Reply to "Comment on 't/U expansion for the Hubbard model'"

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It is shown that the unitary transformation used as the basis of a t/U expansion for the Hubbard model in an earlier Brief Report is *not* equivalent to the canonical perturbation expansion (CPE) discussed in the preceding Comment. The CPE is valid only to leading order in t/U. More evidence is offered in support of our suggestion that higher-order terms in the t/U expansion tend to favor spin-liquid states over antiferromagnetically ordered states.

The Hubbard model,[1-3]

$$H=-t\sum_{i,\ldots,1\sigma}N_{i,j}C_{i\sigma}^{\dagger}C_{j\sigma}+U\sum_{i}n_{i\uparrow}n_{i\downarrow}\,, \tag{1}$$

captures the essential physics of electronic systems for which the dominant interactions are on site (N_{ij} is one if i and j are neighboring sites). In the localized limit ($t=0$) of this model, the spectrum of H consists of macroscopically degenerate subspaces characterized by a given number of doubly occupied sites. For a half-filled band, the lowest-energy subspace has each site singly occupied and the only degree of freedom is the electron's spin. Since the discovery[4] of high-T_c superconductivity, there has been interest in the possible existence of a novel type of superfluidity which evolves from strongly correlated spin-liquid ground states at half-filling (see Refs. 5–8 for discussions of some of the ideas being explored).

For small t/U the Hubbard model may be replaced by an effective Hamiltonian which treats the hopping term perturbatively and describes how it breaks the degeneracy of each subspace. It has long been realized[1] that to leading order this expansion leads (at half-filling) to an effective antiferromagnetic coupling between spins on neighboring sites and, therefore, for the two-dimensional square lattice relevant[9] to high T_c, to a spin crystal[10] (antiferromagnetic) ground state. Superconductivity based on spin-liquid ground states remains a possibility, however, if the antiferromagnetic coupling becomes frustrated, for example, by the introduction of holes.[11] In our Brief Report[12] we pointed out that beyond-leading-order terms in the t/U expansion couple non-neighboring spins and frustrate the antiferromagnetic states. We argued on the basis of comparisons of the energies of the Néel state (the prototypical antiferromagnetic state) and several valence-bond states (the prototypical spin-liquid states) that these corrections are important at realistic values of t/U and can help to tip the delicate balance between spin-liquid and antiferromagnetic states. (A large part of the preceding Comment[13] merely repeats this calculation and obtains the same results except as noted below.) In response to the Comment[13] we first point out some incorrect and some misleading statements contained therein, and then present some additional evidence in favor of the thesis of our Brief Report.

The t/U expansion of the Hubbard model has a long history. A summary of early work[1,14-16] is contained in the paper of Takahashi,[17] who was the first to successfully carry out the expansion beyond leading order for the case of two or three dimensions. The approach taken to the t/U expansion in our Brief Report, was to seek a unitary transformation which eliminated, order by order, terms which couple subspaces with a different number of doubly occupied sites. For this purpose it was useful to rewrite Eq. (1) as

$$H=V+T_0+T_1+T_{-1}\,, \tag{2}$$

where V is the interaction term and T_m comes from hops which increase the number of doubly occupied sites by m. The transformed Hamiltonian,

$$H'=e^{iS}He^{-iS}=H+\frac{[iS,H]}{1!}+\frac{[iS,[iS,H]]}{2!}+\cdots\,, \tag{3}$$

where the kth-order contributions of S are proportional to U^{-k} times the product of k T_m's. S is determined iteratively[12] with the kth-order term fixed so as to eliminate the unwanted terms of order t^k/U^{k-1} from H'. The approach was inspired by the Foldy-Wouthuysen transformation[18] of atomic physics in which a relativistic expansion is obtained by eliminating terms coupling positive and negative energy solutions of the Dirac equation. We found that this approach was very convenient for carrying out the expansion to high order, although other[16,17,19] (equivalent) approaches could have been used. At half-filling the Hamiltonian is equivalent to a spin Hamiltonian which can be expressed in the form

$$H'=\sum_{N,l}\sum_{i_1,\ldots,i_N}L_{N,l}(i_1,\ldots,i_N)H_{N,l}(\sigma_{i_1},\sigma_{i_2},\ldots,\sigma_{i_N})\,, \tag{4}$$

where $i_1,\ldots,i_N$ range over the lattice sites, l labels the distinct ways in which N sites can be linked by near-neighbor hops, $L_{N,l}(i_1,\ldots,i_N)$ specifies the lth linkage of N sites, and $H_{N,l}(\sigma_{i_1},\sigma_{i_2},\ldots,\sigma_{i_N})$ is a spin operator. The linkage functions, which have an obvious graphical analog, are given in Table I and the corresponding spin operators are given[20] in Table II out to order t^6/U^5. These results are valid for any lattice in any number of dimensions. For the case of the 2D square lattice and the 3D simple cubic lattice our results for the effective Hamiltonian out to order t^4/U^3 are in agreement with earlier results by Takahashi.[17] For the case of a 1D lattice our result for

TABLE I. Linkage functions appearing in the t/U expansion out to order t^6/U^5.

N	l	$L_{N,l}(1,\ldots,N)$
2	1	N_{12}
3	1	$N_{12}N_{13}$
3	2	$N_{12}N_{23}N_{31}$
4	1	$N_{12}N_{23}N_{34}N_{41}$
4	2	$N_{12}N_{13}N_{14}$
4	3	$N_{12}N_{13}N_{24}$
4	4	$N_{12}N_{23}N_{34}N_{41}N_{31}$
5	1	$N_{12}N_{23}N_{34}N_{41}N_{15}$
5	2	$N_{12}N_{23}N_{31}N_{14}N_{45}N_{51}$
6	1	$N_{12}N_{23}N_{34}N_{45}N_{56}N_{61}$

the effective Hamiltonian out to order t^6/U^5 differs from the one obtained earlier by Klein and Seitz,[16] which we believe to be in error. We remark that, for small N, the $H_{N,l}$ can be determined to all orders in t/U by comparing the spectrum of H' with the spectrum of the full Hubbard model for small clusters. For example, it follows from diagonalizing the Hubbard Hamiltonian for a two-site cluster that

$$H_{2,1}(\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2)=\frac{U}{16}\left[1-\left[1+\frac{16t^2}{U^2}\right]^{1/2}\right](1-\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2)\,, \quad (5)$$

in agreement with Table II.

Our approach to the t/U expansion has some similarity to earlier work[21] which should have been cited in our Brief Report, including that of Chao, Spałek, and Oleś[22] cited in the preceding Comment. The approach of the latter authors, which they call a canonical perturbation expansion (CPE), is to make a unitary transformation [Eq. (9) of the Comment] with S chosen to satisfy [Eq. (10) of the Comment],

$$i[S,U+T_0]=-T_1-T_{-1}\,. \quad (6)$$

The S defined by this equation is *not* the same as ours. Equation (6) defines the transformation used by many authors[1,23] to obtain the t/U expansion to leading order. In order to carry the expansion to high order, S must be specified order by order so as to eliminate unwanted terms from the transformed Hamiltonian. This procedure is described in detail in our Brief Report.[12] With S defined by Eq. (6) the transformed Hamiltonian has unwanted terms appearing at third and higher order in the hopping. Further errors are introduced because the transformed Hamiltonian is in the end constructed using a low-order approximation to this S [Eq. (12) of the Comment]. It follows that in the CPE, terms beyond leading order are based on uncontrolled approximations. Unlike the CPE, our approach takes the steps necessary to carry the unitary-transformation approach to the t/U expansion beyond leading order. The CPE is *not* equivalent to the approach used in our Brief Report.

Contrary to what is stated in the paragraph below Eq.

TABLE II. Spin operators appearing in the t/U expansion of the Hubbard model out to order t^6/U^5. Here $i\cdot j$ stands for $\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j$ and $x=t^2/U^2$.

N	l	$H_{N,l}(\boldsymbol{\sigma}_1,\ldots,\boldsymbol{\sigma}_N)/U$
2	1	$(-x/2+2x^2-16x^3)(1-1\cdot2)$
3	1	$-\frac{x^2}{2}+9x^3-2x^3(1\cdot2+1\cdot3)+\left[\frac{x^2}{2}-5x^3\right]2\cdot3$
3	2	$x^3[15-5(1\cdot2+1\cdot3+2\cdot3)]/6$
4	1	$x^2/8-3x^3+(-x^2/8+21x^3/4)(1\cdot2+1\cdot4+2\cdot3+3\cdot4)+(5x^2/8-69x^3/4)(1\cdot23\cdot4+1\cdot42\cdot3)$ $-(5x^2/8-54x^3/4)1\cdot32\cdot4+(-x^3/8+3x^2/2)(1\cdot3+2\cdot4)$
4	2	$x^3[15-25(1\cdot2+1\cdot3+1\cdot4)+27(2\cdot3+2\cdot4+3\cdot4)-7(1\cdot23\cdot4+1\cdot32\cdot4+1\cdot42\cdot3)]/24$
4	3	$x^3[-6+5(1\cdot2+3\cdot4)+1\cdot23\cdot4-1\cdot42\cdot3]/2$
4	4	$x^3[7-21(1\cdot2+1\cdot4+2\cdot3+3\cdot4)+152\cdot4+231\cdot3+37(1\cdot23\cdot4+1\cdot42\cdot3)-351\cdot32\cdot4]/8$
5	1	$x^3[3+3(1\cdot2+1\cdot3+1\cdot4)-2(2\cdot5+3\cdot5+4\cdot5)-9(2\cdot3+2\cdot4+3\cdot4)+11(1\cdot23\cdot4+1\cdot42\cdot3-1\cdot32\cdot4)$ $+10(-1\cdot23\cdot5+1\cdot32\cdot5+1\cdot34\cdot5-1\cdot43\cdot5+2\cdot34\cdot5-2\cdot43\cdot5+2\cdot53\cdot4)]/4$
5	2	$x^3[1+1\cdot2+1\cdot3+1\cdot4+1\cdot5+2\cdot3+4\cdot5+5(2\cdot4+2\cdot5+3\cdot4+3\cdot5)$ $-19(1\cdot24\cdot5+1\cdot34\cdot5+1\cdot42\cdot3+1\cdot52\cdot3)+492\cdot34\cdot5]/8$
6	1	$x^3[-3+3(1\cdot2+1\cdot3+1\cdot4+1\cdot5+1\cdot6+2\cdot3+2\cdot4+2\cdot5+2\cdot6+3\cdot4+3\cdot5+3\cdot6+4\cdot5+4\cdot6+5\cdot6)$ $-7(1\cdot23\cdot4+1\cdot23\cdot5+1\cdot23\cdot6+1\cdot24\cdot5+1\cdot24\cdot6+1\cdot25\cdot6+1\cdot34\cdot5+1\cdot34\cdot6+1\cdot35\cdot6+1\cdot42\cdot3$ $+1\cdot45\cdot6+1\cdot52\cdot3+1\cdot52\cdot4+1\cdot53\cdot4+1\cdot62\cdot3+1\cdot62\cdot4+1\cdot62\cdot5+1\cdot63\cdot4+1\cdot63\cdot5+1\cdot64\cdot5$ $+2\cdot34\cdot5+2\cdot34\cdot6+2\cdot35\cdot6+2\cdot45\cdot6+2\cdot53\cdot4+2\cdot63\cdot4+2\cdot63\cdot5+2\cdot64\cdot5+3\cdot45\cdot6+3\cdot64\cdot5)$ $+7(1\cdot32\cdot4+1\cdot32\cdot5+1\cdot32\cdot6+1\cdot42\cdot5+1\cdot42\cdot6+1\cdot43\cdot5+1\cdot43\cdot6+1\cdot52\cdot6+1\cdot53\cdot6+1\cdot54\cdot6$ $+2\cdot43\cdot5+2\cdot43\cdot6+2\cdot53\cdot6+2\cdot54\cdot6+3\cdot54\cdot6)+63(1\cdot23\cdot45\cdot6+1\cdot23\cdot64\cdot5+1\cdot32\cdot54\cdot6$ $+1\cdot42\cdot35\cdot6+1\cdot42\cdot63\cdot5+1\cdot52\cdot43\cdot6+1\cdot62\cdot34\cdot5+1\cdot62\cdot53\cdot4)-63(1\cdot23\cdot54\cdot6$ $+1\cdot32\cdot45\cdot6+1\cdot32\cdot64\cdot5+1\cdot42\cdot53\cdot6+1\cdot52\cdot34\cdot6+1\cdot52\cdot63\cdot4+1\cdot62\cdot43\cdot5)]/48$

(12) of the Comment, the effective Hamiltonian does, in general, involve hops which do not change the number of doubly occupied sites. The fact that such terms are absent in the CPE is a consequence of the approximation used for their S matrix [Eq. (12) of the Comment]. In addition the Comment suggests that odd-order terms vanish from the t/U expansion. This is true only for the case of a half-filled band for which it is a consequence of the particle-hole symmetry of the full Hubbard model and not of some property of the t/U expansion. (For example, one can show that $T_{-1}T_0T_1 \equiv 0$ at half-filling.) Again, the fact that such terms are absent at all band fillings in the CPE reflects the fact that it is valid only to second order in t. Similarly, the coefficient of t^4/U^3 for the expectation value of the effective Hamiltonian in the Néel state is in error in the CPE [see Eq. (14) of the Comment]. We see no basis for the argument following Eq. (14) that this error in the CPE in some way approximates the effects of quantum fluctuations about the Néel state. In any event, it is easy to include quantum fluctuations, at the level of spin-wave theory, using the correct effective Hamiltonian.

Having clarified the relation between the CPE and the unitary transformation discussed in our Brief Report, we turn to a discussion of our suggestion that higher-order terms in the t/U expansion favor spin-liquid states over antiferromagnetically ordered states. We felt that the frustrating effect of coupling to non-neighboring spins would clearly have this effect, and illustrated the tendency by comparing the shifts in the energies (Hamiltonian expectation values) of Néel and valence-bond states. We felt that this was a fair comparison since the two states may be regarded as resulting from the neglect of quantum fluctuations in antiferromagnetic states and spin-liquid states, respectively. While spin-wave theory can be used to describe quantum fluctuations in the ferromagnetic state there is no simple approximation available for spin-liquid states. Here, we provide further evidence in support of our expectation by showing that higher-order corrections reduce the spin-wave energies of the antiferromagnetic state.

Using the information in Tables I and II, we see that for the 2D square lattice, the effective Hamiltonian is

$$H=\frac{t^2}{U}\left(-2N+\tfrac{1}{2}\sum_{i,\tau}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_{i+\tau_1}\right)+\frac{t^4}{U^3}\left(3N-3\sum_{i,\tau_1}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_{i+\tau_1}+\tfrac{1}{2}\sum_{i,\tau_2}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_{i+\tau_2}+\tfrac{1}{2}\sum_{i,\tau_3}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_{i+\tau_3}\right.$$
$$\left.+\tfrac{5}{4}{\sum_{i,\tau_1,i''}}'(\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_{i+\tau_1})(\boldsymbol{\sigma}_{i''}\cdot\boldsymbol{\sigma}_{i''+\tau_1})-\tfrac{5}{4}{\sum_{i,\tau_2}}'(\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_{i+\tau_2})(\boldsymbol{\sigma}_{i''}\cdot\boldsymbol{\sigma}_{i''+\tau_2'})\right)+O(t^6/U^5)\,. \tag{7}$$

Here $i+\tau_1$ denotes one of the four near neighbors of site i which is separated from site i by a distance a, $i+\tau_2$ denotes one of the four second neighbors of site i which is separated from site i by a distance $\sqrt{2}a$, and $i+\tau_3$ denotes one of the four third neighbors of site i which is separated from site i by a distance $2a$. (The lattice constant is a which we now use as a length unit.) Note that for the antiferromagnetic state, sites i and $i+\tau_1$ are on opposite magnetic sublattices while sites i and $i+\tau_2$ and i and $i+\tau_3$ are on the same magnetic sublattice. The primes on the sums in the terms involving spin operators on four sites restrict the sites to an elementary square plaquette, so that there are two choices for i' in the fourth t^4/U^3 term whereas both i' and $i'+\tau_2'$ are fixed in the fifth t^4/U^3 term. Making a Holstein-Primakoff[24] transformation referenced to the Néel state, linearizing the resulting boson Hamiltonian[1,25] and replacing the site representation by a wave-vector representation leads to the Hamiltonian,

$$H=\frac{U}{t^2}H^{(2)}+\frac{t^4}{U^3}H^{(4)}+\frac{t^6}{U^5}H^{(16)}+\cdots\,, \tag{8a}$$

where

$$H^{(n)}=E_I^{(n)}+\sum_{\mathbf{k}}\left(\varepsilon^{(n)}(\mathbf{k})a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}+\frac{\lambda^{(n)}(\mathbf{k})}{2}(a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger}+a_{-\mathbf{k}}a_{\mathbf{k}})\right)\,, \tag{8b}$$

$$E_I^{(2)}=-4,\quad E_I^{(4)}=24,\quad \varepsilon^{(2)}(\mathbf{k})=8\,,$$
$$\varepsilon^{(4)}(\mathbf{k})=-104+4(\cos 2k_x+\cos 2k_y)-16[\cos(k_x+k_y)+\cos(k_x-k_y)]\,,$$
$$\lambda^{(2)}(\mathbf{k})=4[\cos(k_x)+\cos(k_y)]\,,$$
$$\lambda^{(4)}(\mathbf{k})=-64[\cos(k_x)+\cos(k_y)]\,.$$

Here the $E_I^{(n)}$ are the coefficients in the expansion of the Ising energy. Making a Bogoliubov transformation[1,25] we obtain

$$H=H_I-\tfrac{1}{2}\sum_{\mathbf{k}}[\varepsilon(\mathbf{k})-E(\mathbf{k})]+\sum_{\mathbf{k}}d_{\mathbf{k}}^{\dagger}d_{\mathbf{k}}E(\mathbf{k})\,, \tag{9}$$

where $\varepsilon(\mathbf{k})=\sum_k\varepsilon^{(2k)}(\mathbf{k})$, $\lambda(\mathbf{k})=\sum_k\lambda^{(2k)}(\mathbf{k})$, and the spin-wave energies are given by

$$E(\mathbf{k})=\sqrt{\varepsilon^2(\mathbf{k})-\lambda^2(\mathbf{k})}$$
$$=\frac{t^2}{U}E^{(2)}(\mathbf{k})+\frac{t^4}{U^3}\left[\frac{\varepsilon^{(2)}(\mathbf{k})\varepsilon^{(4)}(\mathbf{k})-\lambda^{(2)}(\mathbf{k})\lambda^{(4)}(\mathbf{k})}{E^{(2)}(\mathbf{k})}\right]+O(t^6)$$
$$=\frac{8t^2}{U}\sqrt{1-\gamma^2(\mathbf{k})}-\frac{t^4}{U^3}\left[128\sqrt{1-\gamma^2(\mathbf{k})}+\frac{24[\delta(\mathbf{k})-1]}{[1-\gamma^2(\mathbf{k})]^{1/2}}\right]+O(t^6)\,. \tag{10}$$

In Eq. (10) $\gamma(\mathbf{k}) = [\cos(k_x) + \cos(k_y)]/2$ and

$$\delta(\mathbf{k}) = \{16[\cos(k_x + k_y) + \cos(k_k - k_y)] - 4[\cos(2k_x) + \cos(2k_y)]\}/24 .$$

In the long-wavelength limit Eq. (10) leads to

$$E(\mathbf{k}) = \frac{4t^2\sqrt{2}k}{U}\left[1 - \frac{14t^2}{U^2} + \cdots\right] \tag{11}$$

showing that the spin-wave velocity is reduced by the t^4 terms. Similarly on the boundary of the magnetic Brillouin zone,

$$E(k_x = \pi, k_y = 0) = \frac{8t^2}{U}\left[1 - \frac{8t^2}{U^2} + \cdots\right] \tag{12}$$

and

$$E(k_x = \pi/2\,,\ k_y = \pi/2) = \frac{8t^2}{U}\left[1 - \frac{14t^2}{U^2} + \cdots\right] \tag{13}$$

so that the spin-wave energies are reduced throughout the Brillouin zone.

The reductions in excitation energy discussed above reflect a weakening of the stability of the antiferromagnetic state. The above calculation provides additional evidence in support of the conclusion of our Brief Report, namely that higher-order terms in the t/U expansion can "play a role" in favoring spin-liquid states over Néel states. Contrary to the impression left in the Comment we did *not* attempt to make any conclusions concerning what happens far from the localized limit where the t/U expansion is very slowly convergent. We *did* point out that corrections to the Heisenberg model for the half-filled localized limit become important at values of t/U relevant to models for high-T_c superconductors. The corrections tend to frustrate the antiferromagnetic order of the ground state and will make it easier for the order to be destroyed by the additional frustration introduced by holes.

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