Dynamics of vibrational excitation in the C_{60} single-molecule transistor

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We investigate the vibrational excitations recently observed in the single C_{60} molecule transistor [Park *et al.*, Nature (London) **47**, 57 (2000)]. There can be two mechanisms for this: (a) the displacement of the equilibrium position, as the electron hops onto C_{60} to form C_{60}^- and (b) the position dependence of the hopping matrix element. We find that if the two electrodes are planar with the C_{60} sitting symmetrically between the two, then mechanism (a) is not possible, though (b) is, but the results are not in agreement with experiments. Considering C_{60} to be trapped between a protrusion and a flat electrode, both the mechanisms contribute and the contribution from the second can be large. For example, in the case of C_{60} trapped between the a protrusion and a flat electrode, the contribution can be as large as 20%. Though our results do qualitatively explain the results, for quantitative agreement with experiments, it seems necessary to consider perhaps the nonuniformity of the charge distribution in C_{60}^- caused by the image interaction or more complex electrode geometries.

I. INTRODUCTION

In a very interesting experiment, Park *et al.*[1] reported a three electrode transistor made using a single C_{60} molecule (see Fig. 1). As in ordinary silicon field-effect transistors, the voltage on a ''gate'' electrode controls the current flowing from the ''source'' electrode through the C_{60} molecule to the ''drain'' electrode. The source and drain electrodes are believed to be at about a separation of 1 nm. The size of C_{60} and consequent coulombic interaction, prevent the formation C_{60}^{2-}. Experiment shows that nanomechanical oscillations of C_{60} trapped between the two electrodes can be excited by passage of electrons through the system. There have been few attempts to model the process.[2–4] Attention has been paid to electronic structure[3] and also to the center-of-mass motion.[4] However, there are several things that are not clear, as discussed below. The nature of the potential for center of mass motion is not known, owing to a lack of detailed knowledge of the electrode geometry near the molecule, so quantitative support is still lacking for the theoretical approach.[4] The experimental and theoretical work lead to the conclusion that the formation of C_{60}^- results in a shift of the equilibrium position by about 3–4 pm. It was suggested that this shift arises due to the image interaction, though the details of the geometry that would lead to such a shift was not discussed.[4] In this paper, we elaborate on the work of Refs. 1,4 and investigate: (i) How much is the shift in equilibrium position of C_{60} when one electron is transferred to it to form C_{60}^-? (ii) Are there any other contributing factors to the probability of vibrational excitation? Unfortunately, there is no experimental information available on the way C_{60} is trapped between the two electrodes. If the two electrodes are planar (see Fig. 2), and if C_{60} is sitting symmetrically between the two, then the formation of C_{60}^- cannot lead to a shift in the equilibrium position. Even in the symmetric situation, the distance dependence of the hopping matrix element can lead to the excitation of ''center of mass'' oscillations, a mechanism that has not been considered earlier.[1–4] We make rough estimates for this and find that this is equivalent to having a displacement of roughly 0.165 pm in the equilibrium position, and this is not at all enough to explain the experimental observations. Therefore, we consider a situation where one of the electrodes has a hemispherical protrusion as a model for nonplanar electrodes (see Fig. 2). We find that if C_{60} is trapped between a hemispherical protrusion of radius 3.5 Å (roughly the radius of C_{60}), then on forming C_{60}^- a shift in equilibrium position of about 1.7 pm results. This can explain the experimental results qualitatively.

II. THE MODEL

We consider the simplest possible model, which describes the physics of the problem. C_{60} molecule sits in the combined potential of the two electrodes. Adding an extra electron to C_{60} can change C_{60}-metal equilibrium distance due to the image interaction. When C_{60}^- ion gives the extra electron to the drain electrode, the former equilibrium position is regained and the molecule may be left in a ''center of mass'' oscillation excited state. This is reminiscent of two photon processes encountered in light-matter interactions (for example, in resonance Raman scattering). Here we derive a Kramers-Heisenberg-Dirac type formula[5,6] for current. We consider a simple Hamiltonian, which contains all the physically relevant interactions. It is

$$H = H_0 + H', \tag{1}$$

where

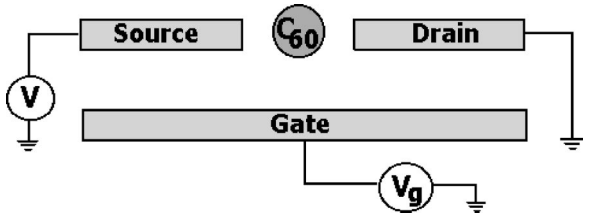

FIG. 1. Diagram of a single C_{60} transistor.

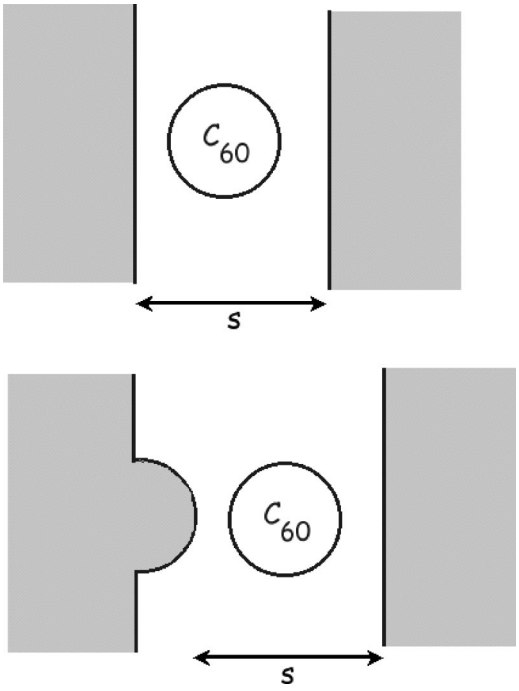

FIG. 2. Two models for C_{60} trapped between two electrodes.

$$H_0=\frac{p^2}{2m}+V(x)+\sum_k\ (\varepsilon_{k_L}n_{k_L}+\varepsilon_{k_R}n_{k_R})$$
$$+[\varepsilon_a-V_Ge+V_{im}(x)]n_a. \qquad (2)$$

x denotes the position of the "center of mass" of C_{60} with respect to the left-hand side electrode and p is the momentum operator corresponding to this motion. $V(x)$ is the potential energy for center-of-mass motion of C_{60}. We do not include any other vibrational mode of C_{60} in our Hamiltonian as there is no evidence of them being excited in the electron transfer process. We have assumed the source and drain electrodes to be described by a one-electron Hamiltonian. In H_0, ε_{k_L} is the energy of k_L-th one-electron state in the source (left-hand side) electrode, ε_{k_R} is the energy of k_Rth one-electron state in the drain (right-hand side) electrode. $c_i(c_i^+)$ is the annihilation (creation) operator for the one-electron state $|i\rangle$. n_i denotes the corresponding occupation number operator and is equal to $c_i^+c_i$. $|a\rangle$ is the lowest unoccupied molecular orbital on C_{60}, which can accept one electron. $-V_Ge$ is the shift in the energy of the orbital due to the gate voltage. $V_{im}(x)$ is the potential that C_{60}^- would experience, due to the images in the metal electrodes. The gate voltage and the image interaction would shift the energy of orbital $|a\rangle$ by $-V_Ge+V_{im}(x)$. The hopping of electrons to and from C_{60} is described by the interaction term

$$H'=\sum_k\ [V_{ak_L}(x)c_a^+c_{k_L}+V_{ak_L}^*(x)c_{k_L}^+c_a]$$
$$+\sum_k\ [V_{ak_R}(x)c_a^+c_{k_R}+V_{ak_R}^*(x)c_{k_R}^+c_a]. \qquad (3)$$

$V_{ak_L}(x)[V_{ak_R}(x)]$ is the matrix element responsible for electron transfer from $|k_L\rangle$ ($|k_R\rangle$) to orbital $|a\rangle$ on C_{60}. Written more explicitly, $V_{ak_L}(x)=\langle a|H_{\text{electronic}}|k_L\rangle$, where $H_{\text{electronic}}$ is the electronic part of the Hamiltonian. The state $|k_L\rangle$ ($|k_R\rangle$) decays exponentially in vacuum, as a result of which, the matrix element would depend upon the distance of C_{60} from the surfaces of the electrodes. As a result, these matrix elements are x dependent. Treating H' as a perturbation, we can use second-order perturbation theory, to derive an expression for the current. We follow an approach appropriate for resonant tunneling through the orbital $|a\rangle$. Assuming this orbital to have an inverse lifetime $\Gamma\hbar$, we get a Kramers-Heisenberg-Dirac type formula for the current

$$I=\sum_F\frac{2e\pi}{\hbar^2}\left|\sum_L\frac{\langle F|H'|L\rangle\langle L|H'|I\rangle}{E_L-E_I-i\Gamma\hbar/2}\right|^2\delta(E_F-E_I). \qquad (4)$$

$|I\rangle$ is the initial state (of all the electrons in the two electrodes and of the center-of-mass motion of C_{60}), $|L\rangle$ is the intermediate state, in which one electron from the left-hand electrode has jumped on to C_{60}, and $|F\rangle$ is the final state, in which the electron has jumped onto the right-hand electrode, leaving C_{60}, perhaps in an excited center-of-mass state. E_F, E_I, and E_L denote the energies of these states. We ignore the temperature effects (the experiment was done at 1.5 K, which justifies this). We assume that $|I\rangle$, $|L\rangle$, and $|F\rangle$ may be written as products of appropriate electronic parts and center-of-mass oscillation parts as $|I\rangle=|G\rangle\,|\psi_i\rangle$, $|F\rangle=c_{k_R}^+c_{k_L}|G\rangle|\psi_f\rangle$, and $|L\rangle=c_a^+c_{k_L}|G\rangle|\tilde{\psi}_l\rangle$, where $|G\rangle$ denotes the ground state of the two electrodes, with all the levels occupied, up to their respective Fermi levels $\varepsilon_{F,L}$ and $\varepsilon_{F,R}$. Obviously, $|k_L\rangle$ must be an occupied orbital in $|G\rangle$ and $|k_R\rangle$ must be unoccupied. $|\psi_i\rangle$, $|\psi_f\rangle$ are the initial and final wave functions for center-of-mass motion of C_{60}, having energies $\varepsilon_{\text{vib}}^i$ and $\varepsilon_{\text{vib}}^f$ respectively. $|\tilde{\psi}_l\rangle$ denotes the wave function for the l-th center-of-mass state of C_{60}^- which is the intermediate state in the transfer. Its energy is denoted by $\tilde{\varepsilon}_{\text{vib}}^l$. Thus $(E_F-E_I)=\Delta\varepsilon_{\text{vib}}^{fi}+\varepsilon_{k_R}-\varepsilon_{k_L}$ and $E_L-E_I=\varepsilon_{a,\text{eff}}+\tilde{\varepsilon}_{\text{vib}}^l-\varepsilon_{\text{vib}}^i-\varepsilon_{k_L}$, with $\varepsilon_{a,\text{eff}}=\varepsilon_a+V_Ge+V_{im}(x_{\text{eq}})$. (We neglect the position dependence of $\varepsilon_{a,\text{eff}}$ and evaluate it at x_{eq}, which denotes the equilibrium position of C_{60}.) $\Delta\varepsilon_{\text{vib}}^{fi}$ is the energy difference between initial and final vibrational states, and is equal to $\varepsilon_{\text{vib}}^f-\varepsilon_{\text{vib}}^i$. Using all these, we get

$$I=\sum_f\sum_{k_R(\varepsilon_{k_R}>\varepsilon_{F,R})}\sum_{k_L(\varepsilon_{k_L}<\varepsilon_{F,L})}\frac{2e\pi}{\hbar^2}\left|\sum_l\frac{\langle\psi_f|V_{ak_R}|\tilde{\psi}_l\rangle\langle\tilde{\psi}_l|V_{ak_L}|\psi_i\rangle}{(\varepsilon_{a,\text{eff}}+\tilde{\varepsilon}_{\text{vib}}^l-\varepsilon_{\text{vib}}^i-\varepsilon_{k_L}-i\Gamma\hbar/2)}\right|^2\delta(\varepsilon_{k_R}-\varepsilon_{k_L}+\Delta\varepsilon_{\text{vib}}^{fi}). \qquad (5)$$

In order to simplify the above equation, we assume that the matrix elements V_{ak_R} (V_{ak_L}) have the same dependence on the distance from the electrodes. That is, $V_{ak_R}(x)=g^R_{k_R}f(s-x)$ and $V_{ak_L}(x)=g^L_{k_L}f(x)$, where s is the separation between the electrodes (see Fig. 2). $f(x)$ determines how the matrix elements decrease with separation from the electrodes. We now define $\rho^R(\varepsilon)=\Sigma_{k_R}|g^R_{k_R}(\varepsilon_{k_R})|^2\delta(\varepsilon-\varepsilon_{k_R})$ and $\rho^L(\varepsilon)=\Sigma_{k_L}|g^L_{k_L}(\varepsilon_{k_L})|^2\delta(\varepsilon-\varepsilon_{k_L})$. $\rho^R(\varepsilon)[\rho^L(\varepsilon)]$ is a ''density of states'' for the right (left) hand electrode, that determines its ability to give (take away) electrons to C_{60}. Further, as the range of ε values is rather small we neglect the energy dependence of these density of states, and get

$$I=\frac{2e\pi}{\hbar^2}|\rho^R|^2|\rho^L\Bigg|^2\sum_f\int_{\varepsilon_{F,R}+\Delta\varepsilon^{fi}_{\rm vib}}^{\varepsilon_{F,L}}d\varepsilon\Bigg|\times\Bigg|\sum_l\frac{\langle\psi_f|f(x)|\tilde{\psi}_l\rangle\langle\tilde{\psi}_l|f(s-x)|\psi_i\rangle}{(\varepsilon_{a,\rm eff}+\tilde{\varepsilon}^l_{\rm vib}-\varepsilon^i_{\rm vib}-\varepsilon-i\Gamma\hbar/2)}\Bigg|^2. \quad (6)$$

We use this ''sum over state'' expression for further analysis. For convenience, we take $\varepsilon_{F,R}$ as our zero of energy and denote $\varepsilon_{F,L}$ as eV_b, where V_b is the magnitude of the bias voltage. We use a simple approach to calculate $f(x)$, which determines the position dependence of the hopping matrix element. As C_{60} moves away from an electrode, the matrix element for electron hopping would decrease. Since the potential difference between the electrodes is only a few meV, the electron has to come from a state whose energy is close to the Fermi level. So it is sensible to neglect the energy dependence of the matrix element and to approximate its distance dependence by the decay of the wave function for an electron at the Fermi energy. We therefore make the natural assumption that the matrix element has the same dependence as the wave function for an electron at the Fermi level of gold, when it extends out into the vacuum. Further, we estimate this dependence using a step function model for the surface. The step height is determined by the work function of gold, which is 5.2 eV. The result is that $f(x)=Ae^{-ax}$, where $a=1.1664/\text{Å}$. The pre-exponential factor A can be absorbed into the definition of $g^R_{k_R}$ or $g^L_{k_L}$. This would lead only to a modification of the product $|\rho^R|^2|\rho^L|^2$, in Eq. (6). In the following, we do not try to reproduce the magnitude of the current, but only the ratios of steps in the current (see below), and consequently, we do not need the value of the product $|\rho^R|^2|\rho^L|^2$. $\Gamma\hbar$ is the inverse lifetime of the extra electron in C^-_{60} and is due to hopping to the two electrodes, which we have taken to be 0.1 μeV[4].

III. THE INTERACTION BETWEEN C_{60} AND GOLD ELECTRODES

Now we calculate the interaction potential between C_{60} and surface of Au metal. We consider the case that is shown in Fig. 2. For this, we make use of method already available in the literature.[7] We take the interaction between a C atom in C_{60} and a gold atom in the metal to be given by the Buckingham potential[8]

$$U(x)=A\exp(Bx)-\frac{C}{x^6}\quad\text{for}\quad x>1.3\ \text{Å},$$

$$U(x)=\frac{D}{x^2}\quad\text{for}\quad x<1.3\ \text{Å}$$

with

$$A=138341.28\ \text{kJ mol}^{-1},\quad B=-3.034\ \text{Å}^{-1},$$

$$C=5249.244\ \text{Å}^{-6}\ \text{kJ mol}^{-1},$$

and

$$D=4288.68\ \text{Å}^{-2}\ \text{kJ mol}^{-18},$$

where x is the distance between the two atoms. We model C_{60} as a hollow sphere of radius b, with carbon atoms smeared into a continuum of density $\sigma=N/(4\pi b^2)$. Here N is the number of carbon atoms in C_{60}. Further, the semi-infinite metal also is assumed to form a continuum, with density appropriate to the bulk.[9] This kind of approach has been successfully used in a variety of problems like thermal expansion of C_{60},[10] cohesive and anharmonic properties of solid C_{70}, interaction of C_{60} with graphite surface,[7] and a variety of other problems.[11–14]

Denoting the distance of center of C_{60} from the metal surface as R, we get the interaction potential to be

$$V(R)=\frac{CN\ \pi\ R\ \rho}{6\ (b-R)^2\ (b+R)^2}+\frac{\rho Ae^{B(-b+R)}N\pi[bB(1+e^{2\ bB})+(-1+e^{2\ bB})(-3+BR)]}{bB^4} \quad (7)$$

ρ is the number density of the metal. Using Eq. (7), we have calculated the binding energy as well as the frequency of the center-of-mass oscillation of the molecule. We find the binding energy to be 0.782 eV at an equilibrium distance of 5.911 Å. The frequency of vibration is found to be $7.02\times10^{11}\,\text{sec}^{-1}$. Also, using the potential of the Eq. (7), we can find the best distance between the two electrodes, which will lead to the maximum binding energy for a C_{60} molecule trapped in between. It is 11.823 Å and this distance leads to a binding energy of 1.56 eV. Results of our calculation indicate that ''C_{60}-gold'' binding near the equilibrium position can be approximated very well by a harmonic potential with a force constant of $k=46.491\ \text{kg sec}^{-2}$. This force constant and the mass of the C_{60} molecule yield a center-of-mass oscillation frequency of $9.925\times10^{11}\,\text{sec}^{-1}$ and a center-of-mass oscillation quantum of 4.10 meV, which is close to the

experimental result (5 meV).

For C_{60}^-, the interaction potential has an additional contribution from the image interaction. To calculate this, we use simple electrostatics[15] and put the negative charge at the center of C_{60}^-. Our calculations show that the force constant for vibration is affected only slightly, by the image interaction (the new value is $k=46.488$ kg sec^{-2} and the frequency is changed only in the fourth decimal place). As this change is very small, we neglect it. In this and in all the calculations reported in the paper, for the sake of clarity in the plots, we take the gate voltage to be such that the acceptor orbital on the fullerene is above the Fermi level of the electrodes by $h\nu/2$ when there is no potential difference applied between the donor and acceptor electrodes. Here ν is the frequency of vibration of C_{60} in the potential well due to the electrodes. This value is arbitrary as changing the value of the gate voltage can be used to move this orbital up or down in energy and the only reason for taking this value is to show the steps in the $I-V$ plots clearly. As the electrodes are located symmetrically, transferring an electron to form C_{60}^- would not lead to any change in its equilibrium position. Still, because of position dependence of the hopping matrix element, there can be net vibrational excitation. We have done a calculation for this case and results are given in Table I. For a fullerene molecule, which is in the ground vibrational level of the center of mass oscillation, the passage of the electron may put it in the v-th vibrational level. We refer to this as the 0-v channel. We find that the current step due to 0-1 channel is only 1/800th of the current due to 0-0 channel and this is far too small to explain the steps seen in the experiment. Therefore we consider a model in which we account for nonflatness of the surface. Thus we take C_{60} molecule to be sitting between a planar electrode and an electrode with a surface protrusion as shown in Fig. 2. The surface protrusion is taken to be a hemisphere of radius r. In principle, the hopping matrix element would depend on the curvature of the protrusion, but as a first approximation, we have neglected this. We now calculate the van der Waals interaction potential between C_{60} and the protruded electrode, and obtain

TABLE I. Ratio of current steps.

	Ratio of $(n+1)$-th step to nth step		
	Without protrusion	With protrusion	
n	$a=1.16636$ per Å	$a=1.16636$ per Å	$a=0$
0	0.0012	0.0953	0.0776
1	0.0023	0.0478	0.0390
2	0.3204	0.0322	0.0255
3	0.5555	0.0281	0.0272

$$
\begin{aligned}
V(R)=&\frac{CN\ \pi\ R\ \rho}{6(b-R)^2\ (b+R)^2}+\frac{Ae^{B(-b+R)}N\pi[bB\ (1+e^{2\ bB})+(-1+e^{2\ bB})(-3+BR)]\rho}{bB^4}\\
&+\frac{Ae^{B(-b+\sqrt{r^2+R^2})}N\ \pi[bB\ (1+e^{2\ bB})(-1+B\ \sqrt{r^2+R^2})]\rho}{bB^5\ R}\\
&+\frac{AN\ \pi\{-[r^2\ B^2\ (-1+e^{2\ bB})]+bB\ (1+e^{2\ bB})+(-1+e^{2\ bB})(-4+BR)\}\rho}{bB^5\ e^{B(r+b-R)}R}\\
&+\frac{r^3\ CN\ \pi\ [-(r^5\ R)+4\ r^4\ R^2+4\ r^3\ R(b^2-2\ R^2)+4\ (b^2-R^2)^2\ (b^2+R^2)]\rho}{6\ (b^2-R^2)^2\ (r^2-b^2+R^2)^2\ (r^2-b^2-2\ rR+R^2)^2}\\
&+\frac{r^3\ CN\ \pi\ [rR\ (b^4+6\ b^2\ R^2-7\ R^4)-4\ r^2\ (b^4+b^2\ R^2-2\ R^4)]\rho}{6\ (b^2-R^2)^2\ (r^2-b^2+R^2)^2\ (r^2-b^2-2\ rR+R^2)^2}\\
&-\frac{Ae^{B(-b+R)}N\ \pi\ [bB\ (1+e^{2\ bB})+(-1+e^{2\ bB})(-4+BR)]\rho}{bB^5\ R}\\
&-\frac{Ae^{B(-b+R)}N\ \pi\ [(-1+e^{2\ bB})(-2+BR)^2+bB\ (1+e^{2\ bB})(-1+BR)]\rho}{bB^5\ R}\\
&+\frac{AN\ \pi\{rB\ [bB\ (1+e^{2\ bB})+(-1+e^{2\ bB})(-4+BR)]\}\rho}{bB^5\ e^{B(r+b-R)}R}\\
&+\frac{Ae^{B(-b+\sqrt{r^2+R^2})}N\ \pi[-(-1+e^{2\ bB})(-4-r^2\ B^2-B^2\ R^2+4\ B\ \sqrt{r^2+R^2})]\rho}{bB^5\ R}.
\end{aligned}
$$

This expression is valid only if C_{60} is allowed to move along a line perpendicular to the flat portion of the surface and passing through the hemisphere. $R=Z-r$, where Z is the distance of the center of C_{60} from the center of the hemisphere. In further calculations we use only $r=3.5$ Å. The reason for taking this radius, which is the same as that of C_{60} is as follows: For a larger radius, the effect of the protrusion will be less, while for a smaller radius, the C_{60} would not be stable on top of the protrusion. Thus, the protrusion that we have chosen would have maximum effect on the process, but at the same time, C_{60} would be able to sit on top of it. With this protrusion, we find a binding energy of 0.267 eV and an equilibrium distance of 9.430 Å between the C_{60} center and the center of the hemisphere. The frequency of vibration is $4.08\times10^{11}\,\mathrm{sec}^{-1}$. Now, using this potential, we can find the best distance between this protruded electrode and a planar electrode, which will lead to maximum binding energy for a C_{60} molecule trapped in between. We find it (s as shown in Fig. 2) to be 11.841 Å with a binding energy of 1.05eV. The equilibrium distance is 5.911 Å between the C_{60} center and the planar electrode. Results of our calculations indicate that C_{60}-gold binding near the equilibrium position can be approximated very well by a harmonic potential with a force constant of $k=31.1136\ \mathrm{kg\,sec}^{-2}$. This force constant and the mass of the C_{60} molecule yield a vibration frequency of $8.1196\times10^{11}\,\mathrm{sec}^{-1}$ and vibrational quantum of 3.36 meV. For C_{60}^{-}, the interaction potential has contribution from the image interaction, which is calculated as follows.

To calculate this contribution, we use simple electrostatics and put a negative charge at the center of C_{60}^{-}. It has not been possible to obtain a closed form for the image potential. Hence we adopted a simple procedure, in which we generated images on a computer and calculated the forces due to them. If one had only one electrode with the hemispherical protrusion, and if one places a charge $-e$ at a distance Z from the center of the hemisphere, then three images, located at positions $(-Z,r^2/Z,-r^2/Z)$ having charges $(e,-eR/Z,eR/Z)$ are required, to make the surface of the electrode an equipotential. These images may be thought of as arising from the plane of the electrode (the image that has the charge e) and from a sphere of radius r whose center coincides with that of the hemisphere [these images have the charges $(-eR/Z,eR/Z)$]. Thus one now has four charges (one original and three images). If one more planar electrode is added, then images of these four charges in it. Then there would be images of the images and so on, *ad infinitum*. We generated these images successively and calculated the forces due to them. As we are calculating the force, this leads to a convergent series due to three reason.

(1) Each reflection on the sphere, reduces the image charge by a factor r/r_d, where r_d is the distance of the charge from the center of the sphere. This leads to a reduction in the magnitude of charges roughly by at least a factor of 1/3 with each reflection on the sphere.

(2) With each reflection, the images change sign and hence, forces from the images tend to cancel each other.

(3) Further, images generated by repeated reflections between the parallel electrodes would be placed at larger and larger distances. The contributions of these to the force decrease rapidly, because of the inverse square law.

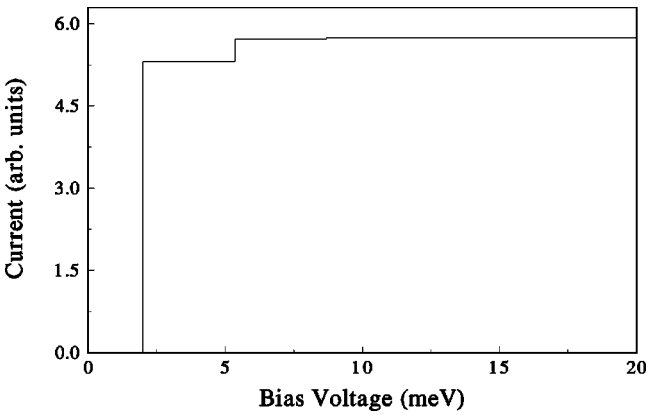

FIG. 3. Current vs voltage plot for a single C_{60} transistor at $T=1.5$ K. in the case where C_{60} molecule is sitting between a planar electrode and an electrode with spherical surface imperfection. We have put $a=0$.

We took 32 760 images, which resulted from a set of six successive reflections between the two electrodes and the answer was checked for convergence by comparing it with results from five and seven successive reflections. The answer was found to be accurate to within six decimal places. Our numerical calculations show that the force constant for vibration is changed only slightly, due to the image interaction. The additional electron on C_{60} results in a shortening of the C_{60}-planar surface equilibrium distance by 1.705 pm but it does not significantly change the vibration frequency. The C_{60}^{-}-gold binding near this new equilibrium position can be approximated very well by a harmonic potential with a force constant of $k=31.1136\ \mathrm{kg\,sec}^{-2}$. The results for the current steps are shown in Table I. In Fig. 4 we show the $I-V$ characteristics in this case. If one compares Fig. 3 and 4, then we find that the step heights are more in Fig. 4. In Fig. 3 only one step is visible at this scale of plotting while in Fig. 4 a second step is barely visible. Therefore, we have presented the numerical values in Table I. If we neglect the dependence

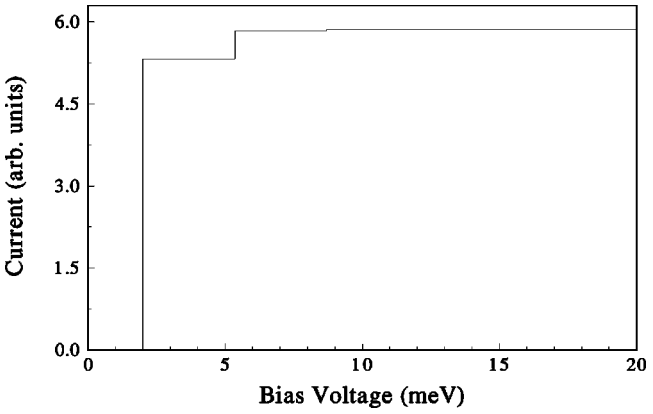

FIG. 4. Current vs voltage plot for a single C_{60} transistor at $T=1.5$ K in the case where C_{60} molecule is sitting between a planar electrode and an electrode with spherical surface imperfection. We have taken $a=1.1664$ per Å.

of hopping matrix element by putting $a=0$, then we find that the current steps Fig. 3 are significantly altered.

Our results indicate that the distance dependence of the hopping matrix element can contribute significantly (up to 20%) to the observed current steps. Even though we have taken a protrusion such that it has the maximum possible effect, the results are only in qualitative agreement with the experiment, the explanation of which requires a displacement of equilibrium position by about 3–4 pm. This can be due to two reasons. (1) In the presence of image interaction, the negative charge on C_{60}^- would not be distributed uniformly. It is energetically more favorable for the charge to concentrate in the vicinity of the electrodes. Even in the case of planar electrodes, as one displaces C_{60}^- from the equilibrium position of C_{60}, charge would move towards the the nearer electrode, resulting in perhaps a double well type of potential energy curve. This effect is not included in our analysis. This is an attractive possibility[16] that needs further investigation. If this were the mechanism, then it is likely that internal modes of the C_{60} can also be excited in the process. The lowest such mode (sphere to ellipsoid oscillations of C_{60}) is around 33 meV and the probability of the excitation would depend on the actual displacement of this mode from its equilibrium value, when C_{60}^- is formed. In the experiments, one needs to look in this region carefully, for this excitation. (2) The electrode geometry is more complex than what we have considered. Further investigations are required on these two aspects.

IV. CONCLUSIONS

We have investigated theoretically the excitation of center-of-mass oscillations in the single C_{60} transistor. In this, there can be two possible mechanisms for vibrational excitation. The first is the displacement of the equilibrium position of C_{60} when an electron is transferred to it and the second is the position dependence of hopping matrix element responsible for the electron transfer. If C_{60} is trapped between two planar electrodes, then, hopping of an electron onto it does not change the equilibrium position. Still, due to the position dependence of hopping matrix element, vibrational excitation can occur. Using the decay of the wave function at the Fermi level to model the decay of matrix element, we find that this effect is roughly equivalent to displacing the equilibrium position of the oscillator by 0.165 pm. However, to reproduce the experimental results, the displacement of the equilibrium position has to be about 3–4 pm. Within a flat trapping geometry for both the electrodes, this does not seem to be possible. Therefore, we investigated a geometry in which the C_{60} is trapped between a planar electrode and one with a surface protrusion. In this case, the formation of C_{60}^- does lead to a change in the equilibrium position. If the protrusion is modeled as a hemisphere, then this change is roughly 1.7 pm. In this case, we find that the distance dependence of hopping matrix element contributes significantly to the observed step heights (maximum being 20%). However, though the results are in qualitative agreement with experiment, quantitative agreement is not good. This may be due to the nonuniformity of charge distribution in C_{60}^- caused by the image interaction or perhaps the electrode geometry may be more complex than what we have envisioned.

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