Instability of the chiral d-wave RVB state for the Heisenberg model on triangular lattice and an improvement of the Gutzwiller approximation

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(Dated: May 18, 2010)

Through Variational Monte Carlo simulation we show the d-wave RVB pairing in the Heisenberg model on triangular lattice can be better described in terms of a two component order parameter. The fully gapped chiral d-wave RVB state, which is predicted by the mean field theory to be the unique minimum of variational energy in the two dimensional representation space of d-wave pairing, is found to be actually a local maximum and the true minimum of energy is reached by the non-chiral \( d_{xy} \) state with line nodes. We also find that the usual Gutzwiller approximation, which enjoys great success for the square lattice system, fails badly on the triangular lattice as a result of the geometric frustration inherent of the system. An improved version of the Gutzwiller approximation is proposed and is found to give a much better results than the usual one.

PACS numbers:

The search for spin liquid ground state on geometrically frustrated quantum magnet is a central issue in the study of the strongly correlated electron systems. On the one hand, the spin liquid state represents a novel state of matter beyond the Landau-Ginzburg description. On the other hand, the study of the spin liquid state is closely related to the study of exotic superconductivity in strongly correlated electron system. The most famous example in this respect is the high-\( T_c \) cuprates. Although the parent compounds of the cuprates(an ideal quantum antiferromagnet described by the Heisenberg model on square lattice) exhibit magnetic order, the study of spin liquid state for the Heisenberg model on square lattice nevertheless predicts unambiguously the d-wave pairing symmetry for the superconducting order at finite doping and help to clarify the nature of the pseudogap phase.[1]

The discovery of superconductivity in hydrated \( Na_xCoO_2 \)[2] triggered another round of intensive researchs in this field as a result of the geometric frustration and the strong electron correlation inherent of the system which has a triangular lattice[3, 4]. Historically, it is first on the triangular lattice that the very concept of spin liquid state was first proposed[5]. However, after many year’s of intensive investigation the nature of the superconducting state remains illusive. While NMR measurement suggests a spin singlet pairing with line node, theories of the t-J model on triangular lattice predicts a chiral d-wave state with full gap and time reversal symmetry breaking. In recent years, the possibility of spin liquid state and exotic superconducting state(with self-doped charge carrier) in half-filled systems with an anisotropic triangular lattice and reduced strength of local correlation also arose great interest in the field[6]. Here the same uncertainty on the nature of the spin liquid state exist[6, 10, 12]. While some theory predicts a spin liquid state with a open Fermi surface for spin excitation, others argue spin pairing is unavoidable in the spin liquid state.

As compared to the square lattice, the triangular lattice is more complex in that there are more choices for the spin pairing pattern on the triangular lattice. According to group theory, spin pairing between neighboring sites on the square lattice can be either extended s-wave or d-wave, both of which belong to a one dimensional irreducible representation of the \( C_4 \) group. However, on the triangular lattice with a point group of \( C_{6v} \), the d-wave pairing channel becomes two dimensional and it is a non-trial problem to decide what kind of pairing pattern is actually realized within the two dimensional representation space of the d-wave pairing(the extended s-wave pairing is in general less stable than d-wave pairing for system with strong local correlation).

The slave Boson mean field theory and the Gutzwiller approximation based on it is widely adopted to study the problem of spin liquid and the pairing symmetry of the strongly correlated system[1]. Such a approach receives great succuss on the square lattice. For example, it predicts correctly the d-wave pairing symmetry of the high-\( T_c \) cuprates[13]. With the Gutzwiller approximation, the mean field theory can even give a quantitative correct estimation of the variational energy with an error less than 5 percent[14, 15]. On the triangular lattice, the slave boson mean field theory predicts unambiguously that the chiral d-wave pairing with time reversal symmetry breaking and a full gap is the most stable in two dimensional representation space and its condensation energy is more than 30 percent lower than that of the non-chiral(real) d-wave pairing state. The chiral d-wave state is widely believed to be the best choice for the triangular lattice.

In this paper, we show the usual Gutzwiller approximation fails badly for the Heisenberg model on the triangular lattice. It overestimates the condensation energy by more than 50 percent and predicts incorrect order for the relative stability of the chiral d-wave and non-chiral d-wave state. Through direct variational Monte Carlo simulation, we find the chiral d-wave RVB state is actu-
ally a local maximum in the two dimensional representation space of d-wave pairing and the true minimum of variational energy is reached by the non-chiral $d_{xy}$ state with line nodes. The anisotropy of the variational energy in the representation space is found to be much smaller than that predicted by the mean field theory and the RVB pairing should be more appropriately described in terms of a two component order parameter. We also find the failure of the usual Gutzwiller approximation on triangular lattice can be attributed to the geometric frustration inherent of the system. Based on this understanding, we propose an improved Gutzwiller approximation scheme and find it works much better than the usual one.

The Heisenberg model under consideration reads

$$H = J \sum_{<i,j>} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

here $\sum_{<i,j>}$ means sum over nearest neighbors on the triangular lattice. The slave Boson mean field theory of the RVB state is established by introducing the slave particle representation of the spin operator $\mathbf{S}_i^\dagger = \frac{1}{2} f_{i,\alpha}^\dagger \mathbf{\sigma}_{\alpha,\beta} f_{i,\beta}$ and then use the mean field order parameter $\chi_{ij} = \langle f_{i,\alpha}^\dagger f_{j,\alpha} \rangle$ and $\Delta_{ij} = \langle \epsilon_{\alpha,\beta} f_{i,\alpha}^\dagger f_{j,\beta}^\dagger \rangle$ to decouple the Hamiltonian written in terms of the slave particles. The slave particles must be subjected to the no double occupancy constraint to represent the spin operator faithfully. In mean field treatment, such local constraint is relaxed to a constraint on the average particle number on each site.

Assuming translational invariance for the RVB order parameter $\chi_{i,j}$ and $\Delta_{i,j}$, the mean field Hamiltonian for the slave particle has the form,

$$H_{MF} = \sum_{k,\alpha} \xi_k f_{k,\alpha}^\dagger f_{k,\alpha} + \sum_k (\Delta_k f_{k,\uparrow}^\dagger f_{-k,\downarrow} + h.c.) \quad (2)$$

whose ground state reads

$$|BCS\rangle = \prod_k (u_k + v_k f_{k,\uparrow}^\dagger f_{-k,\downarrow})|0\rangle, \quad (3)$$

in which $u_k = \frac{\Delta_k}{\xi_k + E_k}$, $E_k = \sqrt{\xi_k^2 + \Delta_k^2}$. Instead of determining $\chi_{i,j}$ and $\Delta_{i,j}$ from the self-consistent equation, we will take the wave function Eq.(3) as a variational description of the system at the mean field level. For simplicity, $\chi_{i,j}$ will be assumed to be a real constant and be nonzero for nearest neighboring sites only. The hopping energy $\xi_k$ then reads

$$\xi_k = -2(\cos k_x + 2 \cos \frac{k_x}{2} \cos \frac{\sqrt{3} k_y}{2}) - \mu, \quad (4)$$

here we have set the hopping integral as one. The chemical potential is determined by the mean field equation for the particle number and is not treated as an independent variational parameter. Thus the magnitude of the gap function, $\Delta$, is the only variational parameter in the theory.

The physical wave function of the spin system should satisfy the no double occupancy. This can be achieved by taking the Gutzwiller projection on the mean field wave function. The Gutzwiller projected mean field state takes the form

$$|\Psi\rangle = P_G \left( \sum_{i,j} a_{i-j} c_{i,\uparrow}^\dagger c_{j,\downarrow} \right) |0\rangle, \quad (5)$$

where the Cooper pair wave function $a_{i-j}$ is given by

$$a_{i-j} = \sum_k \frac{\Delta_k}{c_k \sqrt{\epsilon_k^2 + |\Delta_k|^2}} \exp[i(k\mathbf{R}_i - \mathbf{R}_j)], \quad (6)$$

and $P_G$ is the projection into the subspace of no double occupancy. The Gutzwiller projected wave function is usually treated by the variational Monte Carlo simulation method as analytical calculation is difficult for it. An estimate of variational energy in the Gutzwiller projected wave function can be given by the so called Gutzwiller approximation, in which the expectation value of a physical observable in the Gutzwiller projected state is approximated by its expectation value in the mean field state multiplied with a Gutzwiller factor. For the Heisenberg system, the Gutzwiller factor for spin correlation is usually taken to be $g_k = 4$.

Now we determine the structure of the pairing order parameter. Here we will consider the d-wave pairing only. On the triangular lattice, the d-wave pairing belongs to a two dimensional irreducible representation of the point group of $C_6$, and is thus more complex than its counterpart on the square lattice, which has a unique basis function of the form of $d_{x^2-y^2}$. The two basis functions of this two dimensional representation have the form of $d_{x^2-y^2}$ and $d_{xy}$. The symmetry of the system guarantee the invariance of the free energy under rotation in the two dimensional representation space at the quadratic level. Any anisotropy in the representation space must be attributed to higher order terms in the Landau expansion. Thus, if such anisotropy is small, one should better describe the pairing state with a multi-component order parameter.

In momentum space, these two basis functions of the d-wave representation have the form of $\Phi_1(\mathbf{k}) = \cos k_x - \cos \frac{\sqrt{3} k_y}{2}$ and $\Phi_2(\mathbf{k}) = \sqrt{3} \sin k_x \sin \frac{\sqrt{3} k_y}{2}$. In real space, they behave as $\cos 2\varphi$ and $\sin 2\varphi$ respectively (see Fig.1), where $\varphi$ is the angle the bond made with $x$-axis. The general order parameter is given by

$$\Delta_k = \Delta \eta_1 \Phi_1(\mathbf{k}) + \eta_2 \Phi_2(\mathbf{k})$$

where $\eta_1$ and $\eta_2$ are two complex numbers satisfying $|\eta_1|^2 + |\eta_2|^2 = 1$. Up to a global phase, the gap function can then be parameterized as follows

$$\Delta_k = \Delta \cos \theta \Phi_1(\mathbf{k}) + \sin \theta \exp(i\phi) \Phi_2(\mathbf{k})$$

in which $\Delta$, $\theta$ and $\phi$ are all real numbers. In this parameterization, the chiral d-wave state is given by $(\theta, \phi) =$
The two basis functions of d-wave pairing on triangular lattice. (a)$d_{x^2−y^2}$, (b)$d_{xy}$.

Owing to the gauge symmetry and symmetry of space inversion and time reversal, it is easy to see that we only need to consider the value of $\theta$ and $\phi$ in the region $(\theta, \phi) \in [0, \pi/2] \times [0, \pi/2]$.

At the mean field level, the chiral $d_{x^2−y^2} + id_{xy}$ state gives the lowest energy in this space while the maximum in energy is reached by the non-chiral $d_{xy}$ state. Note also that the energy is periodic in $\theta$ when $\phi = 0$ with a period of $\pi/3$, as a result of the six-fold rotational symmetry of the triangular lattice. A full scan of the variational energy (optimized with respect to $\Delta$) estimated from the usual Gutzwiller approximation with $g_s = 4$ is shown in Fig. 2. For any given $\theta$, a real gap function always gives higher variational energy than a complex gap function.

The chiral $d_{x^2−y^2} + id_{xy}$ state and the non-chiral $d_{xy}$ state as a function of $\Delta$. It is found that the chiral d-wave state enjoys a more than 30 percent lower condensation energy than its non-chiral counterpart. For this reason, the two component nature of the order parameter is irrelevant and we can take the chiral d-wave state as the unique choice for spin pairing.

Now we present the results of Variational Monte Carlo simulation. The simulation is done on a $14 \times 14$ lattice with periodic-antiperiodic boundary condition. $10^7$ statistically independent samples are used to estimate the variational energy. The result for the chiral $d_{x^2−y^2} + id_{xy}$ state and the non-chiral $d_{xy}$ state are shown in Figure 4. The result differs from the mean field result in three important aspects. Firstly, the order of relative stability between the chiral $d_{x^2−y^2} + id_{xy}$ state and the non-chiral $d_{xy}$ state is now reversed. Secondly, the difference in condensation energy is much smaller than that predicted by the mean field theory, being below 3 percent. Lastly, the variational energy estimated from the Gutzwiller approximation is about 50 percent lower than that calculated directly from the Monte Carlo simulation. We thus conclude that the usual Gutzwiller approximation fails badly on the triangular lattice.

A full scan of the variational energy (optimized with respect to $\Delta$) as a function of $\theta$ and $\phi$ is shown in Figure 5. The simulation is done on a $16 \times 16$ lattice with periodic-antiperiodic boundary condition. The variational energy is approximately periodic in $\theta$ with a period of $\pi/3$ when $\phi = 0$, as required by the six-fold rotational symmetry of the triangular lattice. The small deviation from such a periodicity, especially the small extra peaks around $(\theta, \phi) = (\pi/12, 0)$ and $(\theta, \phi) = (5\pi/12, 0)$, are caused by finite size effect which is the strongest for the non-chiral state with line node. From the Figure we see the

FIG. 2: A full scan of the variational energy (optimized with respect to $\Delta$) calculated from the Gutzwiller approximation with $g_s = 4$ in the representation space of d-wave pairing.

FIG. 3: The variational energy as a function of $\Delta$ calculated from the Gutzwiller approximation with $g_s = 4$ for the chiral $d_{x^2−y^2} + id_{xy}$ state and the non-chiral $d_{xy}$ state.
true minimum of the variational energy is reached by the non-chiral $d_{xy}$ state, while the chiral $d_{x^2-y^2}+id_{xy}$ state now becomes a local maximum. At the same time, the anisotropy of the condensation energy in the representation space, being less than 5 percent, is much smaller than that predicted by the mean field theory. For this reason, it is more appropriate to describe to the $d$-wave state and the non-chiral $xy$ state calculated from VMC simulation. The simulation is done on a $14 \times 14$ lattice with periodic-antiperiodic boundary condition.

Then, why does the usual Gutzwiller approximation, which perform well for the square lattice, fails so badly on the triangular lattice? To understand this, we rewrite the expectation value $\langle s_i^z s_j^z \rangle$ in the Gutzwiller projected state as follows,

$$\langle s_i^z s_j^z \rangle = \sum_\alpha W_\alpha s_i^z(\alpha)s_j^z(\alpha) = \sum_\alpha W_\alpha W_0^\alpha s_i^z(\alpha)s_j^z(\alpha),$$

(8)

in which $\alpha$ denotes one of the four spin configurations ($\uparrow\uparrow$, $\uparrow\downarrow$, $\downarrow\downarrow$) on site $i$ and $j$. $W_\alpha$ and $W_\alpha^0$ denote the probabilities for the spin configuration $\alpha$ to appear in the projected and mean field state. If we neglect the $\alpha$ dependence of the ratio $W_\alpha W_0^\alpha$, we get

$$\langle s_i^z s_j^z \rangle \approx g_s \sum_\alpha W_\alpha^0 s_i^z(\alpha)s_j^z(\alpha) = g_s \langle s_i^z s_j^z \rangle_0,$$

(9)

in which $g_s = \sum_\alpha W_\alpha^0 \langle \rangle_0$ denotes the expectation value in the mean field state. We note that such a approximation scheme becomes exact if $\frac{W_\alpha}{W_\alpha^0}$ is indeed $\alpha$ independent. In the BCS mean field state studied in this paper, the Gutzwiller factor $g$ is easily calculated to be given by $g_s = \frac{1}{\sum_\alpha |W_\alpha^0|^2 |\Delta_\alpha|^2}$, where $\chi_{ij} = \sum_\sigma \langle c_{i,\sigma}^\dagger c_{j,\sigma}^\dagger \rangle_0$, $\Delta_{ij} = \sum_\sigma \epsilon_{\sigma} \langle c_{i,\sigma}^\dagger c_{j,\sigma}^\dagger \rangle_0$. This is nothing but the two site version of the(improved) Gutzwiller approximation derived by Hsu [13]. The usual Gutzwiller approximation with $g_s = 4$ can be derived by simply neglecting the inter-site correlation and will be called the single site version of Gutzwiller approximation in the following. In Figure 6, we show the variational energy calculated from both the two-site and the single site version of Gutzwiller approximation and compared them to the result of VMC simulation. Although the two-site Gutzwiller approximation improve the result of the single site Gutzwiller approximation, none of them is truly satisfactory.

The failure of the above approximation schemes can be attributed to the assumption that the ratio $\frac{W_\alpha}{W_\alpha^0}$ being
\(\alpha\) independent. On the triangular lattice, a given pair of nearest neighboring sites \(i\) and \(j\) are neighbored by a third site \(k\). In the mean field state, the ratio between the probabilities for parallel and antiparallel spin alignment on site \(i\) and \(j\), namely \(R = \frac{W_\alpha}{W_0}\), depends crucially on the occupation of their common neighbor site \(k\). For example, if site \(k\) is empty, then site \(i\) and \(j\) can take the full advantage of forming singlet pair without being frustrated by the spin on site \(k\). Similarly, if site \(k\) is doubly occupied, then two antiparallel spins on site \(i\) and \(j\) can each form singlet pair with the electron on site \(k\). On the other hand, if site \(k\) is singly occupied, no matter what the spin it has, its coupling to the two antiparallel spins on site \(i\) and \(j\) is frustrated. For this reason, the ratio \(R\) is larger when site \(k\) is either empty or doubly occupied than it is singly occupied. This explains the breakdown of the assumption that \(\frac{W_\alpha}{W_0}\) being \(\alpha\) independent. To substantiate these arguments, we plot in Figure 7 the ratio \(R\) for the chiral \(d_{x^2-y^2} + id_{xy}\) state as a function of \(\Delta\) when site \(k\) is either empty \((R_e)\), doubly occupied \((R_d)\), or singly occupied \((R_s)\). From the figure we see \(R_s\) is always higher than \(R_e\) and \(R_d\) grows much faster than \(R_s\) and exceeds it at large \(\Delta\). All these observations are consistent with the qualitative arguments raised above.

The above reasoning also suggest a way to improve the Gutzwiller approximation. To reach this goal, we simply extend the two site version of the approximation to a three site version and use \(\alpha\) to denotes the eight possible spin configurations on a elementary triangle of the lattice. In this scheme, the variational energy per triangle reads

\[
(s_i^z s_j^z + s_i^z s_k^z + s_j^z s_k^z) = g_s((s_i^z s_j^z + s_i^z s_k^z + s_j^z s_k^z))_0,
\]

in which \(g_s = \frac{1}{\sum_{\alpha} W_\alpha}\). \(i, j, k\) denotes the three neighboring sites on an elementary triangle. The calculation of \(W_\alpha\) is straightforward but tedious and we will not present analytical expression for it here. For illustrative purpose, we show here the calculation of \(W_\alpha\) for one of the eight spin configuration, namely the configuration with all three spins aligned up. It is given by

\[
W_\alpha^{\uparrow\uparrow\uparrow} = ((1 - n_{i\uparrow})(1 - n_{j\uparrow})(1 - n_{k\uparrow})n_{k\uparrow})_0,
\]

which can be evaluated with the Wick expansion. The expression for other \(W_\alpha\) can be similarly constructed and calculated.

In Figure 6, we compare the variational energy calculated from the one site, two site and the three site version of the Gutzwiller approximation with the VMC results.

In conclusion, we have studied the problem of RVB pairing symmetry for the Heisenberg model on the triangular lattice, which is relevant for both the study of nature of the Mott insulating state of the half filled Hubbard model on the triangular lattice and the superconducting state in the doped system. Unlike the square lattice system, the d-wave pairing on the triangular lattice require a description in terms of a two component order parameter. Contrary to the previous study based on usual Gutzwiller approximation, we find the chiral d-wave state is actually a local maximum rather than local minimum in the two dimensional representation space of the d-wave pairing. We also find the anisotropy of condensation energy in the representation space is very small (less than 5 percent) and the true minimum is reached by the non-chiral \(d_{xy}\) with line nodes. We find the usual Gutzwiller approximation, which perform well on square lattice, fails badly on the triangular lattice. The failure of the usual Gutzwiller approximation scheme is traced back to the geometric frustration inherent of the triangular lattice and an improved version of the Gutzwiller approximation is proposed based on this understanding and is found to result in substantial improvement over the usual approximation scheme.

The author is supported by NSFC Grant No. 10774187 and National Basic Research Program of China No. 2007CB925001 and Beijing Talent Program.
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