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Spin excitations in the Kitaev-Heisenberg model: Popov-Fedotov fermionization

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Abstract.

We study a Kitaev-Heisenberg model of spin 1/2 using the Popov-Fedotov fermionization proccedure. We derive the free energy of the quantum spin system on a Bravais lattice within one-loop approximation with exact on- site constraint. We discuss the obtained magnetic excitation spectrum for a particular case of square lattice in relation to the data derived by the equation of motion method and with the result of the linear spin wave theory in Holstein - Primakoff representation.

1. Introduction

Recently the Kitaev model has been attracted a lot of attention because it may effectively describe complicated magnetic properties of the strongly correlated transition metal compounds with entangling spin and orbital degrees of freedom in 5d electrons [1]. A strong spin orbital interaction in these materials results in an effective antiferromagnetic model for the pseudospin s = 1/2 with the bond dependent Kitaev interaction [2]. Iridium oxide compounds such as $\alpha\_Li_2IrO_3, \mathrm{Na}_2IrO_3, \alpha\_RaCl_3$... may be realistically described by Kitaev interaction together with isotropic Heisenberg interation, so called Kitaev-Heisenberg model [3]. From a theoretical viewpoint the Kitaev-Heisenberg model reveals very rich physics due to SU (2) broken symmetry [1]. It may lead to quantum spin liquid states. Also a multitude of unconventional ordered states may exist in Kitaev-Heisenberg model [1, 2]. In this paper we would like to see whether the functional integral approach proposed by Popov-Fedotov may be applied for studying the Kitaev-Heisenberg model. It is well known that the commutation relations for the spin operators are neither fermionic nor bosonic, leading to the absence of the Wicks theorem, so it is impossible to use an ordinary perturbation theory [4]. To resolve this problem one use different representations of the spin operators in terms of the canonical auxiliary Fermi or Bose ones. However the unphysical states in the Hilbert space, where the auxiliary operators are acting appear. These spurious states should be excluded by some constraint on every site, which is difficult to treat exactly. In 1988, Popov-Fedotov proposed a new fermionization procedure for quantum spin systems, in which the spin operators are represented in terms of the bilinear combinations of the

Fermi operators and an imaginary chemical potential is introduced for ensuring the exact local constraint [5]. The Popov-Fedotov trick has been used for studying ordered phases in different quantum spin systems [6, 7, 8]. It has been applied successfully also to the spin glass model [9], the negative U-Hubbard model [10].

In this paper we derive a general analytical expression of free energy in s = 1/2 Kitaev-Heisenberg model on a Bravais lattice. As an example, we discuss the result for the case of square lattice in comparison with the magnon energy obtained by the equation of motion method and by the approach using Holstein - Primakoff representation for the spin operators. This paper is organized as follows. In Sec.2 we formulate the model and give formalism. The general results are presented in Sec.3. Application to a square lattice and discussions are given in Sec.4.

2. Model and formalism

We start from the spin-1/2 Kitaev-Heisenberg (K-H) model:

$$H = \frac{1}{2}\sum_{ij} J_{ij}\vec{S}_i.\vec{S}_j + \frac{1}{2}\sum_{ij\nu}\Gamma^{\nu}_{\mathrm{ij}} S^{\nu}_i S^{\nu}_j. \tag{1}$$

There are two types of interactions along bonds $\nu\,(\nu = x, y, \mathrm{z})$: Heisenberg interactions J_{ij} and Kitaev interactions which couple S^{ν}_i, S^{ν}_j components along oriented bond. The strength and sign of the coupling J_{ij} and $\Gamma^{\nu}_{\mathrm{ij}}$ vary from material to material but a common feature in Kitaev material is that $\left|\Gamma^{\nu}_{\mathrm{ij}}\right| > |J_{ij}|$. It is convenient to rewrite Hamiltonian (1) in the form:

$$H = \frac{1}{2}\sum_{ij\nu} J^{\nu}_{\mathrm{ij}} S^{\nu}_i S^{\nu}_j, \tag{2}$$

with

$$J^{\nu}_{\mathrm{ij}} = J_{\mathrm{ij}} + \Gamma^{\nu}. \tag{3}$$

For definity we suppose $|\Gamma^x|, |\Gamma^z| > |\Gamma^y|$ so Oxz is the magnetic easy plane. In the classical limit of $S = \infty$ we can parameterize the magnetic state by some magnetic ordering vector $\vec{Q}$ as [11]:

$$\vec{S}_i = S\left(\vec{u}\sin\vec{Q}\vec{r}_i + \vec{v}\cos\vec{Q}\vec{r}_i\right). \tag{4}$$

with $\vec{u}$ and $\vec{v}$ being two orthogonal unit vectors in the Oxz plane. Inserting $\vec{S}_i$ into Hamiltonian (2) and minimizing obtained expression we can find for the given set of J^{ν}_{ij} with a particular lattice structure. Next step is to introduce a local reference frame where the local z axis is along the classical magnetization direction, defined by ordering $\vec{Q}$ [10]. In result in the local coordinate system K-H Hamiltonian reads:

$$H = \frac{1}{2}\sum_{i,j,\alpha,\beta} J^{\alpha\beta}_{\mathrm{ij}} S^{\alpha}_i S^{\beta}_j, \tag{5}$$

with the following non-zero components $J^{\alpha\beta}_{\mathrm{ij}}$:

$$\begin{cases} J^{yy}_{\mathrm{ij}} = J^{y}_{\mathrm{ij}}, \\ J^{xx}_{\mathrm{ij}} = J_{\mathrm{ij}}\cos\vec{Q}\delta_{ij} + \Gamma^{x}_{\mathrm{ij}} g_{1ij} + \Gamma^{z}_{\mathrm{ij}} g_{2ij}, \\ J^{zz}_{\mathrm{ij}} = J_{\mathrm{ij}}\cos\vec{Q}\delta_{ij} + \Gamma^{z}_{\mathrm{ij}} g_{1ij} + \Gamma^{x}_{\mathrm{ij}} g_{2ij}, \\ J^{xz}_{\mathrm{ij}} = -J_{\mathrm{ij}}\sin\vec{Q}\delta_{ij} + \Gamma^{x}_{\mathrm{ij}} g_{3ij} + \Gamma^{z}_{\mathrm{ij}} g_{3ji}, \\ J^{zx}_{\mathrm{ij}} = J_{\mathrm{ij}}\sin\vec{Q}\delta_{ij} + \Gamma^{x}_{\mathrm{ij}} g_{3ji} + \Gamma^{z}_{\mathrm{ij}} g_{3ij}. \end{cases} \tag{6}$$

where:

$$\begin{cases} g_{1ij} = \frac{1}{2}\left[\cos\vec{Q}\vec{\delta}_{ij} + \cos\vec{Q}\left(2r_i + \vec{\delta}_{ij}\right)\right], \\ g_{2ij} = \frac{1}{2}\left[\cos\vec{Q}\vec{\delta}_{ij} - \cos\vec{Q}\left(2r_i + \vec{\delta}_{ij}\right)\right], \\ g_{3ij} = -\frac{1}{2}\left[\sin\vec{Q}\vec{\delta}_{ij} - \sin\vec{Q}\left(2r_i + \vec{\delta}_{ij}\right)\right]. \end{cases} \tag{7}$$

$\vec{\delta}_{ij}$ being a vector connecting site i and site j. According to the Popov-Fedotov [5] the spin operators are represented in term of auxiliary Fermi operators $a^+_{i\sigma}, a_{i\sigma}$:

$$S^\alpha_i = \frac{1}{2}\sum_{i\sigma\sigma'} a^+_{i\sigma}\tau^\alpha_{\sigma\sigma'}a_{i\sigma'}. \tag{8}$$

where $\tau = (\tau^x, \tau^y, \tau^z)$ are the Pauli matrices. The unphysical states have to be eliminated by the constraint:

$$\hat{N}_i = \sum_\sigma a^+_{i\sigma}a_{i\sigma} = 1. \tag{9}$$

It can be enforced by inserting the projection operator $\hat{\mathrm{P}} = \frac{1}{i^N}e^{i\frac{\pi}{2}\hat{N}}$ to the partition function:

$$Z = Tr\left[e^{-\beta\hat{H}_{PF}}\hat{P}\right]. \tag{10}$$

where $\hat{H}_{PF}$ is the Hamiltonian (5) in auxiliary fermion representation (8). As a result, the fermionic Matsubara frequences are modified:

$$\omega = \frac{2\pi}{\beta}\left(n + \frac{1}{4}\right). \tag{11}$$

The next step is to represent the partition function (10) in a functional integral over Grassmann variables, where the products of four Grassmann variables are excluded by a Hubbard-Stratonovich transformation. After integrating out the Grassmann variables the partition function is given in the functional integral over the Bose auxiliary vector field $\vec{\varphi}_i$, introduced by the Hubbard-Stratonovich transformation. In order to apply a perturbation technique, we decompose the auxiliary field $\vec{\varphi}_i$:

$$\vec{\varphi}_i(\Omega) = \vec{\varphi}_{io} + \delta\vec{\varphi}_i(\Omega). \tag{12}$$

with the mean field value $\vec{\varphi}_{io}(\Omega = 0) \equiv \vec{\varphi}_{io}$ and the fluctuation component $\delta\vec{\varphi}_i(\Omega)$.
The further calculations closely following Ref. [8] are straightforward but lengthy and are not given in detail here.

3. General results

The zero order in the fluctuations gives rise to the mean-field free energy. The first order in the fluctuation term is cancelled due to the least action principle. The mean-field value of the auxiliary fields $\vec{\varphi}_{io}$ is related to the mean-field magnetization per site $\vec{m}_{io}$ as follows:

$$\varphi^\alpha_{io} = -\sum_{j\beta} m^\beta_{jo}J^{\beta\alpha}_{ij}. \tag{13}$$

In a Bravais lattice all sites are equivalent and in the local reference frame only the z-component of $\vec{m}_{io}$ and $\vec{\varphi}_{io}$ are non-zero $m_{io}^{\alpha} = m_{io}\delta_{\alpha,z}; \varphi_{io}^{\alpha} = \varphi_{io}\delta_{\alpha,z.}$. Then the mean-field magnetization is given by:

$$m_o = \frac{1}{2}\tanh\frac{\beta m_o}{2}\sum_j J_{ij}^{zz}. \tag{14}$$

The mean-field free energy reads:

$$F_{MF} = \frac{m_o^2}{2}\sum_{ij} J_{ij}^{zz} m_{io} + \ln 2\cosh\left(\frac{\beta\varphi_o}{2}\right). \tag{15}$$

In one loop approximation the fluctuation contribution to the free energy is given by:

$$\delta F_{fl} = \frac{1}{2\beta}\ln\det\hat{D}_{ij}\left(\Omega\right), \tag{16}$$

where:

$$\hat{D}_{ij}\left(\Omega\right) = \hat{I} + \hat{J}_{ij}\hat{K}_{ij}\left(\Omega\right). \tag{17}$$

It is convenient to evaluate (16) in a circular basic $(+,-,z)$ instead of the Decartes one (x, y, z). The elements of the interaction matrix $\hat{J}_{ij}$ in the circular basic are written in the following form:

$$\hat{J} = \begin{pmatrix} J_{\rm ij}^{xx} - J_{\rm ij}^{yy} & J_{\rm ij}^{xx} + J_{\rm ij}^{yy} & J_{\rm ij}^{xz} \\ J_{\rm ij}^{xx} + J_{\rm ij}^{yy} & J_{\rm ij}^{xx} - J_{\rm ij}^{yy} & J_{\rm ij}^{xz} \\ J_{\rm ij}^{zx} & J_{\rm ij}^{zx} & -J_{\rm ij}^{zz} \end{pmatrix} \tag{18}$$

The none-zero components of the matrix $\hat{K}_{ij}\left(\Omega\right)$ in the circular basics read:

$$\begin{cases} K_{ij}^{+-}\left(\Omega\right) = \left(K_{ij}^{-+}\left(\Omega\right)\right)^* = \frac{\beta}{2}\frac{m_o}{\varphi_o + i\Omega}\delta_{ij}, \\ K_{ij}^{zz}\left(\Omega\right) = -\frac{1}{4}\left(1 - 4m_o^2\right)\delta_{ij}\delta_{\Omega,0}. \end{cases} \tag{19}$$

To calculate $\det\hat{D}_{ij}\left(\Omega\right)$ we perform a Fourier transformation so:

$$\det\hat{D}_{ij}\left(\Omega\right) = \det\hat{D}\left(\vec{p},\Omega\right). \tag{20}$$

From (18) - (20) it is straightforward to derive the fluctuation contribution to the free energy (16).

From the expressions for the interaction matrix (6) - (7) one can see that for the isotropic Heisenberg part J_{ij} depends only on the neighboring vectors $\vec{\delta}_{ij} = \vec{r}_i - \vec{r}_j$, but the Kitaev part depends on $2\vec{r}_i + \vec{\delta}_{ij}$. As a consequence, for the Heisenberg model $\hat{D}\left(\vec{p},\Omega\right)$ is 3×3 matrix but for K-H model $\hat{D}\left(\vec{p},\Omega\right)$ is 6×6 matrix because the fluctuation of boson field $\delta\vec{\varphi}\left(\vec{p}\right)$ is coupled with $\delta\vec{\varphi}\left(\vec{p}+\vec{Q}\right)$. The determinant of is 6×6 matrix may be reduced to a determinant of 3×3 matrices by the following formula [13]:

$$\det\begin{pmatrix} \hat{\rm A}\hat{\rm B} \\ \hat{\rm C}\hat{\rm D} \end{pmatrix} = \det\left(\hat{\rm A}\hat{\rm D} - \hat{\rm B}\hat{\rm D}^{-1}\hat{\rm C}\hat{\rm D}\right). \tag{21}$$

In result we get:

$$\delta F = \frac{1}{2\beta} \sum_{\vec{p}\in BZ} \prod_{\Omega} \det \hat{E}, \tag{22}$$

The component of the 3×3 matrix $\hat{E}$ is given by:

$$\begin{cases} E_{11} = 1 + (X^o + Y^o)\, k^* - X^+X^- k^* b - W^+W^- k^* k_z, \\ E_{12} = (X^o - Y^o)\, k - X^+X^- kb - W^+W^- k k_z, \\ E_{13} = -W^o k_z - X^+W^- b k_z + W^+X^- k_z^2, \\ E_{21} = (X^o - Y^o)\, k^* - X^+X^- b k^* - W^+W^- k^* k_z, \\ E_{22} = 1 + (X^o + Y^o)\, k - X^+X^- bk - W^+W^- k k_z, \\ E_{23} = -W^o k_z - X^+W^- b k_z + W^+X^- k_z^2, \\ E_{31} = W^o k^* - W^+X^- b k^* + X^+W^- k^* k_z, \\ E_{32} = W^o k - W^+X^- bk + X^+W^- k k_z, \\ E_{33} = 1 + X^o k_z - W^+W^- b k_z - X^+X^- k_z^2. \end{cases} \tag{23}$$

here:

$$\begin{cases} k = \frac{1}{2}\frac{m_o}{\varphi_o - i\Omega}, \\ k_z = -\frac{1}{4}\left(1 - 4m_o^2\right)\delta_{\Omega,0}, \\ b = \frac{m_o \varphi_o}{\varphi_o^2 - (i\Omega)^2}, \\ \varphi_o = -m_o \sum\limits_j J_{ij}^{zz}. \end{cases} \tag{24}$$

$$\begin{cases} X^o = -\frac{1}{2}\left[2J_+(\vec{p}) + \Gamma_+^x(\vec{p}) + \Gamma_+^z(\vec{p})\right], \\ Y^o = -J(\vec{p}), \\ Z^o = -\frac{1}{4}\left[2J_+(\vec{p}) + \Gamma_+^x(\vec{p}) + \Gamma_+^z(\vec{p})\right], \\ W^o = -\frac{i}{4}\left[2J_-(\vec{p}) + \Gamma_-^x(\vec{p}) - \Gamma_-^z(\vec{p})\right], \\ X^\pm = -\frac{1}{4}\left[\Gamma^x(\vec{p} \pm \vec{Q}) - \Gamma_+^z(\vec{p} \pm \vec{Q})\right], \\ W^\pm = iX^\pm. \end{cases} \tag{25}$$

$$\begin{cases} J_\pm(\vec{p}) = J(\vec{p} + \vec{Q}) \pm J(\vec{p} - \vec{Q}), \\ \Gamma_\pm^\alpha(\vec{p}) = \Gamma^\alpha(\vec{p} + \vec{Q}) \pm \Gamma^\alpha(\vec{p} - \vec{Q}). \end{cases} \tag{26}$$

$J(\vec{p})$ is Fourier transformation of J_{ij} and $\Gamma^\alpha(\vec{p})$ is Fourier transformation of Γ_{ij}^α.

The Kitaev-Heisenber model (1) may possess many different classical ordered states: Neel type antiferromagnetic, ferromagnetic, columnar antiferromagnetic phases [3]. Each of the ordered states may be characterized by some ordering vector $\vec{Q}$, depending on the lattice structure and on the interaction strengths. Hence, in principle the above results give us a possibility to study the K-H s = 1/2 model on any Bravai lattice by means of Popov-Fedotov trick.

4. Application to a square lattice and discussion

As an example we apply the obtained results to the K-H model on a square lattice, investigated in [14, 15, 16]. We consider only the nearest neighbor interaction so the Heisenberg interaction reads:

$$J_{ij} = J\delta_{\vec{r}_j, \vec{r}_i + \vec{\delta}.} \tag{27}$$

And the Kitaev interaction is given by:

$$\Gamma^{\alpha}_{\mathrm{ij}} = \Gamma_{\alpha}\delta_{\vec{r}_j,\vec{r}_i+\vec{\delta}_{\alpha}}(\alpha = x, z). \tag{28}$$

$\vec{\delta}$ is the nearest neighbor vector, $\vec{\delta} = (\pm 1, \pm 1)$ and $\vec{\delta}_{\alpha}$ is the nearest neighbor vector along α direction, $\vec{\delta}_x = (\pm 1, 0); \vec{\delta}_y = (0, \pm 1)$ and the lattice constants a is taken to be 1. As it was shown in [13] the classical ground states of K-H model may be antiferromagnetic, ferromagnetic, columnar antiferromagnetic phases [3]. We study the case where $\Gamma^z > \Gamma^x > J > 0$ and the classical ordered phase is Neel-type antiferromagnetic with the ordering vector $\vec{Q} = (\pi, \pi)$ [14-16]. The Fourier components of the interaction are written in the form:

$$\begin{cases} J(\vec{p}) = 2J\left(\cos p_x + \cos p_z\right), \\ \Gamma^{\alpha}(\vec{p}) = 2\Gamma^{\alpha}\cos p_{\alpha}. \\ (\alpha = x, z) \end{cases} \tag{29}$$

From (14) the equations for the mean-field magnetization and free energy read:

$$m_o = \frac{1}{2}\tanh\beta m_o\left(2J + \Gamma_x + \Gamma_z\right). \tag{30}$$

$$F_{MF} = -\frac{m_o^2}{2}\left(2J + \Gamma_x + \Gamma_z\right) + \ln 2\cosh\left[\beta m_o\left(2J + \Gamma_x + \Gamma_z\right)\right]. \tag{31}$$

The fluctuation contribution to the free energy in one-loop approximation is found to be:

$$\delta F = \frac{1}{2\beta}\sum_{\vec{p}\in BZ}\ln A_o\left(\vec{p}\right) + \frac{1}{2\beta}\sum_{\substack{\vec{p}\in BZ \\ \nu=1,2}}\ln\frac{sh\frac{\beta E_{\nu}(\vec{p})}{2}}{sh\beta m_o\left(2J + \Gamma_x + \Gamma_z\right)}, \tag{32}$$

The first term in right hand of Eq.(32) is the contribution from the longitudinal fluctuation which is given by:

$$A_o = 1 + a(p)\left(1 - \frac{1}{4}m_o^2\right), \tag{33}$$

where $a(p)$ is some function of $\vec{p}$ and m_o satisfies Eq.(14). At low temperature $m_o \simeq \frac{1}{2}$ so the longitudinal contribution is negligible in comparison with the tranverse one given by second term in right hand of Eq.(32). The magnon energy $E_{\gamma}\left(\vec{p}\right)$ reads:

$$E_{1,2}\left(\vec{p}\right) = \left[\varphi_o^2 - P\left(\vec{p}\right) \pm \left[P^2\left(\vec{p}\right) + Q\left(\vec{p}\right)\right]^{1/2}\right]^{1/2}, \tag{34}$$

where:

$$P\left(\vec{p}\right) = \frac{1}{4}m_o\varphi_o\left(\Gamma^x\left(\vec{p}\right) + \Gamma^z\left(\vec{p}\right)\right) - \frac{1}{2}m_o^2 J\left(\vec{p}\right)\left[J\left(\vec{p}\right) + \frac{1}{2}\Gamma^x\left(\vec{p}\right) + \frac{1}{2}\Gamma^z\left(\vec{p}\right)\right], \tag{35}$$

$$Q\left(\vec{p}\right) = \frac{1}{4}(m_o\varphi_o)^2\left(\Gamma^x\left(\vec{p}\right) - \Gamma^z\left(\vec{p}\right)\right)^2\left(1 - \frac{m_o}{\varphi_o}J\left(\vec{p}\right)\right). \tag{36}$$

$$\varphi_o = 2m_o\left[2J\left(\vec{p}\right) + \Gamma^x\left(\vec{p}\right) + \Gamma^z\left(\vec{p}\right)\right]. \tag{37}$$

In the limit $\Gamma^{\alpha} = 0$, Eqs.(29)-(32) reduce to the results obtained by means of Popov-Fedotov approach for the Heisenberg model on the square lattice [7, 8]. In the limit of zero temperature Eqs.(34)-(37) give the similar magnon spectrum derived in linear spin wave theory based on Hostein-Primakoff representations of spin operators [13, 14]. In comparison with the results in [15] obtained by spin rotation invariant Green function method we list the following points.

i) The equations for magnon spectrum are similar if in our equations we replace m_o by σ of Ref. [15], which is the sublattice magnetization at $T = 0K$ renormalized due to zero point fluctuations.

ii) Note that in our results m_o depends on temperature by Eq.(14). Then the temperature dependence of the thermodynamic quantities in one loop approximation within Popov-Fedotov formalism is not only due to Bose distribution function of magnon but also the sublattice magnetization m_o. As a consequence, at finite temperature the Popov Fedotov method improves the thermodynamic quantities in comparison with the other approaches, where the exact constraint is relaxed by the average one.

In summary, we showed that functional integral method with exact constraint may be developed for anisotropic magnetic systems described by Kitaev-Heisenberg model. The essential point is that the fluctuations at momentum $\vec{p}$ are coupled with the fluctuations at momentum $\vec{p}+\vec{Q}$ making the calculation complicate. We sketched a common scheme for studying different ordered phases of the model in any Bravais lattices. As an example we studied the Neel type antiferromagnetic phase in a square lattice and compared with the other methods. The above results may be applied to study other ordered states or may be extended for Kitaev materials on non-Bravais lattices or with $S > \frac{1}{2}$. It should be left for future.

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