Comment on ''Density-matrix renormalization-group method for excited states''

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In a recent paper [Phys. Rev. B **59**, 9699 (1999)], Chandross and Hicks claim to present a new density-matrix renormalization group (DMRG) method for dealing with excited-states of quantum lattice models. The proposed improvement to the DMRG—the inclusion of excited-state wave functions *in addition* to the ground state in the density matrix when calculating excitations—is in fact standard practice, is clearly stated in White's original papers, and has been used repeatedly by many groups to study excited states. The authors apply the method to the extended, dimerized Hubbard model for conjugated polymers. The criteria for determining whether states are bound or not are assessed. The authors claim that their results show that the optically important ''$1B_u$'' state is bound (excitonic), in contrast to a previous study. However, the discussion is qualitative, and the authors arrive at conclusions on the basis of results for one lattice size only. We show that when the criterion of Chandross and Hicks is developed into a quantitative definition of particle-hole separation, with the finite-size dependence analyzed, the implication is that the $1B_u$ state is unbound, at least in the sense of the density-density correlation function, in keeping with the conclusions of a previous study. [S0163-1829(00)01343-6]

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In a recent paper,[1] Chandross and Hicks claim to present a new density-matrix renormalization-group (DMRG) method[2,3] for dealing with excited states of quantum lattice models. They apply the method to the dimerized, extended Hubbard model for conjugated polymers. They claim that a previous study[5] of this model is flawed because it uses a ''conventional'' DMRG method which does not handle excitations correctly. The improvement that they suggest is to form a density matrix not only from the ground state, but from all the states being targeted in the calculation. This is in fact standard practice in DMRG calculations of excited states and the structure of the density matrix required to target excited states is given in White's original papers on the method.[3,4] It has been used by many authors to target excitations in a variety of quantum lattice models (see, e.g., Ref. 6) and was *certainly* used in Ref. 5 when various excitation energies and correlation functions were calculated for the extended Hubbard model. The comparisons presented in Figs. 1 and 2 of Ref. 1, between the ''conventional'' DMRG and Chandross and Hick's ''improvement'' are therefore of limited value, as, to the best of our knowledge, all DMRG studies of excited states to date have incorporated the targeted excitations into the density matrix.[7] Unfortunately, a slightly different value for the Coulomb V is used in Ref. 1 so a direct comparison with the results (e.g., for energies) tabulated in Ref. 5 is not possible. We have run a DMRG program which uses the algorithm used in Ref. 5 for targeting excited states with the parameters $U=3t$, $V=t$, $\delta=0.1$, used in Ref. 1, and found good agreement for the energies and correlation functions with the results plotted in Figs. 1(a) and 2(a) of Ref. 1. For instance, we plot the $1B_u$ and mA_g (Ref. 8) energies as functions of the lattice size N in Fig. 1. The results compare very well with Fig. 1(a) of Ref. 1.

In Ref. 1 Chandross and Hicks also examine criteria for deciding whether a particular excitation is bound (excitonic)

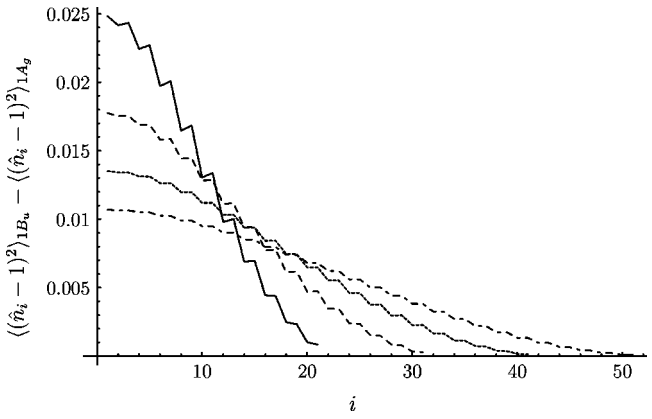

FIG. 2. The average number of doubly occupied sites of the $1B_u$ state relative to the ground state at distance i from the center of the chain for various lattice sizes N.

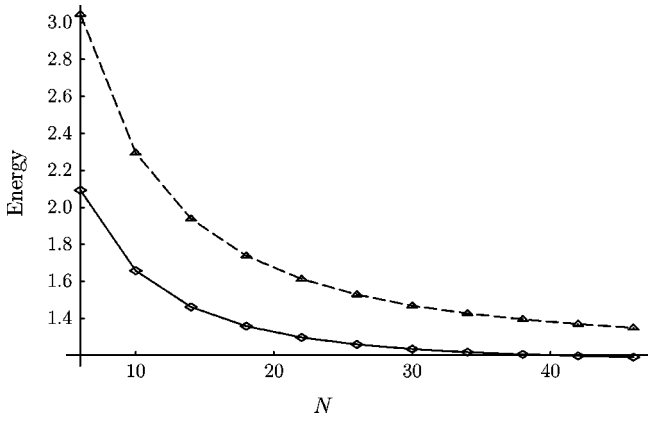

FIG. 1. The energies of the $1B_u$ (diamonds) and mA_g (triangles) states of the dimerized, extended Hubbard model as a function of the lattice size N for the parameter set used in Ref. 1. The number of states retained per block (Refs. 2 and 3) is $m=270$.

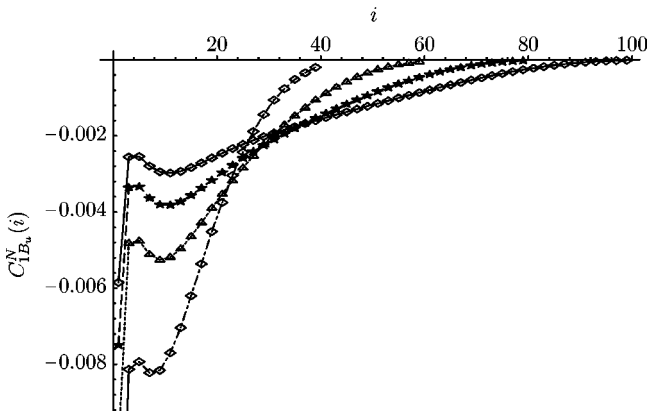

FIG. 3. The averaged, centered, odd-site correlation function (relative to the ground-state value, as defined in Ref. 5) for the $1B_u$ state for $N=42$ (diamonds), 62 (triangles), 82 (stars), and 102 (solid diamonds).

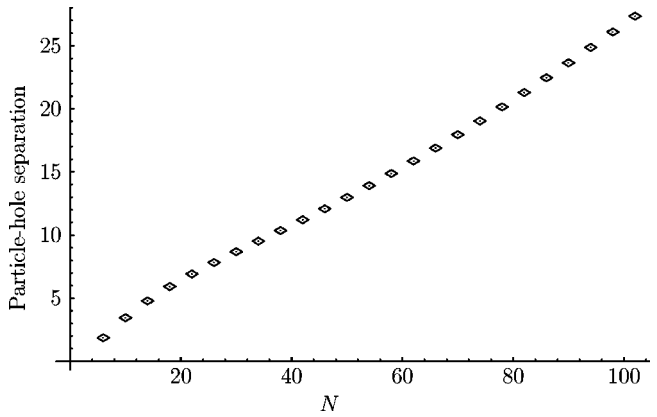

FIG. 4. The (reduced) average particle-hole separation, as defined in Ref. 5 by using $|C^N_{1B_u}(i)|$ as a probability distribution, for the $1B_u$ state. Note the linear increase with N.

or not. They claim that the average particle-hole separation, defined in Ref. 5 in terms of the density-density correlation function, is ''too approximate'' a quantity to determine whether a state is bound or not. They argue that by inspecting the centered correlation function as a function of distance (together with the profile of doubly occupied sites along the chain), for one particular lattice size ($N=36$ sites), one can see that the $1B_u$ and mA_g states are ''different'' in that the $1B_u$ (mA_g) has its strongest particle-hole correlations at short (long) distances. However, Chandross and Hicks do not present an alternative quantitative definition of the particle-hole separation, based on this observation. In Ref. 5, on the other hand, it is argued that a definition of particle-hole binding must take into account the way in which correlations *scale* with lattice size N. In Ref. 5 it is argued that this scaling is different for bound and unbound excitations, and that the scaling of the average particle-hole separation with N is but one manifestation of this.

Suppose we wish to take the average double occupancy of the $1B_u$ state (relative to the ground state) along the chain $\langle(\hat{n}_i-1)^2\rangle_{1B_u}-\langle(\hat{n}_i-1)^2\rangle_{1A_g}$ as an example [Fig. 2(a) in Ref. 1]. In Fig. 2 we plot this quantity for various lattice sizes N. We see that, although the concentration of doubly occupied sites is greatest in the middle of the chain, the distribution spreads out as N is increased. The area under these curves rapidly converges to a nonzero value (≈ 0.538) as $N\rightarrow\infty$. This shows that the number of pairs of particles and holes in the $1B_u$, relative to the number in the ground state, approaches a constant. Our results could indicate that particle-hole pairs separate as N is increased and are hence unbound, or they may simply indicate dispersion of a bound exciton in the $1B_u$.

To address this we again consider the averaged, centered, odd-site correlation function $C^N_{1B_u}(i)$ (again relative to the ground-state value), defined in Ref. 5 and plotted for $N=36$ in the inset to Fig. 2(a) in Ref. 1. In Fig. 3 we plot this quantity for a number of values of N. We see that, although the correlations are generally strongest at short distances, they become increasingly spread out, and hence the particle-hole pair becomes increasingly separated, as N is increased. Indeed, if one utilizes $|C^N_{1B_u}(j)|$ to define a probability distribution for the particle-hole separation, as in Ref. 5, then one finds that the average particle-hole separation grows linearly with N, as shown in Fig. 4. We note that any use of the density-density correlation function to describe particle-hole separation and the nature of exciton binding of excited states in the extended Hubbard model is merely plausible rather than rigorous,[9] but Chandross and Hicks[1] do not offer an alternative *quantitative* definition of the particle-hole separation to the ones provided in Ref. 5.

To summarize, Chandross and Hicks claim that because the $1B_u$ and mA_g have their greatest particle-hole correlations at short and long distances, respectively (on the $N=36$ lattice), the $1B_u$ is bound and the mA_g is unbound. We would argue that it indicates that the particle-hole potential is more strongly attractive for the $1B_u$ state than for the mA_g. However, from the plausible, quantitative definition of the particle-hole separation given above, it would appear that the attraction between the particle and hole in the $1B_u$ state is not strong enough to bind them, and their separation increases throughout the range of lattice sizes studied.

Finally, we consider the structure of the density matrix when targeting excitations such as the mA_g and nB_u. Chandross and Hicks argue that only four states—the $1A_g$ (ground state), the $1B_u$, the mA_g, and the nB_u—need be included in the density matrix. Our examinations of the dipole moments between the A_g states and the $1B_u$ indicate that this approach is probably reasonable for the mA_g which is well defined.

TABLE I. Transition moments with the mA_g states for the first five B_u states (i.e., $\langle jB_u|\hat{\mu}|mA_g\rangle$ for $j=1,\ldots,5$) for $N=6$, 10, 14, and 18. Note that there is no clearly defined nB_u state.

N	$j=1$	$j=2$	$j=3$	$j=4$	$j=5$
6	2.32	0.76	0.30	1.87	1.39
10	3.48	1.77	0.38	3.30	0.06
14	4.45	3.15	0.04	3.98	0.10
18	5.33	4.73	0.67	4.24	1.73

That is, there is a reasonably abrupt jump in the magnitude of the dipole moment $\langle 1B_u|\hat{\mu}|jA_g\rangle$ at $j=m$. As shown in Ref. 5, this coincides with jump in the ionicity (the average number of doubly occupied sites) and in the particle-hole separation. However, the nB_u state is less well defined in that there can be a number of B_u excitations that have a strong dipole moment with the mA_g. This can be seen in Table I where we list the dipole moments $\langle jB_u|\hat{\mu}|mA_g\rangle$ for $N=6$, 10, 14, and 18, for the first five B_u states. Note that in no case is the nB_u state clearly defined, though there is a general trend whereby the $2B_u$ increases its relative dipole strength with the mA_g at the expense of the $4B_u$. Our contention here, as proposed in Ref. 5, is that, at least in terms of dipole moments or the density-density correlation function, the $1B_u$ state is the threshold of unbound states in the B_u sector and the ''nB_u'' is not well defined for this model.

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[7] We note further that Chandross and Hicks (Ref. 1) claim that including only the ground state in the density matrix leads to ''grossly incorrect quantitative'' results for excited states. Although it is illogical to include only the ground state in the density matrix when calculating excited states, for a given lattice size (e.g., $N=36$) it is in fact a mathematical certainty that the DMRG results for the excitations with a simple density matrix formed solely from the ground-state projection operator will converge to the exact results as the number of states retained per block, m, is increased. The phrase ''less rapidly converging'' is therefore more appropriate, as DMRG results should not be accepted if they have not converged with m.
[8] The mA_g state is defined to be the excited state in the ground-state symmetry sector that has the largest dipole moment with the $1B_u$ state, which is the lowest-energy state in the dipole-allowed symmetry sector.
[9] M. Yu. Lavrentiev and W. Barford, Phys. Rev. B **59**, 15 048 (1999). In this paper an alternative quantitative definition of the particle-hole separation is proposed using molecular orbital operators. In terms of this definition there are indications that the $1B_u$ particle-hole separation grows less rapidly than linearly with N.