Isotropic spin-1 dipolar and quadrupolar systems: A Green's-function approach

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The isotropic spin-1 dipolar and quadrupolar coupled 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. In contrast to current mean-field theories, we obtain structure-dependent critical curves separating paramagnetic, dipolar, and dipolar-quadrupolar phases. We obtain the ground-state ($T=0$) order parameters for both the dipolar and quadrupolar phases, and specifically determine the dependence of the latter on the relative dipolar and quadrupolar coupling strengths.

I. INTRODUCTION

Physical systems which include both bilinear (dipolar) and biquadratic (quadrupolar) exchange interactions are usually characterized by the presence of unquenched orbital angular momenta which can couple through some form of superexchange. Such systems include the rare-earth intermetallics,[1,2] the rare-earth arsenates and phosphates[3] as well as UO_2 (Ref. 4) among others. These systems generally exhibit two ordering parameters.

The simplest Hamiltonian which can include both bilinear and biquadratic exchange is the isotropic spin-1 model which has been extensively studied using effective field calculational techniques. Chen and Levy[5] used the mean-field approximation (MFA), Ferrer and Pintanel[6] used the MFA with the Oquchi pair approximation and Chakraborty[7] used the MFA with a Lagrange multiplier method.

This and related models have also been treated by double-time Green's-function (DTGF) methods[8] which have been generally criticized for being based on weakly defined decoupling schemes and for producing ambiguous results. Recently, Bloomfield and Brown[9] have developed a DTGF decoupling scheme based on the concepts of cumulant averages and statistical independence. This scheme has produced well-defined, unambiguous results for the transverse Ising model (TIM)[9] and spin-1 isotropic quadrupolar coupled systems.[10] Their scheme is here extended to treat a model which includes competing isotropic bilinear and biquadratic exchange interactions for all temperatures and exchange ratios.

II. DOUBLE-TIME GREEN'S FUNCTIONS

The retarded commutator ($\eta=-1$) or anticommutator ($\eta=+1$) DTGF is defined[11] as

$$\langle\langle A(t);B(t')\rangle\rangle^{(\eta)}=-i\Theta(t-t')\langle[A(t),B(t')]_\eta\rangle\;, \tag{2.1}$$

with

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

and

$$\Theta(t)=\begin{cases}1, & t>0\\ 0, & t<0\;.\end{cases} \tag{2.3}$$

The single angular brackets denote a thermal average. It follows that $\langle\langle A(t);B(t')\rangle\rangle^{(\eta)}$ is a function of $t-t'$ only.

The Fourier transform of the DTGF is defined by

$$\langle\langle A;B\rangle\rangle^{(\eta)}_{E+i\epsilon}=\int_{-\infty}^{\infty}dt\,e^{i(E+i\epsilon)t}\langle\langle A(t);B\rangle\rangle^{(\eta)},\quad \epsilon\to 0^+ \tag{2.4}$$

and satisfies the equation of motion,

$$E\langle\langle A;B\rangle\rangle^{(\eta)}_E=\langle[A,B]_\eta\rangle+\langle\langle[A,H]_-;B\rangle\rangle^{(\eta)}_E\;. \tag{2.5}$$

The DTGF equation of motion represents a hierarchical series which must be decoupled to obtain a closed system of equations. Note that the Fourier-transformed DTGF, Eq. (2.4), is sectionally holomorphic with the retarded DTGF analytic in the upper half of the complex E plane.[12,13]

It has been shown[12] that the commutator DTGF cannot have a pole at $E=0$, i.e.,

$$C^{(-)}=0\;, \tag{2.6}$$

where

$$C^{(\eta)}=\lim_{E\to 0^+}E\langle\langle A;B\rangle\rangle^{(\eta)}_E\;, \tag{2.7}$$

and that the correlation $\langle BA(t)\rangle$ can be calculated as

$$\langle BA(t)\rangle=\tfrac{1}{4}(1-\eta)C^{(-\eta)}+\frac{i}{2\pi}\int_{-\infty}^{\infty}dE\left[\frac{e^{-iEt}}{e^{\beta E}+\eta}\right]\lim_{\epsilon\to 0^+}(\langle\langle A;B\rangle\rangle^{(\eta)}_{E+i\epsilon}-\langle\langle A;B\rangle\rangle^{(\eta)}_{E-i\epsilon})\;, \tag{2.8}$$

with $\beta=1/kT$. The response of the system to an external field, the generalized susceptibility, is determined by the commutator DTGF and is given by[14,15]

$$\chi_{AB}(E) = -\lim_{\epsilon\to 0^+} \langle\langle A;B\rangle\rangle^{(-)}_{E+i\epsilon}\ . \tag{2.9}$$

The $\epsilon\to 0^+$ poles of DTGF represent the excitation energies of the many particle system and must, therefore, be real.

III. MODEL—ISOTROPIC BILINEAR AND BIQUADRATIC INTERACTIONS

We use the $S=1$ operator basis consisting of the three dipole operators S_i^α, $\alpha\in\{x,y,z\}$ and the five quadrupole operators Q_i^ρ, $\rho\in\{0,1,xy,xz,yz\}$ where

$$Q_i^0=\sqrt{3}[(S_i^z)^2-\tfrac{2}{3}]\ ,\quad Q_i^1=(S_i^x)^2-(S_i^y)^2\ ,\quad Q_i^{xy}=[S_i^x,S_i^y]_+\ ,\quad Q_i^{xz}=[S_i^x,S_i^z]_+\ ,\quad Q_i^{yz}=[S_i^y,S_i^z]_+\ . \tag{3.1}$$

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

$$H_0=-\frac{\lambda}{2}\sum_{i,j,\alpha} J_{ij}S_i^\alpha S_j^\alpha-\frac{\mu}{2}\sum_{j,\rho} J_{ij}Q_i^\rho Q_j^\rho\ , \tag{3.2}$$

and uniform field couplings to Q_i^0 and S_i^z are described by

$$H_1=-\Omega_1\sum_i Q_i^0\ ,\quad \text{and } H_2=-\Omega_2\sum_i S_i^z\ . \tag{3.3}$$

We consider two cases:

(A) the possibility of an ordered phase with $\langle Q_i^0\rangle\neq 0$ by studying the full Hamiltonian $H=H_0+H_1$ in the limit of $\Omega_1\to 0$.

(B) the possibility of an ordered phase where $\langle S_i^z\rangle\neq 0$ by studying the full Hamiltonian $H=H_0+H_2$ in the limit of $\Omega_2\to 0$.

We consider case (A) first, and write the equations of motion for the eight basis operators,

$$[S_i^x,H]_-=i\sqrt{3}\Omega_1 Q_i^{yz}-i\lambda\sum_l J_{il}(S_i^zS_l^y-S_i^yS_l^z)$$
$$+i\mu\sum_l J_{il}[Q_i^{yz}(\sqrt{3}Q_l^0-Q_l^1)-Q_i^{xz}Q_l^{xy}-Q_i^{xy}Q_l^{xz}-(\sqrt{3}Q_i^0+Q_i^1)Q_l^{yz}]\ , \tag{3.4a}$$

$$[S_i^y,H]_-=-i\sqrt{3}\Omega_1 Q_i^{xz}+i\lambda\sum_l J_{il}(S_i^zS_l^x-S_i^xS_l^z)$$
$$-i\mu\sum_l J_{il}[Q_i^{xz}(\sqrt{3}Q_l^0-Q_l^1)-Q_i^{yz}Q_l^{xy}+Q_i^{xy}Q_l^{yz}-(\sqrt{3}Q_i^0-Q_i^1)Q_l^{xz}]\ , \tag{3.4b}$$

$$[S_i^z,H]_-=-i\lambda\sum_l J_{il}(S_i^yS_l^x-S_i^xS_l^y)-i\mu\sum_l J_{il}(2Q_i^{xy}Q_l^1-2Q_i^1Q_l^{xy}+Q_i^{yz}Q_l^{xz}-Q_i^{xz}Q_l^{yz})\ , \tag{3.4c}$$

$$[Q_i^0,H]_-=-i\sqrt{3}\lambda\sum_l J_{il}(Q_i^{yz}S_l^x-Q_i^{xz}S_l^y)-i\sqrt{3}\mu\sum_l J_{il}(S_i^yQ_l^{xz}-S_i^xQ_l^{yz})\ , \tag{3.4d}$$

$$[Q_i^1,H]_-=-i\lambda\sum_l J_{il}(Q_i^{yz}S_l^x-2Q_i^{xy}S_l^z+Q_i^{xz}S_l^y)-i\mu\sum_l J_{il}(2S_i^zQ_l^{xy}-S_i^yQ_l^{xz}-S_i^xQ_l^{yz})\ , \tag{3.4e}$$

$$[Q_i^{xy},H]_-=i\lambda\sum_l J_{il}(Q_i^{xz}S_l^x-Q_i^{yz}S_l^y-2Q_i^1S_l^z)+i\mu\sum_l J_{il}(2S_i^zQ_l^1-S_i^xQ_l^{xz}+S_i^yQ_l^{yz})\ , \tag{3.4f}$$

$$[Q_i^{xz},H]_-=i\sqrt{3}\Omega_1 S_i^y-i\lambda\sum_l J_{il}[Q_i^{xy}S_l^x-(Q_i^1-\sqrt{3}Q_i^0)S_l^y-Q_i^{yz}S_l^z]$$
$$+i\mu\sum_l J_{il}[S_i^xQ_l^{xy}+S_i^y(\sqrt{3}Q_l^0-Q_l^1)-S_i^zQ_l^{yz}]\ , \tag{3.4g}$$

$$[Q_i^{yz},H]_-=-i\sqrt{3}\Omega_1 S_i^x+i\lambda\sum_l J_{il}[(Q_i^1+\sqrt{3}Q_i^0)S_l^x+Q_i^{xy}S_l^y-Q_i^{xz}S_l^z)$$
$$-i\mu\sum_l J_{il}[(Q_l^1+\sqrt{3}Q_l^0)S_i^x+S_i^yQ_l^{xy}-S_i^zQ_l^{xz}]\ . \tag{3.4h}$$

Invoking translational invariance we define $\langle S_i^\alpha\rangle\equiv\alpha$, $\alpha\in\{x,y,z\}$ and

$$\langle Q_i^\rho\rangle=q_\rho\ ,\quad \rho\in\{0,1,xy,xz,yz\}\ .$$

Taking the thermal average of both sides of Eqs. (3.4a) and (3.4b) we have

$$\Omega_1 q_{xz}=\Omega_1 q_{yz}=0\ . \tag{3.5a}$$

From Eqs. (3.4d) and (3.4e) we obtain the correlation identities

$$0=(\lambda-\mu)\sum_l J_{il}(\langle Q_i^{yz}S_l^x\rangle-\langle Q_i^{xz}S_l^y\rangle)\,, \tag{3.5b}$$

$$0=(\lambda-\mu)\sum_l J_{il}(\langle Q_i^{yz}S_l^x\rangle+\langle Q_i^{xz}S_l^y\rangle-2\langle Q_i^{xy}S_l^z\rangle)\,, \tag{3.5c}$$

$$0=(\lambda-\mu)\sum_l J_{il}(\langle Q_i^{xz}S_l^x\rangle-\langle Q_i^{yz}S_l^y\rangle-2\langle Q_i^1S_l^z\rangle)\,, \tag{3.5d}$$

$$0=\sqrt{3}\Omega_1 y-(\lambda-\mu)\sum_l J_{il}[\langle Q_i^{xy}S_l^x\rangle-\langle(Q_i^1-\sqrt{3}Q_i^0)S_l^y\rangle-\langle Q_i^{yz}S_l^z\rangle]\,, \tag{3.5e}$$

$$0=\sqrt{3}\Omega_1 x-(\lambda-\mu)\sum_l J_{il}[\langle(Q_i^1+\sqrt{3}Q_i^0)S_l^x\rangle+\langle Q_i^{xy}S_l^y\rangle-\langle Q_i^{xz}S_l^z\rangle]\,. \tag{3.5f}$$

In addition we note that due to the rotational symmetry of the full H, all single site correlations vanish with the exception of q_0.

We define

$$\begin{aligned}&G_{ij}^{\alpha,R(\eta)}\equiv\langle\langle S_i^\alpha;R_j\rangle\rangle^{(\eta)},\quad K_{ij}^{\alpha,R(\eta)}\equiv\langle[S_i^\alpha,R_j]_\eta\rangle\,,\\&G_{ij}^{\rho,R(\eta)}\equiv\langle\langle Q_i^\rho;R_j\rangle\rangle^{(\eta)},\quad K_{ij}^{\rho,R(\eta)}\equiv\langle[Q_i^\rho,R_j]_\eta\rangle\,,\end{aligned} \tag{3.6}$$

for R_j any member of the basis set of operators and $R\equiv\langle R_j\rangle$. Using the equations of motion of the basis operators, Eq. (3.4), the eight DTGF equations of motion are

$$\begin{aligned}EG_{ij}^{x,R(\eta)}=K_{ij}^{x,R(\eta)}&+i\sqrt{3}\Omega_1G_{ij}^{yz,R(\eta)}-i\lambda\sum_l J_{il}(\langle\langle S_i^zS_l^y;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^yS_l^z;R_j\rangle\rangle_E^{(\eta)})\\&+i\mu\sum_l J_{il}[\langle\langle Q_i^{yz}(\sqrt{3}Q_l^0+Q_l^1);R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^{xz}Q_l^{xy};R_j\rangle\rangle_E^{(\eta)}+\langle\langle Q_i^{xy}Q_l^{xz};R_j\rangle\rangle_E^{(\eta)}-\langle\langle(\sqrt{3}Q_i^0+Q_i^1)Q_l^{yz};R_j\rangle\rangle_E^{(\eta)}]\,,\end{aligned} \tag{3.7a}$$

$$\begin{aligned}EG_{ij}^{y,R(\eta)}=K_{ij}^{y,R(\eta)}&-i\sqrt{3}\Omega_1G_{ij}^{xz,R(\eta)}-i\lambda\sum_l J_{il}(\langle\langle S_i^zS_l^x;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xS_l^z;R_j\rangle\rangle_E^{(\eta)})\\&-i\mu\sum_l J_{il}[\langle\langle Q_i^{xz}(\sqrt{3}Q_l^0-Q_l^1);R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^{yz}Q_l^{xy};R_j\rangle\rangle_E^{(\eta)}+\langle\langle Q_i^{xy}Q_l^{yz};R_j\rangle\rangle_E^{(\eta)}-\langle\langle(\sqrt{3}Q_i^0-Q_i^1)Q_l^{xz};R_j\rangle\rangle_E^{(\eta)}]\,,\end{aligned} \tag{3.7b}$$

$$\begin{aligned}EG_{ij}^{z,R(\eta)}=K_{ij}^{z,R(\eta)}&-i\lambda\sum_l J_{il}(\langle\langle S_i^yS_l^x;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xS_l^y;R_j\rangle\rangle_E^{(\eta)})\\&-i\mu\sum_l J_{il}(2\langle\langle Q_i^{xy}Q_l^1;R_j\rangle\rangle_E^{(\eta)}-2\langle\langle Q_i^1Q_l^{xy};R_j\rangle\rangle_E^{(\eta)}+\langle\langle Q_i^{yz}Q_l^{xz};R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^{xz}Q_l^{yz};R_j\rangle\rangle_E^{(\eta)})\,,\end{aligned} \tag{3.7c}$$

$$EG_{ij}^{0,R(\eta)}=K_{ij}^{0,R(\eta)}-i\sqrt{3}\lambda\sum_l J_{il}(\langle\langle Q_i^{yz}S_l^x;R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^{xz}S_l^y;R_j\rangle\rangle_E^{(\eta)})-i\sqrt{3}\mu\sum_l J_{il}(\langle\langle S_i^yQ_l^{xz};R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xQ_l^{yz};R_j\rangle\rangle_E^{(\eta)})\,, \tag{3.7d}$$

$$\begin{aligned}EG_{ij}^{1,R(\eta)}=K_{ij}^{1,R(\eta)}&-i\lambda\sum_l J_{il}(\langle\langle Q_i^{yz}S_l^z;R_j\rangle\rangle_E^{(\eta)}-2\langle\langle Q_i^{xy}S_l^z;R_j\rangle\rangle_E^{(\eta)}+\langle\langle Q_i^{xz}S_l^y;R_j\rangle\rangle_E^{(\eta)})\\&-i\mu\sum_l J_{il}(2\langle\langle S_i^zQ_l^{xy};R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^yQ_l^{xz};R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xQ_l^{yz};R_j\rangle\rangle_E^{(\eta)})\,,\end{aligned} \tag{3.7e}$$

$$\begin{aligned}EG_{ij}^{xy,R(\eta)}=K_{ij}^{xy,R(\eta)}&+i\lambda\sum_l J_{il}(\langle\langle Q_i^{xz}S_l^x;R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^{yz}S_l^y;R_j\rangle\rangle_E^{(\eta)}-2\langle\langle Q_i^1S_l^z;R_j\rangle\rangle_E^{(\eta)})\\&+i\mu\sum_l J_{il}(2\langle\langle S_i^zQ_l^1;R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^xQ_l^{xz};R_j\rangle\rangle_E^{(\eta)}+\langle\langle S_i^yQ_l^{yz};R_j\rangle\rangle_E^{(\eta)})\,,\end{aligned} \tag{3.7f}$$

$$\begin{aligned}EG_{ij}^{xz,R(\eta)}=K_{ij}^{xz,R(\eta)}&+i\sqrt{3}\Omega_1G_{ij}^{y,R(\eta)}-i\lambda\sum_l J_{il}[\langle\langle Q_i^{xy}S_l^x;R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^{yz}S_l^z;R_j\rangle\rangle_E^{(\eta)}-\langle\langle(Q_i^1-\sqrt{3}Q_i^0)S_l^y;R_j\rangle\rangle_E^{(\eta)}]\\&+i\mu\sum_l J_{il}[\langle\langle S_i^xQ_l^{xy};R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^zQ_l^{yz};R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^y(\sqrt{3}Q_l^0-Q_l^1);R_j\rangle\rangle_E^{(\eta)}]\,,\end{aligned} \tag{3.7g}$$

$$\begin{aligned}EG_{ij}^{yz,R(\eta)}=K_{ij}^{yz,R(\eta)}&-i\sqrt{3}\Omega_1G_{ij}^{x,R(\eta)}+i\lambda\sum_l J_{il}[\langle\langle(Q_i^1+\sqrt{3}Q_i^0)S_l^x;R_j\rangle\rangle_E^{(\eta)}+\langle\langle Q_i^{xy}S_l^y;R_j\rangle\rangle_E^{(\eta)}-\langle\langle Q_i^{xz}S_l^z;R_j\rangle\rangle_E^{(\eta)}]\\&-i\mu\sum_l J_{il}[\langle\langle(Q_l^1+\sqrt{3}Q_l^0)S_i^x;R_j\rangle\rangle_E^{(\eta)}+\langle\langle S_i^yQ_l^{xy};R_j\rangle\rangle_E^{(\eta)}-\langle\langle S_i^zQ_l^{xz};R_j\rangle\rangle_E^{(\eta)}]\,.\end{aligned} \tag{3.7h}$$

IV. DECOUPLING SCHEME—CASE A

In order to obtain a closed soluble set of equations, the equations of motion for the DTGF's must be approximately decoupled. A decoupling scheme based on the concepts of cumulants and statistical independence has been used by Brown and Bloomfield[9,10] for the TIM and quadrupolar coupled systems and we follow their scheme here.

On the right-hand side of the equations of motion there are thermal averages of three operator products all of which contain the operator R_j and are of the form

$$\langle Q_i^\rho(t)S_l^\alpha(t)R_j\rangle\ ,\quad \langle R_jQ_i^\rho(t)S_l^\alpha(t)\rangle\ ,\quad \langle S_i^\alpha(t)S_l^{\alpha'}(t)R_j\rangle\ ,$$
$$\langle R_jS_i^\alpha(t)S_l^{\alpha'}(t)\rangle\ ,\quad \langle Q_i^\rho(t)Q_l^{\rho'}(t)R_j\rangle\ ,\quad \langle R_jQ_i^\rho(t)Q_l^{\rho'}(t)\rangle\ ,$$

which appear in

$$\langle\langle Q_i^\rho S_l^\alpha;R_j\rangle\rangle_E^{(\eta)}\ ,\quad \langle\langle S_i^\alpha S_l^{\alpha'};R_j\rangle\rangle_E^{(\eta)}\ ,\quad \langle\langle Q_i^\rho Q_l^{\rho'};R_j\rangle\rangle_E^{(\eta)}\ .$$

The cumulant averages[16] of the first two three-operator products are

$$\begin{aligned}\langle Q_i^\rho(t)S_l^\alpha(t)R_j\rangle_c &= \langle Q_i^\rho(t)S_l^\alpha(t)R_j\rangle - q_\rho\langle S_l^\alpha(t)R_j\rangle\\ &\quad -\alpha\langle Q_i^\rho(t)R_j\rangle - R(\langle Q_i^\rho S_l^\alpha\rangle - 2\alpha q_\rho)\ ,\\ \langle R_jQ_i^\rho(t)S_l^\alpha(t)\rangle_c &= \langle R_jQ_i^\rho(t)S_l^\alpha(t)\rangle - q_\rho\langle R_jS_l^\alpha(t)\rangle\\ &\quad -\alpha\langle R_jQ_i^\rho(t)\rangle - R(\langle Q_i^\rho S_l^\alpha\rangle - 2\alpha q_\rho)\ ,\\ R &\equiv \langle R_j\rangle\ .\end{aligned} \tag{4.1}$$

The decoupling is based on the assumption that one can choose R_j such that at least one of the operators in every three operator products to be decoupled is statistically independent of the others. This allows us to set[16]

$$\langle Q_i^\rho(t)S_l^\alpha(t)R_j\rangle_c = \langle R_jQ_i^\rho(t)S_l^\alpha(t)\rangle_c = 0\ , \tag{4.2}$$

and obtain the approximation

$$\langle[Q_i^\rho(t)S_l^\alpha(t),R_j]_\eta\rangle = q_\rho\langle[S_l^\alpha(t),R_j]_\eta\rangle + \alpha\langle[Q_i^\rho(t),R_j]_\eta\rangle + (1+\eta)R(\langle Q_i^\rho S_l^\alpha\rangle - 2\alpha q_\rho) \tag{4.3}$$

to obtain the decoupling approximation

$$\langle\langle Q_i^\rho S_l^\alpha;R_j\rangle\rangle_E^{(\eta)} = q_\rho G_{lj}^{\alpha,R(\eta)} + \alpha G_{ij}^{\rho,R(\eta)} + \frac{(1+\eta)}{E}R(\langle Q_i^\rho S_l^\alpha\rangle - 2\alpha q_\rho)\ . \tag{4.4}$$

Proceeding in a similar fashion we obtain the decoupling approximations

$$\langle\langle Q_i^\rho Q_l^{\rho'};R_j\rangle\rangle_E^{(\eta)} = q_\rho G_{lj}^{\rho',R(\eta)} + q_{\rho'} G_{ij}^{\rho,R(\eta)} + \frac{(1+\eta)R}{E}(\langle Q_i^\rho Q_l^{\rho'}\rangle - 2q_\rho q_{\rho'}) \tag{4.5}$$

and

$$\langle\langle S_i^\alpha S_l^{\alpha'};R_j\rangle\rangle_E^{(\eta)} = \alpha G_{lj}^{\alpha',R(\eta)} + \alpha' G_{ij}^{\alpha,R(\eta)} + \frac{(1+\eta)R}{E}(\langle S_i^\alpha S_l^{\alpha'}\rangle - 2\alpha\alpha')\ . \tag{4.6}$$

The statistically independent operators will be self-consistently identified as those with diagonal susceptibilities [in the approximation of Eqs. (4.4)—(4.6)] that diverge as q_0 orders.

Using the decoupling approximations in the DTGF equations of motion and imposing the correlation identities [Eqs. (3.5)] together with the fact that the only nonvanishing single site correlation is q_0, we obtain, after performing a spatial Fourier transform, the approximately decoupled equations,

$$EG_{\mathbf{k}}^{x,R(\eta)} = K_{\mathbf{k}}^{x,R(\eta)} + i\sqrt{3}\Omega_1 G_{\mathbf{k}}^{yz,R(\eta)} + i\sqrt{3}\mu q_0 J_{0\mathbf{k}} G_{\mathbf{k}}^{yz,R(\eta)}\ , \tag{4.7a}$$

$$EG_{\mathbf{k}}^{y,R(\eta)} = K_{\mathbf{k}}^{y,R(\eta)} - i\sqrt{3}(\Omega_1 + \mu q_0 J_{0\mathbf{k}})G_{\mathbf{k}}^{xz,R(\eta)}\ , \tag{4.7b}$$

$$EG_{\mathbf{k}}^{z,R(\eta)} = K_{\mathbf{k}}^{z,R(\eta)}\ , \tag{4.7c}$$

$$EG_{\mathbf{k}}^{0,R(\eta)} = K_{\mathbf{k}}^{0,R(\eta)}\ , \tag{4.7d}$$

$$EG_{\mathbf{k}}^{1,R(\eta)} = K_{\mathbf{k}}^{1,R(\eta)}\ , \tag{4.7e}$$

$$EG_{\mathbf{k}}^{xy,R(\eta)} = K_{\mathbf{k}}^{xy,R(\eta)}\ , \tag{4.7f}$$

$$EG_{\mathbf{k}}^{xz,R(\eta)} = K_{\mathbf{k}}^{xz,R(\eta)} + i\sqrt{3}[\Omega_1 + q_0(\mu J_0 - \lambda J_{\mathbf{k}})]G_{\mathbf{k}}^{y,R(\eta)}\ , \tag{4.7g}$$

$$EG_{\mathbf{k}}^{yz,R(\eta)} = K_{\mathbf{k}}^{yz,R(\eta)} - i\sqrt{3}[\Omega_1 + q_0(\mu J_0 - \lambda J_{\mathbf{k}}]G_{\mathbf{k}}^{x,R(\eta)}\ , \tag{4.7h}$$

where, e.g.,

$$\begin{aligned} G_{\mathbf{k}}^{x,R(\eta)} &= \frac{1}{N}\sum_{i,j} e^{i\mathbf{k}\cdot\mathbf{r}_{ij}} G_{ij}^{x,R(\eta)}\ ,\\ K_{\mathbf{k}}^{x,R(\eta)} &= \frac{1}{N}\sum_{i,j} e^{i\mathbf{k}\cdot\mathbf{r}_{ij}} k_{ij}^{x,R(\eta)}\ ,\end{aligned} \tag{4.8}$$

and

$$\begin{aligned} J_{\mathbf{k}} &= \frac{1}{N}\sum_{i,j} e^{i\mathbf{k}\cdot\mathbf{r}_{ij}} J_{ij}\ ,\\ J_{0\mathbf{k}} &= J_0 - J_{\mathbf{k}}\ .\end{aligned} \tag{4.9}$$

Equations (4.7c)—(4.7f) determine

$$\begin{aligned} &G_{\mathbf{k}}^{z,R(\eta)} = K_{\mathbf{k}}^{z,R(\eta)}/E\ ,\quad G_{\mathbf{k}}^{0,R(\eta)} = K_{\mathbf{k}}^{0,R(\eta)}/E\ ,\\ &G_{\mathbf{k}}^{1,R(\eta)} = K_{\mathbf{k}}^{1,R(\eta)}/E\ ,\quad G_{\mathbf{k}}^{xy,R(\eta)} = K_{\mathbf{k}}^{xy,R(\eta)}/E\ ,\end{aligned} \tag{4.10}$$

while Eqs. (4.7a), (4.7b), (4.7g), and (4.7h) may be solved to obtain

$$G_{\mathbf{k}}^{x,R(\eta)} = \frac{EK_{\mathbf{k}}^{x,R(\eta)} + i\sqrt{3}(\Omega_1 + q_0\mu J_{0\mathbf{k}})K_{\mathbf{k}}^{yz,R(\eta)}}{E^2 - \omega_{\mathbf{k}}^2}\ , \tag{4.11}$$

$$G_{\mathbf{k}}^{y,R(\eta)} = \frac{EK_{\mathbf{k}}^{y,R(\eta)} - i\sqrt{3}(\Omega_1 + q_0\mu J_{0\mathbf{k}})K_{\mathbf{k}}^{xz,R(\eta)}}{E^2 - \omega_{\mathbf{k}}^2}\ , \tag{4.12}$$

$$G_{\mathbf{k}}^{xz,R(\eta)} = \frac{EK_{\mathbf{k}}^{xz,R(\eta)} + i\sqrt{3}[\Omega_1 + q_0(\mu J_0 - \lambda J_{\mathbf{k}})]K_{\mathbf{k}}^{y,R(\eta)}}{E^2 - \omega_{\mathbf{k}}^2}\ , \tag{4.13}$$

$$G_{\mathbf{k}}^{yz,R(\eta)}=\frac{EK_{\mathbf{k}}^{yz,R(\eta)}-i\sqrt{3}[\Omega_1+q_0(\mu J_0-\lambda J_{\mathbf{k}})]K_{\mathbf{k}}^{x,R(\eta)}}{E^2-\omega_{\mathbf{k}}^2}\,, \tag{4.14}$$

with

$$\omega_{\mathbf{k}}=\sqrt{3}[\Omega_1+q_0\mu J_0(1-\gamma_{\mathbf{k}})]^{1/2}\times[\Omega_1+q_0\mu J_0(1-\lambda\gamma_{\mathbf{k}}/\mu)]^{1/2}\,, \tag{4.15}$$

where

$$\gamma_{\mathbf{k}}\equiv J_{\mathbf{k}}/J_0\;. \tag{4.16}$$

In the limit $\Omega_1\to 0$,

$$\omega_{\mathbf{k}}=\sqrt{3}q_0\mu J_0(1-\gamma_{\mathbf{k}})^{1/2}(1-\lambda\gamma_{\mathbf{k}}/\mu)^{1/2}\;. \tag{4.17}$$

Since (4.17) represents the excitation energy of the system and is real only for $\lambda/\mu\le 1$, this solution is restricted to the region $\lambda\le\mu$.

Using (2.9), the diagonal susceptibilities are given by

$$\chi_\mu=-G_0^{\mu,\mu(-)}(E=0)\;. \tag{4.18}$$

Defining

$$\chi_0\equiv q_0/\Omega_1\;, \tag{4.19}$$

and using Eqs. (4.10)–(4.14), we obtain as the only nonvanishing diagonal susceptibilities

$$\chi_0=\chi_{xz}=\chi_{yz}\;, \tag{4.20}$$

$$\chi_x=\chi_y=\frac{\chi_0}{1+\chi_0 J_0(\mu-\lambda)}\;. \tag{4.21}$$

For q_0 ordering,

$$\lim_{\Omega_1\to 0} q_0\neq 0\;, \tag{4.22}$$

and from Eqs. (4.20)–(4.21) χ_0, χ_{xz}, and χ_{yz} are the only diagonal susceptibilities which diverge as q_0 orders. We identify Q_i^0, Q_i^{xz}, and Q_i^{yz} as the members of the set of basis operators [Eq. (3.1)], which are statistically independent of every other member of the set.

For self-consistency, we require that at least one of these statistically independent operators appear in each of the "higher-order" DTGF's on the right-hand side of the equations of motion, Eqs. (3.7). To meet this requirement we must[10] *choose R_j to be one of the statistically independent operators.*

Defining

$$a_{ij}^{R,\nu}=\begin{cases}\langle R_i S_j^\nu\rangle\,, & \nu\in\{x,y,z\}\\ \langle R_i Q_j^\nu\rangle\,, & \nu\in\{0,1,xy,xz,yz\}\end{cases} \tag{4.23}$$

and

$$a_{\mathbf{k}}^{R,\nu}\equiv\frac{1}{N}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_{ij}}a_{ij}^{R,\nu}\,, \tag{4.24}$$

we obtain, by using Eqs. (4.11)–(4.14) in Eq. (2.8),

$$a_{\mathbf{k}}^{R,x}=\frac{-K_{\mathbf{k}}^{x,R(-)}}{2}+\frac{i\sqrt{3}}{2}\frac{(\Omega_1+\mu q_0 J_{0\mathbf{k}})}{\omega_{\mathbf{k}}}\times K_{\mathbf{k}}^{yz,R(-)}\coth\left[\frac{\beta\omega_{\mathbf{k}}}{2}\right]\,, \tag{4.25}$$

$$a_{\mathbf{k}}^{R,y}=\frac{-K_{\mathbf{k}}^{y,R(-)}}{2}-\frac{i\sqrt{3}}{2}\frac{(\Omega_1+\mu q_0 J_{0\mathbf{k}})}{\omega_{\mathbf{k}}}\times K_{\mathbf{k}}^{xz,R(-)}\coth\left[\frac{\beta\omega_{\mathbf{k}}}{2}\right]\,, \tag{4.26}$$

$$a_{\mathbf{k}}^{R,xz}=\frac{-K_{\mathbf{k}}^{xz,R(-)}}{2}+\frac{i\sqrt{3}}{2}\frac{[\Omega_1+q_0(\mu J_0-\lambda J_{\mathbf{k}})]}{\omega_{\mathbf{k}}}\times K_{\mathbf{k}}^{y,R(-)}\coth\left[\frac{\beta\omega_{\mathbf{k}}}{2}\right]\,, \tag{4.27}$$

$$a_{\mathbf{k}}^{R,yz}=\frac{-K_{\mathbf{k}}^{yz,R(-)}}{2}-\frac{i\sqrt{3}}{2}\frac{[\Omega_1+q_0(\mu J_0-\lambda J_{\mathbf{k}})]}{\omega_{\mathbf{k}}}\times K_{\mathbf{k}}^{x,R(-)}\coth\left[\frac{\beta\omega_{\mathbf{k}}}{2}\right]\,. \tag{4.28}$$

Using Eq. (4.10) in Eq. (2.8) yields a series of identities. It is important to note that the correlation Eqs. (4.25)–(4.28) are obtained from both the $\eta=+1$ and $\eta=-1$ versions of Eq. (2.8).

Using $R_j=Q_j^0$, Q_j^{xz}, and Q_j^{yz} in Eqs. (4.25)–(4.28) gives

$$a_{\mathbf{k}}^{0,x}=a_{\mathbf{k}}^{0,y}=a_{\mathbf{k}}^{0,xz}=a_{\mathbf{k}}^{0,yz}=a_{\mathbf{k}}^{xz,x}=a_{\mathbf{k}}^{xz,yz}=a_{\mathbf{k}}^{yz,xz}=a_{\mathbf{k}}^{yz,y}=0\,, \tag{4.29}$$

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

$$a_{\mathbf{k}}^{yz,x}=-i\sqrt{3}q_0/2\,, \tag{4.30}$$

$$a_{\mathbf{k}}^{xz,y}=i\sqrt{3}q_0/2\,, \tag{4.31}$$

$$a_{\mathbf{k}}^{xz,xz}=a_{\mathbf{k}}^{yz,yz}=\tfrac{3}{2}q_0\frac{[\Omega_1+q_0(\mu J_0-\lambda J_{\mathbf{k}})]}{\omega_{\mathbf{k}}}\coth\left[\frac{\beta\omega_{\mathbf{k}}}{2}\right]\,. \tag{4.32}$$

Performing the sum over all $\mathbf{k}$ for Eqs. (4.30) and (4.31) and using the $S=1$ identities,

$$Q_l^{xz}S_l^y=i(\sqrt{3}Q_l^0-Q_l^1)/2\,,\qquad Q_l^{yz}S_l^x=-i(\sqrt{3}Q_l^0+Q_l^1)/2\,, \tag{4.33}$$

we find $\langle Q_l^1\rangle=q_1=0$. Summing Eq. (4.32) over all $\mathbf{k}$ and using the $S=1$ identities,

$$(Q_l^{xz})^2=\frac{1}{2}\left[\frac{4}{3}-\frac{Q_l^0}{\sqrt{3}}-Q_l^1\right]\,,\qquad (Q_l^{yz})^2=\frac{1}{2}\left[\frac{4}{3}-\frac{Q_l^0}{\sqrt{3}}+Q_l^1\right]\,, \tag{4.34}$$

we obtain

$$\frac{4}{3}-\frac{q_0}{\sqrt{3}}=\frac{3q_0}{N}\sum_{\mathbf{k}}\frac{[\Omega_1+q_0(\mu J_0-\lambda J_{\mathbf{k}})]}{\omega_{\mathbf{k}}}\coth\left[\frac{\beta\omega_{\mathbf{k}}}{2}\right]\,, \tag{4.35}$$

with $\omega_{\mathbf{k}}$ given by Eq. (4.15).

For the nonordering region we use $q_0=\chi_0\Omega_1$ to write Eq. (4.35) in the form

$$\frac{4}{3}-\frac{q_0}{\sqrt{3}}=\frac{\sqrt{3}\chi_0\Omega_1}{N}\sum_{\mathbf{k}}\left[\frac{1+\chi_0(\mu J_0-\lambda J_{\mathbf{k}})}{1+\chi_0\mu J_{0\mathbf{k}}}\right]^{1/2} \times\coth\left[\frac{\beta\sqrt{3}\Omega_1}{2}(1+\chi_0\mu J_{0\mathbf{k}})^{1/2} \times[1+\chi_0(\mu J_0-\lambda J_{\mathbf{k}})]^{1/2}\right], \tag{4.36}$$

and take Ω_1 and $q_0 \to 0$ to obtain

$$\frac{2}{3}=\frac{\chi_0}{\beta}\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{1+\mu\chi_0 J_{0\mathbf{k}}}. \tag{4.37}$$

At the critical temperature χ_0 diverges and we obtain

$$\frac{kT_c}{\mu J_0}=\frac{\frac{2}{3}}{F(-1)}, \tag{4.38}$$

where the Watson sum is defined by[17]

$$F(n)\equiv\frac{1}{N}\sum_{\mathbf{k}}(1-\gamma_{\mathbf{k}})^n. \tag{4.39}$$

We find the critical temperature to be structure dependent with

$$\left[\frac{kT_c}{\mu J_0}\right]_{\rm sc}=0.4396, \quad \left[\frac{kT_c}{\mu J_0}\right]_{\rm bcc}=0.4785, \quad \left[\frac{kT_c}{\mu J_0}\right]_{\rm fcc}=0.4956. \tag{4.40}$$

In the notation of Chen and Levy,[5] who use the MFA, we find

$$\frac{kT_c}{I_{20}}=\frac{1}{F(-1)}=\begin{cases}0.7434, & \text{fcc},\\ 0.7178, & \text{bcc},\\ 0.6594, & \text{sc},\end{cases} \tag{4.41}$$

while their structure-independent result is

$$\frac{kT_c}{I_{20}}=0.72. \tag{4.42}$$

In agreement with mean-field theory, however, we find that T_c is independent of λ. Thus, in the special case of $\lambda=0$ our Hamiltonian, Eq. (3.2), reduces to the isotropic quadrupolar model and our results are identical to those obtained by applying this scheme to that case.[10]

For the ordering region ($T<T_c$), q_0 does not vanish as $\Omega_1 \to 0$ and in this limit Eq. (4.35) becomes

$$\frac{4}{3}-\frac{q_0}{\sqrt{3}}=\frac{\sqrt{3}q_0}{\mu N}\sum_{\mathbf{k}}\left[\frac{1-\lambda\gamma_{\mathbf{k}}/\mu}{1-\gamma_{\mathbf{k}}}\right]^{1/2} \times\coth\left[\frac{\beta}{2}\sqrt{3}\mu q_0 J_0(1-\gamma_{\mathbf{k}})^{1/2} \times(1-\lambda\gamma_{\mathbf{k}}/\mu)^{1/2}\right]. \tag{4.43}$$

TABLE I. Ground state q_0 vs the ratio λ/μ ($=I_{10}/I_{20}$ in Chen and Levy) for bcc lattice at $T=0$ [from Eq. (4.44)].

λ/μ	$-\sqrt{3}q_0$
0.0	1.779 886
0.1	1.794 279
0.2	1.808 121
0.3	1.823 370
0.4	1.840 210
0.5	1.858 854
0.6	1.879 644
0.7	1.903 104
0.8	1.929 907
0.9	1.961 336
1.0	2.00

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

$$\sqrt{3}q_0=-\frac{4}{\frac{3}{N}\sum_{\mathbf{k}}\left[\frac{1-\lambda\gamma_{\mathbf{k}}/\mu}{1-\gamma_{\mathbf{k}}}\right]^{1/2}-1}, \tag{4.44}$$

where $\sqrt{3}q_0=\langle O_0^{[2]}\rangle$ in the notation of Chen and Levy. The ground state q_0 for a bcc lattice structure is tabulated as a function of the ratio λ/μ in Table I and shown in Fig. 1.

V. DECOUPLING SCHEME—CASE B

In this case we study the Hamiltonian,

$$H=-\Omega_2\sum_i S_i^z+H_0, \tag{5.1}$$

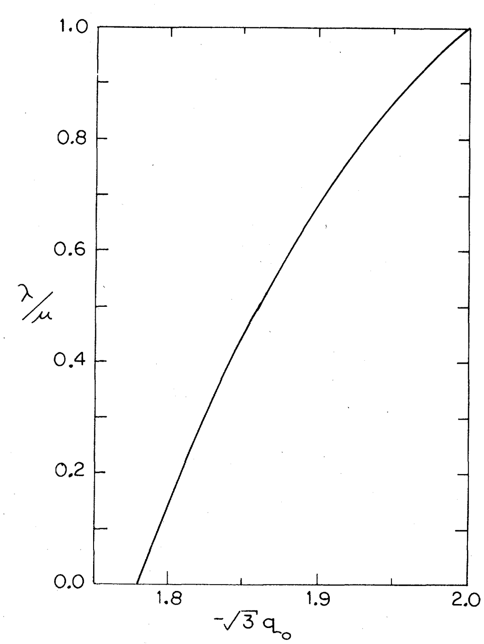

FIG. 1. $\lambda/\mu=I_{10}/I_{20}$ vs $-\sqrt{3}q_0=-\langle O_0^2\rangle$ at $T=0$ for bcc lattice from Eq. (4.44) and Table I.

in the limit $\Omega_2 \to 0$ in order to study the possibility of z ordering. From the symmetry of (5.1), the only nonvanishing correlations are z and q_0. Proceeding in a manner identical to that followed in Sec. III we obtain the correlation identities

$$\Omega_2 x = \Omega_2 y = 0 , \tag{5.2a}$$

$$0 = (\lambda-\mu) \sum_l J_{il} (\langle Q_i^{yz} S_l^x \rangle - \langle Q_i^{xz} S_l^y \rangle) , \tag{5.2b}$$

$$0 = 2\Omega_2 q_{xy} - (\lambda-\mu) \sum_l J_{il} (\langle Q_i^{yz} S_l^x \rangle - \langle Q_i^{xz} S_l^y \rangle - 2\langle Q_i^{xz} S_l^z \rangle) , \tag{5.2c}$$

$$0 = 2\Omega_2 q_1 - (\lambda-\mu) \sum_l J_{il} (\langle Q_i^{xz} S_l^x - \langle Q_i^{yz} S_l^y \rangle - 2\langle Q_i^{1} S_l^z \rangle) , \tag{5.2d}$$

$$0 = \Omega_2 q_{yz} - (\lambda-\mu) \sum_l J_{il} [\langle Q_i^{xy} S_l^x \rangle - \langle Q_i^{yz} S_l^z \rangle - \langle (Q_i^1 - \sqrt{3} Q_i^0) S_l^y \rangle] , \tag{5.2e}$$

$$0 = \Omega_2 q_{xz} - (\lambda-\mu) \sum_l J_{il} [\langle Q_i^{xy} S_l^y \rangle - \langle Q_i^{xz} S_l^z \rangle + \langle (Q_i^1 + \sqrt{3} Q_i^0) S_l^x \rangle] , \tag{5.2f}$$

and the approximate Green's functions,

$$G_{\mathbf{k}}^{z,R(\eta)} = \frac{K_{\mathbf{k}}^{z,R(\eta)}}{E} , \quad G_{\mathbf{k}}^{0,R(\eta)} = K_{\mathbf{k}}^{0,R(\eta)} , \tag{5.3}$$

$$G_{\mathbf{k}}^{1,R(\eta)} = \frac{E K_{\mathbf{k}}^{1,R(\eta)} + 2iB_{\mathbf{k}} K_{\mathbf{k}}^{xy,R(\eta)}}{E^2 - 4B_{\mathbf{k}}^2} , \quad G_{\mathbf{k}}^{xy,R(\eta)} = \frac{E K_{\mathbf{k}}^{xy,R(\eta)} - 2iB_{\mathbf{k}} K_{\mathbf{k}}^{1,R(\eta)}}{E^2 - 4B_{\mathbf{k}}^2} , \tag{5.4}$$

$$\begin{aligned} G_{\mathbf{k}}^{x,R(\eta)} = \frac{1}{\Delta_{\mathbf{k}}(E)} \{ & E^3 K_{\mathbf{k}}^{x,R(\eta)} + iE^2 (A_{\mathbf{k}} K_{\mathbf{k}}^{y,R(\eta)} + C_{\mathbf{k}} K_{\mathbf{k}}^{yz,R(\eta)}) - E[(B_{\mathbf{k}}^2 + C_{\mathbf{k}} D_{\mathbf{k}}) K_{\mathbf{k}}^{x,R(\eta)} - (A_{\mathbf{k}} + B_{\mathbf{k}}) C_{\mathbf{k}} K_{\mathbf{k}}^{xz,R(\eta)}] \\ & - i(A_{\mathbf{k}} B_{\mathbf{k}} - C_{\mathbf{k}} D_{\mathbf{k}})(B_{\mathbf{k}} K_{\mathbf{k}}^{y,R(\eta)} - C_{\mathbf{k}} K_{\mathbf{k}}^{yz,R(\eta)}) \} , \end{aligned} \tag{5.5}$$

$$\begin{aligned} G_{\mathbf{k}}^{y,R(\eta)} = \frac{1}{\Delta_{\mathbf{k}}(E)} \{ & E^3 K_{\mathbf{k}}^{y,R(\eta)} - iE^2 (A_{\mathbf{k}} K_{\mathbf{k}}^{x,R(\eta)} + C_{\mathbf{k}} K_{\mathbf{k}}^{yz,R(\eta)}) - E[(B_{\mathbf{k}}^2 + C_{\mathbf{k}} D_{\mathbf{k}}) K_{\mathbf{k}}^{y,R(\eta)} - (A_{\mathbf{k}} + B_{\mathbf{k}}) C_{\mathbf{k}} K_{\mathbf{k}}^{yz,R(\eta)}] \\ & + i(A_{\mathbf{k}} B_{\mathbf{k}} - C_{\mathbf{k}} D_{\mathbf{k}})(B_{\mathbf{k}} K_{\mathbf{k}}^{x,R(\eta)} - C_{\mathbf{k}} K_{\mathbf{k}}^{xz,R(\eta)}) \} , \end{aligned} \tag{5.6}$$

$$\begin{aligned} G_{\mathbf{k}}^{xz,R(\eta)} = \frac{1}{\Delta_{\mathbf{k}}(E)} \{ & E^3 K_{\mathbf{k}}^{xz,R(\eta)} + iE^2 (D_{\mathbf{k}} K_{\mathbf{k}}^{y,R(\eta)} + B_{\mathbf{k}} K_{\mathbf{k}}^{yz,R(\eta)}) + E[(A_{\mathbf{k}} + B_{\mathbf{k}}) D_{\mathbf{k}} K_{\mathbf{k}}^{x,R(\eta)} - (A_{\mathbf{k}}^2 + C_{\mathbf{k}} D_{\mathbf{k}}) K_{\mathbf{k}}^{xz,R(\eta)}] \\ & + i(A_{\mathbf{k}} B_{\mathbf{k}} - C_{\mathbf{k}} D_{\mathbf{k}})(D_{\mathbf{k}} K_{\mathbf{k}}^{y,R(\eta)} - A_{\mathbf{k}} K_{\mathbf{k}}^{yz,R(\eta)}) \} , \end{aligned} \tag{5.7}$$

$$\begin{aligned} G_{\mathbf{k}}^{yz,R(\eta)} = \frac{1}{\Delta_{\mathbf{k}}(E)} \{ & E^3 K_{\mathbf{k}}^{yz,R(\eta)} - iE^2 (D_{\mathbf{k}} K_{\mathbf{k}}^{x,R(\eta)} + B_{\mathbf{k}} K_{\mathbf{k}}^{xz,R(\eta)}) + E[(A_{\mathbf{k}} + B_{\mathbf{k}}) D_{\mathbf{k}} K_{\mathbf{k}}^{y,R(\eta)} - (A_{\mathbf{k}}^2 + C_{\mathbf{k}} D_{\mathbf{k}}) K_{\mathbf{k}}^{yz,R(\eta)}] \\ & - i(A_{\mathbf{k}} B_{\mathbf{k}} - C_{\mathbf{k}} D_{\mathbf{k}})(D_{\mathbf{k}} K_{\mathbf{k}}^{x,R(\eta)} - A_{\mathbf{k}} K_{\mathbf{k}}^{xz,R(\eta)}) \} , \end{aligned} \tag{5.8}$$

where

$$A_{\mathbf{k}} = \Omega_2 + \lambda z J_{0\mathbf{k}} , \tag{5.9a}$$

$$B_{\mathbf{k}} = \Omega_2 + z(\lambda J_0 - \mu J_{\mathbf{k}}) , \tag{5.9b}$$

$$C_{\mathbf{k}} = \sqrt{3} q_0 \mu J_{0\mathbf{k}} , \tag{5.9c}$$

$$D_{\mathbf{k}} = \sqrt{3} q_0 (\mu J_0 - \lambda J_{\mathbf{k}}) , \tag{5.9d}$$

and

$$\Delta_{\mathbf{k}}(E) = E^4 - E^2 (A_{\mathbf{k}}^2 + B_{\mathbf{k}}^2 + 2C_{\mathbf{k}} D_{\mathbf{k}}) + (A_{\mathbf{k}} B_{\mathbf{k}} - C_{\mathbf{k}} D_{\mathbf{k}})^2 . \tag{5.10}$$

Defining

$$\chi_z \equiv z/\Omega_2 , \tag{5.11}$$

and using Eqs. (5.3)—(5.8), we obtain as the only nonvanishing diagonal susceptibilities,

$$\chi_x = \chi_y = \chi_z , \tag{5.12}$$

$$\chi_{xz} = \chi_{yz} = \frac{\chi_z}{1+(\lambda-\mu)\chi_z J_0}[1+3\chi_z\phi^2(\mu-\lambda)J_0] , \tag{5.13}$$

$$\chi_1 = \chi_{xy} = \frac{\chi_z}{1+(\lambda-\mu)\chi_z J_0} , \tag{5.14}$$

where $\phi \equiv \sqrt{3}q_0/z$. We shall see self-consistently that

$$\lim_{\Omega_2} \lim_{z\to 0} \phi = 0 , \tag{5.15}$$

so that in this limit, Eq. (5.13) becomes

$$\chi_{xz} = \chi_{yz} = \frac{\chi_z}{1+(\lambda-\mu)\chi_z J_0} . \tag{5.16}$$

The critical temperature for z ordering is defined by $\chi_z \to \infty$ and we see from Eqs. (5.12), (5.14), and (5.16) that the only divergent diagonal susceptibilities in the $\chi_z \to \infty$ limit are χ_x, χ_y, and χ_z. Reasoning as in Sec. IV we thus conclude that R_j must be chosen from the set $\{S_j^x, S_j^y, S_j^z\}$.

Using Eqs. (5.3) and (5.4) in Eq. (2.8) yields a series of identities while Eqs. (5.5)—(5.18) give [for both the $\eta=+1$ and $\eta=-1$ versions of Eq. (2.8)]

$$a_{\mathbf{k}}^{R,x} = -\frac{K_{\mathbf{k}}^{x,R(-)}}{2} + i(A_{\mathbf{k}}K_{\mathbf{k}}^{y,R(-)} + C_{\mathbf{k}}K_{\mathbf{k}}^{yz,R(-)})I_{\mathbf{k}}^{(1)} + i(A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(C_{\mathbf{k}}K_{\mathbf{k}}^{xz,R(-)} - B_{\mathbf{k}}K_{\mathbf{k}}^{y,R(-)})I_{\mathbf{k}}^{(-1)} , \tag{5.17}$$

$$a_{\mathbf{k}}^{R,y} = -\frac{K_{\mathbf{k}}^{y,R(-)}}{2} - i(A_{\mathbf{k}}K_{\mathbf{k}}^{x,R(-)} + C_{\mathbf{k}}K_{\mathbf{k}}^{xz,R(-)})I_{\mathbf{k}}^{(1)} - i(A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(C_{\mathbf{k}}K_{\mathbf{k}}^{xz,R(-)} - B_{\mathbf{k}}K_{\mathbf{k}}^{x,R(-)})I_{\mathbf{k}}^{(-1)} , \tag{5.18}$$

$$a_{\mathbf{k}}^{R,xz} = -K_{\mathbf{k}}^{xz,R(-)} + i(D_{\mathbf{k}}K_{\mathbf{k}}^{y,R(-)} + B_{\mathbf{k}}K_{\mathbf{k}}^{yz,R(-)})I_{\mathbf{k}}^{(1)} + i(A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(D_{\mathbf{k}}K_{\mathbf{k}}^{y,R(-)} - A_{\mathbf{k}}K_{\mathbf{k}}^{yz,R(-)})I_{\mathbf{k}}^{(-1)} , \tag{5.19}$$

$$a_{\mathbf{k}}^{R,yz} = -\frac{K_{\mathbf{k}}^{yz,R(-)}}{2} - i(D_{\mathbf{k}}K_{\mathbf{k}}^{x,R(-)} + B_{\mathbf{k}}K_{\mathbf{k}}^{xz,R(-)})I_{\mathbf{k}}^{(1)} - i(A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(D_{\mathbf{k}}K_{\mathbf{k}}^{x,R(-)} - A_{\mathbf{k}}K_{\mathbf{k}}^{xz,R(-)})I_{\mathbf{k}}^{(-1)} , \tag{5.20}$$

where

$$I_{\mathbf{k}}^{(n)} \equiv \frac{\omega_{1\mathbf{k}}^n \coth(\beta\omega_{1\mathbf{k}}/2) - \omega_{2\mathbf{k}}^n \coth(\beta\omega_{2\mathbf{k}}/2)}{2(\omega_{1\mathbf{k}}^2 - \omega_{2\mathbf{k}}^2)} , \tag{5.21}$$

and $\omega_{1\mathbf{k}}^2$ and $\omega_{2\mathbf{k}}^2$ are the roots of $\Delta_{\mathbf{k}}(E)$, i.e.,

$$\Delta_{\mathbf{k}}(E) = (E^2 - \omega_{1\mathbf{k}}^2)(E^2 - \omega_{2\mathbf{k}}^2) . \tag{5.22}$$

Using $R_j = S_j^x, S_j^y, S_j^z$ in Eqs. (5.17)—(5.21) gives

$$a_{\mathbf{k}}^{z,x} = a_{\mathbf{k}}^{z,y} = a_{\mathbf{k}}^{z,xz} = a_{\mathbf{k}}^{z,yz} = 0 , \tag{5.23}$$

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

$$a_{\mathbf{k}}^{x,y} = \frac{iz}{2} = -a_{\mathbf{k}}^{y,x} , \tag{5.24}$$

$$a_{\mathbf{k}}^{x,yz} = \frac{i\sqrt{3}q_0}{2} = -a_{\mathbf{k}}^{y,xz} , \tag{5.25}$$

and

$$a_{\mathbf{k}}^{x,x} = (zA_{\mathbf{k}} + \sqrt{3}q_0 C_{\mathbf{k}})I_{\mathbf{k}}^{(1)} + (A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(\sqrt{3}q_0 C_{\mathbf{k}} - zB_{\mathbf{k}})I_{\mathbf{k}}^{(-1)} = a_{\mathbf{k}}^{y,y} , \tag{5.26}$$

$$a_{\mathbf{k}}^{x,xz} = (zD_{\mathbf{k}} + \sqrt{3}q_0 B_{\mathbf{k}})I_{\mathbf{k}}^{(1)} + (A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(zD_{\mathbf{k}} - \sqrt{3}q_0 A_{\mathbf{k}})I_{\mathbf{k}}^{(-1)} = a_{\mathbf{k}}^{y,yz} . \tag{5.27}$$

Summing Eq. (5.24) over all $\mathbf{k}$ gives $q_{xy}=0$, which is true from the symmetry of H. Summing Eq. (5.25) over all $\mathbf{k}$ yields identities. Summing Eqs. (5.26) and (5.27) over all $\mathbf{k}$ and using the $S=1$ identities

$$(S_i^x)^2 = \frac{1}{2}\left[\frac{4}{3} - \frac{Q_i^0}{\sqrt{3}} + Q_i^1\right] , \tag{5.28}$$

$$(S_i^y)^2 = \frac{1}{2}\left[\frac{4}{3} - \frac{Q_i}{\sqrt{3}} - Q_i^1\right] , \tag{5.29}$$

$$S_i^x Q_i^{xz} = \tfrac{1}{2}(S_i^z - iQ_i^{xy}) , \tag{5.30}$$

$$S_i^y Q_i^{yz} = \tfrac{1}{2}(S_i^z + iQ_i^{xy}) , \tag{5.31}$$

we obtain

$$\frac{2}{3} - \frac{q_0}{2\sqrt{3}} = \frac{1}{N}\sum_{\mathbf{k}} (zA_{\mathbf{k}} + \sqrt{3}q_0 C_{\mathbf{k}})I_{\mathbf{k}}^{(1)} + \frac{1}{N}\sum_{\mathbf{k}} (A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(\sqrt{3}q_0 C_{\mathbf{k}} - zB_{\mathbf{k}})I_{\mathbf{k}}^{(-1)} , \tag{5.32}$$

$$\frac{z}{2} = \frac{1}{N}\sum_{\mathbf{k}} (zD_{\mathbf{k}} + \sqrt{3}q_0 B_{\mathbf{k}})I_{\mathbf{k}}^{(1)} + \frac{1}{N}\sum_{\mathbf{k}} (A_{\mathbf{k}}B_{\mathbf{k}} - C_{\mathbf{k}}D_{\mathbf{k}})(zD_{\mathbf{k}} - \sqrt{3}q_0 A_{\mathbf{k}})I_{\mathbf{k}}^{(-1)} . \tag{5.33}$$

Setting $z=\chi_z\Omega_2$ and letting $z,\Omega_2\rightarrow 0$, we obtain from Eq. (5.33)

$$\lim_{\Omega_2,z\rightarrow 0}\phi=0\ , \tag{5.34}$$

thus confirming self-consistently Eq. (5.15). In the $z,\Omega_2\rightarrow 0$ limit, Eq. (5.32) gives, using Eq. (5.34),

$$\frac{2}{3}=\frac{1}{\beta}\frac{1}{N}\sum_{\mathbf{k}}\frac{\chi_z}{1+\lambda\chi_z J_{0\mathbf{k}}} \tag{5.35}$$

thus determining χ_z above the dipolar ordering temperature, T_D. As $T\rightarrow T_D$, $\chi_z\rightarrow\infty$ and we obtain

$$\frac{kT_D}{\lambda J_0}=\frac{\frac{2}{3}}{F(-1)}\ . \tag{5.36}$$

Proceeding as in Sec. IV we also obtain from Eqs. (5.32) and (5.33) the ground-state solutions $z=1$ and $q_0=1/\sqrt{3}$. From Eqs. (4.38) and (5.36) we see that $T_D<T_c$ for $\lambda<\mu$. Thus, for $\lambda<\mu$, q_0 orders with $z=0$ and z cannot order as described in this section since inequality (5.15) cannot be met. The results of this section are therefore restricted to $\lambda>\mu$.

We find the transition temperature T_D depends on the lattice structure and (as in mean-field theory) is independent of the coupling parameter μ. In the special case of $\mu=0$, the Hamiltonian, Eq. (3.2), reduces to that of the isotropic Heisenberg model and our result for T_D, Eq. (5.36), reduces to that of Tahir-Kheli.[18] These results for T_D in the $\mu=0$ case are within 1.5% (bcc), 0.6% (sc), and 0.8% (fcc) of the Padé approximant results.[19]

VI. CONCLUSIONS

We have self-consistently identified the operators whose susceptibilities diverge in the ordered phase as the statistically independent operators in the spin one basis set. Using this approximation to decouple the Green's function hierarchy, we have obtained results which contain no ambiguities and obey all relevant spin-1 identities.

In this particular application to isotropic competing bilinear and biquadratic exchange, our scheme produces results which are valid for *all* values of temperature and couplings. We find the phase space of the system partitions itself into two regions which we label case (A) and case (B). In terms of the ratio of the couplings λ/μ, case (A) is restricted to $\lambda<\mu$ with $q_0\neq 0$, $z=0$ in the ordered region while case (B) is restricted to $\lambda>\mu$ with both z and $q_0\neq 0$ in the ordered region. The topography of the phase space is therefore the same as in the MFA but with phase boundaries that are lattice structure dependent. In addition we have found the ground-state ($T=0$) order parameters for both cases as a function of the ratio of the couplings.

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