Excitations and long-range order in quadrupolar-coupled system at zero temperature

I. P. Fittipaldi*†
Instituto de Física, Universidade Federal de Pernambuco, 50.000 Recife, Brazil and Department of Physics, Temple University, Philadelphia, Pennsylvania 19122

Raza A. Tahir-Kheli†
Department of Physics, Temple University, Philadelphia, Pennsylvania 19122
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A spin-1 system with isotropic quadrupolar coupling is analyzed by the method of double-time Green's functions. A consistent decoupling procedure, which exactly preserves two important spin-correlation sum rules, is constructed. The resulting self-consistent integral expressions are solved numerically for the case of a simple-cubic lattice with nearest-neighbor coupling and the ground-state energy, the long-range-order parameter, and the dispersion relation of the elementary excitations are evaluated. Our result for the ground-state energy is found to be somewhat lower than that obtained by using the decoupling procedures of Raich-Etters and Barma. On the other hand, our result for the zero-point defect for the quadrupolar long-range order is very slightly greater than that given by the Raich and Etters's random-phase-approximation (RPA) procedure but is distinctly smaller than that found by properly evaluating the expressions obtained by using the Barma procedure. Similarly, our results for the elementary excitation dispersion are also closer to the RPA predictions than those obtained via the Barma procedure.

I. INTRODUCTION

Much like the isotropic Heisenberg antiferromagnet, the ground state of the simplest of the dynamical quadrupolar systems is not exactly known. Here the mean-field-theory prediction of a perfectly ordered ground state, with Q equal to -1, where

$$Q=\tfrac{3}{2}[\langle(S_i^z)^2\rangle-\tfrac{2}{3}]\,, \tag{1.1}$$

is as suspect as the corresponding prediction of saturated sublattice magnetization for the antiferromagnet. The reason, of course, is that the relevant mean-field state is not an eigenfunction of the Hamiltonian and as such zero-point fluctuations are expected, which give rise to a finite zero-point defect for the order parameter.

A variety of physical situations can be described in terms of effective quadrupolar coupling.[1,2] Our interest in this paper is, however, to examine the ground-state properties of the simplest of these (dynamical) Hamiltonians. Such a Hamiltonian is described in Sec. II. In order to study its dynamics, double-time Green's functions are introduced and their equations of motion given in Sec. III.

In Sec. IV, we describe the Raich and Etters[3] and the Barma[1,2] approximations. The crucial role played by the conservation of the off-diagonal self-correlation sum rule has been clarified by Barma.[1,2] The inconsistencies that appear in random-phase-approximation (RPA) theories, which are nonconserving in such a sense, can be rendered relatively harmless by defining a symmetrized version of the RPA. Interestingly, the Raich and Etters[3] approximation is equivalent to just such a symmetrized RPA. The Barma decoupling,[1,2] in this context, can be viewed as an improved approximation which corrects the nonconservation of the crucial off-diagonal sum rule.

To improve the decoupling approximation still further, a higher-order correlation-function sum rule is then identified and finally, in Sec. IV, the resulting equations are recorded and their solution is presented in Sec. V.

In the concluding section, VI, the philosophy of the correlation-function sum rule conserving decouplings of Barma and that used in the present paper is discussed with reference to the ground-state energy. It is shown that the symmetrized RPA estimate for the ground-state energy is a few percent lower than that given by the molecular-field theory. When the ground-state energy is computed self-consistently within the Barma scheme, we find it to be slightly lower than the corresponding prediction of the symmetrized RPA. The interesting feature of this result is that while the lowering of the energy, from the RPA result, is relatively quite small, within the Barma scheme the relative increase in the elementary excitation dispersion relation as well as in the zero-point defect of the long-range-order parameter (LRO) is rather substantial. Finally, within the decoupling approximation of the present paper—which in addition to conserving the off-diagonal self-correlation sum rule also preserves a correlation function sum rule associated with a higher-order frequency moment of the basic spectral function—we find that the ground-state energy estimate is the lowest of the four approximations. Yet, the results for the elementary excitation energy as well

as the zero-point defect in the quadrupolar LRO are found to be very close to those given by the symmetrized RPA. These features are reminiscent of the behavior of oscillatory, though convergent, successive approximations.

II. HAMILTONIAN

Following Barma,[1,2] we study the simplest of the quadrupolar dynamical Hamiltonians, i.e., the isotropic nearest-neighbor Hamiltonian in the spin-1 subspace

$$\mathcal{H} = -\sum_{i<j} J_{ij} \sum_{m=-2}^{2} L_{2,m}(i) L_{2,-m}(j) , \tag{2.1}$$

where

$$L_{2,-m}(i) = [L_{2,m}(i)]^{\dagger} , \tag{2.2a}$$

$$L_{2,0}(i) = \sqrt{\tfrac{3}{2}}[(S_i^z)^2 - \tfrac{2}{3}] , \tag{2.2b}$$

$$L_{2,1}(i) = \tfrac{1}{2}(S_i^z S_i^+ + S_i^+ S_i^z) , \tag{2.2c}$$

$$L_{2,2}(i) = \tfrac{1}{2}(S_i^+)^2 , \tag{2.2d}$$

and where

$$J_{ij} = \begin{cases} J_{ji} = J > 0 \text{ when } i \text{ and } j \text{ are nearest neighbors} \\ 0 \text{ otherwise.} \end{cases} \tag{2.2e}$$

The lattice sum in Eq. (2.1) is over the N sites such that each neighboring pair is counted only once.

Within a mean-field approximation,[4] the ground state of this system is completely ordered and the order parameter,

$$Q = \sqrt{\tfrac{3}{2}} \langle L_{2,0}(i) \rangle , \tag{2.3}$$

attains its minimum value of -1. Because such an ordered state breaks the rotational symmetry of the system, we expect to find a Goldstone soft mode. Because the total order parameter $L_{2,0}(K=0) = \sum_i L_{2,0}(i)$ does not commute with the Hamiltonian, hence a long-wavelength behavior of the type $E_K \propto |K|^{1+\epsilon}$, $\epsilon \geq 0$, is consistent with Bogolyubov's inequality.[5]

Following Raich and Etters[3] (and as in the recent work of Barma), we introduce a set of creation and annihilation operators which are particularly useful for dealing with spin algebra of $S=1$ and are natural operators for describing creation and annihilation of excitations in quadrupolar systems,

$$a_i^{\dagger} = (1/\sqrt{2}) S_i^+ [1 - (S_i^z)^2], \quad a_i = (a_i^{\dagger})^{\dagger} , \tag{2.4}$$

$$b_i^{\dagger} = (1/\sqrt{2}) S_i^- [1 - (S_i^z)^2], \quad b_i = (b_i^{\dagger})^{\dagger} . \tag{2.5}$$

These operators obey the following commutation relations:

$$\begin{aligned} &[a_i, a_j^{\dagger}]_- = (1 - 2n_i - m_i)\delta_{ij} , \\ &[b_i, b_j^{\dagger}]_- = (1 - n_i - 2m_i)\delta_{ij} , \\ &[a_i, b_j^{\dagger}]_- = -b_i^{\dagger} a_i \delta_{ij} , \\ &[b_i, a_j^{\dagger}]_- = -a_i^{\dagger} b_i \delta_{ij} , \\ &[a_i, b_j]_- = [a_i^{\dagger}, b_j^{\dagger}]_- = 0 , \end{aligned} \tag{2.6}$$

where

$$n_i = a_i^{\dagger} a_i , \quad m_i = b_i^{\dagger} b_i . \tag{2.7}$$

(The operators n and m are readily identified to be the number operators for excitations with $M_S = +1$ and $M_S = -1$.)

A basic property of the spin-1 subspace is the following:

$$a_i a_i = b_i b_i = a_i^{\dagger} a_i^{\dagger} = b_i^{\dagger} b_i^{\dagger} = 0 , \tag{2.8a}$$

$$a_i b_i = a_i^{\dagger} b_i^{\dagger} = 0 . \tag{2.8b}$$

Using these operators, the quadrupolar Hamiltonian takes the form

$$\begin{aligned} \mathcal{H} = -\sum_{i,j} J_{ij} \{ &\tfrac{3}{4}[(n_i + m_i) - \tfrac{2}{3}][(n_j + m_j) - \tfrac{2}{3}] \\ &+ \tfrac{1}{2}(a_i^{\dagger} - b_i)(a_j - b_j^{\dagger}) + a_i^{\dagger} b_i b_j^{\dagger} a_j \} . \end{aligned} \tag{2.9}$$

Note that in terms of the a and b operators, the various spin operators are

$$\begin{aligned} &S_i^z = n_i - m_i , \\ &(S_i^z)^2 = n_i + m_i , \\ &S_i^+ = \sqrt{2}(a_i^{\dagger} + b_i) , \\ &S_i^- = \sqrt{2}(a_i + b_i^{\dagger}) . \end{aligned} \tag{2.10}$$

III. GREEN'S FUNCTIONS

In order to analyze the dynamics of the system represented by the Hamiltonian (2.9), we study, the following Green's functions:

$$\langle\langle a_i^{\dagger}(t); a_j(0) \rangle\rangle \equiv G_{ij}^{a^{\dagger}a}(t) , \tag{3.1a}$$

$$\langle\langle b_i(t); a_j(0) \rangle\rangle \equiv G_{ij}^{ba}(t) , \tag{3.1b}$$

$$\langle\langle b_i(t); b_j^{\dagger}(0) \rangle\rangle \equiv G_{ij}^{bb^{\dagger}}(t) , \tag{3.1c}$$

$$\langle\langle a_i^{\dagger}(t); b_j^{\dagger}(0) \rangle\rangle \equiv G_{ij}^{a^{\dagger}b^{\dagger}}(t) . \tag{3.1d}$$

Here we have used the following notation:

$$\begin{aligned} G_{ij}^{AB}(t) &\equiv \langle\langle A_i(t); B_j(0) \rangle\rangle \\ &= -2\pi i \Theta(t) \langle [A_i(t), B_j(0)]_- \rangle , \end{aligned} \tag{3.2}$$

$$\langle \cdots \rangle = \frac{\mathrm{Tr}(e^{-\beta\mathcal{H}} \cdots)}{\mathrm{Tr}(e^{-\beta\mathcal{H}})} , \tag{3.3}$$

$$A(t) = e^{i\mathcal{H}t} A(0) e^{-i\mathcal{H}t} , \tag{3.4}$$

$\Theta(t)$, as usual, denotes the Heaviside unit step function which is unity for $t > 0$ and is zero otherwise.

Whenever convenient we shall use a Fourier-transformed representation such that

$$G_{ij}^{AB}(t) = \int_{-\infty}^{+\infty} G_{ij}^{AB}(E) e^{-iEt}\, dE \ , \tag{3.5a}$$

$$G_{ij}^{AB}(E) \equiv \langle\langle A_i ; B_j \rangle\rangle_E \ . \tag{3.5b}$$

Consequently[6]

$$\langle B_j(0) A_i(t) \rangle = \lim_{\epsilon \to +0} (-1/\pi) \times \mathrm{Im} \int_{-\infty}^{+\infty} \frac{G_{ij}^{AB}(\omega + i\epsilon)\, e^{-i\omega t}\, d\omega}{e^{\beta\omega} - 1} \ . \tag{3.6}$$

Here $\beta = (k_B T)^{-1}$. Because in the present work we focus our attention on the ground state, which is thermodynamically relevant only at $T = 0$, we will proceed to the limit $\beta \to \infty$, once the expressions for the correlation functions have been obtained.

The energy Fourier transform of the equations of motion of the various Green's functions are readily obtained from the relation

$$G_{ij}^{AB}(E) = \langle [A_i, B_j]_- \rangle + G_{ij}^{CB}(E) \ , \tag{3.7a}$$

where

$$C_i = [A_i, \mathcal{H}]_- . \tag{3.7b}$$

We get

$$E G_{ij}^{a^\dagger a}(E) = (2\bar{n} + \bar{m} - 1)\delta_{ij} + \Gamma_{ij}^{a^\dagger ; a}(E) \ , \tag{3.8a}$$

with the notation:

$$\langle \Omega_i \rangle = \bar{\Omega} \ , \tag{3.8b}$$

$$\Gamma_{ij}^{a_i^\dagger ; a}(E) = \sum_p J_{pi} \{ \tfrac{3}{2} [\langle\langle a_i^\dagger (n_p + m_p) ; a_j \rangle\rangle_E - \tfrac{2}{3} G_{ij}^{a_i^\dagger a}(E)] + \tfrac{1}{2} [G_{pj}^{a_i^\dagger a}(E) - 2\langle\langle a_p^\dagger n_i ; a_j \rangle\rangle_E - \langle\langle a_p^\dagger m_i ; a_j \rangle\rangle_E]$$
$$- \tfrac{1}{2} [G_{pj}^{ba}(E) - 2\langle\langle b_p n_i ; a_j \rangle\rangle_E - \langle\langle b_p m_i ; a_j \rangle\rangle_E] + \tfrac{1}{2} \langle\langle a_i^\dagger b_i a_p ; a_j \rangle\rangle_E - \tfrac{1}{2} \langle\langle a_i^\dagger b_i b_p^\dagger ; a_j \rangle\rangle_E + \langle\langle a_p^\dagger b_p b_i^\dagger ; a_j \rangle\rangle_E \} \ . \tag{3.8c}$$

Similarly, we find

$$E G_{ij}^{b b^\dagger}(E) = (1 - \bar{n} - 2\bar{m})\delta_{ij} + \Gamma_{ij}^{b_i ; b^\dagger}(E) \ , \tag{3.9a}$$

$$\Gamma_{ij}^{b_i ; b^\dagger}(E) = \sum_p J_{pi} \{ -\tfrac{3}{2} [\langle\langle b_i (n_p + m_p) ; b_j^\dagger \rangle\rangle_E - \tfrac{2}{3} G_{ij}^{b b^\dagger}(E)] + \tfrac{1}{2} [G_{pj}^{a_i^\dagger b^\dagger}(E) - \langle\langle a_p^\dagger n_i ; b_j^\dagger \rangle\rangle_E - 2\langle\langle a_p^\dagger m_i ; b_j^\dagger \rangle\rangle_E]$$
$$- \tfrac{1}{2} [G_{pj}^{b b^\dagger}(E) - \langle\langle b_p n_i ; b_j^\dagger \rangle\rangle_E - 2\langle\langle b_p m_i ; b_j^\dagger \rangle\rangle_E] + \tfrac{1}{2} \langle\langle a_i^\dagger b_i a_p ; b_j^\dagger \rangle\rangle_E - \tfrac{1}{2} \langle\langle a_i^\dagger b_i b_p^\dagger ; b_j^\dagger \rangle\rangle_E - \langle\langle a_p^\dagger b_p a_i ; b_j^\dagger \rangle\rangle_E \} \ , \tag{3.9b}$$

$$E G_{ij}^{a_i^\dagger b^\dagger}(E) = \Gamma_{ij}^{a_i^\dagger ; b^\dagger}(E) \ , \tag{3.10}$$

$$E G_{ij}^{ba}(E) = \Gamma_{ij}^{b_i a}(E) \ . \tag{3.11}$$

[Note that expressions for $\Gamma_{ij}^{a_i^\dagger ; b^\dagger}(E)$ and $\Gamma_{ij}^{b_i ; a}(E)$ are identical with those of $\Gamma_{ij}^{a_i^\dagger ; a}(E)$ and $\Gamma_{ij}^{b_i ; b^\dagger}(E)$, given in Eqs. (3.8c) and (3.9b), except for the substitution of the appropriate operator on the right of the semicolon in the superfix.]

IV. DECOUPLING

The standard RPA type of decoupling for the various higher-order Green's functions contained in the functions $\Gamma_{ij}^{A_i ; B}(E)$, where $A \equiv a^\dagger, b$ and $B \equiv a, b^\dagger$ is of the following form:

$$\langle\langle A_i W_p ; B_j \rangle\rangle_E = \bar{W} \langle\langle A_i ; B_j \rangle\rangle_E \ , \quad i \neq p \ , \tag{4.1a}$$

$$\langle\langle a_i^\dagger b_i C_p ; B_j \rangle\rangle_E = 0, \quad i \neq p \tag{4.1b}$$

where

$$W_p \equiv n_p, m_p ; \quad C_p \equiv a_p, b_p^\dagger \ . \tag{4.1c}$$

A theory based on such a decoupling cannot conserve the off-diagonal self-correlation sum rule referred to earlier and consequently is subject to inconsistency and nonuniqueness.[1,2]

To remedy this problem, we follow Barma,[1,2] who introduced a Callen-like[7] procedure from which a suitable, conserving decoupling was extracted. We proceed as follows: From the first two commutators given in Eq. (2.6), we can readily obtain the following identities:

$$\begin{aligned} n_i + m_i &= \alpha (n_i + m_i) - \tfrac{1}{3}(1 - \alpha)([a_i, a_i^\dagger]_- + [b_i, b_i^\dagger]_- - 2) \ , \\ n_i - m_i &= \lambda (n_i - m_i) - (1 - \lambda)([a_i, a_i^\dagger]_- - [b_i, b_i^\dagger]_-) \ . \end{aligned} \tag{4.2}$$

(Here α and λ are arbitrary parameters.)

We now introduce these identities into the right hand side of Eqs. (3.8a), (3.9a), (3.10), and (3.11). Next, in the *resultant*, we carry out the following truncation:

$$\begin{aligned} \langle\langle a_i^\dagger a_p^\dagger a_p ; a_j \rangle\rangle_E &= \langle a_p^\dagger a_p \rangle \langle\langle a_i^\dagger ; a_j \rangle\rangle_E + \langle a_i^\dagger a_p \rangle \langle\langle a_p^\dagger ; a_j \rangle\rangle_E, \quad i \neq p \ , \\ \langle\langle a_i^\dagger b_p^\dagger b_p ; a_j \rangle\rangle_E &= \langle b_p^\dagger b_p \rangle \langle\langle a_i^\dagger ; a_j \rangle\rangle_E + \langle a_i^\dagger b_p^\dagger \rangle \langle\langle b_p ; a_j \rangle\rangle_E, \quad i \neq p \ , \\ \langle\langle a_i^\dagger a_p a_p^\dagger ; a_j \rangle\rangle_E &= \langle a_p a_p^\dagger \rangle \langle\langle a_i^\dagger ; a_j \rangle\rangle_E + \langle a_i^\dagger a_p \rangle \langle\langle a_p^\dagger ; a_j \rangle\rangle_E, \quad i \neq p \ , \\ \langle\langle a_i^\dagger b_p b_p^\dagger ; a_j \rangle\rangle_E &= \langle b_p b_p^\dagger \rangle \langle\langle a_i^\dagger ; a_j \rangle\rangle_E + \langle a_i^\dagger b_p^\dagger \rangle \langle\langle b_p ; a_j \rangle\rangle_E, \quad i \neq p \ . \end{aligned} \tag{4.3}$$

[Note that these relations do not constitute a decoupling. Rather, Eqs. (4.7)–(4.11) do.] Here we have ignored the nondiagonal contractions, because they are identically vanishing, i.e.,

$$\langle a_i^\dagger b_p\rangle = \langle a_i^\dagger a_p^\dagger\rangle = 0 \,. \tag{4.4}$$

Using the above prescription, i.e., Eqs. (4.2) and (4.3), the relevant decoupling becomes:

$$\langle\langle a_i^\dagger(n_p + m_p); a_j\rangle\rangle_E = (\bar{n}+\bar{m})\,\langle\langle a_i^\dagger; a_j\rangle\rangle_E + \alpha(\langle a_i^\dagger a_p\rangle\,\langle\langle a_p^\dagger; a_j\rangle\rangle_E + \langle a_i^\dagger b_p^\dagger\rangle\,\langle\langle b_p; a_j\rangle\rangle_E), \quad i\neq p\,, \tag{4.5a}$$

$$\langle\langle a_i^\dagger(n_p - m_p); a_j\rangle\rangle_E = (\bar{n}-\bar{m})\,\langle\langle a_i^\dagger; a_j\rangle\rangle_E + \lambda(\langle a_i^\dagger a_p\rangle\,\langle\langle a_p^\dagger; a_j\rangle\rangle_E - \langle a_i^\dagger b_p^\dagger\rangle\,\langle\langle b_p; a_j\rangle\rangle_E)\,, \quad i\neq p\,, \tag{4.5b}$$

and similarly,

$$\langle\langle b_p(n_i + m_i); a_j\rangle\rangle_E = (\bar{n}+\bar{m})\,\langle\langle b_p; a_j\rangle\rangle_E + \alpha(\langle b_p a_i\rangle\,\langle\langle a_i^\dagger; a_j\rangle\rangle_E + \langle b_p b_i^\dagger\rangle\,\langle\langle b_i; a_j\rangle\rangle_E), \quad i\neq p\,, \tag{4.6a}$$

$$\langle\langle b_p(n_i - m_i); a_j\rangle\rangle_E = (\bar{n}-\bar{m})\,\langle\langle b_p; a_j\rangle\rangle_E + \lambda(\langle b_p a_i\rangle\,\langle\langle a_i^\dagger; a_j\rangle\rangle_E - \langle b_p b_i^\dagger\rangle\,\langle\langle b_i; a_j\rangle\rangle_E), \quad i\neq p\,. \tag{4.6b}$$

Another set of four pseudo-Boson-operator Green's functions, which occurs in Eqs. (3.8)–(3.11) is truncated similarly to Eq. (4.3) by introducing the Boson-like contraction, e.g.,

$$\langle\langle a_i^\dagger b_i b_p^\dagger; a_j\rangle\rangle_E = \langle b_i b_p^\dagger\rangle\,\langle\langle a_i^\dagger; a_j\rangle\rangle_E + \langle a_i^\dagger b_p^\dagger\rangle\,\langle\langle b_i; a_j\rangle\rangle_E, \quad i\neq p\,. \tag{4.7}$$

(Again, in the above, we have omitted writing terms containing correlation functions of non-diagonal operator combinations, e.g. $\langle a_i^\dagger b_i\rangle$, because they are vanishing.)

The net result of these decouplings is identical to that given by Barma, i.e.,

$$\langle\langle a_i^\dagger n_p; a_j\rangle\rangle_E = \bar{n}\langle\langle a_i^\dagger; a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger a_p\rangle\,\langle\langle a_p^\dagger; a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger b_p^\dagger\rangle\,\langle\langle b_p; a_j\rangle\rangle_E, \quad i\neq p\,, \tag{4.8}$$

$$\langle\langle a_i^\dagger m_p; a_j\rangle\rangle_E = \bar{m}\langle\langle a_i^\dagger, a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger a_p\rangle\,\langle\langle a_p^\dagger; a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger b_p^\dagger\rangle\,\langle\langle b_p; a_j\rangle\rangle_E, \quad i\neq p\,, \tag{4.9}$$

$$\langle\langle b_p n_i; a_j\rangle\rangle_E = \bar{n}\langle\langle b_p; a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha+\lambda)\langle b_p a_i\rangle\,\langle\langle a_i^\dagger; a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha-\lambda)\langle b_p b_i^\dagger\rangle\,\langle\langle b_i, a_j\rangle\rangle_E\,, \quad i\neq p\,, \tag{4.10}$$

$$\langle\langle b_p m_i; a_j\rangle\rangle_E = \bar{m}\langle\langle b_p, a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha-\lambda)\langle b_p a_i\rangle\,\langle\langle a_i^\dagger; a_j\rangle\rangle_E + \tfrac{1}{2}(\alpha+\lambda)\langle b_p b_i^\dagger\rangle\,\langle\langle b_i; a_j\rangle\rangle_E\,, \quad i\neq p\,. \tag{4.11}$$

Barma[1,2] has emphasized the importance of preserving the identity (2.8b), which is the basic property of the $S=1$ subspace. As noted by Barma, the conservation of this identity places an important restriction[8] on the Green's functions that occur in Eqs. (3.10) and (3.11). Indeed, it can be shown (see Sec. VI) that a certain frequency moment sum rule has to be satisfied by the corresponding spectral functions. Similarly, we find that the same identity also plays a central role in specifying a null sum rule for the spectral functions relating to Green's functions appearing in Eqs. (4.10) and (4.11). This is best displayed by looking at the correlation functions that follow from Eqs. (4.10) and (4.11), i.e.,

$$\langle a_j b_p n_i\rangle = \bar{n}\langle a_j b_p\rangle + \tfrac{1}{2}(\alpha+\lambda)\langle b_p a_i\rangle\,\langle a_j a_i^\dagger\rangle + \tfrac{1}{2}(\alpha-\lambda)\langle b_p b_i^\dagger\rangle\,\langle a_j b_i\rangle, \quad i\neq p\,, \tag{4.12}$$

$$\langle a_j b_p m_i\rangle = \bar{m}\langle a_j b_p\rangle + \tfrac{1}{2}(\alpha-\lambda)\langle b_p a_i\rangle\,\langle a_j a_i^\dagger\rangle + \tfrac{1}{2}(\alpha+\lambda)\langle b_p b_i^\dagger\rangle\,\langle a_j b_i\rangle, \quad i\neq p\,. \tag{4.13}$$

Consequently, when $j=p$, we have for $i\neq j$

$$\alpha(\langle a_i b_j\rangle\,\langle a_i^\dagger a_j\rangle + \langle b_i^\dagger b_j\rangle\,\langle a_j b_i\rangle) = 0, \quad i\neq j\,, \tag{4.14}$$

$$\lambda(\langle a_i b_j\rangle\,\langle a_i^\dagger a_j\rangle - \langle b_i^\dagger b_j\rangle\,\langle a_j b_i\rangle) = 0, \quad i\neq j\,. \tag{4.15}$$

Hence,

$$\alpha(\langle a_i^\dagger a_j\rangle + \langle b_i^\dagger b_j\rangle) = 0, \quad i\neq j\,, \tag{4.16a}$$

$$\lambda(\langle a_i^\dagger a_j\rangle - \langle b_i^\dagger b_j\rangle) = 0, \quad i\neq j\,. \tag{4.16b}$$

Because for the quadrupolar system under investigation, the identity of the correlation functions, i.e.,

$$\langle a_i^\dagger a_j\rangle = \langle b_i^\dagger b_j\rangle\,, \tag{4.17}$$

holds and, in general, these functions are non-vanishing, therefore α should be taken to be zero. It is clear that the remaining parameter, λ, need not be zero to preserve the sum rules (4.16a) and (4.16b). Indeed, λ can be chosen so as to satisfy the basic sum rule referring to the vanishing of the the correlation $\langle a_i b_i\rangle = \langle a_i^\dagger b_i^\dagger\rangle$, as will be made clear in Sec. V.

In what follows, we shall assume that $\alpha = 0$. To conclude this section, we recast the Green's functions equations of motion, (3.8)–(3.11), into a compact form suitable for computation. To this end we introduce the inverse-lattice Fourier transformation,

$$G_{ij}^{AB}(E) = \frac{1}{N}\sum_{\vec{K}} G_{\vec{K}}^{AB}(E)\, e^{i\vec{K}\cdot(\vec{i}-\vec{j})}\,,$$

$$\langle A_i B_j\rangle = \frac{1}{N}\sum_{\vec{K}} \psi_{AB}(K)\, e^{i\vec{K}\cdot(\vec{i}-\vec{j})}\,, \tag{4.18}$$

$$J_{ij} = \frac{1}{N}\sum_{\vec{K}} J(K)\, e^{i\vec{K}\cdot(\vec{i}-\vec{j})}\,.$$

Hence, for arbitrary range of the quadrupolar coupling we get

$$
\begin{aligned}
E G_{\vec{K}}^{a^\dagger a}(E) = {} & (3\bar{n}-1)\{1+[J(0)-\tfrac{1}{2}J(\vec{K})]G_{\vec{K}}^{a^\dagger a}(E)+\tfrac{1}{2}J(K)G_{\vec{K}}^{ba}(E)\} \\
& +\tfrac{3}{2}\alpha\left(\frac{1}{N}\sum_{\vec{q}}\psi_{a^\dagger a}(\vec{q})J(\vec{K}+\vec{q})G_{\vec{K}}^{a^\dagger a}(E)+\frac{1}{N}\sum_{\vec{q}}\psi_{a^\dagger b^\dagger}(\vec{q})J(\vec{K}+\vec{q})G_{\vec{K}}^{ba}(E)\right) \\
& +\frac{3\alpha+\lambda}{4}\left(\frac{1}{N}\right)\sum_{\vec{q}}J(\vec{q})[\psi_{ba}(\vec{q})-\psi_{a^\dagger a}(\vec{q})]G_{\vec{K}}^{a^\dagger a}(E)+\frac{3\alpha-\lambda}{4}G_{\vec{K}}^{ba}(E)\frac{1}{N}\sum_{\vec{q}}J(\vec{q})[\psi_{b^\dagger b}(\vec{q})-\psi_{a^\dagger b^\dagger}(\vec{q})] \\
& +G_{\vec{K}}^{a^\dagger a}(E)\left(\frac{1}{2N}\right)\sum_{\vec{q}}\{J(\vec{q})[\psi_{ba}(\vec{q})-\psi_{b^\dagger b}(\vec{q})]+2J(\vec{K}+\vec{q})\psi_{b^\dagger b}(\vec{q})\} \\
& +G_{\vec{K}}^{ba}(E)\left(\frac{1}{2N}\right)\sum_{\vec{q}}\{J(\vec{q})[\psi_{a^\dagger a}(\vec{q})-\psi_{a^\dagger b^\dagger}(\vec{q})]+2J(\vec{K}+\vec{q})\psi_{a^\dagger b^\dagger}(\vec{q})\}\,. \qquad (4.19)
\end{aligned}
$$

When the spatial range of the coupling is limited to the nearest-neighbor separation and the lattice coordination number is z ($z=6$ for sc lattice) we have

$$J_{ij}=\frac{zJ}{N}\sum_{\vec{K}}\gamma(\vec{K})\,e^{i\vec{K}\cdot(\vec{i}-\vec{j})}\;;$$

$$\gamma(\vec{K})=z^{-1}\sum e^{i\vec{K}\cdot\vec{\delta}}=\tfrac{1}{3}[\cos(K_x a)+\cos(K_y a)+\cos(K_z a)]. \qquad (4.20)$$

[Consistent with the dictates of Eqs. (3.8b) and (4.17), here we have used the notation $\langle n_i\rangle=\langle m_i\rangle=\bar{n}$.]

For lattices with inversion symmetry, such as cubic lattices, we have the symmetry

$$\psi_{AB}(\vec{K})=\psi_{AB}(-\vec{K})\,. \qquad (4.21)$$

The Brillouin zone also manifests the same symmetry for such lattices; hence we get

$$\frac{1}{N}\sum_{\vec{q}}\psi_{AB}(\vec{q})\gamma(\vec{K}+\vec{q})=\gamma(\vec{K})Qf_{AB}\,, \qquad (4.22)$$

where

$$f_{AB}=\frac{1}{QN}\sum_{\vec{q}}\psi_{AB}(\vec{q})\gamma(\vec{q})\,. \qquad (4.23)$$

(Note that $Q=3\bar{n}-1$.) Using the abbreviated notation,

$$f_{a^\dagger a}=f_{b^\dagger b}\equiv u\,,\qquad f_{a^\dagger b^\dagger}=f_{ab}\equiv v\,, \qquad (4.24)$$

we can recast Eq. (4.19) into the following convenient form:

$$[E-C(\vec{K})]G_{\vec{K}}^{a^\dagger a}(E)-D(\vec{K})G_{\vec{K}}^{ba}(E)=Q\,, \qquad (4.25)$$

where

$$C(\vec{K})=zJQ\{1-\tfrac{1}{2}\gamma(\vec{K})+[\gamma(\vec{K})-\tfrac{1}{2}]u+\tfrac{1}{2}v+\tfrac{1}{4}\lambda(v-u)\}\,, \qquad (4.26a)$$

$$D(\vec{K})=zJQ\{\tfrac{1}{2}\gamma(\vec{K})+[\gamma(\vec{K})-\tfrac{1}{2}]v+\tfrac{1}{2}u+\tfrac{1}{4}\lambda(v-u)\}\,. \qquad (4.26b)$$

Following the same procedure, we can also obtain the corresponding expressions from the three remaining Green's-function equations of motion, i.e., Eqs. (3.9)–(3.11). The resultant three expressions, along with Eq. (4.25), are displayed in a convenient matrix form below:

$$\begin{pmatrix} G_{\vec{K}}^{a^\dagger a}(E) & G_{\vec{K}}^{a^\dagger b^\dagger}(E) \\ G_{\vec{K}}^{ba}(E) & G_{\vec{K}}^{bb^\dagger}(E)\end{pmatrix}=\frac{Q}{E^2-E^2(\vec{K})}\begin{pmatrix} E+C(\vec{K}) & -D(\vec{K}) \\ -D(\vec{K}) & -E+C(\vec{K})\end{pmatrix}\,, \qquad (4.27)$$

where

$$E(\vec{K})=zJQ\{[1-\gamma(\vec{K})](1-u+v)[1+\gamma(\vec{K})(u+v)-\tfrac{1}{2}\lambda(u-v)]\}^{1/2}\,. \qquad (4.28)$$

With the help of the spectral relationship embodied in Eq. (3.6), these Green's functions yield the following result for the corresponding correlation functions:

$$\begin{pmatrix} \psi_{aa^\dagger}(\vec{K}) & \psi_{b^\dagger a^\dagger}(\vec{K}) \\ \psi_{ab}(\vec{K}) & \psi_{b^\dagger b}(\vec{K})\end{pmatrix}=\frac{Q}{2}\begin{pmatrix} \dfrac{C(\vec{K})}{E(\vec{K})}\coth\left(\dfrac{\beta E(\vec{K})}{2}\right)-1 & -\dfrac{D(\vec{K})}{E(\vec{K})}\coth\left(\dfrac{\beta E(\vec{K})}{2}\right) \\ -\dfrac{D(\vec{K})}{E(\vec{K})}\coth\left(\dfrac{\beta E(\vec{K})}{2}\right) & \dfrac{C(\vec{K})}{E(\vec{K})}\coth\left(\dfrac{\beta E(\vec{K})}{2}\right)+1\end{pmatrix} \qquad (4.29)$$

V. CORRELATION FUNCTIONS

The matrix expression (4.29) embodies our result for the four correlation functions $\langle A_i B_j \rangle$, where $A \equiv a, b^\dagger$ and $B \equiv a^\dagger$ and b. We note that there are as of now only four unknowns in our theory, i.e., λ, u, v, and Q. The basic sum rule relating to the off-diagonal self-correlation, i.e.,

$$\langle a_i b_i \rangle = \langle b_i^\dagger a_i^\dagger \rangle = 0 , \tag{5.1a}$$

specifies one condition that these quantities have to obey, namely

$$-\frac{Q}{2N} \sum_{\vec{K}} D(\vec{K}) \Delta(\vec{K}) = 0 , \tag{5.1b}$$

where

$$\Delta(\vec{K}) = \coth[\tfrac{1}{2}\beta E(\vec{K})] / E(\vec{K}) . \tag{5.1c}$$

The second condition is provided by the relationship which follows from Eq. (4.29), i.e.,

$$\langle a_i^\dagger a_i \rangle = \langle b_i^\dagger b_i \rangle = \bar{n} = \tfrac{1}{3}(Q+1) , \tag{5.2a}$$

or in other words

$$Q = 2\left[1 + \frac{3}{N} \sum_{\vec{K}} C(\vec{K}) \Delta(\vec{K})\right]^{-1} . \tag{5.2b}$$

The remaining two constraints, which help specify the four unknowns completely, are contained in Eqs. (4.23) and (4.24) as follows:

$$u = \frac{1}{2N} \sum_{\vec{K}} \gamma(\vec{K}) C(\vec{K}) \Delta(\vec{K}) , \tag{5.3}$$

$$v = -\frac{1}{2N} \sum_{\vec{K}} \gamma(\vec{K}) D(\vec{K}) \Delta(\vec{K}) . \tag{5.4}$$

In order to find a solution at general temperatures, it is necessary to solve the four equations, (5.1)–(5.4), self-consistently. This is a complicated task, for each of these expressions is itself a three dimensional integral, involving rather awkward kernels.

At zero temperature, on the other hand, the situation is considerably simpler. Here $\beta \to \infty$ and as such

$$\coth[\tfrac{1}{2}\beta E(\vec{K})] \to \pm 1 \tag{5.5}$$

depending on whether Q is positive or negative [see Eq. (4.28)]. Knowing that the molecular-field approximation gives $Q < 0$, it is eminently reasonable to choose the lower of the two signs in Eq. (5.5) and then numerically solve for the remaining three unknowns using Eqs. (5.1), (5.3), and (5.4). In what follows we shall treat only the case for which $T = 0$.

Our computations were performed for a simple-cubic lattice. First we used only 8000 Gaussian points in the Brillouin zone by choosing an irreducible section in the first octant. Such a section has a total of $\frac{1}{48}$ of the zone volume but contains 220 relevant Gaussian points in it. Employing Newton-Raphson iteration procedure, the following set of results were obtained for the three unknowns:

$$u = -0.0092; \quad v = 0.0595; \quad \lambda = -1.9461. \tag{5.6}$$

Inserting this set of values into Eq. (5.2b) we found Q to be self-consistently negative, thus justifying our original choice,

$$Q = -0.9357 . \tag{5.7}$$

To test whether other solutions to this set of three self-consistent equations, i.e., Eqs. (5.1), (5.3), and (5.4), existed, we performed the computation for a variety of initial conditions chosen within the limits

$$0.5 \geq \nu \geq -0.5, \quad \nu \equiv \lambda, u, v.$$

In about one hundred such runs, i.e., with different initial conditions, we found no other solution except for that given in Eq. (5.6).

Having identified the correct solution in this manner, we set out to improve the accuracy of our computation by increasing the number of Gaussian points within the zone to 512 000. Moreover, the convergence threshold was now decreased to 10^{-12}, or better (compared with 10^{-6}, or better, previously). The final results were found to be only slightly altered by this procedure, i.e.,

$$\begin{aligned} &u = -0.009\,14, \quad v = 0.059\,46, \\ &\lambda = -1.946\,05, \quad Q = -0.935\,72. \end{aligned} \tag{5.8}$$

Comparing Eqs. (5.6) and (5.7) with (5.8) and noting the level of accuracy obtained in the computation of the Watson sum by the same method, we were able to place error limits on the computational accuracy of Q, i.e.,

$$Q = -0.9357 \pm 0.0001 . \tag{5.9}$$

Having determined Q, we can immediately find the ground-state average of $(S^z)^2$, i.e.,

$$\langle (S^z) \rangle = \tfrac{2}{3}(1+Q) = 0.0429 \pm 0.0001 . \tag{5.10}$$

(We emphasize that the errors quoted above reflect only the level of accuracy of the numerical computational procedure used here.)

It is interesting to compare these results with those given by the RPA and the procedure used by Barma. As mentioned earlier, because of the nonconservation of the off-diagonal self-correlation sum rule, the RPA is not a uniquely defined decoupling prescription and it can lead to inconsistent and even totally erroneous results, e.g., the results obtained by carrying out the relevant computations (for a simple-cubic lattice) using the nonsymmetrical RPA decoupling of Ueyama and Matsubara.[9,10] Nevertheless, the symmetrized RPA (as recently defined by Barma and first used by Raich and Etters) is a uniquely defined procedure,

even though it does not conserve the off-diagonal sum rule referred to above and using this we have found[10]

$$Q=-0.9367, \quad \langle (S^z)^2 \rangle = 0.0422 \ . \tag{5.11}$$

We have also performed the computation of the expressions given by the Barma scheme. Here too, for negative Q, we found only one solution of the three self-consistent equations (involving α, u, and v). Moreover, much like the solution to our equations, on further increase in the computational accuracy this solution was also found to be unaffected to more than a few parts in the fifth figure. The relevant results are

$$\begin{aligned} &\alpha = 0.6889, \quad u = -0.0094, \quad v = 0.0620 \ , \\ &Q = -0.9316, \quad \langle (S^z)^2 \rangle = 0.0456 \ . \end{aligned} \tag{5.12}$$

(Recall that Barma's decoupling procedure consists in making an *ad hoc* choice for λ, i.e., $\lambda = 0$, and then α is chosen from the requirement that the off-diagonal self-correlation be zero. Also note that the given results were computed by us and they are substantially different from those originally reported by Barma. This is largely due to the presence of an error in his original expressions. The error has been rectified in Ref. 2.)

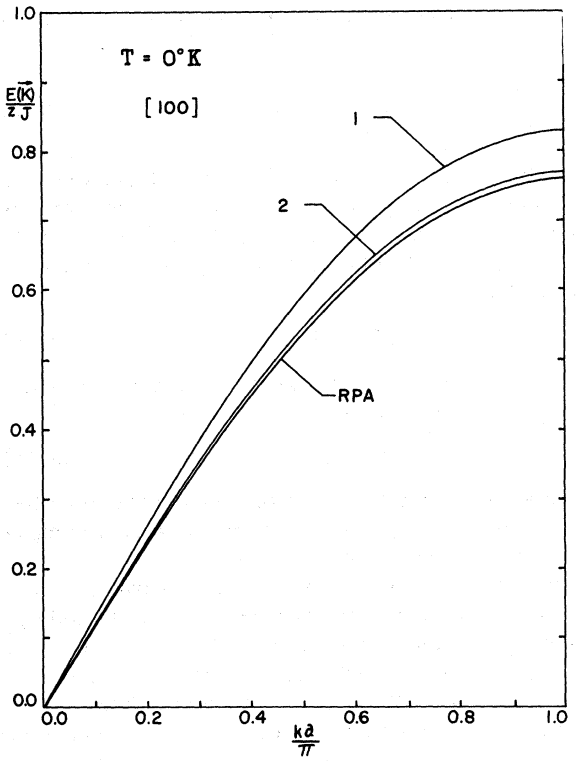

FIG. 1. Zero-temperature excitation spectrum for $\vec{K} = (k, 0, 0)$, is plotted for a simple-cubic lattice with cube edge a. The curve marked RPA is obtained by the use of the relation $E(\vec{K}) = QzJ[1-\gamma(\vec{K})]^{1/2}$, which follows from the symmetrical RPA procedure. Curves 1 and 2 refer, respectively, to the results obtained by using the Barma decoupling and those obtained in the present work.

Let us compare the three sets of results for $\langle (S^z)^2 \rangle$ given in Eqs. (5.10)–(5.12). We note that the symmetrized RPA, i.e., Eq. (5.11), already gives a good description of the quantum-mechanical nature of the ground state for it tells us that, unlike the prediction of the molecular-field approximation, the zero-point fluctuations lead to a nonvanishing value for $\langle (S^z)^2 \rangle$. The next-order approximation, which gives a correctly symmetrized structure for the decoupling used in the symmetrized RPA which, in addition, also conserves the off-diagonal correlation sum rule of Eq. (5.1a), is that of Barma. This predicts [see Eq. (5.12)] a somewhat larger zero-point fluctuation than that given by the RPA.

An even higher-order approximation (see Sec. VI for an amplification of this point), which, in addition to retaining the above-mentioned conserving features of the Barma theory, also preserves the first dominant set of moment-conserving correlation function sum rules,[8,11] is that described in the present work. This approximation is found to lead to a result for the zero-point fluctuation which is only nominally larger [see Eq. (5.10)] than that given by the RPA. Such an oscillatory behavior of successive approximations is not an uncommon occurrence. A satisfying feature of these three sets of results is that they are all in reasonable agreement with each other.

Having compared the results for the LRO in these theories, we examine next the behavior of the excitation spectrum. For simplicity we have carried out the relevant computations only for the symmetry directions $\vec{K} = (k, 0, 0)$, $(k, k, 0)$, and (k, k, k) in the simple-cubic lattice.

In Fig. 1, we have plotted the elementary excitation energy for $\vec{K} = (k, 0, 0)$. Our computation of the expression obtained in the Barma scheme, i.e.,

$$\begin{aligned} [E(\vec{k})]_{\text{Barma}} = zJQ \ \{&[1-\gamma(\vec{k})][1-(1+\tfrac{3}{2}\alpha)(u-v)] \\ &\times [1+(1+\tfrac{3}{2}\alpha)(u+v)\gamma(\vec{k})]\}^{1/2} , \end{aligned} \tag{5.13}$$

is found to lead to a result which lies well above that given by the symmetrical RPA,[2,4] i.e.,

$$[E(\vec{k})]_{\text{RPA}} = zJQ\,[1-\gamma(\vec{k})]^{1/2} \ . \tag{5.14}$$

The result obtained in the present work is close to that given in the RPA and lies a little above it throughout the zone (in this direction).

The behavior along the (1, 1, 0) and (1, 1, 1) directions (see Figs. 2 and 3) is similar except that here the RPA results become larger than those obtained in the present work somewhere past half-way to the zone boundary. This "level inversion" is

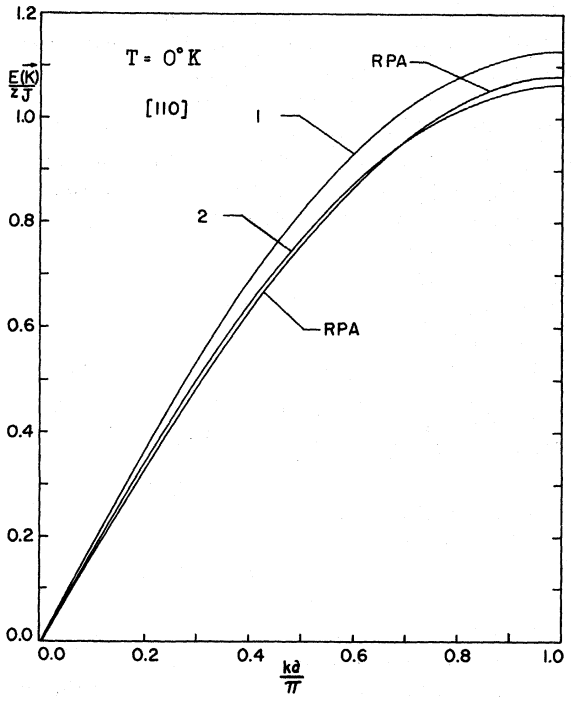

FIG. 2. The same as in Fig. 1 except for the difference that here $\vec{K}=(k,k,0)$.

most noticeable near the (1, 1, 1) zone edge where our results begin to differ appreciably from those given by the RPA decoupling.

VI. GROUND-STATE ENERGY

As explained in the preceding sections, the quadrupolar coupled system has now been solved in a hierarchy of approximations. Although, the successive nature of these approximations was mentioned in the introduction, the relevant statements were rather vague. In particular, no substantive criterion for the establishment of the hierarchy was described.

A few years ago, Roth[11] introduced a procedure which was nominally variational. After making, what was in effect a physically motivated choice of a set of basis operators, a stationarity procedure was invoked which helped specify an appropriate linear combination of the basis operators used for decoupling the Green's function. At about the same time, and independently, Tahir-Kheli and Jarrett[12] presented a decoupling procedure, based upon the conservation of a given number of frequency moments of the spectral function of time-dependent correlation functions, which was later[8,13] shown to lead to identical results to the Roth variational procedure. Thus, the moment conservation procedure was, *a posteriori*, provided with a basis in a variational scheme.

It should, however, be emphasized that such a variational scheme has not, to our knowledge, been shown to be equivalent to any thermodynamic minimum principle.[14] Therefore, it cannot, *per se*, be assumed to guarantee a physically more relevant solution.[15]

On the other hand, a weak, but quite possibly physically meaningful, proviso can be attached to the moment-conserving[8] stationary principle[11,13]: namely, when a self-consistent moment-conserving approximation leads to a lowering of the system free energy, then it is a more accurate approximation than a corresponding decoupling which does not preserve the same moment.

In this spirit, in the following we shall examine the results for the free energy at zero temperature (it is equal to the system energy at $T=0$) that are obtained self-consistently within the context of the four approximations referred to in this paper. These are the molecular-field,[3] the symmetrized[2,4] RPA, the Barma approximation,[2] and the present approximation.

Within the molecular-field approximation (MFA), the system energy is quite straightforward to calculate. Writing the Hamiltonian (2.9) as

$$\mathcal{H}=\mathcal{H}^0+\mathcal{H}' , \tag{6.1}$$

$$\mathcal{H}^0=-\tfrac{3}{4}\sum_{i,j} J_{ij}(n_i+m_i-\tfrac{2}{3})(n_j+m_j-\tfrac{2}{3}) , \tag{6.2}$$

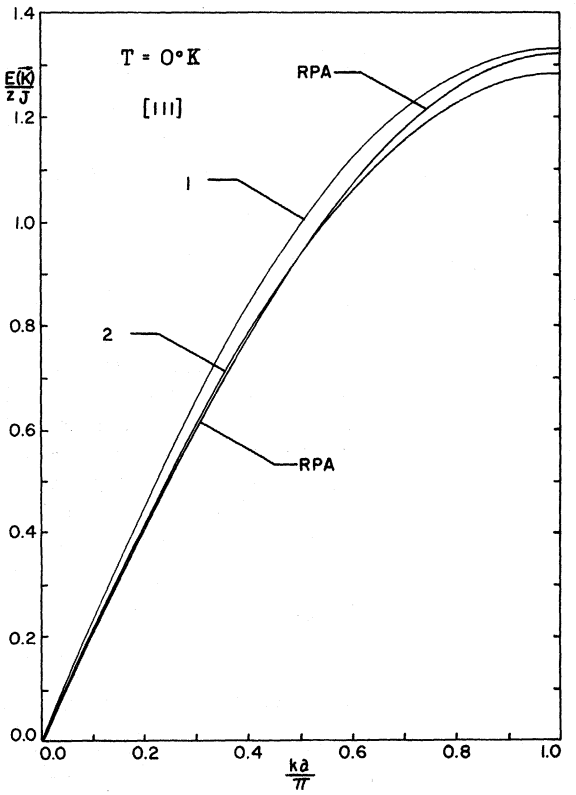

FIG. 3. Same as in Fig. 1, but with wave vector along the cube diagonal, i.e., $\vec{K}=(k,k,k)$.

$$\mathcal{H}' = -\sum_{i,j} J_{ij}[\tfrac{1}{2}(a_i^\dagger - b_i)(a_j - b_j^\dagger) + a_i^\dagger b_i b_j^\dagger a_j] , \tag{6.3}$$

we readily see that

$$\langle \mathcal{H}' \rangle_{\mathrm{MFA}} \to 0 \tag{6.4}$$

and

$$\langle \mathcal{H}^0 \rangle_{\mathrm{MFA}} = -(\tfrac{1}{3}JzN)Q^2 . \tag{6.5}$$

Because $0 \leq \langle (S_i^z)^2 \rangle \leq 1$, therefore $\frac{1}{2} \geq Q \geq -1$. Hence, the ground-state energy is the lowest when $Q = -1$, i.e.,

$$\langle \mathcal{H}^0 \rangle_{\mathrm{MFA}} = -(\tfrac{1}{3}JzN) \equiv -\epsilon_0 . \tag{6.6}$$

To calculate the ground-state energy within the three set of approximations, i.e., RPA, Barma, and ours, we proceed as follows: First we recast Eq. (6.2) into the form

$$\langle \mathcal{H}^0 \rangle = -\frac{3}{4}\sum_{i,p} J_{ip}(\langle n_i n_p \rangle + \langle m_i m_p \rangle + \langle n_i m_p \rangle + \langle m_i n_p \rangle) - \epsilon_0(1 - 6\bar{n}) . \tag{6.7}$$

Next, to calculate the sum of the four correlation functions appearing on the right-hand side of Eq. (6.7) self-consistently with the corresponding decoupling procedures, we take the decoupling specified by Eqs. (4.8) and (4.9) and rewrite it as follows:

$$\langle\langle a_i^\dagger n_p ; a_l \rangle\rangle_E = \bar{n}\langle\langle a_i^\dagger ; a_l \rangle\rangle_E + \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger a_p \rangle \langle\langle a_p^\dagger ; a_l \rangle\rangle_E + \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger b_p^\dagger \rangle \langle\langle b_p ; a_l \rangle\rangle_E , \quad i \neq p , \tag{6.8}$$

$$\langle\langle a_i^\dagger m_p ; a_l \rangle\rangle_E = \bar{m}\langle\langle a_i^\dagger ; a_l \rangle\rangle_E + \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger a_p \rangle \langle\langle a_p^\dagger ; a_l \rangle\rangle_E + \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger b_p^\dagger \rangle \langle\langle b_p ; a_l \rangle\rangle_E , \quad i \neq p . \tag{6.9}$$

Using the usual spectral theorems, [6] this gives

$$\langle a_l a_i^\dagger n_p \rangle = \bar{n}\langle a_l a_i^\dagger \rangle + \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger a_p \rangle \langle a_l a_p^\dagger \rangle + \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger b_p^\dagger \rangle \langle a_l b_p \rangle , \quad i \neq p , \tag{6.10}$$

$$\langle a_l a_i^\dagger m_p \rangle = \bar{m}\langle a_j a_i^\dagger \rangle + \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger a_p \rangle \langle a_l a_p^\dagger \rangle + \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger b_p^\dagger \rangle \langle a_l b_p \rangle , \quad i \neq p . \tag{6.11}$$

For the case when $l = i$, Eqs. (6.10) and (6.11) yield

$$\langle n_i n_p \rangle + \langle m_i n_p \rangle = 2(\bar{n})^2 - \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger a_p \rangle \langle a_i a_p^\dagger \rangle - \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger b_p^\dagger \rangle \langle a_i b_p \rangle , \quad i \neq p , \tag{6.12}$$

$$\langle m_i m_p \rangle + \langle n_i m_p \rangle = 2(\bar{n})^2 - \tfrac{1}{2}(\alpha-\lambda)\langle a_i^\dagger a_p \rangle \langle a_i a_p^\dagger \rangle - \tfrac{1}{2}(\alpha+\lambda)\langle a_i^\dagger b_p^\dagger \rangle \langle a_i b_p \rangle , \quad i \neq p . \tag{6.13}$$

(Here we have made use of the identity $a_i a_i^\dagger = 1 - n_i - m_i$.) Adding Eqs. (6.12) and (6.13) gives us the desired sum appearing in Eq. (6.7), i.e.,

$$\langle n_i n_p \rangle + \langle m_i m_p \rangle + \langle n_i m_p \rangle + \langle m_i n_p \rangle = 4(\bar{n})^2 - \alpha(\langle a_i^\dagger a_p \rangle \langle a_i a_p^\dagger \rangle + \langle a_i^\dagger b_p^\dagger \rangle \langle a_i b_p \rangle) , \quad i \neq p . \tag{6.14}$$

Now, we introduce Eq. (6.14) into Eq. (6.7) and write:

$$\langle \mathcal{H}^0 \rangle = -\epsilon_0[1 - 6\bar{n} + 9(\bar{n})^2 - \tfrac{9}{4}q\alpha Q^2(u^2+v^2)] . \tag{6.15}$$

(Note that $\alpha = 0$ within the MFA, the RPA, and the present approximation. However, $\bar{n}$ is implicitly dependent upon the original decoupling and hence is different in each of these theories.)

Next we examine the average of $\mathcal{H}'$, given in Eq. (6.3). The only interesting correlation here is that referring to the four operators in the last term in Eq. (6.3). Here we take our cue from Eq. (4.7). Using the spectral theorem invoked earlier, this yields:

$$\langle a_i^\dagger b_i b_j^\dagger a_j \rangle = \langle b_i b_j^\dagger \rangle \langle a_i^\dagger a_j \rangle + \langle b_i a_j \rangle \langle a_i^\dagger b_j^\dagger \rangle , \quad i \neq j . \tag{6.16}$$

Thus, using Eq. (6.3) we finally get

$$\langle \mathcal{H}' \rangle = -\epsilon_0[3Q(u-v) + 3Q^2(u^2+v^2)] . \tag{6.17}$$

Hence

$$\langle \mathcal{H} \rangle = -\epsilon_0[Q^2 + 3Q(u-v) + 3\eta Q^2(u^2+v^2)] , \tag{6.18}$$

where

$$\eta = \begin{cases} 0, \text{ for RPA} \\ 1 - \frac{3}{4}\alpha \text{ for the Barma decoupling} \\ 1 \text{ for our decoupling} . \end{cases} \tag{6.19}$$

Upon comparing the above result with that derived by Raich and Etters[3] (within their symmetrical RPA procedure for the fcc lattice) from somewhat different considerations, we find that our result for the RPA is completely analogous to that given by them.

We have carried out the computation of Eq. (6.18) self-consistently within the three decoupling approximations. The result (given by our most elaborate numerical procedure described in Sec. V) is the following:

$$\langle \mathcal{H} \rangle \to \begin{cases} -\epsilon_0 , & \text{MFA} \\ -(1.0715 \pm 0.0002)\epsilon_0 , & \text{RPA} \\ -(1.0731 \pm 0.0002)\epsilon_0 , & \text{Barma} \\ -(1.0790 \pm 0.0002)\epsilon_0 , & \text{this work} . \end{cases} \tag{6.20}$$

(The quoted error bounds refer only to the numerical computations). Hence, our procedure, in addition to satisfying the symmetry features of the RPA and the conservation of the off-diagonal self-correlation identity, also helps self-consistently preserve one additional frequency moment—beyond that of the zeroth order which only specifies the weight under the spectral function and thus is automatically preserved by the commutator appearing in the inhomogeneous term occurring in the Green's-function equation of motion—for the case of the nearest-neighbor spectral functions. Moreover, it yields the lowest estimate for the ground state energy. [See Eqs. (4.12) and (4.13) and note that putting $j=p$, makes these equations relate to the equations of motion of the basic Green's functions referring to two neighboring sites. Also compare with Ref. 8.] We believe that this fact lends some credence to the present results.

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[10]The RPA decoupling used in Ref. 9 would lead to the result $Q=-2\{(3/N)\sum_{\vec{k}}[1-\gamma(\vec{k})]-1\}^{-1}$. We have computed this for the sc lattice with 512 000 Gaussian points in the zone and found that it leads to the unphysical result $\langle(S^z)^2\rangle=-0.026$. On the other hand, the symmetrized RPA approximation of Raich and Etters (Ref. 3) leads to the result $\langle(S^z)^2\rangle=0.0422$.

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