Green's-function theory of quadrupolar coupled systems

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The isotropic quadrupolar coupling Hamiltonian is studied by means of double-time Green's functions. The equations-of-motion hierarchy is decoupled by using the concepts of cumulant averages and self-consistently identifying the statistically independent operators of the system. Our results satisfy all relevant spin-1 identities. We obtain the transition temperature and the ground-state order parameter for the sc, fcc, and bcc lattices. Our result for the ground-state order parameter is larger than that obtained by current decoupling schemes.

I. INTRODUCTION

Systems in which quadrupolar interactions dominate include molecular solids,[1] liquid crystals,[2] and the Jahn-Teller ferroelectric system.[3] Attempts to develop double-time Green's-function (DTGF) theories of even the simplest (i.e., isotropic) quadrupolar coupled system have met with many difficulties. Barma[4] pointed out an ambiguity in the random-phase-approximation (RPA) treatment of the system and proposed a "trace-invariance criterion" to resolve the ambiguity and obtain a single value of the order parameter. In addition, Barma proposed a new (DTGF) treatment which provided a larger value of the $T=0$ order parameter than RPA. Fittipaldi and Tahir-Kheli[5] pointed out that Barma's new treatment did not preserve certain spin-1 identities. Using a Barma-type DTGF treatment which preserved some (but not all) relevant spin-1 identities, they obtained an order-parameter value between those of RPA and Barma, ascribing this result to "oscillatory, though convergent, successive approximation." Ritchie and Mavroyannis[6] proposed another, more complex, DTGF scheme which did not preserve the spin-1 identities, dismissing this shortcoming as being equivalent to the expected failure of an approximate scheme to provide exact results for the correlations of the system. However, the identities in question were assumed valid throughout the entire treatment and were found not to be satisfied at the end, thus violating self-consistency. Ritchie and Mavroyannis did not obtain numerical results with their scheme.

In this paper we consider the isotropic quadrupolar system for all T. Our treatment is based upon the concepts of cumulant averages and statistical independence, is unambiguous, and satisfies all spin-1 identities. We obtain the susceptibility for $T>T_c$, the critical temperature, and the ground-state value of the order parameter for the simple-cubic (sc), face-centered-cubic, and body-centered-cubic lattices. Our value for the sc order parameter in the ground state is *larger* than that obtained by Barma.

II. DOUBLE-TIME GREEN'S FUNCTIONS

The retarded ($\rho=+1$) or advanced ($\rho=-1$) commutator ($\eta=-1$) or anticommutator ($\eta=+1$) double-time Green's function (DTGF) is defined by[7]

$$\langle\langle A(t);B(t')\rangle\rangle^{(\eta)}_{(\rho)}=-\frac{i}{2}[(\rho+1)\Theta(t-t')+(\rho-1)\Theta(t'-t)]\times\langle[A(t),B(t')]_\eta\rangle\ , \quad (2.1)$$

where

$$A(t)=e^{iHt}Ae^{-iHt},\quad [A,B]_\eta=AB+\eta BA\ , \quad (2.2)$$

and $\Theta(t)$ is unity for $t>0$ and zero for $t<0$. The single angular brackets in Eq. (2.1) denote thermal average. It follows from Eq. (2.1) that $\langle\langle A(t);B(t')\rangle\rangle^{(\eta)}_{(\rho)}$ is a function of $t-t'$ only.

The Fourier transform of $\langle\langle A(t);B\rangle\rangle^{(\eta)}_{(\rho)}$ is defined by

$$\langle\langle A;B\rangle\rangle^{(\eta)}_{E+i\rho\epsilon}=\int_{-\infty}^{\infty}dt\, e^{i(E+i\rho\epsilon)t}\langle\langle A(t);B\rangle\rangle^{(\eta)}_{(\rho)}\ ,\quad \epsilon\to 0^+ \quad (2.3)$$

and satisfies the equation of motion

$$E\langle\langle A;B\rangle\rangle^{(\eta)}_E=\langle[A,B]_\eta\rangle+\langle\langle[A,H]_-;B\rangle\rangle^{(\eta)}_E\ . \quad (2.4)$$

The GF on the right-hand side of Eq. (2.4) is generally of "higher order" and must be decoupled so that a closed system of equations is obtained. Note that the Fourier-transformed GF as defined in Eq. (2.3) is sectionally holomorphic; the retarded (or advanced) GF is analytic in the upper (respectively, lower) half of the complex E plane.[8,9]

It has been shown[8,10] that the commutator GF cannot have a pole at $E=0$, i.e.,

$$C^{(-)}=0\ , \quad (2.5)$$

where

$$C^{(\eta)}=\lim_{E\to 0}E\langle\langle A;B\rangle\rangle^{(\eta)}_E \quad (2.6)$$

and that the correlation $\langle BA(t)\rangle$ may be calculated from

$$\langle BA(t)\rangle = \tfrac{1}{4}(1-\eta)C^{(-\eta)} + \frac{i}{2\pi}\int_{-\infty}^{\infty} dE(e^{\beta E}+\eta)^{-1}e^{-iEt} \times \lim_{\epsilon\to 0^+}(\langle\langle A;B\rangle\rangle_{E+i\epsilon}^{(\eta)} - \langle\langle A;B\rangle\rangle_{E-i\epsilon}^{(\eta)}) . \quad (2.7)$$

Also, the response of the system to an external field is described by the generalized susceptibility,[8,11]

$$\chi_{AB}(E) = -\lim_{\epsilon\to 0^+}\langle\langle A;B\rangle\rangle_{E+i\epsilon}^{(-)} . \quad (2.8)$$

III. QUADRUPOLAR COUPLED SYSTEMS

We consider the spin-1 operator basis consisting of the dipolar operators S_i^x, S_i^y, S_i^z, and the quadrupolar operators

$$\begin{aligned} Q_i^0 &= \sqrt{3}[(S_i^z)^2 - \tfrac{2}{3}] ,\\ Q_i^1 &= (S_i^x)^2 - (S_i^y)^2 ,\\ Q_i^2 &= S_i^x S_i^y + S_i^y S_i^x ,\\ Q_i^3 &= S_i^x S_i^z + S_i^z S_i^x ,\\ Q_i^4 &= S_i^y S_i^z + S_i^z S_i^y . \end{aligned} \quad (3.1)$$

In this basis, the isotropic nearest-neighbor coupling of the quadrupolar operators is described by

$$H_0 = -\tfrac{1}{2}\sum_{\nu=0}^{4}\sum_{ij} J_{ij} Q_i^\nu Q_j^\nu \quad (3.2)$$

and a uniform field coupling to Q_i^0 is described by

$$H_i = -\Omega \sum_i Q_i^0 . \quad (3.3)$$

We consider the full Hamiltonian

$$H = H_0 + H_1 \quad (3.4)$$

and consider the possibility of an ordered phase in which $\langle Q_i^0\rangle \neq 0$ by allowing Ω to approach zero.

Defining and invoking translational symmetry,

$$\mu \equiv \langle S_i^\mu\rangle, \quad q_\nu \equiv \langle Q_i^\nu\rangle , \quad (3.5)$$

we note that due to simple rotational symmetries of H all such single-site correlations vanish except q_0.

The equations of motion of our basis operators are given by

$$[S_i^x,H] = i\sqrt{3}\Omega Q_i^4 - i\sum_l J_{il}[(\sqrt{3}Q_i^0+Q_i^1) \times Q_l^4 - Q_i^4(\sqrt{3}Q_l^0+Q_l^1) + Q_i^3 Q_l^2 - Q_i^2 Q_l^3] , \quad (3.6)$$

$$[S_i^y,H] = -i\sqrt{3}\Omega Q_i^3 + i\sum_l J_{il}[(\sqrt{3}Q_i^0 - Q_i^1)Q_l^3 - Q_i^3(\sqrt{3}Q_l^0 - Q_l^1) + Q_i^4 Q_l^2 - Q_i^2 Q_l^4] , \quad (3.7)$$

$$[S_i^z,H] = -i\sum_l J_{il}[2(Q_i^2 Q_l^1 - Q_i^1 Q_l^2) - Q_i^3 Q_l^4 + Q_i^4 Q_l^3] , \quad (3.8)$$

$$[Q_i^0,H] = -i\sqrt{3}\sum_l J_{il}(S_i^y Q_l^3 - S_i^x Q_l^4) , \quad (3.9)$$

$$[Q_i^1,H] = -i\sum_l J_{il}(2S_i^z Q_l^2 - S_i^y Q_l^3 - S_i^x Q_l^4) , \quad (3.10)$$

$$[Q_i^2,H] = i\sum_l J_{il}(2S_i^z Q_l^1 - S_i^x Q_l^3 + S_i^y Q_l^4) , \quad (3.11)$$

$$[Q_i^3,H] = i\sqrt{3}\Omega S_i^y + i\sum_l J_{il}[S_i^y(\sqrt{3}Q_l^0 - Q_l^1) - S_i^x Q_l^2 + S_i^z Q_l^4] , \quad (3.12)$$

$$[Q_i^4,H] = -i\sqrt{3}\Omega S_i^x - i\sum_l J_{il}[S_i^x(\sqrt{3}Q_l^0 + Q_l^1) + S_i^y Q_l^2 - S_i^z Q_l^3] . \quad (3.13)$$

Taking the thermal average of both sides of each member of Eqs. (3.6)—(3.13) yields the correlation identities

$$\Omega q_3 = \Omega q_4 = 0 , \quad (3.14)$$

$$\sum_l J_{il}(\langle S_i^y Q_l^3\rangle - \langle S_i^x Q_l^4\rangle) = 0 , \quad (3.15)$$

$$\sum_l J_{il}(2\langle S_i^z Q_l^2\rangle - \langle S_i^y Q_l^3\rangle - \langle S_i^x Q_l^4\rangle) = 0 , \quad (3.16)$$

$$\sum_l J_{il}(2\langle S_i^z Q_l^1\rangle - \langle S_i^x Q_l^3\rangle + \langle S_i^y Q_l^4\rangle) = 0 , \quad (3.17)$$

$$\sqrt{3}\Omega y + \sum_l J_{il}[\langle S_i^y(\sqrt{3}Q_l^0 - Q_l^1)\rangle - \langle S_i^x Q_l^2\rangle + \langle S_i^z Q_l^4\rangle] = 0 , \quad (3.18)$$

$$\sqrt{3}\Omega x + \sum_l J_{il}[\langle S_i^x(\sqrt{3}Q_l^0 + Q_l^1)\rangle + \langle S_i^y Q_l^2\rangle - \langle S_i^z Q_l^3\rangle] = 0 . \quad (3.19)$$

Defining

$$\begin{aligned} G_{ij}^{\mu,R(\eta)} &\equiv \langle\langle S_i^\mu;R_j\rangle\rangle_E^{(\eta)}, \quad L_{ij}^{\mu,R(\eta)} \equiv \langle[S_i^\mu,R_j]_\eta\rangle ,\\ G_{ij}^{\nu,R(\eta)} &\equiv \langle\langle Q_i^\nu;R_j\rangle\rangle_E^{(\eta)}, \quad L_{ij}^{\nu,R(\eta)} \equiv \langle[Q_i^\nu,R_j]_\eta\rangle , \end{aligned} \quad (3.20)$$

and using Eqs. (3.6)—(3.13), we obtain the Green's-function equations of motion

$$EG_{ij}^{x,R(\eta)} = L_{ij}^{x,R(\eta)} + i\sqrt{3}\Omega G_{ij}^{4,R(\eta)} - i\sum_l J_{il}[\langle\langle(\sqrt{3}Q_i^0+Q_i^1)Q_l^4;R_j\rangle\rangle_E^{(\eta)} - \langle\langle Q_i^4(\sqrt{3}Q_l^0+Q_l^1);R_j\rangle\rangle_E^{(\eta)} + \langle\langle Q_i^3 Q_l^2;R_j\rangle\rangle_E^{(\eta)} - \langle\langle Q_i^2 Q_l^3;R_j\rangle\rangle_E^{(\eta)}] , \quad (3.21)$$

$$EG_{ij}^{y,R(\eta)}=L_{ij}^{y,R(\eta)}-i\sqrt{3}\Omega G_{ij}^{3,R(\eta)}+i\sum_l J_{il}[\langle\langle(\sqrt{3}Q_i^0-Q_i^1)Q_l^3;R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^3(\sqrt{3}Q_l^0-Q_l^1);R_j\rangle\rangle_E^{(\eta)}$$
$$+\langle\langle Q_i^4Q_l^2;R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^2Q_l^4;R_j\rangle\rangle_E^{(\eta)}]\ , \quad (3.22)$$

$$EG_{ij}^{z,R(\eta)}=L_{ij}^{z,R(\eta)}-i\sum_l J_{il}(2\langle\langle Q_i^2Q_l^1;R_j\rangle\rangle_E^{(\eta)}-2\langle\langle Q_i^1Q_l^2;R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^3Q_l^4;R_j\rangle\rangle_E^{(\eta)}+\langle\langle Q_i^4Q_l^3;R_j\rangle\rangle_E^{(\eta)})\ , \quad (3.23)$$

$$EG_{ij}^{0,R(\eta)}=L_{ij}^{0,R(\eta)}-i\sqrt{3}\sum_l J_{il}(\langle\langle S_i^yQ_l^3;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xQ_l^4;R_j\rangle\rangle_E^{(\eta)})\ , \quad (3.24)$$

$$EG_{ij}^{1,R(\eta)}=L_{ij}^{1,R(\eta)}-i\sum_l J_{il}(2\langle\langle S_i^zQ_l^2;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^yQ_l^3;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xQ_l^4;R_j\rangle\rangle_E^{(\eta)})\ , \quad (3.25)$$

$$EG_{ij}^{2,R(\eta)}=L_{ij}^{2,R(\eta)}+i\sum_l J_{il}(2\langle\langle S_i^zQ_l^1;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xQ_l^3;R_j\rangle\rangle_E^{(\eta)}+\langle\langle S_i^yQ_l^4;R_j\rangle\rangle_E^{(\eta)})\ , \quad (3.26)$$

$$EG_{ij}^{3,R(\eta)}=L_{ij}^{3,R(\eta)}+i\sqrt{3}\Omega G_{ij}^{y,R(\eta)}+i\sum_l J_{il}[\langle\langle S_i^y(\sqrt{3}Q_l^0-Q_l^1);R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xQ_l^2;R_j\rangle\rangle_E^{(\eta)}+\langle\langle S_i^zQ_l^4;R_j\rangle\rangle_E^{(\eta)}]\ , \quad (3.27)$$

$$EG_{ij}^{4,R(\eta)}=L_{ij}^{4,R(\eta)}-i\sqrt{3}\Omega G_{ij}^{x,R(\eta)}-i\sum_l J_{il}[\langle\langle S_i^x(\sqrt{3}Q_l^0+Q_l^1);R_j\rangle\rangle_E^{(\eta)}+\langle\langle S_i^yQ_l^2;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^zQ_l^3;R_j\rangle\rangle_E^{(\eta)}]\ , \quad (3.28)$$

IV. DECOUPLING SCHEME

As expected, the Green's functions on the right-hand sides of Eqs. (3.21)—(3.28) are of higher order than our basic Green's functions in Eq. (3.20) and must be approximately decoupled to provide a closed, soluble set of equations to replace Eqs. (3.21)—(3.28). We propose a decoupling scheme based upon the concepts of cumulants and statistical independence. The cumulant averages of $\langle Q_i^\alpha(t)Q_l^\gamma(t)R_j\rangle$ and $\langle S_i^\alpha(t)Q_l^\gamma(t)R_j\rangle$, the correlations appearing in $\langle\langle Q_i^\alpha Q_l^\gamma;R_j\rangle\rangle_E^{(\eta)}$ and $\langle\langle S_i^\alpha Q_l^\gamma;R_j\rangle\rangle_E^{(\eta)}$, are defined by[12]

$$\langle Q_i^\alpha(t)Q_l^\gamma(t)R_j\rangle=q_\alpha\langle Q_l^\gamma(t)R_j\rangle+q_\nu\langle Q_i^\alpha(t)R_j\rangle+R(\langle Q_i^\alpha Q_l^\gamma\rangle-2q_\alpha q_\nu)+\langle Q_i^\alpha(t)Q_l^\gamma(t)R_j\rangle_c\ , \quad (4.1)$$

$$\langle S_i^\alpha(t)Q_l^\gamma(t)R_j\rangle=\alpha\langle Q_l^\gamma(t)R_j\rangle+q_\nu\langle S_i^\alpha(t)R_j\rangle+R(\langle S_i^\alpha Q_l^\gamma\rangle-2\alpha q_\nu)+\langle S_i^\alpha(t)Q_l^\gamma(t)R_j\rangle_c\ , \quad (4.2)$$

where the subscript c denotes cumulant average and $R=\langle R_j\rangle$.

Our decoupling is based upon the assumption that at least one of the operators in $\langle Q_i^\alpha(t)Q_l^\gamma(t)R_j\rangle$ and $\langle S_i^\alpha(t)Q_l^\gamma(t)R_j\rangle$ is *statistically independent* of the others. This allows us to set[12]

$$\langle Q_i^\alpha(t)Q_l^\gamma(t)R_j\rangle_c=\langle S_i^\alpha(t)Q_l^\gamma(t)R_j\rangle_c=0 \quad (4.3)$$

and obtain the approximations

$$\langle Q_i^\alpha(t)Q_l^\gamma(t)R_j\rangle=q_\alpha\langle Q_l^\gamma(t)R_j\rangle+q_\nu\langle Q_i^\alpha(t)R_j\rangle+R(\langle Q_i^\alpha Q_l^\gamma\rangle-2q_\alpha q_\nu)\ , \quad (4.4)$$

$$\langle S_i^\alpha(t)Q_l^\gamma(t)R_j\rangle=\alpha\langle Q_l^\gamma(t)R_j\rangle+q_\nu\langle S_i^\alpha(t)R_j\rangle+R(\langle S_i^\alpha Q_l^\gamma\rangle-2\alpha q_\nu)\ . \quad (4.5)$$

Proceeding in identical fashion for $\langle R_jQ_i^\alpha(t)Q_l^\gamma(t)\rangle$ and $\langle R_jS_i^\alpha(t)Q_l^\gamma(t)\rangle$ we obtain the *decoupling approximations:*

$$\langle\langle Q_i^\alpha Q_l^\gamma;R_j\rangle\rangle_E^{(\eta)}=q_\alpha G_{lj}^{\gamma R(\eta)}+q_\nu G_{ij}^{\alpha R(\eta)}+\frac{(1+\eta)}{E}R(\langle Q_i^\alpha Q_l^\gamma\rangle-2q_\alpha q_\nu)\ , \quad (4.6)$$

$$\langle\langle S_i^\alpha Q_l^\gamma;R_j\rangle\rangle_E^{(\eta)}=\alpha G_{lj}^{\gamma R(\eta)}+q_\nu G_{ij}^{\alpha R(\eta)}+\frac{(1+\eta)}{E}R(\langle S_i^\alpha Q_l^\gamma\rangle-2\alpha q_\nu)\ . \quad (4.7)$$

We will self-consistently identify the statistically independent operators as those whose diagonal susceptibilities in the approximation, Eqs. (4.6) and (4.7), diverge in the ordered phase.

Using Eqs. (4.6) and Eq. (4.7) in the hierarchy, Eqs. (3.21)—(3.28), and using the correlation identities, Eqs. (3.14)—(3.19), and the fact that the only nonvanishing single-site correlation is q_0 yields the decoupled hierarchy (after performing a spatial Fourier transform)

$$EG_{\vec{k}}^{x;R(\eta)}=L_{\vec{k}}^{x,R(\eta)}+i\sqrt{3}(\Omega+q_0J_{0\vec{k}})G_{\vec{k}}^{4,R(\eta)}\ , \quad (4.8)$$

$$EG_{\vec{k}}^{4,R(\eta)}=L_{\vec{k}}^{4,R(\eta)}-i\sqrt{3}(\Omega+q_0J_0)G_{\vec{k}}^{x,R(\eta)}\ , \quad (4.9)$$

$$EG_{\vec{k}}^{y,R(\eta)}=L_{\vec{k}}^{y,R(\eta)}-i\sqrt{3}(\Omega+q_0J_{0\vec{k}})G_{\vec{k}}^{3,R(\eta)}\ , \quad (4.10)$$

$$EG_{\vec{k}}^{3,R(\eta)}=L_{\vec{k}}^{3,R(\eta)}+i\sqrt{3}(\Omega+q_0J_0)G_{\vec{k}}^{y,R(\eta)}\ , \quad (4.11)$$

$$EG_{\vec{k}}^{z,R(\eta)}=L_{\vec{k}}^{z,R(\eta)},\quad EG_{\vec{k}}^{1,R(\eta)}=L_{\vec{k}}^{1,R(\eta)}\ , \quad (4.12)$$

$$EG_{\vec{k}}^{0,R(\eta)}=L_{\vec{k}}^{0,R(\eta)},\quad EG_{\vec{k}}^{2,R(\eta)}=L_{\vec{k}}^{2,R(\eta)}\ , \quad (4.13)$$

where

$$G_{\vec{k}}^{\mu,R(\eta)}\equiv\frac{1}{N}\sum_{ij}e^{i\vec{k}\cdot\vec{r}_{ij}}G_{ij}^{\mu,R(\eta)}\ ,$$
$$L_{\vec{k}}^{\mu,R(\eta)}\equiv\frac{1}{N}\sum_{ij}e^{i\vec{k}\cdot\vec{r}_{ij}}L_{ij}^{\mu,R(\eta)},\quad J_{\vec{k}}\equiv\frac{1}{N}\sum_{ij}e^{i\vec{k}\cdot\vec{r}_{ij}}J_{ij} \quad (4.14)$$

and

$$J_{0\vec{k}}\equiv J_0-J_{\vec{k}}\ . \quad (4.15)$$

Solving Eqs. (4.8)–(4.13) we obtain

$$G_{\vec{k}}^{x,R(\eta)}=\frac{EL_{\vec{k}}^{x,R(\eta)}+i\sqrt{3}(\Omega+q_0J_{0\vec{k}})L_{\vec{k}}^{4,R(\eta)}}{E^2-\omega_{\vec{k}}^2}, \tag{4.16}$$

$$G_{\vec{k}}^{y,R(\eta)}=\frac{EL_{\vec{k}}^{y,R(\eta)}-i\sqrt{3}(\Omega+q_0J_{0\vec{k}})L_{\vec{k}}^{3,R(\eta)}}{E^2-\omega_{\vec{k}}^2}, \tag{4.17}$$

$$G_{\vec{k}}^{3,R(\eta)}=\frac{EL_{\vec{k}}^{3,R(\eta)}+i\sqrt{3}(\Omega+q_0J_0)L_{\vec{k}}^{y,R(\eta)}}{E^2-\omega_{\vec{k}}^2}, \tag{4.18}$$

$$G_{\vec{k}}^{4,R(\eta)}=\frac{EL_{\vec{k}}^{4,R(\eta)}-i\sqrt{3}(\Omega+q_0J_0)L_{\vec{k}}^{x,R(\eta)}}{E^2-\omega_{\vec{k}}^2}, \tag{4.19}$$

$$\begin{aligned}G_{\vec{k}}^{z,R(\eta)}&=\frac{L_{\vec{k}}^{z,R(\eta)}}{E}, \quad G_{\vec{k}}^{0,R(\eta)}=\frac{L_{\vec{k}}^{0,R(\eta)}}{E},\\ G_{\vec{k}}^{1,R(\eta)}&=\frac{L_{\vec{k}}^{1,R(\eta)}}{E}, \quad G_{\vec{k}}^{2,R(\eta)}=\frac{L_{\vec{k}}^{2,R(\eta)}}{E},\end{aligned} \tag{4.20}$$

where

$$\omega_{\vec{k}}^2=3(\Omega+q_0J_0)(\Omega+q_0J_{0\vec{k}}) .$$

Using Eq. (2.8), the diagonal susceptibilities are given by

$$\chi_\mu=-G_0^{\mu,\mu(-)}(E=0) . \tag{4.21}$$

Defining

$$\chi_0\equiv q_0/\Omega \tag{4.22}$$

and using Eqs. (4.16)–(4.20) in Eq. (4.21), we obtain

$$\chi_0=\chi_3=\chi_4 , \tag{4.23}$$

$$\chi_x=\chi_y=\frac{\chi_0}{1+\chi_0J_0} , \tag{4.24}$$

$$\chi_z=\chi_1=\chi_2=0 . \tag{4.25}$$

For q_0 ordering,

$$\lim_{\Omega\to 0} q_0\neq 0 \tag{4.26}$$

and, from Eqs. (4.22)–(4.25), χ_0, χ_3, and χ_4 are the only divergent diagonal susceptibilities. We identify Q_i^0, Q_i^3, and Q_i^4 as the members of the set of Eqs. (3.1), which are statistically independent of every other member of the set.

Having determined the statistically independent operators under the approximations, Eq. (4.6) and Eq. (4.7), we must, for self-consistency, require that at least one of these statistically independent operators appears in every Green's function that we have approximated as in Eq. (4.6) and Eq. (4.7). This requirement is clearly satisfied for Green's functions of the form $\langle\langle Q_i^\alpha Q_l^\gamma;R_j\rangle\rangle_E^{(\eta)}$ if Q_i^α and/or Q_l^γ is one of the statistically independent operators and for Green's functions of the form $\langle\langle S_i^\alpha Q_l^\gamma;R_j\rangle\rangle_E^{(\eta)}$ if Q_l^γ is one of the statistically independent operators. All of the Green's functions we have decoupled fall into one of these categories, except

$$\begin{aligned}&\langle\langle Q_i^2Q_l^1;R_j\rangle\rangle_E^{(\eta)},\ \langle\langle S_i^zQ_l^2;R_j\rangle\rangle_E^{(\eta)},\\ &\langle\langle S_i^zQ_l^1;R_j\rangle\rangle_E^{(\eta)},\ \langle\langle S_i^yQ_l^1;R_j\rangle\rangle_E^{(\eta)},\\ &\langle\langle S_i^xQ_l^2;R_j\rangle\rangle_E^{(\eta)},\ \langle\langle S_i^xQ_l^1;R_j\rangle\rangle_E^{(\eta)},\\ &\langle\langle S_i^yQ_l^1;R_j\rangle\rangle_E^{(\eta)} .\end{aligned} \tag{4.27}$$

In order to assure that at least one statistically independent operator appears in every Green's function in the set of Eqs. (4.27), we must *require that R_j be one of the statistically independent operators.*

Defining

$$a_{ij}^{R,\nu}\equiv\begin{cases}\langle R_iS_j^\nu\rangle, & \nu=x,y,z\\ \langle R_iQ_j^\nu\rangle, & \nu=0,1,2,3,4\end{cases} \tag{4.28}$$

and

$$a_{\vec{k}}^{R,\nu}\equiv\frac{1}{N}\sum_{ij}e^{i\vec{k}\cdot\vec{r}_{ij}}a_{ij}^{R,\nu} , \tag{4.29}$$

we obtain by using Eqs. (4.16)–(4.19) in Eq. (2.7)

$$a_{\vec{k}}^{R,x}=-\frac{L_{\vec{k}}^{x,R(-)}}{2}+\frac{i\sqrt{3}}{2}\frac{\Omega+q_0J_{0\vec{k}}}{\omega_{\vec{k}}}\times L_{\vec{k}}^{4,R(-)}\coth\left[\frac{\beta\omega_{\vec{k}}}{2}\right], \tag{4.30}$$

$$a_{\vec{k}}^{R,y}=-\frac{L_{\vec{k}}^{y,R(-)}}{2}-\frac{i\sqrt{3}}{2}\frac{\Omega+q_0J_{0\vec{k}}}{\omega_{\vec{k}}}\times L_{\vec{k}}^{3,R(-)}\coth\left[\frac{\beta\omega_{\vec{k}}}{2}\right], \tag{4.31}$$

$$a_{\vec{k}}^{R,3}=-\frac{L_{\vec{k}}^{3,R(-)}}{2}+\frac{i\sqrt{3}}{2}\frac{\Omega+q_0J_0}{\omega_{\vec{k}}}\times L_{\vec{k}}^{y,R(-)}\coth\left[\frac{\beta\omega_{\vec{k}}}{2}\right], \tag{4.32}$$

$$a_{\vec{k}}^{R,4}=-\frac{L_{\vec{k}}^{4,R(-)}}{2}-\frac{i\sqrt{3}}{2}\frac{\Omega+q_0J_0}{\omega_{\vec{k}}}\times L_{\vec{k}}^{x,R(-)}\coth\left[\frac{\beta\omega_{\vec{k}}}{2}\right]. \tag{4.33}$$

Using Eq. (4.20) in Eq. (2.7) yields a series of identities. It is important to note that Eqs. (4.30)–(4.33) are obtained from both $\eta=+1$ and $\eta=-1$ versions of Eq. (2.7).

Using $R_j=Q_j^0,Q_j^3,Q_j^4$ in (4.30)–(4.33) gives

$$a_{\vec{k}}^{0,x}=a_{\vec{k}}^{0,4}=a_{\vec{k}}^{0,y}=a_{\vec{k}}^{0,3}=a_{\vec{k}}^{3,x}=a_{\vec{k}}^{3,4}=a_{\vec{k}}^{4,y}=a_{\vec{k}}^{4,3}=0 , \tag{4.34}$$

which are exactly true due to the symmetry of H, and

$$a_{\vec{k}}^{3,y}=i\sqrt{3}q_0/2 , \tag{4.35}$$

$$a_{\vec{k}}^{4,x}=-i\sqrt{3}q_0/2 , \tag{4.36}$$

$$a_{\vec{k}}^{3,3}=\tfrac{3}{2}q_0\frac{\Omega+q_0J_0}{\omega_{\vec{k}}}\coth\left[\frac{\beta\omega_{\vec{k}}}{2}\right], \tag{4.37}$$

$$a_{\vec{k}}^{4,4}=\tfrac{3}{2}q_0\frac{\Omega+q_0J_0}{\omega_{\vec{k}}}\coth\left[\frac{\beta\omega_{\vec{k}}}{2}\right]. \tag{4.38}$$

Summing Eq. (4.35) and Eq. (4.36) over $\vec{k}$ and using the spin-1 identities

$$Q_l^3S_l^y=\frac{i}{2}(\sqrt{3}Q_l^0-Q_l^1), \tag{4.39}$$

$$Q_l^4S_l^x=-\frac{i}{2}(\sqrt{3}Q_l^0+Q_l^1), \tag{4.40}$$

gives

$$q_1=0, \tag{4.41}$$

which is true by the symmetry of H. Summing Eq. (4.37) and Eq. (4.38) over $\vec{k}$ and using the spin-1 identities

$$(Q_l^3)^2=\frac{1}{2}\left[\frac{4}{3}-\frac{Q_l^0}{\sqrt{3}}-Q_l^1\right], \tag{4.42}$$

$$(Q_l^4)^2=\frac{1}{2}\left[\frac{4}{3}-\frac{Q_l^0}{\sqrt{3}}+Q_l^1\right], \tag{4.43}$$

yields, using Eq. (4.41)

$$\frac{4}{3}-\frac{q_0}{\sqrt{3}}=3q_0(\Omega+q_0J_0)\frac{1}{N}\sum_{\vec{k}}\frac{1}{\omega_{\vec{k}}}\coth\left[\frac{\beta\omega_{\vec{k}}}{2}\right]. \tag{4.44}$$

For the nonordering region, we write Eq. (4.44) in the form (using $q_0=\chi_0\Omega$)

$$\frac{4}{3}-\frac{q_0}{\sqrt{3}}=\sqrt{3}\Omega\chi_0\frac{1}{N}\sum_{\vec{k}}\left[\frac{1+\chi_0J_0}{1+\chi_0J_{0\vec{k}}}\right]^{1/2}\times\coth\left[\frac{\beta\sqrt{3}\Omega}{2}(1+\chi_0J_0)^{1/2}\times(1+\chi_0J_{0\vec{k}})^{1/2}\right] \tag{4.45}$$

and take Ω and $q_0\to 0$ to obtain

$$\frac{2}{3}=\frac{\chi_0}{\beta}\frac{1}{N}\sum_{\vec{k}}\frac{1}{1+\chi_0J_{0\vec{k}}}, \tag{4.46}$$

thus obtaining χ_0. At the critical temperature χ_0 diverges and we obtain

$$\frac{kT_c}{J_0}=\frac{\frac{2}{3}}{F(-1)}, \tag{4.47}$$

where the Watson sum is defined by[13]

$$F(n)=\frac{1}{N}\sum_{\vec{k}}(1-\gamma_{\vec{k}})^n,\quad \gamma_{\vec{k}}\equiv J_{\vec{k}}/J_0. \tag{4.48}$$

From Eq. (4.47) we obtain

$$\left[\frac{kT_c}{J_0}\right]_{\rm sc}=0.4396,\quad \left[\frac{kT_c}{J_0}\right]_{\rm bcc}=0.4785,\quad \left[\frac{kT_c}{J_0}\right]_{\rm fcc}=0.4956. \tag{4.49}$$

For the region $T<T_c$, q_0 does not vanish as $\Omega\to 0$ and in this limit Eq. (4.44) becomes

$$\frac{4}{3}-\frac{q_0}{\sqrt{3}}=\sqrt{3}q_0\frac{1}{N}\sum_{\vec{k}}(1-\gamma_{\vec{k}})^{-1/2}\coth\times\left[\frac{\beta}{2}[\sqrt{3}q_0J_0(1-\gamma_{\vec{k}})^{1/2}]\right] \tag{4.50}$$

Taking $\beta\to\infty$ and anticipating q_0 negative, we obtain an expression for q_0 in the ground state

$$q_0=-\frac{4/\sqrt{3}}{3F(-\frac{1}{2})-1}. \tag{4.51}$$

For ready comparison to previous work we calculate the parameter L defined by

$$L=\frac{\sqrt{3}}{2}q_0 \tag{4.52}$$

and obtain

$$L=-\frac{2}{3F(-\frac{1}{2})-1}. \tag{4.53}$$

We thus obtain the ground-state values of L,

$$L_{\rm sc}=-0.8532,\quad L_{\rm bcc}=-0.8843,\quad L_{\rm fcc}=-0.9006. \tag{4.54}$$

For the simple-cubic case, Barma obtained $L_{\rm sc}=-0.9316$, while Fittipaldi and Tahir-Kheli obtained $L_{\rm sc}=-0.9356$. Our result is substantially larger than these.

V. CONCLUSIONS

Green's function decoupling schemes are generally criticized for being based upon unclear approximations and for failing to satisfy relevant operator identities. Our scheme is based upon the *self-consistent approximation* that those operators whose diagonal susceptibilities diverge in the ordered phase are statistically independent of all other operators. The results obtained with this scheme satisfy all relevant spin-1 identities. Also, the ground-state order parameter determined by this scheme is larger than those obtained by previous decoupling schemes.

[1]J. C. Raich and R. D. Etters, Phys. Rev. **168**, 425 (1968).
[2]K. K. Kobayashi, J. Phys. Soc. Jpn. **29**, 101 (1970).
[3]J. K. Kjems, G. Shirane, R. J. Birgeneau, and L. G. Van Uitert, Phys. Rev. Lett. **31**, 1300 (1973).
[4]M. A. Barma, Phys. Rev. B **10**, 4650 (1974).
[5]I. P. Fittipaldi and R. A. Tahir-Kheli, Phys. Rev. B **12**, 1839 (1975).
[6]D. S. Ritchie and C. Mavroyannis, Phys. Rev. B **17**, 1679 (1978).
[7]D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [Sov. Phys.—Usp. **3**, 320 (1960)].
[8]P. E. Bloomfield and N. Nafari, Phys. Rev. A **5**, 806 (1972).
[9]P. E. Bloomfield, R. Hecht, and P. Sievert, Phys. Rev. B **2**, 3714 (1970).
[10]J. G. Ramos and A. A. Gomes, Nuovo Cimento **3A**, 441 (1971).
[11]K. M. Van Vliet, J. Math. Phys. **19**, 1345 (1978).
[12]R. Kubo, J. Phys. Soc. Jpn. **17**, 1100 (1962).
[13]G. N. Watson, Q. J. Math. **10**, 266 (1939).