Long-range Coulomb repulsion and finite-size approximations

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We show that when sizable long-range Coulomb repulsion is included in numerical studies of small clusters, different sizes and shapes should be considered to obtain a ground state which represents correctly the physics of the ground state (GS) in the thermodynamic limit. Also cutting off the interaction at small distances might lead to a spurious GS. In particular the GS reported in the literature for the two-dimensional three-band Hubbard model in the static limit at hole concentration $x = 0.25$ is in fact a slightly excited state when an important (screened or not) Coulomb repulsion at all distances is included.

In spite of the fact that the number of carriers in high-T_c compounds is very low and therefore the screening of the Coulomb interactions is expected to be poor, very few studies of the electronic structure of these materials include electron repulsions beyond nearest neighbors.[1–5] This is due to the difficulty of the theoretical study of the strongly correlated systems even without long-range Coulomb interactions. The effect of the latter is expected to reduce or suppress the regions of parameters for which phase separation takes place, favoring superconductivity in some cases.[1–6] However, a recent numerical study shows that for certain doping levels, the regions of phase separation are replaced by charge-density waves instead of superconductivity.[5] Classical models including long-range Coulomb repulsions were successful in explaining the essential features of the ordering of oxygen atoms in $YBa_2Cu_3O_{6+x}$.[7–10]

In a recent paper,[4] Riera and Dagotto have studied the effect of long-range repulsions in the three-band Hubbard model.[11,12] This model is believed to contain the essential ingredients of high-T_c superconductors, and has been the starting point in the derivation of effective one-band models,[14] like the widely studied t-J model[15] and generalizations of it.[3,14] The interaction included is a screened Coulomb interaction:

$$H_c = \frac{V}{2} \sum_{lm} \frac{n_l n_m}{d_{lm}} e^{-d_{lm}/\lambda} , \qquad (1)$$

where d_{lm} is the distance between sites l and m, n_l and n_m are the hole occupations at these sites, and λ is the screening length. The competition among superconductivity, charge-density waves, and phase separation has been studied in chains of six unit cells. The atomic limit (zero hopping energy) was studied in chains of 12, 24, and 48 CuO cells and in periodic square clusters containing 4×4 and 6×6 CuO_2 unit cells.[4]

It is important to note that to capture the correct physics in the thermodynamic limit, for certain hole concentrations, it is necessary to consider other cluster sizes. For example, in the two-dimensional (2D) case, in the atomic limit, for hole doping $x = 0.2$ and small V, the ground state has unit cell $\sqrt{5} \times \sqrt{5}$, and cannot be described by the calculations of Ref. 4. In fact, for small V in the atomic limit, all Cu atoms have one hole and the problem reduces to distribute x unit charges per unit cell in the square sublattice of oxygen atoms. The ground state of this problem has been studied in detail in Refs. 16 and 17. In general, for $x \leq 2/3$, the added charges tend to arrange themselves into the smallest possible deformation of a perfect hexagonal simple lattice (type $p6m$), needed to place these charges onto the oxygen sublattice (see Table I of Ref. 16). The ground state in 1D has been obtained exactly by Pokrovsky and Uimin[18] and Hubbard.[19] For $x = 1/3$ or $x = 2/3$, the unit cell has length 3 and is contained exactly in the chains studied in Ref. 4. Thus the problem of the correct choice of the cluster size does not affect the ground state of these chains, as a representation of the system in the thermodynamic limit.

Since the on-site oxygen Coulomb repulsion U_p is believed to be larger than two times the nearest-neighbor CuO repulsion,[20] the regime of small V discussed above in the atomic limit is probably the more realistic one, even if the Cu-O charge-transfer energy Δ were very small. For increasing V and small λ there is a second regime in 2D, not discussed in Ref. 4 in which holes in nearest-neighbor Cu and O atoms are avoided, but no double occupancy of O atoms exists.[21] For sufficiently large λ and V, and small or negative U_p, the holes enter in pairs in O atoms, and the problem in the static limit reduces to distribute $y = (1+x)/2$ charges of magnitude $2e$ in the oxygen sublattice, minimizing Eq. (1). Again, this problem is equivalent to that studied in Refs. 16 and 17 and in Refs. 18 and 19 in 1D. According to Ref. 16, the structure of the doubly occupied O atoms in the ground state for $x = 1/4$ has the form shown in Fig. 5 of Ref. 16, which is reproduced in Fig. 1(a) here. Instead, for large V and λ, the structure shown in Fig. 1(d) of Ref. 4, reproduced here in Fig. 1(b), has been obtained by simulated annealing in a periodic 4×4 cluster (the structures of Fig. 1 cannot be obtained in a periodic 6×6 cluster). In the following we denote the structures shown in Figs. 1(a) and 1(b) as A and B, respectively.

One might suspect that the simulated annealing procedure led to a metastable state, as it happened in similar problems,[22] but this is not the case. Our investigation (explained below) allows us to conclude that the structure B is a slightly excited state, but it becomes

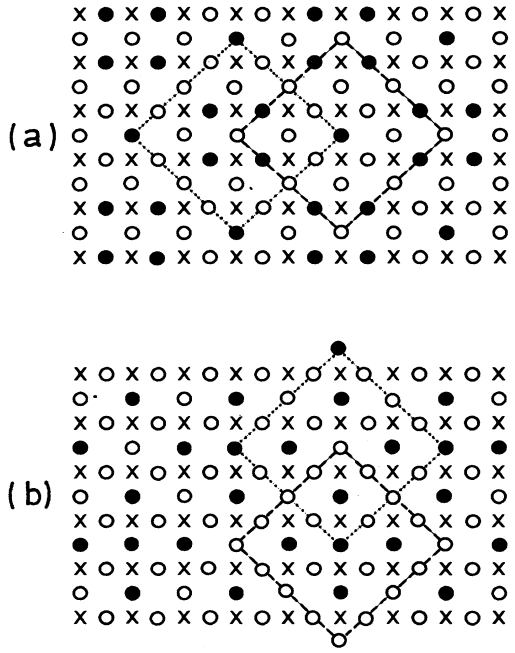

FIG. 1. (a) Ground state of the three-band Hubbard model (Refs. 6 and 12) supplemented with long-range repulsions [Eq. (1)] in the atomic limit (zero hopping), for doping level $x = 0.25$ and large V and λ. Crosses denote Cu atoms without holes (Cu^{+1}), empty circles correspond to empty O atoms [O^{-2} and solid circles denote the double-occupied O^0 atoms (DOO)]. Dashed and dotted lines show possible choices of the unit cell. (b) Same as in (a) when all interactions at distances $\mathbf{R} = (R_x, R_y)$ lying out of the square $|R_x| = |R_y| = 2$ lattice parameters, are neglected and those lying on the boundary are reduced by a factor 1/2.

the ground state if all the interactions between one atom and any other lying outside a 4×4 square centered at the first are neglected, while the interactions with atoms lying on the boundary are counted with a factor 1/2. This is actually equivalent to the procedure used in Ref. 4 to

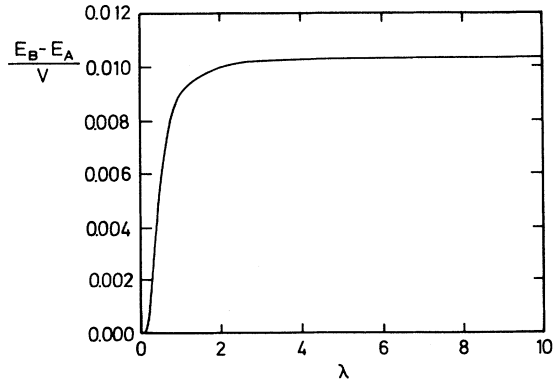

FIG. 2. Difference between the energy of the structures shown in Figs. 1 (b) and 1(a) (denoted B and A, respectively) as a function of the screening length λ for the model including all interactions [see caption of Fig. 1(a)].

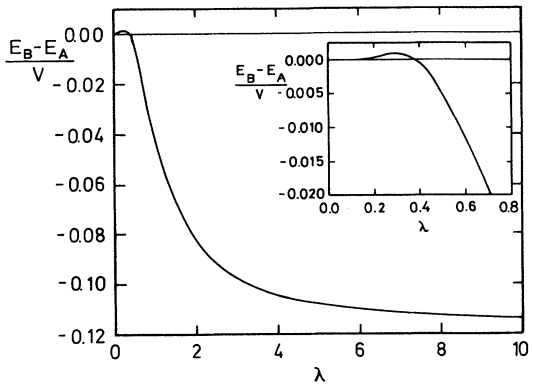

FIG. 3. Same as in Fig. 2 when the interactions are cut off as in Ref. 4 [see caption of Fig. 1(b) and text]. The inset shows the region of small λ in more detail.

count the interactions as explained at the end of Sec. I of the paper.[4] We had already found in thermodynamic studies of $YBa_2Cu_3O_{6+x}$ that cutting off the interactions beyond fifth nearest-neighbor O atoms might lead to spurious ground states.[23]

In Fig. 2 we show the difference between the energies of structures B and A when all interactions are included. Figure 3 displays the corresponding result using the procedure of Ref. 4 to cutoff interactions. The difference is apparent and can be understood in simple terms: when all interactions are included, it has been found previously that for $y < 2/3$, and any λ, the ground state is obtained minimizing successively the number of first, second, third, and further nearest-neighbor (NN) double-occupied O atoms (DOO).[16,17] Both structures A and B have no first NN DOO and half second NN DOO per Cu. The number of third NN DOO (at distance $\sqrt{2}$) is 1/2 for structure A and 1 for B and this explains why the energy of A is lower. A has one fourth NN DOO per Cu [distance $(5/2)^{1/2}$] and no fifth NN DOO (distance 2) and these numbers are permuted for structure B. With

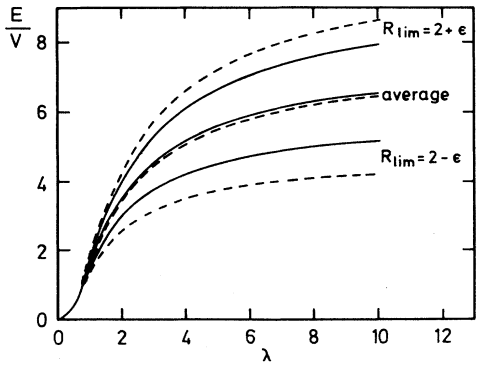

FIG. 4. Energy of the structure A (full line) and B (dashed line) as a function of λ, when all interactions at distances $\mathbf{R} = (R_x, R_y)$ lying out of the square $|R_x| = |R_y| = R_{\rm lim}$ are neglected, for two different quite close values of $R_{\rm lim}$ (ϵ is a positive infinitesimal). The average of the energies corresponds to the cutoff procedure of Riera and Dagotto (Ref. 4).

the method of Ref. 4, only half of the interaction between fifth NN DOO is added. In spite of the fact that B has a larger number of third NN, it has replaced the fourth NN DOO of A by fifth NN DOO with a considerable (artificial) reduction of energy. Also the interactions at distance $\sqrt{5}$ and $2\sqrt{2}$ of B are affected by a factor 1/2 with the method of Ref. 4 (they also lie at the boundary of the 4×4 square of included interactions). As a consequence, for large λ, the energy of B is smaller than that of A. For sufficiently small λ, the effect of the third NN DOO dominates and A has lower energy (see Fig. 3).

In previous studies including all interactions, for fixed composition, the ground state was always the same for all values of λ.[16,17] Thus it is not surprising that the energy of B is always larger than that of A. However, for small values of λ phase separation can take place.[16]

To show how the energy difference is affected when an abrupt cutoff is used (as in Ref. 5), we show in Fig. 4 the energy as a function of λ for two different but very similar areas of included interactions (AII), such that for each O atom, the repulsions between it and any charge lying outside the AII centered at the atom is neglected. In agreement with previous studies,[23] structures with O atoms close to the border of each of the AII but outside them are considerably reduced in energy and artificially favored by the cutoff procedure.

In 1D the convexity of the interaction is essential to obtain the correct ground state[18,19]: each interaction V_n at a distance n should satisfy the inequality $2V_n < V_{n-1} + V_{n+1}$. If all interactions are replaced by zero for $n \geq m$, then this condition is violated for $n = m - 1$ or $n > m$ and the ground state for certain doping levels is modified.

Although the method of counting the interactions used in Ref. 4 has led to a ground state which does not correspond to the correct one in the thermodynamic limit (the structure B instead of A), the conclusion that phase separation is inhibited by the long-range repulsions is not affected. We believe also that the 1D results of Sec. III of Ref. 4 are not affected seriously either by the choice of the cluster or the above mentioned method. However, several aspects of the physics of the three-band Hubbard model which might be important in 2D are missing in 1D. For example to represent the physics of 2D-excitonic states in 1D, one has to assume an unrealistic very low on-site oxygen repulsion U_p.[13]

In summary, to obtain the ground state of a model with long-range repulsions in cluster calculations, which reproduces correctly the physics of the thermodynamic limit it is necessary to study clusters with different shapes and to include the interactions between an atom and all periodic images of the second one. The atomic limit is a good guide to determine the right shape of the clusters.

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