Length-dependence of light-induced currents in graphene

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Abstract
We investigate the transport of optically injected currents in graphene, a (semi-) metal with exceptional optical and electronic properties. We have recently shown that ultrashort laser pulses with low temporal symmetry drive coupled intraband motion and interband (Landau–Zener) transitions resulting in residual ballistic currents in graphene. Here we show experimentally how this current scales as a function of the distance between the light-induced current injection region and the adjacent metal contact electrodes and propose an approach to model the results based on diffusive and field driven charge transport. We expect this study to contribute to ongoing discussions on the propagation of light-field-controlled currents, a requirement for future lightwave electronics, operating at petahertz clock rates.

Keywords: strong field physics, light field electronics, graphene, 2D materials, optoelectronics, charge dynamics

(Some figures may appear in colour only in the online journal)

Graphene-based structures with their unique optical and electric properties represent an ideal platform for light-induced current generation and detection. When asymmetry is introduced to a symmetric system, there is a preferential direction for charge flow of photoexcited carriers. Spatial asymmetry has been applied at material interfaces [1–9], contact electrodes [10, 11] or pn-like junctions [12–14], where electrons and holes are separated.

In contrast, few-cycle laser pulses facilitate the injection and control of currents in solids relying only on a waveform that is asymmetric with respect to time [15]. We consider a Fourier-limited waveform \( E(t) = E_0(t) \cos(\omega t + \varphi_{\text{CEP}}) \) with \( E_0(t) \), a Gaussian envelope, and with a carrier frequency \( f = \frac{\omega}{2\pi} \). Maximal asymmetry is introduced by adjusting the carrier-envelope phase (CEP) to \( \varphi_{\text{CEP}} = \pm \pi/2 \). Recently we have shown that such few-cycle laser pulses with an electric field amplitude larger than 1 V nm\(^{-1}\) drive electrons in graphene fully coherently across the band structure. When an electron approaches the Dirac point, a Landau–Zener transition may occur (figure 1(a)) [16–25]. The repeated competition of intraband motion and interband Landau–Zener transitions during a few-cycle laser pulse may result in a residual asymmetric conduction band population \( \rho_{\text{CB}} \) in graphene, a highly spatial–symmetric system [26–28]. From simulations based on the time-dependent Schrödinger equation we derive, that the asymmetry of this population is maximized for a carrier-envelope phase \( \varphi_{\text{CEP}} = \pm \pi/2 \) and gives rise to a ballistic current in the plane of graphene (figure 1(b)) [29].

For future attosecond-fast electronics it is mandatory to investigate the behavior of the ballistic current and the ensuing dynamics: the transfer of charge from the light–matter
focused down to a $1/31$. The graphene stripes are contacted at their ends to gold is approached, a positive current of a few nA is measured of the structure, see figure 2. When the right ground contact $E$ close to the Dirac point. Under the presence of a strong electric field $E(t)$, electrons undergo intraband motion (solid line) and interband transitions (dashed line), i.e. Landau–Zener transitions. With a carrier envelope phase of $\varphi_{\text{CEP}} = \pi/2$ the asymmetry in residual conduction band population is maximized. (b) We illustrate the difference in conduction band population $\Delta \rho_{\text{CB}} = \rho_{\text{CB}}(\varphi_{\text{CEP}} = \pi/2) - \rho_{\text{CB}}(\varphi_{\text{CEP}} = -\pi/2)$ between x-polarized excitations with the two extrema $\varphi_{\text{CEP}} = \pm \pi/2$ for a peak electric field strength of 2.5 V nm$^{-1}$ ($F_0 = 17$ mJ cm$^{-2}$) as obtained from a time-dependent Schrödinger equation simulation [26]. The population asymmetry with respect to $k_x = 0$ translates into a CEP-dependent ballistic current in $x$-direction.

interaction region toward electrodes required for the detection and processing of currents. Here we show the experimentally measured scaling of such currents in graphene as a function of the electrode distance.

In the experiment, we detect light-field-induced currents in graphene generated by near-infrared CEP-stable laser pulses from a titanium–sapphire oscillator with a pulse duration of 5.5 fs (2.1 cycles at 800 nm) and a repetition rate of 80 MHz. Using epitaxially grown graphene on silicon carbide (SiC), graphene stripes with virtually no restrictions in size can be fabricated via e-beam lithography and plasma etching. Here we pattern stripes with a width of 2 $\mu$m and various lengths from 4 $\mu$m to 13 $\mu$m via e-beam lithography and plasma etching [30]. Due to the substrate coupling the graphene is n-doped with a Fermi energy of 300 meV above the Dirac energy [30, 31]. The graphene stripes are contacted at their ends to gold electrodes with a titanium adhesive layer. The laser pulses are focused down to a $1/e^2$ intensity beam radius of $w_0 = 1.5$ $\mu$m on the graphene stripe with linear polarization parallel to the stripe.

By scanning the focus across the structure with a weak average power of 1 mW (fluence of 0.3 mJ cm$^{-2}$), the total photocurrent reveals its contact potential landscape at the contact electrodes [10, 11], which we use hereinafter for alignment of the structure, see figure 2. When the right ground contact is approached, a positive current of a few nA is measured indicating that electrons are attracted by the higher potential of the adjacent gold contact and are thus extracted from the device. Excited electrons are equally attracted by the built-in potential toward the other gold contact when the opposite interface is illuminated. The measured current thus reverses its sign. From this behavior we infer that the work function of the titanium/gold contacts is lower than that of the graphene stripe, which leads to the observed directional current [11].

For all measurements of CEP-dependent currents only, the center of each structure is illuminated to suppress the photocurrents induced by the discussed spatial symmetry breaking. We use a lock-in detection scheme with the carrier-envelope offset frequency as a reference and the first harmonic of the current response is detected [26–28].

In figure 3 (a), the CEP-dependent current is shown as a function of the electrode distance $L$ for a peak laser fluence of 10 mJ cm$^{-2}$, 17 mJ cm$^{-2}$ and 20 mJ cm$^{-2}$. These fluences correspond to peak electric field strengths of 2.0 V nm$^{-1}$, 2.5 V nm$^{-1}$ and 2.8 V nm$^{-1}$, respectively. Note that the dielectric reduction of the field strength is already taken into account here [26]. We do not illuminate the contacts to avoid plasmonic effects due to excitation of free electrons in the metal [7]. Clearly, increasing $L$ results in a steep decrease of the current for all shown electric field strengths. It is remarkable that for the maximum measured distance of $L = 13$ $\mu$m and $E_0 = 2.5$ V nm$^{-1}$ we still measure a current as large as $(5 \pm 2)$ pA. As shown in figure 3 (b) the current as a function of $L$ follows approximately a second-order power law.

Another degree of freedom for symmetry breaking is obtained by changing the direction of the polarization. In figure 3 (c) we show measurement results of the CEP-dependent current as a function of the polarization angle $\theta$ (see figure 2), rotated by a half-wave plate. Light pulses with a peak electric field strength of 3.1 V nm$^{-1}$ are focused on a 7 $\mu$m long graphene stripe. Under parallel polarization ($\theta = 0$) we measure the same current as for anti-parallel polarization ($\theta = \pm \pi$) except for a reversed sign. Here the wave plate imposes a $\pi$ shift in the CEP which effectively mirrors

![Figure 1](image1.png)

Figure 1. Current generation and current transport in graphene. (a) Schematic illustration of graphene’s cone-shaped band structure close to the Dirac point. Under the presence of a strong electric field $E(t)$, electrons undergo intraband motion (solid line) and interband transitions (dashed line), i.e. Landau–Zener transitions. With a carrier envelope phase of $\varphi_{\text{CEP}} = \pi/2$ the asymmetry in residual conduction band population is maximized. (b) We illustrate the difference in conduction band population $\Delta \rho_{\text{CB}} = \rho_{\text{CB}}(\varphi_{\text{CEP}} = \pi/2) - \rho_{\text{CB}}(\varphi_{\text{CEP}} = -\pi/2)$ between x-polarized excitations with the two extrema $\varphi_{\text{CEP}} = \pm \pi/2$ for a peak electric field strength of 2.5 V nm$^{-1}$ ($F_0 = 17$ mJ cm$^{-2}$) as obtained from a time-dependent Schrödinger equation simulation [26]. The population asymmetry with respect to $k_x = 0$ translates into a CEP-dependent ballistic current in $x$-direction.

![Figure 2](image2.png)

Figure 2. Photocurrent map of a $2 \times 10^5 \mu$m$^2$ graphene stripe. Scanning the laser focus (radius of $w_0 = 1.5$ $\mu$m, not shown) over the structure reveals photocurrents resulting from the excitation of charge carriers in graphene. The current is measured with an optical chopper as a reference for a lock-in amplifier. Due to a mismatch of the work-functions of graphene and the gold electrodes, a predominant photocurrent is measured when the interfaces are illuminated. All CEP-dependent current measurements are performed in the center, marked by a star, using polarized light as indicated by the dark red double arrow. For the measurement of current as a function of the polarization direction (shown in figure 3 (c)), the polarization is rotated by an angle $\theta$, while the focus position and $w_0$ are kept constant (light red double arrow).
the waveform with respect to the electrodes. Clearly, when the polarization is perpendicular to the graphene stripe (\(\theta = \pm \pi/2\)), spatial symmetry is preserved with respect to the electrodes and the measured current is zero. From the sinusoidal modulation in between we derive that the current measured at the electrodes predominantly depends on the projection of the direction of the initial ballistic current injection.

The following mechanisms will play a role for the charge transport toward the electrodes and are essential for understanding the observed length dependence of photocurrent: initially ballistically injected carriers thermalize and lose their predominant momentum within 10–80 fs due to intraband scattering \[32, 33\]. Since the electron group velocity in graphene is the Fermi velocity of 1 nm fs\(^{-1}\) at maximum \[34\], ballistic transport vanishes on a length scale below 100 nm. The initial ballistic kick leaves electron and hole distributions behind to which diffusive and field driven currents respond in the following \[35\] until all excited charge carriers recombine due to interband scattering on time scales of 1–3 ps \[36–38\]. Those electrons and holes that reach the contacts constitute the DC current measured in our quasi-static experiment. Those electron–hole pairs that recombine with each other inside the sample only create a transient alternating current due to their intermediate charge displacement. The DC current reflects a net excess of electrons versus holes reaching the one contact and an equal net excess of holes versus electrons reaching the other contact.

A scheme for modeling the observed length-dependence of the CEP-dependent current incorporating the above sketched mechanisms on the basis of electron and hole mobilities, their diffusion constants, recombination rates and momentum relaxation times is certainly beyond the scope of this work and remains a future task.

Previous studies on length-dependent current measurements in fused silica, an insulator, could serve as a comparison \[39\]. Here, a power law-dependence with an exponent of \((-1.70 \pm 0.68)\) was found. In this case, too, the mechanism for charge transfer has not been finally clarified, but due to the insulating property of fused silica a dipole mediation of ballistic current was proposed. Still, we believe that the experimental data presented will help future work to uncover transport behavior in this intriguing system. In particular, clarifying the roles of diffusive and field mediated charge transport will be insightful, for fused silica, an insulator \[39, 40\], semiconductors \[41\], and for graphene, a semi-metal.

In summary, we have shown the length-dependence of light-field-injected currents and propose a power law-scaling to the order of \((-2.20 \pm 0.05)\) as a function of the electrode distance. Interestingly, this in good agreement with length-scaling of previously reported light-injected currents in fused silica \[39\]. By changing the polarization direction we demonstrated that the current measured at the electrodes directly depends on the direction of initial ballistic current injection. A quantitative model on charge transport has yet to be developed to fully describe our experimental observations.

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