Variational coupled-cluster study of plasmonlike excitations in quantum antiferromagnets

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We study plasmonlike excitations in quantum antiferromagnets by the recently proposed variational coupled-cluster method. Within our approximation, we find that these spin-zero excitations have a nonzero energy gap in a cubic lattice and are gapless in a square lattice, similar to the plasmons in quantum electron gases.

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Since 1952, Anderson's spin-wave theory (SWT) has been a key in our understanding of quantum antiferromagnets in three- and two-dimensional (3D and 2D) bipartite lattices.[1] For many purposes, such an antiferromagnet at zero temperature can be considered as a gas of weakly interacting quasiparticles (equal number of spin $\pm$ magnons, the transverse spin-flip wave excitations with respect to the classical Néel state); also present in this gas are the spin-zero, longitudinal fluctuations consisting of multimagnon continuum.[2–5] This is similar to the better-known quantum electron gases (metals at low temperature) which can also be considered as a gas of weakly interacting quasiparticles (equal number of quasielectrons and holes, the transverse excitations near the Fermi surfaces) and the charge-neutral, longitudinal fluctuations consisting of quasielectron-hole continuum.[6] The interesting question is whether or not the well-known plasmon excitations in metals, which show sharp peaks over the quasielectron-hole continuum,[6] also have a counterpart in an antiferromagnet. This is the focus of our theoretical investigation.

In its present form, SWT does not seem able to derive the plasmonlike excitations, including the more recently modified spin-wave theories.[7–9] We investigate plasmonlike excitations in an antiferromagnet by adapting Feynman's excitation theory of phonon-roton for quantum helium liquid[10] to our recently proposed variational coupled-cluster method (VCCM) which has been successfully applied to antiferromagnetic lattices.[11,12] In these earlier papers, we showed that a simple approximation reproduces the complete ground-state properties of SWT; improvements over SWT by higher-order calculations were also obtained; a close relation between our VCCM and the other well-established variational method, the method of correlated basis functions (CBF),[13] was also established and exploited. Since Feynman's excitation theory is an integrated part of the CBF method, adapting the Feynman's theory in our VCCM is therefore a natural extension. Indeed, by following Feynman, we are able to obtain spectra of spin-zero excitations in antiferromagnets, which are clearly not the multimagnon excitations discussed before,[2–4] but show very much plasmonlike features, a nonzero energy gap in a cubic lattice and a gapless spectrum in a square lattice.

We first briefly describe the application of VCCM to an antiferromagnetic lattice for the ground state as these results will be used in discussion of its plasmonlike excitations. Details are in Refs. 11 and 12. The antiferromagnetic Heisenberg Hamiltonian is given by

$$H=\frac{1}{2}\sum_{l,n}H_{l,l+n}=\frac{1}{2}\sum_{l,n}\mathbf{s}_{\mathbf{l}}\cdot\mathbf{s}_{\mathbf{l+n}}, \tag{1}$$

where the index l runs over all N bipartite lattice sites, n runs over all z nearest-neighbor sites. In VCCM, the ground state $|\Psi_g\rangle$ of Eq. (1) is given by the so-called Coester representation,

$$|\Psi_g\rangle=e^S|\Phi\rangle,\quad S=\sum_I F_I C_I^\dagger \tag{2}$$

with its Hermitian conjugate $\langle\tilde{\Psi}_g|=\langle\Phi|e^{\tilde{S}}$, $\tilde{S}=\Sigma_I\tilde{F}_IC_I$ as the bra ground state. In Eq. (2), the model state $|\Phi\rangle$ is given by the Néel state with alternating spin-up sublattice (denoted by i index) and spin-down sublattice (denoted by j index), and $C_I^\dagger$ with nominal index I is given by the spin-flip operators over the Néel model state,

$$\sum_I F_I C_I^\dagger=\sum_{k=1}^{N/2}\sum_{i_1\cdots,j_1\cdots} f_{i_1\cdots,j_1\cdots}\frac{s_{i_1}^-\cdots s_{i_k}^- s_{j_1}^+\cdots s_{j_k}^+}{(2s)^k}, \tag{3}$$

with s as spin quantum number. The bra state operators are given by the corresponding Hermitian conjugate of Eq. (3), using notation $\tilde{F}_I=\tilde{f}_{i_1\cdots,j_1\cdots}$ for the bra-state coefficients. The coefficients $\{F_I,\tilde{F}_I\}$ are then determined by the usual variational equations as

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

Evaluation of $\langle H\rangle$ is through the important bare distribution functions, $g_I\equiv\langle C_I\rangle$ and $\tilde{g}_I\equiv\langle C_I^\dagger\rangle$, which can be straightforwardly expressed in a self-consistency equation as

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

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 $\tilde{F}_J$). In Refs. 11 and 12, we considered a truncation approximation in which the correlation operators S and $\tilde{S}$ of Eqs. (2) and (3) retain only the two-spin-flip operators as

$$S \approx \sum_{ij} f_{ij} \frac{s_i^- s_j^+}{2s}, \quad \tilde{S} \approx \sum_{ij} \tilde{f}_{ij} \frac{s_i^+ s_j^-}{2s}. \tag{6}$$

The spontaneous magnetization (order parameter) in this two-spin-flip approximation is obtained by calculating the one-body density function as

$$\langle s_i^z \rangle = s - \rho_i, \quad \rho_i = \sum_j \rho_{ij} = \sum_j f_{ij} \tilde{g}_{ij}, \tag{7}$$

where $\rho_i = \rho$ due to translational invariance of the lattice system. For the spin-down j sublattice, $\langle s_j^z \rangle = -s + \rho_j = -\langle s_i^z \rangle$. As demonstrated in Ref. 12, the contributions to the one-body distribution function of Eq. (7) can be represented by diagrams. The results of SWT were obtained by a further approximation (partial resummations of the diagrams). The one-body bare distribution function $\tilde{g}_{ij} = \langle s_i^- s_j^+ \rangle / (2s)$ in this further approximation is given by, after the sublattice Fourier transformation,

$$\tilde{g}_q \approx \frac{\tilde{f}_q}{1 - \tilde{f}_q f_q}, \tag{8}$$

where $\tilde{f}_q$ is the Fourier component of correlation coefficient $\tilde{f}_{ij}$ with q restricted in the magnetic zone, etc. Variational Eq. (4) then reproduces the SWT result for this coefficient as

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

Finally, the two-body distribution functions, $\tilde{g}_{ij,i'j'} = \langle s_i^- s_j^+ s_{i'}^- s_{j'}^+ \rangle / (2s)^2$, are approximated by, in the same order as approximation of Eq. (9) of the large-s expansion,

$$\tilde{g}_{ij,i'j'} \approx \tilde{g}_{ij} \tilde{g}_{i'j'} + \tilde{g}_{ij'} \tilde{g}_{i'j}. \tag{10}$$

Systematic improvements over SWT for the ground state by including an infinite set of diagrams (i.e., resummation of an infinite $1/s$ expansion series) was obtained as detailed in Ref. 12. In the following, we will restrict ourselves to approximations of Eqs. (6)–(10) for the ground state to investigate plasmonlike excitations as required by Feynman's excitation theory.[10]

We also need to discuss one important ground-state property involving two-body correlation functions before discussing excitations. Order-parameter of Eq. (7) can also be calculated through two-body functions as

$$(\langle s_i^z \rangle)^2 = \frac{\langle \tilde{\Psi}_g | (s_a^z)^2 | \Psi_g \rangle}{\langle \tilde{\Psi}_g | \Psi_g \rangle}, \tag{11}$$

where $s_a^z = \Sigma_l (-1)^l s_l^z / N$ is the staggered spin operator.[8] Equation (11) is in fact the sum rule for the two-body distribution function as in the CBF method.[13] This can seen by introducing the total magnon-density operator $\hat{n}_i$ as

$$2\hat{n}_i = 2s - s_i^z + \frac{1}{z} \sum_{n=1}^{z} s_{i+n}^z, \tag{12}$$

where as before summation over n is over all z nearest neighbors. Hence the sum rule for the one-body function is simply $\frac{2}{N}\Sigma_i \langle \hat{n}_i \rangle = \rho$. The two-body counterpart, Eq. (11), can now be written in the following familiar sum rule equation:

$$\frac{2}{N} \sum_{i'=1}^{N/2} \langle \hat{n}_i \hat{n}_{i'} \rangle = \rho \rho_i = \rho^2, \tag{13}$$

where in the last equation, translational invariant property $\rho_i = \rho$ has been used. In the approximation of Eqs. (6)–(10), 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 $\left(\frac{2}{N}\Sigma_{i'} \langle \hat{n}_i \hat{n}_{i'} \rangle - \rho^2\right) \propto 1/N$ in a cubic lattice and $\propto (\ln N)/N$ in a square lattice.[14] These asymptotic properties are important in the corresponding excitation states as discussed later. However, Eq. (13) is violated in the one-dimensional model, showing the deficiency of the two-spin-flip approximation of Eq. (6) for the one-dimensional model. We therefore leave more detailed discussion and a possible cure elsewhere[14] and focus on the cubic and square lattices in the following.

For the quasiparticle excitations, briefly, we follow Emrich in the traditional CCM,[15,16] and write excitation ket state $|\Psi_e\rangle$ involving only $C_I^\dagger$ operators as, $|\Psi_e\rangle = X|\Psi_g\rangle$ with $X = \Sigma_I x_I C_I^\dagger$. However, unlike the traditional CCM, our bra excitation state is the corresponding Hermitian conjugate $\langle \tilde{\Psi}_e| = \langle \tilde{\Psi}_g | \tilde{X} = \langle \Phi | e^{\tilde{S}} \tilde{X}$. Choosing a single spin-flip operator $C_I^\dagger = s_i^-$, we have $X \approx \Sigma_i x_i s_i^-$ with coefficient chosen as $x_i = x_i(q) = \sqrt{\frac{2}{N}} e^{i\mathbf{q}\cdot\mathbf{r_i}}$ to define a linear momentum $\mathbf{q}$. State $|\Psi_e\rangle$ has therefore spin $s_{\text{total}}^z = -1$. The energy difference between this excitation state and the variational ground state of Eqs. (2)–(4), $\epsilon_q = \langle \tilde{\Psi}_g | \tilde{X} H X | \Psi_g \rangle / \langle \tilde{\Psi}_e | \Psi_e \rangle - \langle H \rangle$, can be derived as, to the order of $(2s)$,

$$\epsilon_q \approx sz \frac{1 + \rho_q + \gamma_q \tilde{g}_q}{1 + \rho_q} = sz\sqrt{1 - (\gamma_q)^2}, \tag{14}$$

where $\rho_q = f_q \tilde{g}_q$ and we have used Eqs. (6)–(10). This agrees with SWT (Ref. 1) in this order. The spectrum of Eq. (14) is gapless in all dimensions because $\epsilon_q \propto q$ as $q \to 0$. Similar calculations using spin-flip operators $C_I^\dagger = s_j^\dagger$ of the j sublattice in excitation operator X will produce the same spectrum as Eq. (14) except that the corresponding excitation state has spin $s_{\text{total}}^z = +1$. These excitations are often referred to as magnons.[1]

We now focus on the plasmonlike excitations. As mentioned earlier, Feynman's excitation theory provides an excellent description of longitudinal density-wave excitation in quantum helium liquid, the famous phonon-roton spectrum.[10,13] Feynman employed particle density operator to obtain the spectrum. It is interesting to note that Feynman's excitation theory was also successfully applied to fractional quantum Hall effects to obtain the gapped magnetoplasmon excitations,[17] to valence-bond-solid antiferromagnetic chains for the collective excitation spectrum which was very close to numerical values of exact finite-size calculations,[18] Feynman excitation formula was also derived by Pines for the plasmon spectrum of 3D metals.[6] The 2D plasmon spectrum first derived by Stern[19] can also be derived by using a density operator as shown in a Ph.D. thesis.[20] In order to discuss spin-zero excitations of antiferromagnets, we first notice that the order parameter $\rho_i=\rho_j=\rho$ of Eq. (7) in fact represents the average density of quasiparticle magnons of spin ±1, respectively; the spin operators $(s-s_i^z)$ and (s_j^z+s) are the corresponding magnon-density operators. Magnons of spin ±1 interact with one another with an attractive potential proportional to $-(s-s_i^z)(s_j^z+s)$ in the Hamiltonian of Eq. (1), where j is the nearest neighbor of i. The total magnon-density operator is given by $\hat{n}_i$ of Eq. (12), which nicely obeys the important two-body sum rule of Eq. (13) in our approximation of Eqs. (6)–(10) in the limit $N\to\infty$. Furthermore, for the case of $s=1/2$, magnons of same spin (+1 or −1) repel each other on a lattice site. With all these considerations, we believe it is suitable to apply Feynman's excitation theory to investigate the plasmonlike excitation in antiferromagnets. We therefore write this spin-zero excitation state using $\hat{n}_i$ as

$$|\Psi_e^0\rangle = X_q^0|\Psi_g\rangle, \quad X_q^0=\sum_i x_i(q)\hat{n}_i, \quad q>0 \tag{15}$$

and its Hermitian counterparts for the bra state, $\langle\tilde{\Psi}_e^0| = \langle\tilde{\Psi}_g|\tilde{X}_q^0$. The coefficient, $x_i(q)=\sqrt{\frac{2}{N}}e^{i\mathbf{q}\cdot\mathbf{r_i}}$, defines the linear momentum $\mathbf{q}$. The condition $q>0$ in Eq. (15) ensures the orthogonality between this excited state with the ground state. We notice that the density operator $\hat{n}_i$ in Eq. (15) is a Hermitian operator. This property can be used to derive a double commutation formula for the energy difference between the above excitation state and the variational ground states of Eqs. (2)–(4) as

$$\epsilon_{\mathbf{q}}^0 = \frac{\langle\tilde{\Psi}_g|\tilde{X}^0HX^0|\Psi_g\rangle}{\langle\tilde{\Psi}_e^0|\Psi_e^0\rangle} - \langle H\rangle = \frac{N(q)}{S^0(q)}, \quad q>0, \tag{16}$$

where $N(q)\equiv\langle[\tilde{X}_q^0,[H,X_q^0]]\rangle/2$, $S^0(q)\equiv\langle\tilde{X}_q^0X_q^0\rangle$ is the structure function, and the notation $\langle\cdots\rangle$ is the ground-state expectation value as before. The double commutator $N(q)$ corresponds to the f-sum rule.[6,10] Both $N(q)$ and $S^0(q)$ can be straightforwardly calculated as, using approximations of Eqs. (6)–(10),

$$N(q) = -\frac{sz}{2}\sum_{q'}(\gamma_{q'}+\gamma_q\gamma_{q-q'})\tilde{g}_{q'}, \tag{17}$$

and

$$S^0(q)=\frac{1}{4}(1+\gamma_q^2)\rho+\frac{1}{4}\sum_{q'}[(1+\gamma_q^2)\rho_{q'}\rho_{q-q'}+2\gamma_q\tilde{g}_{q'}\tilde{g}_{q-q'}], \tag{18}$$

where $\rho_q\equiv f_q\tilde{g}_q$ as before, f_q and $\tilde{g}_q$ are as given by Eqs. (8) and (9). Substituting Eqs. (17) and (18) into Eq. (16), we can then calculate the energy spectrum ϵ_q^0 numerically. We notice that Eq. (18) is closely related to the sum rule Eq. (13). It is not difficult to see from Eq. (17) that $N(q)$ has a nonzero, finite value for all values of q. Any special feature such as gapless in the spectrum ϵ_q^0 therefore comes from the structure function of Eq. (18), and hence is determined by the asymptotic behaviors of the sum rule Eq. (13) mentioned earlier. For a cubic lattice, we find that the spectrum ϵ_q^0 has a nonzero gap everywhere. The minimum gap is about $\epsilon_q^0\approx 0.99sz$ at $q\to 0$. This gap is about the same as the largest magnon energy, $\epsilon_q=sz$ at $\mathbf{q}=(\pi/2,\pi/2,\pi/2)$ from Eq. (14). At $\mathbf{q}=(\pi/2,\pi/2,\pi/2)$, we have the largest energy $\epsilon_q^0\approx 2.92sz$. This is nearly three magnons' energy at this $\mathbf{q}$. At $\mathbf{q}=(\pi,0,0)$, we obtain $\epsilon_q^0\approx 2.56sz$.

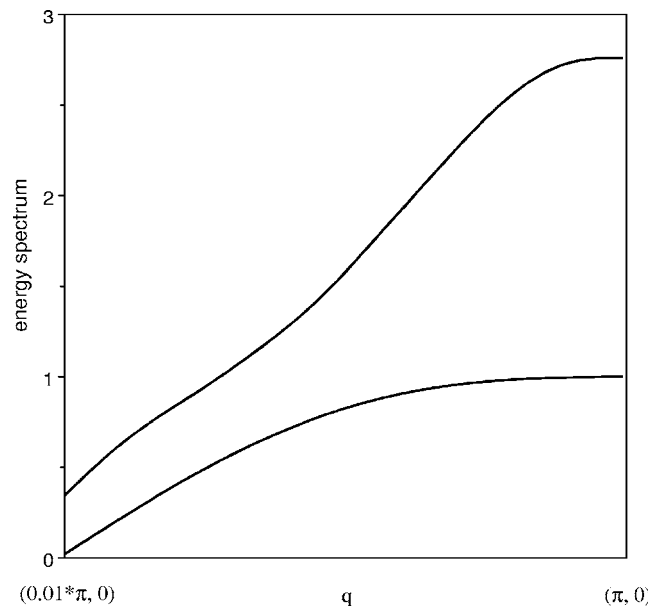

FIG. 1. Excitation energy spectra for the values of $\mathbf{q}=(0.01\pi,0)-(\pi,0)$ in a square lattice. The higher branch is for the plasmonlike excitation of Eq. (16) and the lower one is for the magnon excitation of Eq. (14).

For the square lattice the structure function $S^0(q)$ of Eq. (18) has a logarithmic behavior $\ln q$ as $q\to 0$. This is not surprising as discussed earlier in the sum rule Eq. (13), where occurs the asymptotic behavior $(\ln N)/N$ as $N\to\infty$. For small values of q, $N(q)$ approaches to a finite value, $N(q)\approx 0.276sz$ as $q\to 0$. The corresponding energy spectrum of Eq. (16) is therefore gapless as $q\to 0$. We plot the spectrum for the values of $\mathbf{q}$ between $(0.01\pi,0)$ and $(\pi,0)$ in Fig. 1, together with the corresponding magnon energies for comparison. As can be seen from Fig. 1, the spectrum energy of Eq. (16) is always much larger than the corresponding magnon energy of Eq. (14). At small values of q_x ($q_x<0.05\pi$), we find a good approximation by numerical calculations for the structure function, $S^0(q)\approx 0.25-0.16\ln q_x$ with $q_y=0$. The energy spectrum of Eq. (16) can therefore be approximated by

$$\epsilon_q^0\approx\frac{0.276sz}{0.25-0.16\ln q_x}, \quad q_x\to 0 \tag{19}$$

for a square lattice (with $q_y=0$). This spectrum is 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_x is about $q_x\approx 10^{-10}\pi$ and we have energy $\epsilon_q^0\approx 0.07sz$. Comparing this value with the corresponding magnon energy $\epsilon_q\approx 10^{-10}sz$, we conclude that the energy spectrum of Eq. (16) is "nearly gapped" in a square lattice. We also notice that the largest energy in a square lattice $\epsilon_q^0 \approx 2.79sz$ 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 $\epsilon_q^0 \approx 2.62sz$ for the square lattice.

In summary, we have applied the recently proposed VCCM by adapting the Feynman's excitation theory to investigate the plasmonlike excitations in quantum antiferromagnets, using magnon density operator s^z. The energy spectra obtained indeed appear very much plasmonlike as in quantum electron gases: a large energy gap in Three dimensions and the gapless spectrum in two dimensions. It is also interesting to note that, in a 2D electron gas, the plasmon spectrum ($\propto\sqrt{q}$ at small q limit[19,20]) is also "harder" than the corresponding quasiparticle excitations ($\propto q^2$) near the its Fermi surface. We also notice that recently modified spin-wave theories (SWTs) were applied to finite systems with results in reasonable agreement with exact finite-size calculations.[7–9] As pointed out in Ref. 8, however, a major deficiency in this modified SWT is the missing spin-zero excitation as the low-lying excitations for a finite lattice Heisenberg model are always triplet with spin equal to $0,\pm 1$. We believe the plasmonlike 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. (16) reflect the nature of long-ranged Néel order in the ground states of infinite systems. As observations of both the magnon excitations and multimagnon continuum in 3D and 2D antiferromagnets have been reported,[4,5] it will be interesting to see further experiments on these systems at very low temperature and high energy for observation of plasmonlike excitations as described here. We also believe that these plasmonlike excitations, if confirmed, may play a role in our understanding the physics of high-temperature superconducting cuprates.

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