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Pairing in C_{60}: A Gutzwiller approach

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Going beyond both a simple perturbation theory and Hartree-Fock treatment, we employ the Gutzwiller variational scheme to clarify the possibility of electron pairing in C_{60}. It is shown that the pairing can be induced by electron-phonon interaction (EPI) rather than the pure electron-electron interaction (EEI), although a perturbation scheme showed that the pairing could be introduced by pure *e-e* repulsion. EPI can introduce a bipolaron if the on-site EEI U is not too strong. Our results are supported by electron-paramagnetic-resonance (EPR) experiments both in solid and in solution, which evidenced that the disproportional reactions such as ${C_{60}}^- + {C_{60}}^- \to C_{60} + {C_{60}}^{2-}$ do not occur. From the EPR results we anticipate that the on-site U is larger than 2.0 eV. It can be concluded that the perturbation prediction for small clusters may not hold for some larger clusters.

I. INTRODUCTION

The discovery of superconductivity in potassium doped C_{60} (Refs. 1 and 2) has spurred many research works experimentally as well as theoretically.[3–7] The transition temperature T_c, which is 18 K first observed in K_3C_{60} (Ref. 1) and later 28 K in Rb_3C_{60},[2] is much higher than that of the traditional metal superconductors. However, a universal mechanism for it is still controversial. Earlier experiments involving the isotope effect have revealed phonon-mediated pairing, $T_c \propto M^\alpha$, where M is the ionic mass and $\alpha = 0.30\pm0.06$,[8] $\alpha = 0.37\pm0.05$,[9] [$\alpha = 1.4\pm0.5$ (Ref. 10) may be not accurate] although in the simple BCS model α is 0.5. Recently in single-crystal K_3C_{60} and Rb_3C_{60}, Degiorgi *et al.*[11] obtained $2\Delta/k_bT_c$ of 3.44 and 3.45, respectively, consistent with predictions for a mechanism based on standard BCS mechanism with the electron-phonon coupling to intramolecular modes. All of these constitute strong evidence that superconductivity in alkali doped C_{60} is primarily driven by the phonon-mediated process; however, very strong Coulomb interaction puts many doubts on the applicability of the McMillan equation as it is generally used in the traditional superconductor. Hence, a well-defined phonon-mediated mechanism should cast light on how the strong electron-electron interaction can affect BCS results and how effective phonon-mediated-electron-electron attraction can efficiently overcome the strong on-ball electron repulsion and therefore create an effective electronic pairing attraction in the Fermi surface. Different from the traditional mechanism, a pure electronic mechanism has been put forward by Chakravarty, Gelfand, and Kivelson (CGK).[3] They introduced the pair-binding energy as

$$E^N_{\rm pair} = (E_{N+1} + E_{N-1}) - 2E_N, \tag{1}$$

where E_N is the ground-state energy of an isolated C_{60} molecule with N added electrons. $E^N_{\rm pair}$ measures the tendency toward pair formation at a doping of N electrons per fullerene. Our sign convention is opposite that of CGK but the same as Goff and Phillips (see below).[12] Using second-order perturbation theory, CGK discovered that for intermediate on-site electron repulsion U, pure repulsive interactions on atomic length scales can remarkably generate effective attraction between electrons on a single cluster. Their primary work motivated many research works. Goff and Phillips have developed CGK's earlier work perturbatively and confirmed that the pure attraction does exist. Lammert and Rokhsar[13] found out that the uncrossed ladder diagrams account for the stabilization of the singlet pairing; however, the exact summation of these uncrossed diagrams predict, surprisingly, no sign of pairing, which severely contradicts the perturbation results. In fact, Friedberg *et al.*[14] commented on CGK's work and doubted such a second-order perturbation formula can be meaningfully applied to large U in C_{60}. Simultaneously electron-paramagnetic-resonance (EPR) tests[15,16] of the resonating-valence-bond theory (i.e., CGK work) of superconductivity in $C_{60}K_n$ has shown that disproportional reactions, such as

$${C_{60}}^- + {C_{60}}^- \to C_{60} + {C_{60}}^{2-}, \tag{2}$$

do not occur in solution. While in solid, less than 3% of the radical ions disproportionate according to (2), which again contradicts the CGK perturbation results. Theoretically the conventional perturbation theory is ill suited for strong interaction systems. To reconcile this obvious contradiction, one should go beyond both second-order

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perturbation theory and a simple ladder summation.

In this paper, we employ the Gutzwiller scheme to access this controversial system. Earlier application of the Gutzwiller scheme to C_{60} has been performed by many authors.[17–20] Joyes *et al.*[17] examined the stability of spin-density wave and charge-density wave, but were ignorant of the electronic-phonon interaction. Scherrer *et al.*[18] investigated the effects of the electron interaction on the electron-lattice coupling in C_{60}. Metal-insulator transitions and magnetism in C_{60} have been studied by Lu[19] and Sheng *et al.*,[20] respectively. Traditionally, the Gutzwiller scheme has successfully reproduced the correct bond alteration and the dimerization amplitude in polymer, especially in polyacetylene.[21] Our calculation clarifies several points. (1) The on-site repulsion U does not induce the effective attraction between electrons. (2) Electron-phonon interaction (EPI) will introduce an effective attraction and result in a bipolaron on C_{60}; however, this attraction will be decreased with the enhancement of the on-site repulsion U.

After this work was accomplished, we found that a recent paper[22] addressed a similar problem. They employed the variational Monte Carlo method to access this problem. Although their result is similar to ours, as we point out in Sec. III, the statistical error is also strongly rooted in their calculation. Since the pairing energy is a small difference between two large values, the result is very sensitive to numerical errors (see Refs. 20 and 22) and even the sign of the pairing energy can be changed. Therefore, none of their results can distinctively give out where the pairing energy becomes positive or negative, but we can give out a transition point clearly. Second, none of them noticed the above important experimental tests. They cannot explain why in solid materials about 3% of the radical anions are disproportionate according to (2). We find that the reason for the above disproportional reaction is that EPI induces the effective attraction. None of the authors considered electron-phonon interaction. Actually, EPI plays a critical role in the pairing energy. With EPI the lattice will be relaxed to a stable state and the configuration of C_{60} will be distorted with the symmetry reduction from I_h to D_{5d}. This distortion induces an effective electronic attraction. One may notice that the effective electronic attraction induced by EPI is the same amount as the repulsion induced by the on-site U. These two important factors compete with each other and determine whether the effective attraction exists or not. Third, our Hamiltonian can reproduce two kinds of experimental bond length and the optical gap very well; References 20 and 22 have not been concerned with them. Lastly, one may notice that the strong-coupling limit calculation in Ref. 22 actually has been finished by Ref. 20 already.

This paper is arranged as follows. In Sec. II, the Guztwiller variational procedure is outlined. The main results and a comparison to other schemes appear in Sec. III. The conclusion and discussion are given in Sec. IV.

II. FORMALISM

C_{60}, with its uniqueness, is a quasi-two-dimensional system with π electrons moving mainly on the spherical surface. The on-ball electron hopping t_{ij} typically at 1.8 eV, is many orders of magnitude larger than the interball hopping, typically at 0.01 eV;[7] therefore, the system is well featured by the on-ball hopping. Thus, one can write the Hamiltonian as

$$\hat{H} = -\sum_{i,j,\sigma} t_{ij}(\hat{C}^{\dagger}_{i,\sigma}\hat{C}_{j,\sigma} + \text{H.c.}) + U\sum_{i} n_{i\uparrow}n_{i\downarrow} + \frac{K}{2}\sum_{i,j}(|\vec{r_i} - \vec{r_j}| - d_0)^2, \quad (3)$$

where $\hat{C}^{\dagger}_{i\sigma}$ creates a π electron at site i with spin projection σ and $n_{i\sigma} = \hat{C}^{\dagger}_{i\sigma}\hat{C}_{i\sigma}$, t_{ij} is the hopping integral between the nearest neighbor (NN) atoms sitting at $\vec{r_i}$ and $\vec{r_j}$. $t_{ij} = t_0 - \alpha(|\vec{r_i} - \vec{r_j}| - d_0)$, while t_0 is the average hopping constant. U is the on-site electronic repulsion. The third term in (3) is the lattice elastic energy. In our previous work,[23] we determined the above parameters $t_0 = 1.8$ eV, $\alpha = 3.5$ eV/Å, and $K = 15.0$ eV/Å^2, through fitting the optical energy gap and two kinds of bond length.

The treatment of the Gutzwiller variational scheme is rather conventional. First, we consider the ground-state wave function using the variational ansatz[21]

$$|\psi\rangle = e^S|\psi_0\rangle, \quad (4)$$

where $S = -\frac{1}{2}\eta\sum_i n_{i\uparrow}n_{i\downarrow}$ and $|\psi_0\rangle$ is the uncorrelated Fermi sea in Hartree-Fock space, while η is the variational parameter, which is determined by minimizing the electronic total energy

$$E_e(\text{total}) = \frac{\langle\psi|H_e|\psi\rangle}{\langle\psi|\psi\rangle}, \quad (5)$$

where H_e is the electron part in the Hamiltonian (3) (i.e., first two parts).

Using the linked-cluster technique and expanding e^S to the second order in η, we can explicitly rewrite the electronic energy as

$$\begin{aligned}\frac{\langle\psi|H_e|\psi\rangle}{\langle\psi|\psi\rangle} &= \langle\psi|H_e|\psi\rangle_c \\ &= \langle\psi_0|H_e|\psi_0\rangle_c - \frac{1}{2}\eta\langle\psi_0|\{\omega, H_e\}|\psi_0\rangle_c \\ &\quad + \frac{1}{8}\eta^2\langle\psi_0|\{\omega,\{\omega, H_e\}\}|\psi_0\rangle_c, \end{aligned} \quad (6)$$

where $\{A, B\} = AB + BA$ and $\omega = \sum_i n_{i\uparrow}n_{i\downarrow}$. Here the subscript of $\langle\cdots\rangle_c$ means that only the connected diagrams have to be taken into account. For convenience, we introduce the correlation functions as below,

$$UP_{ij} = \langle\psi_0|C^+_{i\uparrow}C_{j\uparrow}|\psi_0\rangle, \quad UQ_{ij} = \langle\psi_0|C_{i\uparrow}C^+_{j\uparrow}|\psi_0\rangle, \quad (7)$$

$$DP_{ij} = \langle\psi_0|C^+_{i\downarrow}C_{j\downarrow}|\psi_0\rangle, \quad DQ_{ij} = \langle\psi_0|C_{i\downarrow}C^+_{j\downarrow}|\psi_0\rangle. \quad (8)$$

Hence the total electronic energy is

$$E_e(\text{Total}) = T_1 + V_1 - \eta(T_2 + V_2) + \eta^2(T_3 + V_3), \quad (9)$$

where explicit expressions for $T_1, V_1, T_2, V_2, T_3, V_3$ are shown in the Appendix. Therefore, the total energy of the system can be expressed as

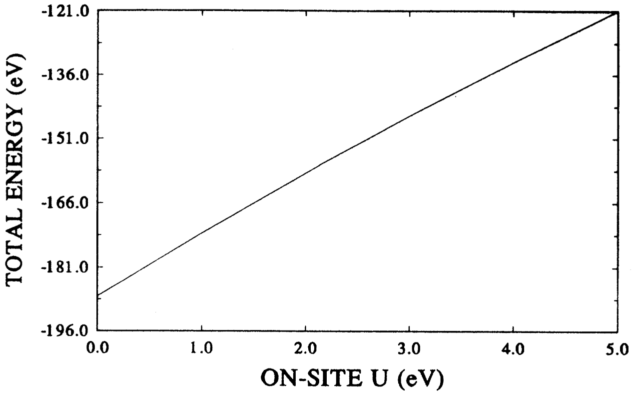

FIG. 1. Total energy of neutral C_{60} changes as U.

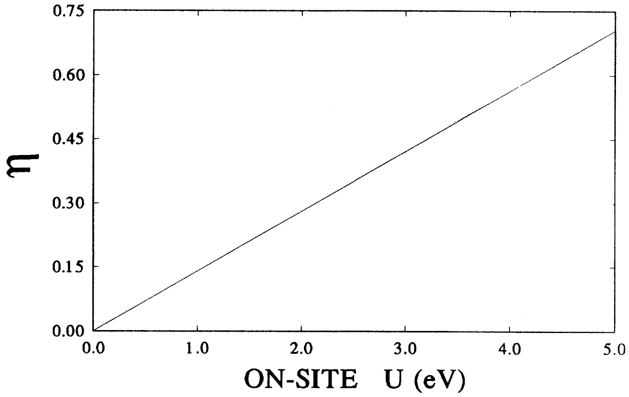

FIG. 3. Variational parameter η via U.

$$E(\text{Total}) = E_e(\text{Total}) + E_L(\text{Total}), \qquad (10)$$

where E_L(Total) refers to the elastic energy.

By minimizing the total energy of the system ground state with respect to η and the bond length, the real total energy and the bond length can be uniquely decided.

III. RESULTS

A. Undoped case

In the undoped case, C_{60} has 60 electrons and its symmetry is I_h. In Fig. 1 we plot the total energy E(Total) as a function of U. Clearly one may discover that E increases sharply as U enhances, the reminiscence of Joyes *et al.*,[17] where they didn't include EPI. We find the on-site U will prolong the bond length but the radius of the ball changes very little. The difference between the long bond and the short bond increases with increasing U (see Fig. 2). The physical reason is that the enhancement of on-site repulsion U makes π electrons far apart, hence through electron-phonon coupling the lattice or the cage will be expanded but later balanced by the sharp increase of the elastic energy.

To check out our variational scheme, we picture η as function of U (see Fig. 3); clearly if $U < 5$ eV, η is rather small, which indicates the expansion is valid.

B. Electron doped case

In the electron doped C_{60}, C_{60} contains more than 60 π electrons. Earlier electron-spin-resonance experiments[24,25] and theories[23,26,27] have shown that in one or two electrons doped C_{60} the bond structure is Jahn-Teller distorted, and the symmetry is reduced from I_h to D_{5d}. Our aim is to determine which state is more favorable: two additional electrons go to one single C_{60} leaving the other C_{60} undoped, or two C_{60}'s get one electron each. Then, the energy difference of these two states $E^1_{\text{pair}} = (E_2 + E_0) - 2E_1$ is the measure of the pairing and called as pairing energy. Obviously, if E^1_{pair} becomes negative, the electrons intend pairing. The numerical results of E^1_{pair} versus U are shown in Fig. 4. It can be seen that the pairing does exist when $U < U_c$=2.0 eV. Especially, the EPI alone (i.e., U=0 eV) will induce the pairing. Actually such pairing forms a bipolaron, which is stable for $C_{60}{}^{-2}$.[14] Also from Fig. 4 one may notice that the pairing energy is reduced with the enhancement of on-site U.

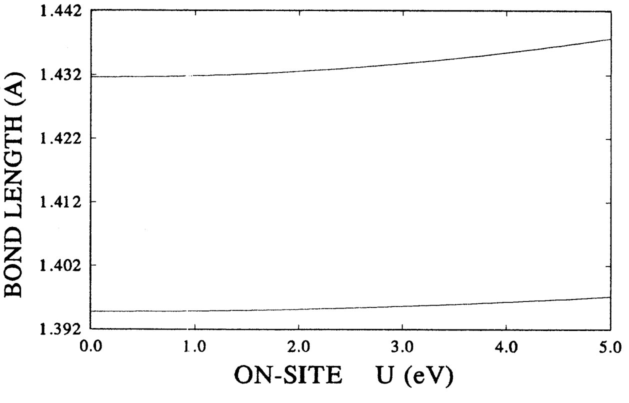

FIG. 2. Effect of on-site interaction U on the lengths of short and long bonds in neutral C_{60}.

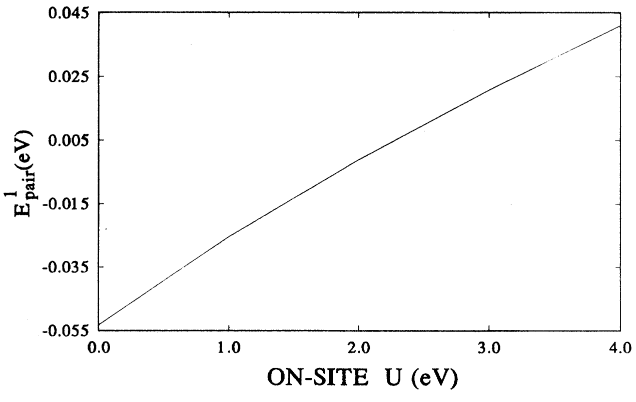

FIG. 4. Pairing energy changes with on-site U.

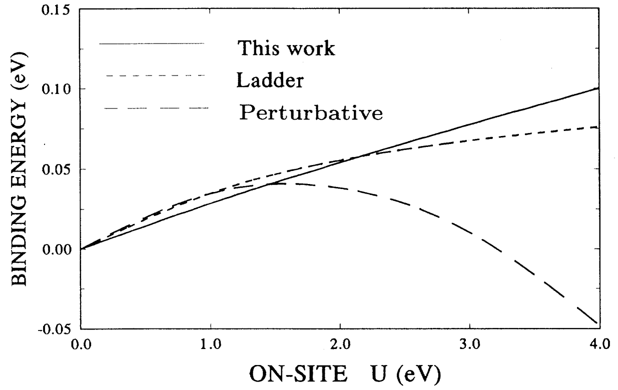

FIG. 5. Typical pairing energies from different theoretical approaches. The solid line, the short dash line, and the long dash line are for this work, ladder summation, and perturbation approach, respectively.

Since in C_{60} the on-site U is much larger than U_c, the on-ball pairing is unlikely. This is why in solution, the disproportional reaction $C_{60}{}^{-} + C_{60}{}^{-} \rightarrow C_{60} + C_{60}{}^{2-}$ is not favored since in solution C_{60} is much like a free molecule. In a bulk material, the on-site U is smaller than that of a single cluster, as reported U =1.6 eV;[28] thus, the possibility of the disproportional reaction for the bulk material is slightly larger than that of a single C_{60}, but the corresponding EPI is slightly smaller, which makes the reaction less plausible. That is why in solid materials less than 3% of the radical anions disproportionate according to (2). The net result for the bulk material indicates that the current values of U are just at the limit where pairing becomes stable. After all, we should emphasize here again that the pairing is induced by EPI rather than electron-electron interaction U.

To make our results more convincing, we compare our results with those of other schemes, such as perturbation theory, whole ladder summation, and a variational Monte Carlo method. The typical pairing energies are shown in Fig. 5. Remember our results in Fig. 5 are from the fixed C_{60}'s configuration and for comparison convenience, we set t of other works to 1 eV. Although different parameters in these different methods make a direct and quantitative comparison unlikely, our results, in small U, can reproduce the correct trend of the perturbation results very nicely, where the perturbation theory works well. A small deviation from the perturbation results is not surprising since in our case we include EPI and distinguish two different kinds of bond length, while in Ref. 13 e-ph coupling is excluded and only one bond length is considered. If two kinds of bonds are considered, the line of the perturbation results will be pulled down a little, which is exactly the CGK original case (see the curve for E^1_{pair}, in Fig. 3 of their paper), and will be very close to our results (but their parameters are still different from ours). At large U, we compare our results with those of other variational schemes[20] because the perturbation results are not reliable. In Ref. 20, Sheng *et al.* employed a variational Monte Carlo technique in the t-J-like Hamiltonian and illustrated their main results in Fig. 8 of their paper. One should understand that at large U, our Hamiltonian is very similar to the t-J model. Thus, a comparison to their outcome is meaningful, and the t-J model can predict the correct trend at large U. Although they used J as the unit of energy along with the same reasons previously mentioned, which makes a quantitative comparison very hard, a qualitative comparison is nontrivial. The real tendency of our results is similar to theirs, i.e., in large U pairing does not appear (see the curve for $n = 1$ in Fig. 8 of their paper). However, we would like to remind readers that we have opposite sign convention of pairing energies from theirs, namely in their paper $E^N_{\text{pair}} = 2E_N - (E_{N+1} + E_{N-1})$, and t/J is just $|U|/2t$.[29] In comparison to their Monte Carlo method, our calculation shows several advantages. Our scheme can be analytically carried out while a possible error is strongly rooted in their numerical results. Last we would like to point out their understanding of pairing is problematic.

In comparison with our results, the whole ladder summation reproduces the correct tendency, which may open a new way to access the strong correlation system. But for the whole ladder summation, the correct selection of specific diagrams is critical and is also very difficult: The Gutzwiller variational method has no such difficulties and is easily performed. In the Guztwiller scheme, one should constantly be aware of the variational parameter η. Only if η is smaller than 1, is the expansion scheme meaningful. The only difference between our results and the perturbation upshot occurs at $U > 2.3$ eV, where the perturbation theory is ill suited since many high-order terms become significant; while in the small U case, they can be properly ignored. In Sec. IV, prove this again, by quoting direct evidence from recently published papers.

IV. DISCUSSION AND CONCLUSION

In this article we employ the Gutzwiller method to clear out the possible on-ball pairing in electron doped C_{60}. We find the on-ball pairing is possible only if on-site $U \leq U_c$. On-site U never introduces the so-called pure electronic attraction, and contrarily U breaks the possible pairing. The electron-phonon interaction is responsible for the pairing. At $U = 0.0$ eV, EPI introduces a negative E^1_{pair} as large as -0.050 eV and may result in a bipolaron. Our results are different from the CGK perturbation ones. Although in a later paper[30] they proved that the perturbation theory could predict the right tendency in small clusters and tried to draw some inferences, that are sufficiently general to hold for C_{60}. However, in that paper, they did not confirm their results beyond the perturbation treatment in large clusters. Recently in a truncated basis, by double configuration interactions technique, Gammel and Mazumdar[31] found that potential pitfalls exist in the extrapolation from small system calculations to C_{60} and stressed the necessity of having the correct intuitive picture. Additionally, as an example, in a four-site chain, one will find that the bond alteration via U is very different from the typical alteration in infinite chain.[32] Of course an exact and more direct judge comes from the experiments.

Since experiments confirm that $C_{60}{}^{-} + C_{60}{}^{-} \rightarrow C_{60}{}^{2-} + C_{60}$ does not occur in solution, we can estimate that the on-site U is larger than $U_c = 2.0$ eV, which is consistent with many experimental and theoretical estimations.[7,33–35] Therefore, on-ball pairing, in fact, does not exist on the C_{60} cage, which brings us back to the original question: what causes pairing. This pushes one to pursue other mechanisms. Naturally the phonon-mediated pairing is the first candidate, but it is believed that a universal theory should include the strong on-site electron-electron repulsion. Recently one interesting exchange-mediated pairing was proposed.[36] However, a complete and convincing theory has not been formed yet.

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APPENDIX A: THE EXPLICIT EXPRESSIONS FOR T_1, T_2, T_3, V_1, V_2, V_3

In this appendix, we demonstrate several details about the distinctive expressions in (9).

$$
\begin{aligned}
T_1 &= \sum_{i,j} t_{ij}(UP_{ij} + DP_{ij}),\\
T_2 &= \frac{1}{2}\sum_{i,j;m} t_{ij}[DP_{mm}(UP_{mm}UQ_{mi} + UP_{im}UQ_{jm})\\
&\quad +UP_{mm}(DP_{mj}DQ_{mi} + DP_{im}DQ_{jm})],\\
T_3 &= \frac{1}{8}\sum_{i,j;m,n} t_{ij}2(DA)(UB) + 4(DC) + (UA)(DB)\\
&\quad +4(UC) + (DA)(UD) + (UA)(DD),
\end{aligned}
\tag{A1}
$$

where

$$
\begin{aligned}
DA &= DP_{mm}DP_{nn} + DP_{mn}DQ_{mn},\\
UB &= UP_{mj}UQ_{mn}UQ_{ni} - UP_{mn}UQ_{mi}UP_{nj},\\
UA &= UP_{mm}UP_{nn} + UP_{mn}UQ_{mn},\\
DB &= DP_{mj}DQ_{mn}DQ_{ni} - DP_{mn}DQ_{mi}DP_{nj},\\
DC &= DP_{mn}DQ_{mn}UP_{mm}UP_{nj}UQ_{ni},\\
DD &= DP_{nm}DQ_{ni}DQ_{jm} - DP_{nj}DQ_{nm}DP_{im},\\
UC &= UP_{mn}UQ_{mn}DP_{mm}DP_{nj}DQ_{ni},\\
UD &= UP_{nm}UQ_{ni}UQ_{jm} - UP_{nj}UQ_{nm}UP_{im},
\end{aligned}
\tag{A2}
$$

and the summations over i,j are taken over NN only while the summations for n,m run over 1 to 60.

$$
\begin{aligned}
V_1 &= U\sum_{i} UP_{ii}DP_{ii},\\
V_2 &= U\sum_{ij}(UP_{ii}UP_{jj} + UP_{ij}UQ_{ij})DP_{ij}DQ_{ij}\\
&\quad +UP_{ij}UQ_{ij}DP_{ii}DP_{jj},\\
V_3 &= U\sum_{ijk} DP_0UP_1 + DP_4(UP_1 + UP_3 + UP_2)\\
&\quad +DP_3(UP_1 + UP_2 + UP_4) + DP_2(UP_1\\
&\quad +UP_3 + UP_4)\\
&\quad +DP_1(UP_0 + UP_1 + UP_2 + UP_3 + UP_4),
\end{aligned}
\tag{A3}
$$

where

$$
\begin{aligned}
DP_0 &= DP_{ii}DP_{jj}DP_{kk}, & UP_0 &= UP_{ii}UP_{jj}UP_{kk},\\
DP_2 &= DP_{jk}DQ_{jk}DP_{ii}, & UP_2 &= UP_{jk}UQ_{jk}UP_{ii},\\
DP_3 &= DP_{ik}DQ_{ik}DP_{jj}, & UP_3 &= UP_{ik}UQ_{ik}UP_{jj},\\
DP_4 &= DP_{ij}DQ_{ij}DP_{kk}, & UP_4 &= UP_{ij}UQ_{ij}UP_{kk},
\end{aligned}
$$

$$
\begin{aligned}
DP_1 &= DP_{ik}DQ_{ij}DQ_{jk} - DP_{ij}DQ_{ik}DP_{jk},\\
UP_1 &= UP_{ik}UQ_{ij}UQ_{jk} - UP_{ij}UQ_{ik}UP_{jk},
\end{aligned}
$$

and the summations over i,j,k run over 1–60.

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