Phonon assisted dynamical Coulomb blockade in a thin suspended graphite sheet.

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The differential conductance in a suspended few layered graphene sample is found to exhibit a series of quasi-periodic sharp dips as a function of bias at low temperature. We show that they can be understood within a simple model of dynamical Coulomb blockade where energy exchanges take place between the charge carriers transmitted through the sample and a dissipative electromagnetic environment with a resonant phonon mode strongly coupled to the electrons.

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One of the great challenges of molecular electronics is to access electron-phonon coupling at the single molecule level. Mechanically tunable atomic break junctions with trapped small molecules such as $(H_2, D_2, H_2O)$ have been shown to exhibit a spectroscopic signature of their characteristic phonon modes $[1]$. The signature of phonons is also spectacular in the Coulomb blockade regime for a molecular single electron transistor: The typical resonant tunneling peaks as a function of gate or source-drain voltage are surrounded by satellites, which correspond to the emission or absorption of one or several phonons. Specific vibrational modes were identified in this way in fullerenes and suspended carbon nanotubes $[2, 3, 4]$. Theoretical models $[5, 6, 7]$ were developed to describe these characteristic phonon modes $[1]$. The signature of phonons shown to exhibit a spectroscopic signature of their characteristic phonon modes $[1]$.

The graphite foil sample itself constitutes the electromagnetic environment. More original, on the thinnest sample (with 30 graphene layers) a series of periodic dips was observed at high bias (above 0.15 $V$, see fig$[1]$) can be related to the linear dependence of the density of states $\nu(E)$, characteristic of the band structure of graphene as well as of graphite at high enough energy $[11]$. Indeed, the electronic transmission between the graphite sample and the underlying electrodes is low, so that the voltage drop occurs mainly at the contacts. The differential conductance can then be written as:
$dI/dV \propto \Gamma_L \Gamma_R (\alpha \nu (EF + \alpha eV) + (1 - \alpha) \nu (EF - (1 - \alpha) eV))$

(1)

where $\Gamma_L$ and $\Gamma_R$ are the transmissions of the left and right contacts respectively. The parameter $1/2 \leq \alpha \leq 1$ characterizes the asymmetry of the contact resistances and voltage drop, $\alpha = 1/2$ corresponds to symmetrical contacts with $V/2$ voltage drop at each contact. The asymmetry observed between positive and negative bias is attributed to a combination of a slight doping of the sample together with some asymmetry in the transmission of the electrodes.

We now focus on the conductance at low voltage (below 0.12 V) which exhibits a pronounced dip at zero bias. This behavior is characteristic of Coulomb blockade through a small capacitance tunnel junction in series with a dissipative electromagnetic environment which can exchange energy with the tunneling quasi particles on a scale much smaller than the charging energy. This yields the so called Dynamical Coulomb Blockade (DCB) [9]. The differential conductance data is expected to follow a scaling behavior as a function of bias and temperature:

$$G(V) = dI/dV = T^z f(eV/k_B T)$$

(2)

with $\lim_{x \to 0} f(x) = Cst$ and $\lim_{x \to \infty} f(x) = x^z$, and the exponent $z$ is expected to be $\alpha^2 R/R_Q$ in the case of two asymmetric junctions, where $R$ is the resistance of the environment and $R_Q = h/2e^2$ is the resistance quantum. The data shown in fig. 2 yields $z$ of the order of 0.25 ± 0.05. Such a power law dependence also was found in the two other thicker samples, with similar exponents. It is thus reasonable to assume that the dissipative ohmic environment in the present case is constituted by the top graphene layers of the graphite samples. Note also that a similar behavior was already observed on multilayer carbon nanotubes with low conductance contacts [12].

More original, as shown on fig. 3, is the bias dependence of the differential conductance measured on the thinnest sample investigated which is 10 nm thick and contains thirty graphene layers. It exhibits a series of eight sharp dips resembling the zero bias one and nearly equally spaced by $20 \pm 2$ mV. Their amplitude decreases with increasing voltage except for the broader dip at 50 mV which can be decomposed into 2 overlapping negative peaks centered around 40 and 60 mV as suggested by the data taken at 1 nA ac excitation. The energy scale of 20 mV does not correspond to any simple electronic energy scale in the sample, whose charging energy is in the meV range and level spacing in the 10 μeV range. On the other hand the lowest energy optical phonon in graphite (so-called Z0') has an energy of 15 meV [8]. This mode, which emerges from the out of plane transverse acoustic mode of graphene [8, 12], is only present in graphite and corresponds to the two neighboring, non equivalent, planes vibrating in phase opposition along the c axis.

This phonon mode has been observed experimentally in graphite by inelastic X-ray scattering [13] and scanning tunneling spectroscopy [14] with an energy of 15 ± 1 meV. The observed peak positions at multiple values of 20 mV instead of 15 mV can be attributed to the asymmetry of the contacts which corresponds to the parameter $\alpha \simeq 0.75$ in eq. [11]. These dips are only observed on the 10 nm thick foil and were not detected on the two thicker...
(more than 100 nm) samples. This can be understood considering that the conversion from electric energy (depending only on the resistance of the tunneling barriers) into mechanical vibrations leads to an induced vibration amplitude inversely proportional to the number of layers in the graphite foil. The suspended character of the sample is also essential, since interaction with a substrate suppresses considerably the amplitude of induced vibrations as already demonstrated on carbon nanotubes [3]. Note that STM spectroscopy on bulk graphite [14] also reveals inelastic contributions due to plasmons which are not detected in the present experiment.

In order to explain the data more quantitatively we extended the work of Mitra et al. [7] on the phonon assisted Coulomb staircase observed in the transport through fullerene molecules. The coupling between the ZO'-phonon mode and the electrons in the graphite sample is described using a Holstein Hamiltonian [15]. In the absence of disorder this Hamiltonian $H_G$ reads:

$$H_G = \sum_k \epsilon_k c_k^\dagger c_k + \lambda \hbar \omega \sum_k c_k^\dagger c_k (b^+ + b) + \hbar \omega b^+ b$$  (3)

where $c_k^\dagger$ and $c_k$ are the fermionic electron creation and annihilation operators in momentum space, $\epsilon_k$ the electronic energy, and $b^+ + b$ the creation and annihilation operators of the bosonic ZO'-phonon of frequency $\omega$. In contrast with previous work [7], the electronic energy level spacing is small compared to the phonon energy $\hbar \omega$. The parameter $\lambda$ is the dimensionless electron phonon coupling constant which we assume to be of the order of unity like in carbon nanotubes [16]. The coupling to the leads is then described in a dynamical Coulomb-blockade formalism [2], by a Hamiltonian of the form $H_T = \sum_{k,k'} T_{k,k'} a_k^\dagger c_{k'}^\dagger e^{-i\phi} + h.c$. Here $a_k$ are the electron annihilation operators in the leads, $T_{k,k'}$ are the tunnel amplitudes and $\phi$ is a phase operator describing the electromagnetic environment of the junction.

The Hamiltonian $H_G$ can be diagonalized with a canonical Lang-Firsov transformation: $b^' = e^{-S} b e^S$, $H_G = e^{-S} H_G e^S$ where $S = \lambda \sum_k \epsilon_k^2 c_k^\dagger c_k (b + b^')$. In the limit when the charging energy of the sample is negligible [7] the transformed Hamiltonian reads simply $H_G = \sum_k \epsilon_k c_k^\dagger c_k + \hbar \omega b^+ b$ where we have omitted the primes for the transformed operators. In the transformed basis the transfer Hamiltonian $H_T$ is given by:

$$H_T = \sum_{k,k'} T_{k,k'} a_k^\dagger c_{k'}^\dagger e^{-i\phi + \lambda (b^+ b)} + h.c$$  (4)

This expression is obtained by expanding the product $e^{-S} H_T e^S$ under the assumption that the environment phase $\phi$ commutes with the phonon operators. It shows that the coupling to phonons essentially changes the phase operator of the junction. As a result the current through the junction can be expressed with an effective $P(E)$ function describing the probability of electrons to loose an energy $E$ in a tunnel transition as in usual DCB theory. Since the electromagnetic environment and phonon operators commute, this function can be expressed as a convolution

$$P(E) = \int dE' P_{env}(E') P_{ph}(E - E')$$  (5)

where $P_{env}(E)$ is the probability of emitting a photon of energy $E$ in the $RC$ environment of the junction and $P_{ph}$ is the probability of emitting a phonon in the sample. The probability distribution $P_{ph}$ can be obtained by noticing that the corresponding phase operator $i\lambda (b^+ b)$ is analogous to that of an electromagnetic $LC$ circuit with resonant frequency $1/\sqrt{LC} = \omega$ [9] (this result can also be obtained directly by tracing out the phonon degrees of freedom in eq(4):

$$P_{ph}(E) = e^{-\lambda^2 \coth(\Delta \hbar \omega/2)} \sum_k \delta(E - k\hbar \omega) e^{k\hbar \omega/2} I_k(\lambda \hbar \omega/2)$$  (6)

where $\beta$ is the inverse of the phonon temperature $T_{ph}$. Using Eqs. (5) and (6) and standard expressions for current as a function of $P(E)$ [8] it is possible to compute the current through the sample. For example in the case of symmetric contacts.
(\alpha = 1/2) and constant density of states, \(dI/dV = \frac{1}{R_T} \int dE \left( P(eV/2 - E) + P(-eV/2 - E) \right) \) where \(R_T\) is the tunnel resistance of each contact. Note the similarity with the DCB in a tunnel junction in series with a LC resonator [10]. The case of an energy dependent density with the DCB in a tunnel junction in series with a LC resonator \[10\]. The case of an energy dependent density with the DCB in a tunnel junction in series with a LC resonator \[10\].

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\frac{dI}{dV}(E) &= \frac{\lambda}{R_T} \int dE \left( P(eV/2 - E) + P(-eV/2 - E) \right) \end{align*} \]

This formula is exact for bilayer graphene with \(\Delta \approx 400 \text{meV} [17] \) and \(|E| < \Delta\). We also take the values of \(R_T\), environment resistance \(R\) and charging energy deduced from the geometry and conductance data at low bias. The only free adjustable parameter is \(\lambda = 0.7\). As shown on fig[3] the agreement with experimental data is only qualitative especially at zero temperature, where theory predicts zero conductance at zero bias which is not observed experimentally. Better agreement is found when a finite phonon temperature is introduced. Since the sample is suspended the populations of phonons are supposed to be strongly out of equilibrium. We have tentatively introduced a phonon temperature increasing linearly with bias which leads to better agreement with experimental data. However, surprisingly, the best fit is obtained by imposing a bias-independent phonon temperature which does not seem a priori very physical. We have also in our model neglected the electronic temperature which is expected to be in the K range. Moreover we have not included the expected broadening of the phonon modes (even at very low temperature) due to the strong coupling to electrons. This may explain why the best fit corresponds to a finite phonon temperature.

In the following we discuss the magnetic field dependence of the dips in the differential conductance (fig. 4). They vary both in amplitude and position between 0 and 5 T. Whereas the first two dips and the forth one are shifted toward lower frequency with increasing magnetic field (as seen both at positive and negative bias) the third peak is shifted toward high frequency (we consider here the negative bias data since the second and third dips at positive bias can barely be resolved as discussed above). The amplitude of the dips (see inset in fig[4]) may decrease or increase with magnetic field (peak 2 and 3), or vary in a non monotonic way (peak 0 and 1). The relative shifts of the dips with magnetic field by an typical amount of 5 to 20% are of the same order of magnitude as the relative variations of their magnitudes. We attribute these effects to the field dependent density of states of the graphite foil in the field range where Shubnikov de Haas oscillations just start to show up. Even if these observations are not yet understood in detail they indicate a strong electron phonon coupling and justify the value of \(\lambda = 0.7\) since a relationship such as \(\Delta \omega(B) / \omega(B = 0) = \lambda^2 \omega(E_F) \Delta G(B)/G(B = 0)\) between the typical phonon frequency magnetic dependence and magneto conductance, is expected to hold [18]. High electron phonon coupling has already been reported in graphene concerning in plane optical modes [19]. In the present case however, strong electron phonon coupling between transverse \(\text{ZO}'\) vibrations and strongly anisotropic transport in the thin graphite layer is not straight-forward but can be understood if the electrical contacts between the two electrodes and the graphite foil take place through distinct graphene mono-layers which is highly probable.

In conclusion we have shown evidence of differential conductance sharp dips in a suspended thin layer of graphite with 30 graphene foils. These dips can be interpreted within a simple model of dynamical Coulomb blockade with an environment strongly coupled to the lowest energy optical phonon mode \(\text{ZO}'\) of graphite. The magnetic field dependence of the effect corroborates this interpretation.

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