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Off-diagonal interactions, Hund's rules, and pair binding in C_{60}

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We have studied the effect of including nearest-neighbor electron-electron interactions, in particular the off-diagonal (non-density-density) terms, on the spectra of truncated tetrahedral and icosahedral Hubbard molecules, focusing on the relevance of these systems to the physics of doped C_{60}. Our perturbation theoretic and exact diagonalization results agree with previous work in that the density-density term suppresses pair binding. However, we find that for the parameter values of interest for C_{60}, the off-diagonal terms *enhance* pair binding, though not enough to offset the suppression due to the density-density term. We also find that the critical interaction strengths for the Hund's rules violating level crossings in C_{60}^{-2}, C_{60}^{-3}, and C_{60}^{-4} are quite insensitive to the inclusion of these additional interactions.

I. INTRODUCTION

To account for the superconductivity of the alkali fullerides A_3C_{60} (A=K, Rb, Cs) Chakravarty, Gelfand, and Kivelson[1] (CGK) suggested a mechanism involving purely electronic (*e-e*) interactions. They showed how this might come about by studying a simplified (Hubbard) model of a C_{60} molecule and arguing that for a range of the atomic parameters the molecule exhibits "pair binding:"[2] that is, a pair of uncoupled monoanions, $C_{60}^- + C_{60}^-$, is unstable with respect to charge disproportionation into $C_{60} + C_{60}^{-2}$ (and likewise for trianions and quintanions, *mutatis mutandis*). Weak electronic overlaps between molecules would then lead to superconductivity. The pair-binding phenomenon was apparently related to an unusual feature of the low-lying spectra of C_{60} di-, tri-, and quadranions (all of which have three icosahedral multiplets of states which are themselves degenerate in the $U = 0$ limit): namely, that Hund's rules were violated and the ground states were those of *minimum* spin and *minimum* orbital degeneracy.

The mechanism of superconductivity in the fullerides is currently not fully understood[3] and the relevance of the work of CGK to that problem is controversial; we will not shed any light on that question directly. Our interest lies in examining the robustness of the pair binding and violations of Hund's rules found by CGK to *e-e* interactions not included in their model but expected to exist in the real materials. This issue is by no means straightforward since the quantities of interest are small differences of large energies. Our work was stimulated in part by work of Goff and Phillips,[4] who studied the effect of further-neighbor density-density interactions on pair binding.

We begin by defining the extension of the Hubbard model studied in this paper and the quantities of interest. Next, we present the results of perturbative calculations for C_{60}. We discuss the assignments of values for the various parameters and the corresponding values for the pair-binding energy and level splittings. To assess the validity of our approach we follow the lead of White *et al.*[5] and present both perturbative and exact diagonalization results for C_{12}, a mythical homolog of C_{60} that has a similar level structure at the Fermi energy. We conclude by summarizing our results and their implications for understanding the properties of the fullerides.

II. THE EXTENDED HUBBARD MODEL

We study the Hamiltonian

$$H = -t\sum_{\langle ij\rangle}(h_{ij}+h_{ji}) + U\sum_i n_{i\uparrow}n_{i\downarrow} + V\sum_{\langle ij\rangle} n_i n_j + W\sum_{\langle ij\rangle}(h_{ij}+h_{ji})^2 + X\sum_{\langle ij\rangle}(h_{ij}+h_{ji})(n_i+n_j)\ , \tag{1}$$

where $\langle ij\rangle$ runs over nearest-neighbor pairs, $h_{ij} = \sum_\sigma c^\dagger_{i\sigma}c_{j\sigma}$, $n_i = \sum_\sigma c^\dagger_{i\sigma}c_{i\sigma}$ and the electronic orbitals are arranged either on the vertices of a truncated tetrahedron (C_{12}) or on those of a truncated icosahedron (C_{60}). The interaction terms are, implicitly, normal ordered. Hence, appearances notwithstanding, the W and X terms do *not* renormalize the hopping and chemical potential at zeroth order. For our qualitative purposes we ignore the difference in the hopping and nearest-neighbor terms between the "short" and "long" bonds, except that in the kinetic energy they are not taken to be exactly equal (but rather, with a ratio of 1.001), so as to split some accidental degeneracies and thus simplify the computer programs somewhat.[6]

The quantities of interest for us are the energies of the low-lying states and the resulting set of pair-binding energies,

$$E^{(i)}_{\rm pair} = 2E_i - E_{i-1} - E_{i+1}. \tag{2}$$

Here E_i is the ground state energy of the molecule with i electrons in excess of charge neutrality; positive values of the $E_{\rm pair}$ indicate that charge disproportionation is favored. (The reader is warned that, in a slight abuse of this notation, we will also use $E_i/E^{(i)}_{\rm pair}$ with further elaboration, such as "$E_2/E^{(1)}_{\rm pair}$ for the ^1A_g state of C_{60}^{-2}," to indicate that energy/energy difference even when the ^1A_g state is not the ground state.)

The model with $V = W = X = 0$ was studied by CGK using second-order perturbation theory in U/t; they found that the $E^{(i)}_{\rm pair}$ with odd i became positive at intermediate values of $U \geq 3.3$. The tendency of an added V to suppress pair-binding was noted by White *et al.*[5] and by Goff and Phillips[4] who also included longer ranged density-density interactions.

Our inclusion of the leading off-diagonal (non-density-density) terms is principally motivated by the observation that energy differences such as $E^{(i)}_{\rm pair}$ and level splittings are much more sensitive functions of the interaction parameters than the energies of the molecular states involved in their definition, and hence one may well need to keep track of interactions that nominally enter with small coefficients.[7] These terms arise when the underlying density-density interaction is reexpressed in the Wannier basis necessary to deriving the extended Hubbard model. The general matrix element of the interaction $\mathcal{V}(\mathbf{x})$ in the Wannier basis $\phi_i(\mathbf{x})$ is

$$\langle ij|\mathcal{V}|kl\rangle = \int d\mathbf{x}d\mathbf{y}\, \phi_i^*(\mathbf{x})\phi_j^*(\mathbf{y})\mathcal{V}(\mathbf{x}-\mathbf{y})\phi_k(\mathbf{y})\phi_l(\mathbf{x}) \quad (3)$$

which then specifies

$$\begin{aligned} U &= \langle ii|\mathcal{V}|ii\rangle\ , \\ V &= \langle ij|\mathcal{V}|ji\rangle\ , \\ W &= \langle ij|\mathcal{V}|ij\rangle\ , \\ X &= \langle ii|\mathcal{V}|ij\rangle\ , \end{aligned} \quad (4)$$

where i and j are nearest-neighbor sites. The magnitudes of these terms have been discussed in the literature, for example, by Campbell, Gammel, and Loh.[8] The qualitative conclusions are that while $U > V, W, X$, the relative magnitudes of the latter are sensitive to the detailed structure of the Wannier function and of the effective (screened) interaction. To illustrate the origin of this sensitivity we show in Fig. 1 a model calculation of the π-band Wannier function for C_{60}. Generally $V > X > W$, but they become comparable when the interaction is screened on length scales shorter than the localization length of the Wannier function; indeed X can even change sign.[8] We will rely upon these estimates when we return below to the question of assigning values relevant to C_{60}.

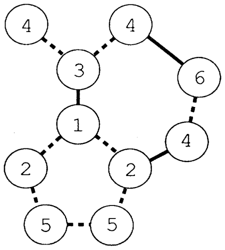

FIG. 1. A model π-band C Wannier function calculated by Löwdin orthogonalization keeping only a nearest-neighbor overlap of 0.35. The amplitudes at the labeled sites are (1) 1.324, (2) −0.359, (3) −0.384, (4) 0.169, (5) 0.092, (6) −0.123. Note the oscillations in the signs of the amplitudes.

III. RESULTS FOR C_{60}

Exact diagonalization of our Hamiltonian for C_{60} is currently out of the question.[9] Therefore, we have followed previous work in computing the energies of the low-lying states (those which constitute the degenerate ground state manifold in the absence of interactions) for the anions C_{60}^{-i} (i =0–6) perturbatively to second order in the various interactions. Our procedure was to calculate *total* energies for each ion. While this requires more computer time than a more sophisticated approach (such as employed in Ref. 1) that calculates energy differences with respect to C_{60} or C_{60}^{-6}, it has the advantage of being straightforward to code. Comparing our results for the U terms with previous work gives a nontrivial check of the programs' validity. We have not attempted to estimate directly the neglected higher order corrections; instead we compare, below, perturbation theory with exact diagonalization results for the smaller homolog, C_{12}.

Our results for C_{60}^{-i} constitute 13 tables of coefficient matrices, one each for $i = 0$, 1, 5, and 6 and three each for $i = 2$, 3, and 4. (Each entry in the tables is the coefficient of the term involving the product of its row and column labels in the expansion to second order.) The degeneracies of the various anions can be understood, literally, in a spherical approximation for C_{60}. The neutral molecule and C_{60}^{-6} consist entirely of filled shells and have a unique ($L = 0, S = 0$) ground state. The three degenerate lowest unoccupied molecular orbitals (LUMO's) of the neutral C_{60} molecule can be treated as an $L = 1$ triplet. Consequently, the low-lying states of C_{60}^{-1} and C_{60}^{-5} form an ($L = 1$, $S = 1/2$) multiplet, those of C_{60}^{-2} and C_{60}^{-4} consist of the multiplets ($L = 0$, $S = 0$), ($L = 1$, $S = 1$) and ($L = 2$, $S = 0$) and finally those of C_{60}^{-3} consist of the multiplets ($L = 0$, $S = 3/2$), ($L = 1$, $S = 1/2$) and ($L = 2$, $S = 1/2$).[1] We note that the $L = 0$, 1, 2 states correspond, respectively, to the icosahedral representations ^1A_g, $^3T_{1g}$, ^1H_g for $n = 2$ and 4, and to ^4A_u, $^2T_{1u}$, and ^2H_u for $n = 3$. Due to space considerations, the tables are not reproduced here but have been deposited with the Physics Auxiliary Publication Service.[10]

Exploring the four-dimensional phase diagram implicit in these tables is a formidable but unnecessary task, for the values of the parameters are constrained by the requirement that perturbation theory be valid and that they derive from a single underlying interaction. It is convenient to consider families of interactions (U, V, W, X) with variable U and fixed ratios V/U, W/U, X/U where the first constraint is incorporated (very roughly) by requiring that U is bounded by the π-orbital single-particle bandwidth ($\approx 5t$). Based on the totality of the parameter values reviewed in Ref. 8 we estimate that the second constraint is incorporated by considering $V/U \leq 0.6$,

$X/U \leq 0.2$, and $W/U \leq 0.04$. At the high end these values are consistent with estimates for benzene[11] where the bare interaction is screened only by the σ bands but in that limit one would need to keep track of longer range interactions.[12] The problem of interest, however, is that of pair binding in the metallic phase of doped C_{60}, and (*modulo* self-consistency) we take the interaction to be the effective, screened interaction appropriate to this system, which will tend to give somewhat smaller values of the parameters.

Before discussing our data, two remarks are in order. First, a caveat: most of the estimates that we have cited in assessing the relevant parameter ranges are from calculations using atomic orbitals and not using Wannier functions. Campbell, Gammel, and Loh[8] carried out a model calculation for a one-dimensional Kronig-Penney model where they calculated the Wannier function and found systematic deviations from the atomic orbital estimates as the screening length was varied. For our purposes the more interesting aspect of their data is that they suggest that W and X might be substantially smaller than we have supposed reasonable. Nevertheless, it is not at all obvious whether this feature of their work, which is certainly sensitive to details of the Wannier function and of the interaction, would carry over to a "first principles" calculation for C_{60}. Hence we have chosen, pending a careful estimate of the parameter values relevant to C_{60}, to use the atomic orbital values as the appropriate ones. (Also, see our concluding discussion.) Second, the reader should note that particular results that do not specify the values of V, W, and X will correspond to a "canonical" set $V/U = 0.5$, $W/U = 0.04$, and $X/U = 0.12$.

A. Hund's rules violations

We are interested here in Hund's first rule which implies that any degeneracies arising from a partially filled shell in excess of those dictated by symmetry be lifted in favor of the states with maximal spin. For C_{60}^{-n}, $n = 0$, 1, 5, and 6, the states are either nondegenerate (0, 6) or degenerate by symmetry (1, 5). For the remaining anions we find the following.

(1) C_{60}^{-2}: At sufficiently small values of U/t, the ground state is always the ${}^3T_{1g}$ state consistent with Hund's rule. For the pure Hubbard model $V = W = X = 0$, Hund's rule is violated beyond $U_c = 2.8t$ as the ground state crosses over to the ${}^1A_g$ state; the ${}^1H_g$ state is always an excited state. (Here and elsewhere the results for the pure Hubbard case are due to CGK and are listed for comparison.) The inclusion of a nonzero V does not shift the location of the ${}^1A_g/{}^3T_{1g}$ level crossing very much and for $V/U \leq 0.5$ it remains at $U_c \approx 2.8t$. The same is true of the off-diagonal terms; including both W and X causes a modest downward shift in U_c/t to about 2.7.

(2) C_{60}^{-3}: Here Hund's rule correctly predicts that the ${}^4A_u$ state is the ground state at small values of U/t. For the pure Hubbard case there is a crossing to the ${}^2T_{1u}$ state at $U/t \approx 2.9$. We find that this crossing is robust; even with the further inclusion of V, W, and X the crossing is always to the ${}^2T_{1u}$ state and the critical value of U/t is constant to within about 0.1.

(3) C_{60}^{-4}: This is, roughly, the particle-hole conjugate (within the t_{1u} manifold) of C_{60}^{-2} and the Hund's rule state at small U/t is again the ${}^3T_{1g}$ state. The pure Hubbard result of a ${}^1A_g/{}^3T_{1g}$ level crossing at about $U/t \approx 2.8$ is mildly decreased by the addition of further interactions, with values of $U_c = 2.5$ (for UV), $= 2.6$ (for UVW), $= 2.4$ (for UVX), and $= 2.6$ (for $UVWX$).

B. Pair-binding energies

The pair-binding energies for even dopings, i.e., for the disproportionations $C_{60}^{-2} \rightarrow C_{60}^{-1} + C_{60}^{-3}$ and $C_{60}^{-4} \rightarrow C_{60}^{-3} + C_{60}^{-5}$, are always negative and hence pair binding does not occur in these cases. For the odd dopings the situation is as follows.

(1) $E_{\text{pair}}^{(1)}$: In the pure Hubbard model pair binding takes place only into the ${}^1A_g$ state of C_{60}^{-2} and $E_{\text{pair}}^{(1)}$ becomes positive for $U > U_{\text{pair}} = 3.2t$. The value of U_{pair} is *extremely* sensitive to the inclusion of further interactions. The inclusion of the next-neighbor repulsion at the values $V/U = 0.2$, 0.5 changes U_{pair}/t to 4.2 and 10, respectively. Of course, finding within perturbation theory a value of $U_{\text{pair}} = 10t$ is meaningless, except to suggest that E_{pair} is *never* positive. This tendency of further neighbor density-density ("diagonal") interactions to suppress pair binding was the basis of Goff and Phillips's conclusion that going beyond the pure Hubbard model was, for practical purposes, fatal to the correlation mechanism for superconductivity. For $E_{\text{pair}}^{(1)}$, we find that the inclusion of the off-diagonal terms, more precisely W, can substantially affect U_{pair}. For example, at $V/U = 0.5$, the inclusion of W at the entirely plausible level of 4% of U brings U_{pair} down to $5.2t$, just outside our nominal "perturbative" range. The further effect of an added X is mildly suppressive of pair binding, e.g., $X = 0.12$ yields $U_{\text{pair}} = 5.4t$. These results are illustrated in Figs. 2, 3, and 4. Note that the ordering of

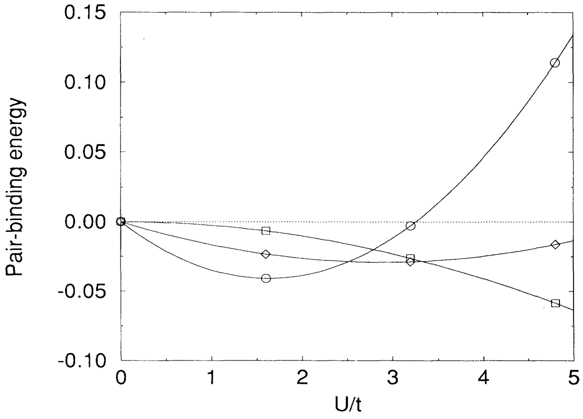

FIG. 2. $E_{\text{pair}}^{(1)}/t$ as a function of U/t for the ${}^1A_g$ (circles), ${}^3T_{1g}$ (squares), and ${}^1H_g$ (diamonds) states for $V/U = W/U = X/U = 0$.

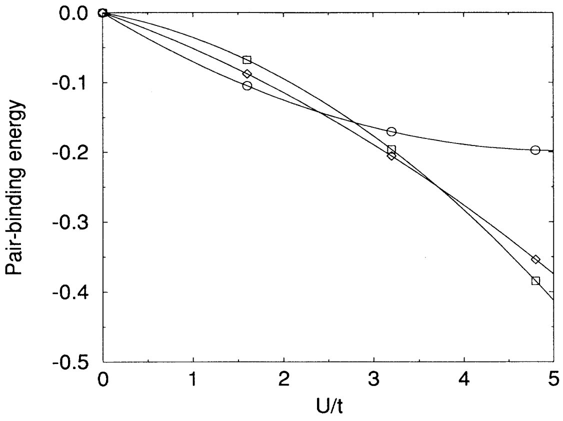

FIG. 3. $E_{\text{pair}}^{(1)}/t$ as a function of U/t for the ${}^1A_g$ (circles), ${}^3T_{1g}$ (squares), and ${}^1H_g$ (diamonds) states for $V/U = 0.5$ and $W/U = X/U = 0$.

the different $E_{\text{pair}}^{(1)}$ is also the ordering of the states of the C_{60}^{-2}.

(2) $E_{\text{pair}}^{(3)}$: Here there are potentially several choices for disproportionation, but in the pure Hubbard case and in the extensions studied here, both the doubly and quadruply charged anions are in the ${}^1A_g$ state *when* the pair-binding energy goes positive. The values of U_{pair}/t for this channel show modest variation. It is suppressed from the pure Hubbard value of 3.3 to the value 4.1 by the inclusion of $V = 0.5$. Note that V appears to be substantially less effective in suppressing pair-binding here than in the case of $E_{\text{pair}}^{(1)}$. Again the inclusion of W enhances the pairing and at $W/U = 0.04$ changes U_{pair}/t to about 3.6. The further inclusion of X has a marginal effect.

(3) $E_{\text{pair}}^{(5)}$: For the pure Hubbard case $U_{\text{pair}} = 3.3t$ for the ${}^1A_g$ state (of C_{60}^{-4}) and the value of U_{pair} for the ${}^1H_g$ state lies slightly outside the physical range. We find that while additional interactions greatly suppress pair binding into the latter, their effect on pair binding into the ${}^1A_g$ state is minor. In particular, we find the values $U_{\text{pair}}/t = 4$, 3.9, 3.8, 3.7 on including V, VX, VW, and VWX, respectively. This is in some contrast to the behavior of the (clearly approximately) particle-hole conjugate quantity $E_{\text{pair}}^{(1)}$.

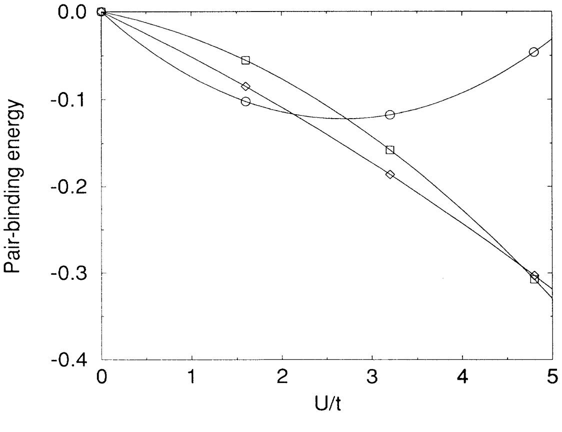

FIG. 4. $E_{\text{pair}}^{(1)}/t$ as a function of U/t for the ${}^1A_g$ (circles), ${}^3T_{1g}$ (squares), and ${}^1H_g$ (diamonds) states for $V/U = 0.5$, $W/U = 0.04$, and $X/U = 0.12$.

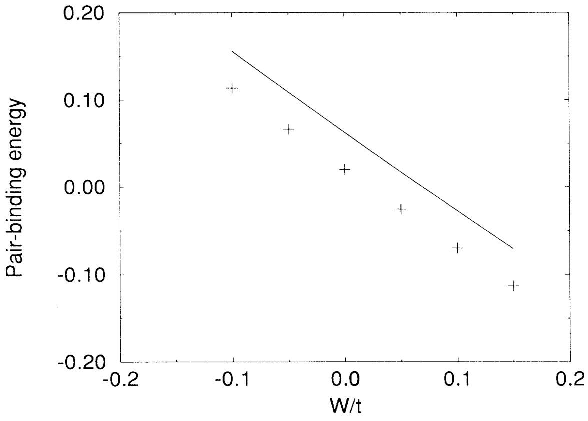

FIG. 5. Effect of W on pair binding for C_{12} at $U = 2$. The solid curve is the perturbative result for $E_{\text{pair}}^{(1)}/t$ for the ${}^1E$ state as a function of W for $U = 2$ and the individual points are exact diagonalization results.

IV. RESULTS FOR C_{12}

To assess the validity of second order perturbation theory, particularly with regard to the sign of the E_{pair}, we have followed White *et al.*[5] and compared perturbative and exact results for C_{12}. The latter has many features in common with C_{60},[5] most notably a degenerate triplet of LUMO's. (Lammert and Rokhsar[13] have argued that the reasonable success of second-order perturbation theory for C_{12}, in the sense of agreement with exact diagonalization at modest values of U, may not carry over to C_{60}. While their argument may well be correct, we believe it may not be relevant to the issue that is addressed below.) The coefficient matrices for $E_{\text{pair}}^{(1)}$ in C_{12} have been calculated, but again to save space they are not reproduced here.[10]

White *et al.* showed that for the pure Hubbard model for C_{12}, while perturbation theory reliably predicted U_{pair} (and even more reliably predicted the value of U for level crossings) the typical value of E_{pair} was considerably overestimated for $U > U_{\text{pair}}$. We find that the *additional* changes in the E_{pair} due to V, W, and X are fairly well described by the perturbative results even when perturbation theory overestimates $E_{\text{pair}}(U; V = W = X = 0)$ by a factor of 3. This is illustrated in Fig. 5 for the case of an added W term. It should be emphasized that this result is not entirely expected: the additional change has three terms, one coming from first-order perturbation theory, $-2W/3$, and two from second order in perturbation theory, $-UW/8$ and $0.2338W^2$. The last term is entirely negligible for the range of parameters in the plot. For $U = 2t$, as in Fig. 5, the second term is 3/8 of the first, but since U is already large enough that the $O(U^2)$

term alone substantially overestimates E_{pair} [and so there are important contributions from $O(U^3)$ and higher], it is not obvious that $O(U^2W)$ and higher terms can be neglected in a calculation of the changes in E_{pair} due to the addition of a W term.

V. CONCLUSIONS

Our results show that the critical values of U/t for the Hund's rules violating level crossings in the doubly, triply, and quadruply charged anions are quite insensitive to the inclusion of all the nearest-neighbor interactions. This suggests that the effects of electron correlations might be significant even for isolated anions for which pair binding is certainly ruled out, since the energy differences between different charge states are dominated by the "molecular capacitance" energy. In this connection the work of Negri *et al.*[14] merits a close examination. It is interesting they find that the ground state of C_{60}^{-2} is a ^1A_g state rather than a $^3T_{1g}$ state. However, they also find that the next-lowest state is of ^1A_u symmetry, which suggests that any useful perturbative treatment may need to employ nearly degenerate perturbation theory and allow for occupancy of the t_{1g} molecular orbitals (that lie roughly 1 eV above the t_{1u} LUMO's) in the unperturbed states.

On the question of pair binding we find that the W term reduces (using the crude measure of U_{pair}) by about 50% the suppression of pair binding produced by the V term. We note that in contrast to U and V, the W term favors pair binding already at *first* order and also favors pair binding in second order, in each case with large coefficients. However, these large *a priori* effects are offset by the small numerical value of W expected on physical grounds in C_{60}. The net effect of the X term is weakly suppressive for the values of interest.[15]

Goff and Phillips had argued that going beyond the Hubbard model necessarily suppressed pair binding and that for parameter values relevant to the fullerides E_{pair} was always negative. At a minimum we have shown that there are interactions beyond the Hubbard approximation that do favor pair binding and, to use the framework of Ref. 4, open a narrow window of parameters for which this could actually take place in the doped fullerides. However, we feel that such purely microscopic considerations ought not be taken too seriously for two reasons. First, we believe that our calculations illustrate that the issue of pair binding is quite delicate and to draw phase diagrams with any confidence one would need to be certain that all relevant interactions had been kept, their effects calculated accurately, and the parameter values known to high precision. These are daunting challenges for current theory. Second, the problem of physical interest involves *intra*molecular interactions that are necessarily renormalized by *inter*molecular dynamics (e.g., screening) in a self-consistent fashion.[16] Consequently, absent a solution of the full problem, it is difficult to assign properly the relevant parameter values; e.g., a modest enhancement of W could greatly enhance pair binding. We are *not* arguing that microscopics can never settle these sorts of issues, merely that in this particular problem the existence of a region of pair binding in parameter space has been clearly demonstrated in model calculations[17] and that the additional problem of locating precisely the parameters of the physical system does not appear amenable to first principles solution. Consequently, it would appear that consistency of the scenario with the totality of experiments is perhaps a better approach.

ACKNOWLEDGMENTS

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[1] S. Chakravarty and S. Kivelson, Europhys. Lett. **16**, 751 (1991); S. Chakravarty, M. P. Gelfand, and S. Kivelson, Science **254**, 970 (1991).

[2] Actually their work (and ours) is intended to apply to a molecule assumed to be embedded, self-consistently, in a metallic background.

[3] Recent reviews of experiments and theory are, respectively, A. P. Ramirez, Supercond. Rev. **1**, 1 (1994); M. P. Gelfand, *ibid.* **1**, 103 (1994).

[4] W. E. Goff and P. Phillips, Phys. Rev. B **46**, 603 (1992); **48**, 3491 (1993).

[5] S. R. White, S. Chakravarty, M. P. Gelfand, and S. A. Kivelson, Phys. Rev. B **45**, 5062 (1992).

[6] We note that the data reported in Ref. 1 are for a kinetic energy ratio of 1.2 and thus are slightly different from those in this paper for the pure Hubbard model.

[7] In recent years these terms have been of interest in at least two fields. In the study of one-dimensional systems such as polyacetylene their impact upon the dimerization has been debated, e.g., in Ref. 8 and A. J. Heeger, S. Kivelson, J. R. Schrieffer, and W. P. Su, Rev. Mod. Phys. **60**, 781 (1988). Their role in high-temperature superconductivity is the subject of J. E. Hirsch, Phys. Lett. A **134**, 451 (1989); J. E. Hirsch and F. Marsiglio, Phys. Rev. B **39**, 11515 (1989); A. Zawadowski, *ibid.* **39**, 4682 (1989). Also, exactly soluble extended Hubbard models with restricted values of these interactions have been presented in F. Essler, V. Korepin, and K. Schoutens, Phys. Rev. Lett. **70**, 73 (1993);

R. Strack and D. Vollhardt, *ibid.* **70**, 2637 (1993).

[8] D. K. Campbell, J. T. Gammel, and E. Y. Loh, Phys. Rev. B **42**, 475 (1990).

[9] However, there have been very recent developments using a numerical renormalization group method which makes possible controlled calculations of pair-binding energies in Hubbard and extended Hubbard models for C_{60} up to moderate interaction strengths. See N. Berdenis and G. Murthy (unpublished), and, for previous work along these lines, T. Tokuyasu *et al.*, Phys. Rev. Lett. **71**, 4202 (1993).

[10] See AIP document no. PAPS PRBMD-51-5943-6 for 6 pages of material containing 17 tables of coefficient matrices, 13 corresponding to the total energy of C_{60} anions and 4 corresponding to the pair-binding energies $E^{(1)}_{\rm pair}$ for C_{12}. Order by PAPS number and journal reference from American Institute of Physics, Physics Auxiliary Publication Service, Carolyn Gehlbach, 500 Sunnyside Boulevard, Woodbury, New York 11797. Fax: 516-576-2223, e-mail: janis@aip.org. The price is $1.50 for each microfiche (98 pages) or $5.00 for photocopies of up to 30 pages, and $0.15 for each additional page over 30 pages. Airmail additional. Make checks payable to the American Institute of Physics. The tables are also available directly from the authors.

[11] R. G. Parr, D. P. Craig, and I. G. Ross, J. Chem. Phys. **18**, 1561 (1950).

[12] In that case we expect the longer ranged diagonal interactions to dominate (Ref. 11) and hence to find an attenuated version of the pair-binding suppression found by Goff and Phillips.

[13] P. E. Lammert and D. S. Rokhsar, Phys. Rev. B **48**, 4103 (1993).

[14] F. Negri, G. Orlandi, and F. Zerbetto, J. Am. Chem. Soc. **114**, 2909 (1992).

[15] For illustration we note that, $E^{(1)}_{\rm pair}\,({}^1A_g) = -0.05\,U + 0.015\,364\,U^2 - 0.055\,560\,2\,V + 0.0045\,U\,V - 0.039\,903\,V^2 - 0.411\,12\,W + 0.267\,823\,U\,W - 0.079\,106\,V\,W + 0.933\,268\,W^2 + 0.027\,7128\,X - 0.014\,621\,U\,X - 0.018\,581\,V\,X - 0.142\,065\,W\,X + 0.153\,486\,X^2$.

[16] In this context we note that there is an additional complication, the dynamics of the screening, which has been discussed recently by P. E. Lammert, D. S. Rokhsar, S. Chakravarty, S. Kivelson, and M. I. Salkola (unpublished). They conclude that this works in favor of pair binding.

[17] All of this does assume that, as with smaller molecules, second-order perturbation theory for C_{60} is not qualitatively unreliable.