THE JOURNAL OF PHYSICAL CHEMISTRY LETTERS
A JOURNAL OF THE AMERICAN CHEMICAL SOCIETY

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Magnetic-Field-Driven Electron Dynamics in Graphene

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Cite This: *J. Phys. Chem. Lett.* 2021, 12, 4749–4754 Read Online

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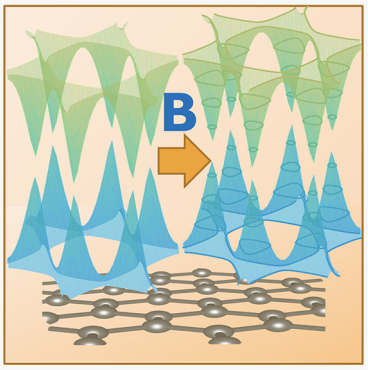
ABSTRACT: Graphene exhibits unique optoelectronic properties originating from the band structure at the Dirac points. It is an ideal model structure to study the electronic and optical properties under the influence of the applied magnetic field. In graphene, electric field, laser pulse, and voltage can create electron dynamics which is influenced by momentum dispersion. However, computational modeling of momentum-influenced electron dynamics under the applied magnetic field remains challenging. Here, we perform computational modeling of the photoexcited electron dynamics achieved in graphene under an applied magnetic field. Our results show that magnetic field leads to local deviation from momentum conservation for charge carriers. With the increasing magnetic field, the delocalization of electron probability distribution increases and forms a cyclotron-like trajectory. Our work facilitates understanding of momentum resolved magnetic field effect on non-equilibrium properties of graphene, which is critical for optoelectronic and photovoltaic applications.



Layered materials offer great potential for advanced technological applications. When layered materials thinned to their atomic limits, often referred to as two-dimension (2D) materials,[1] they exhibit different novel properties compared to their bulk counterpart. Historically, 2D materials have been one of the most extensively studied classes of materials due to their unusual physical properties. Many materials whose properties are dominated by their two-dimensional structural units, for example, the layered metal dichalcogenides (LMDCs) and copper oxides, exhibit correlated electronic phenomena such as charge density waves and high-temperature superconductivity.[2−4] The discovery[5] of single-layer graphene has shown that it is possible to exfoliate stable, single-atom, or single-polyhedral-thick 2D materials from van der Waals solids, yielding unique and fascinating physical properties. Graphene as a 2D zero-band-gap semiconductor attracted high interest.[6−9]

In recent years, different methods have been used for graphene synthesis, such as mechanical cleaving (exfoliation),[5] chemical exfoliation,[10,11] chemical synthesis,[12] and thermal chemical vapor deposition[13] (CVD) as the most common ones.[8] Its electronic band structure displays a zero-energy gap in momentum space at the six corners of the Brillouin zone (BZ) consisting three K and three K' points. It has linear energy dispersion around those points[14] which gives rise to novel phenomena, such as the anomalous room-temperature quantum Hall effect (QHE), and has opened a new category of Fermi−Dirac physics.[2] Since electrons are confined in 2D materials, graphene exhibits quantum mechanically enhanced transport phenomena driven by voltage, where QHE is observed.[6] Under a perpendicular magnetic field, $\vec{B}$, at the apex of the cones (Dirac point), electrons and holes are degenerate and Landau levels (LLs) are formed for electrons in such systems.[15,16] These quantum states have the potential to serve as carriers of quantum information for quantum computing.

The exceptional electronic and optical properties of graphene have attracted tremendous attention both theoretically and experimentally.[17−28] Several theoretical and experimental[29−31] studies of graphene have been conducted in the last years. In particular, a photoexcitation drives the system away from the equilibrium. The non-stationary and non-equilibrium dynamics of Dirac electrons in graphene have been investigated with time-resolved optical techniques.[32−42] In the presence of a magnetic field, B, the particular dispersion relations of graphene generate unique LL spectra. Recently, it has been observed that the linear dispersion of single-layer graphene leads to distinctive $\sqrt{B}$- and index-dependent energies for the LLs[43] in infrared (IR) cyclotron resonance (CR).[44,45] Non-equilibrium Dirac carrier dynamics has been investigated with time- and angle-resolved photoemission spectroscopy to understand the influence of many-body interactions on the Dirac carrier dynamics in graphene.[46] However, it is critical to study the non-equilibrium dynamics of electrons in momentum space to understand electronic transport and optical properties of graphene.

There is limited insight into the magnetic field interaction and interplay of such process on the non-equilibrium electron dynamics in graphene.[42] As a result, it is unclear how the probability density of electron dynamics is distributed in

Received: March 31, 2021
Accepted: April 29, 2021
Published: May 13, 2021

https://doi.org/10.1021/acs.jpclett.1c01020
J. Phys. Chem. Lett. 2021, 12, 4749−4754

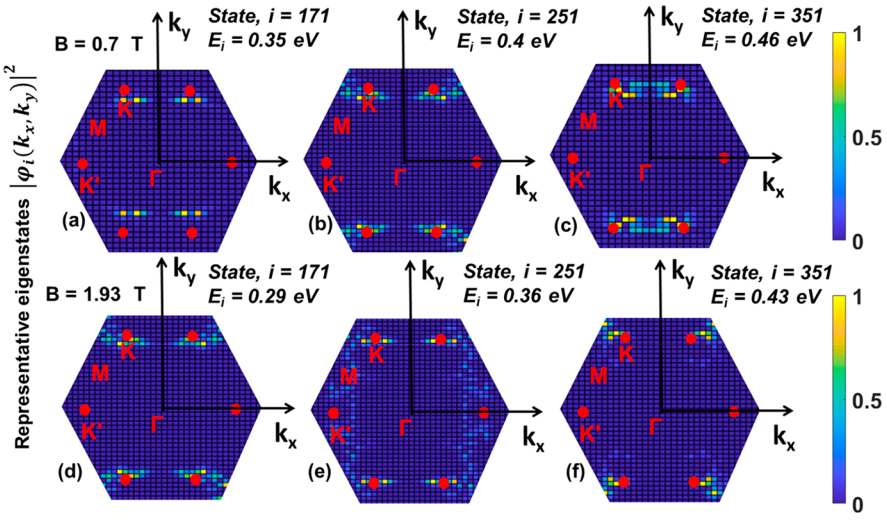


Figure 1. Eigenstates at first Brillouin zone of reciprocal space in the presence of magnetic field of different intensity. Probability density is plotted as a function of momentum vector. Probability density in different eigenstates as a function of wave vector. Here, natural sequence of rainbow colors from blue to yellow corresponds to values of probability density in range from 0 to 1, representing no zero probability density and maximum probability density, respectively.

momentum space when the magnetic field is applied. In our previous research, we have studied the electron dynamics in momentum space for silicon nanowires grown in different crystallographic directions.[47−52] The aim of this present study is to describe and understand the electron dynamics in graphene under the influence of an external magnetic field. Electron dynamics of probability density is mapped with time propagation in 2D momentum space, allowing for a separation of the different microscopic steps. Moreover, the trajectories of the expectation values of momentum are traced in the presence of magnetic field.

Graphene forms a honeycomb lattice consisting of a layer of carbon atoms. An infinite graphene sheet in the absence of external potential and magnetic fields forms the energy band structure, which is gapless in six points of the reciprocal space. The dependence of the energy spectrum on the wave vector $\vec{k}$ is almost linear in the vicinity of each of these points.[30] We are focusing only on the top of the valence band (VB) and the bottom of the conduction band (CB) well represented in terms of tight-binding (TB) Hamiltonian $H_{TB} = E^{+}(k_x,k_y)|VB\rangle\langle VB| + E^{-}(k_x,k_y)|CB\rangle\langle CB|$, with details provided in the Supporting Information (SI). In the presence of a magnetic field, the Hamiltonian includes the interaction term $\hat{H} = \hat{H}_{TB} - \mu_B \hat{L}\cdot\vec{B}$. In the following, we focus only on the CB and assume magnetic field and angular momentum oriented orthogonal to the plane of a graphene sheet $\vec{B} = (0, 0, B_z)$, $\vec{L} = (0, 0, L_z)$, with all wavefunctions and operators expressed in momentum space. The corresponding time-independent Schrödinger equation for one electron is

$$(H_{TB} - \mu_B B_z \hat{L}_z)\varphi_i(k_x, k_y) = \varepsilon_i \varphi_i(k_x, k_y) \quad (1)$$

In eq 1, ε_i is the eigenenergy and $\varphi_i(k_x,k_y)$ is the eigenstate in k-space. At zero magnetic field, eigenstates of Hamiltonian are at the same time eigenstates of the momentum operator. However, at nonzero values of a magnetic field, the eigenstates $\varphi_i(k_x,k_y)$ are defined at several values of momentum, while momentum is not a good quantum number anymore. The solution of this equation provides stationary eigenstates, which do not change in time.

In case the system is prepared in the initial state which does not coincide with any of the stationary state $\psi(k_x,k_y, t = 0) \neq \varphi_i(k_x,k_y)$, one uses the time-dependent Schrödinger equation (TDSE) for the description of the time evolution of non-stationary states:

$$i\hbar\frac{d}{dt}\psi(k_x, k_y, t) = (H_{TB} - \mu_B B_z \hat{L}_z)\psi(k_x, k_y, t) \quad (2)$$

One solves the TDSE by applying the time evolution operator $\boldsymbol{U}(\Delta\tau) = \exp\left(-\frac{i}{\hbar}(H_{TB} - \mu_B B_z \hat{L}_z)\Delta\tau\right)$ as well as by propagating wave function during infinitesimal time step $\Delta\tau$ as $\psi(k_x, k_y, t + \Delta\tau) = \boldsymbol{U}(\tau)\,\psi(k_x, k_y, t)$. On account of the initial conditions, we explore a specific class of the initial wavefunctions in 2D momentum space represented as 2D Gaussian functions, specified by the values of initial projections of momentum k_{x_0},k_{y_0}, initial positions x_0,y_0, and width parameter σ, with details provided in the SI. One can monitor electron dynamics of probability density as a function of the magnetic field by analyzing the absolute value square of the computed wavefunction $|\psi(k_x,k_y,t)|^2$.

Electron dynamics in 2D momentum space in the presence of a normally incident magnetic field is calculated by implementing the above equations using MATLAB software-based code. We consider a 2D rectangular lattice with a finite number of grid points $N = 55$ along both k_x and k_y and the total number of grid points $N^2 = 3025$. For the dynamics calculations, the total number of time steps is 2040 and the duration of each time step is 0.5 au. So, the duration of the whole trajectory is 24.67 fs. Both the total number of time steps and the duration of each time step are selected in such a way that the non-equilibrium dynamics of electron probability distribution is visible. The electron probability distribution does not appear if the time step is less than 0.5 au.

The energy band structure of graphene exhibit Dirac points, where the VB and CB touch each other.[9] In graphene, three valence electrons form σ-bonds with three neighboring carbon atoms, and the fourth electron forms π-bond normal to the plane of the σ-bonds. Since the π-bond electrons are relatively weakly bound to the nuclei and are delocalized over the graphene structure, they contribute to the electronic properties of graphene.[1]

https://doi.org/10.1021/acs.jpclett.1c01020
J. Phys. Chem. Lett. 2021, 12, 4749−4754

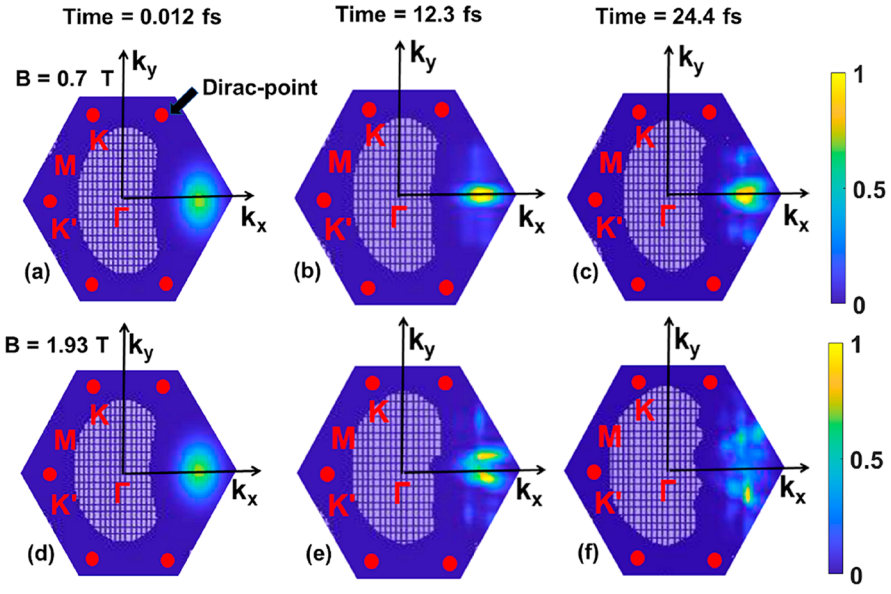


Figure 2. Electron dynamics of probability density with time propagation in 2D momentum space in the presence of magnetic field. The values of magnetic field are (a−c) B = 0.7 T and (d−f) B = 1.93 T. The electron distribution center starts from one of the D-points at the bottom of the CB. The duration of trajectory is 24.67 fs. Snapshots demonstrate probability density for electrons in CB at three instants of time, such as 0.012, 12.3, and 24.4 fs, as a function of wave vector. Here, the first BZ is shown in reciprocal space. The shape of the probability distribution extends over as time passes.

The external magnetic field affects both energies and spatial distribution of electronic states. It causes inhomogeneous carrier distribution in graphene meaning that carrier distribution is denser in the vicinity of the Dirac points. Figure 1 illustrates the solution of the time-independent Schrödinger equation, eq 1, and shows the representative eigenstates in momentum space in the BZ under applied magnetic field at two different values, i.e., B = 0.7 and 1.93 T. Most typical eigenstates for each value of magnetic field are shown here. Each panel represents eigenstate number i by probability distribution as functions of momentum $|\varphi_i(k_x,k_y)|^2$.

It is noticeable that states with lower values of eigenenergies have their maximum probability distributions near the Dirac points. At lower values of magnetic field, these states form plane waves and are localized at fixed values of momentum. As the value of applied magnetic field increases, the eigenstates are experiencing the formation of a delocalized pattern over a broader range of momentum. The electron probability distribution is minimal in the region around the gamma point. It appears that spatial distributions of some eigenstates form closed loop patterns similar to those of Landau levels. Eigenstates are either localized near Dirac points or delocalized in a closed loop manner depending on the energy of orbitals. In case of applied magnetic field B = 0.7 T, most of the eigenstates are localized at Dirac points as shown in Figure 1(a)−(c). On the other hand, for the applied magnetic field 1.93 T, most of the eigenstates are delocalized away from the Dirac points and form a closed-loop pattern as shown in Figure 1(d)−(f).

In case a light pulse is applied to the graphene, electrons are promoted from VB to the CB and the system is perturbed away from the equilibrium. The external magnetic field is expected to affect the non-stationary electron dynamics in the CB and to facilitate the formation of a cyclotron-like pattern. Magnetic field can split the energy offset between energy levels. This phenomenon affects resonance condition between electronic transitions and phonon frequencies. Match or mismatch among energies of transitions between electronic and nuclear subsystem is a key factor that speeds up or slows down electron relaxation. Moreover, the applied magnetic field is expected to facilitate the violation of momentum conservation for electrons. Figure 2 shows computed non-equilibrium electron dynamics of probability density with time propagation in 2D momentum space in the presence of a magnetic field. It shows the snapshots of the probability density dynamics at different time steps at two different values of an applied magnetic field, B = 0.7 and 1.93 T as shown in Figure 2(a)−(c) and 2(d),(e), respectively. The most representative magnetic field values and eigenstates are shown here. The full list of eigenstates is provided in the SI as movies. In this calculation, one considers electron dynamics of probability distribution starting at one Dirac point although there are six degenerate Dirac points. It is expected that the initial condition should be chosen as a superposition of six peaks in the vicinity of each Dirac point, which will result in overall electron dynamics of probability distribution resembling cyclotron trajectory, related to the formation of LLs. However, for simplicity, in what follows, the center of electron distribution starts from one of the Dirac points at the bottom of the CB.

As time goes by, the initial distribution of electron probability density delocalizes over a broader range of momentum space. Although ideal cyclotron orbits are not observed, there is formation of fringes while denser fringes imply the measure of electron motion. For larger values of magnetic field, B, the fringes are spreading in the area of Dirac points. At the lower value of the magnetic field, an electron cannot penetrate the energy barrier between Dirac points. Consequently, eigenstates are superimposed and form fringes close to the Dirac points. It is apparent that at the lower value of the magnetic field, both the higher value of the magnetic field and longer time trajectory cause delocalization of electron probability density distribution.

Linear momentum dispersion appears in the energy band structure of 2D materials. In graphene, the non-stationary electron dynamics driven by photoexcitation is able to carry on signatures of this special dispersion. In this study, we analyze eigenstates of Hamiltonian and wave-packet dynamics of probability density of electrons in graphene as a function of magnitude of external magnetic field. Our modeling results show

https://doi.org/10.1021/acs.jpclett.1c01020
J. Phys. Chem. Lett. 2021, 12, 4749−4754

that magnetic field leads to deviation from local static and dynamic momentum conservation for charge carriers. With increasing magnetic field, an increase of delocalization of electron probability distribution is observed in momentum space making step toward formation of cyclotron-like trajectory. The electrons in CB demonstrate difference in dynamical behavior at stronger and weaker magnetic fields. Electron dynamics monitored for the first 25 fs shows noticeable development in time with changing distribution as a function of momentum, which exhibits fringes with the probability distribution depending on the magnetic field amplitude. The formation of fringes reflects the vividness of the motion of electronic state while denser fringes correspond to quicker motion and correlate to cyclotron trajectories.

We study photoexcited electron dynamics in graphene using the time evolution operator for wavefunction in momentum space under interaction with applied magnetic fields. We found that an external magnetic field leads to local deviation from momentum conservation for charge carriers. It is also found that the delocalization of electron probability distribution increases with the magnetic field, forming a cyclotron-like trajectory in the momentum space. Our results contribute to the understanding of magnetic field effect on properties of 2D materials. Future research will address the photoexcitation and relaxation of charge carriers, formation of excitons, and impact of excitons on cooling and recombination in graphene. Electron dynamics in momentum space under the influence of a weak magnetic field has the potential to play an important role in graphene-based optoelectronic and information storage devices.[53]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jpclett.1c01020.

Details of methodology (PDF)

Animated movie of eigenstates, very slow scan (MP4)

Animated movie of eigenstates and their corresponding energies at first Brillouin zone of reciprocal space in the presence of magnetic field B = 0.7 T (MP4)

Animated movie of eigenstates and their corresponding energies at first Brillouin zone of reciprocal space in the presence of magnetic field B = 1.93 T (MP4)

Animated movie of electron dynamics of probability density with time propagation in 2D momentum space in the presence of magnetic field B = 0.7 T (MP4)

Animated movie of electron dynamics of probability density with time propagation in 2D momentum space in the presence of magnetic field B = 1.93 T (MP4)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work is supported by the National Aeronautics and Space Administration (NASA) under award number of 80NSSC19M0177. W.X. acknowledges the support from the North Dakota Established Program to Stimulate Competitive Research (ND EPSCoR) through the New Faculty Award No. FAR0021960. The authors thank Aaron Forde, Yulun Han, Zhaofan Li, Yangchao Liao, and Amirhadi Alesadi for collective editing. The supercomputing resources of the National Energy Research Scientific Computing Center (NERSC), a U.S. Department of Energy (DOE) Office of Science User Facility located at Lawrence Berkeley National Laboratory, under DE-AC02-05CH11231, via allocation award "Computational Modeling of Photo-catalysis and Photo-induced Charge Transfer Dynamics on Surfaces" and CCAST Thunder Cluster at NDSU are acknowledged. D.K. thanks Svetlana Kilina, David Micha, Sergei Tretiak, Oleg Prezhdo, and Yuri Dahnovski for inspiring discussions. D.K. acknowledges support from NSF CHE-1944921 and DOE DE-SC0021287. This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. DOE Office of Science by Los Alamos National Laboratory (Contract 89233218CNA000001) and Sandia National Laboratories (Contract DE-NA-0003525) along user allocation 2019BU0186: "Magnetic and Nonadiabatic Driven Dynamics in 2D Materials". The work of T.M.I. is performed under the state assignment of IGM SB RAS.

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