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Carrier multiplication in graphene under Landau quantization

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Carrier multiplication is a many-particle process giving rise to the generation of multiple electron-hole pairs. This process holds the potential to increase the power conversion efficiency of photovoltaic devices. In graphene, carrier multiplication has been theoretically predicted and recently experimentally observed. However, due to the absence of a bandgap and competing phonon-induced electron-hole recombination, the extraction of charge carriers remains a substantial challenge. Here we present a new strategy to benefit from the gained charge carriers by introducing a Landau quantization that offers a tunable bandgap. Based on microscopic calculations within the framework of the density matrix formalism, we report a significant carrier multiplication in graphene under Landau quantization. Our calculations reveal a high tunability of the effect via externally accessible pump fluence, temperature and the strength of the magnetic field.

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In 1961 Shockley and Queisser¹ predicted a fundamental limit of $\sim 30\%$ for the power conversion efficiency of single-junction solar cells. Their calculation is based on a simple model, in which the excess energy of absorbed photons is assumed to dissipate as heat. This is a good assumption for most conventional semiconductors, whereas for low-dimensional nanostructures with strong Coulomb interaction, the excess energy can be exploited to generate additional electron-hole pairs. In such structures with highly efficient Auger processes, the Shockley–Queisser limit can be exceeded up to $60\%^2$. Furthermore, the gained multiple electron-hole pairs can also increase the sensitivity of photodetectors and other optoelectronic devices.

Carrier multiplication (CM) was first theoretically predicted³ and measured⁴ for semiconductor quantum dots exhibiting a strong spatial confinement in all three directions. Multiplication of photo-excited carriers has also been found in one-dimensional nanostructures, such as carbon nanotubes^{5,6}. In graphene, the carrier confinement combined with the linear bandstructure was predicted to account for highly efficient Auger processes⁷ giving rise to a considerable CM^{8,9}. Just recently, these predictions were confirmed by Brida *et al.* within a joint experiment–theory study also including new insights into collinear scattering in graphene¹⁰, which has been controversially discussed in the literature. However, to realize graphene-based photovoltaic devices, the key problem of charge carrier extraction in gapless nanostructures needs to be solved. Here we suggest a strategy based on Landau quantization of graphene to address this substantial challenge, where a magnetic field is used to induce gaps between discrete Landau levels. Graphene in high magnetic fields has already attracted an immense interest in recent years^{11–14}. Unlike an ordinary two-dimensional electron gas, it exhibits a unique Landau level structure, where the energy is proportional to the square root of the Landau level index $n=0, \pm 1, \pm 2, \dots$ ($E_n \propto \pm \sqrt{|n|}$), cf. Fig. 1. This is a direct consequence of the linear density of states in the low-energy region and results in an anomalous quantum Hall effect^{11,12}.

In this article, we predict the occurrence of CM in Landau-quantized graphene, where the energy gap between the Landau levels is of advantage for the extraction of gained charge carriers. The magnetic field can be tuned so that the inter-Landau level spacings do not fit the energy of the most relevant optical phonons resulting in a strongly suppressed carrier-phonon scattering. Although at first sight the non-equidistant Landau level spectrum seems to suppress Auger processes, a number of energetically degenerate inter-Landau level transitions exists providing the basis for efficient Auger scattering, cf. Fig. 1. This allows to exploit the advantage of graphene having a strong Coulomb interaction while the acoustic phonon coupling is weak in comparison to an ordinary two-dimensional electron gas in a semiconductor heterostructure. As a result, graphene under Landau quantization presents optimal conditions for the appearance of a significant carrier multiplication.

Results

Theoretical approach. While the carrier dynamics in graphene has been intensively investigated over the last few years^{15–19}, there has been only a single study in the presence of a strong magnetic field by Plochocka *et al.*²⁰ In the latter study, the relaxation dynamics between higher Landau levels ($n \approx 100$) is investigated in a differential transmission experiment revealing the importance of Auger scattering in a magnetic field and an overall suppression of scattering rates in comparison to the no-field case. Based on the density matrix formalism^{21,22}, we perform time-resolved microscopic calculations of the ultrafast carrier

Figure 1 | Auger scattering in Landau-quantized graphene. Sketch of the nine energetically lowest Landau levels of graphene with the Dirac cone in the background. The optical excitation via a linearly polarized laser pulse induces the transitions $-4 \rightarrow +3$ and $-3 \rightarrow +4$, cf. the yellow arrows. The energy-conserving process of IE involves the inter-Landau level transition $+4 \rightarrow +1$, which provides the energy to excite an electron from $n=0$ to $n=+1$, resulting in an increase of the number of charge carriers, cf. the red arrows. The inverse process of AR is shown by the green arrows.

relaxation dynamics within the energetically lowest Landau levels. We exploit a correlation expansion in second order Born–Markov approximation^{21,22} to derive Bloch equations for graphene under Landau quantization:

$$\dot{\rho}_n(t) = -2 \sum_{n'} \text{Re}[\Omega_{nn'}(t)p_{nn'}(t)] + S_n^{\text{in}}(t)[1 - \rho_n(t)] - S_n^{\text{out}}(t)\rho_n(t), \quad (1)$$

$$\dot{p}_{nn'}(t) = i\Delta\omega_{nn'}p_{nn'}(t) + \Omega_{nn'}(t)[\rho_n(t) - \rho_{n'}(t)] - \frac{\Gamma(t)}{\hbar}p_{nn'}(t). \quad (2)$$

It is a coupled system of differential equations for the carrier occupation probability $\rho_n(t)$ in the Landau level n and the microscopic polarization $p_{nn'}(t)$ being a measure for optical inter-Landau level transitions according to optical selection rules, cf. Fig. 1. The equations explicitly include the carrier–light interaction, all energy conserving electron–electron scattering processes, as well as the coupling with most relevant optical phonons. The carrier–light interaction enters the equations via the Rabi frequency $\Omega_{nn'}(t) = \frac{e_0}{m_0} \mathbf{M}_{nn'} \cdot \mathbf{A}(t)$ with the elementary charge e_0 , the electron mass m_0 , the optical matrix element $\mathbf{M}_{nn'}$ and the vector potential $\mathbf{A}(t)$. The energy difference $\Delta\omega_{nn'} = (\epsilon_n - \epsilon_{n'})/\hbar$ between the involved Landau levels defines the resonance condition. The magnetic field is incorporated into the equations by exploiting the Peierls substitution (see Supplementary Note 1), accounting for the change of electron momentum induced by the confinement into cyclotron orbits^{14,23}. The appearing in- and out-scattering rates $S_n^{\text{in/out}}$ describe Coulomb- and phonon-induced many-particle scattering processes. The rates include Pauli-blocking terms and the corresponding matrix elements, cf. Supplementary Notes 2–4 for more details. Owing to the energy mismatch of the optical phonon modes with the inter-Landau level transitions at the assumed magnetic field strength, optical phonon scattering is strongly suppressed. Processes involving acoustic phonons and two-phonon relaxation processes can occur^{24–26}, however, both

are much slower compared with the Coulomb-induced dynamics and are neglected in the following. The scattering processes not only change the carrier occupation $\rho_n(t)$ but also contribute to the dephasing Γ of the microscopic polarization $p_{nm}(t)$. The main contribution of Γ stems from electron-impurity scattering, which also induces a broadening of the Landau levels. This broadening is explicitly calculated in the self-consistent Born approximation following the approach of Ando²⁷, cf. Supplementary Note 5, where the electron-impurity scattering strength is chosen in agreement with previous theoretical studies^{28,29}. The validity of the applied Markov approximation has been tested by performing calculations taking also into account non-Markovian effects. The latter reflect the quantum-mechanical energy-time uncertainty principle giving rise to a weak temporal oscillation of carrier occupations. However, the effect is small and does not qualitatively change the investigated inter-Landau level carrier dynamics, cf. Supplementary Note 6.

In order to account for momentum-dependent dynamical screening, we consider the dielectric function $\epsilon(q, \omega)$ in the random phase approximation following the approach of refs 14,30. While the real part of the dielectric function closely resembles the static limit (cf. refs 14,30), taking a finite energy transfer $\hbar\omega$ into account gives rise to a non-vanishing imaginary part, cf. Supplementary Note 3 for more details. Then, the Fourier transform of the Coulomb potential V_q appearing in the Coulomb matrix element

$$V_{34}^{12} = \sum_q V_q \Gamma_{13}(q) \Gamma_{24}(-q), \Gamma_{ij}(q) = \int dr \Psi_i^*(r) e^{iqr} \Psi_j(r), \quad (3)$$

is substituted by the screened potential $V_q/\epsilon(q, \omega)$. Here, q and $\hbar\omega$ are the momentum and energy transfers of the scattering events and Ψ_i denotes the wave function of a state i . Since in the presence of a magnetic field, the momentum is not a well-defined quantity, the vanishing screening in the long-wavelength limit does not imply a divergence of the Coulomb interaction. Furthermore, the singularities for collinear scattering processes appearing in graphene in the absence of a magnetic field^{10,31} do not occur in Landau-quantized graphene due to the absence of a strictly defined momentum conservation in such a discrete system. Moreover, note that excitonic effects of the Coulomb interaction can be neglected, since the Landau level broadening is generally larger than the Coulomb-induced finite excitonic dispersion, cf. Supplementary Note 3.

Carrier relaxation dynamics. Using tight-binding wave functions, we analytically determine the electronic bandstructure and the matrix elements appearing in the Bloch equations. Then, we have all ingredients at hand to evaluate the equations and obtain a microscopic access to the time-resolved ultrafast relaxation dynamics of optically excited carriers. The initial occupation probabilities $\rho_n(t=0)$ before the optical excitation are given by the Fermi function at room temperature. We apply an external magnetic field of $B=4\text{ T}$ and an optical excitation pulse with a width of 1 ps, an applied pump fluence of $\epsilon_{\text{pf}} = 10^{-2} \mu\text{J cm}^{-2}$, and an energy of $\hbar\omega_L \approx 280 \text{ meV}$ matching the inter-Landau level transitions $-3 \rightarrow +4$ and $-4 \rightarrow +3$, cf. Fig. 1. As illustrated in Fig. 2, the carrier occupation probability ρ_4 increases up to 8% during the optical excitation (blue shaded area), while ρ_{-3} decreases for the same amount (not shown). Interestingly, the build up of occupation in the Landau level $n = +4$ is stopped during the optical excitation and ρ_{+4} starts to slightly decrease, cf. the orange solid line in Fig. 2. This effect can be unambiguously traced back to the Coulomb-induced relaxation process, cf. the orange dashed line in Fig. 2 illustrating the temporal evolution of ρ_{+4} without Coulomb scattering. At the same time, the

occupation in Landau level $n = +1$ that is not driven at all by the optical pulse, increases until saturation is reached. The occupation ρ_1 remains constant if the Coulomb interaction is switched off, cf. the blue dashed line in Fig. 2. This behavior can be understood as following: first, the exciting laser pulse generates a non-equilibrium carrier distribution by transferring electrons from $n = -3$ to $n = +4$ (yellow arrows in Fig. 1). Then, Auger-type processes including impact excitation (IE) and Auger recombination (AR) redistribute the carriers between the equidistant Landau levels $n = +4$, $+1$ and 0 (red and green arrows in Fig. 1). Our calculations clearly reveal that IE is the predominant channel at the beginning of the relaxation dynamics due to the Pauli blocking terms in the scattering rates $S_n^{\text{in/out}}$ in the Bloch equations. During the optical excitation, the occupation of $n = +4$ is enhanced, whereas $n = +1$ contains only few electrons corresponding to the thermal distribution at room temperature. As a result, the electron-electron interaction favors the transition $+4 \rightarrow +1$ over the inverse process and provides the energy to excite an additional electron into $n = +1$ from the energetically equidistant zeroth Landau level. This explains the observed increase of ρ_1 and the decrease of ρ_4 , cf. Fig. 2. Interestingly, the occupation ρ_0 does not change and remains 0.5 during the entire relaxation dynamics (not shown). This is related to the peculiarity of the zeroth Landau level being half valence half conduction band¹⁴. Owing to the symmetry between electrons and holes in the considered undoped graphene, each transition evolving electrons can also occur with holes with exactly the same probability. As a result, the electron transition $0 \rightarrow +1$ is always accompanied by a hole transition $0 \rightarrow -1$ corresponding to the excitation of an electron into the zero Landau level. This results in an unconventional IE, where effectively only one charge carrier is created in every scattering event. The result is a constant ρ_0 , while ρ_4 decreases and ρ_1 increases, just as our theory predicts.

Carrier multiplication. To examine the total impact of Auger scattering processes, we investigate the temporal evolution of the carrier density that is composed of electrons in the conduction

Figure 2 | Landau level occupations. Temporal evolution of the occupation probability $\rho_4(t)$ involved in the optical excitation characterized by an ultrashort pulse with a width of 1 ps and $\rho_1(t)$ reflecting the importance of Auger processes in the presence of a magnetic field with $B=4\text{ T}$. The dashed lines illustrate the dynamics in the absence of Coulomb scattering. The yellow shaded region denotes the width of the excitation pulse. The occupation $\rho_2(t)$ is not involved in the relaxation dynamics, $\rho_0(t)$ remains constant and $\rho_3(t)$ shows an increase during pumping just like $\rho_4(t)$. The study is performed for undoped graphene at room temperature.

Figure 3 | Carrier density. Temporal evolution of the carrier density $\mathcal{N}$ including the thermal equilibrium distribution $\mathcal{N}_{\text{eq}}$, and the optically excited carrier density $\mathcal{N}_{\text{opt}}$. The Auger-induced carrier dynamics results in a CM of ≈ 1.3 at the considered pump fluence of $10^{-2} \mu\text{J cm}^{-2}$.

and holes in the valence band Landau levels. The electron-hole symmetry allows a convenient definition of the carrier density $\mathcal{N} = 2(N_{\text{deg}}/A)\sum_{n \geq 1}\rho_n$ with the factor 2 accounting for the electron-hole symmetry, the total degeneracy of each Landau level N_{deg} and the area A . Under the investigated conditions, the zeroth Landau level, being equally divided between both bands, has a temporally constant occupation and does not contribute. Owing to the finite temperature, the initial value of the carrier density is non-zero and we start with an equilibrium value $\mathcal{N}_{\text{eq}}$ (yellow line in Fig. 3). The optical pulse excites carriers and therewith leads to an ascent of the carrier density on a picosecond time scale. If no many-particle interactions are taken into account, $\mathcal{N}_{\text{opt}}$ reaches a value of $\sim 1.8 \times 10^{11} \text{ cm}^{-2}$, cf. the orange line in Fig. 3. The full dynamics including Coulomb-induced carrier scattering reveals a further increase of the carrier density up to $2.3 \times 10^{11} \text{ cm}^{-2}$, reflecting the appearance of a significant carrier multiplication. The CM factor is defined by

$$\text{CM} = \frac{\mathcal{N} - \mathcal{N}_{\text{eq}}}{\mathcal{N}_{\text{opt}} - \mathcal{N}_{\text{eq}}}, \quad (4)$$

and corresponds to 1.3 at the given characteristics of the excitation pulse. It is a direct consequence of the IE, where more carriers are generated in $n = +1$ than are lost in $n = +4$. This is apparent when comparing the drop-off in ρ_4 (5%) with the increase of ρ_1 (10%) after two picoseconds in Fig. 2.

To further prove the occurrence of CM as direct implication of an efficient IE, we determine the time-resolved scattering rates $\gamma_{\text{AR/IE}}(t) = 1/A \sum_i \dot{\rho}_i(t)|_{\text{AR/IE}}$ of the two competing Auger channels. Our calculations show a much more efficient IE as a consequence of the non-equilibrium distribution created by the optical excitation, cf. Fig. 4. The ongoing IE during and after the excitation leads to a more efficient AR until both rates are equal after roughly 2 ps. This is the time frame, in which CM takes place, cf. Fig. 3. Its value corresponds to the area between both curves, which is ultimately defined by the asymmetry between the two competing processes. Their efficiency as Coulomb channels strongly depends on the pump fluence determining the number of available scattering partners. Furthermore, the electronic temperature and the strength of the magnetic field have an influence on the predicted asymmetry between IE and AR.

Discussion

Addressing the optimization of CM, we investigate its dependence on the externally controllable parameters, such as pump

Figure 4 | Coulomb scattering rates. Scattering rates $\gamma_{\text{IE/AR}}$ for the IE and AR. The yellow area represents the excitation region. The blue shaded area illustrates the asymmetry region in which IE prevails over AR resulting in the occurrence of CM.

fluence, temperature and magnetic field, cf. Fig. 5. Although the initial asymmetry between IE and AR is larger at enhanced pump fluences, it also leads to a faster equilibration of both rates leaving a shorter time frame for CM to occur⁹. As a result, the CM decreases with the pump fluence until a saturation of pumping at higher fluences is reached, as illustrated in Fig. 5a. Then, CM starts to slightly increase again reflecting the inefficient optical pumping at saturation conditions. The dependence on temperature is governed by occupation effects. A higher temperature results in less-efficient IE, since an increased initial occupation of $n = +1$ lowers the efficiency of the electronic transition $0 \rightarrow +1$. The qualitative temperature trend resembles a Fermi distribution (cf. Fig. 5b) resulting in the largest CM for temperatures lower than 200 K. Finally, the dependence on the magnetic field is more complex as it is a combination of both effects discussed above, namely the excitation strength and occupation effects. Increasing the magnetic field, the Landau levels (except for $n=0$) shift to higher energies according to the relation $E_n \propto \pm \sqrt{B|n|}$. Therefore, the initial thermal occupation probability ρ_n for $n=1, 2, 3, \dots$ is reduced with increasing magnetic field and IE becomes more efficient. Moreover, the effective excitation strength is determined by the amplitude of the laser pulse and is inverse proportional to its frequency. As the latter increases with the magnetic field, the pumping becomes less efficient giving rise to a more pronounced CM, as already shown in Fig. 5a. We observe a saturation value for the CM of 1.5, cf. Fig. 5c. This value is characteristic for the investigated undoped graphene within the energetically lowest Landau levels. Here IE can only occur once via scattering involving the energetically equidistant Landau levels $n = +4, +1$ and 0. Furthermore, the peculiar nature of the zeroth Landau level being a conduction and valence band at the same time allows only the generation of one additional charge carrier per excitation, as discussed above. Therefore, an optically excited electron-hole pair can yield in total up to three charge carriers resulting in a maximal CM of 1.5. Note that at higher magnetic fields, certain inter-Landau level transitions can become resonant with optical phonon modes creating so called magneto-phonon resonances. This is expected to reduce the observed CM, since the phonons open up a new relaxation channel directly competing with Auger processes, and even the electron-phonon interaction is modified^{32–35}. Therefore, the high-magnetic field region of Fig. 5c should be understood as a trend of the CM as a function of the magnetic field. At very low magnetic fields, the spacing between Landau levels decreases, resulting in an overlapping of levels. Here the

Figure 5 | Optimization of CM. CM dependence on (a) pump fluence, (b) temperature, (c) magnetic field and (d) Landau level broadening. The red lines represent the values used in Figs 2–4.

applied theoretical approach is not valid anymore. Furthermore, note that the CM does not significantly depend on the Landau level broadening, as illustrated in Fig. 5d. On the one hand, the Lorentzian appearing in the scattering rates decreases for larger broadenings. On the other hand, the dielectric function is reduced leading to an enhancement of the Coulomb interaction. Additionally, due to an increased dephasing of $p_{nn'}$, the optical pumping is less efficient. Overall these effects nearly cancel out resulting in a CM that is to a large extent independent of Landau level broadening.

Including energetically higher Landau levels will increase the CM, since the excess energy of a highly excited electron-hole pair can provide enough energy for more than one IE scattering event. In this very first study, we have provided clear evidence of CM in the low-energy limit. Taking into account the findings of Plochocka *et al.*²⁰ that experimentally prove the importance of Auger scattering even for higher Landau levels, we expect a pronounced CM to occur in a broad spectral range. Although they suspect that Auger scattering may be suppressed for lower Landau levels, their findings are indeed compatible with our results. A multi-color pump-probe experiment addressing the carrier dynamics between the discrete low-energy Landau levels should confirm our theoretical predictions in a straightforward way.

In summary, we have gained new fundamental insights into the relaxation dynamics in graphene under Landau quantization, in

particular revealing the appearance of a considerable carrier multiplication that can be traced back to an efficient impact excitation. We have shown that Landau quantization offers a promising strategy to address the key challenge of carrier extraction in a gapless structure, which might be beneficial for the design of graphene-based optoelectronic devices.

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Author contributions

F.W. performed the calculations. All authors analysed and discussed the results. F.W. and E.M. wrote the paper with major input from A.K.

Additional information

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