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AC transport in carbon-based devices: challenges and perspectives

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Abstract

Time-dependent fields are a valuable tool to control fundamental quantum phenomena in highly coherent low dimensional electron systems. Carbon nanotubes and graphene are a promising ground for these studies. Here we offer a brief overview of driven electronic transport in carbon-based materials with the main focus on carbon nanotubes. Recent results predicting control of the current and noise in nanotube based Fabry-Pérot devices are highlighted.

Résumé

Les champs dépendant du temps sont des outils précieux pour le contrôle des phénomènes quantiques dans les systèmes d'électrons fortement cohérents è base dimentionalitée. Les nanotubes de carbone et le graphène constituent des objets de base prometteurs pour ces études. Ici, nous proposons une brève synthèse des phénomènes de transport électronique induits dans les matériaux carbonés, en se focalisant sur les nanotubes de carbone. Nous mettons en avant des résultats récents prédisant le contrôle du courant et du bruit dans les résonateurs Fabry-Pérot à base de nanotubes.

I. INTRODUCTION

During recent decades the study of electronic transport has flourished in the arena of low dimensional systems. This was mainly due to the advent of miniaturization techniques that allowed the exploration of transport properties of semiconductor heterostructures and quantum dots, and single molecules contacted to electrodes. Most of the experimental and theoretical studies were focused on time-independent transport. In contrast, much less attention has been devoted to the influence of time-dependent excitations by electromagnetic fields or gate voltages. Despite that, interest has been steadily growing[1,2] and many captivating phenomena such as photon-assisted tunnelling, coherent destruction of tunnelling[3] and quantum charge pumping[4,5,6] have been unveiled. Time-dependent excitations provide an opportunity to achieve control through selective excitations, opening an avenue for both fundamental research and practical applications. From the point of view of fundamental research, the study of the interplay between external fields and the transport mechanisms offer a rich prospective to improve the understanding of electron dynamics at the molecular scale. On the other hand, the use of these external fields as means to control currents in coherent conductors may offer innovative alternatives for practical applications such as switches[7], interconnects[8] and sensors[9].

The feasibility of the implementation for a given material being the ultimate limitation to the actual realization of such innovations, it is therefore important to carefully select the physical objects for study. Carbon based materials such as polymers, graphitic structures and fullerenes are promising materials. Within the family of fullerenes, single-walled carbon nanotubes[10,11], hollow cylinders of nanometer scale diameter formed by rolling a graphene sheet (nanotubes hereafter), stand as one of the major driving forces of both fundamental and applied research due to their outstanding mechanical and electrical properties[10,11]. Besides nanotubes, graphene is also emerging as a preferred choice due to its peculiar electronic structure and the better possibilities for building reproducible contacts to electrodes[12,13,14] and could reveal remarkable phenomena when exposed to AC fields.

In this paper we focus on the case of ac transport through single walled carbon nanotubes, being most of the effects that we show generalizable to other classes of low-dimensional systems. First, we point out some of the novel phenomena unique to AC fields and then mention the main challenges as well as the tools for the description of driven transport in carbon-based devices. This adresses the questions: why AC fields and how. In Sec. 3 we offer a brief review of our recent research in this promising area giving a concrete example for the case of Fabry-Pérot devices[15].

A. Novel phenomena under AC fields

In the context of quantum transport, time-dependent phenomena can be classified according to their origin[16]: they can arise as a result of an external driving (applied voltage, time-dependent magnetic flux, etc.) or may occur spontaneously (frequency dependent shot noise[17]). Our main interest is on the former situation.

One of the most studied phenomena produced by external driving is *photon-assisted tunnelling* (PAT). The first experiments showing PAT were carried out during the 1960s in superconductor-insulator-superconductor structures[18].

Motivated by these experiments, Tien and Gordon[19] proposed a simple theory for PAT, the main idea is that a time-dependent potential can induce inelastic tunnelling events by allowing the exchange of energy quanta (photons) between electrons and the oscillating field. Since then, photon-assisted tunnelling has proven to be ubiquitous in electronic transport and it has been observed in double barrier devices, molecules and, more recently, in carbon nanotubes[20].

Another captivating effect is *quantum charge pumping*[4,5]. A direct current (dc) is usually associated to a dissipative flow of the electrons in response to an applied bias voltage. However, in systems of mesoscopic scale a dc current can be generated even at zero bias. This intriguing quantum coherent effect is called quantum pumping and a device capable of providing such effect is termed a quantum pump. Quantum pumping has attracted much attention[21,22,23,24,25,26,27] and experiments aimed at observing this phenomenon have been carried out in semiconductor quantum dots[6], quantum wires[28,29] and also in carbon nanotubes[30]. Quantum spin pumping is also a very active area of research[31,32,33].

The operational regime of a pump can be characterized according to the relative magnitude between the driving frequency Ω and the inverse of the traversal time through the sample, $1/\tau_T$. When $\Omega \ll 1/\tau_T$, the pump is in the so-called adiabatic regime, whereas the opposite case, $\Omega \gg 1/\tau_T$, the pump is in the nonadiabatic regime. For a cyclic adiabatic change of the conductor parameters, the charge pumped per cycle is determined by the area enclosed in parameter space[21]. Beyond the adiabatic regime, pumping has been also studied both theoretically[22,26,34,35,36,37,38] and experimentally[29,39]. Although currents obtained in the nonadiabatic regime are naturally higher, keeping a low current noise becomes crucial[26] to obtain useful devices.

Finally, another issue of interest that we would like to point out is the *phase sensitivity of the current noise*. While for the case of a static conductor the noise depends only on the transmission probability and not on the phase, for a driven conductor the current noise is phase sensitive[2]. Later on we will show an example of our recent research highlighting this issue.

B. Challenges in the description of driven transport through nanotubes and beyond

Transport through carbon nanotubes in the presence of time-dependent excitations remains much less explored than its time-independent counterpart. Experimental studies include the response to microwave fields[20,40,41] and quantum pumping in response to surface acoustic waves[30]. Although some theoretical studies addressing these topics are available in the literature[42,43,44,45,46], the present understanding is not complete. Besides, as we will see later, many of the results obtained for carbon nanotubes could apply to the case of driven transport in graphene devices, a topic of much current interest[47,48,49].

The main challenges for a proper theoretical description stem from the fact that, besides the need of having an adequate modelling of the electronic structure, the most interesting regime lies beyond the scope of either low driving frequencies (adiabatic limit) or perturbative approaches. Although ambitious, any theoretical advance in this direction will be useful not only in the context of carbon nanotubes, but in the broader realm of molecular electronics[50] and therefore we decided to address this problem.

C. Fabry-Pérot regime in carbon nanotubes and the influence of AC fields

Low resistance contacts are very difficult to achieve in molecular systems. Indeed, most of the experiments in molecular electronics are in the Coulomb-Blockade regime where coupling between the molecule and the electrodes is weak. However, carbon nanotubes offer a unique opportunity to experimentally explore high quality low resistance contacts[51]. Indeed, transport experiments carried out on metallic tubes with low resistance contacts show ballistic transport and Fabry-Pérot (FP) interference at low temperatures[52]. In this regime, coherence plays a major role and is manifested as oscillations in the conductance as a function of the bias voltage. Besides the conductance properties, the current noise has also been experimentally probed in this regime[53,54,55] as well as the effect of strong magnetic fields[56].

Motivated by these experiments, we decided to consider the case of ac gating of a metallic nanotube-based Fabry-Pérot resonator[15]. Our main question was whether control of the current and noise could be achieved by tuning the parameters (such as intensity and frequency) of a minimal ac field. To our surprise, our subsequent research not only gave a precise and detailed answer to this question but also pointed out a striking manifestation of the phase sensitivity of the current noise[15]. We will give a closer look to this topic in Sec. 3 but before we will briefly present our theoretical tools.

II. TIGHT-BINDING MODEL AND FLOQUET SOLUTION

The theoretical approaches capable of describing time-dependent transport include the Keldysh or non-equilibrium Green functions formalism[57,58,59,60], schemes that use density functional theory[61], the equation of motion method[62], and schemes that exploit the time-periodicity of the Hamiltonian through Floquet theory[23,63,64].

The main advantage in the Keldysh formalism is that interactions (electron-electron and electron-phonon for example) can be included more easily than in the other schemes. On the other hand, in the Floquet approaches the picture is that of a single particle but the time-periodicity of the Hamiltonian is fully exploited through the use of Floquet theorem thereby simplifying the problem.

In our current research as a general framework we use the Floquet scheme[63] combined with the use of Floquet-Green functions[36]. Within this formalism, the dc component of the time dependent current $I(t)$ can be computed as:

$$\bar{I} = \frac{2e}{h} \sum_n \int \left[T^{(n)}_{R \longleftarrow L}(\varepsilon) f_L(\varepsilon) - T^{(n)}_{L \leftarrow R}(\varepsilon) f_R(\varepsilon) \right] d\varepsilon, \tag{1}$$

where the transmission probabilities from left (L) to right (R) involving the emission (absorption) of n photons, $T^{(n)}_{R \longleftarrow L}(\varepsilon)$, can be fully written in terms of the Green functions for the system[2,36]. The current noise can be obtained from the correlation function $S(t,t^{'}) = \langle [\Delta I(t) \Delta I(t') + \Delta I(t') \Delta I(t)] \rangle$, $\Delta I(t) = I(t) - \langle I(t) \rangle$ being the current fluctuation operator. The noise strength can be characterized by the zero frequency component of this correlation function averaged over a driving period, $\bar{S}$, which can be casted in a convenient way within this formalism[2]. Further simplifications can be achieved by using the broad-band approximation and an homogenous gating of the tube[2].

For simplicity we consider an infinite CNT described through a standard π-orbitals Hamiltonian[10]

$$H_e = \sum_i E_i c_i^\dagger c_i - \sum_{\langle i,j \rangle} [\gamma_{i,j} c_i^\dagger c_j + \text{H.c.}], \tag{2}$$

where $c_i^\dagger$ and c_i are the creation and annihilation operators for electrons at site i, E_i are the site energies and $\gamma_{i,j}$ are nearest- neighbors carbon-carbon hoppings. The modelling of the Fabry-Pérot interferometer is done by connecting a central part of length L (the "sample") to the rest of the tube through matrix elements γ_t smaller than the hoppings in the rest of the tube which are taken to be equal to $\gamma_0 = 2.7$ eV. In the vicinity of the charge neutrality point, the dispersion relation is linear and the mean level spacing of the isolated sample scales as $\Delta \propto 1/L$. A uniform ac gating of the sample can be modeled as an additional on site energy $E_{j \in \text{CNT}} = eV_g + eV_{\text{ac}} \cos(\Omega t)$. In the following section we will comment on some results obtained using this general framework.

III. CONTROL OF THE CONDUCTANCE AND NOISE IN DRIVEN FABRY-PÉROT DEVICES

Recently, we showed that the interplay between the ac field parameters (field intensity and frequency) and the typical energy scales of the nanotube (such as level spacing and position of van Hove singularities) can lead to strong modifications of the conductance and current noise[15]. As compared to the conductance interference pattern observed in static conditions, by tuning the field intensity and frequency it can either be suppressed, exhibit a revival, or show an ac-intensity independent behavior. The former situation occurs whenever the ac field intensity attains specific values that maximize the smoothing of the interference pattern introduced by the inelastic processes, while the latter takes place whenever the driving frequency is commensurate with the mean level spacing Δ. This is illustrated by the condutance interference patterns shown in Fig. 1a-d. These patterns correspond to a numerical calculation for a 2 nm diameter nanotube of 0.44 μm length. The color scale (from dark to light) is in the conductance range $(0.64, 1)$ in units of the quantum limit for the conductance of such a system, i.e., $4e^2/h$.

The overall behavior of the conductance as we move in the driving amplitude-frequency space is captured in 2a, where the half amplitude of the conductance oscillations is shown in a color scale (white corresponds to maximum amplitude and black to vanishing amplitude). There, the vertical light regions indicate the regions where the interference pattern is unaffected by the applied ac field. For low and intermediate frequencies, we observe an alternating suppression and revival of the interference.

The situation in Fig. 1-d and 2a reminds us of the wagon-wheel, stroboscopic or aliasing effect. The results of Fig. 1 hint that something similar occurs with the conductance patterns for our Fabry-Pérot device, although here the energies or time-scales involved belong to the quantum domain.

Based on the previous picture one would be tempted to think that whenever the wagon-wheel condition is met both the current *and* the noise should behave as in the static case. However, numerical and analytical calculations[15] of the

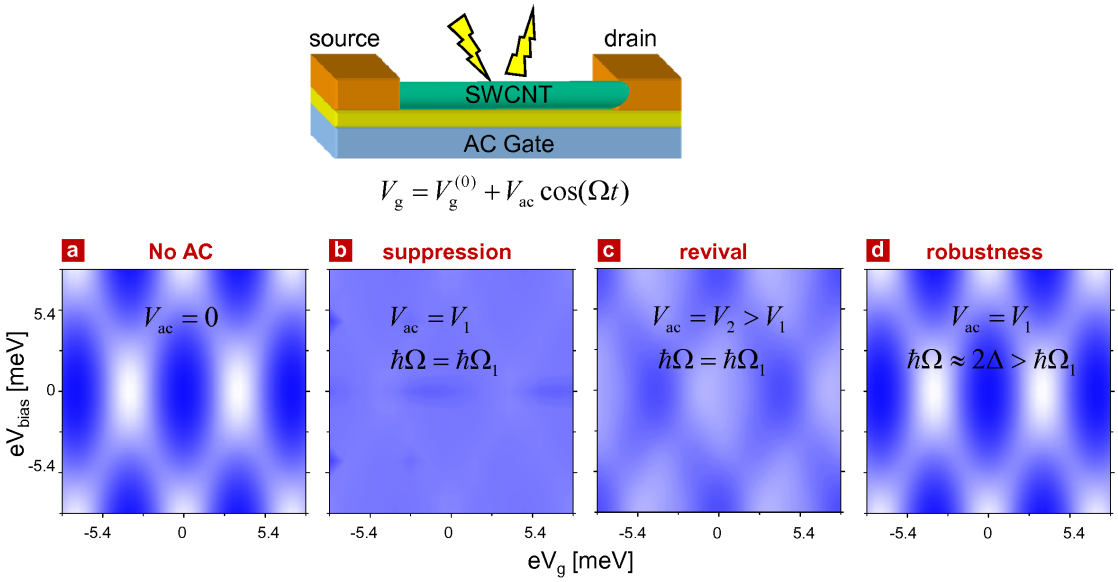

FIG. 1: (color online) Top: scheme of the system considered in the text, namely a SWCNT connected to source and drain electrodes and in the presence of an ac gating. Bottom: a-d are the Fabry-Pérot conductance patterns calculated for different values of the ac field frequency and amplitude (blue and white correspond to low and high conductances respectively).

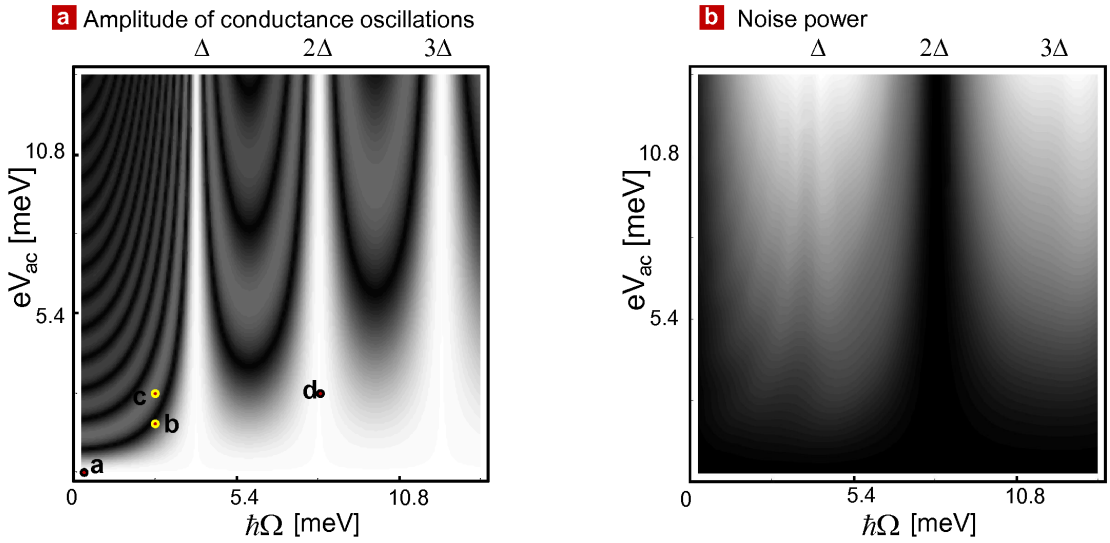

FIG. 2: (color online) a) Contour plot showing the amplitude of the Fabry-Pérot oscillations as a function of the driving amplitude and frequency. White stands for maximum amplitude and black for vanishing amplitude. b) Same contour plot for the current noise $\bar{S}$, black is for vanishing noise.

current noise show that this is not the case. Interestingly, the current noise in the low temperature and low bias limit (which is purely due to the ac field) is suppressed whenever the frequency is commensurate with *twice* the mean level spacing as can be appreciated in the contour plot of the current noise as a function of the ac amplitude and frequency Fig. 2b. This happens due to the fact that the noise under ac conditions is sensitive to the phase of the transmission amplitude, which changes only by π (and not by 2π) from one resonance to the next one. As emphasized in[15], this constitutes a striking manifestation of the phase sensitivity of the noise under ac conditions leading to what we call the *quantum wagon-wheel effect*.

Although the frequencies required to achieve the wagon-wheel condition are here quite high (of about 100 GHz), they are still in the order of what can be experimentally achieved[65] and can be reduced by further tuning of the tube length and temperature. Besides, we must emphasize that the main features on the left side of these amplitude-frequency maps (low frequencies) persist even in the adiabatic limit. We point out that a similar noise suppresion was reported in[66] for a different system, namely two barriers of varying strength and a uniform varying potential in between. Noise suppression with maximum current was also discussed for a different system in Ref.[26].

Again, we would like to stress that although we considered only the case of carbon nanotubes our results remain valid for materials where the dispersion relation may deviate from linear as discussed in the appendix and elsewhere[67].

IV. CONCLUSIONS

An overview of some of the challenges and perspectives in the field of driven quantum transport in carbon-based devices was presented. The study of driven quantum transport in nanoscale devices is surely necessary (after all if these devices are going to be integrated in everyday electronics they will probably have to work under ac conditions). Moreover, ac fields also allow for a wealth of novel phenomena which can help to achieve control of the conductance, current noise and may be even of the energy dissipation.

A particular example for the case of driven nanotube based Fabry-Pérot devices was highlighted. We have shown that by tailoring the parameters of an ac gating, important modifications can be achieved in both the conductance and the current noise in all the frequency range. For frequencies commensurate with twice the mean level spacing, both the conductance and the noise behave as in the static case in a kind of *quantum wagon-wheel effect*.

On the other hand, the phase sensitivity of the current noise under ac conditions may offer interesting perspectives for the quantification of the decoherence time[68,69] in these low dimensional systems. This would require the modelling of decoherent processes.

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V. APPENDIX: THE INFLUENCE OF NON-LINEAR DISPERSION

In this appendix we address the issue of the effect of nonlinearities in the energy dispersion in the phenomena reported in Sec. 3. A first observation is that Fermi energies very close to the precise point where the dispersion relation has a vanishing first derivative are not of interest because the system does not behave as a proper metal and further gating is needed to observe Fabry-Pérot interference. That said, we concentrate on metallic systems with a generic dispersion relation close to the Fermi energy.

For the case of carbon nanotubes, the dispersion relation at the charge neutrality point is linear. As we move away from this point, the dispersion relation acquires a non-linear component which naturally introduces dispersion in the level spacings (as the levels of the finite nanotube are not all equally spaced in the energy range of interest). This dispersion introduces corrections to the simple picture presented in the manuscript to understand the numerical results.

Indeed, a closer scrutiny at our numerical results[15] (which were obtained for a full π-orbitals model for the nanotube) reveals that the dips in the oscillations of the half amplitude of the FP conductance are not perfectly zero as it would be for a system with constant level spacing. The half amplitude of the FP oscillations at a frequency commensurate with the level spacing is not perfectly constant neither.

When the relevant energies are further away from the perfectly linear dispersion region, our simple picture will still be a good approximation provided that the energy range effectively probed by the field ($\sim \max(eV_{ac}, N\hbar\Omega)$, where N is the typical number of photons excited by the field) is small enough. This can be realized by making a Taylor expansion of the energy dispersion around the Fermi wave-vector:

$$\varepsilon(k) = \varepsilon_F + \left(\frac{\partial\varepsilon}{\partial k}\right)_{k=k_F}(k-k_F) + \left(\frac{\partial^2\varepsilon}{\partial k^2}\right)_{k=k_F}(k-k_F)^2 + ...,$$

and requiring for the quadratic term to be smaller than the linear term. This is always fulfilled in the low frequency/low ac-amplitude limit provided that $(\partial\varepsilon/\partial k)(k=k_F) \neq 0$. In more formal terms, $\max(eV_{ac}, N\hbar\Omega) \ll \varepsilon_F$ is a sufficient

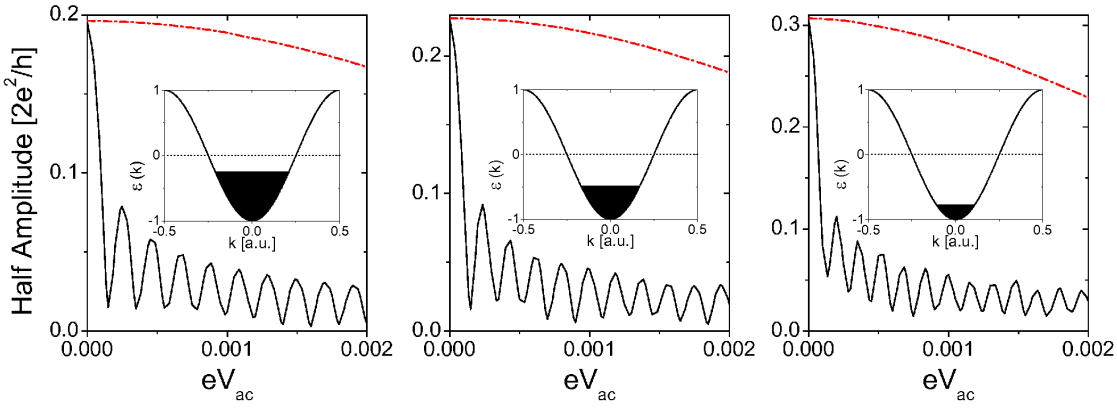

FIG. 3: (color online) Half amplitude of the Fabry-Pérot conductance oscillations as a function of the amplitude of the ac-gating computed for a 1d chain connected to electrodes. The panels are for different Fermi energies: $\varepsilon = -0.25$ (left), $\varepsilon = -0.5$ (center) and $\varepsilon = -0.75$ (right). The solid lines are for $\hbar\Omega = 1.8 \times 10^{-4}$ and the dash-dotted lines correspond to frequencies equal to the mean level spacing. All the energies are given in units of half the band width (W). The position of the Fermi energies are schematized in the insets. The change in the vertical scale of the different panels is because at fixed coupling to the leads, the escape rates to the leads change with energy, thereby modifying the amplitude of the Fabry-Pérot oscillations.

requirement.

To illustrate this point we show in Fig. 3 the results of calculations for an ac-gated 1d system at different Fermi energies. Solid black curves correspond to a frequency smaller than the mean level spacing whereas the dash-dotted red lines are for a frequency equal to the level spacing. As can be appreciated, the effect is quite robust even for Fermi energies very close to the minimum of the dispersion relation.

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