Excited states of quantum many-body interacting systems: A variational coupled-cluster description

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Abstract

We extend recently proposed variational coupled-cluster method to describe excitation states of quantum many-body interacting systems. We discuss, in general terms, both quasiparticle excitations and quasiparticle-density-wave excitations (collective modes). In application to quantum antiferromagnets, we reproduce the well-known spin-wave excitations, i.e. quasiparticle magnons of spin ±1. In addition, we obtain new, spin-zero magnon-density-wave excitations which has been missing in Anserson’s spin-wave theory. Implications of these new collective modes are discussed.

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I. INTRODUCTION

In the preceding papers (Ref. 1 and 2, hereafter referred to as paper I and II), we proposed a general variational theory for ground states of quantum many-body interacting systems. Our analysis extends the well-established coupled-cluster method (CCM) [3, 4, 5] to a variational formalism in which bra and ket states are now hermitian to one another, contrast to the traditional CCM where they are not [6]. Ever since the CCM was first proposed, attempts have been made to extend it to a standard variational formalism, for examples, in the seventies in nuclear physics [7] and later in quantum chemistry [8]. It is perhaps fair to say that progress of this variational approach is slow, particularly when comparing with a plethora of applications made by the traditional CCM over the last 35 years [9]. Main difficulties in this variational approach include ad hoc approximation truncations and slow convergent numerical results. In I and II, we provided a new systematic scheme to overcome these difficulties. In particular, we introduced two sets of important bare distribution functions and derived self-consistency equations for these functions; calculations of physical quantities can all be done in terms of these functions. This strategy is similar to that employed by another well-established variational theory, the method of correlated basis functions (CBF) [10], where density distribution functions are key ingredients. We showed that the traditional CCM is a simple linear approximation to one set of bare distribution functions. We introduced diagrammatic techniques to calculate those distribution functions to high orders for achieving convergent results; resummations of infinite (reducible) diagrams can now be done by a practical, self-consistent technique. Furthermore, in our diagrammatic approach, a close relation with the CBF method was established and exploited; a possible combination of these two methods was also proposed. We demonstrated the efficacy of our variational method by applying to quantum antiferromagnets. The ground-state properties of spin-wave theory (SWT) [11] was reproduced in a simple approximation. Approximation beyond SWT by including higher-order, infinite sets of reducible diagrams produced convergent, improved numerical results for square and cubic lattices and, interestingly, it also cures the divergence by SWT in one dimensional system.

In this article we extend our variational CCM to describe excitation states. A brief report of some preliminary results has been published [12]. We investigate two different types of excitation states using two approaches. In the first approach, we follow the traditional
CCM [13, 14] to investigate quasiparticle excitations, but keeping our ket and bra excited states hermitian to one another. We then investigate collective modes by adapting Feynman’s excitation theory of phonon-roton spectrum of helium liquid [15] to our method. In application to antiferromagnets, we find that quasiparticle excitations correspond to Anderson’s spin-wave excitations which are often referred to as magnons with spin +1 or −1 [11]. We find collective modes in these quantum antiferromagnets as longitudinal, spin-zero magnon-density-wave excitations which have been missing in Anderson’s theory. In our approximation, energy spectra of these spin-zero excitations show a large gap for a cubic lattice (3D) and are gapless in a square lattice (2D). These spectra are similar to those of charge-density-wave excitations (plasmons) in quantum plasmas such as electron gases at low temperature [16]. More discussion on these collective modes will be given in the final section of this article.

II. GROUND STATES BY VARIATIONAL COUPLED-CLUSTER METHOD

We briefly summarize in this section our variational approach for the ground state of a many-body interacting system. Details can be found in I and II. We take a spin-\(s\) antiferromagnetic Heisenberg model on a bipartite lattice as our model system. The Hamiltonian is given by

\[ H = \frac{1}{2} \sum_{l,n} \mathbf{s}_l \cdot \mathbf{s}_{l+n}, \tag{1} \]

where index \(l\) runs over all lattice sites, \(n\) runs over all \(z\) nearest-neighbor sites. We use Coester representation for both ket and bra ground-states, and write

\[ |\Psi_g\rangle = e^S |\Phi\rangle, \quad S = \sum_I F_I C_I^\dagger; \quad \langle\tilde{\Psi}_g| = \langle\Phi| e^{\tilde{S}}, \quad \tilde{S} = \sum_I \tilde{F}_I \tilde{C}_I, \tag{2} \]

where model state \(|\Phi\rangle\) is given by the classical Néel state, \(C_I^\dagger\) and \(C_I\) with nominal index \(I\) are the so-called configurational creation and destruction operators and are given by, for the spin lattice of Eq. (1),

\[ \sum_I F_I C_I^\dagger = \sum_{k=1}^{N/2} \sum_{i_1, j_1, \ldots} f_{i_1, j_1, \ldots} s_{i_1}^z s_{j_1}^z s_{j_2}^+ s_{j_3}^+ \cdots s_{j_k}^+ \left(2s\right)^k, \tag{3} \]

for the ket state. The bra state operators are given by the corresponding hermitian conjugate of Eq. (3), using notation \(\tilde{F}_I = \tilde{f}_{i_1, j_1, \ldots}\) for the bra-state coefficients. As before, we have
used index $i$ exclusively for the spin-up sublattice of the Néel state and index $j$ for the spin-down sublattice. The coefficients $\{F_I, \tilde{F}_I\}$ are then determined by the standard variational equations as

$$\frac{\delta \langle H \rangle}{\delta F_I} = \frac{\delta \langle H \rangle}{\delta \tilde{F}_I} = 0, \quad \langle H \rangle \equiv \frac{\langle \tilde{\Psi}_g | H | \Psi_g \rangle}{\langle \tilde{\Psi}_g | \Psi_g \rangle}.$$  (4)

The important bare distribution functions, $g_I \equiv \langle C_I \rangle$ and $\tilde{g}_I \equiv \langle C_I^\dagger \rangle$, can be expressed in self-consistency equations as

$$g_I = G(\tilde{g}_J, F_J), \quad \tilde{g}_I = G(g_J, \tilde{F}_J),$$  (5)

where $G$ is a function containing up to linear terms in $\tilde{g}_J$ (or $g_J$) and finite order terms in $F_J$ (or $\tilde{F}_J$). The Hamiltonian expectation $\langle H \rangle$ of Eq. (4) can be expressed as, in general, a function containing up to linear terms in $g_I$ and $\tilde{g}_I$ and finite order polynomial in $F_I$ (or in $\tilde{F}_I$),

$$\langle H \rangle = \mathcal{H}(g_I, \tilde{g}_I, F_I) = \mathcal{H}(\tilde{g}_I, g_I, \tilde{F}_I).$$  (6)

In I and II, as a demonstration, we considered a simple truncation approximation in which the correlation operators $S$ and $\tilde{S}$ of Eqs. (2) and (3) retain only the two-spin-flip operators as

$$S \approx \sum_{i,j} f_{ij} C_{ij}^\dagger = \sum_{i,j} f_{ij} \frac{s_i^- s_j^+}{2s}, \quad \tilde{S} \approx \sum_{i,j} \tilde{f}_{ij} C_{ij} = \sum_{i,j} \tilde{f}_{ij} \frac{s_i^+ s_j^-}{2s}.$$  (7)

The spontaneous magnetization (order parameter) in this two-spin-flip approximation is given by the one-body density function $\rho_{ij}$ as

$$\langle s_i^z \rangle = s - \rho, \quad \rho = \sum_j \rho_{ij} = \sum_j f_{ij} \tilde{g}_{ij},$$  (8)

where we have taken the advantage of translational invariance of the lattice system. For $j$-sublattice, $\langle s_j^z \rangle = \rho - s$. Within this approximation the SWT result for the correlation coefficient can be derived from Eq. (4) as

$$f_q = \tilde{f}_q = \frac{1}{\gamma_q} \left[ \sqrt{1 - (\gamma_q)^2} - 1 \right], \quad \gamma_q = \frac{1}{z} \sum_n e^{i\mathbf{q} \cdot \mathbf{r}_n},$$  (9)

where $f_q$ is the sublattice Fourier transformation of $f_{ij}$ with $\mathbf{q}$ restricted to the magnetic zone, $z$ is the coordination number of the lattice, and $n$ is the nearest-neighbor index. Fourier component of the one-body bare distribution function is derived as,

$$\tilde{g}_q = \frac{\tilde{f}_q}{1 - \tilde{f}_q f_q}.$$  (10)
Finally, the two-body distribution functions is approximated by, in the same order,

$$\tilde{g}_{ij,i'j'} \approx \tilde{g}_{ij}\tilde{g}_{i'j'} + \tilde{g}_{ij'}\tilde{g}_{i'j}.$$  \hfill (11)

Approximation beyond these SWT formulas produced improved results and were given in details in II. For simplicity of our first attempt to discuss excitation states, we shall restrict ourselves to these approximations of Eqs. (7-11) in the following.

### III. QUASIPARTICLE EXCITATIONS

As mentioned in Sec. I, inspired by the close relation between our approach and the CBF method, we can investigate quasiparticle-density-wave excitations by adapting Feynman’s excitation theory, as well as usual quasiparticle excitations by similar approach as in the traditional CCM. One well-known example of a quantum system exhibiting similar two kind of excitations is quantum electron gases [16], where quasiparticle excitations are electron or hole excitations and collective modes are plasmon excitations representing longitudinal, charge-neutral density fluctuations of those quasielectron and holes. In this section we focus on quasiparticle excitations and leave discussion of collective modes in the next section. We will first discuss these excitations in a general term and then apply to the spin-lattice model of Eq. (1) as a demonstration.

Following Emrich in the traditional CCM [13, 14], we express excitation ket-state $|\Psi_e\rangle$ by a linear operator $X$ constructed from creation operators acting onto the ground state $|\Psi_g\rangle$ as

$$|\Psi_e\rangle = X|\Psi_g\rangle = X e^S|\Phi\rangle,$$

$$X = \sum_L x_L C_L^\dagger,$$  \hfill (12)

and, unlike the traditional CCM, our bra excitation state is the corresponding hermitian conjugate, involving only destruction operators as

$$\langle \tilde{\Psi}_e | = \langle \tilde{\Psi}_g | \tilde{X} = \langle \Phi | e^{\tilde{S}} \tilde{X},$$

$$\tilde{X} = \sum_L \tilde{x}_L C_L.$$  \hfill (13)

In Eqs. (12) and (13), the ground-state operators $S$ and $\tilde{S}$ are as given by Eqs. (2), $x_L$ and its hermitian conjugate $\tilde{x}_L$ are excitation coefficients. For quasiparticle creation and destruction operators $C_L^\dagger$ and $C_L$ of Eqs. (12-13), we use index $L$ to mark the following important difference to the ground-state counterparts $C_I^\dagger$ and $C_I$ of Eqs. (2). Due to symmetry consideration, some configuration operators are not included in the correlation operator $S$.
and $\tilde{S}$ of the ground states but they are important in the excited states. In our spin lattice example of Eq. (1), the ground-state operators of Eq. (3) always contain even number of spin-flip operators (each spin-flip-up operator for the $i$-sublattice always pairs up with one spin-flip-down operator for the $j$-sublattice) to ensure the total $z$-component of angular momentum $s_{\text{total}}^z = 0$. For the excitation operators, however, the constraints are different. The single spin-flip operator $s_i^-$ for the $i$-sublattice (or $s_j^+$ for the $j$-sublattice) will be the important first term in Eq. (12) to be discussed in the followings; the corresponding excitation state $|\Psi_e\rangle$ is in the $s_{\text{total}}^z = -1$ sector (or $+1$ if $s_j^+$ is used). Therefore, these excitations are referred to as quasiparticles carrying spin $\pm 1$. For our spin lattice models, we expect that these quasiparticles are the well-known magnons of spin-wave excitations [11].

If the ground state $|\Psi_g\rangle$ is exact with energy $E_0$, the energy difference between excitation state of Eqs. (12) and the ground state can be written as,

$$
\epsilon = \frac{\langle \tilde{\Psi}_g | XH \tilde{X} | \Psi_g \rangle}{\langle \tilde{\Psi}_e | \Psi_e \rangle} - E_0 = \frac{\langle \tilde{\Psi}_g | X[H, X] | \Psi_g \rangle}{\langle \tilde{\Psi}_e | \Psi_e \rangle},
$$

which involves a commutation. In general, $|\Psi_g\rangle$ is not exact but calculated by approximations. For our variational ground states of Eqs. (2), Eq. (14) can be shown to remain valid after replacing the exact energy $E_0$ by the variational energy $E_g$ which obeys the following optimal conditions,

$$
E_g = \langle H \rangle = \frac{\langle HC_i^\dagger \rangle}{g_I} = \frac{\langle C_I H \rangle}{g_I},
$$

derived from Eqs. (4).

To prove Eq. (14) after replacing the exact $E_0$ by the variational $E_g$, we first express the normalization of excited states of Eqs. (12) and (13) as an expectation value in the ground-states of Eqs. (2) as,

$$
I_e = \langle \tilde{\Psi}_e | \Psi_e \rangle = I_g \langle \tilde{X} X \rangle = I_g \sum_{L,L'} \tilde{x}_{L'} x_L \langle C_{L'} C_I^\dagger \rangle,
$$

where $I_g = \langle \tilde{\Psi}_g | \Psi_g \rangle$. We now consider a general linear operator $O = O(C_I^\dagger, C_I)$ (a polynomial of $C_I^\dagger$ and/or $C_I$), and write

$$
O|\Psi_g\rangle = Oe^S|\Phi\rangle = e^S \tilde{O}|\Phi\rangle,
$$

where the similarity-transformed operator, $\tilde{O} \equiv e^{-S}Oe^S = O(\tilde{C}_I^\dagger, \tilde{C}_I)$, $\tilde{C}_I = C_I^\dagger$ and

$$
\tilde{C}_I = e^{-S}Oe^S = C_I + [C_I, S] + \frac{1}{2!}[C_I, [S, C_I]] + \cdots,
$$
which always terminates for a finite order operator \( C_I \). In each term of such \( O \) expansion series, by shifting all destruction operators \( C_I \) to the right, and using the property \( C_I \Phi = 0 \), we conclude that only terms containing constants or only creation operators survive. We therefore have a general expression

\[
O(C_I^\dagger, C_I)|\Psi_g\rangle = O(C_J^\dagger, F_J)|\Psi_g\rangle ,
\]
where \( O(C_J^\dagger, F_J) \) is a function containing up to linear terms in \( C_J^\dagger \) and finite-order terms in \( F_J \). We shall refer Eq. (19) as linear theorem in our variational approach as it is useful for general analysis. In fact, the important Eqs. (5) and (6) in Sec. II are two specific application of this linear theorem. Therefore we can write, for a special case of Eq. (19),

\[
C_{L'} C_I^\dagger |\Psi_g\rangle = Y_{L'L}(C_I^\dagger, F_I)|\Psi_g\rangle ,
\]
where \( Y_{L'L}(C_I^\dagger, F_I) \) is a function containing up to linear terms in \( C_I^\dagger \) and finite-order terms in \( F_I \). Using Eq. (20), Eq. (16) can be written as

\[
I_e = I_g \sum_{L,L'} \bar{x}_{L'} Y_{L'L}(\bar{g}_I, F_I) x_L .
\]
Combining with the optimal condition of Eq. (15), it is easy to show

\[
\frac{1}{I_e} \langle \tilde{\Psi}_g | H \tilde{X} X | \Psi_g \rangle = E_g .
\]
Hence, we obtain similar equation to Eq. (14) for the energy difference,

\[
\epsilon = \frac{1}{I_e} \langle \tilde{\Psi}_g | \tilde{X} H X | \Psi_g \rangle - E_g = \frac{I_g}{I_e} \langle \tilde{X} [H, X] \rangle .
\]

We now apply the above formulas to discuss quasiparticle excitations of spin systems of Eq. (1). For simplicity, we consider an approximation in which we retain only single-spin-flip operators in \( X \) and \( \tilde{X} \) of Eqs. (12) and (13),

\[
X \approx \sum_i x_i s^-_i , \quad \tilde{X} \approx \sum_i \tilde{x}_i s^+_i ,
\]
with coefficients chosen as

\[
x_i = x_i(q) = \sqrt{\frac{2}{N}} e^{i q \cdot r_i} , \quad \tilde{x}_i = \tilde{x}_i(q) = \sqrt{\frac{2}{N}} e^{-i q \cdot r_i} ,
\]
to define a linear momentum $q$. Such an excited state, $|\Psi_e\rangle = X|\Psi_g\rangle$, is therefore in the sector of $s_{\text{total}}^z = -1$ and has a linear momentum $q$. The normalization integral of Eq. (24) is easily calculated as

$$I_e \left/ I_g \right. = \langle \tilde{X} X \rangle = 2 \sum_i \tilde{x}_i x_i \langle s_i^z \rangle + 2s \sum_{i,i',j} \tilde{x}_i x_i f_{i'j} \tilde{g}_{ij} - \sum_{i,i',j,j'} \tilde{x}_i x_i f_{i'j} f_{i'j'} \tilde{g}_{ij} \tilde{g}_{ij'},$$

and using Eqs. (8) and (11), we derive,

$$I_e \left/ I_g \right. = (s - \rho)(1 + \rho_q),$$

where $\rho_q \equiv f_q \tilde{g}_q$. Using approximations of Eqs. (9-11), we obtain, for isotropic point $A = 1$,

$$I_e \propto \frac{1}{q}, \quad q \rightarrow 0,$$

in all dimensions.

Calculation of the numerator in Eq. (23) is slightly more complicated. We quote the result here as, to the order of $(2s)^2$,

$$\langle \tilde{X}[H, X] \rangle \approx 2s^2(1 + (1 + 1 + \rho_q + \gamma_q g_q)).$$

The energy spectrum of Eq. (23) is therefore given by, to the order of $(2s)$,

$$\epsilon_q = \frac{I_g}{I_e} \langle \tilde{X}[H, X] \rangle \approx sz \frac{1 + 1 + \rho_q + \gamma_q g_q}{1 + \rho_q}.$$

Using Eqs. (9-11), we obtain the energy spectrum as

$$\epsilon_q = sz \sqrt{1 - (\gamma_q)^2},$$

which agrees exactly with the spin-wave theory [11]. Spectrum of Eq. (31) is gapless in any dimension because $\epsilon_q \propto q$ as $q \rightarrow 0$. Similar calculations using spin-flip operators $s_j^\pm$ for the $j$-sublattice in Eq. (24) will produce the same spectrum as Eq. (31) except that the corresponding excitation state has spin $s_{\text{total}}^z = +1$. These spin-wave excitations are often referred to as magnons.

IV. QUASIPARTICLE-DENSITY-WAVE EXCITATIONS

In previous section, by using quasiparticle operators (i.e., spin-flip operators $s_j^\pm$), we have reproduced the magnon excitations with spin equal to $+1$ or $-1$. These quasiparticles in
general interact with one another, thus producing quasiparticle density fluctuations. Excitation states due to these fluctuations are usually best discussed in terms of corresponding density operator. For our spin models, density operators are clearly given by operators $s^z$ as it measures the number of spin-flips with respect to the Néel model state and its expectation value is the order parameter as given by Eq. (8). For general purpose, we use notation $C_L^0$ for the quasiparticle density operators as opposed to the quasiparticle operators $C_L^\pm$ used earlier. The efficiency of using density operators to investigate collective modes of a quantum interacting system was demonstrated by Feynman for the phonon-roton spectrum of quantum fluid helium-4, who extended Bijl’s theory in a much simpler and clearer fashion. Feynman’s excitation formula was also derived by Pines for the plasmon spectrum of 3D metals. The 2D plasmon spectrum first derived by Stern can also be derived by using density operator as shown in a PhD thesis. It is interesting to note that both the CBF method for the ground state and Feynman’s theory for excitation states have been successfully applied to fractional quantum Hall effects. Feynman’s excitation theory is now often referred to as single-mode approximation.

Following Feynman, we write the quasiparticle density-wave excitation state as

$$|\Psi_0^e\rangle = X^0|\Psi_g\rangle, \quad X^0 = \sum_L x_L C_L^0,$$

where, as defined earlier, $C_L^0$ are the quasiparticle density operators. The bra state is given by the hermitian conjugate of Eq. (32), $\langle \tilde{\Psi}_e | = \langle \tilde{\Psi}_g | X^0$. Using the same argument as before for the quasiparticle excitation of Eq. (23), we obtain a similar equation for the energy difference for our collective modes as,

$$\epsilon^0 = \frac{I_g^e}{I_g^0} \langle \tilde{X}^0[H, X^0]\rangle ,$$

where $I_g^0 = \langle \tilde{\Psi}_g | \Psi_g^0 \rangle$. We notice that, by definition, density operator $C_L^0$ is a hermitian operator, $(C_L^0)^\dagger = C_L^0$. By considering a similar excited state $\tilde{X}^0|\Psi_g\rangle$, it is straightforward to derive the following double commutation formula,

$$\epsilon^0 = \frac{I_g^e}{2I_g^0} \langle [\tilde{X}^0, [H, X^0]]\rangle .$$

The double commutation in the above equation is the key to the efficiency of Feynman’s excitation theory. It is often referred to as $f$-sum rule in other quantum systems such as electron gases.
Before we apply Eq. (34) for collective modes in spin lattices, it is useful to discuss sum rules in our spin models as density operators normally obey sum rule equations \[10, 22\]. The order parameter of Eq. (8) can also be calculated through two-body functions as

\[
\langle \langle s_i^z \rangle \rangle^2 = \frac{\langle \tilde{\Psi}_g | (s_i^z)^2 | \tilde{\Psi}_g \rangle}{\langle \tilde{\Psi}_g | \tilde{\Psi}_g \rangle},
\]

(35)

where \( s_a^z = \sum_i (-1)^i s_i^z / N \) is the staggered spin operator. We introduce total magnon-density operator \( \hat{n}_i \) as

\[
2\hat{n}_i = 2s - s_i^z + \frac{1}{z} \sum_{n=1}^{z} s_{i+n}^z,
\]

(36)

where as before summation over \( n \) is over all \( z \) nearest neighbors. Hence, the sum rule for the one-body function is simply \( \frac{2}{N} \sum_i \langle \hat{n}_i \rangle = \rho \). The two-body Eq. (35) can now be written as, using translational invariant property \( \rho_i = \rho \),

\[
\frac{2}{N} \sum_{i'=1}^{N/2} \langle \hat{n}_i \hat{n}_{i'} \rangle = \rho \rho_i = \rho^2,
\]

(37)

which is the familiar two-body sum rule equation \[10, 22\]. In the approximation of Eq. (7-11), we find that this sum rule is obeyed in both cubic and square lattices in the limit \( N \to \infty \). In particular, we find that \( \frac{2}{N} \sum_i \langle \hat{n}_i \rangle - \rho^2 \propto 1/N \) in a cubic lattice and \( \propto (\ln N)/N \) in a square lattice. These asymptotic properties are important in the corresponding excitation states as will be discussed later. However, Eq. (37) is violated in the one-dimensional model, showing the deficiency of the two-spin-flip approximation of Eq. (7) for the one-dimensional model. We therefore leave further investigation elsewhere and focus on the cubic and square lattices in the followings, using approximations of Eqs. (7-11).

We therefore write our magnon-density-wave excitation state using total magnon density operator \( \hat{n}_i \) of Eq. (36) as,

\[
|\Psi_e^0 \rangle = X_q^0 |\Psi_g \rangle, \quad X_q^0 = \sum_i x_i(q) \hat{n}_i, \quad q > 0
\]

(38)

and its hermitian counterparts for the bra state, \( \langle \tilde{\Psi}_e^0 | = \langle \tilde{\Psi}_g | \tilde{X}_q^0 \rangle \). The coefficient, \( x_i(q) = \sqrt{\frac{2}{N}} e^{i q r_i} \), etc. The condition \( q > 0 \) in Eq. (38) ensures the orthogonality between this excited state with the ground state. The excitation energy difference is given by Eq. (34) as

\[
\epsilon_q^0 = \frac{N(q)}{S_0(q)}, \quad q > 0
\]

(39)
where $N(q) \equiv \langle [\tilde{X}_q^0, [H, X_q^0]] \rangle / 2$ and $S^0(q) \equiv \langle \tilde{X}_q^0 X_q^0 \rangle$ is the structure function. Both $N(q)$ and $S^0(q)$ can be straightforwardly calculated as, using approximations of Eqs. (7-11),

$$ N(q) = -\frac{sz}{2} \sum_{q'} (\gamma_{q'} + \gamma_{q'-q'}) \tilde{g}_{q'}, $$

and

$$ S^0(q) = \frac{1}{4}(1 + \gamma_q^2) \rho + \frac{1}{4} \sum_{q'} [(1 + \gamma_{q'}^2) \rho_{q-q'} + 2 \gamma_{q} \tilde{g}_{q'} \tilde{g}_{q-q'}], $$

where $q > 0$. The energy spectrum $\epsilon_q^0$ of Eq. (39) can then be calculated numerically. We notice that Eq. (41) is closely related to the the sum rule Eq. (37) which correspond to $q = 0$ case [with an additional term in Eq. (41) when $q \rightarrow 0$]. Using the approximation of Eqs. (9) and (10), it is not difficult to show that $N(q)$ of Eq. (40) has a nonzero, finite value for all values of $q$. Any special feature such as gapless in the spectrum $\epsilon_q^0$ therefore comes from the structure function of Eq. (41), and hence is determined by the asymptotic behaviors of the sum rule Eq. (37) mentioned earlier.

For a cubic lattice, we plot $N(q)$ and $S^0(q)$ for two regions of $q$ in Fig. 1. In Fig. 2 we plot the corresponding spectra of Eq. (39), together with that of magnon excitations of Eq. (31) for comparison. As can be seen from Fig. 2, the spectrum $\epsilon_q^0$ has a nonzero gap everywhere. The minimum gap is about $\epsilon_q^0 \approx 0.96sz$ at $q = (q_0, q_0, q_0)$ with $q_0 \approx 0.04\pi$. (This is slightly different to that reported in Ref. 12 where detailed calculations in this region had no been were done.) This gap is about the same as the largest magnon energy, $\epsilon_q = sz$ at $q = (\pi/2, \pi/2, \pi/2)$ from Eq. (31). At $q = (\pi/2, \pi/2, \pi/2)$, we have the largest energy $\epsilon_q^0 \approx 2.92sz$. This is nearly three magnons’ energy at this $q$. At $q = (\pi, 0, 0)$, we obtain $\epsilon_q^0 \approx 2.56sz$.

For a square lattice, the structure function $S^0(q)$ of Eq. (41) has a logarithmic behavior $\ln q$ as $q \rightarrow 0$. This is not surprising as discussed earlier in the sum rule Eq. (37), where occurs the asymptotic behavior $(\ln N)/N$ as $N \rightarrow \infty$. For small values of $q$, $N(q)$ approaches to a finite value, $N(q) \approx 0.275sz$ as $q \rightarrow 0$. The corresponding energy spectrum of Eq. (39) is therefore gapless as $q \rightarrow 0$. Similar to the cubic lattice, we plot $N(q)$ and $S^0(q)$ of a square lattice in Fig. 3 and the corresponding spectra of Eq. (39) and Eq. (31) in Fig. 4. As can be seen from Fig. 4, magnon-density-wave energy is always larger than the corresponding magnon energy. At small values of $q$ ($q < 0.05\pi$), we find a good approximation by numerical calculations for the structure function, $S^0(q) \approx 0.31 - 0.16 \ln q$ with $q_x = q_y$. Similar behavior
holds near \( \mathbf{q} = (\pi, \pi) \). The energy spectrum of Eq. (39) in these region can therefore be approximated by

\[
\epsilon_0^q \approx \frac{0.275sz}{0.31 - 0.16 \ln q}, \quad q \to 0
\]  

(42)

for a square lattice with \( q_x = q_y \). We notice the slight difference for the coefficients of Eq. (42) to that of Eq. (19) of Ref. 12 where we focused in the region with \( q_y = 0 \). Although our calculations clearly show this spectrum of a square lattice is gapless at \( q = 0 \) and \( \mathbf{q} = (\pi, \pi) \), it is nevertheless very "hard" when comparing with the magnon’s soft mode \( \epsilon_q \propto q \) at small \( q \). For example, we consider a system with lattice size of \( N = 10^{10} \), the smallest value for \( q \) is about \( q \approx 10^{-10}\pi \) and we have energy \( \epsilon_0^q \approx 0.07sz \). Comparing this value with the corresponding magnon energy \( \epsilon_q \approx 10^{-10}sz \), we conclude that the energy spectrum of Eq. (39) is "nearly gapped" in a square lattice. We also notice that the largest energy in a square lattice \( \epsilon_0^q \approx 2.79sz \) at \( \mathbf{q} = (\pi, 0) \), not at \( \mathbf{q} = (\pi/2, \pi/2) \) as the case in a cubic lattice. At \( \mathbf{q} = (\pi/2, \pi/2) \), we obtain \( \epsilon_0^q \approx 2.62sz \) for the square lattice. We will discuss physical implications of these excitations in the next section.

V. DISCUSSION

We have obtained in this article two main results. Firstly, we have succeeded in extending our recently proposed variational approach to describe, in general terms, excitation states of a quantum many-body system. Secondly, we have applied our technique to quantum antiferromagnets thus reproducing the well-known magnon excitations and, in addition, we have obtained a new, spin-zero longitudinal collective modes which have been missing in spin-wave theory of Anderson [11]. In the followings, we shall discuss further physical implications of these new excitations and we conclude this article with a summary in the end.

It is interesting to notice similar behaviors between collective modes of quantum antiferromagnets and plasmon excitations of electron gases as both spectra show a large energy gap in 3D and are gapless in 2D. In fact, further similarity between these two quantum systems can be made. It is generally accepted that, for many purposes, a quantum antiferromagnet at zero temperature can be considered as a gas of weakly interacting, equal numbers of spin \( \pm 1 \) magnons (the transverse spin-flip wave excitations with respect to the classical Néel state); also present in the system are the spin-zero, longitudinal fluctuations consist-
This is similar to quantum electron gases which can also be considered as a gas of weakly interacting, equal numbers of quasielectrons and holes (the transverse excitations near the Fermi surfaces) and the charge-neutral, longitudinal fluctuations producing quasielectron-hole continuum. Plasmon excitation of electron gases have been well observed as sharp peaks over the electron-hole continuum. However, plasmon-like collective modes of quantum antiferromagnets as discussed in this article have so far eluded from observation to our best knowledge. We can only draw some support by considering a finite-size Heisenberg model of Eq. (1). As the ground state of a finite antiferromagnetic Heisenberg lattice is spin-singlet, we expect low-lying excitations are triplet with z-component of spin equal to 0, ±1. As lattice size increases from finite to infinite, for the cubic and square lattices, the spontaneous-symmetry-breaking occurs and the ground state is no longer spin singlet but has a long-ranged antiferromagnetic order.

We expect that the triplet excitation splits into different branches. The magnon spectrum of Eq. (31) with spin ±1 and the spectrum of Eq. (39) for spin-zero magnon-density waves are our approximation for these different branches of excitations. We also notice that recently modified spin-wave theories were applied to finite systems with results in reasonable agreements with exact finite-size calculations. As pointed out in Ref. 28, however, a major deficiency in this theory is the missing spin-zero excitations as the low-lying excitations for a finite lattice Heisenberg model are always triplet as mentioned earlier. We believe our magnon-density-wave excitation as discussed here corresponds to the missing branch; the energy gap in the cubic lattice and the nearly gapped spectrum in the square lattice of Eq. (39) reflect the nature of long-ranged Néel order in the ground states of infinite systems. Improvement for spectra of Eq. (39) can be done in similar fashion as was done for the ground state detailed in paper II, particularly for the square lattice. We will have more motivation to do so if we have experimental evidence of these collective modes.

In any case, this article concludes our general presentation of a new formalism of variational coupled-cluster method for a quantum many-body system. Beginning in paper I, we introduced and discussed bare distribution functions, key ingredient of this formalism. In paper II, we developed diagrammatic techniques for practical, high-order calculations of these functions. Application to quantum antiferromagnets has demonstrated the efficacy of this technique. Present article extends this formalism to excitation states. As discussed earlier, application to quantum antiferromagnets have produced new modes which have been
missing in all spin-wave theories and are yet to be confirmed by experiment. Our next main focus is to combine our present variational approach with the CBF method as first discussed in paper II. Hence we write our new ground state as

\[ |\Psi_u\rangle = e^{S^0} |\Psi_g\rangle = e^{S^0} e^{S} |\Phi\rangle , \]  

(43)

where \( S \) is as given by Eqs. (2) and \( S^0 \) is the generalized Jastrow correlation operator involving quasiparticle density operators as

\[ S^0 = \sum_{ij} f^0_{ij} s^z_i s^z_j , \]  

(44)

with \( f^0_{ij} \) as new variational functions. Using the 2-spin-flip approximation of Eq. (7) for \( S \), the new wave function of Eq. (46) can be understood as including both quasiparticle fluctuations described by operator exp(\( S \)) and quasiparticle-density fluctuations described by operator exp(\( S^0 \)). The results of collective modes obtained in Sec. IV certainly make this combination of Eq. (43) much more appealing and imperative.

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Fig. 1 $N(q)$ and $S^0(q)$ of Eqs. (40) and (41) for a cubic lattice. Shown are the values for two regions $q = (0,0,0)$ to $(\pi,0,0)$ and to $(\pi,\pi,\pi)$.

Fig. 2 Excitation energy spectra in unit of $sz$ in a cubic lattice. The higher branch is for the plasmon-like excitation of Eq. (39) and the lower one is for the magnon excitation of Eq. (31).

Fig. 3 Similar to Fig. 1 but for a square lattice. The divergence of $S^0(q)$ at $q = (0,0)$ and $(\pi,\pi)$ is given in the text.

Fig. 4 Similar to Fig. 2 but for a square lattice. The behavior near $q = (0,0)$ and $(\pi,\pi)$ for magnon-density-waves is given by Eq. (42).
\( S^0(q) \quad N(q) \)
Energy spectrum

Spin-waves

Magnon-density-waves
$S_0^0(q)$

$N(q)$
