Correlated resonating valence bond state and gapless spin liquid state in the spin-1/2 $J_1 - J_2$ Antiferromagnetic Heisenberg model on a square lattice

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We propose a class of variational wavefunctions, namely the correlated resonating valence bond (CRVB) states, for the frustrated Hamiltonians in the paramagnetic phase. This class of wavefunctions allow negative amplitude and the same sublattice pairing when a bipartite lattice is considered, however it suffers from the sign problem if handled via variational Monte Carlo (VMC) method. Fortunately, this class of wavefunction can be approximated written in the language of projected entangled pair states (PEPSs), thus the sign problem can be lifted. We apply this wavefunction to the $J_1 - J_2$ Antiferromagnetic Heisenberg model on a square lattice in the coupling region $J_2 \in [0.45:0.56]$. Two approaches have been taken to determine the nature of the intermediate phase: first, we make extensive VMC simulation to optimize the variational parameters on a relatively small systems up to $8 \times 8$; second, we approximate the optimized wavefunction with a PEPS and calculate the correlation functions on a $32 \times 32$ torus. We find that the paramagnetic phase is a gapless spin liquid in the entire range of $J_2 \in [0.45:0.56]$ with a gapless singlet excitation and a gaped triplet excitation. We then perform several steps (up to three) of ground state projection to these wavefunctions at the optimized parameters, the energies before projection are competitive with the slave fermion projective BCS wavefunction without Lanczos optimization, and the energies after projection are consistent with the state of arts density matrix renormalization group (DMRG) results and the projective BCS wavefunction after Lanczos optimization.

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Introduction— Searching for exotic phases has been an exciting task in the last two decades motivated by the potential application to quantum computation [1-2]. Frustrated magnets can host exotic phases whose excitations carry fractional quantum numbers [3-7]. However due to their frustrated nature, finding the exotic phases among any realistic frustrated magnets has been a long-time challenge both theoretically and experimentally [8-12].

The $J_1 - J_2$ antiferromagnetic (AF) Heisenberg model on a square lattice is an intriguing frustrated magnet due to its close relation to high-temperature superconductor materials [13,14]. The nature of its ground state in the intermediate coupling region has been debated for decades [15-18]. Recently, several possibilities have been re-proposed for this model. A density matrix renormalization group (DMRG) study on long cylinders suggested a gaped $Z_2$ spin liquids from evidences of non-vanishing singlet and triplet gap in the paramagnetic phase [19,20]. Later on, a Gutzwiler-projected BCS wavefunction study indicated a gapless spin liquid state supported by the existence of gapless triplet excitation at momenta $(\pi, 0)$ and $(0, \pi)$ [21]. Most recently, another DMRG study with imposed SU(2) symmetry carefully investigated this model on both torus and long cylinders, and found a diverging dimer correlation length, from which the authors claim to find a plaquette valence bond solid (VBS) state as the ground state [22]. These controversial results raise the need for a careful analysis of numerical convergence and system size dependence [23].

For a frustrated system with a sign problem, beyond the unbiased DMRG and exact diagonalization (ED) method, a good trial wavefunction is the key to all variational numerical methods. Resonating valence bond (RVB) state is a quantum fluctuating valence bond product state, which was proposed as the ground state of spin liquids and suggested to play an important role in the high-temperature superconductivity [24,25]. RVB states are powerful to describe a variety of phases including spin liquids and magnetically ordered states [3,7,26,27]. In this paper, we introduce a class of correlated resonating valence bond states (CRVB) within the context of the nearest neighbor (NN) valence bond (VB) pairs and their local resonating configurations. The fluctuation probabilities between the local correlated pairs are variational parameters in the trial state, which we optimize by minimizing the ground state energy for the $J_1 - J_2$ AF Heisenberg model on the square lattice. Here negative bond amplitudes and same-sublattice pairings are introduced. The need for negative amplitudes was brought out by previous studies of amplitude product (AP) state on a $4 \times 4$ torus, where a negative amplitude $h(1, 2)$ helps to minimize the ground state energy beyond certain coupling strength [28,29]. Another study [30] suggested a gaped $Z_2$ spin liquid on a bipartite lattice by introducing diagonal bond pairs. Our ansatz accommodate both situations, however we find that neither negative amplitudes nor diagonal bond pairs are the signal of gaped $Z_2$ spin liquid.

Hamiltonian and CRVB trial wavefunction— The Hamiltonian of the AF $J_1 - J_2$ model on a square lattice...
any operator $\hat{O}$ is the number of loops.

\begin{equation}
H = J_1 \sum_{(i,j)} S_i \cdot S_j + J_2 \sum_{\langle(i,j)\rangle} S_i \cdot S_j,
\end{equation}

where $(i,j)$ means the NN pair and $\langle(i,j)\rangle$ means the next NN (NNN) pair. Apparently, the short range AP state in 2D on a bipartite lattice is too limited, since it describes only the gapless spin liquids \cite{46, 47}. A generalization is to consider bond bond correlations \cite{48}. Given the NN RVB states on a square lattice, all possible arrangement of bond pairs via a link are shown in Fig. 1(a), noting that lattice rotation symmetry are imposed. Within each of the above four-spin subspace, there are two more VB tilings (w.r.t the same link) as presented in Fig. 1(b,c). The CRVB states are superpositions of all compact packing of NN bonds and shapes (paired-bonds including the link) in Fig. 1(b,c).

\begin{equation}
|\Psi\rangle_{\text{CRVB}} = \prod_{\{V_p\}} b_i^{n_{i_b}} c_j^{n_{i_c}} |V_p\rangle,
\end{equation}

where $\{V_p\}$ means all compact packing configurations, which is an extended space than the VB product states; $b_i, c_j$ are the shape amplitudes, which serves as variational parameters, and $n_{i_b}, n_{i_c}$ are the number of such shapes in a packing configuration $V_p$. We also imply that NN bond has amplitude 1. Shapes as in Fig. 1(a) do not enter into configuration $V_p$ due to identity

\begin{equation}
(i_a, j_a)(i_\beta, j_\beta) - (i_a, j_\beta)(i_\beta, j_a) = 0,
\end{equation}

where $(i_\alpha, j_\alpha)$ represents a singlet directing from site $i_\alpha$ to $j_\alpha$. To approximate the CRVB state using the projected entangled pair state (PEPS), we take a similar construction as in Ref. \cite{49, 50} using a relative large virtual spin space of \(\frac{1}{2} \otimes \frac{1}{2} \oplus \ldots \otimes 0\) and present the detail in the Appendix.

The variational MC method– The expectation value of any operator $\hat{O}$ is given by

\begin{equation}
\langle \Psi | \hat{O} | \Psi \rangle = \frac{\sum_{\alpha\beta} W_{\alpha\beta} \text{sgn}_{\alpha\beta} O_{\alpha\beta}}{\sum_{\alpha\beta} W_{\alpha\beta} \text{sgn}_{\alpha\beta}},
\end{equation}

where the importance weight is

\begin{equation}
W_{\alpha\beta} = |\psi(V_\beta)\psi(V_\alpha)\rangle\langle V_\beta|V_\alpha\rangle|,
\end{equation}

\text{sgn}_{\alpha\beta} is defined by $\text{sgn}_{\alpha\beta} W_{\alpha\beta} = \psi(V_\alpha)\psi(V_\alpha)\langle V_\beta|V_\alpha\rangle$, and

\begin{equation}
O_{\alpha\beta} = \frac{\langle V_\beta|\hat{O}|V_\alpha\rangle}{\langle V_\beta|V_\alpha\rangle}.
\end{equation}

Since our trial wavefunction has negative bond pair amplitude [see Fig. 2(b)] and the same sublattice bond pairs [see Fig. 2(c)], the VMC suffers from the sign problem. The sign factor comes from the wavefunction coefficients $\psi(\alpha)$ and the overlap matrix elements $\langle V_\beta|V_\alpha\rangle$. We define the reference VB sign convention for the AA (BB) bond at distance (1,1) and (2,0) as in Fig. 2(a). Fig. 2(b) gives an example of calculating the overlap matrix element $\langle V_\beta|V_\alpha\rangle$. Given VB configurations $V_\alpha$ and $V_\beta$, flip all singlets arrows in $V_\beta$, and draw the transition graph Fig. 2(b). The matrix element $\langle V_\beta|V_\alpha\rangle = (-1)^{n_v} 2^{n_l - \frac{1}{2}}$, where $n_v$ is the number of arrows that violate the direction of the flow and $n_l$ is the number of loops.

The VMC for wavefunction (2) employs two kinds of update. First update is called the local update; randomly select a link and check the two VBs connected via this link, if they both appear as individual VBs, find out in which relative position they are as in Fig. 1(a) (say $a_i$) and replace them by a shape randomly chosen between $b_i$ and $c_i$ with probability $\min[|b_i| \times 2^{n_l-n_i}, 1]$ and $\min[|c_i| \times 2^{n_l-n_i}, 1]$; if the randomly selected link is exactly the link of any shape in $V_p$, replace the shape ($b_i$ or $c_i$) by two individual VBs as arranged in $a_i$ with probability $\min[\frac{1}{|b_i|} \times 2^{n_l-n_i}, 1]$ or $\min[\frac{1}{|c_i|} \times 2^{n_l-n_i}, 1]$; for any other case, exit the local update. Second update
The loop update: randomly construct a loop composed of alternating singlet bonds and unoccupied links in area excluding shapes, and shift the valence bonds by one site along the loop with probability \( \min[2^{n_l^t}, 1] \). Here \( n_l^t \) is the number of loops in the transition graph as if the trial update is accepted.

We calculate the derivative of the trial energy with respect to a variational parameter \( a \) \( (a \in \{b_1, c_1\}) \) as

\[
\frac{\partial \langle E \rangle}{\partial a} = \frac{n_a}{a} \langle E \rangle - \frac{\langle n_a \rangle}{a} \langle E \rangle,
\]

and update it according to [43]

\[
a^{t+1} = a^t - r \delta t^t \times \text{sign} \left( \frac{\partial \langle E \rangle}{\partial a} \right),
\]

where \( t \) is the iteration index, \( r \) is a random number \( r \in [0, 1] \), and \( \delta = 0.01/t \). The form of \( \delta \) is rather heuristic.

**Results**—We present the well optimized variational parameters for system sizes \( L = 4, 6, 8 \) at \( J_2 \in [0.45 : 0.56] \) for the wavefunction \( \Psi_{\text{CRVB}} \) in Fig. 3. There are strong size dependence for the optimized variational parameters \( b_3, b_4 \). However, parameters \( b_1, b_2 \) and \( c_2 \) are large in absolute value and have less size dependence. Other optimized parameters that are not shown are zero. The absolute energy error per site \( \Delta E(L, J_2) \) compared with the exact diagonalization (ED) results for \( L = 4, 6 \) [27] are presented in Fig. 3(f). For example, \( \Delta E(4, 0.55J_1) \sim 2 \times 10^{-3} J_1 \) and \( \Delta E(6, 0.55J_1) \sim 3 \times 10^{-3} J_1 \). Another comparison is with the Gutzwiller-projected BCS wavefunction results without Lanczos projection [37]: at \( J_2 = 0.55J_1 \), our energy error is lower by 40% than the bare Gutzwiller wavefunction.

We then perform several powers of Hamiltonian projection on the trial wavefunction \( |\Psi_n\rangle \equiv (C - H)^n |\Psi\rangle_{\text{CRVB}} \) (C is a constant) and 1-step Lanczos optimization [51] \( |\tilde{\Psi}_n\rangle = (1 + \alpha H)|\Psi_n\rangle \) (\( \alpha \) is variable, can be optimized) (see Appendix B). We extrapolate the ground state energy as a function of the variance \( \sigma^2(\Psi) \equiv \langle \Psi | H^2 | \Psi \rangle / \langle \Psi | \Psi \rangle - (\langle \Psi | H | \Psi \rangle / \langle \Psi | \Psi \rangle)^2 \) for any |\Psi\rangle defined as above. At \( L = 4, 6, 8 \), we perform a maximum number of \( n = 3 \) (\( n = 2 \)) projection steps, and present energies versus their variance in Fig. 4. For each \( L \) and \( J_2 \), the first \( n \) points with larger variance correspond to \( |\Psi_n\rangle \) and the rest points correspond to \( |\tilde{\Psi}_n\rangle \). At \( L = 4, 6, 8 \), the horizontal lines are ED [27] (DMRG [38]) results. A clear consistency has been reached between them.

The spin correlation function is defined as

\[
C(r) = \frac{(-1)^{x_1 + y_1}}{N} \sum_i \left( \mathbf{S}_{r_i} \cdot \mathbf{S}_{r_i+r} \right),
\]

and the magnetization is written as \( M^2 = \frac{1}{N} \sum_r C(r) \). The dimer correlation is defined as

\[
D_{xx}(r) = \frac{1}{N} \sum_i \left( \mathbf{S}_{r_i} \cdot \mathbf{S}_{r_i+x} \right) \left( \mathbf{S}_{r_i+r} \cdot \mathbf{S}_{r_i+r+x} \right),
\]

where \( e = x, y \). In order to see the long range behavior of either plaquette or columnar VBS pattern, one has to take the appropriate combination [38]

\[
D^e_{xx}(r) = D_{xx}(r) - \frac{1}{2} \left[ D_{xx}(r - \vec{e}) + D_{xx}(r + \vec{e}) \right].
\]

The dimer order parameter is \( D^2 = D_x^2 + D_y^2 \), where

\[
D_x = \frac{1}{N} \sum_i (-1)^{x} \mathbf{S}_{r_i} \cdot \mathbf{S}_{r_i+x},
\]

\[
D_y = \frac{1}{N} \sum_i (-1)^{y} \mathbf{S}_{r_i} \cdot \mathbf{S}_{r_i+y}.
\]
FIG. 5: (a) Magnetization and (b) dimer order as a function of system size at $J_2 = 0.45, 0.5, 0.55$ in the log-log plots. Power law functions $M^2 \sim 1/L^\alpha$, $D^2 \sim 1/L^\beta$ are fitted to the points at $L \geq 10$. The long range (c) spin and (d) dimer correlation functions on a $32 \times 32$ torus using tensor MC simulation of a PEPS representation to wavefunction 2 at $b_1 = 0.35$, $c_2 = 0.27$, $b_2 = -0.15$. The spin correlation is exponentially decaying with decay length $\xi_s = 1.28$, the dimer correlation function is power law decaying $D_{xx}^2(x) \sim 1/x^{\beta_s}$, with $\beta_s = 1.60$.

We take the optimized parameters from $L = 8$ and calculate correlation functions using VMC method for $L > 8$ systems until the signs are no longer manageable. The magnetization and dimer order are presented in a log-log scale in Fig. 5(a,b). The magnetization scales as $M^2 \sim 1/L^{\alpha}$, which is expected for an exponentially decaying spin correlation function. The dimer order follows a power law decay $D^2 \sim 1/L^{\beta}$ with $\beta \approx 1$. We turn to the PEPS wavefunction that approximates the 2 [2] taken at parameters $b_1 = 0.35$, $c_2 = 0.27$, $b_2 = -0.15$ and others 0, which is somewhere deep in the intermediate phase, and use tensor MC method [52] to estimate the correlation function on a $32 \times 32$ torus. The results are plotted in Fig. 5(c,d). The spin correlation function is exponentially decaying in the $x$ direction $C(x, 0) \sim \exp(-x/\xi_s)$ with $\xi_s \approx 1.28$. The dimer correlation function follows a power law decay $D_{xx}^2(x, 0) \sim 1/x^{\beta_s}$ with $\beta_s = 1.60$.

Discussion and conclusions—Using the correlated resonating valence bond (CRVB) state, we minimize the ground state energy of the $J_1 - J_2$ antiferromagnetic (AF) Heisenberg model on a square lattice for a coupling ratio $J_2/J_1 \in [0.45 : 0.56]$ by varying a few bond pair (shape) amplitudes. We further apply Hamiltonian projection and 1-step Lanczos optimization [41] on the optimized CRVB wavefunction, and extrapolate the energy as a function of the variance. The energies are consistent with the exact diagonalization (ED) results on the $4 \times 4$ and $6 \times 6$ tori [27] and the density matrix renormalization group (DMRG) results on the $8 \times 8$ torus [38]. We apply the optimal variational parameters from the $8 \times 8$ system to study a larger torus by extensive valence bond Monte Carlo (MC) simulation. The Neel order parameter has an inverse volume scaling, and the dimer order parameter describing the columnar or plaquette valence bond solid (VBS) phases follows a slow power law decay ($\sim 1/L$) with system size $L$, indicating a critical phase with gapless singlet excitations and gaped triplet excitations in the entire range of $J_2/J_1 \in [0.45 : 0.56]$. The projected entangled pair state (PEPS) that approximates the CRVB variational state is investigated on a $32 \times 32$ torus via tensor MC sampling method [52]. We find that the spin correlation decays exponentially and the dimer correlation decays as a power law function, which again confirm the above conclusion.

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Supplementary material

Trial PEPS wavefunction

Let us first remind the construction of the equal weight NN RVB state with a $D = 3$ PEPS [49, 50], the projector matrix $P$ on the vertex and bond tensor $S$ are

$$P_1 = \sum_{\mu=r,u,l,d} \langle \uparrow \rangle \langle 0|_{\mu} + | \downarrow \rangle \langle 1|_{\mu} \otimes \langle 222|_{\mu}, \quad (14)$$

$$|S_1\rangle = |\epsilon\rangle + |22\rangle, \quad (15)$$

where the virtual spin lives in a $1/2 \oplus 1/2$ space: the virtual state “0,1” represent sub-space of spin 1/2 and “2” represents sub-space 0; “$/\mu$” means all indices other than $\mu$. Now we introduce a product auxiliary space of $(1/2 \oplus 1/2) \otimes (1/2 \oplus 1/2)$ to each bond, indicating two virtual spins each spanning a virtual space $\{0, 1, 2\}$. Fig. 6 graphically draws the projector $P$ and matrix $S$ of a local two VBs state (shapes) corresponding to the left VBs tiling. The $P$ and $S$ read as

$$P_2 = \sum_{\mu=r,u,l,d} \langle \uparrow \rangle \langle 0|_{\mu} + | \downarrow \rangle \langle 1|_{\mu} \otimes \langle 2222222|_{\mu} \otimes \sum_{\mu' \neq \mu=r,u,l,d} g_{\mu\nu}(\langle \uparrow \rangle \langle 0|_{\mu} + | \downarrow \rangle \langle 1|_{\mu} \otimes \langle 2222222|_{\mu'\nu}), \quad (16)$$

$$|S_2\rangle = |\epsilon\rangle_{\mu} \otimes |22\rangle_{\mu\nu} \pm |\epsilon\rangle_{\mu} \otimes |\epsilon\rangle_{\mu\nu} \pm |2222\rangle_{\mu\mu'\nu}, \quad (17)$$

$$|S_3\rangle = |\epsilon\rangle_{\mu} \otimes |22\rangle_{\mu\nu} \pm |\delta\rangle_{\mu} \otimes |\delta\rangle_{\mu\nu} \pm |2222\rangle_{\mu\mu'\nu}, \quad (18)$$

where $|\epsilon\rangle = |01\rangle - |10\rangle$, $|\delta\rangle = |00\rangle + |11\rangle$, $\mu, \nu'$ corresponds to the bottom and top virtual spins of the same bond, the coefficients $g_{\mu\nu}$ in projector $P_2$ serve as the variational parameters. Imposing rotation symmetry, there are three parameters in $g_{\mu\nu}$ that one can vary, and denoted as $g_\circ$, $g_\circ$, and $g_-$. If arranging the virtual index $r, u, l, d$ as 0, 1, 2, 3 respectively, they are defined as

$$g_{\mu\nu} = \begin{cases} 
  g_\circ & \text{if } \nu = (\mu + 1)\%4, \\
  g_\circ & \text{if } \nu = (\mu + 1)\%4, \\
  g_- & \text{if } \nu = (\mu + 2)\%4.
\end{cases} \quad (20)$$
Now we can establish the relation between the trial PEPS states and wavefunction as

$$\begin{align*}
|b_1| &= g_2^2 \\
|b_2| &= g_2^2 = g_2^2 \\
|b_3| &= g_2 g_2 = g_2 g_2 \\
|b_4| &= g_2 g_2 = g_2 g_2 \\
\end{align*}$$

and the sign of different shapes are taken care of by the ± in front of the second term in $S_2$ and $S_3$. In this trial PEPS wavefunction, variable group $b$ and $c$ do not appear simultaneously. Other than that, there are correlations between $b_{1-4}$ or $c_{1-4}$ as in Eq. (A.1) and Eq. (A.2). To remove that correlation, one can introduce one variable at a time, or use multiple direct sum of virtual spin $\frac{1}{2}$ subspaces for the top virtual spin $\mu'$ and each designate for one variable, i.e. taking a virtual space of $(\frac{1}{2} + 0) \otimes (\frac{1}{2} + \frac{1}{2} + \cdots + 0)$. The wavefunction $\prod \mathcal{P}_2 \prod |S_2\rangle$ and $\prod \mathcal{P}_2 \prod |S_3\rangle$ capture the CRVB wavefunctions with only one variational parameter. There is one more difference between wavefunction with the trial PEPS: higher order terms (in terms of $g_{\mu \nu}$) appear in the trial PEPS but do not appear in wavefunction, however the relative weights of these terms are small given $g_{\mu \nu} < 1$. Examples of the forth order terms in $g_{\mu \nu}$ are drawn in Fig. 7(b,c).

Now we give an example of a projector $\mathcal{P}_3$ and bond matrices $S_4$ to generate a trial PEPS that has two kinds of correlated VB pair (two shapes) as appearing in wavefunction (A.2) (suppose one parameter from $b_i$ and one from $c_i$). In order to do that, we take a virtual spin space of $(\frac{1}{2} + 0) \otimes (\frac{1}{2} + \frac{1}{2} + 0)$, and use labels $\mu, \nu$ to denote the bottom layer virtual spins, each of which spans a space of $\{0,1,2\}$, and $\mu', \nu'$ to denote the corresponding top layer virtual spins, each of which spans a space of $\{0,1,2,3,4\}$. $\mu, \nu$ runs over indices $l, r, u, d$ indicating left, right, up and down bond. The projector and bond matrix are written as

$$\mathcal{P}_3 = \sum_{\mu, \nu = r, u, d} \langle \uparrow | \langle 0 | \mu + \downarrow \rangle \langle 1 | \mu \rangle \otimes \langle 222 | \mu \otimes \langle 444 |$$

$$\mathcal{P}_3 = \sum_{\mu, \nu = r, u, d} g_{\mu \nu} \langle \uparrow | \langle 0 | \mu + \downarrow \rangle \langle 1 | \mu \rangle \otimes \langle 22 | \mu \otimes \langle 44 | \mu \rangle$$

$$\mathcal{P}_3 = \sum_{\mu, \nu = r, u, d} g'_{\mu \nu} \langle \uparrow | \langle 0 | \mu + \downarrow \rangle \langle 1 | \mu \rangle \otimes \langle 22 | \mu \otimes \langle 44 | \mu \rangle$$

$$\mathcal{P}_3 = \sum_{\mu, \nu = r, u, d} g_{\mu \nu} \langle \uparrow | \langle 0 | \mu + \downarrow \rangle \langle 1 | \mu \rangle \otimes \langle 22 | \mu \otimes \langle 44 | \mu \rangle$$

Hamiltonian projection and 1-step Lanczos optimization of the CRVB wavefunction

Suppose the trial wavefunction is sufficiently close to the ground state of a microscopic Hamiltonian. Applying a few steps of Hamiltonian projection will improve the observed ground state energy. The projection can be evaluated stochastically,

$$|\Psi_n\rangle = (J_1 \sum_{ij} C_{ij} + J_2 \sum_{ij} C_{ij})^n |\Psi_0\rangle = \sum_{\{C_{ij}\}} J_1^n g^{n_2} \prod_{p=1}^n C_{i(p)j(p)} |\Psi_0\rangle,$$

here $C_{ij} = \frac{1}{\mathbf{S}_i \cdot \mathbf{S}_j}$, $g = J_2/J_1$, $n$ is the number of projection steps, $n_2$ is the number of the NNN operators in expansion series $\prod_{p=1}^n C_{i(p)j(p)}$, and the summation runs over all projector series $\{C_{ij}\}$. Let us expand the CRVB wavefunction $|\Psi_0\rangle$ as

$$|\Psi_0\rangle = \sum_{\alpha} \prod_{i=1}^4 a_{\alpha} b_{\alpha}$$

$$|\Psi_0\rangle = \sum_{\alpha} \prod_{i=1}^4 b_{\alpha} a_{\alpha}$$

$$|\Psi_0\rangle = \sum_{\alpha} \psi(\alpha)|V_\alpha\rangle,$$
Here is consistent with that appeared in Fig. 6.

![Diagrams around text](image)

**FIG. 7:** A demonstration of how the higher order term (in $g_{\alpha\nu}$) is generated in trial PEPS wavefunctions. The notation here is consistent with that appeared in Fig. 6.

$n_{b_i}$ ($n_{c_i}$) are the number of $b_i$ ($c_i$) type of local resonating pairs (shapes as in Fig. 2b, c) in the realization $V_{\alpha}$. Note that $V_{\alpha}$ is a configuration in the extended VB basis. We now plug Eq. (26) into Eq. (25) and apply the following valence bond evolution relations,

$$C_{ij}(i, j) = (i, j),$$

$$C_{jk}(i, j)(k, l) = \frac{1}{2}(k, j)(i, l),$$

$$C_{ik}(i, j)(k, l) = \frac{1}{2}(i, k)(j, l),$$

the expectation value of any observable $\hat{O}$ is

$$\frac{\langle \Psi_{\alpha} | \hat{O} | \Psi_{\alpha} \rangle}{\langle \Psi_{\alpha} | \Psi_{\alpha} \rangle} = \sum_\alpha \sum_\beta \sum_\{C_{ij}^\alpha\} W_{\alpha\alpha'} W_{\beta\beta'} \psi(\alpha) \psi(\beta) \langle V_{\beta} | \hat{O} | V_{\alpha'} \rangle,$$

(30)

here $J_1^a g_{\alpha\nu} \prod_{p=1}^n C_{i(p)j(p)} | V_{\alpha} \rangle = W_{\alpha\alpha'} | V_{\alpha'} \rangle$, $\{C_{ij}^\alpha\}$ and $\{C_{ij}^\beta\}$ denote the right and left projector series $\{C_{ij}\}$.

1-step Lanczos optimization is an energy minimization procedure in a 2-vector space $\{\langle \Psi_0 |, H | \Psi_0 \rangle\}$. The goal is to find a variational parameter $\alpha$ which minimize the energy of the combined wavefunction

$$| \tilde{\Psi} \rangle = | \Psi_0 \rangle + \alpha H | \Psi_0 \rangle.$$  

(31)

The energy expectation value of wavefunction (31) is

$$\langle E \rangle = \langle \Psi_0 | + \alpha \langle \Psi_0 | H | \Psi_0 \rangle \rangle \langle | \Psi_0 \rangle + \alpha H | \Psi_0 \rangle \rangle = H_1 + 2\alpha H_2 + \alpha^2 H_3 \frac{1 + 2\alpha H_1 + \alpha^2 H_2}{},$$

(32)

Here $H_n = \frac{\langle \Psi_0 | H^n | \Psi_0 \rangle}{\langle \Psi_0 | \Psi_0 \rangle}$. The optimized parameter $\alpha$ is given by zeros of $\frac{\partial \langle E \rangle}{\partial \alpha} = 0$, from which we obtained the following quadratic equation for $\alpha$,

$$(H_1 H_3 - H_2 H_2)\alpha^2 + (H_3 - H_1 H_2)\alpha + (H_2 - H_1 H_1) = 0.$$  

(33)

For each Hamiltonian projected wavefunction $| \Psi_m \rangle = H^m | \Psi_0 \rangle$, we make measurement $\frac{\langle \Psi_m | H^m | \Psi_m \rangle}{\langle \Psi_m | \Psi_m \rangle}$ for $m = 1, 2$, thus $H_3$ is obtained via a combined way

$$\frac{\langle \Psi_0 | H^3 | \Psi_0 \rangle}{\langle \Psi_0 | \Psi_0 \rangle} = \frac{\langle \Psi_1 | H | \Psi_1 \rangle}{\langle \Psi_1 | \Psi_1 \rangle} \frac{\langle \Psi_0 | H^2 | \Psi_0 \rangle}{\langle \Psi_0 | \Psi_0 \rangle}.$$  

(34)