SCREENED INTERACTION AND COULOMB PSEUDO-POTENTIAL IN C_{60}

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Abstract

We study the screening for C_{60} in the random phase approximation. For a free molecule we find a substantial reduction of the interaction between close atoms but an "antiscreening" of the interaction between distant atoms. For a C_{60} solid there is an efficient screening of the intraband Coulomb interaction ($U \sim 0.05$ eV), leading to $\mu = UN(0) \sim 0.4$, where $N(0)$ is the density of states per spin and molecule. We then study the renormalization of the Coulomb pseudo-potential μ^* due to higher subbands in a simple two-band model. We find that the renormalization is small, although a traditional approach, summing ladder diagrams in the screened interaction, predicts a large renormalization. This suggests that μ^* may not be much smaller than $\mu \sim 0.4$ in C_{60}, where μ^* has been renormalized down to the t_{1u} band edge. To obtain reasonable transition temperatures, T_c, this value of μ^* (~ 0.4) requires values of the electron-phonon coupling constant λ, which are not very much larger than what has been calculated.

1. INTRODUCTION

C_{60} has attracted particularly much interest in the physics community after it was discovered that C_{60} doped with K or Rb is superconducting[1]. A number of papers[2, 3, 4] have calculated the electron-phonon coupling constant (mass enhancement parameter) λ and found values of the order 1/2. The superconductivity transition temperature T_c was then estimated using the McMillan formula[5]. T_c's of the right order of magnitude were obtained[2, 3, 4], provided that the effective Coulomb pseudo-potential parameter μ^* was assumed to have values $\sim 0.1 - 0.2$, typical for transition metals. 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 ω_{ph} by summing ladder diagrams, one obtains for an electron gas like model[6]

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

where W is a typical electronic energy. For C_{60} ω_{ph} is believed to be of the order 0.1 eV[2, 3, 4]. Band structure calculation for C_{60} show many subbands, each about 0.5 eV broad and spread out over about 30 eV. If $W \sim 30/2 = 15$ eV, which appears to be a widespread opinion, one obtains $\mu^* \sim 0.2$, even if $\mu >> 1$. Recently, Anderson has suggested that it is more appropriate to only consider the partly occupied (t_{1u}) conduction subband[7]. The corresponding small value of $W \sim 0.5/2$ eV together with $\omega_{ph} \sim 0.1$ eV then leads to $\mu^* \approx \mu/(1+\mu)$, and with typical values of $N(0) \sim 10$ states/(eV-spin-C_{60} molecule)[11], $\mu^* \sim 0.1 - 0.2$ 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}[7].

In the traditional theory of μ^*, an electron gas like model is considered[6], and the screened Coulomb interaction is calculated. This model is then mapped onto a model where only states up to some typical energy of the order ω_{ph} enter. This requires a renormalization of μ to μ^*. In this "traditional" theory the renormalization is taken into account by summing ladder diagrams in the screened interaction, describing the scattering of a pair of electrons $\mathbf{k}\uparrow, -\mathbf{k}\downarrow$ into states $\mathbf{k}'\uparrow, -\mathbf{k}'\downarrow$, 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 $|\varepsilon(\mathbf{k}')| \leq W$ equal to U and the others equal to zero and by going beyond perturbation theory, the result in Eq. (1) is obtained. The processes beyond perturbation theory can be considered by solving the equation for the T-matrix. We observe, however, that this "traditional" approach is based on summing a particular set of diagrams, and that it is not obvious that other diagrams are not important.

This approach requires knowledge about the screened interaction, and in Sec. 2 we present our formalism for calculating the polarizability. In Sec. 3 we present the model and in Sec. 4 results for the screened interaction, both for an isolated molecule and for the C_{60} solid. In Sec. 5 we present a simple two-band model, where the higher band can be renormalized away exactly. We show that in this case the "traditional" approach incorrectly predicts a large renormalization of μ^*. In Sec. 6 we discuss the results. A summary of these results have been presented elsewhere[8].

2. POLARIZABILITY

In this section we discuss the method for calculating the polarizability for C_{60}. The irreducible polarizability for the frequency ω is in the Random Phase Approximation (RPA) given by

$$P_0(\mathbf{r},\mathbf{r}',\omega) = 2\sum_{\mathbf{k}n}\sum_{\mathbf{k}'n'} \frac{\psi_{\mathbf{k}n}(\mathbf{r})\psi^*_{\mathbf{k}n}(\mathbf{r}')\psi^*_{\mathbf{k}'n'}(\mathbf{r})\psi_{\mathbf{k}'n'}(\mathbf{r}')}{\varepsilon(\mathbf{k}n) - \varepsilon(\mathbf{k}'n') - \omega}[f(\mathbf{k}n) - f(\mathbf{k}'n')] \tag{3}$$

where $\psi_{\mathbf{k}n}(\mathbf{r})$ is the wave function for a state with the wave vector $\mathbf{k}$, band index n and

energy $\varepsilon(\mathbf{k}n)$. $f(\mathbf{k}n)$ is the Fermi function and a factor 2 comes from summation over spin. The Bloch states $\psi_{\mathbf{k}n}(\mathbf{r})$ are expressed as

$$\psi_{\mathbf{k}n}(\mathbf{r}) = \frac{1}{\sqrt{N}} \sum_{iL\nu} e^{i\mathbf{k}\cdot\mathbf{R}_\nu} c_{iL}(\mathbf{k}n)\phi_L(\mathbf{r} - \mathbf{R}_i - \mathbf{R}_\nu), \tag{4}$$

where $\mathbf{R}_i$ gives the positions of the 60 atoms inside a molecule relative to the position $\mathbf{R}_\nu$ of that molecule. $L \equiv (l, m)$ labels the basis functions ϕ_L (e.g., one $2s$ and three $2p$) on a given atom and N is the number of molecules in the system. We now introduce a new basis set for describing the polarizability

$$\Phi_{\mathbf{q}iLL'} = \frac{1}{\sqrt{N}} \sum_{\nu} e^{i\mathbf{q}\cdot\mathbf{R}_\nu} \phi_L(\mathbf{r} - \mathbf{R}_i - \mathbf{R}_\nu)\phi_{L'}(\mathbf{r} - \mathbf{R}_i - \mathbf{R}_\nu). \tag{5}$$

If we assume that the overlap between ϕ's on different atoms can be neglected, we obtain

$$P_0(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mathbf{q}} \sum_{iLL'} \sum_{jL_1L_2} \Phi^*_{\mathbf{q}iLL'}(\mathbf{r})\tilde{P}_0(iLL', jL_1L_2, \mathbf{q}, \omega)\Phi_{\mathbf{q}jL_1L_2}(\mathbf{r}'), \tag{6}$$

where the sum over $\mathbf{q}$ is limited to the first Brillouin zone and

$$\tilde{P}_0(iLL', jL_1L_2, \mathbf{q}, \omega) =$$

$$\frac{2}{N} \sum_{\mathbf{k}nn'} \frac{c_{iL}(\mathbf{k}n)c^*_{iL'}(\mathbf{k}+\mathbf{q}n')c^*_{jL_1}(\mathbf{k}n)c_{jL_2}(\mathbf{k}+\mathbf{q}n')}{\varepsilon(\mathbf{k}n) - \varepsilon(\mathbf{k}+\mathbf{q}n') - \omega}[f(\mathbf{k}n) - f(\mathbf{k}+\mathbf{q}n')]. \tag{7}$$

In this way we have expressed the irreducible polarizability in terms of matrices with the dimension 60*4*4=960, if there are 4 basis functions per atom and one C_{60} molecule per unit cell.

We now consider the screened interaction $W(\mathbf{r}, \mathbf{r}', \omega)$, which satisfies the equation

$$W(\mathbf{r}, \mathbf{r}', \omega) = v(\mathbf{r} - \mathbf{r}') + \int d^3r'' \int d^3r''' v(\mathbf{r} - \mathbf{r}'')P_0(\mathbf{r}'', \mathbf{r}''', \omega)W(\mathbf{r}''', \mathbf{r}', \omega), \tag{8}$$

where $v(\mathbf{r} - \mathbf{r}')$ is the unscreened Coulomb interaction. We calculate the matrix elements of $W(\mathbf{r}, \mathbf{r}', \omega)$

$$W(iLL', jL_1L_2, \mathbf{q}, \omega) = \int d^3r \int d^3r' \Phi_{\mathbf{q}iLL'}(\mathbf{r})W(\mathbf{r}, \mathbf{r}', \omega)\Phi^*_{\mathbf{q}jL_1L_2}(\mathbf{r}'). \tag{9}$$

For a periodic system, the matrix element vanishes if the two Φ's have different $\mathbf{q}$-arguments (in the first Brillouin zone), and we therefore only consider equal $\mathbf{q}$ arguments. We can then write down a matrix equation for W

$$W(iLL', jL''L''', \mathbf{q}, \omega) = v(iLL', jL''L''', \mathbf{q})$$

$$+ \sum_{mL_1L_2} \sum_{nL_3L_4} v(iLL', mL_1L_2, \mathbf{q})\tilde{P}_0(mL_1L_2, nL_3L_4, \mathbf{q}, \omega)W(nL_3L_4, jL''L''', \mathbf{q}, \omega), \tag{10}$$

where we have also introduced the matrix elements of v. The results for W can be obtained by from the matrix expression

$$W = (1 - v\tilde{P}_0)^{-1}v. \tag{11}$$

We can also write down a similar expression for the polarizability

$$\tilde{P} = (1 - v\tilde{P}_0)^{-1}\tilde{P}_0. \tag{12}$$

If we only consider the intraband screening inside the partly occupied 3-fold degenerate t_{1u} band, this calculation can be reduced to treating 9*9 matrices. We have not followed such an approach here, since the the higher lying bands are important for the screening of the interband scattering Coulomb matrix elements.

3. MODEL

We now introduce a model of C_{60} for which we calculate the screened interaction. We consider the one-particle Hamiltonian describing the states in an isolated molecule

$$H^0_{Mol} = \sum_{iL\sigma} \varepsilon_L n_{iL\sigma} + \sum_{iLjL'\sigma} [t(iL, jL')\psi^\dagger_{iL\sigma}\psi_{jL'\sigma} + h.c.], \tag{13}$$

where ε_L gives the energies of the $2s$ and $2p$ levels and $t(iL, jL')$ gives the hopping integrals between the states. We use the parameters of Tomanek and Schluter[9], and assume nearest neighbor hopping as in [9]. This Hamiltonian gives the discrete levels of an isolated molecule and the corresponding molecular orbitals (MO) $\varphi(\mathbf{r})$. In the solid these levels are broadened to narrow subbands. We neglect the corresponding dispersion for all the levels, except for the t_{1u} level in the doped system, which is partially occupied. The dispersion of the t_{1u} band is important, since it influences the metallic screening in the doped solid. To obtain this dispersion, we have earlier given a 3*3 analytical **k**-dependent Hamiltonian[11], which is used here. We also neglect the mixing in the solid of MO's with different energies. This mixing is weak, since the energy separation of the MO's is large compared with the hopping matrix elements. For instance, for a model which only includes the radial $2p$ (π) orbital[10], we find, after averaging over the **k**-points, that more than 95 % of the weight of the t_{1u} conduction band states consists of t_{1u} MO's.

The Coulomb integrals between the orbitals are defined as

$$\tilde{v}(iL\nu, jL'\mu; mL''\gamma, nL'''\delta) =$$

$$e^2 \int d^3r \int d^3r' \phi_L(\mathbf{r}-\mathbf{R}_i-\mathbf{R}_\nu)\phi_{L'}(\mathbf{r}-\mathbf{R}_j-\mathbf{R}_\mu)\frac{1}{|\mathbf{r}-\mathbf{r}'|}\phi_{L''}(\mathbf{r}'-\mathbf{R}_m-\mathbf{R}_\gamma)\phi_{L'''}(\mathbf{r}'-\mathbf{R}_n-\mathbf{R}_\delta). \tag{14}$$

Since we neglect the overlap of the functions centered on different atoms, $\tilde{v}$ is nonzero only if $(i\nu) = (j\mu)$ and $(m\gamma) = (n\delta)$. We note that if $L = L'$ and if $L'' = L'''$, this integral corresponds to the interaction between two monopoles. The corresponding integral is large. If these two conditions are not fulfilled, the interaction is between multipoles and therefore smaller. We therefore make the assumption

$$\tilde{v}(iL\nu, jL'\mu; mL''\gamma, nL'''\delta) =$$

$$\delta_{L,L'}\delta_{L'',L'''}\delta_{i,j}\delta_{\nu,\mu}\delta_{m,n}\delta_{\gamma,\delta} * \begin{cases} \frac{e^2}{|\mathbf{R}_i+\mathbf{R}_\nu-\mathbf{R}_j-\mathbf{R}_\mu|} & \text{for } |\mathbf{R}_i+\mathbf{R}_\nu-\mathbf{R}_j-\mathbf{R}_\mu| > 0 \\ v_0 & \text{for } |\mathbf{R}_i+\mathbf{R}_\nu-\mathbf{R}_j-\mathbf{R}_\mu| = 0 \end{cases} \tag{15}$$

For the nearest neighbor ($d \sim 1.42$ Å) the monopole interaction is about 10 eV, while the neglected dipole-dipole interaction is about a factor 4 smaller and the quadrupole-quadrupole interaction is still smaller. Since the monopole-monopole interaction decays as the inverse separation, while the dipole-dipole and quadrupole-quadrupole interactions decay as the separation to the inverse third and fifth power, respectively, the approximation (15) is very good for somewhat larger separations. For instance, for the second nearest neighbours ($d \sim 2.4$ Å) the dipole-dipole interaction is a factor of 10 smaller than the monopole-monopole interaction. For the on-site interaction, we have obtained $v_0 = 12$ eV from atomic calculations, by studying how the levels move in a self-consistent calculation when an electron is added to the atom. With the approximation (15) it is then straightforward to calculate $v(iLL, jL'L', \mathbf{q})$ by using the Ewald technique. The dimension of the matrices in (11) is now reduced to 60*4=240. The approximation (15) means that we neglect the polarization of the individual C atoms, but that we allow for polarization through charge transfer from one atom to another. Since the diameter of the C_{60} molecule is large compared with the size of a C atom, we expect the charge transfer across the molecule, and for the doped systems the charge transfer between the molecules, to dominate over the polarization of the individual atoms.

In the calculations for the solid, we consider an fcc lattice with the lattice parameter 14.1 Å and one C_{60} molecule per unit cell.

4. RESULTS FOR THE SCREENED INTERACTION

We first consider a free C_{60} molecule. The formulas in the previous section are still valid, if we take the limit of a large lattice parameter so that all quantities become **k**- and **q**-independent. In particular, we consider the macroscopic molecular polarizability

$$\alpha(\omega) = \frac{e^2}{3} \sum_{k=1}^{3} \sum_{iL} \sum_{jL'} \mathbf{R}_{i,k} \tilde{P}(iLL, jL'L', \mathbf{q}, \omega) \mathbf{R}_{j,k}, \tag{16}$$

where $\mathbf{R}_{i,k}$ is the kth component of the vector $\mathbf{R}_i$. For the static polarizability we obtain the result $\alpha(0) = 50$ Å^3. This result should be compared with a much more sophisticated quantum chemical calculation[12], which gave the result 65 Å^3. This suggests that our approximations give semi-quantitative results.

We have also calculated the screened interaction between the different atoms on a free molecule. The results are compared in Fig. 1 with the unscreened interaction. For short distances the screening reduces the interaction by about a factor of two. For larger separations there is, however, an antiscreening effect and the "screened" interaction is *larger* than the unscreened interaction. The reason is that when one electron is put at one side of the molecule, charge is moved from this side to the other side of the molecule. If we now put a second electron on the other side of the molecule, this second electron sees the screening charge and therefore a more repulsive potential than if there had been no screening. The results furthermore illustrate that it is not very realistic to describe a free C_{60} molecule in a Hubbard model with only an on-site Coulomb interaction.

Figure 1: The screened interaction (dots) as a function of separation for a free C_{60} molecule compared with the unscreened interaction (full line). For distances shorter than the nearest neighbor separation the unscreened interaction has been extended (dashed line) to the atomic integral $v_0 = 12$ eV.

We next study the screened interaction between Bloch-states in the solid,

$$W(\alpha\beta, \gamma\delta, \mathbf{q}) = N < \alpha\mathbf{k} \uparrow \beta(\mathbf{k}+\mathbf{q}) \uparrow |W(\mathbf{r}, \mathbf{r}')|\gamma(-\mathbf{k}) \downarrow, \delta - (\mathbf{k}+\mathbf{q}) \downarrow >, \qquad (17)$$

where

$$|\alpha\mathbf{k}> = \frac{1}{\sqrt{N}} \sum_{\nu} e^{i\mathbf{k}\cdot\mathbf{R}_\nu} \varphi_\alpha(\mathbf{r} - \mathbf{R}_\nu), \qquad (18)$$

is a Bloch sum of molecular orbitals $\varphi_\alpha(\mathbf{r})$. Table 1 gives results for this interaction for the intraband and interband interaction. For the intraband interaction we give matrix elements $W(\alpha\alpha, \alpha\alpha, \mathbf{q})$ between two equal t_{1u} conduction band state and $W(\alpha\alpha, \beta\beta, \mathbf{q})$ between two different t_{1u} states. These matrix elements do not describe scattering processes but give the Coulomb interaction between two electrons. These are of particular interest, since matrix elements of this type determine the value of μ. We also give the largest interband scattering matrix element W^{inter}, which occurs for the scattering of two electrons from the t_{1u} conduction band to the t_{1g} subband at about 1 eV higher energy. Matrix elements of this type are of interest, since they control the scattering into higher bands, which renormalizes μ into μ^*. We show results for the unscreened interaction and the screened interaction in the undoped and in the doped systems. As a comparion we also show results for a free molecule. For the solid we consider $\mathbf{q} = \mathbf{G}/2$ and $\mathbf{q} = \mathbf{G}/20$, where $\mathbf{G} = 2\pi/a(1,1,1)$ is a reciprocal lattice vector.

We first observe that the unscreened interband scattering matrix elements are in general much smaller than the intraband Coulomb interaction. The W^{inter} given in Table 1

Table 1: The intraband Coulomb interactions between two equal $W(\alpha\alpha,\alpha\alpha,\mathbf{q})$ and two different $W(\alpha\alpha,\beta\beta,\mathbf{q})$ t_{1u} conduction band Bloch states as well as the largest interband scattering matrix element W^{inter} $(t_{1u} \rightarrow t_{1g})$. We consider both a free molecule (Mol) and a solid (Sol), where in the latter case $\mathbf{q} = \mathbf{G}/2$ and $\mathbf{q} = \mathbf{G}/20$, where $\mathbf{G} = 2\pi/a(1,1,1)$. The unscreened (Unscr) and screened (Scr) interactions in the undoped (Undop) and, for the solid, doped systems are shown.

	System	$W(\alpha\alpha,\alpha\alpha,\mathbf{q})$		$W(\alpha\alpha,\beta\beta,\mathbf{q})$		W^{inter}	
		$\mathbf{q}=\mathbf{G}/20$	$\mathbf{q}=\mathbf{G}/2$	$\mathbf{q}=\mathbf{G}/20$	$\mathbf{q}=\mathbf{G}/2$	$\mathbf{q}=\mathbf{G}/20$	$\mathbf{q}=\mathbf{G}/2$
Unscr	Mol	3.81		3.63		0.77	
Scr	Mol	3.72		3.67		0.14	
Unscr	Sol	176.8	2.03	176.7	1.86	1.01	0.98
Undop	Sol	78.9	1.68	78.8	1.63	0.15	0.15
Doped	Sol	0.094	0.065	0.060	0.031	0.10	0.10

is the largest element and most other elements are substantially smaller. To understand this, we consider the formula for the integrals between molecular orbitals (MO's) for a free molecule

$$v(\alpha\beta,\gamma\delta) = \int d^3r \int d^3r' \varphi_\alpha(\mathbf{r})\varphi_\beta(\mathbf{r})\frac{1}{|\mathbf{r}-\mathbf{r}'|}\varphi_\gamma(\mathbf{r}')\varphi_\delta(\mathbf{r}') \tag{19}$$

The intraband matrix elements considered here have $\alpha = \beta$ and $\gamma = \delta$, while these indices are different for the interband scattering matrix elements. In the latter case, the MO's with the same $\mathbf{r}$-argument are orthogonal, and therefore the the product must have sign-changes, so that it would integrate to zero if the Coulomb interaction $1/|\mathbf{r}-\mathbf{r}'|$ were not present. In general, there are therefore large cancellations when these integrals are performed for the interband scattering matrix elements. Alternatively, we can see an interband scattering matrix element as describing the interaction between two charge distributions with zero net charge, i.e., a multipole interaction, while for the intraband matrix elements considered here the interaction is between charge distributions where the net charge is unity, i.e., a monopole interaction.

When screening is introduced for the free molecule, the interband elements are reduced substantially, while the intraband elements are essentially unchanged. The reason is that the intraband elements correspond to the interaction between charges which are rather uniformly spread out over the molecule. Redistribution of the other electrons in the molecule can therefore not reduce this interaction very much, in particular in the model used here. For the interband scattering, on the other hand, the corresponding charge is very nonsymmetric and redistribution of charge inside the molecule can give an efficient screening.

For the solid there are two additional effects. Firstly the interaction is now between Bloch states, and there is therefore an interaction with charges on the neighboring molecules. The unscreened interaction therefore has a very strong $\mathbf{q}$ dependence for the monopole intraband interaction but not for the multipole interband scattering. The latter

result follows from the fact that multipoles on different molecules interact weakly due to the large separation. Secondly, the screening can now also take place as a polarization of neighboring molecules and for the doped system screening charge can be transferred from one molecule to another. For the doped case the intraband Coulomb matrix elements are drastically reduced. The reason is that these matrix elements correspond to setting up net charges on the molecules. The system then reacts by transferring screening charge of the opposite sign and a similar magnitude to the molecules. The result is a very efficient screening. For the interband matrix elements, on the other hand, the corresponding charge integrates to zero on each molecule, and the system has no reason to transfer charge between the molecules. The reduction of W^{inter} in the doped system is primarily due to the fact that the partial filling of the t_{1u} band in the doped system allows for new *interband* transitions, which in this case improves the screening.

The intraband interaction for the doped system is of the order $1/[2N(0)]$. This can be traced to the formula (11) $W = (1-v\tilde{P}_0)^{-1}v$. If we transform the matrix $\tilde{P}_0$ in (7) to a basis of Bloch sums over t_{1u} orbitals, the diagonal elements $\tilde{P}_0(mm, mm, \mathbf{q}, \omega = 0) \approx 2N(0)/3$, where m now labels the three t_{1u} orbitals and we have considered small values of $|\mathbf{q}|$. Since the corresponding Coulomb matrix elements $v(mm, m^{'}m', \mathbf{q})$ are large, we have that $v(mm, m^{'}m^{'}, \mathbf{q})2N(0)/3 >> 1$. If we further assume that all elements $v(mm, m^{'}m', \mathbf{q})$ are equal for a fixed value of $\mathbf{q}$ it follows that the screened interaction between two t_{1u} orbitals is $1/[2N(0)]$ for small $|\mathbf{q}|$. This agree well with the weighted average

$$\frac{1}{3}W(\alpha\alpha, \alpha\alpha, \mathbf{q}) + \frac{2}{3}W(\alpha\alpha, \beta\beta, \mathbf{q}), \tag{20}$$

of the two intraband matrix elements in Table 1. The difference between these two matrix elements can be traced back to the difference between $v(mm, m^{'}m^{'}, \mathbf{q})$ for $m = m^{'}$ and $m \neq m^{'}$.

We now consider the value of μ, which is obtained by multiplying the density of states $N(0)$. If we neglect the $\mathbf{q}$ dependence and use the small $\mathbf{q}$ result above we obtain $\mu = 0.5$. We next take the relatively weak $\mathbf{q}$-dependence into account by considering the average (20) for some representative values of $\mathbf{q}$, which allows us to estimate an average over the Fermi surface. Multiplying by the density of states, $N(0) = 7.5$ eV^{-1}, we then obtain $\mu \sim 0.4$.

5. TWO-BAND MODEL

In this section we discuss the renormalization of μ due to higher subbands in a simple two-band model. Thus we consider the Hamiltonian

$$H = \sum_i \sum_{n=1}^{2} \sum_{\sigma} \varepsilon_n n_{in\sigma} + t \sum_{<ij>} \sum_{n=1}^{2} \sum_{\sigma} \psi^{\dagger}_{in\sigma}\psi_{jn\sigma}$$

$$+U_{11} \sum_i \sum_{nn^{'}\sigma\sigma^{'}}{}^{'} n_{in\sigma}n_{in^{'}\sigma^{'}} + U_{12} \sum_i [\psi^{\dagger}_{i2\uparrow}\psi^{\dagger}_{i2\downarrow}\psi_{i1\downarrow}\psi_{i1\uparrow} + h.c.], \tag{21}$$

where i is a site index, ε_n the energies of the two levels, t is the hopping integral, U_{11} is the intraband interaction and U_{12} is the interband scattering. The particularly strong

interband scattering between the t_{1u} and t_{1g} bands in C_{60} is of the same type as the last term in Eq. (21), namely the excitation of *two* electrons from band 1 to band 2. For the scattering between other bands there are also terms of the type $\psi^\dagger_{i2\uparrow}\psi_{i1\uparrow}n_{i1\downarrow}$, namely the excitation of *one* electron. Such terms are not included in our two-band model. We assume that the band separation $\Delta\varepsilon \equiv \varepsilon_2 - \varepsilon_1$ is large compared with U_{12} and the hopping matrix element t

$$\Delta\varepsilon >> U_{12}, \quad t, \tag{22}$$

and that

$$\frac{1}{2N(0)} \sim \frac{U_{12}^2}{\Delta\varepsilon} << U_{11}. \tag{23}$$

In the limit (22) we can renormalize away the higher band using second order perturbation theory. We introduce projection operators

$$Q_i = (1 - n_{i2\uparrow})(1 - n_{i2\downarrow}), \tag{24}$$

and

$$Q = \sum_i Q_i \equiv 1 - P. \tag{25}$$

We now want to transform H to an effective Hamiltonian, H_{eff}, acting only in the space Q with no electrons in the higher band. For this purpose we formally introduce all N electron basis states obtained by occupying the states $|in\sigma>$ in all possible ways. The eigenvalues of the corresponding large matrix are the eigenvalues of the model. We now partition this matrix into subblocks corresponding to the spaces Q and P. Matrix algebra then shows that the Hamiltonian H_{eff} in

$$(z - H_{eff}) = Q(z - H)Q - QHP[P(z - H)P]^{-1}PHQ \tag{26}$$

has the same eigenvalues z as H. We have

$$PHQ = U_{12}\sum_i \psi^\dagger_{2i\uparrow}\psi^\dagger_{2i\downarrow}\psi_{i1\uparrow}\psi_{i1\downarrow}. \tag{27}$$

We now consider low-lying states, for which z is close to the ground-state energy. If we let $P(z - H)P$ act on such states, the diagonal elements are at least of the order $2\Delta\varepsilon$, which is much larger than the nondiagonal elements of the order t or U_{12} in the limit we consider here. We then replace $[P(z - H)P]^{-1}$ by a diagonal matrix, where the diagonal elements are $1/(2\Delta\varepsilon)$ for the parts of the matrix which enter in (26). Then we obtain a new effective interaction U_{eff} in H_{eff}, where

$$U_{eff} = U_{11} - \frac{U_{12}^2}{2\Delta\varepsilon}. \tag{28}$$

For a general Coulomb interaction and degenerate bands, this renormalization procedure would also lead to three-body and one-body terms, while for the model studied here the renormalization only leads to a two-body term of the same type as the one already

present in H. The properties of H_{eff} only depend on U_{eff}/t. In the limit considered here, U_{eff} differs very little form U_{11}. We therefore can conclude that within this model and in the limit (23), the effect of the higher band is negligible. Below we shall demonstrate that if we instead use the "traditional" approach[6], a large renormalization of μ^* is, nevertheless, incorrectly obtained.

The first step in this approach is the calculation of the statically screened interaction. Thus we first calculate the irreducible polarizability P_0 in Eq. (3) in a similar way as in Sec. 2. We introduce the basis states

$$\Phi_{\mathbf{q}nn'} = \frac{1}{\sqrt{N}} \sum_{\nu} e^{i\mathbf{q}\cdot\mathbf{R}_\nu} \phi_n(\mathbf{r}-\mathbf{R}_\nu)\phi_{n'}(\mathbf{r}-\mathbf{R}_\nu), \tag{29}$$

where n, $n^{'}$ label the two bands and $\mathbf{R}_\nu$ are the lattice sites. The polarizability can then be written as

$$P_0(\mathbf{r},\mathbf{r}^{'}) = \sum_{\mathbf{q}nn^{'}} \Phi_{\mathbf{q}nn'}(\mathbf{r})^* \tilde{P}_0(n,n^{'},\mathbf{q})\Phi_{\mathbf{q}nn'}(\mathbf{r}) \tag{30}$$

where

$$\tilde{P}_0(n,n^{'},\mathbf{q}) = \frac{2}{N}\sum_{\mathbf{k}} \frac{f(\mathbf{k}n)-f(\mathbf{k}+\mathbf{q}n^{'})}{\varepsilon(\mathbf{k}n)-\varepsilon(\mathbf{k}+\mathbf{q}n^{'})}. \tag{31}$$

We consider the case when band 1 is half-filled and band two is empty and make the approximations

$$\tilde{P}_0(1,1,\mathbf{q}) \approx -2N(0), \qquad \tilde{P}_0(1,2,\mathbf{q}) \approx -\frac{2}{\Delta\varepsilon}, \qquad \tilde{P}_0(2,2,\mathbf{q}) = 0, \tag{32}$$

where $2N(0)$ is the density of states per atom at the Fermi energy. The matrix element of the Coulomb interaction are defined as in Eq. (9). In the two-band model (21), the nonzero bare Coulomb matrix elements are then given by $v(11,11,\mathbf{q})$, $v(12,12,\mathbf{q})$ and $v(22,22,\mathbf{q})$ together with other matrix elements obtained by obvious permutations of the indices, while, e.g., $v(11,12,\mathbf{q}) \equiv 0$. This simplifies the solution of Eq. (10) for the screened interaction, and we obtain

$$U_{11}^{scr} \equiv W(11,11,\mathbf{q}) = \frac{v(11,11,\mathbf{q})}{1-\tilde{P}_0(1,1,\mathbf{q})v(11,11,\mathbf{q})} = \frac{U_{11}}{1+2N(0)U_{11}} \approx \frac{1}{2N(0)}, \tag{33}$$

where we have used (23) that $2N(0)U_{11} >> 1$. We further obtain

$$U_{12}^{scr} \equiv W(12,12,\mathbf{q}) = \frac{v(12,12,\mathbf{q})}{1-\tilde{P}_0(1,2,\mathbf{q})v(12,12,\mathbf{q})} \approx \frac{U_{12}}{1+2U_{12}/\Delta\varepsilon} \approx U_{12} \tag{34}$$

since we have assumed (22) that $U_{12}/\Delta\varepsilon << 1$. This calculation illustrates how the screening of the intraband interaction U_{11} is much more efficient than the screening of the interband interaction U_{12}.

We now follow the "traditional" approach[6] for renormalizing away the higher band, i.e., first the Coulomb interaction is screened and then the T-matrix is calculated, allowing

Figure 2: Diagrams contributing to the effective interaction in the two-band model. The dashed line represents the screened interaction in the 1-1 channel (U_{11}^{scr}) and the wiggly line the scattering of two electrons from band 1 to band 2 (U_{12}^{scr}).

for scattering into the band 2 but not 1. This corresponds to diagram a and b in Fig. 2. The equation for the T-matrix can be solved in perturbation theory and we obtain

$$U_{11}^{scr} - \frac{(U_{12}^{scr})^2}{2\Delta\varepsilon} \approx \frac{1}{2N(0)} - \frac{U_{12}^2}{2\Delta\varepsilon} << \frac{1}{2N(0)}, \tag{35}$$

since we are considering the limit $1/2N(0) \approx U_{12}^2/\Delta\varepsilon$. It follows that the renormalization of μ due to the higher band is large according to the "traditional" approach, while it is small according to the second order perturbation treatment above. Since we have considered parameters where the second order perturbation treatment is valid, we have to conclude that at least for the model considered here, the "traditional" approach incorrectly gives a large renormalization due to higher bands.

In Fig. 2 c-e we show other diagrams which are also of second order in U_{12}^{scr}. These diagrams give an effective particle-particle interaction in the 1-1 channel, which could be used in the calculation of the T matrix for scattering only in the 1-1 channel, i.e., summing ladder diagrams involving only electrons in band 1. Using the same approximations as above we then obtain the contributions

$$U_{11}^{scr} - \frac{(U_{12}^{scr})^2}{2\Delta\varepsilon} + 2\frac{(U_{12}^{scr})^2}{2\Delta\varepsilon}(2N(0)U_{11}^{scr}) - \frac{(U_{12}^{scr})^2}{2\Delta\varepsilon}(2N(0)U_{11}^{scr})^2$$

$$= U_{11}^{scr} - \frac{(U_{12}^{scr})^2}{2\Delta\varepsilon}(\frac{1}{1+2N(0)U_{11}})^2. \tag{36}$$

This result agrees with the result obtained if we use the effective Hamiltonian (26,28) and screen the corresponding U_{eff}, which gives

$$\frac{U_{eff}}{1+2N(0)U_{eff}} \approx U_{11}^{scr} - \frac{(U_{12}^{scr})^2}{2\Delta\varepsilon}(\frac{1}{1+2N(0)U_{11}})^2. \tag{37}$$

6. DISCUSSION

We first discuss the renormalization of μ within the "traditional" approach[6], where the ladder diagrams in the screened interaction are summed. Within this approach there is an essential difference between electron gas like models and C_{60}, due to the fact that there is no reason why the interband scattering and intraband matrix elements should be

similar for C_{60}. For instance, in the simple formula (2), obtained in perturbation theory, the intraband matrix element enters in the first term but interband matrix elements in the second term on the right hand side. If the interband matrix elements are much smaller than the intraband elements, the renormalization becomes very small. We have discussed this earlier[8] for a simple model of undoped C_{60}. Table 1 illustrates that this is typical for undoped C_{60}. For doped C_{60} the situation is completely different. Now the interband matrix elements are larger than the intraband elements (see Table 1), and we expect the renormalization to be efficient and μ^* to be substantially smaller than $\mu \sim 0.4$. In both cases the simple formula (1) fails to describe the renormalization, since this formula assumes equal intra- and interband matrix elements.

In Sec. 4 we presented results for a simple two-band model, and showed that within this model, the large renormalization of μ predicted by the "traditional" approach is incorrect. In this model the parameters were chosen so that perturbation theory is valid. For C_{60}, the interband matrix elements typically *are* smaller than the separations of the subbands. Because of the large number of subbands, we find that perturbation theory is, nevertheless, not valid for the lower lying subbands. For C_{60} there are furthermore more general Coulomb terms than in the model in Sec.4, which lead to three-body terms in the transformed Hamiltonian. We can therefore not prove that the conclusions for the two-band model apply to C_{60}, but the results for this model, nevertheless, raises strong doubts about using the "traditional" approach for C_{60}.

Even if the renormalization due to the higher bands is inefficient, μ^* is, nevertheless, not very large (~ 0.4), due to the efficient screening. The T_c was calculated using a program extending Ref. [13] and solving the Eliashberg equation, assuming a single phonon mode with $\hbar\omega_{ph} = 0.1$ eV, $\mu^* = 0.4$ and a Lorentzian shaped band with the width 0.4 eV. To obtain a reasonable T_c ($\sim$ 30 K), $\lambda \sim 1$ is needed. This value is not very much larger than the calculated values[2, 3, 4] for λ, given the uncertainties both in the calculation of μ^* and λ as well as possible effects from violations of Migdal's theorem.

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