}{2}J'\right)^3 \sum_{m,k} c_{m,k}^\dagger c_{m,k} (4\cos k + 4\cos 2k), \quad (5.25a)$$

$$[V_{-2}, [V_0, V_2]] = \left(\frac{1}{2}J'\right)^3 \left(-6N + \sum_{m,k} c_{m,k}^\dagger c_{m,k} \right. \\ \left. \times (12 + 8\cos k - 8\cos 2k - 4\cos 3k)\right), \quad (5.25b)$$

$$[V_1, [V_1, V_{-2}]] = -\left(\frac{1}{2}J'\right)^3 \sum_{m,k} c_{m,k}^\dagger c_{m,k} (4\cos k + 4\cos 2k). \quad (5.25c)$$

Using these evaluations we obtain the third-order contribution to \mathcal{H}_{eff} in Eq. (5.11) as

$$\mathcal{H}_{\text{eff},3} = -\frac{3NJ'^3}{64J^2} + \frac{J'^3}{32J^2} \sum_{m,k} c_{m,k}^\dagger c_{m,k} \\ \times (3 + 2\cos k - 2\cos 2k - \cos 3k). \quad (5.26)$$

Collecting the results of Eqs. (5.20), (5.24), and (5.26) we arrive at the following expansion for the exciton energy:

$$E_m(k)/2J = \left(1 - \frac{1}{16}\hat{j}'^2 + \frac{3}{64}\hat{j}'^3\right) - \left(\frac{1}{2}\hat{j}' + \frac{1}{4}\hat{j}'^2 - \frac{1}{32}\hat{j}'^3\right) \cos k \\ - \left(\frac{1}{16}\hat{j}'^2 + \frac{1}{32}\hat{j}'^3\right) \cos 2k - \frac{1}{64}\hat{j}'^3 \cos 3k, \quad (5.27a)$$

which can be written in the form

$$E_m(k)/2J = \left(1 + \frac{5}{64}\hat{j}'^3\right) - \left(\frac{1}{2}\hat{j}' + \frac{1}{4}\hat{j}'^2 - \frac{5}{64}\hat{j}'^3\right) \cos k \\ - \left(\frac{1}{8}\hat{j}'^2 + \frac{1}{16}\hat{j}'^3\right) \cos^2 k - \frac{1}{16}\hat{j}'^3 \cos^3 k. \quad (5.27b)$$

The bandwidth ΔE when $J' \ll J$ is then given as

$$\Delta E = E_m(\pi) - E_m(0) = 2J' \left(1 + \frac{1}{2}\hat{j}' - \frac{1}{32}\hat{j}'^2\right). \quad (5.28)$$

Using Eqs. (5.24) and (5.26) we obtain the ground-state energy to third order in (J'/J) as

$$(E_0/NJ) = -\frac{3}{2} - \frac{3}{16}\hat{j}'^2 - \frac{3}{64}\hat{j}'^3 + \dots \quad (5.29)$$

Several remarks should be made about these results. It may be of interest to compare Eq. (5.27) with what one would get by expanding the linear exciton result of Eq. (2.12):

$$E_m(k)/2J = 1 - \frac{1}{2}\hat{j}' \cos k - \frac{1}{8}\hat{j}'^2 \cos^2 k - \frac{1}{16}\hat{j}'^3 \cos^3 k + \dots \quad (5.30)$$

Thus, harmonic-exciton theory gives the coefficient of $\cos^n k$ correct to leading order in \hat{j}' , as can be proved by general arguments. Our results for $E_m(k)$ differ from those of Ref. 8, but since those

do not agree with Eq. (5.30) as to the coefficient of $(\hat{j}' \cos k)^n$, they are clearly incorrect.

Finally, note that even for $J'/J = \frac{1}{2}$, the terms in the above series decrease rapidly in magnitude, indicating that further terms may not be important for $|J'/J| \leq \frac{1}{2}$. The magnitudes of successive terms we find is not inconsistent with Brinkman's result¹⁷ mentioned earlier, that perturbation theory for short chains converges for $|J'/J| < 1$. In fact, a rather crude extrapolation for the series for the ground-state energy for $J' = J$ (i. e., $\hat{j}' = 1$) can be obtained by writing Eq. (5.29) as

$$\frac{E_0}{NJ} = -\frac{3}{2} - \frac{3}{16} \hat{j}'^2 \frac{1}{1 - \frac{1}{4} \hat{j}'^2} + \dots \quad (5.31)$$

For $\hat{j}' = 1$, this form gives

$$E_0/NJ = -\frac{3}{2} - \frac{1}{4} = -1.75, \quad (5.32)$$

which is rather close to the exact value²²

$$E_0/NJ = -1.77. \quad (5.33)$$

(Note that the chain has $2N$ spins.) This argument encourages us to attempt to extrapolate $E_m(k)$ to larger values of \hat{j}' . Since for the uniform antiferromagnet it is $E^2(k)$ which is analytic in $\cos k$, we have expressed our results as a series for it:

$$[E_m(0)/2J]^2 = 1 - \hat{j}'^2 - \frac{1}{2} \hat{j}'^4 + \frac{7}{16} \hat{j}'^6 + \dots \quad (5.34)$$

Since we know that $E(0) = 0$ for $\hat{j}' = 1$, we write

$$[E_m(0)/2J]^2 = (1 - \hat{j}') \Phi(\hat{j}'), \quad (5.35)$$

where

$$\Phi(\hat{j}') = 1 - \frac{1}{2} \hat{j}'^2 - \frac{1}{16} \hat{j}'^4 + \dots \quad (5.36)$$

This series for $\Phi(\hat{j}')$ may provide a good approximation for $E_m(0)$ for fairly large values of \hat{j}' .

VI. CONCLUSION

We have calculated the susceptibility of the alternating linear Heisenberg antiferromagnet as a function of temperature at low exciton density. This work essentially justifies the use of localized statistics, according to which

$$\frac{\chi T}{NC} = 3\rho N^{-1} \sum_k \frac{e^{\beta J' \cos k}}{1 + 3\rho e^{\beta J' \cos k}}, \quad (2.5a)$$

since our result is almost identical to what one would find by expanding Eq. (2.5a) in powers of ρ . The appropriateness of localized statistics is due to the fact that two excitons coupled into a total-spin-2 or total-spin-1 state interact repulsively. On the other hand, for total spin 0, the interactions are attractive and an attractive bound state occurs for all values of total momentum. Although these attractive bound states do not affect the susceptibility, they could be observed via a light scattering experiment. It would be interesting if our calculations could be verified in this way.

The question of the appropriateness of localized statistics in one dimension is studied in a general way.²³ For a hard-core interaction with repulsive or weakly attractive dynamical interactions localized statistics are shown to obtain at low temperatures. For strongly attractive interactions the two-particle properties are dominated by bound states which no independent-particle statistics can describe. At the boundary between these two regimes only small interference effects occur and Bose statistics are appropriate. Spin waves in a Heisenberg ferromagnet exemplify this latter case.

We have also calculated the single-exciton dispersion relation to order $(J'/J)^3$ and find

$$E_m(k) = (2J + 5J'^3/32J^2) - (J' + J'^2/2J - 5J'^3/32J^2) \cos k - (J'^2/4J + J'^3/8J^2) \cos^2 k - (J'^3/8J^2) \cos^3 k, \quad (6.1)$$

which yields an exciton bandwidth

$$\Delta E = 2J' + J'^2/J - J'^3/16J^2. \quad (6.2)$$

A similar expansion for the ground-state energy is given in Eq. (5.29). The magnitude of successive terms in these expressions is consistent with a radius of convergence $J'/J = 1$. Thus, these results may be useful for the case of tetramethylenediamine-tetracyanoquinodimethan (TMPD)-(TCNQ), where J'/J may be of order $\frac{1}{2}$.²⁴ Furthermore we have given extrapolations of some of our results into the region $J' \sim J$.

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APPENDIX A: USE OF BETHE'S ANSATZ

In this Appendix we show how the use of Bethe's ansatz¹⁴ leads to a convenient eigenvalue condition in the case $m = 2$. This formalism has been investigated in Ref. 3 to which the reader is referred for more details. For $m = 2$, we specify the two-exciton states by

$$|n_1, n_2\rangle \quad (A1)$$

with $n_1 < n_2$. Bethe's ansatz is to look for a solution to the Schrödinger equation of the form

$$\psi(k_1, k_2) = \sum_{n_1 < n_2} (e^{i(k_1 n_1 + k_2 n_2)} + A e^{i(k_1 n_2 + k_2 n_1)}) |n_1, n_2\rangle, \quad (A2a)$$

$$\psi(k_1, k_2) \equiv \sum_{n_1 < n_2} c(n_1, n_2) |n_1, n_2\rangle. \quad (\text{A2b})$$

Translational invariance dictates that $K = k_1 + k_2 = 2\pi m/N$. We set

$$k_1 = \frac{1}{2}K + p, \quad (\text{A3a})$$

$$k_2 = \frac{1}{2}K - p, \quad (\text{A3b})$$

and the first Brillouin zone can be considered to be

$$0 \leq K \leq 2\pi, \quad (\text{A4a})$$

$$-\pi \leq p \leq \pi. \quad (\text{A4b})$$

Further, we also have periodicity in the sense that

$$c(n_1, n_2) = c(n_2, n_1 + N). \quad (\text{A5})$$

It is this relation which is improperly handled in Eq. (10) of Ref. 8, and which invalidates that treatment. From Eq. (A5) and using that $e^{iN(k_1+k_2)} = e^{iNK} = 1$, we obtain $A = e^{-iNk_1}$. Thus

$$c(n_1, n_2) = e^{i(k_1 n_1 + k_2 n_2)} + e^{i(k_2 n_1 + k_1 n_2 - N k_1)}. \quad (\text{A6})$$

Now we substitute this ansatz into the Schrödinger equation. We note that if $n_2 - n_1 > 1$, then

$$\begin{aligned} & (\mathcal{H} - 4J) |n_1, n_2\rangle \\ &= -\frac{1}{2} J' \sum_{\sigma=\pm 1} (|n_1 + \sigma, n_2\rangle + |n_1, n_2 + \sigma\rangle), \end{aligned} \quad (\text{A7a})$$

whereas the case $n_2 - n_1 = 1$ yields

$$\begin{aligned} & (\mathcal{H} - 4J - \frac{1}{2} J') |n, n+1\rangle \\ &= -\frac{1}{2} J' (|n-1, n+1\rangle + |n, n+2\rangle). \end{aligned} \quad (\text{A7b})$$

We now study

$$\mathcal{H} \psi(k_1, k_2) = E_K(p) \psi(k_1, k_2). \quad (\text{A8})$$

In configurations with $n_2 - n_1 > 1$ Eq. (A7a) yields

$$E_K(p) + J' \cos k_1 + J' \cos k_2 = 0 \quad (\text{A9a})$$

or

$$E_K(p) = -2J' \cos \frac{1}{2}K \cos p. \quad (\text{A9b})$$

For configurations with $n_2 - n_1 = 1$, Eq. (A7b) yields

$$\left(\frac{1}{2} J' - E\right) c(n, n+1) = \frac{1}{2} J' [c(n-1, n+1) + c(n, n+2)] \quad (\text{A10})$$

or, by Eqs. (A6) and (A9b),

$$\begin{aligned} & \left(\frac{1}{2} + \cos k_1 + \cos k_2\right) (e^{i(k_1 n + k_2 n + k_2)} + e^{i(k_2 n + k_1 n + k_1 - N k_1)}) \\ &= \frac{1}{2} (e^{i(k_1 n - k_1 + k_2 n + k_2)} + e^{i(k_1 n + k_2 n + 2k_2)} \\ &+ e^{i(k_2 n - k_2 + k_1 n + k_1 - N k_1)} + e^{i(k_2 n + k_1 n + 2k_1 - N k_1)}). \end{aligned} \quad (\text{A11})$$

We may simplify this to the form

$$\cos p + \frac{\sin p}{i} \left(\frac{e^{iN(p+K/2)} - 1}{e^{iN(p+K/2)} + 1} \right) = -2 \cos \frac{1}{2}K, \quad (\text{A12})$$

which agrees with Ref. 3, where only the ground state was considered. For a given value of K , this relation gives the solution for p and hence for

$E_K(p)$.

The bound-state condition is obtained by looking for a solution with complex p . Assume that $\text{Im} p > 0$. Then Eq. (A12) in the limit $N \rightarrow \infty$ becomes

$$\cos p + i \sin p = -2 \cos \frac{1}{2}K \quad (\text{A13a})$$

or

$$e^{ip} = -2 \cos \frac{1}{2}K. \quad (\text{A13b})$$

Since $\text{Im} p > 0$, we see that a bound state occurs only for $|\cos \frac{1}{2}K| \leq \frac{1}{2}$. Then

$$E_K(p) = -2J' \cos \frac{1}{2}K \cos p = \frac{1}{2} J' (1 + 4 \cos^2 \frac{1}{2}K), \quad (\text{A14})$$

in accord with Eq. (3.25).

APPENDIX B: SOLUTION FOR THE $m=0$ t MATRIX

In this Appendix we give some details of the solution for the $m=0$ t matrix. If the two excitons in the intermediate state in Eq. (3.39) correspond to $n''=0$, then D will not appear in the energy denominator. Then we may define a reduced energy variable.

$$x_0 \equiv (z - 4J)/2J'. \quad (\text{B1a})$$

For the case $n''=1$, it is convenient to define

$$x_1 \equiv (z - 4J - 2D)/2J'. \quad (\text{B1b})$$

Correspondingly, and in analogy with Eq. (3.23), we define

$$r_n = (1 - \alpha^2/x_n^2)^{1/2}. \quad (\text{B2})$$

The odd-parity scattering, described by Γ_{00} in Eq. (3.40), leads to

$$\Gamma_{00} = 2\alpha^2(2\alpha^2 - x_1 + x_1 r_1)^{-1}. \quad (\text{B3})$$

For the even-parity scattering it is clear that Eq. (3.40) will lead to a set of inhomogeneous linear equations to determine the coefficients $A^{n,n'}$, etc. The derivation of these equations is straightforward, so we give only the result.

$$\underline{\mathbf{M}} \begin{bmatrix} A^{11} \\ B^{11} \\ A^{01} \\ B^{01} \end{bmatrix} = \begin{bmatrix} T \\ 2J'\alpha \\ 0 \\ 0 \end{bmatrix}, \quad (\text{B4a})$$

$$\underline{\mathbf{M}} \begin{bmatrix} A^{10} \\ B^{10} \\ A^{00} \\ B^{00} \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ T \\ 2J'\alpha \end{bmatrix}, \quad (\text{B4b})$$

$$\underline{\mathbf{M}} \begin{bmatrix} C^{11} \\ D^{11} \\ C^{01} \\ D^{01} \end{bmatrix} = \begin{bmatrix} 2J'\alpha \\ -J' \\ 0 \\ J' \end{bmatrix}, \quad (\text{B4c})$$

$$\underline{\mathbf{M}} \begin{bmatrix} C^{10} \\ D^{10} \\ C^{00} \\ D^{00} \end{bmatrix} = \begin{bmatrix} 0 \\ 2J' \\ 2J' \\ 0 \end{bmatrix}, \quad (\text{B4d})$$

where $\underline{\mathbf{M}}$ is the matrix

$$\begin{bmatrix} 1 - TI_0^1/2J' - \alpha I_1^1 & -TI_1^1/2J' - \alpha I_2^1 & 0 & 0 \\ -\alpha I_0^1 + \frac{1}{2}I_1^1 & 1 - \alpha I_1^1 + \frac{1}{2}I_2^1 & -I_1^0 & -I_2^0 \\ 0 & 0 & 1 - TI_0^0/2J' - \alpha I_1^0 & -TI_1^0/2J' - \alpha I_2^0 \\ -\frac{1}{2}I_1^1 & -\frac{1}{2}I_2^1 & -\alpha I_0^0 & 1 - \alpha I_1^0 \end{bmatrix}, \quad (B5)$$

where

$$I_n^m = N^{-1} \sum_{p''} \frac{\cos^{n p''}}{x_m + \alpha \cos p''}. \quad (B6)$$

Explicit expressions for the I_n^m 's can be obtained from Eq. (3.21), e.g., $I_0^0 = (x_0 r_0)^{-1}$, etc. The solutions to Eq. (B4) are conveniently formulated by giving the matrix \underline{M}^{-1} :

$$\begin{bmatrix} \left(1 - \frac{T}{2J'x_1}\right)^{-1} & \frac{-x_1(r_1-1)}{\alpha \Delta r_1 r_0} & 0 & \frac{x_1 x_0 (r_1-1)(r_0-1)}{\alpha^3 r_1 r_0 \Delta} \\ \frac{\alpha}{x_1 - T/2J'} & \frac{1}{\Delta r_1 r_0} & 0 & \frac{-x_0(r_0-1)}{\alpha^2 r_1 r_0 \Delta} \\ 0 & \frac{x_1 x_0 (r_1-1)(r_0-1)}{2\alpha^3 \Delta r_1 r_0} & \left(1 - \frac{T}{2J'x_0}\right)^{-1} & \frac{-x_0(r_0-1)[2\alpha^2 - x_1(r_1-1)]}{2\alpha^3 \Delta r_1 r_0} \\ 0 & \frac{-x_1(r_1-1)}{2\alpha^2 \Delta r_1 r_0} & \frac{\alpha}{x_0 - T/2J'} & \frac{2\alpha^2 - x_1(r_1-1)}{2\alpha^2 \Delta r_1 r_0} \end{bmatrix}, \quad (B7)$$

where

$$\Delta = \frac{1}{2\alpha^2 r_0 r_1} \left(2\alpha^2 - x_1(r_1-1) - \frac{x_1 x_0}{\alpha^2} (r_1-1)(r_0-1) \right). \quad (B8)$$

Note that T enters the result in an appropriate way: The bound state of two excitons on the same site occurs at an energy independent of J' and the continuum is unaffected by T .

The bound-state condition is that \underline{M}^{-1} be singular. In addition to the trivial case of two excitons on the same site, this condition is simply

$$\Delta = 0. \quad (B9)$$

In general this condition is rather complicated, and we have not analyzed it in detail. For the case $D=0$, the condition $\Delta=0$ is equivalent to

$$x_0(r_0-1) = \frac{1}{2}\alpha^2 \pm \frac{3}{2}\alpha^2. \quad (B10)$$

The choice of the lower sign gives Eq. (3.27) apart from a trivial shift of energy due to the external field. For the upper sign one finds the solution

$$x_0 = -\frac{1}{2}(1 + \alpha^2). \quad (B11)$$

In other words, for $m=0$, for all values of the total momentum there is an attractive bound state below the continuum. This state is shown in Fig. 2.

APPENDIX C: ANALYSIS OF Λ_c

We wish to reduce Λ_c to a sum of two- (or less) dimensional integrals. We write

$$\Lambda_c = \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} \Phi(\alpha, \gamma) \times \int_{-1}^1 dy \frac{(1-y^2)^{1/2}}{1+4\alpha^2-4\alpha y} \sum_{i=1,3} F_i(\gamma, y, \alpha), \quad (C1)$$

where

$$\Phi(\alpha, \gamma) = 3e^{-4\beta J + 2\beta J' \alpha \gamma} [5(2\alpha + \gamma)^2 + \gamma^2 - 1] \pi^{-3} \quad (C2)$$

and the F_i 's are the terms in the final bracket of Eq. (4.23). Following Bloomfield's suggestion,¹⁵ we consider the integrals I_i :

$$I_i = \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} \Phi(\alpha, \gamma) \times \oint_C \frac{iy dy}{2} \frac{(1-y^2)^{1/2}}{1+4\alpha^2-4\alpha y} F_i(\gamma, y, \alpha), \quad (C3)$$

where $(1-y^2)^{1/2}$ is defined like r in Eq. (3.23) for $\alpha=1$. The contour C , shown in Fig. 6, surrounds the branch cut in $(1-y^2)^{1/2}$ but not the simple pole at $y_0 = -(1+4\alpha^2)/4\alpha$. Now deform the contour C into the contour C' , shown in Fig. 6. It can be shown that the contribution from the circle of radius ϵ at $y = -\gamma$ vanishes in the limit $\epsilon \rightarrow 0$. Hence

$$I_i = \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} \Phi(\alpha, \gamma) \times \left(\int_{-1}^{-\gamma-\epsilon} + \int_{-\gamma+\epsilon}^1 \right) \frac{dy(1-y^2)^{1/2}}{1+4\alpha^2-4\alpha y} F_i(\gamma, y, \alpha), \quad (C4)$$

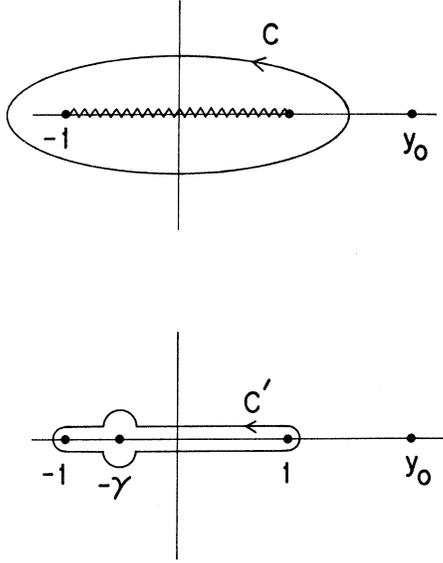


FIG. 6. The contours in the complex y plane C (top) and C' (bottom) used to evaluate the integral of Eq. (C3). The branch cut, shown as the saw-tooth line in the upper diagram, is omitted for clarity in the lower diagram. In the lower diagram the two semicircular arcs of radius ϵ surrounding $y = -\gamma$ are shown.

so that

$$\Lambda_c = \sum_{i=1,3} I_i. \quad (\text{C5})$$

Note that for I_i as defined in Eq. (C3) the integrand has no significant singularities on the contour. (The singularities at $\alpha=1$ or $\gamma=1$ are irrelevant in this context.) We therefore can interchange the order of the γ and y integrations at will. For $i=2$ and $i=3$ we do the y integrals first and use the principal-value representation of Eq. (C4). Then the evaluations of Eq. (4.25) apply and we obtain

$$I_2 = \frac{3}{\pi^2} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} e^{-4\beta J + 2\beta J' \alpha \gamma} \\ \times \frac{\beta J'}{\frac{1}{4} + \alpha^2 + \alpha \gamma} [5(2\alpha + \gamma)^2 + \gamma^2 - 1] \\ \times [\frac{1}{2} \alpha \gamma + \frac{1}{4} + (\alpha^2 - \frac{1}{4}) \theta(1 - 4\alpha^2)], \quad (\text{C6a})$$

$$I_3 = \frac{3}{\pi^2} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} e^{-4\beta J + 2\beta J' \alpha \gamma} \\ \times \frac{|4\alpha^2 - 1|}{(1 + 4\alpha^2 + 4\alpha \gamma)^2} [5(2\alpha + \gamma)^2 + \gamma^2 - 1]. \quad (\text{C6b})$$

For $i=1$ we use the representation of Eq. (C3) and do the y integral first:

$$I_1 = \frac{3}{\pi^3} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \oint_C \frac{iy dy}{2} \frac{(1-y^2)^{1/2}}{1 + 4\alpha^2 - 4\alpha y}$$

$$\times e^{-4\beta J - 2\beta J' \alpha \gamma} \int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} \frac{[5(2\alpha + \gamma)^2 + \gamma^2 - 1]}{(\gamma + y)^2}. \quad (\text{C7})$$

To do the γ integral we use the following evaluations which are valid for complex y :

$$\int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} = \pi, \quad (\text{C8a})$$

$$\int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} (y + \gamma)^{-1} = \frac{\pi}{y(1-y^2)^{1/2}}, \quad (\text{C8b})$$

$$\int_{-1}^1 \frac{d\gamma}{(1-\gamma^2)^{1/2}} (y + \gamma)^{-2} = \frac{\pi}{(y^2 - 1)(1-y^2)^{1/2}}, \quad (\text{C8c})$$

where the radical is defined as above. Thus we obtain

$$I_1 = \frac{3}{\pi^2} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \oint_C \frac{iy dy}{2} \frac{(1-y^2)^{1/2}}{1 + 4\alpha^2 - 4\alpha y} \\ \times \left(6 + \frac{-12y + 20\alpha}{y(1-y^2)^{1/2}} + \frac{5(2\alpha - y)^2 + y^2 - 1}{(y^2 - 1)(1-y^2)^{1/2}} \right) \\ e^{-4\beta J - 2\beta J' \alpha y}. \quad (\text{C9})$$

The result is then

$$e^{4\beta J} I_1 = -\frac{15}{\pi} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} e^{2\beta J' \alpha} \\ + \frac{18}{\pi^2} \int_{-1}^1 \frac{d\alpha}{(1-\alpha^2)^{1/2}} \int_{-1}^1 \frac{dx(1-x^2)^{1/2}}{1 + 4\alpha x + 4\alpha^2} e^{2\beta J' \alpha x}. \quad (\text{C10})$$

The first term in the final bracket of Eq. (C9) yields the two-dimensional integral in Eq. (C10), the second term vanishes, and the third term, having simple pole at $y = \pm 1$, yields the one-dimensional integral. It is this latter contribution which is very easy to lose, if one attempts to interchange principal-value integrations in an ill-defined way. Combining the results of Eqs. (C6) and (C10) yields Eq. (4.26) of the text.

APPENDIX D: EVALUATION OF THIRD-ORDER TERMS IN THE SINGLE-EXCITON DISPERSION RELATION

Referring to Eq. (5.11) we see that a third-order calculation requires the construction of $\Theta_A = [V_{-2}, [V_0, V_2]]$. The ground-state matrix element of Θ_A is simply

$$\langle 0 | \Theta_A | 0 \rangle = \langle 0 | V_{-2} V_0 V_2 | 0 \rangle, \quad (\text{D1})$$

inasmuch as $V_0 | 0 \rangle = 0$. Using Eq. (5.15) for V_2 we have that

$$\langle 0 | \Theta_A | 0 \rangle = \frac{1}{2} J' \langle 0 | V_{-2} V_0 \left(\sum_n |1, n; -1, n+1\rangle \right. \\ \left. - |0, n; 0, n+1\rangle + |-1, n; 1, n+1\rangle \right), \quad (\text{D2})$$

where $|m, n; m', n'\rangle \equiv c_{m,n}^\dagger c_{m',n'} |0\rangle$; this notation will later be generalized in the obvious way. Note

that V_0 as given in Eq. (5.15c) has two kinds of terms. The first term, which causes excitations to hop from site to site, can be dropped because it creates states which do not intersect $\langle 0 | V_{-2}$. Including only the effect of the second term in Eq. (5.15c) we have

$$\langle 0 | \epsilon_A | 0 \rangle = (\frac{1}{2}J')^2 \langle 0 | V_{-2} \left(\sum_n -2 | 1, n; -1, n+1 \rangle + 2 | 0, n; 0, n+1 \rangle - 2 | -1, n; 1, n+1 \rangle \right), \quad (D3a)$$

$$\langle 0 | \Theta_A | 0 \rangle = -6N(\frac{1}{2}J')^3. \quad (D3b)$$

Now we consider the effect of Θ_A on a one-exciton state. Due to the rotational invariance of the Hamiltonian, we only need to study the effect of Θ_A on an ($S^z=1$) exciton. Also, effectively we have $\Theta_A = V_{-2}[V_0, V_2]$, because $V_{-2}|1, n\rangle=0$. Since V_2 creates pairs of excitons on adjacent sites, we have

$$V_{-2}V_0V_2|1, n\rangle = (\frac{1}{2}J')V_{-2}V_0 \sum_{\substack{n' \geq n+1 \\ n' \leq n-2}} (|1, n'; -1, n'+1; 1, n\rangle - |0, n'; 0, n'+1; 1, n\rangle + |-1, n'; 1, n'+1; 1, n\rangle). \quad (D4)$$

Next we compute that

$$\begin{aligned} V_{-2}V_0V_2 &= (\frac{1}{2}J')^2 V_{-2} \sum_{\substack{n' \geq n+2 \\ n' \leq n-3}} \sum_m (-1)^m (2|1, n; m, n'; -m, n'+1\rangle + |1, n-1; m, n'; -m, n'+1\rangle \\ &+ |1, n+1; m, n'; -m, n'+1\rangle + |1, n; m, n'-1; -m, n'+1\rangle + |1, n; m, n'; -m, n'+2\rangle) \\ &+ (\frac{1}{2}J')V_{-2} \sum_m (-1)^m (|1, n-1; m, n+1; -m, n+2\rangle + |1, n; m, n+1; -m, n+3\rangle \\ &+ |1, n+1; m, n-1; -m, n-2\rangle + |1, n; m, n-1, -m, n-3\rangle) \\ &+ (\frac{1}{2}J')^2 V_{-2} (-3|1, n; -1, n+1; 1, n+2\rangle + 2|1, n; 0, n+1; 0, n+2\rangle - |0, n; 1, n+1; 0, n+2\rangle \\ &- |1, n; 1, n+1; -1, n+2\rangle + |0, n; 0, n+1, 1, n+2\rangle - 3|1, n; -1, n-1; 1, n-2\rangle + 2|1, n; 0, n-1; 0, n-2\rangle \\ &- |0, n; 1, n-1; 0, n-2\rangle - |1, n; 1, n-1; -1, n-2\rangle + |0, n; 0, n-1; 1, n-2\rangle), \quad (D5) \end{aligned}$$

which finally leads to the evaluation

$$\begin{aligned} V_{-2}V_0V_2|1, n\rangle &= (\frac{1}{2}J')^3 [(-6N+12)|1, n\rangle + (-3N+9)(|1, n+1\rangle + |1, n-1\rangle) \\ &- 4(|1, n+2\rangle + |1, n-2\rangle) - 3(|1, n+3\rangle + |1, n-3\rangle)]. \quad (D6) \end{aligned}$$

Likewise we have

$$V_{-2}V_2V_0|1, n\rangle = (\frac{1}{2}J')V_{-2}V_2(-|1, n-1\rangle - |1, n+1\rangle), \quad (D7a)$$

$$\begin{aligned} V_{-2}V_2V_0|1, n\rangle &= (\frac{1}{2}J')^2 V_{-2} \sum_{\substack{n' \geq n \\ n' \leq n-3}} (-|1, n'; -1, n'+1; 1, n-1\rangle + |0, n'; 0, n'+1; 1, n-1\rangle \\ &- |-1, n'; 1, n'+1; 1, n-1\rangle) + \sum_{\substack{n' \geq n+2 \\ n' \leq n-1}} (-|1, n'; -1, n'+1; 1, n+1\rangle \\ &+ |0, n'; 0, n'+1; 1, n+1\rangle - |-1, n'; 1, n'+1; 1, n+1\rangle), \quad (D7b) \end{aligned}$$

$$V_{-2}V_2V_0|1, n\rangle = (\frac{1}{2}J')^3 [(-3N+5)(|1, n+1\rangle + |1, n-1\rangle) - |1, n+3\rangle - |1, n-3\rangle]. \quad (D7c)$$

Combining Eqs. (D6) and (D7c) we may write

$$\begin{aligned} [V_{-2}, [V_0, V_2]]|1, n\rangle &= (\frac{1}{2}J')^3 [(-6N+12)|1, n\rangle \\ &+ 4(|1, n+1\rangle + |1, n-1\rangle) - 4(|1, n+2\rangle + |1, n-2\rangle) - 2(|1, n+3\rangle + |1, n-3\rangle)], \quad (D8a) \end{aligned}$$

and in view of Eq. (D3b) we can express Eq. (D8a) as

$$[V_{-2}, [V_0, V_2]] = (\frac{1}{2}J')^3 \left(-6N + \sum_{m,k} c_{m,k}^* c_{m,k} (12 + 8\cos k - 8\cos 2k - 4\cos 3k) \right). \quad (D8b)$$

Next we study $\mathcal{O}_B \equiv [V_{-1}, [V_0, V_1]]$. Since none of these operators have nonzero matrix elements connecting to the ground state, we have effectively that $\mathcal{O}_B = V_{-1}[V_0, V_1]$. As before, we evaluate \mathcal{O}_B acting on $|1, n\rangle$. We have that

$$V_{-1}V_0V_1|1, n\rangle = (\frac{1}{2}J')V_{-1}V_0(|1, n-1; 0, n\rangle - |0, n-1; 1, n\rangle - |1, n+1; 0, n\rangle + |0, n+1; 1, n\rangle). \quad (D9)$$

Note that V_{-1} can destroy an exciton only if two neighboring sites are occupied. This fact implies that we can drop those terms in V_0 causing exciton hopping. Thus,

$$V_{-1}V_0V_1|1, n\rangle = (\frac{1}{2}J')^2V_{-1}(|0, n-1; 1, n\rangle - |1, n-1; 0, n\rangle - |0, n+1; 1, n\rangle + |1, n+1; 0, n\rangle), \quad (D10a)$$

$$V_{-1}V_0V_1|1, n\rangle = -(\frac{1}{2}J')^3(2|1, n+1\rangle + 4|1, n\rangle + 2|1, n-1\rangle). \quad (D10b)$$

The other term in \mathcal{O}_B is

$$V_{-1}V_1V_0|1, n\rangle = (\frac{1}{2}J')V_{-1}V_1(-|1, n-1\rangle - |1, n+1\rangle), \quad (D11a)$$

$$V_{-1}V_1V_0|1, n\rangle = (\frac{1}{2}J')^2V_{-1}(-|1, n-2; 0, n-1\rangle + |0, n-2; 1, n-1\rangle + |1, n; 0, n-1\rangle - |0, n; 1, n-1\rangle - |1, n; 0, n+1\rangle + |0, n; 1, n+1\rangle + |1, n+2; 0, n+1\rangle - |0, n+2; 1, n+1\rangle), \quad (D11b)$$

$$V_{-1}V_1V_0|1, n\rangle = -2(\frac{1}{2}J')^3(|1, n-2\rangle + 2|1, n-1\rangle + 2|1, n\rangle + 2|1, n+1\rangle + |1, n+2\rangle). \quad (D11c)$$

Combining Eqs. (D10) and (D11) we have that

$$[V_{-1}, [V_0, V_1]]|1, n\rangle = (\frac{1}{2}J')^3(2|1, n-2\rangle + 2|1, n-1\rangle + 2|1, n+1\rangle + 2|1, n+2\rangle), \quad (D12)$$

so that, for one or fewer excitons we may write

$$[V_{-1}, [V_0, V_1]] = (\frac{1}{2}J')^3 \sum_{m,k} c_{m,k}^\dagger c_{m,k} (4\cos k + 4\cos 2k). \quad (D13)$$

Finally, we evaluate $\mathcal{O}_C = [V_1, [V_1, V_{-2}]]$. Since V_{-1} does not connect to the ground state, we have effectively that $\mathcal{O}_C = V_{-1}V_{-1}V_2$. Also, in computing $\mathcal{O}_C|1, n\rangle$, we need only consider the terms where V_2 creates excitons next to the n th site. Other states created by V_2 will be annihilated by $V_{-1}V_{-1}$. Hence

$$\mathcal{O}_C|1, n\rangle = (\frac{1}{2}J')V_{-1}V_{-1}(|1, n-2; -1, n-1; 1, n\rangle - |0, n-2; 0, n-1; 1, n\rangle + |-1, n-2; 1, n-1; 1, n\rangle + |1, n; 1, n+1; -1, n+2\rangle - |1, n; 0, n+1; 0, n+2\rangle + |1, n; -1, n+1; 1, n+2\rangle) \quad (D14a)$$

$$\mathcal{O}_C|1, n\rangle = (\frac{1}{2}J')^2V_{-1}(-|1, n-2; 0, n-1\rangle - |1, n+1; 0, n+2\rangle + |0, n+1; 1, n+2\rangle + |0, n-2; 1, n-1\rangle), \quad (D14b)$$

$$\mathcal{O}_C|1, n\rangle = -2(\frac{1}{2}J')^3(|1, n-2\rangle + |1, n-1\rangle + |1, n+1\rangle + |1, n+2\rangle). \quad (D14c)$$

Thus we may write

$$[V_1, [V_1, V_{-2}]] = -(\frac{1}{2}J')^3 \sum_{m,k} c_{m,k}^\dagger c_{m,k} (4\cos k + 4\cos 2k). \quad (D15)$$

The results of Eqs. (D8b), (D13), and (D15) are Eqs. (5.25).

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Decay of Order in Classical Many-Body Systems. III. Ising Model at Low Temperature*

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In this work the decay of correlation in the d -dimensional Ising model is studied at low temperatures as a function of dimensionality of the lattice and magnetic field h . Except for the special case of the two-dimensional zero-field nearest-neighbor lattices, the decay of correlation verifies the Ornstein-Zernike prediction $G_{AB}(\vec{R}) \approx D_{AB}(d, h)R^{-(d-1)/2}e^{-\kappa R}$. For the two-dimensional zero-field case, the Ornstein-Zernike form is replaced by the "anomalous" form $G_{AB}(\vec{R}) \approx D_{AB}R^{-2}e^{-\kappa R}$. This "anomalous" result is shown to arise from the peculiarities of the spectrum of the transfer matrix in this case and is replaced by the Ornstein-Zernike result when further-neighbor forces are present. The results presented herein agree with the previously obtained exact results for the zero-field two-dimensional Ising model.

I. INTRODUCTION

In the first two papers of this series^{1,2} (hereafter referred to as I and II, respectively) the transfer-matrix approach to classical statistical mechanics was developed in a general framework¹ and applied to a study of the decay of pair correlation functions in the d -dimensional Ising model at high temperatures.² It was found that an arbitrary pair correlation function defined on the system decays as

$$G_{LQ}(\vec{R}) \equiv \langle \delta \underline{L}(\vec{F}) \delta \underline{Q}(\vec{F} + \vec{R}) \rangle \\ \approx (A_0 + A_1 R^{-1} + \dots) R^{-(d-1)/2} e^{-\kappa R} \\ + (B_0 + B_1 R^{-1} + \dots) R^{-d} e^{-2\kappa R} + \dots, \quad (1.1)$$

where A_n and B_n factor as $C_n(\underline{L})C_n(\underline{Q})$ and $D_n(\underline{L})D_n(\underline{Q})$, respectively. If \underline{L} is an operator composed of an odd number of closely spaced spins, the coefficients $C_n(\underline{L})$ tend to a finite limit as the magnetic field h tends to zero, while the coefficients $D_n(\underline{L})$ tend to zero. On the other hand, if \underline{L} is composed of an even number of such spins, the coefficients $D_n(\underline{L})$ remain finite and the coefficients $C_n(\underline{L})$ tend to zero as h tends to zero.² The first series of terms corresponds to the Ornstein-Zernike (OZ) result, while the second series is the leading correction to it.

In this paper the analysis of such correlation functions is extended to the d -dimensional Ising model at low temperatures. This problem is both more interesting and more difficult than the high-temperature analysis—more interesting because one is able to treat the spontaneously ordered system, more difficult because of the increased complexity of the transfer-matrix spectrum at low temperatures. Indeed, a major impetus for this work was the interest in understanding the "anomalous" decay of correlation in the two-dimensional model below the critical point.³ That is, whereas at high temperatures the spin-pair correlation function $G_S(\vec{R})$ decays as

$$G_S(\vec{R}) \sim R^{-1/2} e^{-\kappa R}, \quad (1.2)$$

in agreement with the OZ hypothesis,^{2,4} at low temperatures it is found that^{3,4}

$$G_S(\vec{R}) \sim R^{-2} e^{-\kappa R}, \quad (1.3)$$

which does not verify the OZ prediction. However, (1.3) has the form of the first-correction term to the OZ result in (1.1) if 2κ is replaced by κ . The OZ term in (1.1) arises from the single-particle band of the transfer-matrix spectrum,² and the second term arises from the two-particle band.² Thus, one is tempted to speculate that (1.3) reflects the absence of effects due to single-