

Excitation spectrum in quadrupolar systems

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The static and dynamic properties of an isotropic quadrupolar system are studied by the method of double-time Green's functions. Using symmetrized equations of motion, a Dyson equation for the single-particle Green's function is obtained. The self-energy is expressed in terms of three-particle Green's functions, which are determined using an effective Hamiltonian, with the exact commutation relations approximated by quasiboson ones. A detailed expression for the lifetime of the elementary excitations is obtained.

I. INTRODUCTION

Recently there has been considerable interest in systems with quadrupolar interactions. Consider a quadrupole consisting of four point charges suitably placed along a straight line. The potential energy between a pair of such quadrupoles is minimized if the line joining the center of the quadrupoles is parallel to the axis of one quadrupole and perpendicular to the axis of the other. Now consider a sequence of these rigid quadrupoles, with center of mass fixed to a linear chain and each rotating about the center of mass with angular momentum J . Classically, in the ground state, these quadrupoles would line up with adjacent rotation axes perpendicular and $M = \pm J$. The elementary excitations of this system are librational waves in which the axes of rotation perform small oscillations about their ground-state orientation.

The potential energy between two quadrupoles, with centers separated by the distance R_{ij} , is¹

$$V_{ij} = \frac{4\pi\mu^2}{5R_{ij}^5} \sum_{m=-2}^2 \alpha_m Y_2^m(\Omega_i) Y_2^{-m}(\Omega_j), \quad (1.1)$$

where Y_l^m is the spherical harmonic [with sign convention $Y_2^{-1} = (Y_2^1)^*$]; $\alpha_0 = 6$, $\alpha_{\pm 1} = -4$, and $\alpha_{\pm 2} = 1$; and μ is the quadrupole moment of the charge configuration (with four charges along the z axis the quadrupole tensor has only the diagonal elements $-\mu$, $-\mu$, and 2μ). Nakamura¹ has introduced a quantum-mechanical model by expressing the spherical harmonics in terms of spin operators in the $S=1$ subspace

$$Y_l^m(\Omega_i) = -(3/10\pi)^{1/2} L_l^m(i), \quad (1.2)$$

where the spherical tensor operators $L_l^m(i)$ are given by

$$\begin{aligned} L_2^0(i) &= \sqrt{\frac{3}{2}} \left[(S_i^z)^2 - \frac{2}{3} \right], \\ L_2^{\pm 1}(i) &= \frac{1}{2} (S_i^+ S_i^z + S_i^z S_i^+), \\ L_2^{\pm 2}(i) &= \frac{1}{2} (S_i^{\pm})^2. \end{aligned} \quad (1.3)$$

It is convenient to rotate the spin axes at each site so the z axis corresponds to the classical ground state configuration. For instance, in the linear chain the spin axes would be rotated 90° counter-clockwise about the x axis at every alternate site. The Hamiltonian will then take the form

$$\mathcal{H} = - \sum_{(i,j)} \sum_{m,n} J_{ij}^{mn} L_2^m(i) L_2^n(j), \quad (1.4)$$

where the coupling constants J_{ij}^{mn} , between spins at lattice sites i and j , depend on the particular ground-state structure.

The most studied quadrupolar system is molecular hydrogen.² At low temperature, ortho-hydrogen can be modeled as $J=1$ rigid rotators fixed to a rigid lattice with the orientational interaction described by the Hamiltonian (1.4).^{1,3} Infrared absorption⁴ and x-ray diffraction⁵ experiments confirmed that the crystal structure is fcc at sufficiently low temperatures. It is impossible for all molecular axes to be perpendicular. The ordered state, classically, consists of four sublattices, with all molecular axes in a given sublattice pointing along one of the four $[1, 1, 1]$ directions.⁶ The quadrupolar order parameter

$$Q = \left(\frac{3}{2}\right) \langle (S_i^z)^2 \rangle - 1, \quad (1.5)$$

has been measured by NMR techniques,⁷ with the indication that there is probably a first-order transition around 3°K .⁸ The transition to an orientationally disordered phase is marked by a lambda anomaly in the specific heat.⁹ The order-disorder transition is accompanied by a structural phase change from the fcc to a hcp lattice. The exact role of this structural phase transition, which occurs at or near the order-disorder transition, is not clear at this time.²

In the molecular-field ground state all spins are in the $M=0$ state. Librons are collective excitations of the $M=\pm 1$ states. Since there are four sublattices, there will be eight branches to the excitation spectrum $\epsilon_\mu(\vec{k})$ all with a gap due to the anisotropy in the interactions. Green's function

techniques have been used to determine the excitation spectrum.^{10,11} Particular attention has been given to the zero wave-vector case,¹² where the effects of libron-libron interactions can be seen in Raman scattering experiments.¹³ It has been suggested that the full dispersion relation $\epsilon_{\mu}(\vec{k})$ and the libron lifetimes should be measurable in nuclear γ -ray scattering.¹⁴

Harris has developed a complete analogy between hydrogen and a dilute antiferromagnet.¹⁵ The para-hydrogen ($J=0$) take the place of the nonmagnetic ions as there is no orientational interaction between the spherically symmetric $J=0$ molecules and its neighbors. The corresponding percolation problem has also been studied by both theory and experiment.¹⁶

In this paper we follow Barma¹⁷ and Fittipaldi and Tahir-Kheli¹⁸ in studying the simplest of the dynamical quadrupolar Hamiltonians, by restricting $J_{ij}^{mn} = \delta_{m,n} J_{ij}$. The resulting Hamiltonian,

$$\mathcal{H} = - \sum_{\langle i,j \rangle} \sum_m J_{ij} L_2^m(i) L_2^m(j), \quad (1.6)$$

is isotropic; that is S^2 is a good quantum number. This Hamiltonian is to a quadrupolar system what the isotropic Heisenberg model is to a dipolar system. There is only one sublattice and the excitations $M = \pm 1$ are equivalent so there is only one branch to the dispersion relation. The molecular-field ground state $M=0$ is still not an eigenstate of the Hamiltonian (1.6) as with real hydrogen. The existence of an ordered state breaks the rotational symmetry of the Hamiltonian. Thus the excitations will be gapless in the limit of vanishing momentum.¹⁹ Clearly this model does not represent real hydrogen; we take the philosophy that it is nevertheless an interesting mathematical model.

In fact even a simpler nondynamical model, in which $J_{ij}^{mn} = \delta_{m,0} \delta_{n,0} J_{ij}$, has been considered as a model for ortho-hydrogen.^{20,21} Harris has pointed out that it has no critical point as it transforms into a $S = \frac{1}{2}$ Ising model in a nonzero field.²² Whether or not the addition of the fluctuation terms in the Hamiltonian (1.6) gives rise to a critical point remains an open question.

In addition to molecular hydrogen and other molecular solids,²³ quadrupolar interactions arise in several physical situations. Liquid crystals possess similarities with molecular crystals. They are characterized by the presence of an orientational order of rodlike molecules (with disordered centers of gravity). Maier and Suppe²⁴ have presented a microscopic theory for molecules possessing quadrupolar symmetry to describe the first-order nematic-isotropic liquid phase trans-

ition. The Heisenberg model is a suitable description of magnetic systems with no orbital contribution to the magnetic moment. When there is strong spin-orbit coupling, biquadratic terms appear in the pseudospin Hamiltonian.²⁵ Another example is Jahn-Teller ferroelectric system, where there may be a phonon-induced electric quadrupolar interaction.²⁶ In all of these cases, the anisotropic contributions to the Hamiltonian will undoubtedly be important.

In this paper we limit our discussion to a study of the zero-temperature excitation spectrum of the Hamiltonian (1.6). Our approach differs from the previous authors,^{17,18} in that we maintain a symmetrized form for the equations of motion without resorting to a complicated Callen-like decoupling scheme. This enables us to calculate the three-particle Green's function so we can determine the damping of the elementary excitations. We do, however, violate the off-diagonal self-correlation sum rule.^{17,18} We argue that just as the diagonal self-correlation is only calculated approximately in any perturbation scheme, so too is the off-diagonal self-correlation. Hence it does not vanish identically.

In Sec. II we introduce creation and annihilation operators particularly suitable to describe the $S=1$ algebra. Then the equations of motion are derived and written in the form of a Dyson equation for the Green's function. In the zero-order or linear theory the dispersion relation for the excitations is not surprisingly the same as that for an antiferromagnet. In Sec. IV a static renormalization of the excitation energies is calculated using the exact equations of motion. Next, an effective quadratic Hamiltonian is introduced with quasiboson operators. This is diagonalized by a canonical transformation. In Sec. VI we calculate the contribution of three-particle Green's functions to the self-energy, which leads to damping of the excitation. The lifetime is determined in Sec. VII. Finally the effect of cubic anisotropies present in real hydrogen is considered.

II. HAMILTONIAN

Following Raich and Eters¹⁰ we express the $S=1$ spin operators in terms of two creation and annihilation operators. The creation operators

$$a_i^\dagger = (1/\sqrt{2}) S_i^+ [1 - (S_i^z)^2], \quad (2.1)$$

$$b_i^\dagger = (1/\sqrt{2}) S_i^- [1 - (S_i^z)^2] \quad (2.2)$$

connect the $M=0$ spin state to the $M=1$ and $M=-1$ states, respectively. The annihilation operators a_i and b_i are given by the Hermitian conjugates of (2.1) and (2.2). These are natural operators to describe the excitations from the molecular-field

ground state $M=0$ of the quadrupolar system. The number operators

$$n_i = a_i^\dagger a_i, \quad (2.3)$$

$$m_i = b_i^\dagger b_i, \quad (2.4)$$

take the value unity in the excited states $M=+1$ and $M=-1$, respectively, and are otherwise zero. Since these operators are describing a $S=1$ subspace, they give zero if they act twice on a state

$$a_i^2 = b_i^2 = 0. \quad (2.5)$$

They also satisfy the relations

$$a_i b_i = b_i a_i = a_i b_i^\dagger = 0. \quad (2.6)$$

The dynamics of the quadrupolar system are complicated by the fact that the creation and annihilation operators are neither bosons nor paulion operators. In fact they satisfy the commutation relations

$$[a_i, a_j^\dagger] = (1 - 2n_i - m_i) \delta_{i,j}, \quad (2.7)$$

$$[b_i, b_j^\dagger] = (1 - n_i - 2m_i) \delta_{i,j}. \quad (2.8)$$

The other commutation relations between a_i and b_j or b_j^\dagger follow from (2.6).

The transformation (2.1) and (2.2) can be inverted to express the spin operators in terms of the creation and annihilation operators

$$S_i^z = n_i - m_i, \quad (2.9)$$

$$S_i^y = \sqrt{2} (a_i^\dagger + b_i). \quad (2.10)$$

The spherical tensor operators (1.3) are given by

$$Q_i = \sqrt{\frac{3}{2}} L_2^0(i) = (\frac{3}{2})(n_i + m_i) - 1, \quad (2.11)$$

$$L_2^1(i) = \sqrt{\frac{1}{2}} (a_i^\dagger - b_i), \quad (2.12)$$

$$L_2^2(i) = a_i^\dagger b_i. \quad (2.13)$$

The isotropic quadrupolar Hamiltonian (1.6) now takes the simple form

$$\begin{aligned} \mathcal{H} = - \sum_{i,j} J_{ij} \{ (\frac{1}{3}) Q_i Q_j + (\frac{1}{2}) (a_i^\dagger - b_i) (a_j - b_j^\dagger) \\ + a_i^\dagger b_i b_j^\dagger a_j \}. \end{aligned} \quad (2.14)$$

We note that the commutator of the operators a_i and b_i with the quadrupole operator Q_i is diagonal

$$[a_i, Q_j] = (\frac{3}{2}) a_i \delta_{i,j}, \quad (2.15)$$

$$[b_i, Q_j] = (\frac{3}{2}) b_i \delta_{i,j}. \quad (2.16)$$

The other commutators with $L_2^{\pm 1}$ and $L_2^{\pm 2}$ follow directly from Eqs. (2.6)–(2.8).

III. EQUATION OF MOTION OF GREEN'S FUNCTIONS

In order to study the dynamics of the quadrupolar Hamiltonian (1.6), we form the equations of mo-

tion for four one-particle Green's functions. These equations involve higher-order Green's functions which are then usually decoupled to obtain a closed set of equations. Instead, we write equations of motion for the higher-order Green's functions and formally obtain a Dyson's equation for the single-particle Green's functions.

It is convenient to represent the four Green's functions as a 2×2 matrix. Consider the column vector $X_{\mathbf{k}}$, with components

$$X_{\mathbf{k}}^1 = b_{\mathbf{k}}, \quad (3.1)$$

$$X_{\mathbf{k}}^2 = a_{-\mathbf{k}}^\dagger, \quad (3.2)$$

where \mathbf{k} is the momentum vector in reciprocal space, and

$$b_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} b_i, \quad (3.3)$$

$$a_{\mathbf{k}}^\dagger = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{k} \cdot \mathbf{r}_i} a_i^\dagger, \quad (3.4)$$

are the Fourier transforms of the lowering and raising operators. The Green's functions to be studied form a 2×2 matrix with elements $\langle\langle X_{\mathbf{k}}^\alpha; X_{\mathbf{k}}^\beta \rangle\rangle$, ($\alpha, \beta = 1, 2$),

$$g(\vec{\mathbf{k}}, E) = \begin{pmatrix} \langle\langle b_{\mathbf{k}}; b_{\mathbf{k}}^\dagger \rangle\rangle & \langle\langle b_{\mathbf{k}}; a_{-\mathbf{k}} \rangle\rangle \\ \langle\langle a_{-\mathbf{k}}^\dagger; b_{\mathbf{k}} \rangle\rangle & \langle\langle a_{-\mathbf{k}}^\dagger; a_{-\mathbf{k}} \rangle\rangle \end{pmatrix}. \quad (3.5)$$

We shall represent this symbolically simply as $\langle\langle X_{\mathbf{k}}; X_{\mathbf{k}}^\dagger \rangle\rangle$. The Green's function $\langle\langle A; B \rangle\rangle_E$ is the temporal fourier transform of the double-time Green's function $\langle\langle A(t); B(t') \rangle\rangle_{\mathbf{r}}$, taking the retarded and advanced functions in the upper and lower half-plane, respectively,

$$\langle\langle A; B \rangle\rangle_E = \int_{-\infty}^{\infty} dt e^{iE(t-t')} \langle\langle A(t); B(t') \rangle\rangle_{\mathbf{r}}. \quad (3.6)$$

We shall be discussing only the Fourier transform $\langle\langle A; B \rangle\rangle_E$ and shall drop the subscript E which indicates this. For a review of the Green's function method, the reader is referred to Zubarev²⁷ and Mavroyannis.²⁸

The equations of motion of the Green's functions are determined by the commutator of the operators b_i and a_i^\dagger with the Hamiltonian. It is more convenient to calculate these commutators in real space and then transform to momentum space to obtain

$$[b_{\mathbf{k}}, \mathcal{H}] = \bar{\omega}_{\mathbf{k}} b_{\mathbf{k}} + \hat{\omega}_{\mathbf{k}} a_{-\mathbf{k}}^\dagger + F_{\mathbf{k}}^1, \quad (3.7)$$

$$-[a_{-\mathbf{k}}^\dagger, \mathcal{H}] = \bar{\omega}_{\mathbf{k}} a_{-\mathbf{k}}^\dagger + \hat{\omega}_{\mathbf{k}} b_{\mathbf{k}} + F_{\mathbf{k}}^2, \quad (3.8)$$

where

$$\bar{\omega}_{\mathbf{k}} = \mathcal{J}_0 - \frac{1}{2} \mathcal{J}_{\mathbf{k}}, \quad (3.9)$$

$$\hat{\omega}_{\mathbf{k}} = \frac{1}{2} \mathcal{J}_{\mathbf{k}}, \quad (3.10)$$

and $\mathcal{J}_{\mathbf{r}}$ is related to the Fourier transform of the quadrupolar coupling constant

$$\mathcal{J}_{\mathbf{r}} = \sum_j e^{-i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)} J_{ij}. \quad (3.11)$$

The coupling J_{ij} is assumed to depend only on the magnitude of separation $(\mathbf{r}_i - \mathbf{r}_j)$; thus $\mathcal{J}_{\mathbf{r}}$ is an even function of momentum. On the simple-cubic lattice with only nearest-neighbor coupling $J_{ij} = J$, the parameters $\bar{\omega}_{\mathbf{r}}$ and $\hat{\omega}_{\mathbf{r}}$ both take the value $3J$ at the origin $k=0$; $\bar{\omega}_{\mathbf{r}}$ increases to a value of $6J$ at $(\pi/2a, \pi/2a, \pi/2a)$, whereas $\hat{\omega}_{\mathbf{r}}$ decreases to zero and on to a value of $-3J$ at $(\pi/a, \pi/a, \pi/a)$, whereas $\bar{\omega}_{\mathbf{r}}$ increases to $9J$ (a is the nearest-neighbor distance). The higher-order terms in the commutator have been collected into the vector $F_{\mathbf{r}}$ with components

$$F_i^1 = - \sum_j J_{ij} \left(\frac{3}{2} b_i(n_j + m_j) + \frac{1}{\sqrt{2}} (n_i + 2m_i) L_2^1(j) - \frac{1}{\sqrt{2}} L_2^2(i) L_2^{-1}(j) + a_i L_2^2(j) \right), \quad (3.12)$$

$$F_i^2 = - \sum_j J_{ij} \left(\frac{3}{2} a_i^{\dagger}(n_j + m_j) - \frac{1}{\sqrt{2}} (2n_i + m_i) L_2^1(j) + \frac{1}{\sqrt{2}} L_2^2(i) L_2^{-1}(j) + b_i^{\dagger} L_2^2(j) \right). \quad (3.13)$$

From the commutation relations (3.7) and (3.8) follow the equations of motion for the Green's functions

$$\frac{1}{-Q} \begin{pmatrix} E - \bar{\omega}_{\mathbf{r}} & -\hat{\omega}_{\mathbf{r}} \\ -\hat{\omega}_{\mathbf{r}} & -E - \bar{\omega}_{\mathbf{r}} \end{pmatrix} \mathcal{G}(\mathbf{k}, E) = \mathcal{J} + \mathcal{F}(\mathbf{k}, E), \quad (3.14)$$

where \mathcal{J} is the unit matrix, $\mathcal{F}(\mathbf{k}, E)$ is a higher-order 2×2 Green's function

$$\mathcal{F}(\mathbf{k}, E) = \frac{1}{-Q} \langle \langle F_{\mathbf{r}}; X_{\mathbf{r}}^{\dagger} \rangle \rangle, \quad (3.15)$$

and Q , the order parameter (1.5), is the ensemble average of the quadrupolar operator Q_i ,

$$-Q = 1 - 3\langle n_i \rangle = \langle [b_{\mathbf{r}}, b_{\mathbf{r}}^{\dagger}] \rangle = \langle [a_{\mathbf{r}}, a_{\mathbf{r}}^{\dagger}] \rangle. \quad (3.16)$$

In the molecular-field approximation $-Q$ takes the value unity, and we anticipate Q to be negative in this treatment as well.

Before obtaining an equation of motion for the higher-order Green's function $\mathcal{F}(\mathbf{k}, E)$, we consider the zero-order approximation (ZOA) in which it is neglected entirely. Equation (3.14) can then be inverted to obtain

$$\mathcal{G}_0(\mathbf{k}, E) = \frac{-Q}{E^2 - \bar{\omega}_{\mathbf{r}}^2} \begin{pmatrix} E + \bar{\omega}_{\mathbf{r}} & -\hat{\omega}_{\mathbf{r}} \\ -\hat{\omega}_{\mathbf{r}} & -E + \bar{\omega}_{\mathbf{r}} \end{pmatrix}, \quad (3.17)$$

with the simple poles $\pm \omega_{\mathbf{r}}$ occurring on the real axis at

$$\omega_{\mathbf{r}} = \left(\bar{\omega}_{\mathbf{r}}^2 - \hat{\omega}_{\mathbf{r}}^2 \right)^{1/2} = \mathcal{J}_0 (1 - \mathcal{J}_{\mathbf{r}}/\mathcal{J}_0)^{1/2}. \quad (3.18)$$

As expected, the excitation energy shows no gap at $k=0$. On the nearest-neighbor simple-cubic lattice along the $k_x = k_y = k_z$ direction the energy has a sinusoidal behavior reaching a maximum of $6\sqrt{2}J$ at $(\pi/a, \pi/a, \pi/a)$.

The various correlation functions can be obtained by taking the difference of the appropriate Green's function across the real axis branch cut

$$\bar{n}_{\mathbf{r}} = (1/-Q) \langle a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}} \rangle = (1/-Q) \langle b_{\mathbf{r}}^{\dagger} b_{\mathbf{r}} \rangle = \frac{1}{2} \{ (\bar{\omega}_{\mathbf{r}}/\omega_{\mathbf{r}}) \coth \frac{1}{2} \beta \omega_{\mathbf{r}} - 1 \}, \quad (3.19)$$

$$\hat{n}_{\mathbf{r}} = (1/-Q) \langle a_{-\mathbf{r}} b_{\mathbf{r}} \rangle = (1/-Q) \langle b_{\mathbf{r}}^{\dagger} a_{-\mathbf{r}}^{\dagger} \rangle = -(\hat{\omega}_{\mathbf{r}}/2\omega_{\mathbf{r}}) \coth(\beta\omega_{\mathbf{r}}/2), \quad (3.20)$$

where $\beta = 1/k_B T$ is the inverse temperature. We note that in the limit of zero temperature both occupation numbers

$$\bar{n}_{\mathbf{r}} = \frac{1}{2} (\bar{\omega}_{\mathbf{r}}/\omega_{\mathbf{r}} - 1), \quad (3.21)$$

$$\hat{n}_{\mathbf{r}} = -\hat{\omega}_{\mathbf{r}}/2\omega_{\mathbf{r}}, \quad (3.22)$$

are nonzero, reflecting that the molecular-field ground state is not exact. Again considering the nearest-neighbor simple-cubic model, $\bar{n}_{\mathbf{r}}$ and $\hat{n}_{\mathbf{r}}$ both diverge as plus and minus $0.6124/ka$, respectively, near the origin. At $(\pi/2a, \pi/2a, \pi/2a)$ $\bar{n}_{\mathbf{r}}$ has dropped to zero and $\hat{n}_{\mathbf{r}}$ has increased to zero; $\hat{n}_{\mathbf{r}}$ continues to increase to the value 0.1768 at $(\pi/a, \pi/a, \pi/a)$, and $\bar{n}_{\mathbf{r}}$ remains positive increasing to the value 0.0303.

The thermal average of the number operators is obtained by summing the correlation functions over momentum. It is convenient to divide out the factor of $-Q$ in the definition of this average

$$\bar{n} = \frac{1}{-Q} \langle n_i \rangle = \frac{1}{N} \sum_{\mathbf{r}} \bar{n}_{\mathbf{r}}, \quad (3.23)$$

$$\hat{n} = \frac{1}{N} \sum_{\mathbf{r}} \hat{n}_{\mathbf{r}}. \quad (3.24)$$

The expectation value of the quadrupole operator Q_i can be expressed in terms of the diagonal number operator \bar{n}

$$-Q = 1/(1 + 3\bar{n}). \quad (3.25)$$

The off-diagonal number operator \hat{n} should be identically zero by Eq. (2.6). Note that $\hat{n}_{\mathbf{r}}$ takes on both positive and negative values, whereas $\bar{n}_{\mathbf{r}}$ is entirely positive, so that the integral (3.24) for \hat{n} is considerably smaller than (3.23) for \bar{n} . However it is not identically zero. We return to the question whether this is a serious fault of the theory later. In both cases the integrand is singular at $q=0$; however, the integral is convergent in two and three dimensions.

In order to go beyond the zero-order approximation an equation of motion for the higher order Green's function $\mathfrak{F}(\vec{k}, E)$ must be obtained. To do this we take the commutator of the second argument $X_{\vec{r}}^{\dagger}$ of $\mathfrak{F}(\vec{k}, E)$ with the Hamiltonian, rather than the first $F_{\vec{r}}$. Using the Hermitian conjugate of the commutation relations (3.7) and (3.8) we obtain the equation

$$\mathfrak{F}(\vec{k}, E) \mathfrak{g}_0^{-1}(\vec{k}, E) = \mathcal{O}(\vec{k}, E), \quad (3.26)$$

where the right-hand side is the sum of two terms

$$\mathcal{O}(\vec{k}, E) = \mathcal{O}_0(\vec{k}) + \mathcal{O}_1(\vec{k}, E). \quad (3.27)$$

The first is a static term related to the ensemble average of the commutator of $F_{\vec{r}}$ with $X_{\vec{r}}^{\dagger}$,

$$\mathcal{O}_0(\vec{k}) = \frac{1}{Q^2} \begin{pmatrix} \langle [F_{\vec{r}}^1, b_{\vec{r}}^{\dagger}] \rangle - \langle [F_{\vec{r}}^1, a_{-\vec{r}}] \rangle \\ \langle [F_{\vec{r}}^2, b_{\vec{r}}^{\dagger}] \rangle - \langle [F_{\vec{r}}^2, a_{-\vec{r}}] \rangle \end{pmatrix}. \quad (3.28)$$

The second term is a still-higher-order Green's function

$$\mathcal{O}_1(\vec{k}, E) = (1/Q^2) \langle \langle F_{\vec{r}}; F_{\vec{r}}^{\dagger} \rangle \rangle. \quad (3.29)$$

Combining Eq. (3.26) with the equation of motion for the one-particle Green's function

$$\mathfrak{g}_0^{-1}(\vec{k}, E) \mathfrak{g}(\vec{k}, E) = \mathcal{g} + \mathfrak{F}(\vec{k}, E), \quad (3.14')$$

gives

$$\mathfrak{g}(\vec{k}, E) = \mathfrak{g}_0(\vec{k}, E) + \mathfrak{g}_0(\vec{k}, E) \mathcal{O}(\vec{k}, E) \mathfrak{g}_0(\vec{k}, E). \quad (3.30)$$

If a self-energy operator is defined by

$$\Sigma(\vec{k}, E) = \mathcal{O}(\vec{k}, E) [\mathcal{g} + \mathfrak{g}_0(\vec{k}, E) \mathcal{O}(\vec{k}, E)]^{-1}, \quad (3.31)$$

then we obtain formally a Dyson equation for the single-particle Green's function

$$[\mathfrak{g}_0^{-1}(\vec{k}, E) - \Sigma(\vec{k}, E)] \mathfrak{g}(\vec{k}, E) = \mathcal{g}. \quad (3.32)$$

The problem now is to determine the self-energy operator $\Sigma(\vec{k}, E)$.

IV. RENORMALIZED ZERO-ORDER APPROXIMATION

The simplest extension of the zeroth-order approximation is to consider the effect of the static term $\mathcal{O}_0(\vec{k})$ on the self-energy. Since it is time-independent, the poles of the Green's function remain on the real axis. The only effect is a temperature-dependent renormalization of the excitation energies $\omega_{\vec{r}}$. We also approximate the self-energy to the leading term in the expansion (3.31),

$$\Sigma(\vec{k}, E) \approx \mathcal{O}(\vec{k}, E) \approx \mathcal{O}_0(\vec{k}). \quad (4.1)$$

For the Heisenberg ferromagnet this has been shown to be equivalent to the random phase approximation.²⁹

Using the commutation relations (2.7) and (2.8), $\mathcal{O}_0(\vec{k})$ can be determined in a relatively straightforward fashion. Calculating the commutator of F_i with X_r^{\dagger} in real space gives

$$[F_i^1, b_r^{\dagger}] \approx - \left(\delta_{i,r} \sum_j \left(\frac{1}{2} J_{rj} \right) \{ 3(n_j + m_j) + \sqrt{2} [2L_2^1(j) b_r^{\dagger} - L_2^{-1}(j) a_r^{\dagger}] \} + \frac{1}{2} J_{ir} (3b_i b_r^{\dagger} + 2a_i a_r^{\dagger} - n_i - 2m_i) \right) (1 - n_r - 2m_r) \quad (4.2)$$

and

$$[F_i^1, a_r] \approx (1 - 2n_r - m_r) \left(\delta_{i,r} \sum_j \sqrt{2} \left(\frac{1}{2} J_{rj} \right) [a_r L_2^1(j) - b_r L_2^{-1}(j)] + \frac{1}{2} J_{ir} (3a_r b_i + 2b_r a_i + n_i + 2m_i) \right). \quad (4.3)$$

A number of identically zero terms, such as $b_r^{\dagger} a_r^{\dagger} a_r$, have been included in (4.2) and (4.3) in order to obtain $(1 - n_r - 2m_r)$ or $(1 - 2n_r - m_r)$ as a common factor. We have also dropped higher-order terms involving $L_2^2(r) L_2^{-2}(j)$.

Consistent with the random phase approximation, we assume the product of quadrupole operator $(1 - n_r - 2m_r)$ or $(1 - 2n_r - m_r)$ with the other operators [in large parentheses in Eqs. (4.2) and (4.3)] factorizes when the thermal average is taken,

$$\begin{aligned} \langle [F_i^1, b_r^{\dagger}] \rangle \approx Q \left[\delta_{i,r} \sum_j \left(\frac{1}{2} J_{rj} \right) (3 \langle a_j^{\dagger} a_j \rangle + 3 \langle b_j^{\dagger} b_j \rangle - \langle a_r^{\dagger} a_r \rangle - 2 \langle b_r^{\dagger} b_r \rangle + 2 \langle b_r^{\dagger} a_r^{\dagger} \rangle + \langle b_r^{\dagger} a_r \rangle) \right. \\ \left. - \left(\frac{1}{2} J_{ir} \right) (\langle a_i^{\dagger} a_i \rangle + 2 \langle b_i^{\dagger} b_i \rangle - 2 \langle a_r^{\dagger} a_i \rangle - 3 \langle b_r^{\dagger} b_i \rangle) \right], \end{aligned} \quad (4.4)$$

and

$$\langle [F_i^1, a_r] \rangle \approx (-Q) \left[\delta_{i,r} \sum_j \frac{1}{2} J_{rj} (\langle a_j^{\dagger} a_r \rangle + \langle b_j^{\dagger} b_r \rangle - \langle a_r b_j \rangle - \langle a_j b_r \rangle) + \left(\frac{1}{2} J_{ir} \right) (\langle a_i^{\dagger} a_i \rangle + 2 \langle b_i^{\dagger} b_i \rangle + 3 \langle a_r b_i \rangle + 2 \langle a_i b_r \rangle) \right]. \quad (4.5)$$

Finally, taking the Fourier transform, we obtain an expression for the static contribution to the self-energy operator

$$P_0^{11}(\vec{k}) = (1/Q^2) \langle [F_{\vec{k}}^\dagger, b_{\vec{k}}^\dagger] \rangle \\ = -3\bar{\omega}_{\vec{k}} - 5\bar{S}_{\vec{k}} - 3(\hat{S}_0 - \bar{S}_0), \quad (4.6)$$

$$P_0^{12}(\vec{k}) = -(1/Q^2) \langle [F_{\vec{k}}^\dagger, a_{-\vec{k}}] \rangle \\ = -3\bar{\omega}_{\vec{k}} - 5\hat{S}_{\vec{k}} + 2(\hat{S}_0 - \bar{S}_0), \quad (4.7)$$

where $\bar{S}_{\vec{k}}$ and $\hat{S}_{\vec{k}}$ are sums over the diagonal and off-diagonal correlation function, respectively,

$$\bar{S}_{\vec{k}} = \frac{1}{N} \sum_{\vec{q}} \hat{\omega}_{\vec{k}-\vec{q}} \bar{n}_{\vec{q}}, \quad (4.8)$$

$$\hat{S}_{\vec{k}} = \frac{1}{N} \sum_{\vec{q}} \hat{\omega}_{\vec{k}-\vec{q}} \hat{n}_{\vec{q}}. \quad (4.9)$$

Making the same approximations, one can show the matrix $\sigma_0(\vec{k})$ is symmetric, $P_0^{21}(\vec{k}) = P_0^{12}(\vec{k})$, and in addition $P_0^{22}(\vec{k}) = P_0^{11}(\vec{k})$.

The renormalized zeroth-order (RZO) expression for the one-particle Green's function is similar to the ZOA expression

$$g^{-1}(\vec{k}, E) = \frac{1}{-Q} \begin{pmatrix} E - \bar{\Omega}_{\vec{k}} & -\hat{\Omega}_{\vec{k}} \\ -\hat{\Omega}_{\vec{k}} & -E - \bar{\Omega}_{\vec{k}} \end{pmatrix}, \quad (4.10)$$

except that the coupling constants are replaced by their renormalized values

$$\bar{\Omega}_{\vec{k}} = \bar{\omega}_{\vec{k}} + (-Q)P_0^{11}(\vec{k}) \\ = (-Q)\{\bar{\omega}_{\vec{k}} - [5\bar{S}_{\vec{k}} + 3(\hat{S}_0 - \bar{S}_0)]\}, \quad (4.11)$$

$$\hat{\Omega}_{\vec{k}} = \hat{\omega}_{\vec{k}} + (-Q)P_0^{12}(\vec{k}) \\ = (-Q)\{\hat{\omega}_{\vec{k}} - [5\bar{S}_{\vec{k}} - 2(\hat{S}_0 - \bar{S}_0)]\}. \quad (4.12)$$

The poles of the Green's function are shifted but remain on the real axis at $\pm\Omega_{\vec{k}}$, where

$$\Omega_{\vec{k}} = (\bar{\Omega}_{\vec{k}}^2 - \hat{\Omega}_{\vec{k}}^2)^{1/2} \\ = (-Q)(\omega_{\vec{k}}^2 + \Delta_{\vec{k}})^{1/2}. \quad (4.13)$$

The shift $\Delta_{\vec{k}}$ is determined in terms of the sums (4.8) and (4.9). We note that the shift is zero for zero momentum, so there is still no gap in the excitation spectrum.

Equations (3.19)–(3.22) for the correlation functions remain valid in the RZO except that the parameters $\bar{\omega}_{\vec{k}}$, $\hat{\omega}_{\vec{k}}$, and $\omega_{\vec{k}}$ must be replaced by their renormalized values $\bar{\Omega}_{\vec{k}}$, $\hat{\Omega}_{\vec{k}}$, and $\Omega_{\vec{k}}$. However, now they must be solved by an iterative procedure, as the parameters $\bar{\Omega}_{\vec{k}}$, $\hat{\Omega}_{\vec{k}}$, and $\Omega_{\vec{k}}$ depend on the correlation function $\bar{n}_{\vec{k}}$ through the order parameter Q . At the same time the off-diagonal correlations $\hat{n}_{\vec{k}}$ can be calculated and the sum \hat{n} will still presumably be nonzero, violating Eq. (2.6).

Barma¹⁷ as well as Fittipaldi and Tahir-Kheli¹⁸

has introduced a Callen-like decoupling procedure to ensure that the off-diagonal self-correlation sum rule $\hat{n} = 0$ is satisfied. This results in four equations to be solved self-consistently, which severely limits the extension of the theory. They found this necessary in order to symmetrize their random-phase approximation (RPA) decoupling, so as to obtain a unique result for the diagonal self-correlation \bar{n} . However our decoupling scheme is already symmetric and there is no ambiguity in the value of the order parameter, whether it is calculated from $\langle a_i^\dagger a_i \rangle$ or $\langle b_i^\dagger b_i \rangle$. We also note the violation of the self-correlation sum rule is not a fault of the RPA but is inherent in the ZOA, to begin with.

V. EFFECTIVE HAMILTONIAN

In order to determine the lifetime of the collective modes we must calculate the higher-order Green's function $\sigma_1(\vec{k}, E)$. This is a rather formidable task. In order to simplify matters we replace the Hamiltonian (1.6) with an effective Hamiltonian, which contains only quadratic terms. The coefficients of these terms are chosen to reproduce the RZO exactly. The quadratic Hamiltonian can be diagonalized by a canonical transformation. We then determine the Green's function $\sigma_1(\vec{k}, E)$ with the diagonal Hamiltonian.

It is convenient to introduce the new parameters $u_{\vec{k}}$ and $v_{\vec{k}}$, defined in terms of $\bar{\Omega}_{\vec{k}}$ and $\hat{\Omega}_{\vec{k}}$,

$$u_{\vec{k}}^2 = (1 + \bar{\Omega}_{\vec{k}}/\Omega_{\vec{k}})/(-2Q), \quad (5.1)$$

$$v_{\vec{k}}^2 = (-1 + \bar{\Omega}_{\vec{k}}/\Omega_{\vec{k}})/(-2Q). \quad (5.2)$$

These parameters weight the positive and negative poles of the diagonal Green's function in the RZO,

$$G^{11}(\vec{k}, E) = G^{22}(\vec{k}, -E) = Q^2 \left\{ \frac{u_{\vec{k}}^2}{E - \Omega_{\vec{k}}} + \frac{v_{\vec{k}}^2}{-E - \Omega_{\vec{k}}} \right\}. \quad (5.3)$$

The product of the two parameters

$$u_{\vec{k}} v_{\vec{k}} = -\hat{\Omega}_{\vec{k}}/(-2Q\Omega_{\vec{k}}), \quad (5.4)$$

weights both poles of the off-diagonal Green's functions

$$G^{12}(\vec{k}, E) = G^{21}(\vec{k}, E) \\ = Q^2 u_{\vec{k}} v_{\vec{k}} \left\{ \frac{1}{E - \Omega_{\vec{k}}} + \frac{1}{-E - \Omega_{\vec{k}}} \right\}. \quad (5.5)$$

We note that $(-Q)v_{\vec{k}}^2$ equals the diagonal correlation function $\bar{n}_{\vec{k}}$ at $T=0$ and hence is a relatively small parameter. In the nearest-neighbor simple-cubic model, it is zero at $(\pi/2a, \pi/2a, \pi/2a)$ and small throughout most of the Brillouin zone (the divergence at the origin is unimportant in three dimensions). The factor $(-Q)u_{\vec{k}}^2$ is, on the other hand,

close to unity over a large part of the Brillouin zone, since

$$u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2 = \frac{1}{-Q}. \quad (5.6)$$

The product $(-Q)u_{\mathbf{k}}v_{\mathbf{k}}$ equals the off-diagonal correlation function $\hat{n}_{\mathbf{k}}$ at $T=0$ and hence oscillates in sign through the Brillouin zone.

The first step to obtain an effective Hamiltonian is to approximate the operators $a_{\mathbf{q}}$ and $b_{\mathbf{q}}$ by quasiboson operators. If we assume

$$\sum_{\mathbf{k}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}+\mathbf{q}-\mathbf{p}} \approx \delta_{\mathbf{q},\mathbf{p}} \sum_{\mathbf{k}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}}, \quad (5.7)$$

the exact commutation relations

$$[b_{\mathbf{q}}, b_{\mathbf{p}}^\dagger] = \delta_{\mathbf{q},\mathbf{p}} - \frac{1}{N} \sum_{\mathbf{k}} (a_{\mathbf{k}}^\dagger a_{\mathbf{k}+\mathbf{q}-\mathbf{p}} + 2b_{\mathbf{k}}^\dagger b_{\mathbf{k}+\mathbf{q}-\mathbf{p}}), \quad (5.8)$$

$$[a_{\mathbf{q}}, a_{\mathbf{p}}^\dagger] = \delta_{\mathbf{q},\mathbf{p}} - \frac{1}{N} \sum_{\mathbf{k}} (2a_{\mathbf{k}}^\dagger a_{\mathbf{k}+\mathbf{q}-\mathbf{p}} + b_{\mathbf{k}}^\dagger b_{\mathbf{k}+\mathbf{q}-\mathbf{p}}), \quad (5.9)$$

reduce to the quasiboson relations

$$[b_{\mathbf{q}}, b_{\mathbf{p}}^\dagger] = [a_{\mathbf{q}}, a_{\mathbf{p}}^\dagger] = (-Q)\delta_{\mathbf{q},\mathbf{p}}. \quad (5.10)$$

We now replace the exact Hamiltonian

$$\begin{aligned} \mathcal{H} = & -\frac{1}{2}NJ_0 + \sum_{\mathbf{k}} [\bar{\omega}_{\mathbf{k}}(a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + b_{\mathbf{k}}^\dagger b_{\mathbf{k}}) + \hat{\omega}_{\mathbf{k}}(a_{-\mathbf{k}} b_{\mathbf{k}} + a_{\mathbf{k}}^\dagger b_{-\mathbf{k}}^\dagger)] \\ & - \frac{1}{2} \sum_{\mathbf{k}, \mathbf{l}, \mathbf{m}, \mathbf{n}} \hat{\omega}_{\mathbf{k}-\mathbf{l}} \delta_{\mathbf{k}-\mathbf{l}, \mathbf{n}-\mathbf{m}} [3(a_{\mathbf{k}}^\dagger a_{\mathbf{l}}^\dagger a_{\mathbf{m}}^\dagger a_{\mathbf{n}} + b_{\mathbf{k}}^\dagger b_{\mathbf{l}}^\dagger b_{\mathbf{m}}^\dagger b_{\mathbf{n}}) \\ & + 6a_{\mathbf{k}}^\dagger a_{\mathbf{l}}^\dagger b_{\mathbf{m}}^\dagger b_{\mathbf{n}} + 4a_{\mathbf{k}}^\dagger b_{\mathbf{l}}^\dagger b_{\mathbf{m}}^\dagger a_{\mathbf{n}}], \end{aligned} \quad (5.11)$$

where the operators satisfy the commutation relations (5.8) and (5.9) with the effective Hamiltonian

$$\mathcal{H}_{\text{eff}} = \frac{1}{-2Q} \sum_{\mathbf{k}} [\bar{\Omega}_{\mathbf{k}}(a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + b_{\mathbf{k}}^\dagger b_{\mathbf{k}}) + \hat{\Omega}_{\mathbf{k}}(a_{-\mathbf{k}} b_{\mathbf{k}} + a_{\mathbf{k}}^\dagger b_{-\mathbf{k}}^\dagger)], \quad (5.12)$$

where the operators satisfy the quasiboson commutation relations (5.10). In addition we have assumed $a_{\mathbf{q}} = b_{\mathbf{q}}$ (this accounts for the factor of 2 in the denominator). The only effect of the interaction terms in the Hamiltonian (5.11) which has been retained is the renormalization of the coupling constants $\bar{\omega}_{\mathbf{k}}$ and $\hat{\omega}_{\mathbf{k}}$. The one-particle Green's functions for this quadratic Hamiltonian are the same as those for the exact Hamiltonian (1.6) in the RZQA [that is, Eqs. (5.3) and (5.5)].

The quadratic Hamiltonian (5.12) is diagonalized by the canonical transformation

$$\begin{aligned} b_{\mathbf{k}} &= a_{\mathbf{k}} = (-Q)(u_{\mathbf{k}} \xi_{\mathbf{k}} - v_{\mathbf{k}} \xi_{-\mathbf{k}}^\dagger), \\ b_{\mathbf{k}}^\dagger &= a_{\mathbf{k}}^\dagger = (-Q)(u_{\mathbf{k}} \xi_{\mathbf{k}}^\dagger - v_{\mathbf{k}} \xi_{-\mathbf{k}}). \end{aligned} \quad (5.13)$$

In terms of the transformed operators the effective Hamiltonian becomes

$$\mathcal{H}_{\text{eff}} = E_0 + \sum_{\mathbf{k}} \Omega_{\mathbf{k}} \xi_{\mathbf{k}}^\dagger \xi_{\mathbf{k}}, \quad (5.14)$$

where the ground-state energy is

$$\frac{E_0}{N} = -\frac{1}{3}J_0 + \frac{1}{2N} \sum_{\mathbf{k}} (\bar{\Omega}_{\mathbf{k}} - \Omega_{\mathbf{k}}). \quad (5.15)$$

From the inverse of the transformation (5.13), it is easily verified from Eq. (5.10) that the operators $\xi_{\mathbf{k}}$ and $\xi_{\mathbf{k}}^\dagger$ satisfy boson commutation relations

$$[\xi_{\mathbf{q}}, \xi_{\mathbf{p}}^\dagger] = \delta_{\mathbf{q},\mathbf{p}}. \quad (5.16)$$

Henceforth, we shall limit our attention to zero temperature, where the expectation value of the number operator $\langle \xi_{\mathbf{k}}^\dagger \xi_{\mathbf{k}} \rangle$ is identically zero.

VI. POLARIZATION OPERATOR

We are now in a position to include the effect of scattering, described by the time-dependent polarization operator $\mathcal{O}_1(\mathbf{k}, E)$, on the self-energy $\Sigma(\mathbf{k}, E)$. The three-particle Green's function has an imaginary contribution to the self-energy, which pushes the poles of the Green's function off the real axis. Accordingly the δ -function spectrum of the RZOA becomes Lorentzian.

With the approximation $b_i = a_i$, introduced in the preceding section the polarization operator (3.29) reduces to

$$\langle\langle F_{\mathbf{k}}; F_{\mathbf{k}}^\dagger \rangle\rangle = \begin{pmatrix} \langle\langle f_{\mathbf{k}}; f_{\mathbf{k}}^\dagger \rangle\rangle & \langle\langle f_{\mathbf{k}}; f_{-\mathbf{k}} \rangle\rangle \\ \langle\langle f_{-\mathbf{k}}^\dagger; f_{\mathbf{k}}^\dagger \rangle\rangle & \langle\langle f_{-\mathbf{k}}^\dagger; f_{-\mathbf{k}} \rangle\rangle \end{pmatrix}, \quad (6.1)$$

where $f_{\mathbf{k}} = F_{\mathbf{k}}^1 = (F_{\mathbf{k}}^2)^\dagger$ is given by

$$\begin{aligned} f_{\mathbf{k}} = & -\frac{4}{N} \sum_{\mathbf{q}, \mathbf{p}} [2\hat{\omega}_{\mathbf{k}-\mathbf{q}} a_{\mathbf{p}-\mathbf{k}}^\dagger a_{\mathbf{p}-\mathbf{q}} a_{\mathbf{q}} \\ & + \hat{\omega}_{\mathbf{q}} a_{\mathbf{p}-\mathbf{k}}^\dagger a_{\mathbf{p}-\mathbf{q}} (a_{\mathbf{q}}^\dagger - a_{\mathbf{q}})]. \end{aligned} \quad (6.2)$$

This expression is now written in terms of the transformed variable $\xi_{\mathbf{q}}$. Since the effective Hamiltonian is diagonal, the equations of motion of the resulting three-particle Green's functions can be readily determined. There are only two types of terms which contribute to either $\langle\langle f_{\mathbf{k}}; f_{\mathbf{k}}^\dagger \rangle\rangle$ or $\langle\langle f_{\mathbf{k}}; f_{-\mathbf{k}} \rangle\rangle$. The first is the combination of three annihilation operators with three creation operators $\langle\langle \xi_i \xi_m \xi_n; \xi_n^\dagger \xi_m^\dagger \xi_i^\dagger \rangle\rangle$ and its Hermitian conjugate. The second is the nine permutations of $\langle\langle \xi_i^\dagger \xi_m \xi_n; \xi_n^\dagger \xi_m^\dagger \xi_i \rangle\rangle$ and their Hermitian conjugates. We shall show later that the second type does not contribute to the scattering, at zero temperature. It only leads to a further renormalization of the excitation spectrum. At present we consider the

two terms corresponding to the first type; they will contribute to the lifetime of the collective modes.

For bookkeeping purposes it is convenient to define the coupling parameter

$$K_{\vec{q}}^u(\vec{k}) = 2\hat{\omega}_{\vec{k}-\vec{q}}u_{\vec{q}} - \hat{\omega}_{\vec{q}}(u_{\vec{q}} + v_{\vec{q}}). \quad (6.3)$$

$K_{\vec{q}}^v(\vec{k})$ is defined by (6.3) with u replaced by v . Considering only the frequency-dependent contribution the polarization operator can be expressed as

$$P_1^{11}(\vec{k}, E) = \frac{16Q^4}{N^2} \sum_{\vec{q}, \vec{p}, \vec{q}', \vec{p}'} [u_{\vec{p}-\vec{q}}v_{\vec{p}-\vec{q}'}K_{\vec{q}}^u(\vec{k})u_{\vec{p}-\vec{q}'}v_{\vec{p}-\vec{q}}K_{\vec{q}'}^u(\vec{k}) \langle\langle \xi_{\vec{k}-\vec{p}} \xi_{\vec{p}-\vec{q}} \xi_{\vec{q}}^{\dagger}; \xi_{\vec{q}}^{\dagger} \xi_{\vec{p}-\vec{q}'}^{\dagger} \xi_{\vec{k}-\vec{p}'}^{\dagger} \rangle\rangle + u_{\vec{p}-\vec{q}}v_{\vec{p}-\vec{q}'}K_{\vec{q}}^v(\vec{k})u_{\vec{p}-\vec{q}}v_{\vec{p}-\vec{q}'}K_{\vec{q}'}^v(\vec{k}) \langle\langle \xi_{\vec{k}-\vec{p}}^{\dagger} \xi_{\vec{p}-\vec{q}}^{\dagger} \xi_{\vec{q}}^{\dagger}; \xi_{\vec{q}} \xi_{\vec{p}-\vec{q}'} \xi_{\vec{k}-\vec{p}'} \rangle\rangle], \quad (6.4)$$

and

$$P_1^{12}(\vec{k}, E) = -\frac{16Q^4}{N^2} \sum_{\vec{q}, \vec{p}, \vec{q}', \vec{p}'} [u_{\vec{p}-\vec{q}}v_{\vec{p}-\vec{q}'}K_{\vec{q}}^u(\vec{k})u_{\vec{p}-\vec{q}'}v_{\vec{p}-\vec{q}}K_{\vec{q}'}^v(\vec{k}) \langle\langle \xi_{\vec{k}-\vec{p}} \xi_{\vec{p}-\vec{q}} \xi_{\vec{q}}^{\dagger}; \xi_{\vec{q}}^{\dagger} \xi_{\vec{p}-\vec{q}'}^{\dagger} \xi_{\vec{k}-\vec{p}'}^{\dagger} \rangle\rangle + u_{\vec{p}-\vec{q}}v_{\vec{p}-\vec{q}'}K_{\vec{q}}^v(\vec{k})u_{\vec{p}-\vec{q}}v_{\vec{p}-\vec{q}'}K_{\vec{q}'}^u(\vec{k}) \langle\langle \xi_{\vec{k}-\vec{p}}^{\dagger} \xi_{\vec{p}-\vec{q}}^{\dagger} \xi_{\vec{q}}^{\dagger}; \xi_{\vec{q}} \xi_{\vec{p}-\vec{q}'} \xi_{\vec{k}-\vec{p}'} \rangle\rangle]. \quad (6.5)$$

In the diagonal representation, the equations of motion of the three-particle Green's functions follow directly

$$[E - \epsilon_{\vec{q}, \vec{p}}(\vec{k})] \langle\langle \xi_{\vec{k}-\vec{p}} \xi_{\vec{p}-\vec{q}} \xi_{\vec{q}}^{\dagger}; \xi_{\vec{q}}^{\dagger} \xi_{\vec{p}-\vec{q}'}^{\dagger} \xi_{\vec{k}-\vec{p}'}^{\dagger} \rangle\rangle = [-E - \epsilon_{\vec{q}, \vec{p}}(\vec{k})] \langle\langle \xi_{\vec{k}-\vec{p}}^{\dagger} \xi_{\vec{p}-\vec{q}}^{\dagger} \xi_{\vec{q}}^{\dagger}; \xi_{\vec{q}} \xi_{\vec{p}-\vec{q}'} \xi_{\vec{k}-\vec{p}'} \rangle\rangle = \Delta_{\vec{q}, \vec{p}}^{\vec{q}', \vec{p}'}(\vec{k}), \quad (6.6)$$

where the simple pole is at the sum of the three energies

$$\epsilon_{\vec{q}, \vec{p}}(\vec{k}) = \Omega_{\vec{k}-\vec{p}} + \Omega_{\vec{p}-\vec{q}} + \Omega_{\vec{q}}, \quad (6.7)$$

and the right-hand side reduces to a sum of δ -functions at zero temperature

$$\Delta_{\vec{q}, \vec{p}}^{\vec{q}', \vec{p}'}(\vec{k}) = \delta_{\vec{q}', \vec{p}'}(\delta_{\vec{p}, \vec{p}'} + \delta_{\vec{p}', \vec{k}-\vec{q}-\vec{p}}) + \delta_{\vec{q}', \vec{k}-\vec{p}}(\delta_{\vec{p}, \vec{k}-\vec{q}} + \delta_{\vec{p}', \vec{k}-\vec{q}-\vec{p}}) + \delta_{\vec{q}', \vec{p}-\vec{q}}(\delta_{\vec{p}, \vec{p}'} + \delta_{\vec{p}', \vec{k}-\vec{q}}). \quad (6.8)$$

The final expression for the polarization operator follows from inserting the solution (6.6) into Eqs. (6.4) and (6.5). Summing over the momenta \vec{q}' and \vec{p}' , we define

$$U_{\vec{q}, \vec{p}}(\vec{k}) = (u_{\vec{p}-\vec{q}}v_{\vec{k}-\vec{p}} + u_{\vec{k}-\vec{p}}v_{\vec{p}-\vec{q}})K_{\vec{q}}^u(\vec{k}) + (u_{\vec{q}}v_{\vec{p}-\vec{q}} + u_{\vec{p}-\vec{q}}v_{\vec{q}})K_{\vec{q}}^v(\vec{k}) + (u_{\vec{q}}u_{\vec{k}-\vec{p}} + u_{\vec{k}-\vec{p}}v_{\vec{q}})K_{\vec{p}-\vec{q}}^u(\vec{k}), \quad (6.9)$$

and $V_{\vec{q}, \vec{p}}(\vec{k})$ by the same equation, except u and v are interchanged. The weight for the positive pole of the diagonal Green's function $\langle\langle f_{\vec{q}}, f_{\vec{p}}^{\dagger} \rangle\rangle$ is

$$\bar{A}_{\vec{q}, \vec{p}}^{u, v}(\vec{k}) = 16(-Q)^3 u_{\vec{p}-\vec{q}}v_{\vec{k}-\vec{p}}K_{\vec{q}}^u(\vec{k})U_{\vec{q}, \vec{p}}(\vec{k}), \quad (6.10)$$

and for the negative pole $\bar{A}_{\vec{q}, \vec{p}}^{v, u}(\vec{k})$, which is the same expression with u and v interchanged. The weight for the positive pole of the off-diagonal Green's function $\langle\langle f_{\vec{q}}, f_{-\vec{q}} \rangle\rangle$ is

$$\hat{A}_{\vec{q}, \vec{p}}^{u, v}(\vec{k}) = 16(-Q)^3 u_{\vec{p}-\vec{q}}v_{\vec{k}-\vec{p}}K_{\vec{q}}^u(\vec{k})V_{\vec{q}, \vec{p}}(\vec{k}), \quad (6.11)$$

and similarly $\hat{A}_{\vec{q}, \vec{p}}^{v, u}(\vec{k})$ for the negative pole. Quite generally a Green's function obeys the symmetry property $\langle\langle A; B \rangle\rangle_{(E)} = \langle\langle B; A \rangle\rangle_{(-E)}$.²⁸ This relates the two diagonal elements of the polarization matrix

$$P_1^{22}(\vec{k}, -E) = P_1^{11}(\vec{k}, E) = P_{\vec{k}}(E)/(-Q), \quad (6.12)$$

and implies the off-diagonal elements are even functions of frequency

$$P_1^{12}(\vec{k}, E) = P_1^{12}(\vec{k}, -E) = -\hat{P}_{\vec{k}}(E)/(-Q). \quad (6.13)$$

We have assumed a cubic lattice so that all Green's functions and coupling parameters are even functions of momentum \vec{k} . Also the general symmetry relation $\langle\langle B^{\dagger}; A^{\dagger} \rangle\rangle_{(E)} = \langle\langle A; B \rangle\rangle_{(E)}$ implies the two off-diagonal elements are equal,

$$P_1^{21}(\vec{k}, E) = P_1^{12}(\vec{k}, E). \quad (6.14)$$

These general symmetry relations also follow from the explicit form of the coefficients \bar{A} and \hat{A} . For instance, Eq. (6.13) implies the coefficient $\hat{A}_{\vec{q}, \vec{p}}^{u, v}(\vec{k})$ is unchanged by the interchange of u and v , when divided by the energy denominator $[E - \epsilon_{\vec{q}, \vec{p}}(\vec{k})]$ and summed over \vec{q} and \vec{p} . This follows directly from (6.9) and (6.11) by interchanging the first factor $u_{\vec{p}-\vec{q}}v_{\vec{k}-\vec{p}}K_{\vec{q}}^u(\vec{k})$ with the six factors making up $V_{\vec{q}, \vec{p}}(\vec{k})$ and suitably changing the dummy indices in the sum (6.5) over \vec{q} and \vec{p} .

The final expressions for the frequency-dependent contribution to the polarization operator are

$$P_{\vec{k}}(E) = \left(\frac{Q}{N}\right)^2 \sum_{\vec{q}, \vec{p}} \left(\frac{\bar{A}_{\vec{q}, \vec{p}}^{u, v}(\vec{k})}{E - \epsilon_{\vec{q}, \vec{p}}(\vec{k})} + \frac{\bar{A}_{\vec{q}, \vec{p}}^{v, u}(\vec{k})}{-E - \epsilon_{\vec{q}, \vec{p}}(\vec{k})} \right) \quad (6.16)$$

and

$$\hat{P}_{\vec{k}}(E) = \left(\frac{Q}{N}\right)^2 \sum_{\vec{q}, \vec{p}} \hat{A}_{\vec{q}, \vec{p}}^{u, v}(\vec{k}) \left(\frac{1}{E - \epsilon_{\vec{q}, \vec{p}}(\vec{k})} + \frac{1}{-E - \epsilon_{\vec{q}, \vec{p}}(\vec{k})} \right). \quad (6.16)$$

Before proceeding with determining the lifetime of the collective modes, we shall examine more closely the terms we have neglected in the polarization operator. There are nine pairs of three-particle Green's functions of the form $\langle\langle \xi \xi \xi^\dagger; \xi \xi^\dagger \xi^\dagger \rangle\rangle$. At zero temperature, five of these pairs are zero. In the remaining four pairs, the sum of three frequencies $\epsilon_{\vec{q}, \vec{p}}(\vec{k})$ is reduced to the single term $\Omega_{\vec{k}}$ by the δ -function constraints on the right-hand side:

$$\begin{aligned} (E - \Omega_{\vec{k}}) \langle\langle \xi_{\vec{k}-\vec{p}} \xi_{\vec{q}-\vec{p}}^\dagger \xi_{\vec{q}}^\dagger; \xi_{\vec{q}}^\dagger \xi_{\vec{q}-\vec{p}} \xi_{\vec{k}-\vec{p}}^\dagger \rangle\rangle &= \delta_{\vec{q}, \vec{k}} \delta_{\vec{q}', \vec{k}}, \\ (E - \Omega_{\vec{k}}) \langle\langle \xi_{\vec{k}-\vec{p}} \xi_{\vec{q}-\vec{p}}^\dagger \xi_{\vec{q}}^\dagger; \xi_{\vec{q}}^\dagger \xi_{\vec{q}-\vec{p}} \xi_{\vec{k}-\vec{p}}^\dagger \rangle\rangle &= \delta_{\vec{q}, \vec{k}} (\delta_{\vec{p}, 0} + \delta_{\vec{p}, \vec{q} + \vec{k}}), \\ (E - \Omega_{\vec{k}}) \langle\langle \xi_{\vec{k}-\vec{p}} \xi_{\vec{p}-\vec{q}} \xi_{\vec{q}}^\dagger; \xi_{\vec{q}}^\dagger \xi_{\vec{q}-\vec{p}} \xi_{\vec{k}-\vec{p}}^\dagger \rangle\rangle &= \delta_{\vec{q}, \vec{k}} (\delta_{\vec{p}, 0} + \delta_{\vec{p}, \vec{q} + \vec{k}}), \\ (E - \Omega_{\vec{k}}) \langle\langle \xi_{\vec{k}-\vec{p}} \xi_{\vec{p}-\vec{q}} \xi_{\vec{q}}^\dagger; \xi_{\vec{q}}^\dagger \xi_{\vec{p}-\vec{q}} \xi_{\vec{k}-\vec{p}}^\dagger \rangle\rangle &= (\delta_{\vec{p}, 0} + \delta_{\vec{p}, \vec{q} + \vec{k}}) (\delta_{\vec{p}', 0} + \delta_{\vec{p}', \vec{q} + \vec{k}}). \end{aligned} \quad (6.17)$$

The static contributions to the polarization operator $\mathcal{P}_1(\vec{k}, E)$ are given by a sum over these three-particle Green's functions and their Hermitian conjugates. Because the denominator $(E - \Omega_{\vec{k}})$ is independent of the internal momenta \vec{q} and \vec{p} , the expression for these contributions simplifies to

$$\langle\langle f_{\vec{k}}; f_{\vec{k}}^\dagger \rangle\rangle_s = (-Q)^3 \left(\frac{\bar{B}^{u, v}(\vec{k})}{E - \Omega_{\vec{k}}} + \frac{\bar{B}^{v, u}(\vec{k})}{-E - \Omega_{\vec{k}}} \right), \quad (6.18)$$

$$\langle\langle f_{\vec{k}}; f_{-\vec{k}} \rangle\rangle_s = (-Q)^3 \hat{B}^{u, v}(\vec{k}) \left(\frac{1}{E - \Omega_{\vec{k}}} + \frac{1}{-E - \Omega_{\vec{k}}} \right). \quad (6.19)$$

The coefficients $\bar{B}(\vec{k})$ and $\hat{B}(\vec{k})$ are given in the Appendix.

Since the static contribution has poles identical to the RZOA poles, the only effect is to split the poles $\pm\Omega_{\vec{k}}$ of the single-particle Green's function into four levels $\pm\Omega_{\vec{k}}^\pm$. The relation between the splitting and the coefficients $\bar{B}(\vec{k})$ and $\hat{B}(\vec{k})$ is also given in the Appendix. The magnitude of the splitting will be second order in the interaction. We shall not consider these effects further here.

VII. LIFETIME OF THE EXCITATIONS

In this section, we consider the effect of the time-dependent contribution to the polarization operator on the excitation spectrum. From Eqs. (6.15) and (6.16), it is clear that the polarization operator has a branch cut along the real axis. As such, it contributes an imaginary as well as a real part to the single-particle Green's function. Thus the poles of the Green's function move off the real axis. The correlation spectrum turns

from a δ function to a Lorentzian behavior, the width corresponding to the finite lifetime of the excitations.

In terms of the polarization operator $\bar{P}_{\vec{k}}(E)$ and $\hat{P}_{\vec{k}}(E)$, the single-particle Green's function becomes

$$g(\vec{k}, E) = \frac{-Q}{D(E)} \begin{pmatrix} E + [\bar{\Omega}_{\vec{k}} + \bar{P}_{\vec{k}}(-E)] & -[\hat{\Omega}_{\vec{k}} - \hat{P}_{\vec{k}}(E)] \\ -[\hat{\Omega}_{\vec{k}} - \hat{P}_{\vec{k}}(E)] & -E + [\bar{\Omega}_{\vec{k}} + \bar{P}_{\vec{k}}(E)] \end{pmatrix}, \quad (7.1)$$

with the poles determined by

$$\begin{aligned} D(E) &= E^2 - [\bar{P}_{\vec{k}}(E) - \bar{P}_{\vec{k}}(-E)] E \\ &\quad - \{[\bar{\Omega}_{\vec{k}} + \bar{P}_{\vec{k}}(E)][\bar{\Omega}_{\vec{k}} + \bar{P}_{\vec{k}}(-E)] \\ &\quad - [\hat{\Omega}_{\vec{k}} - \hat{P}_{\vec{k}}(E)]^2\}. \end{aligned} \quad (7.2)$$

To obtain the spectral density $J_{\vec{k}}^{\alpha\beta}(\omega)$, the difference of the Green's functions across the real axis branch cut must be taken. Accordingly, we define

$$\bar{P}_{\vec{k}}(\omega \pm i\epsilon) = \bar{R}_{\vec{k}}(\omega) \mp i\bar{I}_{\vec{k}}(\omega), \quad (7.3)$$

$$\hat{P}_{\vec{k}}(\omega \pm i\epsilon) = \hat{R}_{\vec{k}}(\omega) \mp i\hat{I}_{\vec{k}}(\omega), \quad (7.4)$$

where the real part $R_{\vec{k}}(\omega)$ is given by the appropriate principal part integral and

$$\begin{aligned} \bar{I}_{\vec{k}}(\omega) &= \pi \left(\frac{Q}{N}\right)^2 \sum_{\vec{q}, \vec{p}} \{ \bar{A}_{\vec{q}, \vec{p}}^{u, v}(\vec{k}) \delta[\omega - \epsilon_{\vec{q}, \vec{p}}(\vec{k})] \\ &\quad - \bar{A}_{\vec{q}, \vec{p}}^{v, u}(\vec{k}) \delta[\omega + \epsilon_{\vec{q}, \vec{p}}(\vec{k})] \}, \end{aligned} \quad (7.5)$$

$$\begin{aligned} \hat{I}_{\vec{k}}(\omega) &= \pi \left(\frac{Q}{N}\right)^2 \sum_{\vec{q}, \vec{p}} \hat{A}_{\vec{q}, \vec{p}}^{u, v} \{ \delta[\omega - \epsilon_{\vec{q}, \vec{p}}(\vec{k})] \\ &\quad - \delta[\omega + \epsilon_{\vec{q}, \vec{p}}(\vec{k})] \}. \end{aligned} \quad (7.6)$$

The real and imaginary parts of $\bar{P}_{\vec{k}}(-E)$ are obtained by interchanging the parameters u and v ; hence the real part corresponding to $\bar{P}_{\vec{k}}(-E)$ is $\bar{R}_{\vec{k}}(-\omega)$ and the imaginary part is $-\bar{I}_{\vec{k}}(-\omega)$. The real and imaginary parts $\hat{R}_{\vec{k}}(\omega)$ and $\hat{I}_{\vec{k}}(\omega)$ are even and odd in frequency, respectively. The same is not true of $\bar{R}_{\vec{k}}(\omega)$ and $\bar{I}_{\vec{k}}(\omega)$ because of the difference between $\bar{A}_{\vec{q}, \vec{p}}^{u, v}(\vec{k})$ and $\bar{A}_{\vec{q}, \vec{p}}^{v, u}(\vec{k})$.

The real part of the polarization operator results in a frequency-dependent renormalization of the coupling parameters $\bar{\Omega}_{\vec{k}}$ and $\hat{\Omega}_{\vec{k}}$, which determine the position of the poles in the limit of small damping. Consistent with previous approximations, we shall neglect this contribution, which is second order in the interaction. The imaginary part of the polarization operator is also second order in the interaction. However it is the leading-order contribution to the damping and we consider now its effect on the excitation spectrum.

With this approximation the denominator (7.2), when evaluated on both sides of the branch cut along the real axis, simplifies to

$$D(\omega \pm i\epsilon) \simeq \omega^2 - \Omega_{\vec{k}}^2 \pm 2i\Omega_{\vec{k}}\Gamma_{\vec{k}}(\omega). \quad (7.7)$$

The poles are shifted off the real axis an amount determined by

$$\begin{aligned} \Gamma_{\vec{k}}(\omega) &= \{\omega^{\frac{1}{2}}[\bar{I}_{\vec{k}}(\omega) + \bar{I}_{\vec{k}}(-\omega)] + \bar{\Omega}_{\vec{k}}^{\frac{1}{2}}[\bar{I}_{\vec{k}}(\omega) - \bar{I}_{\vec{k}}(-\omega)] \\ &\quad + \hat{\Omega}_{\vec{k}} \hat{J}_{\vec{k}}(\omega)\} / \Omega_{\vec{k}} \\ &= \left(\frac{Q}{N}\right)^2 \sum_{\vec{q}, \vec{p}} \gamma_{\vec{q}, \vec{p}}(\vec{k}) \{\delta[\omega - \epsilon_{\vec{q}, \vec{p}}(\vec{k})] \\ &\quad - \delta[\omega + \epsilon_{\vec{q}, \vec{p}}(\vec{k})]\}, \end{aligned} \quad (7.8)$$

where

$$\begin{aligned} \gamma_{\vec{q}, \vec{p}}(\vec{k}) &= \pi \left[\epsilon_{\vec{q}, \vec{p}}(\vec{k})^{\frac{1}{2}} (\bar{A}_{\vec{q}, \vec{p}}^{\nu, \mu}(\vec{k}) - \bar{A}_{\vec{q}, \vec{p}}^{\mu, \nu}(\vec{k})) \right. \\ &\quad + \bar{\Omega}_{\vec{k}}^{\frac{1}{2}} (\bar{A}_{\vec{q}, \vec{p}}^{\mu, \nu}(\vec{k}) + \bar{A}_{\vec{q}, \vec{p}}^{\nu, \mu}(\vec{k})) \\ &\quad \left. + \hat{\Omega}_{\vec{k}} \hat{A}_{\vec{q}, \vec{p}}^{\mu, \nu}(\vec{k}) \right] / \Omega_{\vec{k}}. \end{aligned} \quad (7.9)$$

The spectral intensity of the two-time correlation function

$$J_{\vec{k}}^{\alpha\beta}(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \frac{\langle X_{\vec{k}}^{\beta\alpha}(0) X_{\vec{k}}^{\alpha\beta}(t) \rangle}{-Q} \quad (7.10)$$

is obtained, as usual, by taking the difference between the Green's function across the real axis branch cut. From Eq. (7.1), it follows that

$$J_{\vec{k}}^{11}(\omega) = e^{-\beta\omega} J_{\vec{k}}^{22}(-\omega) = \bar{J}_{\vec{k}}(\omega), \quad (7.11)$$

$$J_{\vec{k}}^{12}(\omega) = J_{\vec{k}}^{21}(\omega) = \hat{J}_{\vec{k}}(\omega), \quad (7.12)$$

where the spectral intensities show broadened

$$(e^{\beta\omega} - 1) \bar{J}_{\vec{k}}(\omega) \simeq \frac{(1 \pm \bar{\Omega}_{\vec{k}} / \Omega_{\vec{k}}) \Gamma_{\vec{k}}(\Omega_{\vec{k}}) + \epsilon [\Gamma_{\vec{k}}(\Omega_{\vec{k}}) - \bar{I}_{\vec{k}}(\mp \Omega_{\vec{k}})] / \Omega_{\vec{k}}}{\epsilon^2 + \Gamma_{\vec{k}}^2(\Omega_{\vec{k}})}, \quad (7.16)$$

and the off-diagonal intensity to

$$(e^{\beta\omega} - 1) \hat{J}_{\vec{k}}(\omega) \simeq (\mp) \frac{(\hat{\Omega}_{\vec{k}} / \Omega_{\vec{k}}) \Gamma_{\vec{k}}(\Omega_{\vec{k}}) - \epsilon \hat{I}_{\vec{k}}(\Omega_{\vec{k}}) / \Omega_{\vec{k}}}{\epsilon^2 + \Gamma_{\vec{k}}^2(\Omega_{\vec{k}})}, \quad (7.17)$$

at $\omega \simeq \pm \Omega_{\vec{k}}$, respectively. Since the damping is second order we expect $I_{\vec{k}} / \Omega_{\vec{k}}$ to be small so that the spectral intensity reduces to a Lorentzian with half-width $\Gamma_{\vec{k}}(\Omega_{\vec{k}})$. The height of the peak for $\bar{J}_{\vec{k}}(\omega)$ is $2(-Q)u_{\vec{k}}^2 / \Gamma_{\vec{k}}(\Omega_{\vec{k}})$ at $\omega = \Omega_{\vec{k}}$ and $-2(-Q)v_{\vec{k}}^2 / \Gamma_{\vec{k}}(\Omega_{\vec{k}})$ at $\omega = -\Omega_{\vec{k}}$. The value of $\hat{J}_{\vec{k}}(\omega)$ at $\omega = \pm \Omega_{\vec{k}}$ is $\pm 2(-Q)u_{\vec{k}}v_{\vec{k}} / \Gamma_{\vec{k}}(\Omega_{\vec{k}})$.

The integrand $\gamma_{\vec{q}, \vec{p}}(\vec{k})$ in the integral (7.8) over momentum \vec{q} and \vec{p} is singular along the lines $\vec{q} = 0$, $\vec{q} = \vec{p}$, and $\vec{p} = \vec{k}$. The singularity arises from the parameters $u_{\vec{q}}^2$ and $v_{\vec{q}}^2$ which diverge as $1/q$ for small values of the argument. For $\vec{k} \neq 0$, only two of the three parameters in $\gamma_{\vec{q}, \vec{p}}(\vec{k})$ will be singular at the same point so the integral is convergent in two and three dimensions. For $\vec{k} = 0$ all three parameters are singular at $\vec{q} = \vec{p} = 0$; however the coefficient $\gamma_{\vec{q}, \vec{p}}(\vec{k})$ approaches zero as k^2 for small \vec{k} . Thus both the damping $\Gamma_{\vec{k}}(\Omega_{\vec{k}})$ and the excitation en-

ergy $\Omega_{\vec{k}}$ approach zero as the first power of the momentum k for small momentum.

$$\begin{aligned} \bar{J}_{\vec{k}}(\omega) &= 2(e^{\beta\omega} - 1)^{-1} \\ &\quad \times \frac{2(\omega + \bar{\Omega}_{\vec{k}}) \Omega_{\vec{k}} \Gamma_{\vec{k}}(\omega) - (\omega^2 - \Omega_{\vec{k}}^2) \bar{I}_{\vec{k}}(-\omega)}{(\omega^2 - \Omega_{\vec{k}}^2)^2 + 4\Omega_{\vec{k}}^2 \Gamma_{\vec{k}}^2(\omega)}, \end{aligned} \quad (7.13)$$

$$\begin{aligned} \hat{J}_{\vec{k}}(\omega) &= 2(e^{\beta\omega} - 1)^{-1} \\ &\quad \times \frac{2\hat{\Omega}_{\vec{k}} \Omega_{\vec{k}} \Gamma_{\vec{k}}(\omega) - (\omega^2 - \Omega_{\vec{k}}^2) \hat{I}_{\vec{k}}(\omega)}{(\omega^2 - \Omega_{\vec{k}}^2)^2 + 4\Omega_{\vec{k}}^2 \Gamma_{\vec{k}}^2(\omega)}. \end{aligned} \quad (7.14)$$

The detailed balance relation (7.11) between the spectral intensities for $\langle b_{\vec{k}}^{\dagger}(0) b_{\vec{k}}(t) \rangle$ and $\langle a_{\vec{k}}(0) a_{\vec{k}}^{\dagger}(t) \rangle$ indicates there is no ambiguity whether the order parameter is determined using $J_{\vec{k}}^{11}(\omega)$ or $J_{\vec{k}}^{22}(\omega)$. The off-diagonal intensity also satisfies the detailed balance relation $\hat{J}_{\vec{k}}(-\omega) = e^{\beta\omega} \hat{J}_{\vec{k}}(\omega)$.

In the limit of small damping the spectral intensities approximate Lorentzians of width $\Gamma_{\vec{k}}(\Omega_{\vec{k}})$. We expand about the zero-damping poles $\pm \Omega_{\vec{k}}$, letting

$$\epsilon = \pm \omega - \Omega_{\vec{k}}, \quad (7.15)$$

for positive and negative frequencies, respectively. Assuming the damping $\Gamma_{\vec{k}}(\omega)$ to be a smooth function of frequency, it can be replaced by its value at $\omega = \pm \Omega_{\vec{k}}$. The diagonal spectral intensity reduces to

energy $\Omega_{\vec{k}}$ approach zero as the first power of the momentum k for small momentum.

VIII. EFFECT OF ANISOTROPIC TERM ON LIFETIME

The isotropic Hamiltonian (1.6) is not a realistic model for quadrupolar solids, such as hydrogen. It is, however, possible to include the effect of some anisotropic terms within our treatment. In particular, we consider a perturbation of the form

$$V = \sum_{i,j} \{(\alpha_{ij} + i\beta_{ij}) [L_2^{-1}(i) + L_2^1(i)] L_2^2(j) + \text{H.c.}\}, \quad (8.1)$$

which is of cubic order in the creation and annihilation operators. Other possible anisotropies, such as $L_2^0(i) L_2^2(j)$, have a quadratic term and will change the zero-order excitation spectrum.

Our discussion will be limited to the effect the

perturbation (8.1) has on the lifetime of the collective mode. Neglecting the cubic term $F_{\mathbf{k}}$, arising from the isotropic Hamiltonian, the equations of motion (3.7) and (3.8) become

$$[b_{\mathbf{k}}, \mathcal{H}] - \bar{\omega}_{\mathbf{k}} b_{\mathbf{k}} - \hat{\omega}_{\mathbf{k}} a_{-\mathbf{k}}^{\dagger} \approx E_{\mathbf{k}}^1, \quad (8.2)$$

$$-[a_{-\mathbf{k}}^{\dagger}, \mathcal{H}] - \hat{\omega}_{\mathbf{k}} b_{\mathbf{k}} - \bar{\omega}_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} \approx E_{\mathbf{k}}^2. \quad (8.3)$$

We shall not consider the renormalization of the excitation energies by the static term $\langle E_{\mathbf{k}}, X_{-\mathbf{k}}^{\dagger} \rangle$, but rather take the leading-order contribution to the damping from the higher-order Green's function $\langle\langle E_{\mathbf{k}}; E_{\mathbf{k}}^{\dagger} \rangle\rangle$. For this it is sufficient to use the effective Hamiltonian (6.7) with the parameters determined by the ZOA. With this approximation ($a_i = b_i$) the remainder term $E_{\mathbf{k}}^2$ in Eqs. (8.2) and (8.3) reduces to

$$E_{\mathbf{k}}^1 = [b_{\mathbf{k}}, V] = -\sqrt{2} \sum_j \alpha_{ij} (1 - 2n_i) n_j, \quad (8.4)$$

$$E_{\mathbf{k}}^2 = -[a_{\mathbf{k}}^{\dagger}, V] = -E_{\mathbf{k}}^1. \quad (8.5)$$

The diagonal and off-diagonal terms of the frequency-dependent polarization operator $\phi_1^{(V)}(\mathbf{k}, E)$ are identical

$$\begin{aligned} \phi_1^{(V)}(\mathbf{k}, E) &= \frac{1}{Q^2} \langle\langle E_{\mathbf{k}}; E_{\mathbf{k}}^{\dagger} \rangle\rangle \\ &= \frac{1}{-Q} \begin{pmatrix} P_{\mathbf{k}}(E) & -P_{\mathbf{k}}(E) \\ -P_{\mathbf{k}}(E) & P_{\mathbf{k}}(E) \end{pmatrix}. \end{aligned} \quad (8.6)$$

The leading contribution to the polarization operator is a two-particle Green's function

$$P_{\mathbf{k}}(E) \approx \frac{2\alpha_{\mathbf{k}}^2}{(-Q)N} \sum_{\mathbf{q}, \mathbf{q}'} \langle\langle a_{\mathbf{q}-\mathbf{k}}^{\dagger} a_{\mathbf{q}}; a_{\mathbf{q}}^{\dagger} a_{\mathbf{q}-\mathbf{k}} \rangle\rangle. \quad (8.7)$$

The two-particle Green's function is evaluated by transforming to the diagonal representation. At zero temperature the only nonzero two-particle Green's functions are

$$\begin{aligned} (E - \omega_{\mathbf{q}} - \omega_{\mathbf{k}-\mathbf{q}}) \langle\langle \xi_{\mathbf{k}-\mathbf{q}}^{\dagger} \xi_{\mathbf{q}}; \xi_{\mathbf{q}}^{\dagger} \xi_{\mathbf{k}-\mathbf{q}} \rangle\rangle \\ = (-E - \omega_{\mathbf{q}} - \omega_{\mathbf{k}-\mathbf{q}}) \langle\langle \xi_{\mathbf{q}-\mathbf{k}}^{\dagger} \xi_{-\mathbf{q}}^{\dagger}; \xi_{-\mathbf{q}} \xi_{\mathbf{q}-\mathbf{k}} \rangle\rangle \\ = \delta_{\mathbf{q}, \mathbf{q}} + \delta_{\mathbf{q}, \mathbf{k}-\mathbf{q}}. \end{aligned} \quad (8.8)$$

Using the transformation (5.13) and the equations of motion (8.8), the expression (8.7) for the polarization operator reduces to

$$P_{\mathbf{k}}(E) = \frac{-Q}{N} \sum_{\mathbf{q}} A_{\mathbf{q}}^{u,v}(\mathbf{k}) \left(\frac{1}{E - \omega_{\mathbf{q}} - \omega_{\mathbf{k}-\mathbf{q}}} + \frac{1}{-E - \omega_{\mathbf{q}} - \omega_{\mathbf{k}-\mathbf{q}}} \right), \quad (8.9)$$

where the coefficient $A_{\mathbf{q}}^{u,v}(\mathbf{k})$ is given by

$$A_{\mathbf{q}}^{u,v}(\mathbf{k}) = Q^2 \alpha_{\mathbf{k}}^2 u_{\mathbf{k}-\mathbf{q}} v_{\mathbf{k}-\mathbf{q}} (u_{\mathbf{q}} v_{\mathbf{k}-\mathbf{q}} + u_{\mathbf{k}-\mathbf{q}} v_{\mathbf{q}}). \quad (8.10)$$

From (8.10) it is easily seen that $A_{\mathbf{q}}^{u,v}(\mathbf{k})$ is symmetric under interchange of u and v when the sum over \mathbf{q} in Eq. (8.9) is performed. Thus $P_{\mathbf{k}}(E)$ is an even function of the complex energy variable E .

The analysis of the previous section to obtain the spectral intensity of the various correlation functions follows here with the simplification that both the diagonal $\bar{P}_{\mathbf{k}}(E)$ and off-diagonal terms $\hat{P}_{\mathbf{k}}(E)$ of the polarization operator are replaced by $P_{\mathbf{k}}(E)$. Also, the RZO parameters are replaced by their ZOA counterparts. The imaginary part $I_{\mathbf{k}}(\omega)$ of $P_{\mathbf{k}}(\omega - i\epsilon)$ is given by the sum over momentum

$$I_{\mathbf{k}}(\omega) = \frac{\pi(-Q)}{N} \sum_{\mathbf{q}} A_{\mathbf{q}}^{u,v}(\mathbf{k}) [\delta(\omega - \omega_{\mathbf{q}} - \omega_{\mathbf{k}-\mathbf{q}}) - \delta(\omega + \omega_{\mathbf{q}} + \omega_{\mathbf{k}-\mathbf{q}})]. \quad (8.11)$$

The width of the Lorentzian peak in the spectral intensity turns out to be simply

$$\Gamma_{\mathbf{k}}(\omega) = \mathcal{J}_0 I_{\mathbf{k}}(\omega) / \omega_{\mathbf{k}}. \quad (8.12)$$

Equations (7.13), (7.14), (7.16), and (7.17) for the spectral intensity remain valid, with the imaginary part $\bar{I}_{\mathbf{k}}(\omega)$ and $\hat{I}_{\mathbf{k}}(\omega)$ both replaced by $I_{\mathbf{k}}(\omega)$.

IX. CONCLUSION

We have systematically developed a Green's function theory of an isotropic quadrupolar model, through a Dyson's equation approach. Two terms contributing to the self-energy have been calculated: a static term which leads to a renormalization of the excitation spectrum and a dynamic term which results in a Lorentzian spectral intensity. Our approach has led to symmetrized equations so there is no ambiguity in applying the results either to calculate the long-range order or the spectral intensities.

The perturbation theory developed here has assumed the interaction coupling is small compared to the quadratic coupling term ($\hat{\omega}_{\mathbf{k}} \ll \bar{\omega}_{\mathbf{k}}$). The equation of motion formalism expresses a lower-order Green's function in terms of a higher one. At some stage the procedure must be truncated and further Green's functions neglected. We have included the three-particle Green's function in our treatment. As in many cases where the Green's function formalism is applied the interaction term here is not small. However, the coupling parameter does oscillate in sign, being

positive in one half of the Brillouin zone and negative in the other half.

A study of the results developed here requires detailed numerical analysis. The renormalization of the excitation spectrum involves a single sum over momentum and the damping involves a double sum; in both cases the integrand is singular but the integral is convergent in two and three dimensions. We plan to carry out the numerical investigation in a following study.

APPENDIX

The static contribution to the polarization operator $\mathcal{P}_1(\vec{k}, E)$ is determined by the coefficients $\bar{B}^{u,v}(\vec{k})$ and $\hat{B}^{u,v}(\vec{k})$ through equations (6.18) and (6.19). Inserting Eq. (6.2) into (6.1) and collecting the terms corresponding to the four pairs of non-zero three-particle Green's functions (6.17), we obtain the following expression for these coefficients:

$$\bar{B}^{u,v}(\vec{k}) = -Q\{\bar{n}^2[K_{\vec{k}}^u(\vec{k})]^2 + 2\bar{n}[\bar{T}_{\vec{k}}u_{\vec{k}}K_{\vec{k}}^u(\vec{k}) + \hat{T}_{\vec{k}}v_{\vec{k}}K_{\vec{k}}^u(\vec{k})] + [\bar{T}_{\vec{k}}u_{\vec{k}} + \hat{T}_{\vec{k}}v_{\vec{k}}]^2\}; \quad (A1)$$

$$\hat{B}^{u,v}(\vec{k}) = -Q(\bar{n}^2K_{\vec{k}}^u(\vec{k})K_{\vec{k}}^v(\vec{k}) + \bar{n}\{\bar{T}_{\vec{k}}[v_{\vec{k}}K_{\vec{k}}^u(\vec{k}) + u_{\vec{k}}K_{\vec{k}}^v(\vec{k})] + \hat{T}_{\vec{k}}[v_{\vec{k}}K_{\vec{k}}^v(\vec{k}) + u_{\vec{k}}K_{\vec{k}}^u(\vec{k})]\}) + [\bar{T}_{\vec{k}}u_{\vec{k}} + \hat{T}_{\vec{k}}v_{\vec{k}}][\bar{T}_{\vec{k}}v_{\vec{k}} + \hat{T}_{\vec{k}}u_{\vec{k}}]. \quad (A2)$$

In these equations, the parameter $T_{\vec{k}}$ is a sum over momentum. It can be expressed in terms of the sum $S_{\vec{k}}$, defined by equations (4.8) and (4.9)

$$\bar{T}_{\vec{k}} = \frac{-Q}{N} \sum_{\vec{q}} u_{\vec{q}}K_{\vec{q}}^u(\vec{k}) = 2\bar{S}_{\vec{k}} - (\bar{S}_0 + \hat{S}_0), \quad (A3)$$

$$\hat{T}_{\vec{k}} = \frac{-Q}{N} \sum_{\vec{q}} u_{\vec{q}}K_{\vec{q}}^v(\vec{k}) = 2\hat{S}_{\vec{k}} - (\bar{S}_0 + \hat{S}_0). \quad (A4)$$

If only this static contribution to the self-energy is considered, then the only change in the solution

(4.10) for the single-particle Green's functions is that the simple poles at $\pm\Omega_{\vec{k}}$ are each split into two levels $\pm\Omega_{\vec{k}}^{\pm}$,

$$(\Omega_{\vec{k}}^{\pm})^2 = (\Omega_{\vec{k}} \pm \theta_{\vec{k}})^2 - \phi_{\vec{k}}^2, \quad (A5)$$

where

$$\theta_{\vec{k}}^2 = -Q[u_{\vec{k}}^2\bar{B}^{uv}(\vec{k}) + v_{\vec{k}}^2\bar{B}^{vu}(\vec{k}) - 2u_{\vec{k}}v_{\vec{k}}\hat{B}^{uv}(\vec{k})], \quad (A6)$$

$$\phi_{\vec{k}}^2 = -Q[u_{\vec{k}}^2\bar{B}^{vu}(\vec{k}) + v_{\vec{k}}^2\bar{B}^{uv}(\vec{k}) - 2u_{\vec{k}}v_{\vec{k}}\hat{B}^{vu}(\vec{k})]. \quad (A7)$$

The poles remain on the real axis, so these terms do not contribute to the lifetime of the excitations.

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