Valence bond spin liquid state in two-dimensional frustrated spin-1/2 Heisenberg antiferromagnets

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Fermionic valence bond approach in terms of SU(4) representation is proposed to describe the $J_1-J_2$ frustrated Heisenberg antiferromagnetic (AF) model on a bipartite square lattice. A uniform mean field solution without breaking the translational and rotational symmetries describes a valence bond spin liquid state, interpolating the two different AF ordered states in the large $J_1$ and large $J_2$ limits, respectively. This novel spin liquid state is gapless with the vanishing density of states at the Fermi nodal points. Moreover, a sharp resonance peak in the dynamic structure factor is predicted for momenta $q = (0,0)$ and $(\pi,\pi)$ in the strongly frustrated limit $J_2/J_1 \sim 1/2$, which can be checked by neutron scattering experiment.

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A spin liquid ground state without any symmetry breaking is regarded as one of the most fascinating possibilities allowed by the physics of two-dimensional quantum antiferromagnets.\[1\] It is argued that reduced dimensionality, a small spin value, and the presence of competing interactions may lead to strong enough quantum fluctuations to destroy magnetic long-range order (LRO). A realistic prototype is the quantum two-dimensional spin-1/2 Heisenberg model with the nearest neighbor (NN) and next-nearest neighbor (NNN) antiferromagnetic (AF) couplings,\[2\] which has been recently materialized experimentally in Li$_2$VOSiO$_4$ and Li$_2$VOMeO$_4$ compounds.\[2\] The model is defined on a square lattice by

$$H = J_1 \sum_{n,n,n} S_i \cdot S_j + J_2 \sum_{n,n,n} S_i \cdot S_j,$$

where $J_1, J_2 > 0$, and the AF alignment between spins on the NN sites is hindered by the AF coupling of spins between NNN sites.

It is known that, when $J_2/J_1 \ll 1/2$, the ground state has the conventional Néel LRO with magnetic wave vector $Q = (\pi,\pi)$, while in the opposite limit $1 > J_2/J_1 \gg 1/2$, the minimum energy corresponds to a collinear state with $Q = (\pm \pi,0)$ and $(0,\pm \pi)$, consisting of two interpenetrating Néel sublattices with independent staggered magnetizations.\[3\] However, when $J_2/J_1 \sim 1/2$, i.e., in the strongly frustrated limit, the degeneracy of the ground state is large, and there is a consensus that it corresponds to a spin liquid state without LRO.\[3\] What is the exact nature of this nonmagnetic ground state turns out to be one of the most challenging problems for the physics of frustrated spin systems. There have been a number of different proposals, including the columnar, or spin-Peierls state, \[4,5,6,7,8\] where the spin rotational symmetry is preserved but the translational symmetry is broken, \[6,7,8\] the plaquette state, recovering the x-y symmetry, \[8\] a chiral-spin state, breaking the PT symmetry, \[10\] or a truly homogeneous state, not breaking any translational and rotational symmetries.\[11,12\] For some time the spin-Peierls and plaquette states seemed to be favored by numerical studies,\[13,14,15\] but the most recent numerical simulations\[16,17\] on finite size lattices show strong evidence against all states breaking translational symmetry, including spin-Peierls and plaquette states, staggered flux phase, etc. Moreover, Capriotti et al.,\[16\] have argued that the corresponding strongly frustrated ground state may be characterized by a projective BCS-type wave function though the two different AF LRO states in large $J_1$ and large $J_2$ limits failed to be reproduced. In view of this latest development the earlier proposal of homogeneous spin liquid state\[11,12\] appears now as a promising candidate. How to construct such a state, and how can it interpolate the two different AF LRO states in two opposite limits is an outstanding issue.

In this Letter, by using an SU(4) constrained fermion representation to describe the spin-1/2 operators on both sublattices simultaneously, we propose a fermionic valence bond (VB) approach to describe the $J_1-J_2$ frustrated Heisenberg AF model on a bipartite square lattice. A uniform mean field (MF) solution gives rise to a new VB spin liquid state with gapless spin excitations. At zero temperature, due to the inter-band nesting between two quasiparticle dispersions, the dynamic spin structure factor (SSSF) at momenta $q = (0,0)$ and $(\pi,\pi)$ for spins on the same sublattice and on different sublattices are found to be equal to each other up to sign, and a sharp resonance peak is formed in the strongly frustrated limit $J_2/J_1 \sim 0.5$. Although the true AF LRO can not be reproduced in the weakly frustrated limits, a broad peak at $(\pi,\pi)$ for $J_2/J_1 \ll 0.5$ is dominant in the static spin structure factor (SSSF) of spins on different sublattices, while for $J_2/J_1 \gg 0.5$ rather sharp peaks appear at $(0,\pm \pi)$ and $(\pm \pi,0)$ for spins on the same sublattice. It has thus been demonstrated a smooth crossover from the Néel to collinear AF (quasi) LRO when changing $J_2/J_1$ from small to large values, and a uniform VB spin liq-
uid phase interpolates between the two weakly frustrated regimes. The $J_1 - J_2$ model on a bipartite square lattice can be rewritten as

$$H = \frac{J_1}{2} \sum_{<i,j>} (S_{i,A} \cdot S_{j,B} + S_{i,B} \cdot S_{j,A}) + J_2 \sum_{(i,l)} (S_{i,A} \cdot S_{l,A} + S_{i,B} \cdot S_{l,B}),$$  \hspace{1cm} (2)$$

where $<ij>$ denotes summation over NN belonging to different sublattices, while $(i,l)$ means summation over the NN sites of the same sublattice. Each sublattice has $N$ sites. As we know, the bipartite lattice structure is essential for the spin density wave (SDW) theory in describing AF LRO state, where the correspondence between the spin operators on the two sublattices is assumed as $S^z_{i,B} \rightarrow S^z_{i,A}$, $S^z_{i,B} \rightarrow S^z_{i,A}$, and $S^x_{i,B} \rightarrow -S^x_{i,A}$, reflecting the presence of the Néel LRO. Actually, the bipartite lattice structure is also an important setting to study various quantum magnetic systems even in the absence of AF LRO. However, the correspondence between the spin operators on the two sublattices in the VB state is no longer the same as that in the SDW theory.

We generalize the conventional SU(2) constrained fermion to an SU(4) representation [18]. The generators of the SU(4) fermion representation are given by $F_{\beta}^\dagger (i) = C_{\alpha \beta}^\dagger C_{i,\beta}$, satisfying the SU(4) Lie algebra: $[F_{\beta}^\dagger (i), F_{\gamma}^\dagger (j)] = \delta_{\beta \gamma} F_{\alpha}^\dagger (i) - \delta_{\alpha \gamma} F_{\beta}^\dagger (i) \delta_{\beta, j}$. The spin operators on the sublattice $A$ can be expressed as

$$S^+_{i,A} = C_{i,1}^\dagger C_{i,2} + C_{i,3}^\dagger C_{i,4},$$

$$S^-_{i,A} = C_{i,2}^\dagger C_{i,1} + C_{i,4}^\dagger C_{i,3},$$

$$S^z_{i,A} = \frac{1}{2} \left[(C_{i,1}^\dagger C_{i,1} - C_{i,2}^\dagger C_{i,2}) + (C_{i,3}^\dagger C_{i,3} - C_{i,4}^\dagger C_{i,4})\right],$$

while $S^x_{i,B}$ and $S^y_{i,B}$ are given by interchanging $C_{i,2} \leftrightarrow C_{i,3}$, reflecting the symmetry of the model. With this representation, $S^x_{i,A}$ and $S^y_{i,B}$ ($\alpha = x, y, z$) are proved to satisfy their respective commutation relations of the SU(2) Lie algebra and $[S^\alpha_{i,A}, S^\beta_{i,B}] = 0$. By imposing a local constraint $\sum_{\mu} C^\dagger_{i,\mu} C_{i,\mu} = 1$ on each lattice site, we can further prove that $S^z_{i,A} = 3/4$.

In this representation, the $J_1 - J_2$ model can be rewritten as

$$H = -\frac{J_1}{4} \sum_{<i,j>} \left(A_{i,j} A_{i,j} + B_{i,j}^\dagger B_{i,j}\right)$$

$$-\frac{J_2}{2} \sum_{(i,l)} \left(P_{i,l} P_{i,l}^\dagger + Q_{i,l}^\dagger Q_{i,l}\right),$$  \hspace{1cm} (3)$$

where the normal ordering has been taken for the first and third terms, and four composite VB order parameters have been introduced

$$A_{i,j} = [(C_{j,1}^\dagger C_{i,1} + C_{j,4}^\dagger C_{i,4}) + (C_{j,3}^\dagger C_{i,2} + C_{j,2}^\dagger C_{i,3})],$$

$$B_{i,j} = [(C_{j,3}^\dagger C_{i,1} + C_{j,1}^\dagger C_{i,4}) - (C_{j,2}^\dagger C_{i,2} + C_{j,3}^\dagger C_{i,3})],$$

$$Q_{i,l} = [(C_{i,1}^\dagger C_{i,1} + C_{i,1}^\dagger C_{i,4}) - (C_{i,3}^\dagger C_{i,2} + C_{i,2}^\dagger C_{i,3})].$$

When uniform VB order parameters are assumed that $\langle A_{i,j} \rangle = \Delta_{1c}$, $\langle B_{i,j} \rangle = -\Delta_{1s}$, $\langle P_{i,l} \rangle = -\Delta_{2c}$, and the local constraint is replaced by a uniform Lagrangian multiplier, the MF model Hamiltonian is obtained as

$$H_{mf} = \frac{1}{2} \sum_k \Psi_k^\dagger \left[\begin{array}{c} \epsilon(\omega) \\ -\Omega_1 \end{array}\right]_k \Omega_1$$

$$+ \frac{1}{2} \sum_k \Phi_k^\dagger \left[\begin{array}{c} \epsilon(\omega) \\ -\Omega_2 \end{array}\right]_k \Omega_2,$$

where the Nambu spinors are introduced as $\Psi_k = (C_{k,1}^\dagger C_{k,3}^\dagger; C_{k,1}^\dagger C_{k,4}^\dagger)$, $\Phi_k = (C_{k,2}^\dagger C_{k,4}^\dagger; C_{k,2}^\dagger C_{k,3}^\dagger)$, and 4 x 4 matrices are defined by $\Omega_1 = \sigma_z \otimes \sigma_0$, $\Omega_2 = \sigma_y \otimes \sigma_0$, $\Omega_3 = \sigma_x \otimes \sigma_0$, $\Omega_4 = \sigma_y \otimes \sigma_0$. Moreover, $\Delta_{1c}(k) = J_1 \Delta_{1c}(\cos k_x + \cos k_y)$, $\Delta_{2c}(k) = J_2 \Delta_{2c}(\cos k_x + \cos k_y) + \Delta_{1s}(k)$, and $\Delta_{2s}(k) = J_2 \Delta_{2s}(\sin k_x + \sin k_y)$. The fermionic Matsubara Green function matrices are then derived as

$$G_{1,4}(k, i\omega_n) = \left[\begin{array}{c} \epsilon(\omega) - \Delta_{1c}(k) - \Delta_{2s}(k) \\ \Delta_{1c}(k) + \Delta_{2s}(k) \end{array}\right]_k \Omega_1$$

$$+ \left(\Delta_{1c}(k) + \Delta_{2s}(k)\right) \Omega_2^{-1},$$

$$G_{2,3}(k, i\omega_n) = \left[\begin{array}{c} \epsilon(\omega) - \Delta_{2c}(k) \end{array}\right]_k \Omega_3$$

$$- \Delta_{1s}(k) \Omega_4 - \Delta_{2s}(k) \Omega_2^{-1},$$

whose poles give rise to the quasiparticle dispersions

$$\epsilon_k = \sqrt{(\epsilon + \Delta_{1c}(k) - \Delta_{2c}(k))^2 + \Delta_{1s}(k)^2 \pm \Delta_{2s}(k)^2},$$

where $\epsilon_k$ corresponds to the triplet excitations with three-fold degeneracy, and $\epsilon_k$ corresponds to the singlet excitations. However, an inter-band nesting property, namely $\epsilon_k$ with a nesting wave vector $Q = (\pi, \pi)$ is a very important feature for the two quasiparticle bands. The usual intra-band nesting for the half-filled Hubbard model leads to the AF LRO at zero temperature. Similarly, the inter-band nesting will make the AF quasi-long range correlations dominant, excluding any possible incommensurate density wave states.

Hence the ground state energy per site is obtained and simplified as

$$\epsilon = -\frac{1}{N} \sum_k \epsilon_+(k) + J_1 \left(\Delta_{1c}^2 + \Delta_{1s}^2\right) + J_2 \left(\Delta_{2c}^2 + \Delta_{2s}^2\right) + \lambda.$$
and $\lambda$. By solving those equations, we can determine the saddle point parameters as functions of $J_2 / J_1$. We notice that the two pairing order parameters are competing with each other: when $J_2 / J_1 < 0.545$, $\Delta_{1s} > 2\Delta_q$, while for $J_2 / J_1 \geq 0.545$, $\Delta_{1s} \leq 2\Delta_q$.

Due to the inter-band nesting property, only the quasiparticle spectrum $\epsilon_\pm(k)$ is considered for different coupling parameters $J_2 / J_1$. Although the uniform VB order parameters are assumed above, $\epsilon_\pm(k)$ displays two nodal Fermi points at $(\mp k_{0x} , \pm k_{0y})$, and the quasiparticle density of states algebraically vanishes at the Fermi points. Moreover, when $J_2 / J_1 \ll 0.5$, the two nodal Fermi points are very close to the diagonal line $k_x + k_y = 0$, while for $J_2 / J_1 > 0.5$ the two nodal Fermi points are rotated versus the vertical line $k_x = 0$. The position of one nodal Fermi point ($-k_{0x}, k_{0y}$) is plotted as a function of $J_2 / J_1$ in Fig.1.

In order to reveal the nature of the new VB spin liquid state, the DSSFs are calculated. In terms of Nambu spinors $\Psi_i$ and $\Phi_i$, the spin operators on the two sublattices are written as $S_{i,A}^z = (\Psi_i^\dagger \Omega_0 \Psi_{i,A} - \Phi_i^\dagger \Omega_0 \Phi_i)/4$ and $S_{i,B}^z = (\Psi_i^\dagger \Omega_0 \Psi_{i,A} + \Phi_i^\dagger \Omega_0 \Phi_i)/4$, where $\Omega_0 = \sigma_z \otimes \sigma_z$, and in the Fourier space the DSSF for spins on the same and different sublattices are given by

$$
\begin{align*}
\chi_{A,A}(k,i\omega_n) &= \chi_{1,4}(q,i\omega_m) \pm \chi_{2,3}(q,i\omega_m), \\
\chi_{A,B}(q,i\omega_m) &= \frac{1}{16} \sum_{\omega_n} \int \frac{d^2 \mathbf{k}}{(2\pi)^2} 
\text{Tr} \left[ \Omega_0 G_{1,4}(k, i\omega_n) \Omega_0 G_{1,4}(q + k, i\omega_m + i\omega_n) \right],
\end{align*}
$$

where $\chi_{1,4}(q,i\omega_m)$ can be obtained by replacing $G_{1,4}$ with $G_{2,3}$ in $\chi_{1,4}(q,i\omega_m)$. Due to the vanishing density of states of quasiparticles around the nodal Fermi points, the DSSFs are not divergent at zero temperature, yielding only algebraically decaying spin-spin correlations (quasi-LRO).

By considering the inter-band nesting, the following important relations can be further proved

$$
\chi_{A,A}(k,i\omega_n) = \chi_{A,B}(q,i\omega_m)
$$

and

$$
\chi_{A,A}(0, i\omega_n) = -\chi_{A,B}(0, i\omega_m) \equiv \chi(Q, i\omega_m),
$$

which is delineated in Fig.2. Increasing the coupling parameter $J_2 / J_1$, a sharp resonance is gradually formed in the strongly frustrated limit $J_2 / J_1 \sim 0.5$ and disappears quickly away from it. The resonance peak simultaneously appears at momenta $(\pi, \pi)$ and $(0, 0)$, and may be regarded as a signature of the present fermionic VB spin liquid state in the strongly frustrated limit of the model, which is ready to be verified by future experiments.

Moreover, to compare with the AF LRO states in the large $J_1$ and large $J_2$ regimes, respectively, the SSSFs at $T = 0$ are evaluated. A smooth crossover is obtained for the SSSFs $\chi_{A,A}(q)$ and $\chi_{A,B}(q)$ when increasing the coupling parameter through the strongly frustrated limit $J_2 / J_1 = 0.5$. In Fig.3, for $J_2 / J_1 = 0.3$ a rather broad peak is clearly identified at $q = (\pi, \pi)$ in $\chi_{A,B}(q)$, while $\chi_{A,A}(q)$ has no distinctive features. This peak manifests the dominant quasi-long range AF correlations towards the Néel LRO in the limit of small $J_2 / J_1$. In contrary, in Fig.4 for $J_2 / J_1 = 0.7$, the SSSF $\chi_{A,B}(q)$ displays rather sharp peaks at momenta $(\pm \pi, 0)$ and $(0, \pm \pi)$, while the SSSF $\chi_{A,A}(q)$ is very small and spreads over a certain range. These peculiar features indicate the quasi-long range AF correlations towards the collinear LRO in the limit of large $J_2 / J_1$. Therefore, it has been shown that a fermionic VB spin liquid state in terms of the SU(4) representation exists and approximately interpolates between the two AF LRO states in the large $J_1$ and large $J_2$ limits, respectively.

In conclusion, an SU(4) constrained fermion representation has been used to describe the $J_1 - J_2$ frustrated Heisenberg AF model on a bipartite square lattice, and a fermionic uniform MF solution gives rise to a novel VB
spin liquid state with gapless spin excitations, not breaking any translational and rotational symmetries. This MF solution has reproduced correct asymptotic behavior in the weakly frustrated limits with the provision that the true LRO is approximated by quasi-LRO in our scheme. To the best of our knowledge, no other theoretical approach has succeeded in doing so in this complicated problem. The success of our MF theory is mainly due to the appropriate choice of representation which grasps the essential features of the strongly frustrated state. Of course, the eventual “proof” of our theory should come from the experiment. In particular, our explicit prediction of the sharp resonance peak at momenta \((\pi, \pi)\) and \((0, 0)\) in the imaginary part of the DSSF in the strongly frustrated limit \(J_2/J_1 \sim 0.5\) can serve as a crucial experimental test. There are still many open problems, for example, what is the nature of transitions between this VB state and AF LRO states, their possible coexistence, etc. These issues require further studies.

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