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Incommensurate phase of a triangular frustrated Heisenberg model studied via Schwinger-boson mean-field theory

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Abstract
We study a triangular frustrated antiferromagnetic Heisenberg model with nearest-neighbor interactions J_1 and third-nearest-neighbor interactions J_3 by means of Schwinger-boson mean-field theory. By setting an antiferromagnetic J_3 and varying J_1 from positive to negative values, we disclose the low-temperature features of its interesting incommensurate phase. The gapless dispersion of quasiparticles leads to the intrinsic T^2 law of specific heat. The magnetic susceptibility is linear in temperature. The local magnetization is significantly reduced by quantum fluctuations. We address possible relevance of these results to the low-temperature properties of $NiGa_2S_4$. From a careful analysis of the incommensurate spin wavevector, the interaction parameters are estimated as $J_1 \approx -3.8755$ K and $J_3 \approx 14.0628$ K, in order to account for the experimental data.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In two-dimensional (2D) antiferromagnets, it was proposed the 'geometrical frustration' may enhance the quantum spin fluctuation and suppress the magnetic order to form a spin liquid [1]. In this context the triangular- and Kagomé-related lattices are studied extensively to seek a quantum spin liquid [2]. It turns out that the triangular lattice antiferromagnet with nearest-neighbor (NN) coupling exhibits 120° magnetic order [3], while the Kagomé lattice antiferromagnet is still a controversial topic for intriguing exploring [4]. People resort to other interactions, such as longer range and multiple-spin exchange ones, to realize a quantum spin liquid [2]. Experimental evidence in favor of this long-predicted spin-liquid state have emerged in recent years[5], although many aspects are still elusive. The spin disorder at low temperatures found in the compound $NiGa_2S_4$, in which Ni spins ($S = 1$) form a stack of triangular lattices, aroused much attention [6–9]. The crystal structure of the material is highly 2D, since inter-layer interactions are quite weak. Intriguing low-temperature properties of this material include the T^2 law of specific heat, incommensurate short-range spin correlation and lack of divergent behavior of the magnetic susceptibility. A dominant third-nearest-neighbor (3-NN) antiferromagnetic (AFM) interaction J_3 could produce the incommensurate phase in a rough picture: four sublattices will form commensurate 120° magnetic order separately if the NN interaction J_1 is zero, and the system will be driven into an incommensurate order if J_1 is gradually switched on. A first-principles calculation by Mazin [10] suggests a large 3-NN interaction J_3 and a negligible 2-NN interaction. J_3 is confirmed to be AFM, but the sign of J_1 has not yet been identified [10]. The classical

[5] The main evidence comes from the Kagomé-related materials. For recent progress see [5].

spin version of this model was studied in a Monte Carlo simulation [11], which provides some helpful information such as the incommensurability. Up to now, the quantum spin version of this model has not yet been studied very well. Besides the sign of J_1, many aspects of this model, either in agreement or disagreement with the experiment of $NiGa_2S_4$, need further clarification and treatment. In this paper we focus on the low-temperature properties of the quantum spin model and intend to make a contribution to this topic.

The Schwinger-boson mean-field theory (SBMFT) provides a reliable description for both quantum ordered and disordered antiferromagnets based on the picture of the resonant valence-bond (RVB) state [1, 12, 13]. As a merit, it does not prescribe any prior order for the ground state in advance, which should emerge naturally if the Schwinger bosons condense in the lowest energy states. For the Heisenberg antiferromagnets with NN couplings at zero temperature, it successfully captures the (π, π) magnetic order on the square lattice and the 120° magnetic order on the triangular lattice, respectively [12–15]. By means of SBMFT, we will show that the J_1–J_3 model falls into an incommensurate order phase at zero temperature for an AFM J_3 and either an FM J_1 or an AFM J_1. We also show that the T^2 law of specific heat is an intrinsic feature of this phase, the magnetic susceptibility is linear in temperature and the local magnetization is significantly reduced by quantum fluctuations. We address the possible relevance of these results to low-temperature properties of $NiGa_2S_4$. In agreement with previous work [11], we find that the NN interaction J_1 should be in the small FM region to obtain the incommensurate wavevector observed in the experiment. Our results suggests that the J_1–J_3 model is an essential part of the minimal model for $NiGa_2S_4$. In the following, we first present a formalism of the SBMFT scheme for the J_1–J_3 model, then solve the mean-field equations numerically and calculate relevant quantities. Finally we discuss the physical meanings of the results.

2. The Schwinger-boson mean-field theory

The J_1–J_3 model on the triangular lattice is

$$H = J_1 \sum_{\langle ij\rangle \in \mathrm{NN}} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle i'j'\rangle \in 3\mathrm{rd-NN}} \mathbf{S}_{i'} \cdot \mathbf{S}_{j'}. \quad (1)$$

We set $J_3 > 0$, but J_1 can be either AFM or FM. In the Schwinger-boson representation for the spin operators, $S_i^+ = a_i^\dagger b_i$, $S_i^- = b_i^\dagger a_i$, $S_i^z = (a_i^\dagger a_i - b_i^\dagger b_i)/2$ with $[a_i, a_j^\dagger] = [b_i, b_j^\dagger] = \delta_{ij}$, we decompose the NN and 3-NN interactions as [16]

$$J_1 \mathbf{S}_i \cdot \mathbf{S}_j = J_1 {:}F_{ij}^\dagger F_{ij}{:} - J_1 A_{ij}^\dagger A_{ij}, \quad (2)$$

$$J_3 \mathbf{S}_{i'} \cdot \mathbf{S}_{j'} = -J_3 \Pi_{i'j'}^\dagger \Pi_{i'j'}, \quad (3)$$

with $F_{ij} = (a_i^\dagger a_j + b_i^\dagger b_j)/2$, $A_{ij} = (a_i b_j - b_i a_j)/2$ and $\Pi_{i'j'} = (a_{i'} b_{j'} - b_{i'} a_{j'})/2$. Correspondingly, we introduce three competing mean fields, $F = \langle F_{ij}\rangle$, $A = -\mathrm{i}\langle A_{ij}\rangle$ and $\Pi = -\mathrm{i}\langle \Pi_{i'j'}\rangle$, and apply the Hartree–Fock decompositions for the interactions. A Lagrangian multiplier λ is also introduced to impose the constraint on the Schwinger bosons, $+\lambda \sum_i (a_i^\dagger a_i + b_i^\dagger b_i - 2S)$. After performing the Fourier transform, the effective Hamiltonian can be written in a compact form:

$$H_{\mathrm{eff}} = \sum_{\mathbf{k}} \phi_{\mathbf{k}}^\dagger M(\mathbf{k}) \phi_{\mathbf{k}} + \varepsilon_0, \quad (4)$$

where $\phi_{\mathbf{k}}^\dagger = (a_{\mathbf{k}}^\dagger, b_{\mathbf{k}}^\dagger, a_{-\mathbf{k}}, b_{-\mathbf{k}})$, $M(\mathbf{k}) = \epsilon(\mathbf{k})\sigma_0 \otimes \sigma_0 + \Delta(\mathbf{k})\sigma_y \otimes \sigma_y$, $\epsilon(\mathbf{k}) = \lambda - J_1 F \sum_\delta \cos k^{(\delta)}$, $\Delta(\mathbf{k}) = J_1 A \sum_\delta \sin k^{(\delta)} + J_3 \Pi \sum_\delta \sin 2k^{(\delta)}$, $\varepsilon_0 = 3N_\Lambda(-J_1 F^2 + J_1 A^2 + J_3 \Pi^2) - N_\Lambda \lambda (2S+1)$ and $\otimes$ means the Kronecker product, σ_0 is a 2×2 unit matrix and σ_αs ($\alpha = x, y, z$) are Pauli matrices, $k^{(\delta)} = k_x, k_x/2 + \sqrt{3}k_y/2, -k_x/2 + \sqrt{3}k_y/2$ for $\delta = 1, 2, 3$, respectively. The Matsubara Green functions are defined as

$$G(\mathbf{k}, \tau) = -\langle T_\tau \phi_{\mathbf{k}}(\tau) \phi_{\mathbf{k}}^\dagger \rangle, \quad (5)$$

where τ is the imaginary time and $\phi_{\mathbf{k}}(\tau) = \mathrm{e}^{\tau H_{\mathrm{eff}}} \phi_{\mathbf{k}} \mathrm{e}^{-\tau H_{\mathrm{eff}}}$. All physical quantities can be expressed in terms of the matrix elements of the Green function.

The Matsubara Green function in Matsubara frequency $\omega_n = 2n\pi/\beta$ (n is an integer for bosons) can be worked out as

$$G(\mathbf{k}, \mathrm{i}\omega_n) = \frac{\mathrm{i}\omega_n \sigma_z \otimes \sigma_0 - \epsilon(\mathbf{k})\sigma_0 \otimes \sigma_0 + \Delta(\mathbf{k})\sigma_y \otimes \sigma_y}{(\mathrm{i}\omega_n)^2 - \omega^2(\mathbf{k})}. \quad (6)$$

From the poles of the Matsubara Green function, the two degenerate spectra of the quasiparticles can be readily read out:

$$\omega(\mathbf{k}) = \sqrt{\epsilon^2(\mathbf{k}) - \Delta^2(\mathbf{k})}. \quad (7)$$

The mean-field equations are established by the constraint and the introduced mean fields. We omit the details and only present the results here:

$$\frac{1}{N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_{\mathrm{B}}[\omega(\mathbf{k})]) \frac{\epsilon(\mathbf{k})}{\omega(\mathbf{k})} = 2S + 1, \quad (8a)$$

$$\frac{1}{6N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_{\mathrm{B}}[\omega(\mathbf{k})]) \frac{\epsilon(\mathbf{k}) \sum_\delta \cos k^{(\delta)}}{\omega(\mathbf{k})} = F, \quad (8b)$$

$$\frac{1}{6N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_{\mathrm{B}}[\omega(\mathbf{k})]) \frac{\Delta(\mathbf{k}) \sum_\delta \sin k^{(\delta)}}{\omega(\mathbf{k})} = A, \quad (8c)$$

$$\frac{1}{6N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_{\mathrm{B}}[\omega(\mathbf{k})]) \frac{\Delta(\mathbf{k}) \sum_\delta \sin 2k^{(\delta)}}{\omega(\mathbf{k})} = \Pi, \quad (8d)$$

where $n_{\mathrm{B}}[\omega(\mathbf{k})] = [\mathrm{e}^{\omega(\mathbf{k})/k_{\mathrm{B}}T} - 1]^{-1}$ is the Bose–Einstein distribution function. In the thermodynamical limit $N_\Lambda \to \infty$, the momentum sum is replaced by an integral, $(1/N_\Lambda)\sum_{\mathbf{k}} \to (1/A_{\mathrm{BZ}}) \int \mathrm{d}^2 k$, $A_{\mathrm{BZ}} = 8\pi^2/\sqrt{3}$. If the Schwinger-boson condensation occurs at $\mathbf{k}^*$, a condensation term should be extracted in the momentum summation of the first equation, equation (8a):

$$2S + 1 = \rho_0 + \int \frac{\mathrm{d}^2 k}{A_{\mathrm{BZ}}} (1 + 2n_{\mathrm{B}}[\omega(\mathbf{k})]) \frac{\epsilon(\mathbf{k})}{\omega(\mathbf{k})}, \quad (9)$$

where the density of condensates:

$$\rho_0 = \frac{1}{N_\Lambda} \sum_{\mathbf{k}^*} (1 + 2n_{\mathrm{B}}[\omega(\mathbf{k}^*)]) \frac{\epsilon(\mathbf{k}^*)}{\omega(\mathbf{k}^*)}. \quad (10)$$

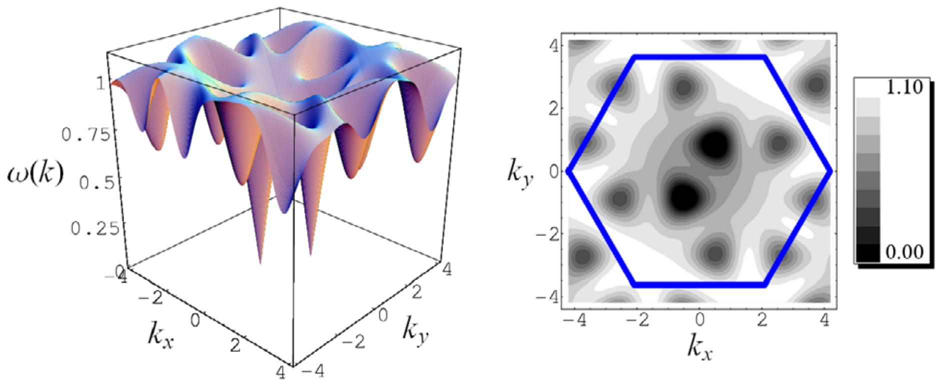

Figure 1. The gapless spectrum with nodal points. To compare with the experiment, we choose the parameter $J_1/J_3 = -0.2756$, so that the gapless nodal points occur at $\mathbf{k}^* = \pm(k^*/2, \sqrt{3}k^*/2)$ with $k^* = 0.158\pi$. The blue hexagon denotes the first Brillouin zone. See more details in the text.

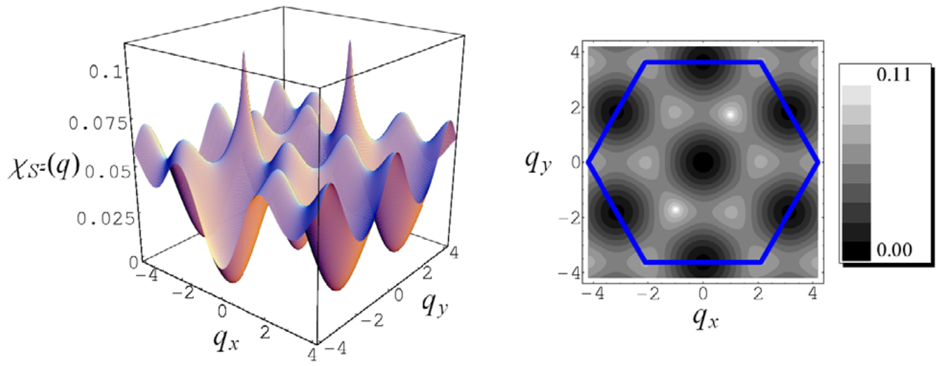

Figure 2. The zero-temperature static spin structure factor at the parameter $J_1/J_3 = -0.2756$. The blue hexagon denotes the first Brillouin zone. The divergent peaks located at $\mathbf{q}^* = 2\mathbf{k}^*$ indicate an incommensurate order.

Our numerical solution demonstrates the condensation occurs at zero temperature for spin $S > S_C$, where S_C varies with the ratio of J_3/J_1. Since we have $S_C < 0.172$ in our concerned range of the ratio of J_3/J_1, we will always consider condensations in the following discussions. The condensation terms in the next three mean-field equations, equations (8*b*)–(8*d*), should also be extracted carefully. It is noticeable the per site ground-state energy can be simplified by utilizing the mean-field equations:

$$E_0/N_\Lambda = \frac{1}{N_\Lambda}\left(\sum_{\mathbf{k}} \omega(\mathbf{k}) + \varepsilon_0\right) = -3J_1(A^2 - F^2) - 3J_3\Pi^2. \tag{11}$$

3. The incommensurate phase solution

The mean-field equations are solved numerically at zero temperature. For our purpose, we set $S = 1$ in the calculation in order to compare the result with the related experiment, although the qualitative conclusion is spin-independent, but the quantitative results vary with the values of spin. One fact that should be noticed is that the mean fields F and A could not exist simultaneously [14][6], so the number of mean-field equations can be reduced from 4 to 3 in both $J_1 > 0$ and $J_1 < 0$ regions. In the two regions, we found the system falls into the incommensurate phases with gapless excitations.

The quasiparticle's spectra become gapless at the nodal points, say $\mathbf{k}^* = (k_x^*, k_y^*) = \pm(k^*/2, \sqrt{3}k^*/2)$ (e.g. see figure 1). Near the nodal points, the spectrum is linear in $|\mathbf{k} - \mathbf{k}^*|$:

$$\omega(\mathbf{k}) \approx \alpha|\mathbf{k} - \mathbf{k}^*| + O(|\mathbf{k} - \mathbf{k}^*|^2). \tag{12}$$

At a finite temperature, a gapful spectrum will develop asymptotically as $\Delta_{\text{gap}} = c_1 \mathrm{e}^{-c_2/T}$ with constants c_1 and c_2, which coincides with the Mermin–Wagner theorem [13]. The incommensurate order at zero temperature of the system is signaled by the divergence in the static spin structure factor:

$$\chi_{S^z}(\mathbf{q}) = \frac{1}{N_\Lambda}\sum_{\mathbf{k}} \frac{1}{2}[P(\mathbf{k}+\mathbf{q})Q(\mathbf{k}) - R(\mathbf{k}+\mathbf{q})R(\mathbf{k})], \tag{13}$$

where $P(\mathbf{k}) = [\epsilon(\mathbf{k})/\omega(\mathbf{k}) + 1]/2$, $Q(\mathbf{k}) = [\epsilon(\mathbf{k})/\omega(\mathbf{k}) - 1]/2$, $R(\mathbf{k}) = \Delta(\mathbf{k})/[2\omega(\mathbf{k})]$. Because the spectra is gapless at $\mathbf{k}^*$, $\omega(\mathbf{k}^*) = 0$, $\chi_{S^z}(\mathbf{q})$ becomes divergent at $\mathbf{q}^* = 2\mathbf{k}^*$ (see figure 2):

$$\chi_{S^z}(\mathbf{q}^*) = \tfrac{1}{16} N_\Lambda \rho_0^2, \tag{14}$$

as it is proportional to the number of lattice sites N_Λ. The local magnetization will be reduced significantly due to strong

[6] Introduction of both F and A mean fields can be found in [14]. Here we find that if one supposes both F and A are nonzero at the same time, then an inconsistent result will be induced. This fact is very useful when one solves the equations. The merit of retaining both mean fields is that quite a good ground-state energy value can be produced.

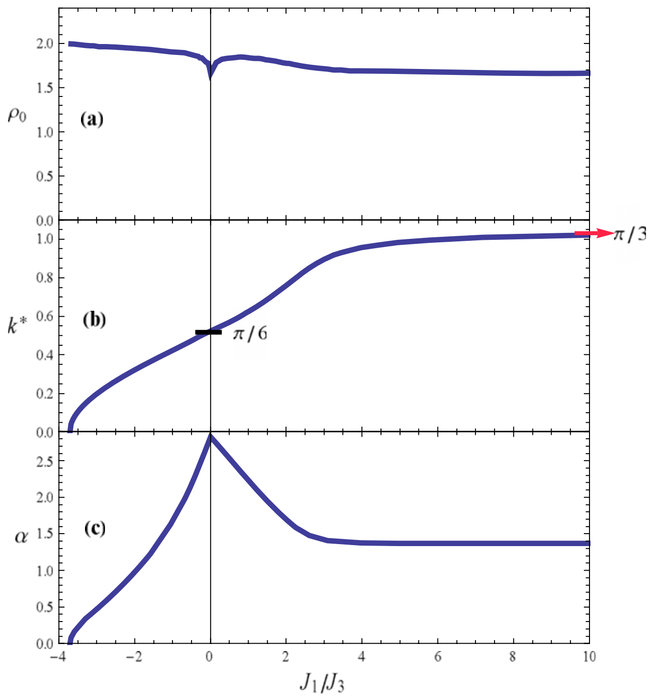

Figure 3. (a) The condensation term ρ_0 versus J_1/J_3. (b) The magnitude of the nodal point's momentum of the spectrum k^* versus J_1/J_3. In the limit $J_1/J_3 \to +\infty$, the result reproduces the 120° commensurate spin order correctly. The incommensurate spin wavevector observed in $NiGa_2S_4$, $k^* \cong 0.158\pi$ lies in the $J_1 < 0$ region. See more details in the text. (c) The coefficient α in equation (12) versus J_1/J_3.

quantum fluctuations:

$$m \approx \sqrt{\frac{\chi_{S^z}(\mathbf{q}^*)}{N_\Lambda |\cos \mathbf{q}^*|}} = \frac{\rho_0}{4\sqrt{|\cos \mathbf{q}^*|}}. \tag{15}$$

The important difference between the regions of $J_1 > 0$ and $J_1 < 0$ is the nodal point's momentum $k^* \in [\pi/6, \pi/3]$ for $J_1 > 0$ and $k^* \in [0, \pi/6]$ for $J_1 < 0$ regions, respectively. In the limit of $J_1/J_3 \to \infty$, $k^* \to \pi/3$, the solution reproduces 120° spin order correctly, while below the critical value $J_1/J_3 \approx -3.71$, the system becomes a saturated ferromagnet, where the linear expansion, equation (12), will be replaced by a parabolic form $\omega(\mathbf{k}) \approx \beta(\mathbf{k} - \mathbf{k}^*)^2$. The plots of k^* versus J_1/J_3 and α versus J_1/J_3 are shown in figure 3.

The incommensurate spin wavevector observed in $NiGa_2S_4$ is $k^* \cong 0.158\pi < \pi/6$ [6]. From this data we estimate that $J_1/J_3 \approx -0.2756$, which is slightly different from the value -0.20 in [6], i.e. we have a considerable FM J_1. Thus we can exclude the possibility of AFM J_1 [10]. The local site-averaged spin at $J_1/J_3 \approx -0.2756$ evaluated by equation (15) is 0.6223 for this spin-1 system, while the experimental data of $NiGa_2S_4$ suggest a larger value, 0.75(8) [6].

The nodal structure of the spectra, equation (12), leads to a linear density-of-states (DOS) in energy E:

$$D(E) = 2\sum_{\mathbf{k}} \delta(E - \omega(\mathbf{k})) \approx \frac{\sqrt{3}}{\pi\alpha^2} E \tag{16}$$

where the factor 2 comes from the degeneracy of the quasiparticle spectra. As a result, a T^2 law of specific heat follows apparently:

$$C_V/N_\Lambda \approx \frac{6\sqrt{3}\zeta(3)k_B^3}{\pi\alpha^2 J_3^2} T^2, \tag{17}$$

where $\zeta(3) = 1.202$. If one supposes that the T^2 law of specific heat of $NiGa_2S_4$ is ascribed to the gapless incommensurate phase, a numerical estimation, $J_1 \approx -3.8755$ K and $J_3 \approx 14.0628$ K, could be obtained.

4. Discussions

Before ending this paper, we point out that the zero-field susceptibility for this incommensurate phase is linear in temperature:

$$\chi_M/N_\Lambda \approx \frac{\sqrt{3}(g\mu_B)^2 k_B}{2\pi\alpha^2 J_3^2} T. \tag{18}$$

Using the parameters noted above, we find that it is $\chi_M \approx 2.77 \times 10^{-4} T$ (emu mol^{-1}), which is not in agreement with the experimental data of $NiGa_2S_4$, $\chi_M \approx A + BT$ with $A \approx 0.009$ (emu mol^{-1}) and $B \approx 0$ below 10 K [6]. The Monte Carlo study also shows the classical version of this model only produces a single peak in the specific heat [11]. These facts indicate that the model in equation (1) could not account for all mysteries in $NiGa_2S_4$. Thus, the solution shows the model equation (1) with AFM J_3 and FM J_1 has captured the main features for an incommensurate correlation in $NiGa_2S_4$, but it is still oversimplified as the minimal model for all low-temperature properties of $NiGa_2S_4$. A biquadratic interaction might be a good candidate for reproducing a finite susceptibility at zero temperature. In the absence of the 3-NN interactions, a biquadratic term can induce a quadrupolar order and totally suppress the spin order. The T^2 law of specific heat is also intact when quadrupolar order sets in [17–19]. It will be interesting to see how the incommensurate spin correlation will be influenced by the biquadratic interactions.

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