Coulomb Pseudopotential, Screening and Superconductivity in C_{60}

O. Gunnarsson and G. Zwicknagl
Max-Planck Institut für Festkörperforschung, D-7000 Stuttgart 80, Germany
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We estimate the Coulomb pseudopotential μ^* for a model of undoped C_{60}, renormalizing away all subbands but the t_{1u} conduction band by summing ladder diagrams in the screened interaction. The renormalization of μ^* is small. For doped C_{60} the screened U is calculated. A very efficient metallic screening is found for the intraband U, but not for the interband U, suggesting a large renormalization of μ^* within a ladder diagram approach. Exact results for a two-band model suggest, however, that the ladder diagrams strongly overestimate the renormalization.

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A number of papers [1–3] have discussed superconductivity in C_{60} using the McMillan formula [4]. According to this approach, the transition temperature T_c can be estimated from the electron-phonon coupling constant (mass enhancement parameter) λ and an effective Coulomb pseudopotential parameter μ^*, which accounts for the residual repulsion among the quasiparticles close to the Fermi energy E_F. Calculations [1–3] give $\lambda \sim \frac{1}{2}$ and if μ^* is assumed to have values typical for transition metals ($\mu^* \sim 0.1 - 0.2$), values for T_c of the right order of magnitude are obtained [1–3]. The parameter μ^* is related to $\mu = UN(0)$, where U is an effective Coulomb interaction and $N(0)$ is the density of states per spin at E_F. Retardation effects reduce μ^* relative to μ, and by renormalizing away electronic states further from E_F than typical phonon energies $\omega_{\rm ph}$, one obtains [5]

$$\mu^* = \mu/[1 + \mu \ln(W/\omega_{\rm ph})], \tag{1}$$

where W is a typical electronic energy. For C_{60} $\omega_{\rm ph}$ is believed to be of the order 0.1 eV [1–3]. Band structure calculations for C_{60} show many subbands, each about 0.5 eV broad and spread out over about 30 eV. If $W \sim \frac{30}{2} = 15$ eV, which appears to be a widespread opinion, one obtains $\mu^* \sim 0.2$, even if $\mu \gg 1$. Recently, Anderson has suggested that it is more appropriate to consider only the partly occupied (t_{1u}) conduction subband [6]. The corresponding small value of $W \sim 0.5/2$ eV together with $\omega_{\rm ph} \sim 0.1$ eV then leads to $\mu^* \approx \mu/(1+\mu)$, and with typical values of $N(0) \sim 10$ states/(eV-spin-C_{60} molecule) [7], $\mu^* \sim 0.1-0.2$ can only be obtained if U ($\sim 0.01-0.02$ eV) is extremely small. Anderson therefore concluded that a phonon mechanism cannot explain T_c for C_{60} [6].

In this paper we estimate the renormalization of μ due to all π subbands but t_{1u} for a model of undoped C_{60}. We find that the reduction of μ is rather small, and not properly described by Eq. (1). The resulting μ is very large. We then calculate the dielectric function for doped C_{60} in the random phase approximation (RPA) and find that the intraband Coulomb interaction is strongly reduced due to the metallic screening ($\mu \sim 0.4$). A model calculation suggests that higher-lying subbands give a small renormalization of μ.

In the traditional theory of μ^*, an electron-gas-like model is considered [5]. Ladder-diagram types of scattering processes of a pair $\mathbf{k}\uparrow, -\mathbf{k}\downarrow$ into states $\mathbf{k}'\uparrow, -\mathbf{k}'\downarrow$ are considered, where the energy $\varepsilon(\mathbf{k}')$ of the state $\mathbf{k}'$ belongs to the range that is renormalized away. In perturbation theory one obtains the renormalization

$$V(\mathbf{k},\mathbf{k}) \to V(\mathbf{k},\mathbf{k}) - {\sum}' \frac{|V(\mathbf{k},\mathbf{k}')|^2}{2|\varepsilon(\mathbf{k}')|}, \tag{2}$$

where $V(\mathbf{k},\mathbf{k}')$ is the matrix element for scattering a pair from $\mathbf{k}$ to $\mathbf{k}'$. By putting all elements for $|\varepsilon(\mathbf{k}')| \le W$ equal to U and the others equal to zero and by going beyond perturbation theory, the result in Eq. (1) is obtained. We now generalize this result to a model for undoped C_{60}, following the traditional approach [5] of summing ladder diagrams in the screened interaction.

C_{60} is characterized by a strong hopping inside the C_{60} molecule and a weak hopping between the molecules. We therefore use the molecular orbitals (MO) of an isolated molecule as a basis set for the solid. The discrete states of the molecule then broaden to narrow subbands, with a small mixing (for the t_{1u} band less than 5% of the weight) of the different MO's, which is neglected here. For the undoped case, we consider intramolecular but not intermolecular Coulomb interactions, using the model

$$H = \sum_{i\mathbf{k}\sigma} \varepsilon(i\mathbf{k}) n_{i\mathbf{k}\sigma} + \frac{1}{2} {\sum_{ijlm\sigma\sigma'}}' \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} U(ij,ml) \psi^\dagger_{i\mathbf{k}\sigma} \psi^\dagger_{j\mathbf{k}'\sigma'} \psi_{l\mathbf{k}'-\mathbf{q}\sigma'} \psi_{m\mathbf{k}+\mathbf{q}\sigma},$$

where i labels a subband. The $\varepsilon(i\mathbf{k})$ are generated from a tight-binding fit with a band-structure calculation [8,9]. We only consider the π-like bands resulting primarily from the $2p$ orbitals pointing radially out of the molecule. The σ-like bands should play a less important role in this context, as discussed below. The Coulomb integrals are

calculated as

$$U(ij,ml)=\sum_{\nu=1}^{60}\sum_{\mu=1}^{60}\alpha_i(\nu)\alpha_m(\nu)v(\nu,\mu)\alpha_j(\mu)\alpha_l(\mu), \quad (3)$$

where $\alpha_i(\nu)$ is the coefficient of the radial $2p$ orbital on the νth atom. The Coulomb integrals $v(\nu,\mu)$ are approximated as $e^2/|\mathbf{R}_\nu-\mathbf{R}_\mu|$, for $\nu\neq\mu$, where $\mathbf{R}_\nu$ and $\mathbf{R}_\mu$ are atomic sites on one C_{60} molecule, and $v(\nu,\nu)$ is set equal to v_0 for $\nu=\mu$. For a free C atom we obtain $v_0=12$ eV, while a lower limit ~ 6 eV was derived from a calculation for C_{60} including correlation effects [10]. We therefore consider $v_0=6$ and 12 eV. The intraband Coulomb integral $U_0\equiv U(ii,ii)\sim 3.7$ eV. This monopole Coulomb integral is heavily screened. Using local density calculations, we have estimated $U_0\sim 2.7$ eV for a *free* molecule [11]. Other calculations have found slightly larger values (~ 3 eV) [12–14]. This value is screened by the polarization of the surrounding molecules, leading to a reduction of U_0 to 0.9–1.2 eV [11]. Here we consider $U_0=1.2$ eV, which is close to experimental estimates (1.6 ± 0.2 eV [15] and 1.1 ± 0.2 eV [16]). For atoms multipole interactions $U(ij,ml)$ with $i\neq m$ and/or $j\neq l$ are less screened and typically reduced by (15–20)%. The polarizability calculated below suggests a more efficient screening for C_{60}, but as a conservative estimate we use a 15% reduction. A larger reduction would make the renormalization of the intraband U even less efficient than what is shown below in Table I. If σ-like states had been included, we would have had to consider $v(\mu,\nu)$ where the two functions with the same argument $\mathbf{r}$ have different symmetry, leading to multipole instead of monopole contributions. The corresponding Coulomb interband scattering from the π-like t_{1u} states into σ-like states should therefore be small.

There are two reasons to expect Eq. (1) to be inappropriate for C_{60}. First, in the integral over energy, leading to the logarithm, only the energies inside the subbands should be considered. For instance, if each of the sixteen subbands is 0.5 eV broad and the centers are separated by 1 eV, the logarithm, describing the renormalization down to the t_{1u} band, is reduced from ln(15.5/0.25)=4.1 to about 2.6. Alternatively, we could consider the higher subbands as smeared out, but the corresponding density of states is then smaller than $N(0)$, leading to a similar effect.

TABLE I. The intraband Coulomb interaction $\bar{U}$ after all subbands but t_{1u} have been renormalized away. The unrenormalized value was $U_0=1.2$ eV, and two values of the atomic on-site integral v_0 are considered. For $i=j$ the spin up and down electrons were forced to be in the same subband, while for i,j no such restriction was enforced. $E_{\min}$ and $E_{\max}$ give the centers of the highest and lowest subband considered. For $E_{\min}=0$ ($E_{\max}=0$) no fully occupied (completely empty) subband was considered.

$E_{\max}$	$E_{\min}$	$v_0=6$ eV		$v_0=12$ eV	
		$i=j$	i,j	$i=j$	i,j
0.00	0.00	1.200	1.200	1.200	1.200
1.17	0.00	1.018	1.018	0.949	0.949
1.17	−1.69	1.013	1.011	0.942	0.944
3.76	−1.69	1.011	0.974	0.938	0.888
3.76	−2.72	0.991	0.948	0.908	0.852
6.98	−7.97	0.988	0.937	0.900	0.820

Second, and more important, while in electron-gas-like models all Coulomb integrals are assumed to be similar, this is a poor approximation for C_{60}. Coulomb integrals of the type $U(ii,jj)$ where i belongs to t_{1u} and $j\neq i$ are usually small, since the factor $\alpha_i(\nu)\alpha_j(\mu)$ entering in Eq. (3) adds to zero when summed over ν, and a nonzero contribution is only obtained due to the relatively slow variation of $1/|\mathbf{R}_\nu-\mathbf{R}_\mu|$. Only the U's for scattering of two electrons from the t_{1u} band into the t_{1g} band, about 1 eV above, or into the g_g subband, about 3 eV below the t_{1u} band, are relatively large.

To estimate the renormalization of μ, we have calculated the T matrix. Thus we solve

$$T(\mathbf{k}i\uparrow,\mathbf{k}'j\downarrow;\mathbf{k}+\mathbf{q}m\uparrow,\mathbf{k}'-\mathbf{q}n\downarrow;\omega)=U(ij,mn)+i\sum_{i'j'}\int\frac{d\omega_2}{2\pi}\int\frac{d^3q'}{(2\pi)^3}G^0_{i'}(\mathbf{k}+\mathbf{q}'\uparrow;\omega-\omega_2)$$
$$\times G^0_{j'}(\mathbf{k}'-\mathbf{q}'\downarrow;\omega_2)U(ij,i'j')$$
$$\times T(\mathbf{k}+\mathbf{q}'i'\uparrow,\mathbf{k}'-\mathbf{q}j'\downarrow;\mathbf{k}+\mathbf{q}m\uparrow,\mathbf{k}'-\mathbf{q}n\downarrow;\omega),$$

where G^0 is the zeroth-order Green's function. The t_{1u} states are excluded in the sum over i' and j', which defines the "cutoff" used in the calculation of μ^*. The two first (the third and fourth) sets of arguments of T give momenta, band indices, and spins of the incoming (outgoing) electrons. For C_{60} i and j can be different, but both must correspond to either occupied or unoccupied states. Since we have neglected the dispersion of the bands, the T matrix then becomes independent of the momenta in this model. We consider the energy $\omega=2\varepsilon(t_{1u})$ for the electrons.

Table I shows the results $\bar{U}$ for the T matrix between two equal t_{1u} orbitals. In the renormalization, we have gradually included more and more subbands, with $E_{\max}$ ($E_{\min}$) giving the center of the highest unoccupied (lowest occupied) subband included. The table illustrates the importance of the t_{1g} band at 1.17 eV, for which the terms $i=j$ dominate. For larger energies the terms $i=j$ give a moderate contribution, and changing $E_{\min}$

from -2.72 to -7.97 eV and $E_{\max}$ from 3.76 to 6.98 eV has practically no effect. If terms $i \neq j$ are included, higher energies become only slightly more important.

We emphasize the importance of having different intraband and interband scattering. If only one Coulomb matrix element and one density of states enters the theory, the renormalization will reduce the Coulomb interaction by a certain *factor*. In the present case, the renormalization due to the higher subbands leads to the *subtraction* of a term, which is rather independent of U_0 [see, e.g., Eq. (2)]. For instance, for $v_0 = 6$ eV and $U_0 = 1.2$ eV, the reduction in Table I is 0.25 eV and for $U_0 = 0.6$ eV (not shown) it is 0.31 eV. This is also illustrated by the failure of an attempt to describe the results in the upper and lower row in the left column of Table I using Eq. (1). For $v_0 = 12$ eV and $i \neq j$, this would require that $\ln(W/W_l) \sim 0.05$, where W_l is the lower cutoff, which is clearly nonsensical.

We now consider the doped system and in particular how efficiently metallic screening can reduce the Coulomb interaction. We use an analytical expression [7] for the 3×3 Hamiltonian generating the t_{1u} bands. The remaining (σ- and π-like) bands are described in a tight-binding model with four (one $2s$ and three $2p$) orbitals per atom and parameters from the literature [17], neglecting dispersion. The polarizability is calculated in the RPA. The Coulomb integrals are calculated as in Eq. (3), except that ν is now a combined site and orbital index, and the long-range interaction between molecules is included. The atomic integral v is assumed to be zero unless the two orbitals with the same $\mathbf{r}$ argument have the same quantum number. This allows for polarization due to charge transfer across the molecules and between the molecules, but not due to polarization of the individual C atoms. The polarizability P is calculated in a basis which consists of Bloch sums over products of two atomic orbitals, which with our approximations leads to 240×240 matrices. The screened Coulomb interaction V is then calculated according to the matrix formula $V = U(1+PU)^{-1}$, where U is the unscreened interaction.

For a free, neutral C_{60} molecule, this leads to the polarizability 50 Å^3, compared with 65 Å^3 in a much more sophisticated quantum chemical calculation [18]. For a free molecule and $v_0 = 12$ eV, screening reduces the on-site interaction from 12 to 5.7 eV and the nearest-neighbor interaction from 10.3 to 5.0 eV. For distant neighbors there is antiscreening, since the introduction of an electron on one atom moves screening charge towards the opposite side of the molecule, and the interaction is increased from about 2.0–2.2 to about 2.9–3.7 eV, depending on the separation (6.5–7.1 Å).

We now consider the statically screened Coulomb interaction between Bloch states for the doped solid

$$V(\alpha\beta, \gamma\delta, \mathbf{q}) = N\langle \alpha\mathbf{k}\uparrow, \beta{-}\mathbf{k}\downarrow | W(\mathbf{r}, \mathbf{r}') | \gamma(\mathbf{k}+\mathbf{q})\uparrow, \delta(\mathbf{k}+\mathbf{q})\downarrow \rangle$$

where $W(\mathbf{r}, \mathbf{r}')$ is the screened interaction, $|\alpha\mathbf{k}\rangle = (1/\sqrt{N}) \sum_{\mathbf{R}} \exp(-i\mathbf{k}\cdot\mathbf{R})|\alpha\mathbf{R}\rangle$, and N is the number of molecules in all of space. $\mathbf{R}$ denotes the lattice sites and α a molecular orbital. We treat an fcc lattice with the lattice parameter 14.1 Å.

The intraband matrix elements have a moderate $\mathbf{q}$ dependence and are drastically reduced due to the screening. For small values of $|\mathbf{q}|$, $V(\alpha\alpha, \alpha\alpha, \mathbf{q})$ is of the order $1.5/[2N(0)]$, where $2N(0)$ is the density of states per molecule at the Fermi energy, and $V(\alpha\beta, \alpha\beta, \mathbf{q})$ ($\alpha \neq \beta$) is of the order $0.75/[2N(0)]$. For instance, for $v_0 = 12$ eV and $\mathbf{q} = (0.05, 0.05, 0.05)2\pi/a$ we obtain the values 0.094 and 0.060 eV for $\alpha = \beta$ and $\alpha \neq \beta$, respectively, while the corresponding values for $\mathbf{q} = (0.5, 0.5, 0.5)2\pi/a$ are 0.065 and 0.031 eV, respectively. The values of μ are obtained by multiplying by $N(0) = 7.5$ eV^{-1}. Considering that large $|\mathbf{q}|$ dominates in an average over the Fermi surface, we estimate such an average to give $\mu \sim 0.4$ for $\omega = 0$. These results can be qualitatively understood from $V = U/(1+PU)$, where $P \sim 2N(0)$ and $PU \gg 1$. Then $V \sim 1/[2N(0)]$. The differences between $\alpha = \beta$ and $\alpha \neq \beta$ follows from the matrix nature of $V = U/(1+PU)$ and the slight differences in the bare Coulomb matrix elements for $\alpha = \beta$ and $\alpha \neq \beta$.

The interband scattering matrix elements, corresponding to multipole interaction, are reduced much less by the metallic screening than the monopole, intraband elements. For instance, for $\mathbf{q} = (0.5, 0.5, 0.5)2\pi/a$ the largest interband scattering matrix element is 0.98 eV without screening, 0.15 eV with nonmetallic screening, and 0.10 eV with the full screening, while for the intraband matrix element ($\alpha \neq \beta$) the corresponding numbers are 1.9, 1.6, and 0.031 eV.

The screened intraband matrix elements have a strong energy dependence, in particular, for small $\mathbf{q}$. For $\mathbf{q} = (0.25, 0.25, 0.25)2\pi/a$ the values of $V(\alpha\beta, \alpha\beta, \mathbf{q})$ are 0.048, 0.083, and -0.33 eV for ω=0.0, 0.1, and 0.2 eV, respectively, and for $\mathbf{q} = (0.5, 0.5, 0.5)2\pi/a$ the corresponding values are 0.031, 0.048, and 0.42 eV.

The energy can be lowered by forming a state $\sum_{\alpha\neq\beta} c_{\alpha\beta}|\alpha\uparrow\beta\downarrow\rangle$, since there are nonzero matrix elements of the type $V(\alpha, \beta, \beta, \alpha, \mathbf{q})$. For instance, for $\mathbf{q} = (0.5, 0.5, 0.5)2\pi/a$ we obtain the energy 0.011 eV.

The electron-phonon coupling in doped C_{60} due to alkali ion optic phonons has been estimated to be large [19], while experiments suggest a small coupling [20]. We find that in RPA the effective metallic screening reduces this coupling by almost 2 orders of magnitude, suggesting that the alkali phonons give a small contribution to T_c but that they could possibly influence other properties.

There are now some intricate questions about how to proceed to include the renormalization due to the higher subbands. We could follow the traditional approach [5] and first screen the Coulomb interaction and then renormalize away the higher states by calculating the T matrix, or we could proceed in the opposite order. In the first case μ^* should be substantially reduced relative to

$\mu \sim 0.4$, because the interband matrix elements are large compared with the intraband elements [cf. Eq. (2)]. In the second case, the unscreened intraband U is reduced but remains large. Introducing metallic screening then again leads to a screened V of the order $1/[2N(0)]$, and in contrast to the first approach, renormalizing away the higher bands hardly changes μ^*. These two approaches differ in the way vertex corrections are included.

This problem can be addressed for a model with two subbands (a half-filled band 1 and an empty band 2) with the coupling

$$U_{12} \sum_i [\psi^\dagger_{i2\uparrow} \psi^\dagger_{i2\downarrow} \psi_{i1\downarrow} \psi_{i1\uparrow} + \text{H.c.}],$$

where i is a site index. We assume that the band separation $\Delta\varepsilon$ is large compared with U_{12} and the hopping matrix element t, and that $1/[2N(0)] \sim U_{12}^2/\Delta\varepsilon \ll U_{11}$, where U_{11} is the intraband scattering. The "traditional" approach then gives a large renormalization of μ due to the higher band. In this limit we can, however, renormalize away the higher band using perturbation theory, which leads to a new effective one-band Hamiltonian, with $U_{11} \to U_{11} - U_{12}^2/\Delta\varepsilon$. Although the corresponding μ should be drastically reduced due to *intraband* processes, we can see that the higher band has a negligible effect. Thus the "traditional" approach gives an incorrect result for this model. The assumptions of the model are not quite valid for C_{60}, but the results are suggestive.

T_c was calculated using a program extending Ref. [21] and solving the Eliashberg equation, assuming a single-phonon mode with $\hbar\omega_{\text{ph}} = 0.1$ eV, $\mu^* = 0.4$, and a Lorentzian-shaped band with width 0.4 eV. To obtain a reasonable T_c (~ 30 K), $\lambda \sim 1$ is needed. Here $\mu^* \sim 0.4$ refers to the t_{1u} band cutoff. Renormalizing this value to the cutoff $\omega_{\text{ph}} = 0.1$ eV (as in the McMillan formula) using Eq. (1) gives $\mu^* \sim 0.3$.

We have studied the renormalization of the Coulomb pseudopotential due to scattering into higher-energy subbands in C_{60}, and the screening of the Coulomb matrix elements in the RPA. As a result of the difference between intraband and interband scattering, there are large differences from calculations for electron-gas-like models, and the traditional formula Eq. (1) is not valid. For our model of the undoped system, the interband matrix elements are small compared with the intraband elements and the renormalization due to higher-energy subbands is small. For the doped system, the metallic screening drastically reduces the intraband elements, leading to $\mu \sim 0.4$, while the interband scattering matrix elements are less reduced. In the traditional approach [5], renormalizing away higher subbands should then give a large reduction of μ. In a simple model, we find, however, that this approach greatly overestimates the effect of the higher subbands, suggesting that μ^* may not be much smaller than μ. Such a result for μ^* (~ 0.4 for the t_{1u} band cutoff) would, however, not require very much larger values of λ (~ 1) than what has been calculated [1–3]. Interesting questions about the effect of vertex corrections, in particular, for the renormalization inside the t_{1u} conduction band remain. Finally, we note that the small value of the statically screened Coulomb interaction does not mean that one should use such a small U in a (positive U) Hubbard model of doped C_{60}, but that this efficient static screening should be a result of solving the model.

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