Time evolution of Floquet states in graphene

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Based on a solution of the Floquet Hamiltonian we have studied the time-evolution of electronic states in graphene nanoribbons driven out of equilibrium by time-dependent electromagnetic fields in different regimes of intensity, polarization and frequency. We show that the time-dependent band structure contains many unconventional features that are not captured by considering the Floquet eigenvalues alone. By analyzing the evolution in time of the state population we have identified regimes for the emergence of time-dependent edge states responsible of charge oscillations across the ribbon.

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If a time periodic field is applied to electrons in a periodic lattice the Bloch theorem can be applied twice, both in space and in time. This is the essence of the Floquet theory10-14 that has recently attracted a renewed interest for its ability to describe topological phases in driven quantum systems15-17. The discovery that circularly polarized light may induce non trivial topological behaviour in materials that would be standard in static conditions18-21 has opened the way to the realization of the so-called Floquet topological insulators, where a topological phase may be engineered and manipulated by tunable controls such as polarization, periodicity and amplitude of the external perturbation.

In the presence of a continuous periodic driving electrons are in a non-equilibrium steady state characterized by a periodic time dependence of the wavefunctions and, consequently, of the expectation values of any observable. In this paper we focus on this time dependence, looking for the time evolution of some relevant observables such as energy and charge density. We will consider the prototypical case of graphene that under the influence of a periodic time dependence of the wavefunctions and, consequently, of the expectation values of any observable.

Under a periodic driving the non-equilibrium steady states, solutions of the time-dependent Schrödinger equation

\[
\left( \hat{H}(r, t) - \frac{\partial}{\partial t} \right) \psi(r, t) = 0,
\]

evolve in time as

\[
\psi(r, t) = e^{-i\epsilon \alpha t} \phi(r, t)
\]

where \(\phi(r, t)\) is periodic in time and \(\epsilon_\alpha\) - the Floquet quasi-energies - are the eigenvalues of an effective Hamiltonian \(\hat{H}^F \equiv \hat{H} - i \frac{\partial}{\partial t}\) - the so-called Floquet Hamiltonian:

\[
\hat{H}^F \phi_\alpha(r, t) = \epsilon_\alpha \phi_\alpha(r, t).
\]

Here \(\hat{H}(r, t)\) is the full Hamiltonian of the driven system

\[
\hat{H}(r, t) = \hat{H}_0(r) + \hat{V}(r, t)
\]

with \(\hat{H}_0(r)\) the static Hamiltonian and \(\hat{V}(r, t)\) the external periodic driving. The factorization in eq. 1 is exact and represents the temporal analogue of the Bloch theorem. Being \(\phi_\alpha(r, t)\) time-periodic it can be expressed as a Fourier series

\[
\phi_\alpha(r, t) = \sum_{n=-\infty}^{\infty} B_{\alpha n}(r) e^{-i n \Omega t}.
\]

where in turn \(B_{\alpha n}(r)\) can be expanded on a complete set, for instance on a localized basis

\[
B_{\alpha n}(r) = \sum_i C_{\alpha i} \chi_i(r)
\]

with \(i\) a site index, \(N\) the number of sites in the unit cell and \(\chi_i(r)\) the localized orbital12. In practice the Fourier expansion is truncated to include a finite number of modes, up to a sufficiently large \(n_{\text{max}}\) whose value depends obviously on \(\Omega\). This allows to formulate the eigenvalue problem in eq. 2 in a standard matrix form whose eigenvalues turn out to be replicas of the static band structure with gaps opening at their crossing points13.
The field-free Hamiltonian of graphene is described in the tight-binding scheme with a single hopping parameter $J \simeq 2.8\text{eV}$ between nearest neighbor sites, reproducing the well-known Dirac-like valence and conduction bands. In the presence of the oscillating field described by the vector potential $\mathbf{A}(t)$, the hopping between neighboring sites is modified according to Peierls’ substitution:

$$\tilde{J}_{ij}(t) = J e^{i\mathbf{A}(t) \cdot (\mathbf{r}_j - \mathbf{r}_i)} \quad (6)$$

We are interested in the effects of reduced dimensionality, namely on the gapless edge states that arise in graphene ribbons; we chose a zig-zag terminated ribbon 50-atom wide (Fig. 1). We consider two frequency values ($\Omega_1 = 5.5\text{eV}, \Omega_2 = 12\text{eV}$) representative of the intermediate and large frequency regime ($\Omega_1/J \simeq 2, \Omega_2/J \simeq 4$). We study also the effect of different amplitudes of the external vector potential ($A_0 = 0.5$ and $A_0 = 1.5$ in units of the inverse carbon-carbon distance). In Figs. 2 we compare the Floquet quasi-energies obtained for the honeycomb lattice in 2D and 1D exposed to a circularly polarized field $\mathbf{A}(t) = A_0(\cos(\Omega t)i + \sin(\Omega t)j)$.

Panels (a), (c), (e) (g) report the Floquet Projected Bulk Band Structure (FPBBS), namely the Floquet eigenvalues obtained for the 2D lattice using the ribbon unit cell but adding an extra lattice vector (see Fig. 1) to restore the 2D translation symmetry. As currently done in standard surface physics, the projected bulk band structure allows to identify straightaway the energy regions that, prohibited in the bulk, can host localized states at the edges. Panels (b), (d), (f), (h) of Fig. 2 report the Floquet quasi-energies obtained in the 1D ribbon geometry clearly showing extra states in the 2D forbidden regions. These states go in pairs being localized either on the upper or on the lower edge of the ribbon. We notice that for the largest frequency $\Omega = 12\text{eV}$ the effect of increasing $A_0$ is to widen the gap between bulk Floquet bands and to increase the edge state dispersion. Gapless edge states appearing around $k_x = 1/2$ (in units of $2\pi/a$) exhibit for both values of $A_0$ the peculiar linear dispersion evocative of a non-trivial topological character.

It is interesting to compare these results with those obtained assuming a linear polarization, with the vector potential oscillating perpendicularly to the ribbon length.
As shown in Fig. (3) 2D Floquet bands are gapless independently on the field strength. Edge states appearing in the middle of the 1D Brillouine Zone (BZ) have no appreciable k-dispersion in the same way as edge states in static conditions.

![FIG. 3. Same as Fig. 2 but for linear polarization.](image)

We may conclude this analysis of Floquet quasi-energies by noticing that only circularly polarized fields of sufficient strength may induce Floquet edge states with a significant linear dispersion. It is interesting now to go one step further and use the solution of the Floquet problem to obtain information on physical observables.

Even if Floquet quasi-energies are used to interpret the photon dressed electronic excitations probed by spectroscopic techniques such as pump-probe experiments and time-resolved photoemission their connection with measurable quantities is somewhat indirect: Floquet quasi-energies are time-independent eigenvalues of an auxiliary Hamiltonian and as such cannot be strictly interpreted as "true" observables of the full time-dependent one. However Floquet eigenvalues and eigenvectors can be used to calculate expectation values exactly, thanks to the exact representation of the time-dependent wave-functions (eq. [4]). In particular the time-dependent single particle energies of the driven system, defined as the expectation values of the time-dependent Hamiltonian over \( \psi(r,t) \), can be expressed in terms of the Floquet eigenstates as follows

\[
E_\alpha(t) \equiv \langle \psi(r,t) | \hat{H}(r,t) | \psi(r,t) \rangle = e^{i(n-m)\Omega t} \sum_{n,m} e^{i(n-m)\Omega t} m \Omega < B_{\alpha n}(r) | B_{\alpha m}(r) >
\]

These time-dependent single-particle energies represent the extension of the band structure to the time domain. Notice that at each time step we are able to populate the single particle levels quite naturally by a straightforward electron counting and so to distinguish between filled and empty states in the same way as in static conditions. On the contrary, this distinction is far from being obvious for Floquet quasi-energies and the issue of Floquet state occupation remains an open question.

For 1D ribbons we are interested in particular on the time evolution of edge states. As shown in Figs. (2,3) Floquet edge states exist only in a narrow portion of the 1D BZ where we now plot their time evolution (Fig. (4)). By looking at the site composition of the wave function we can unambiguously identify states localized either at the upper or at the lower edge and in the following we describe the time evolution of each of them. We consider...
Let us focus first on circular polarization. The high frequency regime (Fig. 4 panels (a) and (b)) is particularly interesting: during the time evolution the two edge states modify their $k$-dispersion keeping however the same positive or negative slope: positive for states localized at the upper edge and left movers on the lower one. This is remarkable since it corresponds to unidirectional edge states that would carry, if occupied, a constant current around the sample. For a smaller frequency (Fig. 4 panels (c), (d)) this is no more completely true and edge states exhibit a more complex $k$-dispersion with kinks and variable slopes. We also stress that for smallest frequency and largest intensity (panel (d) of Fig. 4) the edge localization is less pronounced than in all other cases.

The effect of linearly polarized fields is significantly different: edge states are confined in a reduced $k$-space region and their overall dispersion is drastically reduced by orders of magnitude with respect to the case of circular polarization. Moreover, and even more notably, the right mover/left mover behaviour is lost and for any value of frequency and intensity the two edge bands have a parabolic dispersion, with both positive and negative slope. The width of the parabolas varies with time and the upper/lower edge bands are either fully occupied or empty.

Since in both conditions of circular and linear polarization the edge state occupation varies in time we expect this to affect the local charge density evolution. The charge density at each site $i$ in the unit cell is calculated quite simply as the sum of $|\psi(r,t)|^2$ over the occupied states. The results for circular and linear polarization are reported in Figs. (5) in units of electrons per site (assuming 1 valence electron per site in static conditions). Within the ribbon and at the edges charge oscillates in time as expected; moreover excess charge accumulates at the ribbon edges, moving in time from one edge to the other.

The width of charge oscillations at the edges is more pronounced for linear polarization and grows with field intensity, in agreement with physical intuition. The situation is more complex for circular polarization where the strongest oscillations occur for $\Omega = 5.5eV$, $A_0 = 0.5$ (panel (c) of Fig. 5). For the same frequency but for higher intensity ((panel (d) of Fig. 5)) we notice no charge accumulating at the edges. This is a consequence of the poor edge localization occurring for these parameter values.

This analysis allows us to conclude that fields of appropriate frequency and intensity of both linear and circular polarization induce an oscillating dipole across the ribbon. A quantitative analysis of the spectrum of the emitted radiation that would involve the current density and its expectation value will be the subject of a further study.

FIG. 4. Time evolution of the edge states of a zig-zag ribbon for circular (left panel) and linear polarization (right panel). States localized at upper and lower edge are indicated by black and brown lines respectively. Panels (a), (a’): $\Omega = 12eV$, $A_0 = 0.5$; panels (b), (b’): $\Omega = 12eV$, $A_0 = 1$; panels (c),(c’): $\Omega = 5.5eV$, $A_0 = 0.5$. Snapshots at times $t_n = (n-1)/8T$ are reported, $T$ being the period of the external field.
FIG. 5. Time evolution of the local charge calculated for circularly (left panels) and linearly (right panels) polarized fields of different strengths and frequencies. Panels (a), (a’): $\Omega = 12\,\text{eV}$, $A_0 = 0.5$. Panels (b), (b’): $\Omega = 12\,\text{eV}$, $A_0 = 1$. Panels (c), (c’): $\Omega = 5.5\,\text{eV}$, $A_0 = 0.5$. Panels (d), (d’): $\Omega = 5.5\,\text{eV}$, $A_0 = 1$. Snapshots are given at times $t_n = (n - 1)/8T$.

In summary, we have shown how the time evolution of physical observables in systems driven out of equilibrium by a time-periodic electromagnetic field can be obtained from Floquet eigenstates and eigenvalues. The expectation values of the time-dependent Hamiltonian over the time-dependent single-particle wavefunctions represent an extension of the band structure to the time domain giving information on the time evolution of single-particle energies, on their population and to physical quantities that require a summation over occupied states. In graphene ribbons the effects depend strongly on the polarization of the applied field and in the case of circularly polarized light in a given regime of frequency and intensity, unidirectional edge states are identified that describe electrons moving in opposite directions along the edges.

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