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TITLE: PAIRING ON SMALL CLUSTERS IN THE PEIERLS-HUBBARD MODEL:
IMPLICATIONS FOR C_{60}

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PAIRING ON SMALL CLUSTERS IN THE PEIERLS-HUBBARD MODEL: IMPLICATIONS FOR C₆₀

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ABSTRACT

Inclusion of electron-phonon interactions can substantially modify the conclusions about pairing obtained from models including only electron-electron interactions. We study pairing within the Peierls-Hubbard model for electron- and hole-doped analogs of C₆₀ accessible to exact diagonalization techniques (cube, truncated tetrahedron, *etc.*), and discuss the extrapolation from these small system calculations to C₆₀.

INTRODUCTION

Within the Hubbard model,

$$H_{Hub} = \sum_{\langle ij \rangle, \sigma} t_{ij} c_{i, \sigma}^{\dagger} c_{j, \sigma} + U \sum_i n_{i, \uparrow} n_{i, \downarrow} \quad [1]$$

where $\langle ij \rangle$ are nearest neighbors and $t_{ij} = -t_0$, pairing of the Cooper type is supposed to occur if the electronic energy needed to add one electron (hole) is more than half the energy to add two, *i.e.*, if the quantity

$$\Delta E = 2E(N \pm 1) - E(N) - E(N \pm 2) \quad [2]$$

is positive. As approximate analytic techniques have been known to lead to qualitatively incorrect predictions for solid state systems with strong electron correlation, exact calculation for (small) finite clusters is often employed. Pairing is found for the cube and the truncated tetrahedron (1-3), which has been interpreted as evidence that electron-electron induced pairing occurs for C₆₀. However, it is wellknown that the ground states of one-dimensional Hubbard rings may be substantially altered when electron-phonon interactions are included. To investigate the effects of electron-phonon coupling, we add a variable lattice distortion δ_{ij} from uniform bond length d_0 constrained by an effective spring κ , and use $t_{ij} = -t_0 + \alpha \delta_{ij}$ in the Hamiltonian Eq. 1, yielding the Peierls-extended Hubbard Hamiltonian.

$$H_{PH} = \sum_{\langle ij \rangle, \sigma} [-t_0 + \alpha \delta_{ij}] (c_{i, \sigma}^{\dagger} c_{j, \sigma} + c_{j, \sigma}^{\dagger} c_{i, \sigma}) + U \sum_i n_{i, \uparrow} n_{i, \downarrow} + \frac{1}{2} \kappa \sum_{\langle ij \rangle} (\delta_{ij} - d_0)^2. \quad [1']$$

We consider both δ_{ij} externally fixed and δ_{ij} self-consistently chosen to minimize the total energy (thus depending on filling). Since the numerical exact techniques we employ are limited to small system sizes, we must extrapolate from calculations on small systems to C_{60} . We point out below that straightforward extrapolations are not always valid, and can lead to erroneous theoretical predictions, by examining first the case of pairing in Hubbard rings. We then apply this insight to interpret the implications for C_{60} of the small cluster pairing data.

PAIRING IN 1D HUBBARD RINGS

Pairing as defined in Eq. 2 can occur in one-dimensional periodic rings with $N=4n$ atoms, but not in $N=4n+2$ rings (4). As small $4n$ -rings undergo Jahn-Teller distortions, the restriction to $4n$ rings is very suggestive. As shown in Fig. 1, the pairing disappears for Jahn-Teller distorted rings. To understand this better, we calculated the effect of the Hubbard U on the Jahn-Teller distortion of the $4n$ - and $(4n-1)$ - electron, $4n$ -site systems. These results are shown in Fig. 2. It is seen that U strongly reduces the gain in energy for the Jahn-Teller distortion of the even $(4n)$ electron system, but the distortion of the odd numbered $(4n-1)$ electron system remains virtually unaffected. This then gives us the mechanism of the pairing. The information regarding the tendency to distort is built into the wave function in the form of the various bond order parameters. By not letting the system distort, we are overestimating the energies in each case, but as Fig. 2 indicates, this overestimation is much stronger for the odd electron case than for the even electron case. The calculated energy required to add one hole (or electron) to the undistorted system is therefore too high, thereby giving "pairing". This same mechanism can explain "pairing" in higher dimension.

PAIRING IN 3D HUBBARD CLUSTERS

For the three-dimensional cluster geometries shown in Fig. 3, we have calculated the pair binding energy ΔE . The result is shown for $N=12$ in Fig. 4. Pairing is found in both a region near $U=0$ and a region at large U . Similar to the ring case, the pairing at small U appears to be related to the presence of energy level degeneracies at $U=0$. In Fig. 5 we demonstrate for the truncated tetrahedron that pairing again disappears when a fixed Jahn-Teller distortion is added, clearly indicating the relationship between pairing at small U and the degeneracies in the single particle limit: in the absence of distortion, the energy to add one hole or electron is being overestimated. We also show the results of our calculation for the truncated tetrahedron with the δ_{ij} which self-consistently minimize the total energy and thus depend on filling (d_0 and κ were chosen to depend on U such that the minimum energy distortion at $N_{el}=N$ was independent of U : $\delta_{ij}=0.1$ on the triangular faces and $\delta_{ij}=-0.2$ on the bonds connecting triangular faces). Here we find a greatly enhanced pairing, though maximum at $U=0$, indicating again that it is driven by the Jahn-Teller distortion and not the electron-electron interaction ("bipolaronic" pairing).

IMPLICATIONS FOR C_{60}

Pairing as defined in Eq. 2 occurs at small U for an N site system only if the $(N \pm 1)$ electron system has a strong tendency to have a Jahn-Teller distortion. This pairing is destroyed when a fixed Jahn-Teller distortion is introduced, and is related to the suppression of Jahn-Teller distortion. Since the Jahn-Teller distortion results from the discrete level degeneracies in finite molecules, this small U pairing is a finite size effect. As C_{60} is a finite molecule, this "on-ball" pairing mechanism cannot be ruled out. For a Hubbard model where the "site" is a C_{60} in the molecular crystal rather than a C on an individual ball, the arguments here and in Ref. 3 indicate the absence of pairing: this small U pairing mechanism does not operate in the infinite C_{60} molecular crystal (5).

Pairing at large U is unrelated to the tendency to have Jahn-Teller distortion, and may or may not be accompanied by pairing at small U (3). The occurrence of such large U pairing in nonbipartite systems (*e.g.*, geometry of Fig. 3b) indicates that it is also apparently unrelated to antiferromagnetism. It may be related to energy level crossings. We are investigating whether the large U pairing is also a finite size effect, and how pairing in the presence of long-range Coulomb interactions (1) is affected by inclusion of electron-phonon interactions. Finally, we note that the definition of pairing in Eq. 2 clearly is not synonymous with superconducting pairing; it may indicate *e.g.* phase segregation (3,6) instead.

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Fig. 1 Pair binding energies ΔE for undistorted (solid) and Jahn-Teller distorted (dashed) (a) $N=4$ and (b) $N=8$ rings.

Fig. 2. The gain in electronic energy on Jahn-Teller distortion for the $N = 4$ ring, as a function of Hubbard U . The solid and dashed curves correspond to the 4 and 3 (5) electron cases respectively.

Fig. 3. Three-dimensional cluster geometries for which the pair binding energy has been calculated: (a) drum, (b) twisted drum, and (c) truncated tetrahedron.

Fig. 4 Pairing in undistorted $N=12$ site electron- (solid) and hole- (dashed) doped clusters for the geometries of Fig. 3. Note there are two regions, small and large U , where pairing can occur.

Fig. 5. Pair binding energies for the uniformly distorted truncated tetrahedron for electrons (solid) and holes (dashed): (a) Peierls-distorted and (b) Jahn-Teller distorted. The pair binding energies are also shown for (c) the self-consistently distorted truncated tetrahedron. Notice the absence of pairing at small U for the Jahn-Teller distortion, and that the pairing is strongest at $U=0$ for the self-consistent distortion.

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