SU(3) bosons and the spin nematic state on the spin-1 bilinear-biquadratic triangular lattice

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A bond-operator mean-field theory in the SU(3) bosons representation is developed to describe the antiferronematic phase of the spin-1 bilinear-biquadratic model. The quadruplar order of this phase is delicately shown by the calculated static structure factors. The result shows the quadruple operators may be divided into two nonequivalent types. The type exhibiting both the ferroquadruple and antiferroquadruplar long-range orders is reminiscent of the ferrimagnets or the canted antiferromagnets. To address the possible relevance of this unconventional state to the quasi-two-dimensional triangular material $NiGa_2S_4$, we calculated the quasiparticle density of states, the specific heat, and the uniform magnetic susceptibility. We discussed the coincidences and discrepancies between our results and others.

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I. INTRODUCTION

The spin-1 bilinear-biquadratic model (SBBM)

$$H = J_{\varphi}\sum_{\langle ij\rangle}[\cos\varphi \mathbf{S}_i\cdot\mathbf{S}_j + \sin\varphi(\mathbf{S}_i\cdot\mathbf{S}_j)^2] \tag{1}$$

was put forward long time ago,[1–3] where $\mathbf{S}_i$ is the spin-1 operator. In one dimension, the phase diagram was well established,[4–6] but there are still some controversies.[7,8] The phase diagram in two and higher dimensions may be simpler because of suppression of quantum fluctuations. On square lattice, there are two regimes exhibiting different types of spin nematic orders: (1) the ferronematic phase for $-3\pi/4<\varphi<-\pi/2$, (2) the antiferro-nematic phase for $\pi/4<\varphi<\pi/2$. Recently the first regime with ferro-quadruplar long-range order (LRO) attracts much attention due to the fact that the Mott insulating state was realized in a system of bosonic atoms in an optical lattice.[9–11] Here we shall study the second regime by a bond-operator mean-field theory in SU(3) bosons representation. We study the triangular lattice since the unconventional properties of this nematic state, such as the absence of magnetic LRO and the gapless excitation, are quite instructive for explaining recent experimental observations in $NiGa_2S_4$.[12–15] Notice that the antiferronematic phase on the triangular lattice has the same boundaries as the square lattice.[14]

In the framework of frustrated SU(N) model, we expressed the SBBM in terms of SU(3) generators and proposed an associated bond-operator mean-field theory in both bosonic and fermionic representations.[16,17] The theory is a generalization of the widely used Schwinger-boson mean-field theory (SBMFT).[18] The advantage of the theory is that we can use it to study either the ordered or disordered phases. In this paper, we shall use the bosonic theory to study the unconventional orders of the antiferronematic states on the triangular lattice. It will be shown that the ferroquadruplar and antiferroquadruplar LRO's may coexist at low temperature for the quadruple operators, which is reminiscent of the ferrimagnets or the canted antiferromagnets. And the uniform quadruplar moments may keep nonzero at finite temperatures. These two new features enrich our knowledge of the antiferronematic state of this model. To illustrate the relevance of this state to the observations in $NiGa_2S_4$, we also calculate the ground energy, the specific heat, and the uniform magnetic susceptibility. A similar theory with a different scheme had been applied to the ferronematic phase by one of the authors in a previous work.[8]

The paper is organized as follows. In Sec. II we introduce the SU(3) boson representation for spin-1 system, and express the Hamiltonian of Eq. (1) in terms of SU(3) generators. In Sec. III, we present the formalism of the bond-operator mean-field theory in bosonic language. Then in Sec. IV, we work out the mean-field equations and uncover some properties of the antiferronematic phase. In Sec. V, we present discussions of our results.

II. SU(3) BOSONS REPRESENTATION

In SBBM, each site has three states $|m_\alpha\rangle$ with $m_1=-1$, $m_2=0$, and $m_3=+1$, according to the eigenvalues of the z component of spin S^z. We reorganize the three states and introduce three bosonic creation operators

$$b_1^\dagger|0\rangle = \frac{1}{\sqrt{2}}(|m_1\rangle - |m_3\rangle), \tag{2a}$$

$$b_2^\dagger|0\rangle = \frac{i}{\sqrt{2}}(|m_1\rangle + |m_3\rangle), \tag{2b}$$

$$b_3^\dagger|0\rangle = |m_2\rangle, \tag{2c}$$

where $|0\rangle$ is the vacuum state. In terms of b operators, the eight generators of SU(3) group can be expressed by the three boson operators. They are three spin operators

$$S_i^x = -i(b_{i2}^\dagger b_{i3} - b_{i3}^\dagger b_{i2}), \tag{3a}$$

$$S_i^y = -i(b_{i3}^\dagger b_{i1} - b_{i1}^\dagger b_{i3}), \tag{3b}$$

$$S_i^z = -i(b_{i1}^\dagger b_{i2} - b_{i2}^\dagger b_{i1}) \tag{3c}$$

and five quadrupole operators

$$Q_i^{(0)} = (S_i^z)^2 - \frac{2}{3} = \frac{1}{3}(b_{i1}^\dagger b_{i1} + b_{i2}^\dagger b_{i2} - 2b_{i3}^\dagger b_{i3}), \tag{4a}$$

$$Q_i^{(2)} = (S_i^x)^2 - (S_i^y)^2 = -(b_{i1}^\dagger b_{i1} - b_{i2}^\dagger b_{i2}), \tag{4b}$$

$$Q_i^{xy} = S_i^x S_i^y + S_i^y S_i^x = -(b_{i1}^\dagger b_{i2} + b_{i2}^\dagger b_{i1}), \tag{4c}$$

$$Q_i^{yz} = S_i^y S_i^z + S_i^z S_i^y = -(b_{i2}^\dagger b_{i3} + b_{i3}^\dagger b_{i2}), \tag{4d}$$

$$Q_i^{zx} = S_i^z S_i^x + S_i^x S_i^z = -(b_{i3}^\dagger b_{i1} + b_{i1}^\dagger b_{i3}). \tag{4e}$$

In this case the Hamiltonian in Eq. (1) can be expressed in terms of these generators and has a form of the generalized frustrated SU(3) model[17]

$$H = \sum_{\langle ij\rangle} \Upsilon_1(i,j) + \sum_{\langle ij\rangle} \Upsilon_2(i,j) + \sum_i \lambda_i\left(\sum_\mu b_{i\mu}^\dagger b_{i\mu} - 1\right), \tag{5}$$

where the Lagrangian multipliers λ_i are introduced to realize the single occupancy of the bosons at each lattice site, and

$$\Upsilon_1(i,j) = J_1 \sum_{\mu\nu} \mathcal{J}_\nu^\mu(r_i)\mathcal{J}_\mu^\nu(r_j), \tag{6}$$

$$\Upsilon_2(i,j) = -J_2 \sum_{\mu\nu} \mathcal{J}_\nu^\mu(r_i)\mathcal{J}_\nu^\mu(r_j), \tag{7}$$

with $J_1 = J_\varphi \cos\varphi$ and $J_2 = J_\varphi(\cos\varphi - \sin\varphi)$. $\mathcal{J}_\nu^\mu(r_i) = b_{i\mu}^\dagger b_{i\nu}$ are the original generators of the U(3) group. The SU(3) representation is realized by the imposed constraint $\Sigma_\mu b_{i\mu}^\dagger b_{i\mu} - 1 = 0$, which is the source of the Lagrangian term in Eq. (5). The first term in Eq. (5) serves as the permutation operator

$$\sum_{\mu\nu} \mathcal{J}_\nu^\mu(r_i)\mathcal{J}_\mu^\nu(r_j) \equiv P_{ij}, \tag{8}$$

which swaps two quantum states at sites i and j,

$$P_{ij}|i,\mu;j,\nu\rangle = |i,\nu;j,\mu\rangle. \tag{9}$$

The second term in Eq. (5) breaks the SU(3) symmetry.

III. MEAN-FIELD THEORY

A. Decomposition scheme

Now we concentrate on the regime with $J_1 > 0$ and $J_2 < 0$. In the boson representation, we introduce two types of bond operators

$$\Delta_{ij,\mu\nu} = b_{j\mu} b_{i\nu} - b_{j\nu} b_{i\mu}, \quad (\mu < \nu), \tag{10a}$$

$$\Xi_{ij,\mu\nu} = b_{j\mu}^\dagger b_{i\nu} - b_{j\nu}^\dagger b_{i\mu}, \quad (\mu < \nu), \tag{10b}$$

and the four-operator terms in the Hamiltonian (5) can be written as

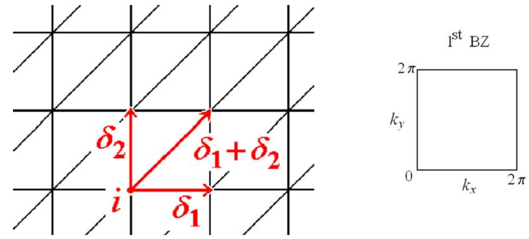

FIG. 1. (Color online) The lattice we used in the calculation, which is topologically equivalent to the triangular lattice given that the interactions along the directions δ_1, δ_2, and $\delta_1+\delta_2$ are equal. On this lattice, the first Brillouin zone is a square with volume $(2\pi)^2$.

$$\Upsilon_1(i,j) = -J_1 \sum_{\mu<\nu} \Delta_{ij,\mu\nu}^\dagger \Delta_{ij,\mu\nu} + J_1, \tag{11a}$$

$$\Upsilon_2(i,j) = -|J_2| \sum_{\mu<\nu} :\Xi_{ij,\mu\nu}^\dagger \Xi_{ij,\mu\nu}: + |J_2|. \tag{11b}$$

Notice the single occupancy constraint $\Sigma_\mu b_{i\mu}^\dagger b_{i\mu} = 1$ is used when the expressions are deduced and the sums do not contain the terms with $\mu = \nu$. Consider that the model is isotropic, one can introduce two real mean-field parameters

$$\Delta = \langle \Delta_{ij,\mu\nu}\rangle = \langle \Delta_{ij,\mu\nu}^\dagger\rangle \ (\mu < \nu), \tag{12a}$$

$$\Xi = \langle \Xi_{ij,\mu\nu}\rangle = \langle \Xi_{ij,\mu\nu}^\dagger\rangle \ (\mu < \nu). \tag{12b}$$

Just like the SBMFT, the assumed mean-field parameters represent the ultra-short-range correlations. The assumption of uniform mean-field parameters may be oversimplified, but we will see that it can capture the dominant features of the antiferronematic phase. When the mean field equations are solved, some physical restrictions should be fulfilled. For instance, the biquadratic term in the SBBM ($N=3$) can be written as

$$(\mathbf{S}_i \cdot \mathbf{S}_j)^2 = -\sum_{\mu<\nu} :\Xi_{ij,\mu\nu}^\dagger \Xi_{ij,\mu\nu}: + 1 \ (\mu,\nu = 1,2,3). \tag{13}$$

Since $(\mathbf{S}_i \cdot \mathbf{S}_j)^2 \geq 0$, one would obtain the restriction

$$\Xi = \langle \Xi_{ij,\mu\nu}\rangle \leq \frac{1}{\sqrt{3}}. \tag{14}$$

Our numerical result shows that this restriction is well satisfied.

B. Mean-field equations

We limit our calculation on the triangular lattice (see Fig. 1). The Hubbard-Stratonovich transformation is performed to decouple the Hamiltonian (5) into a bilinear form

$$H = -J_1 \Delta \sum_{\mu<\nu} \sum_{i,\delta>0} (\Delta_{i,i+\delta;\mu\nu} + \Delta_{i,i+\delta;\mu\nu}^\dagger) - |J_2| \Xi \sum_{\mu<\nu} \sum_{i,\delta} (\Xi_{i,i+\delta;\mu\nu} + \Xi_{i,i+\delta;\mu\nu}^\dagger) + \frac{z}{2} N_\Lambda N (J_1 \Delta^2 + |J_2| \Xi^2) + \lambda \sum_i \sum_\mu b_{i\mu}^\dagger b_{i\mu} - \lambda N_\Lambda, \tag{15}$$

where $\Sigma_{\delta>0}$ means summation over the nearest neighbours in the positive directions of a given site, N_Λ is the total number of lattice sites, z is the coordinate number of the lattice, e.g., $z=6$ for the triangular lattice.

After performing the Fourier transform and introducing the Nambu spinor in the momentum space

$$\Phi_{\mathbf{k}}^\dagger = (b_{\mathbf{k},1}^\dagger, b_{\mathbf{k},2}^\dagger, b_{\mathbf{k},3}^\dagger, b_{-\mathbf{k},1}, b_{-\mathbf{k},2}, b_{-\mathbf{k},3}) \tag{16}$$

one can arrive at the mean-field Hamiltonian in a compact form

$$H = \frac{1}{2}\sum_{\mathbf{k}} \Phi_{\mathbf{k}}^\dagger M_{\mathbf{k}} \Phi_{\mathbf{k}} + \varepsilon_0, \tag{17}$$

where

$$M_{\mathbf{k}} = \lambda \sigma^0 \otimes A_0 + i\Delta_{\mathbf{k}} \sigma^x \otimes A_1 + i\Xi_{\mathbf{k}} \sigma^0 \otimes A_1, \tag{18a}$$

$$A_0 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad A_1 = \begin{pmatrix} 0 & -1 & -1 \\ 1 & 0 & -1 \\ 1 & 1 & 0 \end{pmatrix}, \tag{18b}$$

$$\Delta_{\mathbf{k}} = 2J_1 \Delta\, \eta_{\mathbf{k}}, \quad \Xi_{\mathbf{k}} = 2|J_2| \Xi\, \eta_{\mathbf{k}}, \tag{18c}$$

$$\eta_{\mathbf{k}} = \sum_{\delta>0} \sin k_\delta = \sin k_1 + \sin k_2 + \sin(k_1 + k_2), \tag{18d}$$

$$\varepsilon_0 = \frac{3}{2} z N_\Lambda (J_1 \Delta^2 + |J_2| \Xi^2) - \frac{5}{2} \lambda N_\Lambda. \tag{18e}$$

By diagonalizing the Hamiltonian, we get three spectra

$$\omega_1 = \lambda, \tag{19a}$$

$$\omega_2(\mathbf{k}) = \sqrt{(\lambda - \sqrt{3}\Xi_{\mathbf{k}})^2 - (\sqrt{3}\Delta_{\mathbf{k}})^2}, \tag{19b}$$

$$\omega_3(\mathbf{k}) = \sqrt{(\lambda + \sqrt{3}\Xi_{\mathbf{k}})^2 - (\sqrt{3}\Delta_{\mathbf{k}})^2}. \tag{19c}$$

Notice that the two spectra $\omega_3(\mathbf{k})$ and $\omega_2(\mathbf{k})$ has a relation of $\omega_3(-\mathbf{k}) = \omega_2(\mathbf{k})$. By optimization of the total free energy

$$F = \varepsilon_0 - \frac{1}{\beta}\sum_{\mathbf{k},\mu} \ln\{n_B(\omega_\mu)[n_B(\omega_\mu) + 1]\}, \tag{20}$$

where $n_B(\omega_\mu)$ is the Boltzmann distribution function, three mean-field equations are established

$$2 - n_B(\lambda) = \int \frac{d^2k}{(2\pi)^2} \frac{1 - \tilde{\Xi}\eta_{\mathbf{k}}}{\tilde{\omega}_2(\mathbf{k})} \coth \frac{\beta \tilde{\omega}_2(\mathbf{k})}{2}, \tag{21a}$$

$$\Delta = \frac{1}{3\sqrt{3}} \int \frac{d^2k}{(2\pi)^2} \frac{\tilde{\Delta}\eta_{\mathbf{k}}^2}{\tilde{\omega}_2(\mathbf{k})} \coth \frac{\beta \tilde{\omega}_2(\mathbf{k})}{2}, \tag{21b}$$

$$\Xi = \frac{1}{3\sqrt{3}} \int \frac{d^2k}{(2\pi)^2} \frac{(1 - \tilde{\Xi}\eta_{\mathbf{k}})\eta_{\mathbf{k}}}{\tilde{\omega}_2(\mathbf{k})} \coth \frac{\beta \tilde{\omega}_2(\mathbf{k})}{2}, \tag{21c}$$

in which we have introduced three dimensionless quantities for convenience of calculation

$$\tilde{\omega}_\mu(\mathbf{k}) = \frac{\omega_\mu(\mathbf{k})}{\lambda}, \tag{22a}$$

$$\tilde{\Delta} = \frac{2\sqrt{3}J_1\Delta}{\lambda}, \tag{22b}$$

$$\tilde{\Xi} = \frac{2\sqrt{3}|J_2|\Xi}{\lambda}. \tag{22c}$$

$\beta = 1/k_B T$, and k_B is the Boltzmann constant. There are generally three branches of valid solutions: (i) nonzero solution $\Delta \neq 0$ and $\Xi \neq 0$; (ii) zero solution $\Delta = 0$ and $\Xi \neq 0$; (iii) zero solution $\Delta \neq 0$ and $\Xi = 0$. The one with the lowest energy is picked out as the physically realized state. At zero temperature, the ground energy per site has a simple form

$$\frac{E_0}{N_\Lambda} = -\frac{3}{2} z (J_1 \Delta^2 + |J_2| \Xi^2). \tag{23}$$

C. Green's function and susceptibility

In order to calculate the susceptibility we introduce the Matsubara Green's function in the form of a 6×6 matrix

$$G(\mathbf{k}, \tau) = -\langle T_\tau \Phi_{\mathbf{k}}(\tau) \Phi_{\mathbf{k}}^\dagger(0) \rangle = \frac{1}{\beta}\sum_n G(\mathbf{k}, i\omega_n) e^{-i\omega_n \tau}. \tag{24}$$

The bosonic Matsubara Green's function $G(\mathbf{k}, i\omega_n)$ is generally worked out as

$$G(\mathbf{k}, i\omega_n) = (i\omega_n \sigma_z \otimes A_0 - M_{\mathbf{k}})^{-1}, \tag{25}$$

where $\omega_n = 2n\pi/\beta$. The three spectra (19) can also be read out from the poles of the Green's function.

As we shall study spin order as well as the nematic order in the system, we define two types of correlation functions in Matsubara formalism. The first type is the spin-spin correlation. Due to rotational invariance, we need only to consider the imaginary-time spin-spin correlation for S^z,

$$\chi_{S^z}(\mathbf{q}, \tau) = \langle T_\tau S^z(\mathbf{q}, \tau) S^z(-\mathbf{q}, 0) \rangle. \tag{26}$$

Its Fourier transform is given by

$$\chi_{S^z}(\mathbf{q}, i\omega_n) = \int_0^\beta d\tau e^{i\omega_n \tau} \chi_{S^z}(\mathbf{q}, \tau). \tag{27}$$

The second type is the imaginary-time quadrupole-quadrupole correlation and its Fourier transform defined for the quadrupole operators Q's in Eq. (4) is given by

$$\chi_Q(\mathbf{q}, \tau) = \langle T_\tau Q(\mathbf{q}, \tau) Q(-\mathbf{q}, 0) \rangle, \tag{28}$$

$$\chi_Q(\mathbf{q}, i\omega_n) = \int_0^\beta d\tau e^{i\omega_n \tau} \chi_Q(\mathbf{q}, \tau), \tag{29}$$

$$Q \in \{Q^{(0)}, Q^{(2)}, Q^{xy}, Q^{yz}, Q^{zx}\}. \tag{30}$$

Due to the rotational invariance, here we only present two of them, $\chi_{Q^{(2)}}$ and $\chi_{Q^{xy}}$ ($\chi_{Q^{(0)}}$ is equivalent to $\chi_{Q^{(2)}}$, $\chi_{Q^{yz}}$ and $\chi_{Q^{zx}}$

are equivalent to $\chi_{Q^{xy}}$). According to the single-mode approximation theory,[19] χ_{S^z} is related to the spin order by the single mode $S^z(\mathbf{q})|0\rangle$ with spin density wave

$$S^z(\mathbf{q}) = \sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} S_i^{\alpha}, \tag{31}$$

while χ_Q is related to the nematic order by the single mode $Q(q)|0\rangle$ with quadrupole density wave

$$Q(\mathbf{q}) = \sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} Q_i. \tag{32}$$

The expressions of the susceptibilities at zero temperature can be found in the Appendix.

IV. LONG-RANGE SPIN NEMATIC ORDER ON TRIANGULAR LATTICE

The nonzero solution of the mean field parameters satisfies

$$\tilde{\Delta} + \tilde{\Xi} = \frac{2}{3\sqrt{3}}, \tag{33}$$

at zero temperature and on the triangular lattice. With this relation, the spectrum $\omega_2(\mathbf{k})$ becomes gapless at the nodal point

$$\mathbf{k}^* = (k_x^*, k_y^*) = \left(\frac{\pi}{3}, \frac{\pi}{3}\right), \tag{34}$$

where the boson condensation occurs. A similar nodal structure of the spectrum has been observed by Tsunetsugu *et al.*,[13] but notice that different types of quasiparticles are introduced in their work. As temperature becomes nonzero, the spectrum $\omega_2(\mathbf{k})$ will open a gap and thus no condensation occurs. When the condensation occurs, we should parse the condensation terms and rewrite the equations as

$$\rho_0 = 2 - \int \frac{d^2k}{(2\pi)^2} \frac{1 - \tilde{\Xi}\eta_{\mathbf{k}}}{\tilde{\omega}_2(\mathbf{k})}, \tag{35a}$$

$$\Delta = \frac{1}{2}\rho_0 + \frac{1}{3\sqrt{3}} \int \frac{d^2k}{(2\pi)^2} \frac{\tilde{\Delta}\eta_{\mathbf{k}}^2}{\tilde{\omega}_2(\mathbf{k})}, \tag{35b}$$

$$\Xi = \frac{1}{2}\rho_0 + \frac{1}{3\sqrt{3}} \int \frac{d^2k}{(2\pi)^2} \frac{(1 - \tilde{\Xi}\eta_{\mathbf{k}})\eta_{\mathbf{k}}}{\tilde{\omega}_2(\mathbf{k})}, \tag{35c}$$

where the condensation density is (see numerical result in Fig. 2)

$$\rho_0 = \left[\frac{2n_B[\tilde{\omega}_2(\mathbf{k}^*)] + 1}{N_\Lambda \tilde{\omega}_2(\mathbf{k}^*)}\right] \frac{3\sqrt{3}\tilde{\Delta}}{2}. \tag{36}$$

From the ground energies shown in Fig. 3, we see the nonzero solution is the optimized one in the range $\pi/4 < \varphi < \pi/2$. At the SU(3) point of $\varphi = \pi/4$, the zero solution with $\Delta \neq 0$ and $\Xi = 0$ is degenerate with the nonzero solution. At the point of $\varphi = \pi/2$, the zero solution with $\Delta = 0$ and $\Xi \neq 0$ is degenerate with the nonzero solution. This reflects the fact that the two points are highly symmetric points. In the range $\pi/4 < \varphi < \pi/2$, the condensate is nonzero. So what is the physical effect of the quasiparticle condensation? By probing the possible orders in the system, we find the condensation leads to the nematic LRO while spin moments vanish, i.e., the nematic state is nonmagnetic. This conclusion is drawn from the static spin and quadrupole structure factors shown in Fig. 4. (Please refer to the expressions listed in the Appendix.)

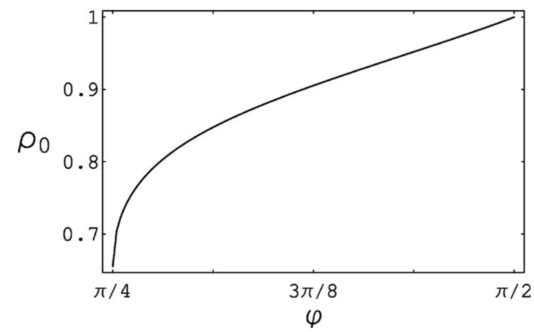

FIG. 2. The condensation density ρ_0 at zero temperature.

Our spin structure factor does not agree with the one by Tsunetsugu *et al.* in that we can not produce the cone singularity behavior.[13] According to the definition of spin structure factor (26), one should have the relation,

$$\chi_{S^z}(\mathbf{q} + \mathbf{G}, \tau = 0^+) = \chi_{S^z}(\mathbf{q}, \tau = 0^+), \tag{37}$$

where $\mathbf{G}$ is the reciprocal lattice vector. Equation (37) means the spin structure factor in the first Brillouin zone is replicative in the whole momentum space.

The static quadrupole structure factor $\chi_{Q^{(2)}}(\mathbf{q}, \tau = 0^+)$ and $\chi_{Q^{xy}}(\mathbf{q}, \tau = 0^+)$ show sharp divergent peaks at the two points $\mathbf{q}^* = \pm 2\mathbf{k}^* = \pm\left(\frac{2\pi}{3}, \frac{2\pi}{3}\right)$ indicating the existence of antiferroquadrupolar LRO. While the static spin structure factor $\chi_{S^z}(\mathbf{q}, \tau = 0^+)$ just shows two small humps at $\mathbf{q}^* = \pm\left(\frac{2\pi}{3}, \frac{2\pi}{3}\right)$. Surprisingly $\chi_{Q^{xy}}(\mathbf{q}, \tau = 0^+)$ also exhibits a divergent peak at

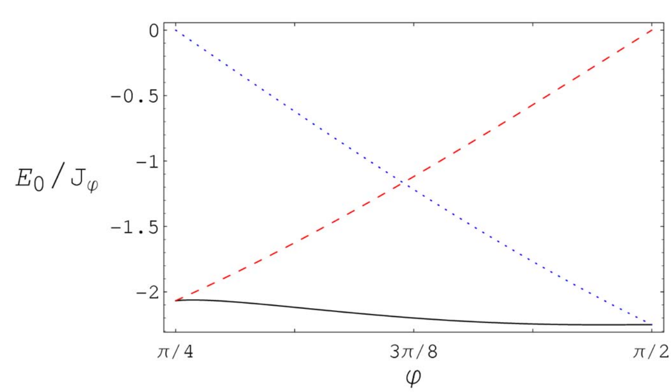

FIG. 3. (Color online) The ground energies. The solid line is the nonzero solution $\Delta \neq 0, \Xi \neq 0$. The dashed line is the zero solution $\Delta \neq 0, \Xi = 0$. The dotted line is the zero solution $\Delta = 0, \Xi \neq 0$. This figure shows the nonzero solution is the optimized one.

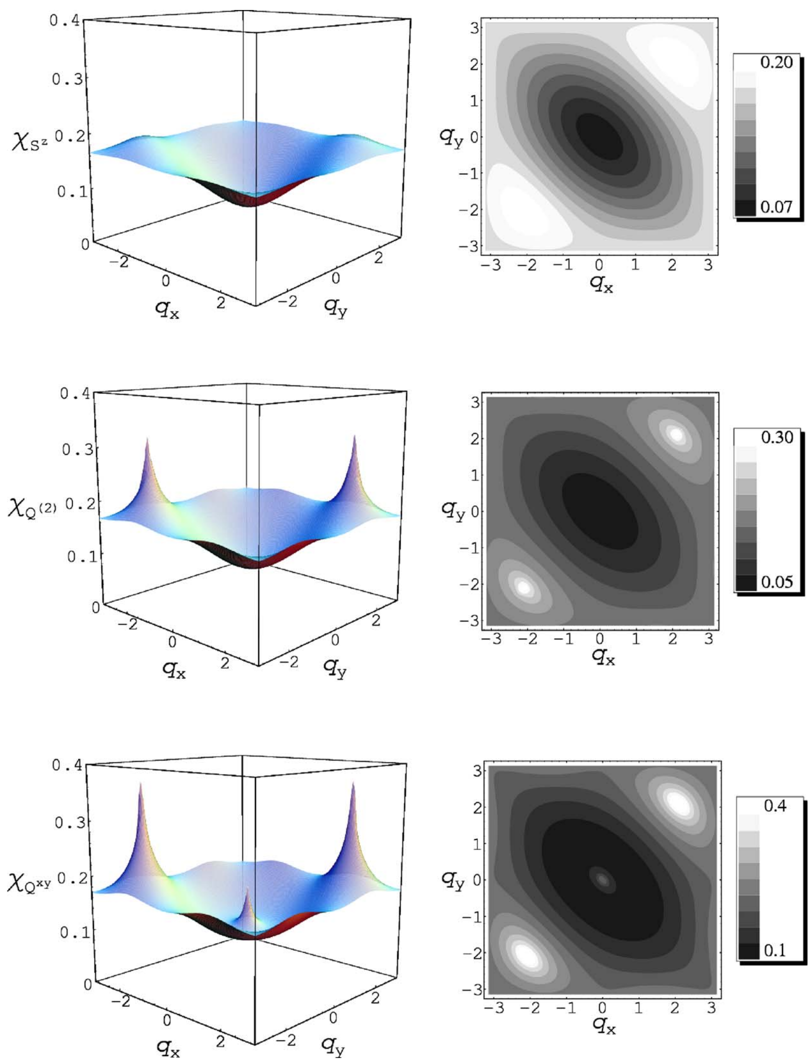

FIG. 4. (Color online) Static structure factors χ_{S^z}, $\chi_{Q^{(2)}}$, and $\chi_{Q^{xy}}$ at a sampled point $\varphi=0.863938$. The images at other point in the regime $\frac{\pi}{4}<\varphi<\frac{\pi}{2}$ are qualitatively the same.

the point $\mathbf{q}^0=(0,0)$. Analytically, at the condensate points, the divergent terms of $\chi_{Q^{xy}}$ are parsed out as

$$\chi_{Q^{xy}}^{\rho_0}(\mathbf{q}^0)=\frac{1}{9}\rho_0^2 N_\Lambda, \tag{38a}$$

$$\chi_{Q^{xy}}^{\rho_0}(\mathbf{q}^*)=\frac{2}{9}\rho_0^2 N_\Lambda. \tag{38b}$$

The ratio of the weights of the ferroquadrupole and antiferroquadrupole divergent peaks is

$$r=\frac{\chi_{Q^{xy}}^{\rho_0}(\mathbf{q}^0)}{\chi_{Q^{xy}}^{\rho_0}(\mathbf{q}^*)}=\frac{1}{2}. \tag{39}$$

$\chi_{Q^{xy}}^{\rho_0}(\mathbf{q}^0)$ and $\chi_{Q^{xy}}^{\rho_0}(\mathbf{q}^*)$ are proportional to the number of the lattice site N_Λ, which indicates that the ferroquadrupole and antiferroquadrupole LRO coexist for Q_i^{xy}. To understand the difference of the two quadruple operators $Q_i^{(2)}$ and Q_i^{xy}, let us write down the eigenstates of $Q_i^{(2)}$ [see also Eq. (2)],

$$|Q_i^{(2)}=-1\rangle=b_{i1}^\dagger|0\rangle, \tag{40a}$$

$$|Q_i^{(2)}=1\rangle=b_{i2}^\dagger|0\rangle, \tag{40b}$$

$$|Q_i^{(2)}=0\rangle=b_{i3}^\dagger|0\rangle. \tag{40c}$$

On the triangular lattice, the LRO arrangement of the nonzero quadrupolar moments $\langle Q_i^{(2)}\rangle$ is a $2\pi/3$ structure as we revealed above. However they are not the eigenstates of Q_i^{xy}, instead

$$|Q_i^{(2)}=-1\rangle=\frac{1}{\sqrt{2}}(|Q_i^{xy}=1\rangle-|Q_i^{xy}=-1\rangle), \tag{41a}$$

$$|Q_i^{(2)}=1\rangle=\frac{-1}{\sqrt{2}}(|Q_i^{xy}=1\rangle+|Q_i^{xy}=-1\rangle). \tag{41b}$$

So the antiferroquadrupolar moment of $Q_i^{(2)}$ means the existence of both the ferroquadrupole and antiferroquadrupolar moments of Q_i^{xy}. This phenomenon is reminiscent of the ferrimagnets or the canted antiferromagnets, which exhibit both ferromagnetic and antiferromagnetic orders. This result reflects the fact that the eight generators, Eqs. (3) and (4) can be divided into three nonequivalent categories due to the SU(3) symmetry breaking of the model. Nevertheless the ferroquadrupolar order may also be an artifact due to the oversimplified assumption of mean-field parameters (12).

The static quadrupole structure factor $\chi_{Q^{(2)}}(\mathbf{q},\tau=0^{+})$ has a minimum value at $\mathbf{q}=0$, which means the ferroquadruplar fluctuation is very weak in the antiferronematic phase. But unfortunately in our mean-field scheme, we cannot get zero result in the whole range of the phase.

Now we address to the two terminals of the range $\pi/4 \leqslant \varphi \leqslant \pi/2$. At the SU(3) point $\varphi=\pi/4$, the solution is highly degenerate, i.e., the zero solution $(\Delta\neq 0, \Xi=0)$ and the nonzero solutions $(\Delta\neq 0, 0<\Xi<0.33)$ are degenerate (please see Fig. 3). The zero solution leads to the divergence of the static spin structure factor at $\mathbf{q}^{*}=\pm(\frac{2\pi}{3},\frac{2\pi}{3})$, which indicates the SU(3) point is a dividing point for the antiferronematic and antiferromagnetic phases.[1,2] At $\varphi=\pi/2$ where the Hamiltonian becomes $H=\Sigma_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j)^2$, the solution is also highly degenerate, i.e., the zero solution $(\Delta=0,\Xi\neq 0)$ and the nonzero solutions $(0<\Delta<0.50,\Xi\neq 0)$ are degenerate (please see Fig. 3). For the zero solution, all of the static structure factors vanish and the system is free of both spin and quadrupole moments, which means the system is totally disordered. Given that this point is a dividing point for the antiferronematic and ferromagnetic phases, our theory may miss the ferromagnetic state at this terminal.

The Matsubara formalism facilitates the evaluation of the expectation values at finite temperatures $(\pi/4<\varphi<\pi/2)$,

$$\langle b_{i1}^{\dagger}b_{i1}\rangle = \langle b_{i2}^{\dagger}b_{i2}\rangle = \frac{1}{3}, \tag{42a}$$

$$\langle b_{i1}^{\dagger}b_{i2}\rangle = \langle b_{i2}^{\dagger}b_{i1}\rangle = \frac{1}{6} - \frac{1}{2}n_B(\lambda). \tag{42b}$$

Thus we obtain

$$\langle S_i^{z}\rangle = -i(\langle b_{i1}^{\dagger}b_{i2}\rangle - \langle b_{i2}^{\dagger}b_{i1}\rangle) = 0, \tag{43a}$$

$$\langle Q_i^{(2)}\rangle = -(\langle b_{i1}^{\dagger}b_{i1}\rangle - \langle b_{i2}^{\dagger}b_{i2}\rangle) = 0, \tag{43b}$$

$$\langle Q_i^{xy}\rangle = -(\langle b_{i1}^{\dagger}b_{i2}\rangle + \langle b_{i2}^{\dagger}b_{i1}\rangle) = -\left[\frac{1}{3} - n_B(\lambda)\right]. \tag{43c}$$

The uniform quadrupole moment $\langle Q_i^{xy}\rangle$ keeps nonzero at zero tempurature, which indicates the existence of quadrupole long-range correlation.

The density of states (DOS) is defined as

$$D(E) = \frac{1}{N_\Lambda}\sum_{\mathbf{k},\mu}\delta[E-\omega_\mu(\mathbf{k})]. \tag{44}$$

Since there exist a flat band $\omega_1=\lambda$ for the quasi-particles, $D(E)$ always has a divergent peak at $E=\lambda$. We find that DOS rises linearly in E from zero in the range of $\pi/4\leqslant\varphi<\pi/2$,

$$D(E) \sim a(\varphi)E + O(E^2), \tag{45}$$

because the gapless spectrum $\omega_2(\mathbf{k})$ exhibits a node at $\mathbf{k}^{*}=(\frac{\pi}{3},\frac{\pi}{3})$. By the DOS in Eq. (45), the low temperature specific heat is shown to exhibit the law of T^2,

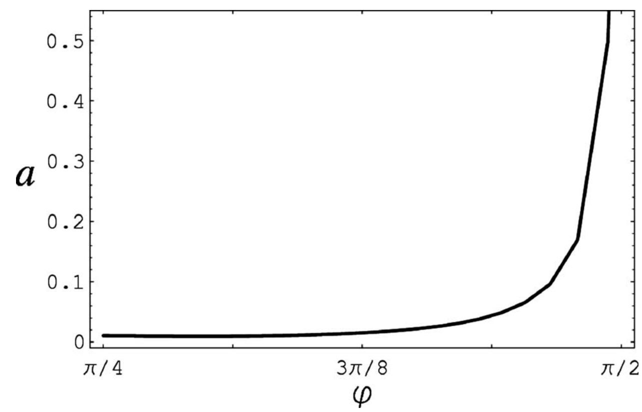

FIG. 5. The coefficient a in Eq. (45).

$$C_V/N_\Lambda = k_B\int dED(E)\left(\frac{E}{2k_BT}\right)^2\left(\sinh\frac{E}{2k_BT}\right)^{-2} \sim 6\zeta(3)a(\varphi)k_B^3T^2. \tag{46}$$

The coefficient $a(\varphi)$ is plotted in Fig. 5. The above results agree qualitatively with the ones by Tsunetsugu *et al.*[13] While at the terminal $\varphi=\pi/2$, the node of the spectra disappears and the DOS has the form $D(E)\sim b+cE$ with $b\neq 0$, then one would get leading term $C_V/N_\Lambda\sim\frac{\pi^2}{3}bk_BT$.

It is noteworthy that, at nonzero temperatures, the spectrum $\omega_2(k)$ is gapful and the DOS always has $D(E)\sim b+cE$. But at very low temperatures, b is quite small and the power law in Eq. (46) can be satisfied asymptotically.

The uniform magnetic susceptibility at zero temperature is obtained by Kramers-Kronig relation[19]

$$\chi_M = \lim_{q\to 0}\frac{1}{\pi}\int_0^{\infty} d\omega\frac{\mathrm{Im}\,\chi_{S^z}(\mathbf{q},\omega)}{\omega} \tag{47}$$

or by analytic continuation[20]

$$\chi_M = \lim_{\mathbf{q}\to 0}\lim_{i\omega_n\to 0}\chi_{S^z}(\mathbf{q},i\omega_n). \tag{48}$$

They give the same result. At zero temperature, χ_M versus φ is illustrated in Fig. 6. χ_M reaches the maximal value at the SU(3) point $\varphi=\pi/4$, while it approaches zero at the end

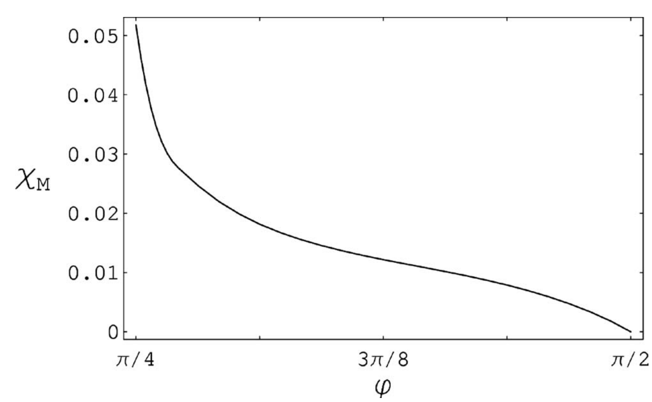

FIG. 6. The magnetic susceptibility χ_M at zero temperature.

point $\varphi=\pi/2$, where the system is insusceptible to the external inspiration.

V. DISCUSSION

Recently the insulating antiferromagnet $NiGa_2S_4$ arouses much attention.[12] The spin disorder observed in the experiment suggest that it may be a realization of the conceptual spin liquid that has long been explored over the past decades. This chalcogenide has a stacked triangular lattice with weak interactions between layers. Strong Hund's coupling in $Ni^{2+}(t_{2g}^6 e_g^2)$ leads to the magnetism with spin $S=1$. Magnetic neutron scattering shows absence of conventional magnetic order and excludes the possibility of bulk spin glass freezing at low temperatures. Its specific heat shows low temperature power law, $C_V \sim T^2$, indicating gapless excitations and linearly dispersive modes in two dimensions. No divergence was observed for the magnetic susceptibility with the temperature decreasing down to 0.35 K. These features can be produced by the nematic state as we studied above, as has been pointed out in Ref. 13. However, the incommensurate short-range order observed in the experiment still remains untouched. It was also proposed by Läuchli *et al.* that the ferronematic phase may be a more plausible candidate.[14,15]

Our bond-operator mean-field theory has the same origin as the SBMFT and is superior to the molecular-field approximation (MFA) because the MFA starts from a prescribed ferro-or antiferro-order and produces the same result regardless of the dimensionality,[2,21] while our theory has no bias on the order or disorder of the ground state in advance. In two dimensions, we got a gapless nematic phase with quadruplar LRO as we illustrated above for the triangular lattice. In one dimension, we got a gapped result. Nevertheless, any mean-field theory cannot be conclusive in its own right, thus other methods for the same problem are demanded for a corroboration. Unlike the spin wave theory, the bond-operator mean-field theory used in this paper does not prescribe an ordered state in advance. It has the same origin as SBMFT, except the species of bosons is altered from 2 to 3. The antiferroquadruplar LRO emerges as a consequence of the condensation of the SU(3) bosons. Our results also show the quadruple operators may be divided into two types. They have different LRO patterns and should be considered differently in a spin wave theory (i.e., for $Q^{(0)}$ and $Q^{(2)}$, one need only to consider their antiferro-orders; while for Q^{xy}, Q^{yz}, and Q^{zx}, one should consider their ferro-orders and antiferro-orders at the same time. The coexistence of ferroquadruplar and antiferroquadruplar LRO's reveals that the quadruple operators cannot be considered as the analogues of spin operators in the magnetic LRO phenomena. As a merit of bosonic language, we also expect that this theory gives good estimation of ground energy values (see Fig. 3), similar to the SBMFT.[22,23] To see how this theory describes the antiferromagnetic phase ($-\pi/4<\varphi<\pi/4$) of the SBBM is also desirable, which will be considered in our future work.

In summary, a SU(3) bosons representation is introduced and the associated bond-operator mean-field theory is established to describe the antiferronematic phase of SBBM. It is revealed delicately that this nematic state may exhibit both the ferroquadruplar and antiferroquadruplar LRO's, which is reminiscent of the ferrimagnets or the canted antiferromagnets. Possible relevance of this unconventional state to the quasi-two-dimensional triangular material $NiGa_2S_4$ is addressed by the calculated specific heat and magnetic susceptibility.

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APPENDIX: STATIC SPIN AND QUADRUPOLE STRUCTURE FACTORS

The static spin and quadrupole structure factors at zero temperature are worked out as

$$\chi_{S^z}(\mathbf{q},\tau=0^+)=\int\frac{d^2k}{(2\pi)^2}[A_2(\mathbf{k})+A_3(\mathbf{k})+3A_2(\mathbf{k})B_2(\mathbf{k}+\mathbf{q}) + 3A_3(\mathbf{k})B_3(\mathbf{k}+\mathbf{q})+3C_2(\mathbf{k})C_2(\mathbf{k}+\mathbf{q}) + 3C_3(\mathbf{k})C_3(\mathbf{k}+\mathbf{q})], \tag{A1a}$$

$$\chi_{Q^{(2)}}(\mathbf{q},\tau=0^+)=\int\frac{d^2k}{(2\pi)^2}[A_2(\mathbf{k})+A_3(\mathbf{k})+3A_2(\mathbf{k})B_3(\mathbf{k}+\mathbf{q}) + 3A_3(\mathbf{k})B_2(\mathbf{k}+\mathbf{q})+3C_2(\mathbf{k})C_3(\mathbf{k}+\mathbf{q}) + 3C_3(\mathbf{k})C_2(\mathbf{k}+\mathbf{q})], \tag{A1b}$$

$$\chi_{Q^{xy}}(\mathbf{q},\tau=0^+)=\int\frac{d^2k}{(2\pi)^2}\left[\frac{1}{3}A_2(\mathbf{k})+\frac{1}{3}A_3(\mathbf{k})+A_2(\mathbf{k})B_2(\mathbf{k} + \mathbf{q})+A_3(\mathbf{k})B_3(\mathbf{k}+\mathbf{q})+4A_2(\mathbf{k})B_3(\mathbf{k}+\mathbf{q}) + 4A_3(\mathbf{k})B_2(\mathbf{k}+\mathbf{q})+4C_2(\mathbf{k})C_3(\mathbf{k}+\mathbf{q}) + 4C_3(\mathbf{k})C_2(\mathbf{k}+\mathbf{q})-C_2(\mathbf{k})C_2(\mathbf{k}+\mathbf{q}) - C_3(\mathbf{k})C_3(\mathbf{k}+\mathbf{q})\right], \tag{A1c}$$

where the abbreviated notations are

$$A_2(\mathbf{k})=\frac{1}{6}\left[1+\frac{\lambda-\sqrt{3}\Xi_{\mathbf{k}}}{\omega_2(\mathbf{k})}\right], \tag{A2a}$$

$$A_3(\mathbf{k})=\frac{1}{6}\left[1+\frac{\lambda+\sqrt{3}\Xi_{\mathbf{k}}}{\omega_3(\mathbf{k})}\right]=A_2(-\mathbf{k}), \tag{A2b}$$

$$B_2(\mathbf{k})=\frac{1}{6}\left[1-\frac{\lambda+\sqrt{3}\Xi_{\mathbf{k}}}{\omega_2(\mathbf{k})}\right], \tag{A2c}$$

$$B_3(\mathbf{k})=\frac{1}{6}\left[1-\frac{\lambda+\sqrt{3}\Xi_{\mathbf{k}}}{\omega_3(\mathbf{k})}\right]=B_2(-\mathbf{k}), \tag{A2d}$$

$$C_2(\mathbf{k})=\frac{1}{6}\frac{\sqrt{3}\Delta_{\mathbf{k}}}{\omega_2(\mathbf{k})}, \tag{A2e}$$

$$C_3(\mathbf{k}) = \frac{1}{6}\frac{\sqrt{3}\Delta_{\mathbf{k}}}{\omega_3(\mathbf{k})} = -C_2(-\mathbf{k}). \tag{A2f}$$

$\Xi_{\mathbf{k}}$ and $\Delta_{\mathbf{k}}$ can be found in Eq. (18c). With these expressions, one can easily judge that $\chi_{Q^{(2)}}(\mathbf{q},0^+)$ is divergent at $\mathbf{q}^* = \pm(\frac{2\pi}{3},\frac{2\pi}{3})$ and $\chi_{Q^{xy}}(\mathbf{q},0^+)$ at $\mathbf{q}^0=(0,0)$ and $\mathbf{q}^*=\pm(\frac{2\pi}{3},\frac{2\pi}{3})$. The divergences indicate the quadrupolar order as discussed in the text.

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