RENORMALIZATION GROUP APPROACH TO THE
COULOMB PSEUDOPOTENTIAL FOR C\textsubscript{60}

Nikos Berdenis and Ganpathy Murthy

Department of Physics, Boston University

Abstract

A numerical renormalization group technique recently developed by one of us is used to analyse the Coulomb pseudopotential ($\mu^*$) in C\textsubscript{60} for a variety of bare potentials. We find a large reduction in $\mu^*$ due to intraball screening alone, leading to an interesting non-monotonic dependence of $\mu^*$ on the bare interaction strength. We find that $\mu^*$ is positive for physically reasonable bare parameters, but small enough to make the electron-phonon coupling a viable mechanism for superconductivity in alkali-doped fullerides. We end with some open problems.
The discovery of the icosahedrally symmetric molecule $C_{60}$ in 1985 [1], and the subsequent discovery of the many interesting electronic properties of doped fullerene lattices has led to an explosion of research work on these compounds. In this Letter we will be concerned with the effects of Coulomb interactions on fullerene superconductivity.

$C_{60}$ forms an FCC lattice which can be doped with up to 6 alkali metal atoms per $C_{60}$. The triply-doped compounds $A_3C_{60}$ (where $A = K, Rb, Cs$) are bad metals but superconduct at relatively high temperatures ($\approx 20K$ for $K_3C_{60}$) [2–4]. They also have a very short coherence length of $\xi \approx 30\,\text{Å}$, which is only about 2 lattice spacings of the FCC lattice. Experimental evidence [5–10], supported by numerous phonon calculations [11–18], indicates that some of the intraball phonon modes are very high in energy and sufficiently strongly coupled to the electrons to account for these high $T_c$ s. Most of the phonon calculations assume that the Coulomb repulsion between the electrons comprising a pair are small, as they are in a usual metallic superconductor. (Effective Coulomb interactions are typically parameterized by a dimensionless number $\mu^* = N(E_F)U^*$ [19], where $N(E_F)$ is the density of states at the Fermi Surface (FS) and $U^*$ is an effective interaction matrix element between electrons at the FS. For a metal $\mu^* \approx 0.2$). However, there are two significant differences in the case of superconducting fullerides which make this correspondence uncertain. Firstly, the coherence length, which is a measure of how close the electrons in a pair are, is an order of magnitude shorter in $A_3C_{60}$ than in metallic superconductors. Furthermore, in metallic superconductors the effective Coulomb interaction between the electrons comprising a pair is reduced by the highly retarded nature of the phonon-mediated attraction, as shown by Anderson and Morel in 1962 [19]

$$\mu^* = \frac{\mu}{1 + \mu \log(W/\omega_d)}$$  \hspace{1cm} (1)

where $\omega_d$ is the Debye frequency, $W$ is the electronic bandwidth, and $\mu$ measures the instantaneous Coulomb repulsion. In metals $\omega_d$ is typically two orders of magnitude lower than $W$, which accounts for the small $\mu^*$. However, the electronic bandwidth of the conduction level of $A_3C_{60}$ is about $W \approx 0.5eV$, while the phonon modes go up to an energy of $0.2eV$
which makes retardation highly ineffective in reducing $\mu^*$.

However, it has been proposed by Baskaran and Tosatti \cite{20}, and simultaneously by Chakravarty and Kivelson \cite{21}, that a repulsive bare interaction between the electrons on a single fullerene molecule can result in an effective attraction between electrons in the conduction level. Second-order perturbation theory (PT) in $U/t$ in the tight-binding model on the truncated icosahedral (TI) lattice with the on-site Hubbard interaction $U$ shows that $\mu^*$ becomes negative beyond $U/t \approx 3$ \cite{21}. Exact diagonalizations on small clusters \cite{22,23} also demonstrate the possibility of $\mu^* < 0$ for intermediate $U/t$. However, increasing the range of the interaction increases $\mu^*$ in second-order PT \cite{24,25}, and the validity of second-order PT is questionable in the region where $\mu^* < 0$ \cite{24}.

A numerical RG method has been developed recently by one of us to address these issues and get a reliable estimate of $\mu^*$ \cite{26}. This is based on the extention of Wilson’s momentum shell integration \cite{27} to the case of condensed fermion systems as developed by Shankar \cite{28}. The crucial distinction between the RG as applied to critical phenomena in statistical mechanics, and the RG as applied to condensed fermion systems is that in the latter case there are an infinite number of flowing coupling constants which are relevant to the low-energy behavior of the system (in infinite volume) \cite{28}. To illustrate the method for finite sizes, let us consider the case of interacting electrons on $C_{60}$. We start with the Hamiltonian

$$H = -t \sum_{\langle ij, s \rangle} (c_{is}^\dagger c_{js} + h.c.) + \sum_{i,j} V(i,j)n_i n_j$$

where the first term is just the tight-binding approximation and the second term represents a generic density-density interaction between the electrons. (We have also performed calculations for two different hopping matrix elements for the purely pentagonal versus the other bonds \cite{9}, but since the results are almost identical to the ones for the above hamiltonian, we report only the latter here). We solve the tight-binding problem and get 60 eigenstates, each belonging to a representation of the icosahedral group $I_h$ \cite{29}. We can then go to the zero-temperature fermionic path integral with the action
\[ S = \sum_{\eta_s} \int_{-\infty}^{\infty} \bar{\eta}_{ks} Z^{-1}(k)(i\omega - \epsilon(k))\eta_{ks}(\omega) \]
\[ - \sum_{\{k_i\}, ss'} \int_{-\infty}^{\infty} d\omega_1 \cdots d\omega_4 \frac{(2\pi)^3}{\delta(\omega_1 + \omega_2 - \omega_3 - \omega_4)} V(\{k_i\})\bar{\eta}_{k_1s}\bar{\eta}_{k_2s'}\eta_{k_3s'}\eta_{k_4s} \]

where \( \eta, \bar{\eta} \) are Grassmann variables representing the fermions, and \( V_{ss'}(k_1, k_2, k_3, k_4) \) is the matrix element of the bare interaction, and \( Z \) is the quasiparticle residue, or the wave function renormalization. For the bare theory \( Z = 1 \) and the energies are the tight-binding energies. This method produces results in excellent agreement with exact diagonalizations for small clusters [26].

We now integrate out the variables corresponding to the high energy levels (levels far from the conduction level) step by step. We go to two-loop order in the interaction matrix elements to compute effective values for the energy, the wave function renormalization, and the interaction. Each integration generates effective interactions for the remaining levels [26]. We continue until only the conduction level remains, and obtain an effective hamiltonian for the conduction level. In accordance with the formalism for infinite fermion systems [28], we allow all the independent couplings to flow separately. In order to reduce computation time we use the full symmetries of \( I_h \) to keep only symmetry-reduced matrix elements, in analogy with the Wigner-Eckhart theorem. This still leads to \( \approx 10^5 \) flowing couplings. We also ignore the frequency dependence of the effective interactions (they are irrelevant in the RG sense for properties near the fermi surface [28]). Apart from keeping track of \( Z \), going to two-loop order enables us to generate an internal validity criterion. As far as the effective interactions in the conduction level are concerned, the smallest energy denominators come from the excitations to levels close to the conduction level, which will be integrated out during the final steps of the RG. We compare the two-loop to the one-loop contribution to the effective interactions at the final RG step. When their ratio becomes \( \approx 1 \) we know that all loops will be important, and that truncating to two-loop is invalid. As will be evident from our numerical results, this method is reliable up to intermediate values of \( U/t \approx 5 \), which fortunately includes the physical regime for \( C_{60} \) [4].

We have considered two classes of models for the bare potential; the extended Hubbard
model with on-site and nearest neighbor interactions, a screened Ohno potential. We also considered the problem of only two additional electrons in the $T_{1u}$ conduction level (as long as the three- and higher-body interactions are negligible compared to the two-body interaction, these results should hold for arbitrary numbers of electrons). The two additional electrons can be in one of three channels $A_g$, $T_{1g}$, $H_g$, which roughly correspond to total angular momentum $L = 0, 1, 2$ respectively [29]. Let us now proceed to the results.

In Fig. 1 we show the effective interaction $U^*/t$ for the on-site Hubbard model as a function of the bare interaction $U/t$. From the non-monotonicity it is very evident that no naive correspondence can be made with the Anderson-Morel formula, eq(1). For $U/t < 5$ the triplet $T_{1g}$ pair state has the lowest energy. When considered in the context of a lattice problem, this will lead to itinerant ferromagnetism. There is also a level crossing between the $A_g$ and the $H_g$ pair states at $U/t \approx 3.5$, and for $U/t > 5$ the $A_g$ state is the ground state. Fig. 2 shows how far one can trust these results by plotting the ratio of the third order to the second order at the final step of RG as a function of $U/t$. We see that these results are reliable up to $U/t \approx 5 - 6$. For $U/t > 6$ the $A_g$ channel shows negative $U^*$, which was precisely the original counterintuitive claim that inspired this work. However, it unfortunately happens in a region where we cannot trust the result. Fig. 3 compares various different approximations for $U^*/t$ for the $A_g$ channel, from which it is clear that perturbation theory fails for physically interesting $U/t \approx 3 - 5$, and even one-loop RG differs significantly from the two-loop result. Finally, the ladder resummation [30], which corresponds in our language to a one-loop RG with only the Cooper channel, is also significantly different from the two-loop result. Qualitatively similar results hold for the other two channels.

The next set of three figures shows the same information for the screened Ohno potential [24] given by

$$V(i, j) = \frac{U \exp(-r_{ij}/\lambda)}{\sqrt{1 + r_{ij}^2/a^2}} \hspace{1cm} (4)$$

with the screening length $(\lambda)$ set to 0.75 of a $C - C$ bond length(= $a$) (this $\lambda$ is somewhat larger than the Thomas-Fermi screening length for $A_3C_{60}$ [4]). The major differences from
the Hubbard model are that $U^*$ is generally higher, and that the region of $U^* < 0$ has been pushed to higher $U/t$, as was predicted on the basis of second-order PT earlier \[24,25\]. It is easy to see from Fig. 5 that the method is trustworthy only to about $U/t \approx 3 - 4$ now, especially for the $T_{1g}$ and $H_g$ channels. Once again Fig. 6 shows the comparison between the different approximation methods. Adding a nearest neighbor interaction to the on-site Hubbard model leads to results very similar to Fig. 4.

A simple physical picture of the reduction of $U$ to $U^*$ is that the renormalized conduction level electrons interact weakly with the core primarily via the lowest $H_g$ and $T_{1g}$ collective modes \[31\] (the second is actually a spin wave), which act very much like phonons in producing an effective interaction between the added electrons. While this picture produces qualitatively correct results, \textit{i.e.}, that the $A_g$ pair state has the highest negative curvature, and that the $T_{1g}$ pair state always has positive curvature, the issue is complicated by nonzero overlaps between occupied and unoccupied levels in the interacting theory. We hope to present a full analysis in a future publication.

To summarize, we have used a novel finite-size RG method to compute the effective Coulomb interactions at the FS for a single $C_{60}$ molecule. The method has an internal self-consistency check, and is reliable in the physically interesting range of bare interaction strengths, as opposed to other approximation schemes. The results show that while the effective interactions are always repulsive for physically relevant bare interaction strengths, there is a dramatic reduction in strength of the repulsion compared to its bare value. It is fascinating to speculate that for $U/t > 6$ in the Hubbard model two added electrons will attract each other on $C_{60}$, although the method has reached the limit of its validity here. Clearly, this renormalization cannot be parametrized by the Anderson-Morel formula.

A useful feature that we have not yet exploited is to reduce the size of the problem, especially for strong couplings. One could run the RG as long as the validity criterion allowed one to, and then exactly diagonalize the much smaller effective problem.

The biggest deficiency of the above results is that since they include lattice screening phenomenologically at best (via Thomas-Fermi static screening \[32\]), they cannot be applied
with much confidence to superconducting $A_3C_{60}$. We are currently working on a RG scheme which will include the effects of polarization and charge fluctuations of neighboring fullerene molecules on $U^*$, which should go some way towards addressing the lattice screening problem \[33\]. In the absence of such a calculation one can believe that lattice screening should only reduce $U^*$, and that the viability of phonon mediated theories would be enhanced. In the Fig. 7 we present a model calculation of the coupled electron-phonon system, where the effective interactions at the conduction level in the Hubbard model have been taken into account. We have used the phonon coupling constants of Ref [12]. Despite the repulsive Coulomb interactions, the phonons succeed in binding the pairs.

There are many directions in which this work could be taken. One could calculate the exciton energies \[34\] for neutral and charged $C_{60}$, one could measure the spin structure of neutral $C_{60}$, which has been the subject of some controversy \[35\], and one could hope to elucidate metallic screening on the lattice.

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FIGURES

FIG. 1. The Coulomb pseudopotential $U^*/t$ for the on-site Hubbard model as a function of bare coupling strength $U/t$ for the $A_g$, $T_{1g}$, and $H_g$ channels.

FIG. 2. The ratio of the two-loop to one-loop contributions at the final RG step as a function of $U/t$ for the various channels for the on-site Hubbard model. Note that renormalized potential matrix elements are used in the loop calculations so that the result is reliable for much greater $U/t$ than perturbation theory.

FIG. 3. A comparison of different approximation methods for $U^*/t$ in the $A_g$ channel. All the methods differ appreciably from our two-loop RG results long before the two-loop RG becomes unreliable.

FIG. 4. The Coulomb pseudopotential $U^*/t$ for the Ohno potential with screening length $\lambda = 0.75a \approx 1\,\AA$ as a function of bare coupling strength $U/t$ for the $A_g$, $T_{1g}$, and $H_g$ channels.

FIG. 5. The ratio of the two-loop to one-loop contributions at the final RG step as a function of $U/t$ for the various channels for the Ohno potential.

FIG. 6. The same comparison as fig. 2 for the Ohno potential. Once again, all the methods differ appreciably from our two-loop RG results long before the two-loop RG becomes unreliable.

FIG. 7. A model calculation of the binding energy of two electrons in $eV$, including the effective $U^*$ from the electronic calculation, and including up to two vibrons \cite{18}. We have used $t = 2\,\text{eV}$ and the electron-phonon couplings from Ref. \cite{12}. It is clear that even in the Ohno case the phonons succeed in binding the electrons.