Best Student Paper Award

# Efficient Auger scattering in Landau-quantized graphene

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## ABSTRACT

We present an analytical expression for the differential transmission of a delta-shaped light field in Landau-quantized graphene. This enables a direct comparison of experimental spectra to theoretical calculations reflecting the carrier dynamics including all relevant scattering channels. In particular, the relation is used to provide evidence for strong Auger scattering in Landau-quantized graphene.

**Keywords:** Landau-quantized graphene, Auger scattering, carrier dynamics, differential transmission spectroscopy

## 1. INTRODUCTION

Since its discovery in 2004,[1] graphene has attracted an enormous amount of interest[2] being considered as a novel material for optoelectronic devices.[3–8] To exploit the fascinating electronic and optical properties of this atomically thin material, it is of crucial importance to understand the non-equilibrium carrier dynamics. There has been a lot of research in this field,[9–27] revealing that carrier-carrier and carrier-phonon scattering dominate the ultrafast relaxation dynamics.[28,29] Remarkably, Auger scattering – which is generally suppressed in ordinary semiconductors – turns out to be of particular importance in graphene, where it gives rise to carrier multiplication occurring at certain experimentally controllable conditions.[23,30–33]

In an external magnetic field, the electronic properties change dramatically, as the energy is quantized into non-equidistant Landau levels.[34] Unlike in the case without a magnetic field, there are only a few studies of the carrier dynamics in Landau-quantized graphene.[35–41] Here, optical selection rules[42] allow the excitation of specific inter-Landau level transitions opening the possibility to address the carrier dynamics of selected levels in a pump-probe experiment. Plochocka et al. have investigated the carrier relaxation in highly energetic Landau levels (with an index $n \sim 100$) which was found to be slowed down by the magnetic field as a consequence of the energy quantization.[35] In a more recent joint experiment theory study, we have investigated the carrier dynamics within the energetically lowest Landau levels.[36] Based on a pump probe experiment and microscopic modeling of the carrier dynamics, we were able to provide evidence for strong Auger-type scattering in Landau-quantized graphene: Including this scattering channel in the theoretical model, an unexpected behavior in the measured differential transmission could be explained.

In this Article, we derive the analytic expression that was used in Ref. [36] to compare the experimental differential transmission spectra (DTS) with the calculations of the Landau level occupations. To this end, we introduce the Bloch equations for Landau-quantized graphene in Section 2, calculate the absorbance for a delta-shaped probe pulse in Section 3, and ultimately derive an analytic expresion for the differential transmission in Section 4.

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## 2. BLOCH EQUATIONS FOR LANDAU-QUANTIZED GRAPHENE

The many-particle Hamilton operator for charge carriers in Landau-quantized graphene subject to an optical light field is given by

$$H = \sum_i \epsilon_i a_i^\dagger a_i + i\hbar \sum_{i,f} \Omega_{if} a_f^\dagger a_i, \tag{1}$$

where the first term is the free energy part that is determined by the Landau level dispersion $\epsilon_n^\lambda = \lambda v_\mathrm{F}\sqrt{2\hbar n e_0 B}$ which depends on the band index $\lambda = \pm 1$, the Fermi velocity $v_\mathrm{F} \approx 1\,\mathrm{nm/fs}$,[43] the Landau level index $n = 0, 1, 2, \ldots$, the elementary charge $e_0$, and the magnetic field $B$. The second term describes the carrier-light coupling expressed through the Rabi frequency $\Omega_{if}(t) = \frac{e_0}{m_0}\mathbf{M}_{if} \cdot \mathbf{A}(t)$ with the free electron mass $m_0$, the vector potential $\mathbf{A}(t)$, and the optical matrix element[38,44]

$$\mathbf{M}_{if} = i\delta_{\xi_i,\xi_f}\delta_{m_i,m_f}\frac{\alpha_{n_i}\alpha_{n_f} m_0 v_\mathrm{F}}{2\sqrt{2}\hbar}\left[\lambda_i \hat{\boldsymbol{\epsilon}}^- \delta_{n_f,n_i-1} + \lambda_f \hat{\boldsymbol{\epsilon}}^+ \delta_{n_f,n_i+1}\right] = -\mathbf{M}_{fi}^*. \tag{2}$$

Here, $\xi = \pm 1$ is the valley index that distinguishes the two equivalent Dirac cones of graphene.[34] Furthermore, $m = 0, 1, \ldots, L^2 e_0 B/(2\pi\hbar) - 1$ is a quantum number expressing the Landau level degeneracy with the area of graphene $L^2$,[34] $\alpha_{n=0}$ is a constant that can take the values $\alpha_{n=0} = \sqrt{2}$ and $\alpha_{n\neq 0} = 1$, and $\hat{\boldsymbol{\epsilon}}^\pm = (\hat{\mathbf{e}}_x \mp i\hat{\mathbf{e}}_y)/\sqrt{2}$ are Jones vectors describing left (+) and right (−) circularly polarized light.[45]

The temporal evolution of the microscopic polarization $p_{if}(t) = \langle a_f^\dagger a_i\rangle(t)$ (where $a_j^\dagger$ and $a_j$ are fermionic creation and annihilation operators of the state $j$) is obtained using the Heisenberg equation of motion $i\hbar\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{O}(t) = [\mathcal{O}, H]$ which provides an equation of motion for the operator $\mathcal{O}$.[28,46] Using the Hamilton operator from Eq. 1, this yields

$$\dot{p}_{if}(t) = \left(i\triangle\omega_{if} - \gamma\right) p_{if}(t) - \Omega_{if}^* \left[\rho_f(t) - \rho_i(t)\right], \tag{3}$$

with the energy difference $\hbar\triangle\omega_{if} = \epsilon_f - \epsilon_i$, a phenomenologically introduced dephasing $\gamma$, and the occupation probability $\rho_j = p_{jj}$ in the state $j$. The dephasing is equivalent to a Landau level broadening and can be caused by disorder[47] or, alternatively by interaction with acoustic phonons.[48,49]

## 3. ABSORBANCE OF A DELTA-SHAPED PULSE

For a two-dimensional nanostructure in the linear optics regime, the absorbance reads[26,28,29]

$$\alpha^\pm(\omega) = \frac{\omega \mathrm{Im}\chi^\pm(\omega)}{c\left|1 + i\omega\chi^\pm(\omega)/2c\right|^2}, \tag{4}$$

where $c$ is the light velocity and

$$\chi^\pm(\omega) = \frac{j^\pm(\omega)}{\epsilon_0 \omega^2 A^\pm(\omega)} \tag{5}$$

is the linear optical susceptibility, with the current density $j$, and the permittivity $\epsilon_0$. The superscript $\pm$ denotes the $\sigma^\pm$-circularly polarized components of the respective quantity in the Jones vector basis ($\hat{\boldsymbol{\epsilon}}^\pm$). The components

$$A^\pm(\omega) = \frac{1}{2\sqrt{\pi}}\left[A_0^+ e^{i(\omega\mp\triangle\omega_{if})\tau} + A_0^- e^{i(\omega\pm\triangle\omega_{if})\tau}\right], \tag{6}$$

of a $\delta$-shaped probe pulse that is in resonance with the transition $i \to f$ is obtained from

$$\mathbf{A}(t) = \delta(t+\tau)\left[A_0^+ \begin{pmatrix} \cos\triangle\omega_{if}t \\ \sin\triangle\omega_{if}t \end{pmatrix} + A_0^- \begin{pmatrix} \cos\triangle\omega_{if}t \\ -\sin\triangle\omega_{if}t \end{pmatrix}\right], \tag{7}$$

where $\tau$ describes the temporal delay of the probe pulse, and $A_0^{\pm}$ are the amplitudes of the left (+) and right (−) circularly polarized contributions. The current density is given by[28]

$$j^{\pm}(\omega) = -i\frac{\hbar e_0}{2m_0 L^2}\sum_{if}\left[M_{if}^{\pm}p_{if}(\omega) + M_{fi}^{\pm}p_{fi}(\omega)\right]. \tag{8}$$

where a term proportional to $A^{\pm}$ was neglected. Using the substitution $p_{if} = p_{if}^{\mathrm{rot}}e^{i\triangle\omega_{if}t}$, we transform Eq. 3 to the rotating frame

$$\dot{p}_{if}^{\mathrm{rot}}(t) = -\gamma p_{if}^{\mathrm{rot}}(t) - \Omega_{if}^{*}\left[\rho_f(t) - \rho_i(t)\right]e^{-i\triangle\omega_{if}t}, \tag{9}$$

and obtain its Fourier transform (FT) exploiting the relation $\mathrm{FT}\left[\dot{p}_{if}^{\mathrm{rot}}(t)\right] = i\omega p_{if}^{\mathrm{rot}}(\omega)$

$$p_{if}^{\mathrm{rot}}(\omega) = \delta_{\xi_i,\xi_f}\delta_{m_i,m_f}\frac{e_0 v_{\mathrm{F}}}{4\hbar}\alpha_{n_i}\alpha_{n_f}\left(\lambda_i\delta_{n_f,n_i-1}A_0^{-} + \lambda_f\delta_{n_f,n_i+1}A_0^{+}\right)\frac{\mathrm{FT}\left[\delta\left(t-\tau\right)\left(\rho_f-\rho_i\right)\right](\omega)}{\omega - i\gamma}. \tag{10}$$

Here, the rotating wave approximation (RWA) was applied by omitting terms $e^{\pm i2\triangle\omega_{if}t}$ in comparison to 1. Since the symmetry relation $p_{if}^{*} = p_{fi}$ effects an inverse rotation of $p_{fi}$ compared to $p_{if}$, cf. $p_{fi} = p_{fi}^{\mathrm{rot}}e^{-i\triangle\omega_{if}t}$, the RWA for $p_{fi}^{\mathrm{rot}}(\omega)$ is not obtained by exchanging $i \leftrightarrow f$ in Eq. 10, but the relation $p_{fi}^{\mathrm{rot}}(\omega) = -p_{if}^{\mathrm{rot}}(\omega)$ must be used instead. Exploiting the convolution theorem, we can write

$$\begin{aligned}\mathrm{FT}\left[\delta\left(t+\tau\right)\left(\rho_f-\rho_i\right)\right] &= \mathrm{FT}\left[\delta\left(t-\tau\right)\right] * \mathrm{FT}\left[\rho_f-\rho_i\right] \\ &= \frac{1}{2\pi}e^{i\omega\tau}\left[\rho_f(\tau)-\rho_i(\tau)\right].\end{aligned} \tag{11}$$

where $*$ denotes the convolution of two functions $f$ and $g$: $(f*g)(\omega) = \int_{-\infty}^{\infty}\mathrm{d}\omega' f(\omega')g(\omega-\omega')$, Using Eqs. 2, 8, 10, 11, as well as the relations $p_{if}(\omega) = p_{if}^{\mathrm{rot}}(\omega-\triangle\omega_{if})$ and $p_{fi}(\omega) = -p_{if}^{\mathrm{rot}}(\omega+\triangle\omega_{if})$, the current density reads

$$j^{\pm}(\omega) = \frac{e_0^3 v_{\mathrm{F}}^2 B}{64\pi^2\hbar^2}\sum_{if}\left(\alpha_{n_i}\alpha_{n_f}\right)^2\left[\rho_f(\tau)-\rho_i(\tau)\right]\left[A_0^{\pm}\delta_{n_f,n_i\pm1}\frac{e^{i(\omega-\Delta\omega_{if})\tau}}{\omega-\Delta\omega_{if}-i\gamma_{if}^{\mathrm{imp}}} - A_0^{\mp}\delta_{n_f,n_i\mp1}\frac{e^{i(\omega+\Delta\omega_{if})\tau}}{\omega+\Delta\omega_{if}-i\gamma_{if}^{\mathrm{imp}}}\right] \tag{12}$$

Here, we performed the summations over the valley and $m$ degrees of freedom. In due consideration of the Kronecker deltas $\delta_{\xi_i,\xi_f}\delta_{m_i,m_f}$ in Eq. 10 this yields a factor of $L^2 e_0 B/(\pi\hbar)$. Omitting terms with a non vanishing rotation $e^{i\tilde{\omega}\tau} \neq 1$, we obtain the linear optical susceptibility (Eq. 5) using the Eqs. 6 and 12

$$\chi^{\pm}(\omega) = \frac{\sqrt{\pi}e_0^3 v_{\mathrm{F}}^2 B}{32\pi^2\hbar^2\epsilon_0\omega^2}\sum_{if}\left(\alpha_{n_i}\alpha_{n_f}\right)^2\frac{\rho_f(\tau)-\rho_i(\tau)}{\omega-\Delta\omega_{if}-i\gamma_{if}^{\mathrm{imp}}}\delta_{n_f,n_i\pm1}. \tag{13}$$

Neglecting the term $i\omega\chi(\omega)/2c$ in the denominator of Eq. 4 which is usually small, we find the final expression for the absorbance of a delta-shaped pulse

$$\alpha^{\pm}(\omega) = \frac{\sqrt{\pi}e_0^3 v_{\mathrm{F}}^2 B}{32\pi^2\hbar^2\epsilon_0 c\omega}\sum_{if}\left(\alpha_{n_i}\alpha_{n_f}\right)^2\left[\rho_f(\tau)-\rho_i(\tau)\right]\frac{\gamma}{\left(\omega-\Delta\omega_{if}\right)^2+\gamma^2}\delta_{n_f,n_i\pm1}. \tag{14}$$

## 4. DIFFERENTIAL TRANSMISSION

The idea of the differential transmission spectroscopy is to optically excite the system under investigation using a strong pump pulse. Then, the differential transmission of a much weaker probe pulse incident after a certain time delay $\tau$ is measured. Based on the assumption that the occupations of the system are not affected by the weak probe pulse, the differential transmission allows to draw conclusions about the carrier dynamics in the system. Omitting possible interference between both pulses, which can lead to the emergence of a so-called coherent artifact, the ansatz

$$\mathrm{DTS}(\omega,\tau) = \alpha_{\mathrm{pump}}(\omega,\tau) - \alpha_0(\omega) \tag{15}$$

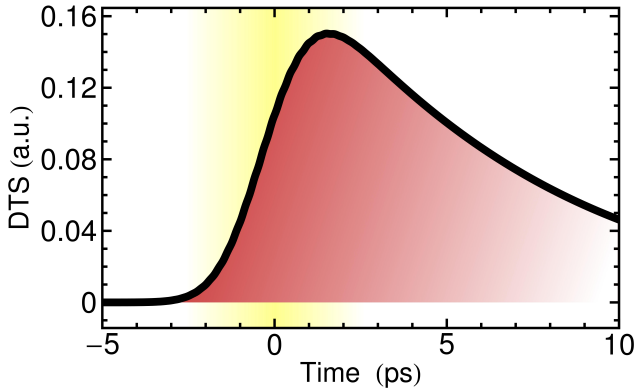


Figure 1. Differential transmission spectrum pumping and probing with linear polarized radiation and an energy matching the inter-Landau level transitions $\mathrm{LL}_0 \to \mathrm{LL}_{+1}$ and $\mathrm{LL}_{-1} \to \mathrm{LL}_0$ at $B = 4\,\mathrm{T}$. The yellow area in the background represents the pump pulse centered around $t = 0$.

for the differential transmission is used, where $\alpha_{\mathrm{pump}}$ ($\alpha_0$) is the absorbance of the probe pulse with (without) a previous pump pulse. Assuming a pump pulse centered around $t = 0$, the initial absorbance of the probe pulse is given by $\alpha_0(\omega) = \alpha(\omega, \tau = -\infty)$, while the absorbance of the probe pulse following the pump pulse after a time $\tau$ is simply given by $\alpha_{\mathrm{pump}}(\omega, \tau) = \alpha(\omega, \tau)$. Thereby, using Eq. 14, the DTS for a single transition $i \to f$ and a resonant excitation ($\omega = \Delta\omega_{if}$) reads

$$\mathrm{DTS}^{\pm}_{if}(\tau) = \frac{\sqrt{\pi} e_0^2 v_{\mathrm{F}}^2}{32\pi^2 \hbar \epsilon_0 c l_B^2 \Delta\omega_{if} \gamma} \left(\alpha_{n_i} \alpha_{n_f}\right)^2 \left[\Delta\rho_f(\tau) - \Delta\rho_i(\tau)\right] \delta_{n_f, n_i \pm 1}, \tag{16}$$

where $\Delta\rho_j(\tau) = \rho_j(\tau) - \rho_j(t = -\infty)$ is the occupation difference induced by the pump pulse. Note that (in RWA) $\sigma^+$-polarized radiation is only capable of exciting transitions where the Landau level index is incremented by one $n \to n + 1$, while transitions with $n \to n - 1$ require $\sigma^-$-polarized radiation (cf. $\delta_{n_f, n_i \pm 1}$ in Eq. 16, respectively). When more than one transition is addressed by the probe pulse at the same time, the DTS is given by the sum of the individual spectra (cf. sum over $i$ and $f$ in Eq. 14). Therefore, the DTS pumping and probing the inter-Landau level transitions $\mathrm{LL}_0 \to \mathrm{LL}_{+1}$ and $\mathrm{LL}_{-1} \to \mathrm{LL}_0$ is given by $\mathrm{DTS}^{+}_{0\to+1}(\tau) + \mathrm{DTS}^{-}_{-1\to0}(\tau)$ which is proportional to $(\Delta\rho_{+1} - \Delta\rho_{-1})$. An example of such a calculated DTS for a linear polarized excitation of the energetically lowest Landau levels is shown in Fig. 1, where an increase during the optical excitation reflects the enhanced Pauli blocking induced by the pumping (absorption bleaching), and the decay results from the many-particle-induced relaxation back to the equilibrium distribution. Investigating the decay behavior (mono- or multi-exponential) and the decay rate allows conclusions on the relaxation channels and their efficiency.

## 5. CONCLUSION

We have presented the derivation of the relation between the differential transmission accessible in pump-probe experiments and the occupation probabilities of Landau-quantized graphene. The obtained analytic expression shows that the differential transmission is proportional to the difference between the pump-induced occupation change in the final and the initial state.

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