Ultrafast intrinsic optical-to-electrical conversion dynamics in a graphene photodetector

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Optical-to-electrical conversion in graphene is a central phenomenon for realizing anticipated ultrafast and low-power-consumption information technologies. However, revealing its mechanism and intrinsic timescale require uncharted terahertz electronics and device architectures. Here we succeeded in resolving optical-to-electrical conversion processes in high-quality graphene via the on-chip electrical readout of an ultrafast photothermoelectric current. By suppressing the time constant of a resistor–capacitor circuit using a resistive zinc oxide top gate, we constructed a gate-tunable graphene photodetector with a bandwidth of up to 220 GHz. Measuring the non-local photocurrent dynamics, we found that the photocurrent extraction from the electrode is quasi-instantaneous without a measurable carrier transit time across several-micrometre-long graphene, following the Shockley–Ramo theorem. The time for photocurrent generation is exceptionally tunable from immediate to >4 ps, and its origin is identified as Fermi-level-dependent intraband carrier–carrier scattering. Our results bridge the gap between ultrafast optical science and device engineering, accelerating ultrafast graphene optoelectronic applications.

For a comprehensive understanding of the O–E conversion mechanism in graphene, we thoroughly investigate non-local photocurrent dynamics while tuning the Fermi energy of several graphene samples with different carrier mobilities and channel lengths that are encapsulated in hexagonal boron nitride (hBN). After photoexcitation, O–E conversion proceeds via four stages: (1) non-equilibrium photoexcited carriers thermalize via intraband relaxation, (2) the photocarrier temperature increases on a picosecond timescale through various phonon interactions, (3) the measured photocurrent directly tracks the development and evolution of the carrier temperature, enabling us to resolve multiple thermalization and cooling pathways that coexist in graphene, and (4) the photocurrent decays via carrier cooling and a sample with a lower mobility shows a faster decay due to supercollision (SC) cooling. Owing to the negligible RC time constant in our devices and the Shockley–Ramo-type response, the measured photocurrent directly tracks the development and evolution of the carrier temperature, enabling us to resolve multiple thermalization and cooling pathways that coexist in graphene.

The quantitative understanding of the above O–E conversion processes now serves as the basis for designing ultrafast graphene optoelectronic devices.

We first present our experimental set-up for ultrafast electrical readout. Then we discuss the time for photovoltage generation and carrier transport in graphene PDs through measuring the non-local photocurrent dynamics. Next, we describe the mechanism of photocurrent decay. Finally, we discuss the difference between our electrical readout and the optoelectrical readout, which is commonly used to deduce the timescale of graphene PDs.

Experimental set-up

The basic concept of our experimental set-up is illustrated in Fig. 1a. We made a high-quality single-layer-graphene transistor via hBN encapsulation with a ZnO top gate and titanium/gold contacts.

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We carried out pump–probe experiments using a pulsed femtosecond laser with a pulse duration of 280 fs. This beam was divided into pump and probe beams to excite the graphene and PC switch. Note that the Goubau line is suitable for investigating one of the Ti/Au electrodes that forms a Goubau-line waveguide (LT-GaAs) PC switch. We fabricated four samples with different graphene channel lengths ($L=5 \mu m$ (two samples), $10 \mu m$ and $15 \mu m$) and carrier mobilities ($\mu=11,000–140,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) (Supplementary Section 2a). We refer to the four samples by their channel length, that is, samples $5 \mu m(1)$, $5 \mu m(2)$, $10 \mu m$ and $15 \mu m$. As shown in the inset to Fig. 1a, one of the Ti/Au electrodes that forms a Goubau-line waveguide is connected to a low-temperature-grown gallium arsenide (LT-GaAs) PC switch. Note that the Goubau line is suitable for investigating O–E conversion processes without the complexity of mode interference and mode coupling between graphene and the waveguide.

We carried out pump–probe experiments using a pulsed femtosecond laser with a pulse duration of 280 fs. This beam was divided into pump and probe beams to excite the graphene and PC switch, respectively, with a controlled time delay to measure the photocurrent in the time domain. The pump beam was tightly focused on the graphene using an objective lens to perform scanning photocurrent microscopy. We selected laser wavelength ($\lambda_{\text{lin}}$) values of $517 \text{ nm}$, for better focus and a higher signal-to-noise ratio, and $1,035 \text{ nm}$, for accurate Fermi-level tuning to avoid unwanted photo-induced doping. Figure 1b,c shows an optical image of sample $5 \mu m(1)$ and the corresponding scanning photocurrent image at zero source–drain bias voltage ($V_{\text{ds}}$), respectively. The photocurrent is maximized with opposite signs at the two graphene–metal interfaces, which is the signature of a PTE current (see Methods for details). Throughout the experiments, we kept $V_{\text{gs}}=0 \text{ V}$ to focus on the O–E conversion via the PTE effect. Figure 1d shows time-resolved photocurrent for sample $5 \mu m(1)$ ($\mu=11,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and sample $5 \mu m(2)$ ($\mu=140,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$). The Fourier transform shows that the 3 dB bandwidth of sample $5 \mu m(1)$ reaches $220 \text{ GHz}$ (Fig. 1e), demonstrating that the cut-off frequency by the RC time constant is higher than $220 \text{ GHz}$, as well as the capability of our system for investigating the ultrafast dynamics of O–E conversion processes. On the other hand, the bandwidth of sample $5 \mu m(2)$, whose carrier mobility is an order of magnitude higher than that of sample $5 \mu m(1)$, is as narrow as $51 \text{ GHz}$. This suggests that the carrier mobility has a strong impact on the bandwidth. In the following, we investigate the intrinsic mechanism behind the ultrafast O–E conversion via a comprehensive study, including its dependence on the Fermi level, mobility, source–drain channel length and pump-spot position.

**Experimental results**

**Tunable thermalization and instantaneous photocurrent flow.** First, we focus on the early stages of the O–E conversion, namely the generation of the photocurrent in the sample with $\mu=140,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (L = $5 \mu m$), that is, the one with the highest mobility (sample $5 \mu m(2)$). Figure 2a shows the measured waveform for different gate bias voltage ($V_{\text{gate}}$) values. The peak current varies with $V_{\text{gate}}$ (inset of Fig. 2a) and is maximized around $E_F=-0.07 \text{ eV}$, which is consistent with the $V_{\text{gate}}$ dependence of the Seebeck coefficient, $\sigma \text{ d}F/\text{ d}V_{\text{gate}}$, where $\sigma$ is the graphene conductivity. This non-monotonic behaviour is characteristic of the PTE effect, whereas the photovoltaic effect should exhibit a monotonic increase of the photocurrent with $V_{\text{gate}}$ (ref. 9). Most strikingly, as highlighted in Fig. 2b, the peak position of the photocurrent is shifted to a larger value of $\sim 4 \text{ ps}$ as the Fermi level is tuned towards the charge neutrality point (CNP). Note that the negative photocurrent for the minimal doping is likely to be the recently observed CNP photocurrent (Supplementary Section 3). It is tempting to explain the substantial delay in the photocurrent generation via the carrier transit time in...
Fig. 2 | Instantaneous Shockley–Ramo response and tunable HCM. a, Temporal profiles of the photocurrent in sample 5 μm with μ = 140,000 cm² V⁻¹ s⁻¹ for several values of V_{gate}. Time origin is set at the current peak of V_{gate} = −2.46 V. The inset shows the corresponding peak-current amplitude (black circles) and two-terminal resistance (red curve). b, Normalized waveform of a around the peak position, highlighting the gate-induced peak shift. c, Schematic of experimental configurations (i)–(iii) for measuring the non-local photocurrent extraction dynamics. α is obtained using configuration (i). d, Peak shift extracted from each configuration in c, as shown by the corresponding colour plots. These values show relative delay from |V_{gate} - V_{CNP}| = 2.8 V. Error bars are due to the uncertainty of determining the peak positions. The inset shows the waveforms obtained using sample 10 μm by pumping the (red) and lower (blue) graphene-metal interfaces (V_{gate} = 0 V, λ_{sat} = 517 nm). V_{CNP}, voltage of the charge neutrality point. e, Rise time of the photocurrent as a function of the Fermi level. The blue curve shows the best fit obtained using the HCM model with the adjustable scaling constant Α = 7.9 ± 0.3 eV ps. The horizontal dotted line shows the pulse duration of the pump pulse. Inset: a schematic of the HCM process. ΔE is the energy loss per step, with ΔE ≈ |E_F|. Data are presented as fit coefficients with vertical error bars corresponding to the estimated standard deviation. Horizontal error bars are due to the uncertainty of the gate capacitance. Experimental parameters: V_{LS} = 0 V, λ_{sat} = 1,035 nm and P_{laser} = 0.1 mW.

graphene⁶,₁₂⁻, which is assumed to limit the bandwidth of graphene PDs. However, we verify below that the delay in the photocurrent generation is due to the slow carrier thermalization time near the CNP and not to the carrier transit time in graphene.

To uncover the cause of the delay, we evaluate the non-local photocurrent extraction dynamics by exciting different positions (interface to upper or lower contact) and samples with different lengths and mobilities, as shown schematically in Fig. 2c. The corresponding peak shift as a function of V_{gate} in the three configurations falls on a single curve (Fig. 2d). This overlap indicates that the carrier transport in the graphene PD is not the cause of the delay but instead is quasi-instantaneous (less than or comparable to the pump pulse duration of 280 fs), as showcased in the inset of Fig. 2d, in contrast to the naive expectation for the carrier transit time of ~18 ps for L = 10 μm with the saturation velocity of 5.5 × 10⁵ m s⁻¹ (ref. ⁶). This quasi-instantaneous response can be explained by the Shockley–Ramo theorem. In conductive materials like graphene, a photocurrent is immediately generated when ambient carriers enter the contact, whereas photoexcited carriers themselves travel to the contact in a semiconductor. In the former case, the timescale of the photocurrent response is determined by the time for the screening of the long-range electric field. This Shockley–Ramo response time is much shorter than the carrier transit time and is predicted to be around 300 fs for L = 10 μm (ref. ¹⁵). This indicates that the carrier transit time is independent of the bandwidth of a graphene PD, which is highly beneficial for developing ultrafast PDs.

An alternative possible cause of the delay is the time for the thermalization of a Fermi–Dirac distribution with an elevated carrier temperature. In graphene, the energy dissipated to the phonon system does not contribute to the PTE current, since electronic heat capacity is orders of magnitude smaller than the lattice, and thus the rise time of the photocurrent is determined by carrier–carrier scattering⁸⁻¹¹,¹³⁻¹⁵,¹⁸⁻²³. Since interband carrier scattering is minor due to the limited phase space⁶⁻¹¹,¹³⁻¹⁵,¹⁸⁻²³, intraband carrier scattering dominates the thermalization dynamics except when very close to the CNP³⁸ (|E_F| < ~0.03 eV) (Supplementary Section 3). To quantitatively discuss the effects of intraband scattering on the thermalization dynamics, we determined the rise time of the photocurrent via exponential fitting of the rise and decay time as shown in Fig. 2e (see Supplementary Section 4 for the detailed procedure). Above |E_F| > 0.12 eV, the rise time is determined by the duration of the pump pulse, whereas it increases with decreasing |E_F| and exceeds 4 ps near the CNP. These slower dynamics can be explained by the smaller energy loss (ΔE) per intraband scattering event for smaller |E_F| during the thermalization. Namely, more scattering events are required to thermalize for a smaller |E_F| (inset of Fig. 2e) because the amount of energy exchanged between the photoexcited carriers and carriers in the Fermi sea is ΔE ≈ |E_F| (ref. ¹¹). This cascade step leads to HCM with the number of secondary electron–hole pairs scaled by E_n/|E_F|, where E_n is the excitation energy (E_n/|E_F| > 1). The HCM model¹¹, which suggests that the thermalization time (Δt) is proportional to the inverse square of the Fermi energy, Δt = α(E_n/E_F)/E_n (with α = 7.9 ± 0.3 eV ps), agrees well.
with the experimental results. The tunable range of the rise time is an order of magnitude larger than that observed using intraband absorption of terahertz pulses$^5$ (from immediate to ~0.4 ps). This colossal tunability of the O–E conversion will be useful for ultrafast signal processing. We note that observing the generation of a tunable photocurrent and identifying its origin become possible due to relaxation of the momentum restriction by the disorder ($q_0$). Experimental parameters are $V_{bi}=0$ V, $\lambda_{min}=1.035$ nm and $P_{sers}=0.1$ mW. $k_B$, Boltzmann constant; $T$, temperature; $e^-$, electron.

**Mobility dependence of cooling.** Next, we discuss the photocurrent decay that is induced by lowering the carrier temperature. We note that, because of the PTE effect, the Shockley–Ramo response and the negligible RC time constant, the photocurrent tracks the time evolution of the carrier temperature directly. The responsivity of graphene PDs is known to be enlarged in a high-mobility device$^b$ due to the large Seebeck coefficient$^b$. On the other hand, the mobility effect on the cooling time has not been quantitatively investigated. We show that the mobility is a crucial parameter for adjusting the cooling time through a systematic study that uses four samples with different mobilities at a common substrate and Fermi energy ($E_F=-0.05$ eV) (Fig. 3a). Faster decay is observed for the samples with lower mobility. This behaviour can be explained by the disorder-assisted SC cooling$^{27,40}$, where three-body collisions between carriers and both phonons and impurities take place. As shown in the bottom part of Fig. 3b, the decay time in the samples with a mobility of $\mu < 51,000$ cm$^2$ V$^{-1}$ s$^{-1}$ (sample 15 µm) is reasonably explained using this SC prediction$^{14,27}$ (Supplementary Section 5). The decay time in sample 5 µm with $\mu = 140,000$ cm$^2$ V$^{-1}$ s$^{-1}$ is far below the SC prediction, suggesting that another pathway dominates the carrier cooling for this mobility regime. The $E_F$ dependence of the decay time in this sample (Supplementary Fig. 7) suggests that coupling to hyperbolic phonons in the hBN substrate$^{17}$ dominates for the high-doping regime. In the low-doping regime, where the decay is slow, the effects of the hyperbolic phonons are weak, and the optical phonons may be another major cooling pathway. The Seebeck coefficient calculated using the Mott formula$^{3,45}$ is also shown in the top part of Fig. 3b to estimate the responsivity. We note that the obtained Seebeck coefficient of up to 350 ± 40 $\mu$V K$^{-1}$ is substantially high among various graphene PDs$^{10}$ owing to the high carrier mobility of our device because of hBN encapsulation. Although there is a trade-off between bandwidth and responsivity, the large Seebeck coefficient with the first picosecond decay indicates the superior performance of hBN-encapsulated graphene. These results provide crucial information for tailoring the performance of graphene PDs depending on their intended application.

**Electrical readout versus optoelectrical readout of decay.** Finally, we further demonstrate the advantage of the electrical readout by comparing it with the commonly used optoelectrical readout$^{12,15,17,40-42}$, which is another method used for measuring the response of an ultrafast photocurrent among various femtosecond pump–probe techniques$^{5,10,12-25}$. The two different methods are compared schematically in Fig. 4a. In the optoelectrical readout, graphene is excited using two laser pulses, and the photocurrent decay
is measured as a function of the delay between them. Because the photocurrent scales non-linearly with the pump intensity, the photocurrent decreases in proportion when two pump pulses overlap. Then, the decay constant is monitored as the recovery time of the photocurrent. Although this local autocorrelation technique enables ultrafast time resolution of up to sub-50 fs (ref. 13), it cannot measure non-local photocurrent dynamics such as the photocurrent extraction from the electrode. Moreover, evaluating the physical quantity deduced from the measured decay constant is challenging because it requires the assumption of a nonlinear response from a system, such as Pauli blocking, temperature-dependent electronic heat capacity and Auger recombination, depending on the experimental conditions.

Figure 4b compares the photocurrent decay obtained via the electrical and optoelectrical readouts for two samples (5 μm(1) and 5 μm(2)). In graphene with a lower mobility (sample 5 μm(1) with μ = 11,000 cm2 V−1 s−1), the two readout methods give similar decay times (upper traces in Fig. 4b). By contrast, in sample 5 μm(2) (μ = 140,000 cm2 V−1 s−1), the electrical readout gives a slower decay time than the optoelectrical readout (lower traces in Fig. 4b). The difference between the electrical and optoelectrical readouts might come from the difference in the sensitive energy region of the two methods. The signal of the electrical readout will reflect the lower-energy region more than that of the optoelectrical readout because the direct-current component is included when the generated photocurrent is electrically probed through the contact. Indeed, a microscopic first-principles calculation has predicted slower decay of the carrier occupation for the lower-energy region in defect-free graphene. By contrast, in low-mobility graphene, the SC cooling will compensate for the energy-dependent decay time by accelerating the decay in the low-energy region because it can effectively relax electrons with energies lower than optical and hyperbolic phonons (≈0.2 eV). These results further demonstrate that the direct readout of photocurrent is essential to understanding the O–E conversion processes and hence evaluating the performance of graphene PDs.

Discussion
We demonstrated the ultrafast non-local electrical readout of the photocurrent in PTE-based graphene PDs. By combining on-chip terahertz spectroscopy and gate-tunable devices with a suppressed RC time constant, we succeeded in resolving O–E conversion processes in graphene with their intrinsic timescale. Contrary to common expectations, we found that the time for photocurrent generation is tunable, from immediate at a large Fermi level to >4 ps when close to the CNP, as determined via the thermalization of photexcited carriers through tunable intraband HCM. We also found that the photocurrent response across the PDs is quasi-instantaneous without a carrier transit time, following the Shockley–Ramo theorem. Once thermalization is attained, the photocurrent decays via the carrier temperature being lowered through phonon interactions. Owing to the quasi-instantaneous photocurrent flow and the negligible RC constant, our method works as the ultrafast thermometry of graphene by directly tracking the time evolution of the carrier temperature. This enables us to obtain a deeper insight into the O–E conversion mechanism in graphene. Our prescriptions for overcoming the bandwidth limitations and gaining a quantitative understanding of the O–E conversion processes can be applied to any PTE-based graphene PDs and will set new standards for the design of graphene optoelectronic devices. Furthermore, our platform as developed here can be easily expanded for two-dimensional van der Waals materials and their heterostructures for investigating the key ultrafast processes for O–E conversion enabled by peculiar interlayer coupling, such as interlayer hot carrier dynamics, interlayer excitons and shift current. Our on-chip ultrafast electrical readout removes the barrier between ultrafast optical science and device engineering by demonstrating how to bring out the functionality (ultrafast O–E conversion) based on a fundamental understanding of ultrafast carrier dynamics, which is beneficial for the development of ultrafast optoelectronic applications.

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Methods

Device fabrication. Photoconductive switches were prepared using an LT-GaAs wafer supplied from BATOP. The wafer consists of a 2.6-μm-thick LT-GaAs surface layer (grown at 300 °C) and a 500-nm-thick Al0.1Ga0.9As sacrificial layer on a semi-insulating GaAs substrate. After the LT-GaAs layer had been etched into 100×100μm squares using citric acid solution, the sacrificial layer was dissolved in hydrochloric acid. The etched LT-GaAs chips on the GaAs substrate were transferred to a sapphire substrate using a thermoplastic methacrylate copolymer (Elvacite 2552C, Lucite International) as an adhesive. Any remaining Elvacite on the sapphire substrate was removed using citric acid.

Graphene was prepared via the mechanical exfoliation of natural graphite on silica (285 nm)/doped silicon substrates. Monolayer graphene was identified via optical contrast under a microscope. Using a separately exfoliated hBN flake, graphene was picked up and transferred onto the sapphire substrate via a typical dry-transfer technique using polydimethylsiloxane and polycarbonate.

Graphene was then patterned via reactive ion etching, and the Ti/Au waveguide structure with side contacts to graphene was deposited via sputtering. Then the whole surface was covered with a 30-nm-thick alumina (Al2O3) insulating layer grown via atomic layer deposition. The 20-nm-thick ZnO top gate layer grown via atomic layer deposition at 140 °C was patterned on the Al2O3 layer using photolithography and liftoff processes. The ZnO top gate was protected by deposition of another Al2O3 layer on top. Finally, for electrical contact with the waveguide, Al2O3 on the bonding pads was removed using Miroposit 351 developer.

On-chip terahertz spectroscopy (electrical readout). We used a Coherent Monaco femtosecond laser as the light source (1,035 nm, 280 fs, 50 MHz), and the second-harmonic wavelength of 517 nm was generated using a beta barium borate (Elvacite 2552C, Lucite International) as an adhesive. Any remaining Elvacite on the sapphire substrate was removed using citric acid.

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As the data were measured in our devices, the time evolution of the photocurrent is available at www.nature.com/reprints. Reprints and permissions information is available at https://doi.org/10.1038/s41566-022-01058-z.

Author contributions

K.Y. and N.K. conceived the experiment. K.Y. designed and built the optical setup, performed the measurements and analysed the data. K.Y. and N.K. designed the experimental apparatus. K.W. and T.T. conducted the experiments. K.Y. and N.K. wrote the manuscript with input from all authors.

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Competing interests

The authors declare no competing interests.

Additional information

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