Variational Monte Carlo method for shell-model calculations in odd-mass nuclei and restoration of symmetry

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We investigate two kinds of extensions for the variational Monte Carlo (VMC) method with the Pfaffian in the nuclear shell-model calculations. One is the extension to odd-mass nuclei, for which we find a new Pfaffian expression of the VMC matrix elements. We can, thereby, give a unified VMC framework both for even and odd mass nuclei. The other is the extension of the variation after angular-momentum projection. We successfully implement the full angular-momentum projected trial state into the VMC method, which can provide us with the precise yrast energies. We also find a unique characteristic, namely that this angular-momentum projection in the VMC can be even "approximately" performed. This characteristic is useful not only for efficient computation but also for precise estimation of the yrast energies through the energy-variance extrapolation.

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I. INTRODUCTION

Variational Monte Carlo is one of the quantum Monte Carlo methods to solve quantum many-body problems numerically. While it is a variational method and the precision of the approximation depends on the quality of the trial wave function and the Hamiltonian, it is applicable to any Hamiltonian without the notorious sign problem. Therefore, it has been intensively developed in various fields, such as condensed matter physics and nuclear physics. Especially, the advent of the stochastic reconfiguration (SR) method enables us to use a large number of variational parameters efficiently. Moreover, as a trial state, a particle-number-projected Hartree-Fock-Bogoliubov (HFB) wave function can be used owing to the Pfaffian, which is known to provide us with the compact and computationally effective wave function. This recent progress broadens the applicability to the configuration-space method, such as the Hubbard model.

In nuclear physics, the large-scale shell model (LSSM) calculation is one of the configuration-space methods and a powerful model to describe the nuclear spectroscopic information precisely. However, the number of the many-body configurations which appear in the LSSM tends to be huge, and the dimension of the Hamiltonian matrix to be diagonalized often surpasses the capability of the state-of-the-art supercomputers. In order to avoid this problem and to describe the shell-model wave function in a sophisticated form, the pair-correlated wave function, or the HFB-type wave function, was suggested in the VAMPIR method. However, the HFB wave function is awkward for treating odd-mass system. We have proposed a new formulation of the variational Monte Carlo (VMC) method for shell model calculations for even-mass nuclei and demonstrated its feasibility for LSSM calculations.

In the present paper, we address two kinds of extensions of the previously-presented VMC method. One extension is to handle odd-mass nuclei in the framework of VMC by a new Pfaffian expression. We present the common VMC framework both for even and odd mass nuclei. The other extension is the implementation of the variation after angular-momentum projection. Since the atomic nucleus is an isolated system, the restoration of symmetry is crucial for the nuclear structure calculations. We successfully implement the trial state with full angular-momentum projection into the VMC method. Unlike other applications of angular-momentum projection, we find a unique characteristic that full angular momentum projection in the VMC can be performed “approximately”. This characteristic is useful not only for efficient computation but also for precise estimation of the yrast energies through the energy-variance extrapolation. In the condensed matter physics, the projection method was introduced in, e.g., Ref. and it was also introduced into the VMC in Ref. The projection method is well-known, but this implementation of the VMC is more flexible than the preceding works. It may be useful to other fields of physics.

This paper is organized as follows: Section is devoted to explaining the theoretical framework of the VMC method and its extension to odd-mass nuclei. The numerical results and “approximate” projection are discussed in Sect. The summary is given in Sect. IV.
II. FORMULATION OF THE VMC

In this section, we briefly describe the formulation of the VMC. We introduce a trial wave function in the subsection II.A and describe the way how to stochastically evaluate the energy expectation value of the wave function in the framework of the Monte Carlo in subsection II.B. The restoration of rotational symmetry by the projection method in the VMC is summarized in subsection II.C. The variational parameters are determined so that the energy is minimized utilizing the SR method, the details of which are given in appendix C.

A. Trial wave function

As a trial wave function for nuclei with N valence particles for the present VMC, we take $|\psi\rangle$ as

$$|\psi\rangle = GP|\phi\rangle,$$

where the $|\phi\rangle$ is a pair-correlated wave function and $P$ is a projection operator, both of which are discussed later. The operator $G$ is the Gutzwiller-like factor as

$$G = e^{\sum_{i<j} a_{ij} n_i n_j}$$

where $n_i$ is the number operator of the single-particle orbit $i$ and $a_{ij}$'s are variational parameters.

For even-mass nuclei, the $|\phi\rangle$ is defined as

$$|\phi\rangle = \left(\sum_{kk'} f_{kk'} c^\dagger_{k'} c_{k}\right)^{N/2} |\rangle$$

where $f$ is a skew-symmetric matrix, $f_{kk'} = -f_{k'k}$, the matrix elements of which are variational parameters. The $|\rangle$ is an inert core and the $c^\dagger_{k}$'s are proton or neutron creation operator of the single-particle state $i$. It corresponds to the number projected Hartree-Fock-Bogoliubov wave function [15], which is advantageous for the description of pairing correlations. Note that this wave function contains the proton-neutron pairing correlations in addition to the proton-proton and neutron-neutron pairing correlations, while the usual HFB method does not include proton-neutron pairing correlations. It plays a crucial role in understanding the nuclear structure of $N = Z$ nuclei [16, 17].

For odd-mass nuclei, we extend the trial wave function $|\phi\rangle$, which is defined as

$$|\phi\rangle = \left(\sum_{i} h_i c^\dagger_{i}\right) \left(\sum_{kk'} f_{kk'} c^\dagger_{k'} c_{k}\right)^{(N-1)/2} |\rangle,$$

where the $h_i$ are additional variational parameters. This form is the simplest for odd-mass nuclei. Hereafter we discuss the VMC formalism for the odd-mass cases. The formulation of the even-mass case can be seen in Ref. [6] and is also obtained easily by omitting the terms containing the $h_i$ parameters in the following formulations, that is, we can give a unified description with this trial wave function for even and odd mass nuclei.

The projection operator $P$ serves to restore the rotational symmetry, parity symmetry and $z$-component of isospin such as

$$P = P_T P^\pi P_M \tag{5}$$

where $P_T$, $P^\pi$ and $P_M$ are projectors of the $z$-component of the isospin, the parity $\pi$, and the total angular momentum $(I,M)$, respectively. The angular momentum operator is decomposed into the $\langle J_z \rangle = M$ projection and the rest such as

$$P_M^I = P_M \tilde{P}_M^I \tag{6}$$

where

$$\tilde{P}_M^I \equiv \frac{2I + 1}{4\pi} \sum_{K'=1}^{I} g_K \int d\gamma d\beta \sin \beta d_{MK}^{K}(\beta) e^{-iK\gamma} e^{iJ_\beta} e^{iJ_\gamma}.$$

The $d_{MK}^{K}(\beta)$ is Wigner’s $d$-function and $g_K$ denotes the $2I + 1$ variational parameters.

B. Markov Chain Monte Carlo

We describe how to estimate the energy expectation value of the trial wave function. First of all, the projection operator of the $z$-component of isospin, parity, and $z$-component of angular momentum is expressed as a linear combination of the complete set in the $m$-scheme basis states as

$$P_M^I P^\pi P^T = \sum_{m \in \{M^\pi\}} |m\rangle \langle m| \tag{8}$$

where the $m$-scheme basis state $|m\rangle$ is defined as

$$|m\rangle = c^\dagger_{m_1} c^\dagger_{m_2} \cdots c^\dagger_{m_N} |\rangle$$

which is parametrized by a set of occupied single-particle states, $m = \{m_1, m_2, \ldots, m_N\}$. The $\sum_{m \in M^\pi}$ denotes the summation of any $|m\rangle$ in the subspace with $J_z = M$ and $\pi$-parity. It is convenient to take $M = I$, especially for the yrast states.

The energy expectation value is obtained as

$$\langle H \rangle = \frac{1}{\sum_{m \in M^\pi} |\langle m|\psi\rangle|^2} \sum_{m \in M^\pi} |\langle m|\psi\rangle|^2 \langle m|H|\psi\rangle \langle m|\psi\rangle \tag{9} = \sum_{m \in M^\pi} p(m) E_l(m)$$

where $p(m)$ is defined as $p(m) = |\langle m|\psi\rangle|^2 / \sum_{m} |\langle m|\psi\rangle|^2$. $E_l(m)$ is called the local energy and defined as

$$E_l(m) = \frac{\langle m|H|\psi\rangle}{|\langle m|\psi\rangle|^2} \tag{10} = \frac{1}{|\langle m|\psi\rangle|^2} \sum_{m' \in M^\pi} \langle m|H|m'\rangle |\langle m'|\psi\rangle|^2.$$
where the matrix \( H_{mm'} = \langle m | H | m' \rangle \) is sparse and the summation concerning \( m' \) can be computed efficiently since the shell-model Hamiltonian \( H \) is a two-body interaction and has good parity and rotational symmetries.

The weighted summation \( \sum_m p(m) \) in Eq. (10) is estimated stochastically using the Markov Chain Monte Carlo (MCMC) method in which \( |m⟩ \) walks randomly in the \( \{ M^\pi \} \) subspace obeying the probability \( p(m) \). Such random walker of the \( m \) scheme basis state was adopted also in Refs. [6, 18, 19]. The energy gradient and the overlap matrix are also estimated stochastically by the SR method.

The overlap between the \( m \)-scheme basis state and the \( |\psi⟩ \) is shown by

\[
\langle m | \psi \rangle = G(m) \langle m | P | \phi \rangle \tag{12}
\]

with \( G(m) = G(m) |m⟩ \). Note that \( G \) is a diagonal operator for the \( m \)-scheme basis representation and is commutable with the projection operator \( P \). This factor usually accelerates the convergence of the SR iterations. While this operator can include many-body correlation beyond the mean-field and pairing correlations, its contribution to the energy gain is limited in the case of shell-model calculations. The projected overlap, \( \langle m | P | \phi \rangle \), is discussed in the following subsection.

### C. Angular-momentum projection

The projected overlap \( \langle m | P | \phi \rangle \) is evaluated as

\[
\langle m | P | \phi \rangle = \langle m | \tilde{P}_M^I | \phi \rangle = \frac{2I + 1}{4\pi} \int d(cos \beta) d(\gamma) d^4 z |I, \gamma \rangle \langle I, \beta, \gamma | \phi \rangle \tag{13}
\]

\[
\approx \frac{2I + 1}{4\pi} \sum_K g_K \sum_a u_a(\beta) e^{-iK \gamma} \sum_b u_b(y) \langle I | \tilde{P}_M^I | \phi \rangle,
\]

where the integrals over \( \cos \beta \) and \( \gamma \) are numerically approximated by weighted sums. The points \( (\gamma_b, \beta_a) \) and its weight factors \( (u_a(\beta), u_b(y)) \) are determined by the trapezoidal rule for \( \gamma \) and the Gauss-Legendre quadrature for \( \beta \). The number of the points for integrals, \( N_z \) and \( N_y \), is usually determined to be large enough to evaluate the correct expectation value of \( J^2 \). The numbers are taken typically as \( (N_z, N_y) = (32, 16) \). The rotation of the correlated-pair wave function \( |\phi⟩ \) is evaluated as

\[
R(\beta, \gamma)|\phi⟩ = e^{iJ_y b_k} e^{iJ_z \gamma \alpha_i} |\phi⟩ \tag{14}
\]

\[
= \left( \sum_i h_i c_i^+ \right) \left( \sum_{kk'} f_{kk'} c_{k'}^+ \right) |\phi⟩ \sim (N-1)/2.
\]

with \( h' = Rh, f' = RfR^T \). The rotation matrix \( R \) is defined as \( R = e^{iJ_y b_k} e^{iJ_z \gamma} \). Thus the rotated wave function is kept of the same form thanks to the Baker-Campbell-Hausdorff theorem [21].

In this paper, we find that the overlap between this form of the wave function \( |\phi⟩ \) and the \( m \)-scheme basis state can be written using the single Pfaffian. This is shown in Appendix [A].

The variational parameters \( h, f, \) and \( g \) are determined so that the energy is minimized utilizing the SR method. In this paper, we show that the angular-momentum projected energy can be minimized in the VAP framework of the VMC, while the unprojected energy is also minimized to determine the wave function and the projected energy can be evaluated in the variation-before-projection (VBP) framework [6]. In the VMC approach, “unprojected” means without full-angular-momentum projector \( P^I_M \), but with the \( J_z \), parity, and \( T_z \) projections.

### III. NUMERICAL RESULTS

We discuss the VMC results with variation after angular-momentum projection (J-VAP) in the even-mass case in subsection [III A] and the odd-mass case in subsection [III B]. The J-VAP calculation can give better yrast energies than those of our previous paper [6], while it requires a more substantial computational cost. In subsection [III C] the “approximation” scheme of angular-momentum projection is introduced to reduce the computational cost. We show that this “approximation” scheme can give a sequence of wave functions, which can be useful for the extrapolation using the energy variance. With the energy variance extrapolation, the exact yrast energies can be computed beyond the limitation of the trial wave function.

#### A. Variation after projection for even-mass nuclei

In this subsection, we demonstrate the VAP calculation with the variation after angular-momentum projection of \( ^{58} \text{Cr} \) in the \( pf \) shell. The GXPF1A interaction is adopted as an effective interaction [22]. For the test of VMC calculation, we use a realistic residual interaction, not a schematic interaction so as to properly judge the feasibility of the method.

Figure [I] shows the convergence of the VMC energy with full angular momentum projection, which is called J-VAP VMC energy later, as a function of the number of the iterations of the SR method. The MCMC procedure generates eight random walkers with 8000 steps with the Gibbs sampler, the details of which are shown in Ref. [6]. This step needs two-fold integration over Euler’s angle as in Eq. (13), which needs heavy numerical computation. The present VMC calculations cost a few hours typically on a PC server with 56 CPU cores. We will show how to reduce the computation later.

The convergence of the J-VAP VMC energies is almost achieved with up to 50 ~ 60 steps. Since the Monte Carlo (MCMC) method in which |m⟩ walks randomly in the {Mπ} subspace obeying the probability p(m). Such random walker of the m scheme basis state was adopted also in Refs. [6, 18, 19]. The energy gradient and the overlap matrix are also estimated stochastically by the SR method.
Carlo error of the energy is typically 2 keV and small enough, the error bars are omitted for simplicity in the figure. The J-VAP VMC energy converges well and close enough to the exact shell-model energies within 160 keV from $0^+$ to $12^+$ states. For comparison, we show the VBP energy as the rightmost levels in the figure. The VMC with VAP improves the energy over VBP as expected. Especially the VBP result underestimates the $2^+$ excitation energy, while the VAP result sufficiently reproduces the exact energies including the isoscalar-pairing correlations. The small energy differences between the exact energies and J-VAP VMC ones will be discussed in subsection III D.

In this subsection, we consider the odd-mass nuclei for a test of the new trial wave function. We calculate the yrast energies of $^{49}$Cr within the $pf$-shell model space and the GXPF1A interaction \cite{22}. In this VMC calculations, we apply the full angular momentum projection to the trial state. In the MCMC process, we adopt the Gibbs sampler with 640 random walkers, each of which contains 500 sample steps after 100 burn-in steps. In order to suppress the biases induced by the initial state of the Markov Chain, we take the last sample of the previous SR iteration as an initial sample of the MCMC process.

Figure 2 shows the convergence of the J-VAP VMC energy of $^{49}$Cr as an example of odd-mass nuclei. The energies of the yrast states $5/2^-$, $7/2^-$, $9/2^-$, and $11/2^-$ are shown in the figure. The difference between the converged energy and the exact one is similar to the one of the even case, which means that our trial wave function Eq.(4) is considerably more proper. However, the number of iterations of the odd case is larger than the one of the even case.

**C. Approximate angular-momentum projection**

Since the correlated-pair wave function $|\psi\rangle$ does not have good rotational and parity symmetries, the solution spontaneously breaks these symmetries and it is crucial to restoring them by the projection method. In general, the J-VAP has a large effect to minimize the energy in the context of the configuration-interaction approach. Various variational calculation after the angular-momentum projection have been, therefore, proposed such as the Monte Carlo shell model \cite{29}, the VAMPIR approach \cite{10}, and the hybrid multideterminant method \cite{24}.

In these J-VAP calculations, since the energy and the energy gradient are computed under the mathematical conditions $[H, P_{MK}^L] = 0$ and $P_{MK}^L P_{L/K}^L = \delta_{L/L'} P_{MK}^L$, the high-precision numerical evaluation of the projection is essential. The insufficient number of points for the integral of the Euler angles causes numerical instability and the angular momentum projection fails in solving the Hill-Wheeler equation. The angular-momentum projection is, therefore, a central bottleneck of the computation.

![FIG. 1: Convergence of energies of $I^n = 0^+, 2^+, 4^+, 6^+, 8^+$ and $12^+$ states of $^{48}$Cr obtained by the J-VAP VMC as functions of the number of the SR iterations. The right column shows the VAP results, exact shell-model energies, and the VBP results.](image1)

![FIG. 2: Convergence of the J-VAP VMC energies of $^{49}$Cr with GXPF1A interaction. The black circles, blue diamonds, orange triangles, and green reverse triangles denote the energy expectation values of $5/2^-$, $7/2^-$, $9/2^-$, $11/2^-$, and $12^+$ states, respectively, as functions of the number of iterations. The exact shell-model energies are shown as the rightmost red triangles.](image2)
of various variational approaches to the nuclear quantum many-body solver [23].

On the other hand, in the VMC formalism, since the conditions \([H, P_{MK}] = 0\) and \(P_{MK}P_{MK} = \delta_{LL}P_{MK}\) are not adopted, high precision calculations for the angular-momentum projection \(P_M^I\) is not necessarily needed, which means that the number of mesh points for numerical integration could be reduced. In fact, even when we use a small number of points for the integrals and the operator \(P_M^I\) is mathematically no longer valid as a projection operator, the \(P_M^I|\phi\rangle\) works as a trial wave function with “approximated” angular momentum, because this wave function is simply a superposition of the rotated wave functions of \(|\phi\rangle\) with appropriate weight coefficients as

\[
P_M^I|\phi\rangle \simeq \sum_{a=1}^{N_a} \sum_{b=1}^{N_b} w_a^{(z)} w_b^{(y)} R(\beta_b, \gamma_a)|\phi\rangle.
\]  

Therefore, as an approximation to the projection operator, we introduce the \(P_M^I\) with a set of the small numbers of \(N_a\) and \(N_b\) and call it \(\tilde{P}_M^I\) hereafter. Note that this \(\tilde{P}_M^I\) is still commutable with the operator \(G\) for any \((N_a, N_b)\).

\[
\begin{align*}
\text{FIG. 3: VMC results with the variation after approximate angular-momentum projection against the total number of the mesh points for the integral, } N_a N_b. \text{ The black circles, squares, and triangles denote the converged results of the } I^z = 0^+, 2^+, 4^+, \text{ and } 6^+ \text{ states of } ^{48}\text{Cr, respectively, with the GXPF1A interaction [24]. The orange symbols denote the full-projected energy of the resultant wave function. These symbols are connected with the dotted lines to guide the eyes. See text for further details.}
\end{align*}
\]

Figure 3 shows the converged VMC energies of the \(0^+, 2^+\) and \(4^+\) states for \(^{48}\text{Cr}\) with the GXPF1A interaction [24] as functions of the number of points for the integral of the projection operator \(\tilde{P}_M^I\). The VMC calculation was performed with variation after the \(\tilde{P}_M^I\) projection. The number of the points is taken as \((N_a, N_b) = (1, 1), (2, 1), (4, 2), (6, 3), (8, 4), (10, 5), \text{ and } (21, 11)\). The converged energies of the variation after the approximated projection are shown as the black symbols in Fig. 3. The case of \((N_a, N_b) = (1, 1)\) corresponds to the variation without the angular-momentum projection. In the figure, the rightmost red triangles denote the exact shell-model energies. The VMC results well reproduce the exact one even with the small number of \(N_a N_b\). In order to improve the precision of the angular-momentum projection so that the expectation value of \(J^2\) equals \(I(I+1)\) exactly to 6 decimal digits, the necessary number of points is higher than the minimal one given by \((N_a, N_b) = (28, 14), (28, 14), (31, 16), \text{ and } (35, 18)\) for \(I^z = 0^+, 2^+, 4^+, \text{ and } 6^+\) states, respectively.

Astonishingly, the approximated projection works well even for \((N_a, N_b) = (6, 3)\). The total number of the points, \(N_a N_b\), is almost proportional to the amount of computations of the projected matrix elements, which is the most time-consuming part of the VMC calculations. Therefore, the computation time is dramatically reduced in comparison with the full projection. The required number of the points is rather constant as a function of the angular momentum \(I\), while in the case of the full angular-momentum projection the necessary number of points increases as \(I\) does. However, we should mention that the numerical calculation is stable as far as no higher spin state exists in the lower-energy region than the target state.

Moreover, we apply the \(\tilde{P}_M^I\) projection, in which the \(N_a N_b\) is large enough to obtain the correct expectation value of \(J^2\) to the resultant wave functions. The orange symbols in Fig. 3 denote the “full” angular-momentum projected energies. It is considered to be the variation after the approximated projection before the full projection. These energies are quite close to those of the variation after full projection. In practice the energies obtained by the \((N_a, N_b) = (6, 3)\) variation agree with those of the J-VAP VMC within 70-keV difference.

\section*{E. Energy-variance extrapolation}

As the VMC is a variational method, it must not necessarily give us exact energies. The obtained energy is an upper limit. To know the exact energy, one useful method is energy-variance extrapolation [7, 26–29], which uses a series of the well-approximated wave functions \(|\psi_1\rangle, |\psi_2\rangle, \cdots\) with monotonically decreasing energies \(|\langle \psi_1 | H | \psi_1 \rangle| > |\langle \psi_2 | H | \psi_2 \rangle| > \cdots\). By evaluating the energy variance as \(|\Delta H^2| = |\langle H^2 \rangle - \langle H \rangle^2|\) for each wave function, we can show a linear or quadratic relation between the energy variances and the energies and show that the energy approaches the exact energy along the sequence. By fitting a second-order polynomial for data points of energy variance and energy, the exact energy can be expected by extrapolating the energy to the limit of \(|\Delta H^2| = 0\).

In the preceding application of the energy-variance extrapolation to the nuclear shell model, we used the trun-
to variation after angular-momentum projection, which applied to odd-mass nuclei. We also extended the VMC element. We demonstrated that the VMC is successfully extended the previous VMC method for odd-mass nuclei, angular momentum projection. In the present paper, we extended only even-mass nuclei and variation before angular-momentum projection method also provides us with a sequence of well-approximated wave functions by changing the number of points for the projection are taken as \((N_x, N_y) = (2, 1), (4, 2), (6, 3), (8, 4), (10, 5), \) and \((21, 11)\). The red lines are chi-square fitted to the symbols. The red squares on the y-axis are the exact shell-model energies.

**FIG. 4:** Energy variance extrapolation by the variation after the approximate angular-momentum projection. The \(0^+, 2^+, 4^+, \) and \(6^+ \) shell-model energies of \(^{48}\text{Cr}\) are obtained with the GXPF1A interaction \([22]\). The energy expectation values against the energy variance are plotted as the black symbols with the approximate projection. The numbers of the points for the projection are taken as \((N_x, N_y) = (2, 1), (4, 2), (6, 3), (8, 4), (10, 5), \) and \((21, 11)\). The red lines are chi-square fitted to the symbols. The red squares on the y-axis are the exact shell-model energies.

**IV. SUMMARY**

We presented the VMC method with the Pfaffian to solve the nuclear shell model in Ref. \([6]\), where we handle only even-mass nuclei and variation before angular-momentum projection. In the present paper, we extended the previous VMC method for odd-mass nuclei, by deriving a new Pfaffian expression for the VMC matrix element. We demonstrated that the VMC is successfully applied to odd-mass nuclei. We also extended the VMC to variation after angular-momentum projection, which enhances the quality of the VMC energy.

In addition to these extensions, we also found that the “approximated” angular-momentum projection can work in the VMC framework. So far, no feasible approximation scheme for full angular momentum projection has been presented, and its numerical calculations have been believed to be performed in quite a strict manner. However, we proposed a novel approximation scheme of angular momentum projection, which reduces the computation drastically and brings about an efficient way to calculate angular momentum projection.

Furthermore, we found that this “approximated” angular-momentum projection also gives a series of well-approximated wave functions, which is useful to the energy variance extrapolation. By this development, we could estimate the exact energies of the shell model beyond the limitation of the VMC.

The form of the trial wave function can be straightforwardly extended to that of a one-broken-pair state, which is used in Tamm-Dancoff approximation and shown in Appendix \([33]\). Its numerical application remains as a future subject.

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**Appendix A: Pfaffian and its relevant formulas**

The Pfaffian plays the main role to evaluate the matrix elements which appear in the present VMC formalism. Some useful formulas relevant to the Pfaffian are given in this appendix. The Pfaffian of a \(2n \times 2n\) skew-symmetric matrix \(A\) is defined as

\[
Pf(A) = \frac{1}{2^n n!} \sum_{\sigma \in S_{2n}} \text{sgn}(\sigma) \prod_{i=1}^{n} A_{\sigma(2i-1)\sigma(2i)}
\]

\[
= \frac{1}{n!} \sum_{\sigma \in S_{2n} \backslash \{\sigma(2i-1) < \sigma(2i)\} \subset \{1, 2, 3, \ldots, 2n\}} \text{sgn}(\sigma) \prod_{i=1}^{n} A_{\sigma(2i-1)\sigma(2i)}
\]

where \(\sigma\) is a permutation of \(\{1, 2, 3, \ldots, 2n\}\), \(\text{sgn}(\sigma)\) is its sign, and \(S_{2n}\) is a group of the permutations.

For preparation, the recursive relation of Pfaffian is

\[
Pf(A) = \frac{1}{2^n n!} \sum_{\sigma \in S_{2n}} \text{sgn}(\sigma) \prod_{i=1}^{n} A_{\sigma(2i-1)\sigma(2i)}
\]
given as
\[ \text{Pf}(A) = \sum_{j=1}^{2n} (-1)^{i+j+1+\theta(i-j)} A_{ij} \text{Pf}(A_{ij}^{-1}) \] (A2)

where \( A_{ij}^{-1} \) denotes the matrix \( A \) with the \( i \)-th and \( j \)-th columns and rows removed, \( \theta(i-j) \) is the Heaviside step function. Its special case with \( i = 1 \) is written as
\[ \text{Pf}(A) = \sum_{j=1}^{2n} (-1)^{j} A_{ij} \text{Pf}(A_{ij}^{-1}). \] (A3)

The differentiation of the Pfaffian is given by
\[ \frac{\partial}{\partial A_{ij}} \text{Pf}(A) = -\text{Pf}(A)(A^{-1})_{ij}. \] (A4)

**Appendix B: Overlap with the trial wave function and \( m \)-scheme basis state**

In the present VMC formalism, the overlap between the trial wave function and the \( m \)-scheme basis state must be computed efficiently. The trial wave function is a product of the Gutzwiller-like operator \( G \) and the pair-correlated wave function. Since the operator \( G \) is diagonal in the \( m \)-scheme basis, the overlap is factorized into the matrix element of \( G \) and the pair-correlated part such as
\[ \langle m|\psi \rangle = G(m)\langle m|\phi \rangle \] (B1)

with
\[ G(m) = \exp \left( \sum_{i<j} \alpha_{ij} n_{i} n_{j} \right) \] (B2)

where \( n_{i} \) is the number operator of the single-particle orbit \( i \) and \( \alpha \)'s are variational parameters. The differential with respect to the variational parameter \( \alpha_{ij} \) is obtained simply as
\[ \frac{1}{\langle m|\psi \rangle} \frac{\partial}{\partial \alpha_{ij}} \langle m|\psi \rangle = n_{i} n_{j}. \] (B3)

The overlap between the pair-correlated wave functions (e.g. Eqs. (3) and (4)) and \( m \)-scheme basis state in Eq. (1) are obtained by using the Pfaffian efficiently. Hereafter we describe the overlap and its derivative concerning the pair-correlated wave functions.

1. **Even-mass nuclei**

It is useful to obtain the overlap between the \( m \)-scheme basis state for the \( 2n \)-valence-particles nuclei in Eq. (3) and the pair-correlated state \( |\phi \rangle \). Using Eq. (A1), it is obtained as
\[ \langle m|\phi \rangle = |m| \left( \sum_{ij} f_{ij} c_{i}^\dagger c_{j}^\dagger \right)^{n-1} |\rangle = n! \text{Pf}(F) \] (B4)

where \( F_{rs} = f_{m_{r}m_{s}} - f_{m_{s}m_{r}} \).

Utilizing Eq. (A3), its differential is obtained as
\[ \frac{1}{\langle m|\psi \rangle} \frac{\partial}{\partial F_{rs}} \langle m|\psi \rangle = -(F^{-1})_{rs}. \] (B5)

2. **Odd-mass nuclei**

The correlated wave function for the odd-mass case is defined in Eq. (1). The number of particles is \( N = 2n-1 \). As a novelty, we show the overlap between this odd wave function and the \( m \)-scheme basis state. Using Eq. (A3), the overlap is obtained as
\[ \langle m|\phi \rangle = \langle m| \left( \sum_{l} h_{l} c_{l}^\dagger \right) \left( \sum_{kk'} f_{kk'} c_{k'}^\dagger c_{k'} \right)^{n-1} |\rangle \]
\[ = n! \text{Pf}(F) \] (B6)

where \( F \) is a \( n \times n \) skew-symmetric matrix and consists of the first row being \( h_{mp} \) and the other being \( f_{ij} = f_{m_{i},m_{j}} - f_{m_{j},m_{i}} \) such as
\[ F = \begin{pmatrix} 0 & h_{m_{1}} & h_{m_{2}} & h_{m_{3}} & \cdots & h_{m_{N}} \\ -h_{m_{1}} & 0 & f_{1,2} & f_{1,3} & \cdots & f_{1,N} \\ -h_{m_{2}} & f_{2,1} & 0 & f_{2,3} & \cdots & f_{2,N} \\ \vdots & \vdots & \vdots & \ddots & \ddots & \vdots \\ -h_{m_{N}} & f_{N,1} & f_{N,2} & f_{N,3} & \cdots & 0 \end{pmatrix}. \] (B7)

Its differentiation is also obtained in a similar manner as in the even-mass case such as
\[ \frac{1}{\langle m|\psi \rangle} \frac{\partial}{\partial h_{m_{k}}} \langle m|\psi \rangle = -(F^{-1})_{1,k+1} \]
\[ \frac{1}{\langle m|\psi \rangle} \frac{\partial}{\partial f_{m_{k},m_{l}}} \langle m|\psi \rangle = -(F^{-1})_{k+1,l+1}. \] (B8)

3. **Tamm-Dancoff wave function**

The wave function used in the Tamm-Dancoff approximation, which is called a one-broken-pair state, is a good approximation to the excited state of the pair-condensed wave function in even-mass nuclei having \( 2n \) valence particles. It can also be used in the VMC formalism, and is defined as
\[ |\phi \rangle = \left( \sum_{l} h_{l} c_{l}^\dagger c_{l}^\dagger \right) \left( \sum_{kk'} f_{kk'} c_{k'}^\dagger c_{k'}^\dagger \right)^{n-1} |\rangle. \] (B9)
Its overlap is obtained using Eq. (A3) as
\[ \langle m| \phi \rangle = \sum_{p,q=1}^{2n} (-1)^{p+q-1} h_{m,m_q} (n-1)! \text{ Pf}(F_{m,m_q}) \]
\[ = \sum_{p=1}^{2n} (-1)^p (n-1)! \text{ Pf}(F_{p}). \] (B10)

The overlap is defined as \( F_{1,1} = 0 \), \( F_{1,r+1} = h_{m,m_r} \), and \( F_{r+1,s+1} = \tilde{f}_{r,s} = f_{m_r,m_s} - f_{m_s,m_r} \) with \( r, s \neq p \) such as
\[ F_{p} = \begin{pmatrix} 0 & h_{m_p,m_1} & h_{m_p,m_2} & h_{m_p,m_3} & \cdots & (p) & h_{m_p,m_2n} \\ -h_{m_p,m_1} & 0 & f_{1,2} & f_{1,3} & \cdots & (p) & f_{1,2n} \\ -h_{m_p,m_2} & f_{2,1} & 0 & f_{2,3} & \cdots & (p) & f_{2,2n} \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ -h_{m_p,m_{2n}} & \tilde{f}_{2n,1} & \tilde{f}_{2n,2} & \tilde{f}_{2n,3} & \cdots & (p) & 0 \end{pmatrix} \] (B11)
where \( (p) \) denotes that the index \( p \) is skipped. The extension to more-broken-pairs states is also expected.

**Appendix C: Stochastic Reconfiguration**

In the present VMC framework, many variables are optimized simultaneously to minimize the energy expectation values stochastically. Although the stochastic estimation of the gradient vector enables us to use the steepest gradient method, it is unstable due to the stochastic error. In order to stabilize the numerical calculation and to accelerate it, S. Sorrellma introduced the stochastic reconfiguration (SR) method \([4]\). In this appendix, we describe the details of the SR method with variation after the angular-momentum projection.

The angular-momentum projection obliges us to introduce complex numbers as variational parameters, while only real numbers are often used as variational parameters in the preceding works in condensed matter physics (e.g. \([1]\)). Here, we describe the extension of the SR method of the projected wave function including complex numbers as variational parameters.

We define a derivative operator \( O_i \), which is diagonal in the \( m \)-scheme basis states, and its conjugate operator \( O_i^\dagger \) such as
\[ O_i = \sum_{m} |m\rangle \frac{1}{\langle m|\psi_\alpha \rangle} \frac{\partial}{\partial \alpha_i} \langle m|\psi_\alpha \rangle \langle m| \]
\[ = \sum_{m} |m\rangle O_i (m,\alpha) \langle m| \]
\[ O_i^\dagger = \sum_{m} |m\rangle \frac{1}{\langle \psi_\alpha|m\rangle} \frac{\partial}{\partial \alpha_i^*} \langle \psi_\alpha|m\rangle \langle m| \]
\[ = \sum_{m} |m\rangle O_i^\dagger (m,\alpha) \langle m|. \] (C1)

with
\[ O_i (m,\alpha) = \frac{1}{\langle m|\psi_\alpha \rangle} \frac{\partial}{\partial \alpha_i} \langle m|\psi_\alpha \rangle \]
\[ O_i^\dagger (m,\alpha) = \frac{1}{\langle \psi_\alpha|m\rangle} \frac{\partial}{\partial \alpha_i^*} \langle \psi_\alpha|m\rangle, \] (C2)
and \( \alpha \) denotes a set of variational parameters which are complex numbers. In the present work for the odd-mass case, the variational parameters are \( \alpha = \{ g_{K\ell}, \alpha_{ij}, h_1, f_{kk} \} \). These operators satisfy the following derivative equations,
\[ \langle m|O_i |\psi_\alpha \rangle = \frac{\partial}{\partial \alpha_i} \langle m|\psi_\alpha \rangle \]
\[ \langle \psi_\alpha|O_i^\dagger |m\rangle = \frac{\partial}{\partial \alpha_i^*} \langle \psi_\alpha|m\rangle = \langle m|O_i |\psi_\alpha \rangle^*. \] (C3)

The normalized trial wave function is written as
\[ |\overline{\psi}_\alpha \rangle = \frac{1}{\sqrt{\langle \psi_\alpha|\psi_\alpha \rangle}} |\psi_\alpha \rangle. \] (C4)

The derivative of the normalized trial wave function with respect to \( \alpha \) can be written as
\[ \frac{\partial}{\partial \alpha_i} |\overline{\psi}_\alpha \rangle = \left( O_i - \frac{1}{2} \langle O_i | \overline{\psi}_\alpha \rangle \right) |\overline{\psi}_\alpha \rangle \]
\[ = \frac{1}{2} \langle O_i | \overline{\psi}_\alpha \rangle |\overline{\psi}_\alpha \rangle \] (C5)
where we use the shorthand notation \( \langle O \rangle = \langle \psi|O|\psi \rangle \).

The energy gradient \( g_i \) is obtained utilizing these derivative operators as
\[ g_i = \frac{\partial}{\partial \alpha_i} \langle \psi|HO|\psi \rangle \]
\[ = \langle O_i^\dagger |H|\psi \rangle - \langle O_i^\dagger |H \rangle, \] (C6)

We evaluate \( \langle O_i^\dagger \rangle, \langle O_i \rangle, \langle O_i^\dagger O_j \rangle \) and \( \langle O_i^\dagger H \rangle \) stochastically by
\[ \langle O_i^\dagger \rangle = \langle O_i^\dagger |\psi \rangle \langle \psi|\psi \rangle^2 \sum_m \langle m|\psi \rangle^2 \]
\[ = \frac{\sum_m \langle m|\psi \rangle^2 O_i^\dagger (m,\alpha) \sum_m |\langle m|\psi \rangle|^2}{\sum_m |\langle m|\psi \rangle|^2} \]
\[ = \sum_m p(m) O_i^\dagger (m,\alpha) \] (C7)

where \( p(m) \) is defined as \( p(m) = |\langle m|\psi \rangle|^2 / \sum_m' |\langle m'|\psi \rangle|^2 \). The weighted summation \( \sum_p p(m) \) is realized by the Markov Chain Monte Carlo (MCMC) process in which \( |m\rangle \) is generated obeying the probability \( p(m) \). The energy is also evaluated in the same manner as
\[ E_i (m) = \frac{\langle m|H|\psi \rangle}{\langle m|\psi \rangle} \]
\[ = \sum_p p(m) E_i (m), \] (C8)
Other relevant values are evaluated as
\[
\langle O_i \rangle = \sum_m p(m) O_i(m, \alpha) = \langle O_i^\dagger \rangle^* \quad (C9)
\]
\[
\langle O_i^\dagger O_j \rangle = \sum_m p(m) O_i^\dagger(m, \alpha) O_j(m, \alpha). \quad (C10)
\]
\[
\langle O_i^\dagger H \rangle = \sum_m \langle \psi | O_i^\dagger | m \rangle \langle m | H | \psi \rangle \frac{1}{\sum_m |\langle m | \psi \rangle|^2} = \sum_m p(m) O_i^\dagger(m, \alpha) E_L(m), \quad (C11)
\]
\[
\langle H O_i \rangle = \sum_m p(m) E_L^\dagger(m) O_i(m, \alpha). \quad (C12)
\]
The derivative concerning the operator \( G \) is evaluated as
\[
O_{\alpha,i}(m, \alpha) = \frac{1}{\langle m | \psi \rangle} \frac{\partial}{\partial \alpha_i} \langle m | e^{-\sum_i \alpha_i n_i} \psi \rangle = \sum_{i \leq j} n_i^{(m)} n_j^{(m)} \quad (C13)
\]
with \( n_i^{(m)} = \langle m | n_i | m \rangle \).

The derivative concerning correlated pairs is
\[
O_{f_{ij}}(m, \alpha) = \frac{1}{\langle m | \psi \rangle} \frac{\partial}{\partial (f_{m})_{ij}} \langle m | \psi \rangle
\]
\[
= \frac{1}{\gamma_m 2^{N/2}(N/2)!} \text{Pf}(f_{m}) \left(- (f_{m})_{ij}^{-1} \text{Pf}(f_{m}) \gamma_m 2^{N/2}(N/2)! \right)
\]
\[
= -(f_{m})_{ij}^{-1} - (f_{m})_{ij}^{-1}
\]
\[
= \frac{1}{2} \left((f_{m})_{ij}^{-1} - (f_{m})_{ij}^{-1}\right) \quad (C14)
\]
The derivative concerning the correlated-pair parameters of the \( J \)-projected energy is
\[
O_{f_{ij}}(m, \alpha) = \frac{1}{\langle m | P_M^f \phi \rangle} \frac{\partial}{\partial X_{ab}} \langle m | P_M^f \phi \rangle \quad (C15)
\]
\[
= \sum_{nK} g_{K} w_{nK} \langle m | R_n | \phi \rangle \sum_{nK} g_{K} w_{nK} \langle m | R_n | \phi \rangle
\]
\[
= \sum_{nK} g_{K} w_{nK} \langle m | R_n | \phi \rangle \sum_{nK} g_{K} w_{nK} \langle m | R_n | \phi \rangle
\]
\[
\times \left(- \sum_{j=1}^N R^T_{\alpha m} \langle (R X R^T)_{m}^{-1} \rangle_{m,j} R_{m,b} \right)
\]
The derivative concerning the \( g_K \) is
\[
O_{g_K}(m, \alpha) = \frac{1}{\langle m | \psi \rangle} \frac{\partial}{\partial g_K} \langle m | \psi \rangle \quad (C16)
\]
\[
= \frac{1}{\sum_{nK} g_{K} w_{nK} \langle m | R_n | \phi \rangle} \sum_{n} w_{nK} \langle m | R_n | \phi \rangle
\]

By combining these equations and the MCMC procedure, we can evaluate the energy gradient of the \( J \)-projected energy.

The norm of the small displacement of the \( | \psi \rangle \) caused by the small change of the variational parameters \( \gamma_i \) is
\[
\Delta_{\text{norm}}^2 = \left| \langle \psi_{\alpha+\gamma} | - \langle \psi_{\alpha} \rangle \right|^2
\]
\[
= \sum_{ij} \gamma_i^* \gamma_j \frac{\partial}{\partial \alpha_i} \frac{\partial}{\partial \alpha_j} \langle \psi | \psi \rangle
\]
\[
= \sum_{ij} \gamma_i^* S_{ij} \gamma_j \quad (C17)
\]

with the overlap matrix \( S_{ij} \),
\[
S_{ij} = \langle O_i^\dagger O_j \rangle - \langle O_i^\dagger \rangle \langle O_j \rangle \quad (C18)
\]

which is Hermitian and positive semidefinite [30].

In the steepest gradient method, the small displacement is taken as the derivative of energy such as
\[
\gamma_i = - \Delta t \frac{\partial (H)}{\partial \alpha_i} = - \Delta t g_i. \quad (C19)
\]

On the other hand, in the SR method, the small displacement is taken as the product of the inverse of \( S_{ij} \) and derivative of energy such as
\[
g_i = - \Delta t \sum_j S_{ij}^{-1} g_j \quad (C20)
\]

By using the inverse of \( S_{ij} \), the direction with the small norm of the \( S_{ij} \), or the direction causing small displacement, is taken as large step width and vice versa. In this work, we typically take \( \Delta t = 0.2 \).

In order to stabilize the SR method further, we apply two modifications to the overlap matrix, \( S_{ij} \) following Ref. [1]. One is the scaling of its diagonal matrix elements. We replace the overlap matrix by the scaled one,
\[
S'_{ij} = (1 + \epsilon \delta_{ij}) S_{ij}, \quad (C21)
\]

where \( \epsilon \) is a small constant. This modification makes the overlap matrix positive definite and stable even if \( S_{ij} \) is calculated stochastically including a certain error [31]. In this work, we typically take \( \epsilon = 0.01/\sqrt{i} \) where \( i \) is the number of iterations.

The other method to stabilize the SR method is the truncation of the redundant directions by introducing the cut off of the small eigenvalues of the overlap matrix. As it is Hermitian, we can diagonalize the overlap matrix such as
\[
S_{ij} = \sum_k U_{ik} \lambda_k U^\dagger_{kj}. \quad (C22)
\]

The redundancy of the variational-parameter space causes zero or small eigenvalues of the overlap matrix. Besides, small eigenvalues with statistical errors cause instability in evaluating the inverse matrix in Eq. (C20). In
order to avoid the problem, we replace $1/\lambda_i$ by 0 for $\lambda_i < \epsilon_{\text{cut}}$. In this work, we typically take $\epsilon_{\text{cut}} = 2/\sqrt{i} \times 10^{-4}$ where $i$ is the number of iterations. Thus,

$$\gamma_k = -\Delta t \sum I S_{kl}^{-1} g_l = -\Delta t \sum il U_{kl} U_{il}^\dagger g_l$$  \hspace{1cm} (C23)

is replaced by

$$\gamma_k = -\Delta t \sum il \Theta(\lambda_i - \epsilon_{\text{cut}}) \frac{1}{\lambda_i} U_{kl} U_{il}^\dagger g_l$$  \hspace{1cm} (C24)

where $\Theta(x)$ is the Heaviside function.

As a summary, we iteratively shift the variational parameters by adding the direction provided by Eq. (C20) in the SR method. It is expected to decrease the energy expectation value and, at the same time, to suppress the norm of the displacement of the wave functions by removing the effect of the redundancy of the variational parameters. This procedure is iterated until the energy converges.

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