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Laser-induced effects on the electronic features of graphene nanoribbons

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We study the interplay between lateral confinement and photon-induced processes on the electronic properties of illuminated graphene nanoribbons. We find that by tuning the device setup (edges geometries, ribbon width, and polarization direction), a laser with frequency Ω may either not affect the electronic structure, or induce bandgaps or depletions at ±hΩ/2, and/or at other energies not commensurate with half the photon energy. Similar features are also observed in the dc conductance, suggesting the use of the polarization direction to switch on and off the graphene device. Our results could guide the design of novel types of optoelectronic nano-devices.

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The extraordinary properties of graphene1–3 led to an unprecedented narrowing in the expected gap between the understanding of phenomena and the development of disruptive applications.4 Though originally focused mainly on pure electronic, mechanical, or optical properties, much attention is now devoted to the interplay between these variables.4 Graphene optoelectronics,4–10 in particular, is one of the most active and promising fields with flagship applications including energy harvesting devices6 and novel plasmonic properties.8,11

Recently, the captivating possibility of controlling the electronic properties of graphene through simple illumination with a laser field12,13 has been re-examined through atomistic calculations,14 calculations of the optical response,15,16 and proposals for tuning the topological properties of the underlying photon-induced states,17–20 among other interesting issues.21–26 The basic idea is that laser illumination may couple states on each side of the charge neutrality point inducing a bandgap at ±hΩ/2, if the field intensity and frequency are appropriately tuned. This non-adiabatic and non-perturbative effect relies crucially on the low dimensionality and peculiar electronic structure of graphene and has attracted much recent attention.17–20 Notwithstanding, most of these predictions were restricted to bulk graphene. One may wonder about the possible consequences of reduced dimensionality and quantum confinement.

Here, we address the effects of laser illumination on graphene nanoribbons and show that lateral confinement plays a crucial role: tuning the sample size and the direction of the laser polarization relative to the sample edges, linearly polarized light may or not induce bandgaps or depletions in the density of states and the conductance spectra. Strikingly, for finite size samples, these features may appear at energies different from integer multiples of ±hΩ/2. This is in stark contrast with bulk graphene where the electrical response is insensitive to the linear polarization direction. Our results fill the gap in the understanding of the laser-induced control of the electrical response and may guide the design of experiments on optoelectronic devices.

Hamiltonian model and solution scheme. We consider an infinite graphene nanoribbon illuminated by a laser only in a finite region of length L and perpendicular to it (as shown schematically in Fig. 1(a)). Electrons in the graphene ribbon are modeled through a nearest neighbours π-orbitals Hamiltonian3,27

\[ H_c = \sum_i \left( E_i c_i^\dagger c_i - \sum_{ij,\pi} t_{ij} c_i^\dagger c_j + H.c. \right), \]

where \( E_i \) and \( c_i \) are the electronic creation and annihilation operators at site \( i \), \( E_i \) are the on-site energies and \( \gamma_{ij} \) the nearest-neighbors carbon-carbon hopping amplitudes, which are taken equal to \( \gamma_0 = 2.7 eV. \)3 Radiation is described through a time-dependent electric field \( \mathbf{E} \). By choosing a gauge such that \( \mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t} \), where \( \mathbf{A} \) is the vector potential, the hopping matrix elements acquire a time-dependent phase according to:

\[ \gamma_{ij} = \gamma_0 \exp(i \frac{2\pi}{\omega} \int_0^t \mathbf{A}(t) \cdot d\mathbf{r}), \]

where \( \Phi_0 \) is the magnetic flux quantum.

Retaining non-perturbative and non-adiabatic corrections to the electrical response is crucial for the results presented hereafter. In this regime, Floquet theory provides an appropriate framework. An efficient solution using this scheme is used to obtain the average density of states and the dc component of the conductance, which is computed from the inelastic transmission probabilities in Floquet space.28 The interested reader may find further generalities of the method in Refs. 28 and 29, while more technical details will be published elsewhere.30 For a periodic modulation of the hoppings, the spectral and transport properties can be derived from the so-called Floquet Hamiltonian: \( H_F = H_c - i \hbar \Omega / \partial t \). Such Hamiltonian has a time-independent representation in the Floquet space, which is the direct product between the usual Hilbert space and the space of time-periodic functions with the same period as the Hamiltonian \( H_c \).28,31 Therefore, on top of the \( k \) label, our states have a second label \( n \), which indicates the number \( n \) of photon excitations in the system. In the absence of radiation, the quasi-energies spectrum of \( H_F \) is given by \( \varepsilon(k, n) = \varepsilon_0(k) + n \hbar \Omega \) (\( \varepsilon_0(k) \) is the spectrum of \( H_c \)).
Electronic properties of irradiated graphene nanoribbons. The first question concerns the response of graphene nanoribbons to a linearly polarized laser excitation. While in the bulk limit, both the conductance and the density of states (DOS) are independent on the polarization direction, the picture turns out to change radically in confined geometries.

Figure 2 shows the average density of states as a function of the Fermi energy for a frequency in the mid-infrared regime ($\hbar\Omega = 140$ meV). Two different armchair nanoribbons widths and polarizations are chosen: $N = 130$ (a) and (b), $N = 129$ (c) and (d); and linear polarization along the $x$ (a) and (c) and $y$ (b) and (d) directions. For $N = 130$, one sees the appearance of strong depletions at $\pm \hbar\Omega/2$. These depletions are located at the same energy as the ones for the bulk system and correspond to the excitation of an electron between the conjugate states at $\pm \hbar\Omega/2$. In striking contrast, the DOS is restored at $\pm \hbar\Omega/2$ for $y$-polarized laser, whereas features occur at energies incommensurate with $\hbar\Omega$.

The DOS for the ribbon with $N = 129$ (c and d) also exhibit a laser-induced fragmentation of the spectrum. In this case, the observation of fully depleted energy regions (bandgap) at $\pm \hbar\Omega/2$. One observes that by increasing the laser intensity, some DOS modifications are further enhanced (see for instance, the DOS depletion around $\hbar\Omega/2$ for Fig. 2(a)), complemented by the emergence of fine structure. Figure 3 shows the dc conductance as a function of the Fermi energy position for the case shown in Fig. 2. Here, we see that the depletions in the DOS yield the same conductance fingerprints. One can see that switching the polarization direction may produce a marked on-off ratio if the Fermi energy is appropriately tuned.

To rationalize these differences, it is instructive to write $H_e$ in a basis of independent transversal channels or modes as discussed in Refs. 33 and 34. In the absence of radiation, the Floquet spectrum of the system is just the sum of the contributions from each of the modes $\varepsilon_0(k)$ plus their Floquet replicas: $\varepsilon(k,n) = \varepsilon_0(k) + n\hbar\Omega$. An illustration for a very small system is shown in Fig. 1(b), each mode contains an electron and a hole branch. At the crossings between the different lines, one may expect stronger effects of the radiation (if it provides the necessary coupling between the corresponding states).

From the Floquet spectrum in Fig. 1(b), one can see that there are two kind of crossings: the ones that connect an electronic (hole) state with a hole (electron) state belonging to the same mode plus or minus an integer number of photons (like the one marked with an open circle); and the ones that connect states in different modes (as the one marked with an open square in Fig. 1(b)). In such cases, a non-vanishing matrix element of the Floquet Hamiltonian will give intra and inter-mode transitions, respectively. Given the electron-hole symmetry of the spectrum, intra-mode transitions always connect states, which are symmetric relative to...
the Dirac point, i.e., integer multiples of $\pm \hbar \Omega/2$. On the other hand, inter-mode transitions always couple states which are not symmetrically located from the Dirac point, as can be seen on Fig. 1(b). A scheme showing these two type of transitions is shown in Fig. 1(b).

For armchair graphene nanoribbons, it turns out that a laser with linear polarization along the transport direction ($x$) does not mix these transversal modes, leading to features in the density of states only at $\pm \hbar \Omega/2$ as can be seen in Fig. 1(b). On the other hand, calculation of the matrix elements shows that when the polarization is along $y$, inter-mode processes are allowed and intra-mode ones are forbidden. Depletions and gaps develop at the crossing between Floquet states corresponding to different modes leading to the features observed in Figs. 2(b) and 2(c).

A complementary approach to this problem is possible by using the $k_p$ model, which could be accurate enough for medium-sized ribbons. A careful analysis shows consistent results: inter-mode processes lead to gaps/depletions located away from $\pm \hbar \Omega/2$ while polarization along $y$ suppresses the depletions at $\pm \hbar \Omega/2$. In the bulk limit, as the energy difference between subbands gets smaller, the crossings between Floquet states accumulate close to $\pm \hbar \Omega/2$ leading to the same behavior for both polarizations (along $x$ and $y$). A flavor of this can be seen in the red dotted lines in Figs. 2(b) and 2(c): The two depletions seen in Fig. 2(d) black line merge when increasing the laser power.

Another interesting feature is that the metallic modes/subbands in armchair ribbons are quite insensitive to the radiation (as seen in Figs. 2 and 3). Hence, very small metallic armchair ribbons containing only one transport channel within the energy range of interest will not experience relevant changes in their electronic properties. We emphasize that this is a peculiar property of armchair graphene nanoribbons.

Figures 2 and 3 correspond to a laser frequency in the mid-infrared, which gives an optimum playground to test these predictions in the laboratory. Going to higher frequencies may help to achieve device miniaturization but the size of the gaps and depletions at constant laser power diminishes, whereas at very low frequencies (THz), the gaps further decrease. Experiments would require temperatures below 20 K for $P \approx 1 \text{mW}/\mu \text{m}^2$.

In summary, we show that the interplay between photon-induced inelastic processes and lateral confinement in graphene nanoribbons leads to diverse modifications in the band structure and transport properties not evident in the bulk limit. In the case of moderate sized nanoribbons (ca. 10 nm), the careful tuning of the polarization direction may widen the opportunities for achieving control of the electrical response in optoelectronic devices.

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