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The possible explanation of electric-field-doped C_{60} phenomenology in the framework of Eliashberg theory

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(Dated: November 15, 2018)

In a recent paper (J.H. Schön, Ch. Kloc, R.C. Haddon and B. Batlogg, Nature 408 (2000) 549) a large increase in the superconducting critical temperature was observed in C_{60} doped with holes by application of a high electric field. We demonstrate that the measured T_c versus doping curves can be explained by solving the (four) s-wave Eliashberg equations in the case of a finite, non-half-filled energy band. In order to reproduce the experimental data, we assume a Coulomb pseudopotential depending on the filling in a very simple and plausible way. Reasonable values of the physical parameters involved are obtained. The application of the same approach to new experimental data (J.H. Schön, Ch. Kloc and B. Batlogg, Science 293 (2001) 2432) on electric field-doped, lattice-expanded C_{60} single crystals (T_c =117 K in the hole-doped case) gives equally good results and sets a theoretical limit to the linear increase of T_c at the increase of the lattice spacing.

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Keywords: Eliashberg equations, fullerenes.

After obtaining the surface field-effect doping of high-quality organic crystals, J.H. Schön, Ch. Kloc, R.C. Haddon and B. Batlogg have carried out the same experiment on C_{60} crystals[1,2,3], achieving superconductivity up to 52 K in hole-doped samples[1]. The fundamental importance of this experiment is due to the fact that the superconductivity is obtained by field-effect doping the original single-crystal material and, thus, without modifying its structural properties. The authors of the experiments believe as plausible that the doping charges are confined only in a single layer of C_{60} molecules[1]. The physical parameters of C_{60} are rather well known; the symmetry of the order parameter appears to be s-wave and the electron-phonon interaction is widely indicated as the main cause for the appearance of superconductivity in this material. In the present paper we show that a theory suitable for reproducing the experimental T_c versus filling data reported in Ref. 1 is the Migdal-Eliashberg theory[4,5,6,7,8,9,10], even though, in fullerenes, the Migdal Theorem probably breaks down[11] due to the large energy of phonons and the rather low energy of the Fermi level[12,13]. As a first approximation, we neglect these complications and see where the theory in its most simple form and with the smallest number of free parameters can lead. Notice that a study of the phenomenology of field-doped C_{60} in the framework of the Eliashberg theory was carried out also in Ref. 14 although in a different way. Figure 1 (a) was obtained by normalizing the original data from Ref. 1 (and from more recent experimental data in lattice-expanded C_{60}, as we will discuss later) so that the maximum of T_c corresponds to half filling (here indicated by $n = 1$). In order to fit these experimental data, we solved the Eliashberg equations in the general case of non-half filling, finite bandwidth and flat normal density of states. The physical quantities that appear in the fit are thus: the bandwidth W, the electron-phonon spectral function $\alpha^2F(\Omega)$ and the Coulomb pseudopotential μ. It is also necessary to set a cut-off energy ω_C related to the renormalization of the Coulomb pseudopotential. We took the value of W from literature[12] : W=250 meV (independent of doping since the lattice structure does not change[1]) and we assumed the cut-off energy ω_C=W as in Ref. 10. The spectral function is unknown but, as a first approximation, one can assume that $\alpha^2F(\Omega)$ is proportional to the phonon density of states[12] $F(\Omega)$ of C_{60} (see figure 1 (b)) and nearly equal to the transport spectral function $\alpha^2F_{tr}(\Omega)$.

To determine the proportionality factor between $\alpha^2F(\Omega)$ and $F(\Omega)$, we took the experimental resistivity data $\rho(T)$ for the electron-doping case from the figure 5 of Ref. 1, in the range between $T = 225$ K and $T = 245$ K. The $\rho(T)$ curves appear to have the same shape and slope at all gate voltages, thus, as a first approximation, the ratio λ_{tr}/ω_P between the transport coupling constant λ_{tr} and the plasma frequency ω_P can be assumed to be doping-independent. The phonon density of states $F(\Omega)$ and the functions $\alpha^2_{e,h}(\Omega)$ (for both electrons and holes) are doping-independent as well, since the lattice parameters do not change[6]. As a result, also the electron-phonon coupling constants $\lambda_{e,h} = 2\int d\Omega\alpha^2_{e,h}F(\Omega)/\Omega$ are independent of doping. From the high-temperature expression of the resistivity: $\rho = 8\pi k_B T\lambda_{tr}/(\omega_P^2\varepsilon_0\hbar^2)$, and by using the value of the plasma frequency reported in literature[12] $\hbar\omega_P = 1.36$ eV, we can deduce the value of the transport electron-phonon coupling constant λ_{tr}[15]. In the electron-doping case we find $\lambda_{tr} = 1.1$ and, as a first approximation, we assume $\lambda_{tr} \equiv \lambda_e$. As far as the hole-doped coupling constant λ_h is concerned, from resistivity data and from calculations of the electron density of states J.H. Schön et al.[2] estimate that $\lambda_h/\lambda_e = 1.5$ and so we find $\lambda_h = 1.5\lambda_e = 1.65$.

In order to obtain the exact maximum value of the crit-

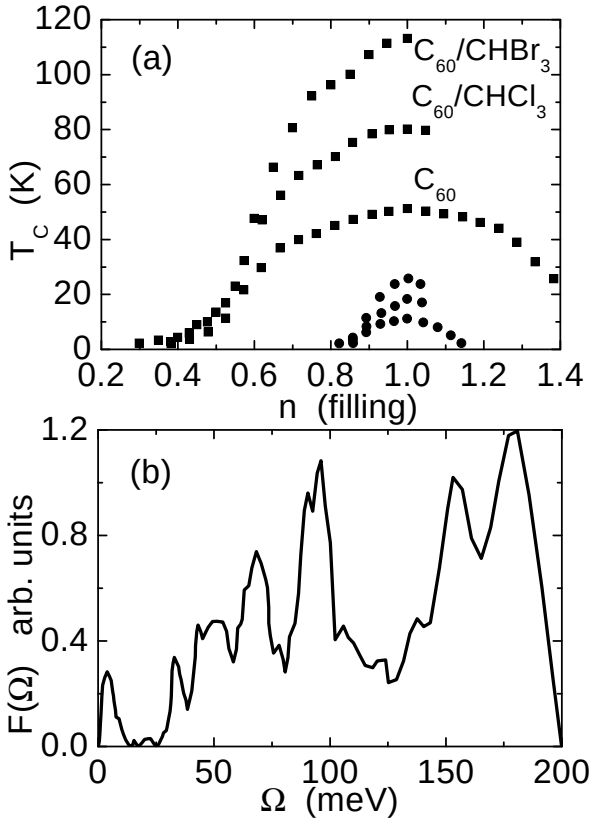

FIG. 1: (a) Experimental T_c versus filling n data for hole doping (full squares) and electron doping (full circles) [from[1,21]]; (b) The C_{60} phonon density of states $F(\Omega)$ [from[12]].

ical temperature in the half-filling case, as shown in figure 1 (a), we solved the Eliashberg equations in the half-filling case by using for the Coulomb pseudopotential μ the values 0.395 and 0.336 in the electron and hole doped case, respectively. This result is in very good agreement with theoretical calculations[12] that predict $\mu = 0.3 - 0.4$.

Two critical remarks can be made to our basic assumptions.

First: due to the degeneracy of C_{60}-orbitals, one has probably to deal with a multi-band problem[16]. It is well known that, in such a case, transport and superconducting properties can be managed by different groups of electrons but, in this way, there would be several free parameters and the model would become too complex. Instead, our simple approximation allows precise quantitative predictions.

Second: we have assumed for simplicity that the normal density of states (NDOS) is flat even if in recent theoretical calculations[17] it is not so. This calculated NDOS can be approximately reproduced by a very simple analytical formula: $N_N(\omega) = 1 + 1.5 exp(-|\omega|/\alpha)$ with $\alpha = 10$ meV. If we use this expression of the NDOS in the Eliashberg equations, with the same values of μ, i.e. 0.395 and 0.336, we find that the exact maximum T_c is obtained by using $\lambda = 0.843$ and $\lambda = 1.58$. The difference with respect to the case where $N_N(\omega) = 1$ is small, so that the I-V curves measured in the tunable SNS weak-link junctions of Ref. 3 can be fitted equally well by both the NDOS. Moreover, if the energy dependence of the NDOS is symmetric and the peak at the Fermi level is not too narrow, its effects on some physical quantities can be approximately simulated by an efficient value[6] of λ. In conclusion, if $\Delta E = (\omega_P^2 \varepsilon_0 \hbar^2)\rho_0/7.5 \geq \hbar\omega_D$ (where ω_D is the Deybe frequency and ρ_0 is the residual resistivity), the effect of a non-flat NDOS can be neglected[6]. In our case[1] $\rho_0 = 250 \div 300$ $\mu\Omega$ cm, $\Delta E = 62 \div 74$ meV and $\hbar\omega_D \cong \hbar\omega_{ln} = 27$ meV, where $\omega_{\log} = \exp(\frac{2}{\lambda}\int_0^{+\infty} d\Omega \frac{\alpha^2 F(\Omega)}{\Omega} \ln\Omega)$.

Since the lattice is not modified by the field-effect doping, we assume the electron-phonon coupling constant λ and the width of the conduction band W independent of filling n. Now we can try to reproduce the experimental values of T_c versus n by solving the Eliashberg equations in the non-half-filling case[8]. This is a hard task because there are three equations to be solved for the calculation of the gap $\Delta(i\omega_n)$, the renormalization function $Z(i\omega_n)$ and the asymmetric part of the self-energy $\chi(i\omega_n)$ – which is always equal to zero in the half-filling case – plus one equation that represents the conservation of the particles' number and is necessary for calculating the shift of the chemical potential $\delta\mu$. Thus the Eliashberg equations in the non-half-filling case, for finite bandwidth and s-wave symmetry of the order parameter read:[8]:

$$\Delta(i\omega_n)Z(i\omega_n) = \pi k_B T \sum_{m=-\infty}^{+\infty} [\lambda(i\omega_n - i\omega_m) - \mu^*/W] \cdot \vartheta(|\omega_C| - \omega_m) P(i\omega_m)\Theta(i\omega_m) \quad (1)$$

$$\omega_n Z(i\omega_n) = \pi k_B T \sum_{m=-\infty}^{+\infty} \lambda(i\omega_n - i\omega_m) N(i\omega_m)\Theta(i\omega_m) \quad (2)$$

$$\chi(i\omega_n) = -\pi k_B T \sum_{m=-\infty}^{+\infty} \lambda(i\omega_n - i\omega_m) M(i\omega_m) \quad (3)$$

$$n = 1 - (\pi k_B T/W) \sum_{m=-\infty}^{+\infty} M(i\omega_m) \quad (4)$$

where ϑ is the Heaviside function, $i\omega_m = i\pi(2m+1)k_B T$ with $m = 0, \pm 1, \pm 2, ...$ and

$$\mu^* = \mu/[1 + \frac{k_B T \mu}{W} \int_{-W}^{W} d\varepsilon \Sigma_{m=0}^{+\infty} \frac{\vartheta(\omega_m - \omega_C)}{\omega_m^2 + (\varepsilon - \delta\mu)^2}] \quad (5)$$

$$\lambda(i\omega_n - i\omega_m) = \int_0^{+\infty} d\Omega \frac{\alpha^2(\Omega)F(\Omega)}{\Omega^2 + (i\omega_n - i\omega_m)^2} \quad (6)$$

$$P(i\omega_m) = \frac{\Delta(i\omega_m)Z(i\omega_m)}{R(i\omega_m)}, N(i\omega_m) = \frac{\omega_m Z(i\omega_m)}{R(i\omega_m)} \quad (7)$$

$$R(i\omega_m) = \sqrt{\omega_m^2 Z^2(i\omega_m) + \Delta^2(i\omega_m) Z^2(i\omega_m)} \quad (8)$$

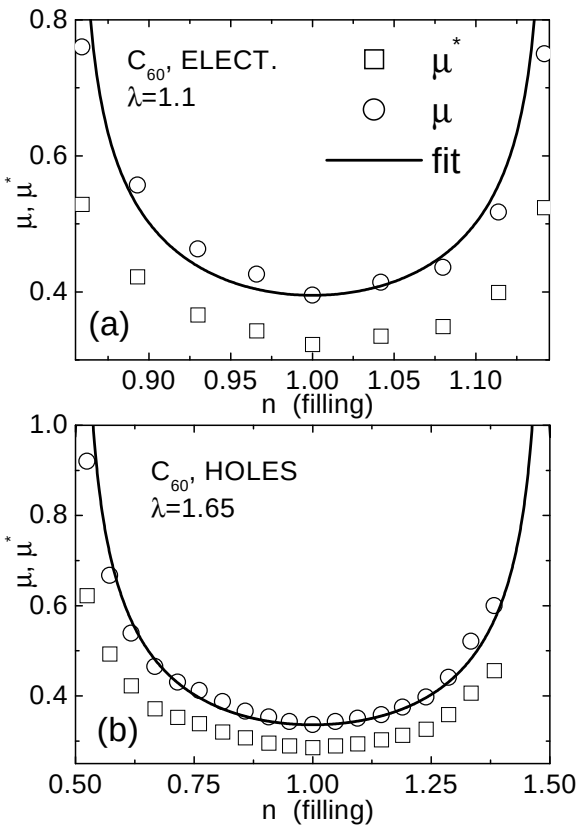

FIG. 2: Coulomb pseudopotential μ (open circle), μ^* (open square) and theoretical fit (solid line) versus filling n: (a) electron-doped case, (b) hole-doped case.

$$\Theta(i\omega_m) = \{\arctan(W_-(i\omega_m)) + \arctan(W_+(i\omega_m))\}/\pi \tag{9}$$

$$M(i\omega_m) = 0.5\ln[\frac{1+W_-^2(i\omega_m)}{1+W_+^2(i\omega_m)}] \tag{10}$$

and $W_\mp(i\omega_m) = \{W \mp [\delta\mu - \chi(i\omega_m)]\}/R(i\omega_m)$.

In order to reproduce the experimental values of T_c versus doping, and having excluded the dependence of λ on the filling, the only possibility is to assume that the Coulomb pseudopotential μ is variable with the filling n: $\mu \equiv \mu(n)$. We find the values of $\mu(n)$ by solving the equations (1)-(4) conditioned to the obtainment of the experimental $T_c(n)$. In figure 2 we show the $\mu(n)$ dependence in the two cases: (a) electron-doped C_{60} and (b) hole-doped C_{60} (open circles). It is important to notice that here the definition of μ^* is more general than usual because it depends on the shift of the chemical potential and on the value of the bandwidth. In the usual and less general definition[5], if $\omega_C = W$ then $\mu = \mu^*$ while in our case it is always $\mu > \mu^*$ as we can see in figure 2 (open squares).

Is it possible to explain this particular dependence of μ on the filling, obtained by the fit of the critical temperature of figure 1 (a)? This can be done in two or three dimensions. In a very recent paper[19] the authors affirm that it is not so sure that the superconductivity in this material is 2D. For completeness we examine both the 2D and 3D cases. From the definition of μ[18] and from the very simple analytical expression of the Thomas-Fermi dielectric function in the 3D case $\varepsilon(q,n) = 1 + k_S^2(n)/q^2$ it follows:

$$\mu(n) = \frac{1}{4\pi^2\hbar v_F}\int_0^{2k_F}\frac{V(q)}{\varepsilon(q,n)}qdq \tag{11}$$

where $V(q) = 4\pi e^2/q^2$. In the half-filling case, the calculation of the integral gives:

$$\mu(n=1) = [\frac{k_S^2(n=1)}{8k_F^2}]\ln[1+(\frac{4k_F^2}{k_S^2(n=1)})] \tag{12}$$

where k_F and k_S are the Fermi and Thomas-Fermi wave vector, respectively. We can expand the dielectric function in the vicinity of the half filling ($n = 1$) by remembering that, since the physical parameters λ, $\alpha^2F(\Omega)$ and W are fixed, T_c is maximum when $\mu(n)$ is minimum, which means that $\varepsilon(n)$ is maximum (see definition of $\mu(n)$) i.e. $\partial\varepsilon/\partial n = 0$ for $n = 1$. For simplicity, and to minimize the number of free parameters, we arrest the expansion to the second-order terms, even though the range of the series expansion is not so small in the hole-doping case:

$$\varepsilon(q,n) = \varepsilon(q,n=1) + \frac{1}{2}[\frac{\partial^2\varepsilon(q,n)}{\partial n^2}]_{n=1}(n-1)^2 + ... \tag{13}$$

and by substituting in the definition of $\mu(n)$ we find

$$\mu(n) = \mu(n=1)\frac{ln(1+a^2/[1+b(n-1)^2])}{ln(1+a^2)} \tag{14}$$

where $a = 2k_F/k_S(n = 1)$ and $b = [(1/k_S)(\partial^2 k_S/\partial n^2)]_{n=1}$. By starting from the values of $\mu(n = 1)$ that give the exact experimental T_c in the electron- and hole-doping case and using eq. 12 we can calculate $2k_F/k_S(n = 1)$. We obtain 1.05 for hole doping and 0.77 for electron doping. Now we are left with only one free parameter, $b = (1/k_S)[\partial^2 k_S/\partial n^2]_{n=1}$, that can be adjusted to fit the μ versus n curves with eq. 14. The results are very good (see figure 2 (a) and (b), solid lines): for electron doping $b = -25$ and for hole doping $b = -4$. Of course, here we used the 3D version of the Thomas-Fermi theory but, as pointed out by the authors of ref. 1-3, the field-effect charge injections is likely to be confined to the first atomic layer of C_{60} crystal, so it is worthwhile to examine also the 2D case. The analytical expression of the Thomas-Fermi dielectric function[20] is now $\varepsilon(q,n) = 1 + k_S(n)/q$ and

$$\mu(n) = \frac{k_F}{\pi\hbar v_F}\int_0^{2k_F}\frac{V(q)}{\varepsilon(q,n)\sqrt{4k_F^2-q^2}}dq \tag{15}$$

where $V(q) = 2\pi e^2/q$[20]. By following the same approach as before we find:

$$\mu(n=1) = 2\frac{[\arctan(\frac{a}{\sqrt{a^2-1}})-1]}{\sqrt{a^2-1}} \tag{16}$$

and finally

$$\mu(n) = \mu(n=1)\frac{\sqrt{a^2-1}}{\sqrt{a^{*2}-1}} \cdot \frac{[\arctan(\frac{a^*}{\sqrt{a^{*2}-1}})-1]}{[\arctan(\frac{a}{\sqrt{a^2-1}})-1]} \quad (17)$$

with $a^* = a[1+0.5b(n-1)^2]$.

In this case, from eq. 16, we can calculate $a \equiv 2k_F/k_S(n=1)$. We find two solutions: 1.9 and 13.8 for hole doping and 2.1 and 10.3 for electron doping. Again we have only one free parameter, $b = (1/k_S)[\partial^2 k_S/\partial n^2]_{n=1}$, that can be adjusted to fit the μ versus n curves with eq. 17. We find $b = -43$ and $b = -73$ for electron doping while for hole doping we have $b = -4$ and $b = -8$, since now two possible values for a are present. In the range of interest, the results are exactly equal to the 3D case and the 2D curves perfectly overlap those obtained in that case. If we compare the 2D and 3D densities of charge carriers obtained from the a values of the fits to the values predicted at optimal doping and corresponding to 3 carriers per C_{60} ($\sim 3 \cdot 10^{18}$ carriers/m^2 and $\sim 4.2 \cdot 10^{27}$ carriers/m^3, respectively) we obtain interesting results. The 3D density and the 2D one obtained from the small a values are of the same order of magnitude of the predicted densities, even if they don't coincide. On the contrary the 2D density obtained from the large a values is more than 2 orders of magnitude greater than the predicted one and, thus, the corresponding solution for a must be rejected on the basis of physical arguments. It follows that the results of the fit by using our model do not allow us to understand whether the system is better described by 2D or 3D approach, even if the experimental results[1,21] clearly point to a 2D nature of the charge injected region.

Note that, surprisingly, the curves $T_c(n)/T_c(n=1)$ and $\mu(n=1)/\mu(n)$ are perfectly superimposed.

Now, since we have the approximate analytical dependence of μ from n, we can solve the Eliashberg equations for any value of the filling and calculate different physical quantities. By using the standard technique of Padé approximants we can carry out the analytical continuation of $Z(i\omega_n)$, $\Delta(i\omega_n)$, $\chi(i\omega_n)$ and of the normalized superconductive density of states (DOS) $N_S(i\omega_n)/N_N(i\omega_n) = [N(i\omega_n)\Theta(i\omega_n) - M(i\omega_n)]/[N_N(i\omega_n)\Theta_N(i\omega_n) - M_N(i\omega_n)]$. Here the quantities with suffix N are calculated for $\Delta(i\omega_n) = 0$. In figure 3, we can see how the shape of $Z(\omega)$, $\Delta(\omega)$ and $\chi(\omega)$ changes for three different values of the doping in the hole-doped case: $n = 1$ ($T_c = 51$ K), $n = 1.189$ ($T_c = 46$ K) and $n = 1.287$ ($T_c = 39$ K). Figure 4 shows the normalized superconducting DOS as a function of the energy, calculated by the analytical continuation of the imaginary-axis DOS for the same doping values as in Fig. 3. In the non-half-filling cases ($n = 1.189$ and $n = 1.287$) the calculated DOS is asymmetric. In principle low-temperature SIN tunneling experiments on this field-doped material would make it possible to observe this asymmetry. The calculated values of the ratio $2\Delta/k_B T_c$ are slightly greater than the experimental results of Ref. 3.

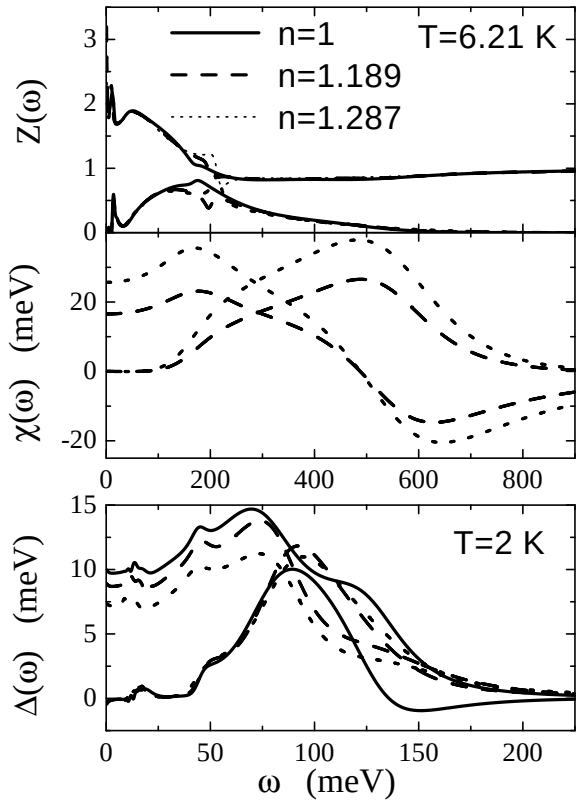

FIG. 3: The real and imaginary part of the functions $Z(\omega)$, $\xi(\omega)$ at $T = 6.21$ K and of $\Delta(\omega)$ at $T = 2$ K for different values of the filling: $n = 1$ (solid line), $n = 1.189$ (dash), and $n = 1.287$ (dot) in the hole-doped case.

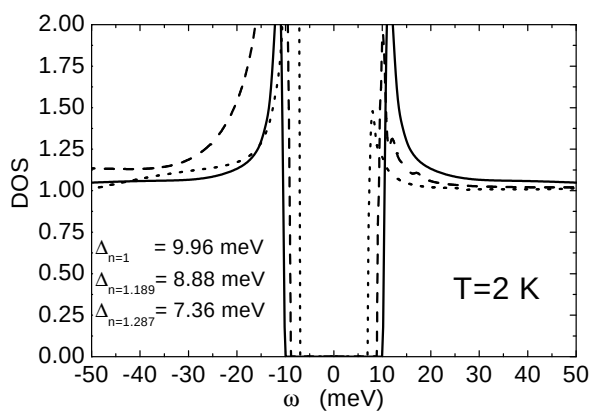

FIG. 4: The normalized DOS, at T=2 K, for different values of the filling: $n = 1$ (solid line), $n = 1.189$ (dash), and $n = 1.287$ (dot) in the hole-doped case.

Very recently J.H. Schön *et al.*[21] have intercalated single crystals of C_{60} with $CHCl_3$ and $CHBr_3$ in order to expand the lattice (see figure 1). The maximum of the critical temperature (onset of resistive transition) in the hole-doped $C_{60}/CHBr_3$ is 117 K. We can try to explain

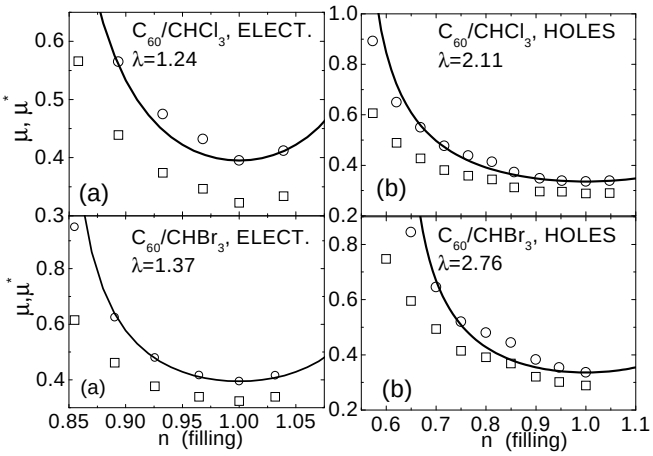

FIG. 5: Coulomb pseudopotential μ (open circle), μ^* (open square) and theoretical fit (solid line) versus filling n: (a) electron-doped case, (b) hole-doped case.

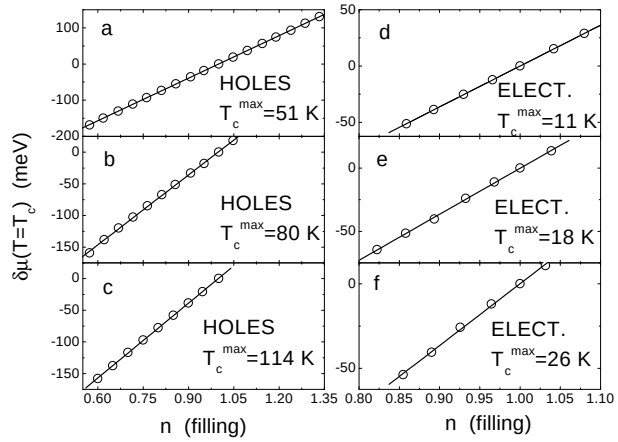

FIG. 6: The chemical potential shift (open circles), at $T = T_c$, calculated from Eliashberg equations and the linear fit (solid line), in the hole-doped cases (a,b,c) and electron-doped cases (d,e,f).

	C_{60}	$C_{60}/CHCl_3$	$C_{60}/CHBr_3$	C_{60}/?
$T_{c,h}^{max}$(K)	51	80	114	135
$T_{c,e}^{max}$(K)	11	18	26	35
λ_h	1.65	2.11	2.76	3.51
λ_e	1.1	1.24	1.37	1.52
Δ_h(meV)	9.9	16.2	25.7	35.5
Δ_e(meV)	1.9	3.2	4.9	6.5
$b_h^{3D}(a = 1.1)$	-4	-5.5	-7.5	-9.6
$b_h^{2D}(a = 1.9)$	-4	-5.5	-7.5	-9.6
$b_h^{2D}(a = 13.8)$	-8	-9.5	-14	-17
$b_e^{3D}(a = 0.77)$	-33	-36	-39	-43
$b_e^{2D}(a = 2.1)$	-43	-46	-49	-53
$b_e^{2D}(a = 10.3)$	-73	-79	-85	-92
d(Å)	14.15	14.28	14.43	14.6

TABLE I: Important quantities for the six cases examined.

these new experimental data by following the approach previously illustrated. We note that the maximum of T_c occurs at the same value of doping in the three hole-doped and electron-doped cases (see figure 1 (a)).

As a first approximation we can assume that the Coulomb pseudopotential is, for $n = 1$, the same as in the bare C_{60}. For simplicity, we also use the electron-phonon spectral function of C_{60} simply multiplied by a constant in order to obtain the correct T_c. These two approximations can be justified by: (i) the presence of an almost constant density of charge carriers[1,21] in the bare and intercalated C_{60}; (ii) the weak dependence of T_c from the details of the actual form of the spectral function[22] (T_c strongly depends on $\omega_{\log}$ and on the total energy range of the spectral function that are likely not to be changed very much by the intercalation). The values of the coupling constant λ necessary to get the new experimental critical temperatures are 2.11 and 2.76 in the hole-doped case (T_c = 80 K and T_c = 114 K, respectively) and 1.24 and 1.37 in the electron-doped case (T_c = 18 K and T_c = 26 K, respectively). In figure 5 the μ and μ^* values calculated by the solution of Eliashberg equations (1)-(4) applied to the $T_c(n)$ data of lattice-expanded C_{60} are shown, as well as the best fit obtained by using eq. 14 and eq. 17. Also now the fitting curves obtained by using the 2D and 3D formula are perfectly superimposed. It is clear that the fits are equally good as those shown in Fig. 2.

Unexpectedly, we find that in all cases the shift of the chemical potential at $T = T_c$, calculated by solving the Eliashberg equations, is a linear function of the filling as we can see in the six panels of figure 6. The reason of this behaviour will be the subject of further investigation.

Figure 7 (a) shows that the calculated coupling constant λ is a linear function of the lattice constant d. We can now extrapolate this behaviour to calculate the coupling constant of a hypothetical material with lattice constant $d \approx 14.6\AA$ (the increase is of 1% with respect to the case where T_c = 117 K)[21]: $\lambda_h = 3.51$ and $\lambda_e = 1.52$. With these λ values and, by using the Eliashberg equations, we determine the corresponding critical temperatures that are 135 K and 35 K, respectively. Finally, figure 7 (b) reports the critical temperature as a function of the coupling constant, for constant μ and $n = 1$, in the hole- and electron-doped case. The critical temperature, in the hole-doped case, does not increase linearly for $\lambda \gtrsim 3$ ($d \gtrsim 14.5\AA$), in disagreement with the theoretical predictions of Ref. 12. On the contrary, T_c drops to zero for $\lambda \lesssim 0.8$ ($d \lesssim 13.9\AA$) as reported in Ref. 20.

In the inset of figure 7 (b) we can observe that also the parameter b is a linear function of the lattice constant d. All the results are finally summarized in table I. In principle, the predictions of Fig. 7(b) and of the last column of Table I concerning T_c and the gap could be experimentally verified in the future. As far as the lattice constant dependence of T_c shown in Table I is concerned, we admit to that the T_c enhancement might be not due

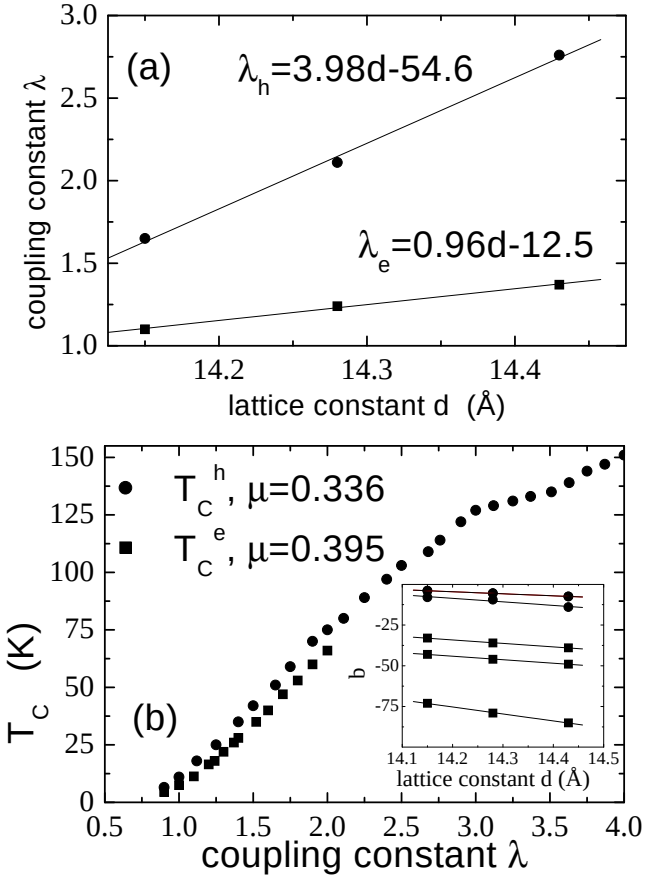

FIG. 7: (a) (a) Coupling constant λ versus lattice constant d in the electron-doped case (full squares), hole-doped case (full circles) and their linear fits (solid lines); (b) T_c versus λ in the electron-doped case (full squares) and hole-doped case (full circles); in the inset, the parameter b is plotted versus d in the electron-doped case (full squares) and hole-doped case (full circles) together with the linear fits (solid lines).

to the increase of the lattice constant but due to the enhanced polarization and the resulting enhanced screening of the Coulomb interaction. Unfortunately, the formalism proposed here is not powerful enough to discriminate between both scenarios.

In conclusion, the experimental results concerning the complete doping dependence of T_c in field-effect hole-doped and electron-doped C_{60} can be naturally explained by the Eliashberg theory generalized to the case where the conduction band is finite, non half-filled and the Coulomb pseudopotential is filling-dependent in a very simple and physically reasonable way. All the values of the physical parameters of the present model appear plausible.

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