Green's-function theory of the spin-1 exchange-interaction model of ferromagnetism

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(Received 17 March 1989)

Double-time Green's functions are used to study dipolar and biaxial ordering in the presence of a uniaxial field for the spin-1 exchange-interaction model of ferromagnetism. The decoupling of the Green's-function equations of motion hierarchy is based upon the identification of the members of the spin-1 basis operator set whose diagonal susceptibilities diverge in the ordered phase as the statistically independent members of the basis set. In contrast to standard double-time Green's-function techniques, the results are unambiguous and the result for the critical temperature in the vanishing field limit compares very favorably with the high-temperature series expansion result.

There have been numerous studies[1-5] of the properties of systems described by the Hamiltonian

$$H=-\frac{\lambda}{2}\sum_{i,j,\alpha}J_{ij}S_i^{\alpha}S_j^{\alpha}-\frac{\mu}{2}\sum_{i,j,\rho}J_{ij}Q_i^{\rho}Q_j^{\rho}, \tag{1}$$

where the $S=1$ operator basis set consists of the three dipolar operators $S_i^{\alpha},\alpha\in\{x,y,z\}$ and the five quadrupolar operators $Q_i^{\rho},\rho\in\{0,1,xy,xz,yz\}$ and

$$\begin{aligned}&Q_i^0=\sqrt{3}[(S_i^z)^2-\tfrac{2}{3}],\quad Q_i^1=(S_i^x)^2-(S_i^y)^2,\\&Q_i^{xy}=S_i^xS_i^y+S_i^yS_i^x,\quad Q_i^{xz}=S_i^xS_i^z+S_i^zS_i^x,\\&Q_i^{yz}=S_i^yS_i^z+S_i^zS_i^y.\end{aligned} \tag{2}$$

The special case $\lambda/\mu=1$, for which H_0 becomes the Schrödinger spin-exchange model,[6-9] has recently recieved attention. Brown[10] has discussed the difficulties involved in obtaining the properties of the Schrödinger model from the $\lambda/\mu\rightarrow 1$ limits of various approximate solutions of H and has proposed a constant-coupling-like approximation to study the $\lambda/\mu=1$ case directly. Typically for approximations of this type, the results depend upon lattice structure only through the coordination number. Also, the effects of applied fields were not considered.

In the present paper, a recently developed double-time Green's-function (DTGF) approximation scheme[4] is used to provide an improved treatment of the Schrödinger spin-exchange model, i.e.,

$$H_0=h-\tfrac{1}{2}\sum_{i,j,\alpha}J_{ij}S_i^{\alpha}S_j^{\alpha}-\tfrac{1}{2}\sum_{i,j,\rho}J_{ij}Q_i^{\rho}Q_j^{\rho}, \tag{3}$$

where the effects of a uniaxial field are included through

$$h=-\Delta\sum_i Q_i^0. \tag{4}$$

The DTGF approximation scheme is based upon the self-consistent identification of the operators whose susceptibilities diverge in the ordered phase as the statistically independent operators of the $S=1$ basis set. The results obtained are unambiguous and satisfy all relevant $S=1$ identities.

The DTGF are defined by

$$\langle\langle A;B\rangle\rangle_{E+i\epsilon}=-i\int_{-\infty}^{\infty}dt\,e^{i(E+i\epsilon)t}\Theta(t)\langle[A(t),B]\rangle,\quad \epsilon\rightarrow 0^+, \tag{5}$$

where $\Theta(t)$ is the step function, $A(t)=e^{iHt}Ae^{-iHt}$, and single angular brackets denote thermal average. The correlation $\langle BA\rangle$ and the DTGF $\langle\langle A;B\rangle\rangle_E$ are related by

$$\langle BA\rangle=\frac{i}{2\pi}\int_{-\infty}^{\infty}dE(e^{\beta E}-1)^{-1}\lim_{\epsilon\rightarrow 0^+}(\langle\langle A;B\rangle\rangle_{E+i\epsilon}-\langle\langle B;A\rangle\rangle_{E+i\epsilon}), \tag{6}$$

where $\beta=1/k_BT$, while the response of $\langle A\rangle$ to a vanishingly small field coupling to A (the diagonal susceptibility) is given by

$$\chi_A=-\lim_{E\rightarrow 0}\langle\langle A;A\rangle\rangle_E. \tag{7}$$

For the Hamiltonians which we are to consider, forming the equations of motion for the DTGF in which A is a member of the $S=1$ basis set leads, as usual, to an infinite, coupled hierarchy of equations. The hierarchy must be decoupled to provide a closed, soluble set of equations. The DTGF to be decoupled are of the form $\langle\langle S_i^{\alpha}Q_j^{\rho};B\rangle\rangle_E$ and $\langle\langle Q_i^{\rho}Q_j^{\rho'};B\rangle\rangle_E$. Recognizing that B is the only operator that appears in every DTGF to be decoupled and assuming that B is statistically independent of the other members of the $S=1$ basis set leads to the symmetric decouplings[4]

$$\begin{aligned}&\langle\langle S_i^{\alpha}Q_j^{\rho};B\rangle\rangle_E=\alpha\langle\langle Q_j^{\rho};B\rangle\rangle_E+q_{\rho}\langle\langle S_i^{\alpha};B\rangle\rangle_E,\\&\langle\langle Q_i^{\rho}Q_j^{\rho'};B\rangle\rangle_E=q_{\rho}\langle\langle Q_j^{\rho'};B\rangle\rangle_E+q_{\rho'}\langle\langle Q_i^{\rho};B\rangle\rangle_E,\end{aligned} \tag{8}$$

where $\alpha=\langle S_i^{\alpha}\rangle$ and $q_{\rho}=\langle Q_i^{\rho}\rangle$.

To study dipolar ordering, we consider the Hamiltonian

$$H_1=-\Omega_1\sum_i S_i^z+H_0 \tag{9}$$

in the limit $\Omega_1 \to 0$. Due to the symmetry of H_1, only z and q_0 do not vanish. Forming the DTGF equations of motion hierarchy, decoupling as described in Eqs. (8), and forming the nonvanishing susceptibilities from Eq. (7) leads to the relations

$$z = \chi_1 \Omega_1 , \tag{10}$$

$$\eta_+ = \chi_+ \Omega_{1+} , \tag{11}$$

$$\eta_- = \chi_- \Omega_{1-} . \tag{12}$$

In Eqs. (10)–(12)

$$\Omega_{1\pm} = \tfrac{1}{2}(\Omega_1 \pm \sqrt{3}\Delta), \quad \eta_\pm = \tfrac{1}{2}(z \pm \sqrt{3} q_0) , \tag{13}$$

and the diagonal susceptibilities χ_1 and $\chi_\pm$ are given by

$$\chi_1 = -\lim_{E\to 0} \sum_{i,j} \langle\langle Q_i^1 ; Q_j^1 \rangle\rangle_E$$
$$= -\lim_{E\to 0} \sum_{i,j} \langle\langle Q_i^{xy} ; Q_j^{xy} \rangle\rangle_E \tag{14}$$

and

$$\chi_\pm = -\lim_{E\to 0} \sum_{i,j} \langle\langle S_i^x \pm Q_i^{xz} ; S_j^x \pm Q_j^{xz} \rangle\rangle_E$$
$$= -\lim_{E\to 0} \sum_{i,j} \langle\langle S_i^y \pm Q_i^{yz} ; S_j^y \pm Q_j^{yz} \rangle\rangle_E . \tag{15}$$

In the z-ordered phase,

$$\lim_{\Omega_1 \to 0} z \neq 0 \tag{16}$$

and, from Eq. (10), χ_1 diverges as $\Omega_1 \to 0$ and Eqs. (11) and (12) become

$$\eta_+ = \sqrt{3}\chi_+ \Delta/2 , \tag{17}$$

$$\eta_- = -\sqrt{3}\chi_- \Delta/2 . \tag{18}$$

We now assume (to be verified self-consistently) a z-ordered phase in which

$$\lim_{\Delta \to 0} \eta_+ \neq 0 \tag{19}$$

and

$$\lim_{\Delta \to 0} \eta_- = 0 . \tag{20}$$

That is, χ_+ diverges as $\Delta \to 0$ but χ_- does not. The divergent susceptibilities are thus χ_1 and χ_+. From Eqs. (14) and (15), Q_i^1, Q_i^{xy}, $S_i^x + Q_i^{xz}$, and $S_i^y + Q_i^{yz}$ are identified as the statistically independent operators of the $S=1$ basis set. Thus, $B = Q_j^1$, Q_j^{xy}, $S_j^x + Q_j^{xz}$, and $S_j^y + Q_j^{yz}$ in the decoupling approximations [Eqs. (8)] and in the resultant DTGF. Requiring that the resultant DTGF satisfy Eq. (6) in common with their exact counterparts, summing over $\mathbf{k}$ and using the $S=1$ identities,

$$\langle (Q_i^1)^2 \rangle = \langle (Q_i^{xy})^2 \rangle = \tfrac{2}{3} + q_0/\sqrt{3} , \tag{21}$$

$$\langle (S_i^x + Q_i^{xz})^2 \rangle = \langle (S_i^y + Q_i^{yz})^2 \rangle = \tfrac{4}{3} - q_0/\sqrt{3} + z , \tag{22}$$

gives

$$\frac{2}{3} + \frac{q_0}{\sqrt{3}} = \frac{z}{N} \sum_{\mathbf{k}} \coth \frac{\beta \omega_{\mathbf{k}}}{2} , \tag{23}$$

$$\frac{4}{3} - \frac{q_0}{\sqrt{3}} + z = \frac{z + \sqrt{3} q_0}{N} \sum_{\mathbf{k}} \coth \frac{\beta \omega_{\mathbf{k},1}^+}{2} , \tag{24}$$

where

$$\omega_{\mathbf{k}} = 2z(J_0 - J_{\mathbf{k}}),$$
$$\omega_{\mathbf{k},1}^+ = \sqrt{3}\Delta + (z + \sqrt{3} q_0)(J_0 - J_{\mathbf{k}}) , \tag{25}$$

and

$$J_{\mathbf{k}} = \frac{1}{N} \sum_{i,j} e^{i\mathbf{k}\cdot\mathbf{r}_{ij}} J_{ij} . \tag{26}$$

Equations (23) and (24) determine z and q_0 in the z-ordered phase in the presence of a field given by Eq. (4). Standard DTGF schemes in which B in Eqs. (8) can be any member of the $S=1$ basis set provide four equations for z and $\sqrt{3} q_0$ and are therefore highly ambiguous. In the $\Delta \to 0$ limit, Eqs. (23) and (24) have the solutions

$$\sqrt{3} q_0 = z \tag{27}$$

and

$$\frac{2}{3} + \frac{z}{3} = \frac{z}{N} \sum_{\mathbf{k}} \coth \frac{\beta \omega_{\mathbf{k}}}{2} . \tag{28}$$

Thus, $\lim_{\Delta \to 0} \eta_+ = z$ and $\lim_{\Delta \to 0} \eta_- = 0$, establishing the consistency of Eqs. (23) and (24) with the assumptions on which they are based, Eqs. (19) and (20). Assuming that $\lim_{\Delta \to 0} \eta_+ = 0$ and $\lim_{\Delta \to 0} \eta_- \neq 0$ in place of Eqs. (19) and (20) produces results which are equivalent to Eqs. (23), (24), (27), and (28) under $z \to -z$ (e.g., a rotation by π about the x axis).

In the ground state, obtained by letting $T \to 0$ in Eqs. (23) and (24), $z = \sqrt{3} q_0 = 1$ for all Δ. The relationship between Δ and the critical temperature for z ordering (denoted by T_0), obtained by letting $z \to 0$ in Eqs. (23) and (24), is shown in Table I for the bcc lattice. Table II compares the results of various approximation schemes for T_0 in the $\Delta \to 0$ limit. The value of T_0 determined by the present treatment is closest to the high-temperature series expansion (HTE) result.

TABLE I. Dependence of the critical temperatures for z ordering (T_0) and q_1 ordering (T_1) on the uniaxial field strength (Δ) for the bcc lattice.

$\sqrt{3}\Delta/J_0$	$k_B T_0/J_0$, $k_B T_1/J_0$
0.0	0.479
0.1	0.562
0.2	0.600
0.3	0.629
0.4	0.653
0.5	0.663
0.6	0.672
0.7	0.679
0.8	0.685
0.9	0.690
1.0	0.694
2.0	0.713
3.0	0.716
⋮	
∞	0.719

TABLE II. Values of $k_B T_0/J_0$ in the $\Delta \to 0$ limit for the fcc lattice as obtained by various approximation schemes.

$k_B T_0/J_0$	Reference	Approximation method
0.515	11	HTE
0.666	12	GF
0.538	10	Constant coupling
0.496	Present	GF

To study biaxial ordering, we consider the Hamiltonian

$$H_2 = -\Omega_2 \sum_i Q_i^1 + H_0$$

in the limit $\Omega_2 \to 0$. Due to the symmetry of H_2, only q_0 and q_1 do not vanish. Proceeding as in the treatment of z ordering leads to the relations

$$q_1 = \chi_z \Omega_2 , \tag{29}$$

$$\mu_+ = \chi_x \Omega_{2+} , \tag{30}$$

$$\mu_- = \chi_y \Omega_{2-} , \tag{31}$$

where

$$\Omega_{2\pm} = \tfrac{1}{2}(\Omega_2 \pm \sqrt{3}\Delta), \quad \mu_\pm = \tfrac{1}{2}(q_1 \pm \sqrt{3} q_0) \tag{32}$$

and

$$\chi_z = -\lim_{E\to 0} \sum_{i,j} \langle\langle S_i^z ; S_j^z \rangle\rangle_E = -\lim_{E\to 0} \sum_{i,j} \langle\langle Q_i^{xy} ; Q_j^{xy} \rangle\rangle_E , \tag{33}$$

$$\chi_x = -\lim_{E\to 0} \sum_{i,j} \langle\langle S_i^x ; S_j^x \rangle\rangle_E = -\lim_{E\to 0} \sum_{i,j} \langle\langle Q_i^{yz} ; Q_j^{yz} \rangle\rangle_E , \tag{34}$$

$$\chi_y = -\lim_{E\to 0} \sum_{i,j} \langle\langle S_i^y ; S_j^y \rangle\rangle_E = -\lim_{E\to 0} \sum_{i,j} \langle\langle Q_i^{xz} ; Q_j^{xz} \rangle\rangle_E . \tag{35}$$

In the q_1-ordered phase,

$$\lim_{\Omega_2 \to 0} q_1 \neq 0 , \tag{36}$$

and, from Eq. (29), χ_z diverges as $\Omega_z \to 0$ and Eqs. (30) and (31) become

$$\mu_+ = \sqrt{3}\chi_x \Delta/2 , \tag{37}$$

$$\mu_- = -\sqrt{3}\chi_y \Delta/2 . \tag{38}$$

We now assume (to be verified self-consistently) a q_1-ordered phase in which

$$\lim_{\Delta \to 0} \mu_+ \neq 0 \tag{39}$$

and

$$\lim_{\Delta \to 0} \mu_- = 0 . \tag{40}$$

That is, χ_x diverges as $\Delta \to 0$ but χ_y does not. The divergent susceptibilities are thus χ_z and χ_x and, from Eqs. (33) and (34), S^z, S^x, Q^{xy}, and Q^{yz} are identified as the statistically independent operators of the $S=1$ basis set. Thus, $B = S_j^z$, S_j^x, Q_j^{xy}, and Q_j^{yz} in the decoupling approximations [Eqs. (8)] and in the resultant DTGF. Requiring that the resultant DTGF satisfy Eq. (6) in common with their exact counterparts, summing over $\mathbf{k}$ and using the $S=1$ identities

$$\langle (S^z)^2 \rangle = \langle (Q^{xy})^2 \rangle = \tfrac{2}{3} + q_0/\sqrt{3} , \tag{41}$$

$$\langle (S^x)^2 \rangle = \langle (Q^{yz})^2 \rangle = \tfrac{2}{3} + (q_1 - \sqrt{3} q_0)/2 , \tag{42}$$

gives

$$\frac{2}{3} + \frac{q_0}{\sqrt{3}} = \frac{q_1}{N} \sum_{\mathbf{k}} \coth \beta q_1 (J_0 - J_{\mathbf{k}}) , \tag{43}$$

$$\frac{4}{3} + q_1 - \frac{q_0}{\sqrt{3}} = \frac{q_1 + \sqrt{3} q_0}{N} \sum_{\mathbf{k}} \coth \frac{\beta \omega_{\mathbf{k},2}^+}{2} , \tag{44}$$

where

$$\omega_{\mathbf{k},2}^+ = \sqrt{3}\Delta + (q_1 + \sqrt{3} q_0)(J_0 - J_{\mathbf{k}}) . \tag{45}$$

Equations (43)–(45) are identical with Eqs. (23)–(25) under the substitution $z \to q_1$. In the $\Delta \to 0$ limit, Eqs. (43) and (44) have the solutions

$$\sqrt{3} q_0 = q_1 , \tag{46}$$

$$\frac{2}{3} + \frac{q_1}{3} = \frac{q_1}{N} \sum_{\mathbf{k}} \coth \beta q_1 (J_0 - J_{\mathbf{k}}) . \tag{47}$$

Thus, $\lim_{\Delta \to 0} \mu_+ = q_1$ and $\lim_{\Delta \to 0} \mu_- = 0$, establishing the consistency of Eqs. (43) and (44) with the assumptions on which they are based, Eqs. (39) and (40). Assumption of a q_1-ordered phase in which $\lim_{\Delta \to 0} \mu_+ = 0$ and $\lim_{\Delta \to 0} \mu_- \neq 0$ in place of Eqs. (43) and (44) produces results which are equivalent to (43) and (44) under $q_1 \to -q_1$ [e.g., a rotation by π about (1,1,0)]. The relationship between Δ and the critical temperature of q_1 ordering (denoted by T_1) is shown in Table I for the bcc lattice. In the ground state, $q_1 = \sqrt{3} q_0 = 1$ for all Δ.

For a rotation by $\pi/2$ about x to a primed coordinate system,

$$q_1' = (q_1 - \sqrt{3} q_0)/2 , \tag{48}$$

$$\sqrt{3} q_0' = -(q_1 + \sqrt{3} q_0) , \tag{49}$$

and, for $\Delta \to 0$, H is invariant. In the primed system, then, using Eq. (46),

$$q_1' = 0, \quad \sqrt{3} q_0' = -2 q_1 , \tag{50}$$

thus extending the mean-field-theory result[1] for $\lambda/\mu \neq 1$ to the $\lambda/\mu = 1$ case.

Green's-function approximation schemes are generally criticized for being based upon unclear decoupling approximations, failing to satisfy relevant operator identities, and providing ambiguous results. The present 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 members of the $S=1$ basis set. The results ob-

tained for dipolar and uniaxial ordering for the Schrödinger model in the presence of a uniaxial field are unambiguous and satisfy all relevant $S=1$ identities. Also, the zero-field dipolar critical temperature for the fcc lattice obtained by this scheme agrees more closely with the high-temperature series expansion result than the critical temperatures obtained by other DTGF, constant-coupling, and mean-field approximation schemes.

This work was supported by Research Corporation Grant No. C-2165.

[1]H. H. Chen and P. M. Levy, Phys. Rev. Lett. **27**, 1383 (1971); Phys. Rev. B **7**, 4267 (1973).
[2]R. Micnas, J. Phys. C **9**, 3307 (1976).
[3]K. G. Chakraborty, Phys. Rev. B **28**, 2827 (1983).
[4]E. B. Brown and P. E. Bloomfield, Phys. Rev. B **31**, 1503 (1985).
[5]E. B. Brown and L. F. Uffer, Phys. Rev. B **31**, 7197 (1985).
[6]C. K. Lai, J. Math. Phys. **15**, 1675 (1974).
[7]B. Sutherland, Phys. Rev. B **12**, 3795 (1975).
[8]H. H. Chen, Y. C. Chen, and F. Lee, Phys. Lett. A **125**, 235 (1987).
[9]Y. C. Chen, H. H. Chen, and F. Lee, Phys. Lett. A **130**, 257 (1988).
[10]H. A. Brown, Phys. Rev. B **31**, 3118 (1985).
[11]G. A. T. Allan and D. D. Betts, Proc. Phys. Soc. London **91**, 941 (1967).
[12]M. Tiwari and R. N. Srivastava, Nuovo Cimento **58B**, 323 (1980).