Coherent THz detection via ultrafast dynamics of hot Dirac fermions in graphene

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Graphene has recently been shown to exhibit ultrafast conductivity modulation due to periodic carrier heating by either terahertz (THz) waves, leading to self-induced harmonic generation, or the intensity beat-note of two-color optical radiation. We exploit the latter to realize an optoelectronic photomixer for coherent, continuous-wave THz detection, based on a photoconductive antenna with multilayer graphene in the gap. While for biased THz emitters, the dark current would pose a serious detriment for performance, we show that this is not the case for bias-free THz detection, and demonstrate a detection bandwidth of at least 700 GHz at room temperature, even without optimized tuning of the doping. We account for the photocurrent and photomixing response using detailed simulations of the time-dependent carrier distribution, which also indicate significant potential for enhancement of the sensitivity, to become competitive with well-established semiconductor photomixers.

**Introduction**

Graphene possesses two key aspects for ultrafast optoelectronic applications. Firstly, one can obtain a drastic change in the carrier temperature $T_{\text{el}}$ and concomitant electronic conductivity, with only modest incident radiation intensities [1, 2], applicable over a huge spectral range due to the (near-)gapless band structure. Secondly, the cooling of the carriers due to energy transfer to the lattice (which has a much higher heat capacity) can take place on a time scale $\tau_e \approx 1$ ps [3–5], allowing for response bandwidths reaching the THz range [6, 7]. In addition to realizing conventional power-law photodetectors, the ultrafast conductivity modulation can interact with incident GHz-/THz-waves to manifest as a strong non-linear wave-mixing effect. This was demonstrated in single-layer graphene with high-field THz pulses [8], derived from a free-electron laser with center frequencies in the range 0.3-0.68 THz. In this case, the THz waves both drive and subsequently mix with the modulation to manifest as harmonic generation, with odd harmonic orders up to $n = 7$ being observed. The nonlinear response was found to be dependent on the doping (Fermi level) of the charge carriers [9], and could be described by a model accounting for the time-dependent conductivity of the hot-carrier distribution and the resulting current density. Due to the high 2D carrier density in a graphene monolayer, the effective bulk non-linear susceptibilities $\chi^{(n)}$ are remarkably high, although this is not specifically due to the Dirac band structure, as one arrives at similar magnitudes of $\chi^{(n)}/N$ per carrier in conventional bulk semiconductors with parabolic bands (as demonstrated recently in weakly $p$-doped Si [10]).

Besides such THz self-mixing, the conductivity modulation can be driven by superposing the light from two continuous-wave (CW) optical lasers (at frequencies $\nu_{1,2}$ respectively) to obtain a beat-note at the difference-frequency $\nu = \nu_2 - \nu_1$. Indeed, this was demonstrated to achieve a strong periodic modulation of $T_{\text{el}}$ [5], albeit at low temperature, where $\tau_e > 10$ ps is much longer than at room temperature. Here, we explore the potential to realize the coherent detection of CW THz waves, as depicted in Fig. 1(a), by photomixing the optically driven conductivity modulation from two near-infrared (IR) lasers with CW THz waves at the same frequency. The conductivity in the photomixer gap is given (to first order) by $\sigma(t) = \sigma_0 + \Delta \sigma \cos(\omega t + \varphi)$ ($\varphi$ being the phase offset between the conductivity modulation and incident THz wave, $\omega = 2\pi\nu$), where the photoconductivity $\Delta \sigma$ can be either positive or negative depending on the initial doping (Fermi level) [1, 4], as discussed in detail below. One then obtains an oscillatory current density $j(t) = \sigma(t)E_{\text{THz}}(t)$ due to the incident THz field $E_{\text{THz}}$, which contains even harmonics $j_{2n}$, where the rectified component $j_0 \propto \Delta \sigma \cdot E_0 \cos(\varphi)$ (coherent and linear in the THz field amplitude $E_0$) can be measured in the external circuit, as per conventional THz photomixers.

We investigate both the photoconductivity and THz photomixing response of a multi-layer graphene device and show that such coherent THz detection is possible. This is complemented with simulations which account near-quantitatively for the experimental results and indicate avenues to improve the photomixing performance.

**Results**

**Graphene photomixer devices**

Fig. 1(b,c) displays two micrographs of devices studied in this work. In (b) we show an array of packaged and bonded graphene photomixer devices fabricated on a...
sapphire substrate, with a magnified view of a single graphene photomixer in (c) (see Methods for details of the fabrication). Not clearly discernible is the gap region (13.6 × 11 μm²) of the bow-tie antenna which contains 6–8 layers of graphene as the photoconductive material, while the graphene outside of the gap region was etched away in an oxygen plasma. The devices have no backgates, with the native graphene doping due to interaction between the substrate and other layers (see section on simulations below).

The photo-response and the THz-photomixing properties of the devices were measured with a TOPTICA Terascan 1550 spectroscopy system, which usually employs a photoconductive InGaAs emitter and receiver, but where the standard receiver unit was replaced by our graphene device, as depicted in Fig. 1(d). All experiments were performed at room temperature.

**Photocurrent measurements**

The dark current was measured vs. bias voltage $U_b$ and found to be perfectly linear vs. bias, indicating essentially ideal ohmic resistance. The extracted dark resistance $R_0$ of one of the graphene photomixers (Device S1) was estimated to be $R_0 = 1.04 \, \text{kΩ}$. Measurements of the photocurrent $I_{pc}$ vs. $U_b$ for the same device S1 are shown in Fig. 2(a) for a range of near-IR pump powers $P$ (also shown for the highest $P$ in Fig. 2(b) for comparison with the dark current). For bias values $|U_b| < 0.6 \, \text{V}$, the photocurrent is found to depend in a fairly linear manner on $U_b$, while it shows a tendency towards saturation at higher bias. The corresponding photoconductivity $\sigma_{pc}$ is obtained from the slope in the linear regime. The absolute value $|\sigma_{pc}|$ is presented in Fig. 2(c) for two exemplary devices S1 and S2. For both, one observes a linear dependence vs. incident power. The negative sign of $\sigma_{pc}$ corresponds to photoinduced differential resistivity, as observed previously [1, 4] for graphene devices sufficiently away from the intrinsic doping regime (where one instead has positive photoconductivity). One also sees in Fig. 2(a) that the sign of the photocurrent changes at $U_b = 0.1 \, \text{V}$ instead of at zero bias. This shift is attributed to a residual asymmetry of the properties at the two gap contacts (e.g. due to electrode-induced doping gradients) which gives rise to an additional unipolar photocurrent component [11, 12].

**THz detection**

If the unbiased device is now illuminated by THz radiation (frequency $\nu$) instead of the two-color near-IR laser radiation, one also obtains a photocurrent – termed “direct signal” in the following – as shown vs. $\nu$ for device S1 in Fig. 3(a) (green curve). The magnitude of the direct signal, unlike the photomixing signal to be discussed next, is proportional to the power of the incoming THz wave (data not shown). The frequency dependence exhibits a strong roll-off towards high frequencies such that the photocurrent reaches the noise floor just below 200 GHz. This direct signal must result from a residual spatial asymmetry in the device, and being proportional to the THz power, likely arises from photovoltaic/photothermoelectric effects due to energy and charge-carrier diffusion at the (unequal) contact regions [13].

If the device is illuminated simultaneously by the THz wave and the near-IR beat-note at the same frequency (with both beams at the maximal available power), one obtains the photocurrent spectrum $\Delta I_{pm}(\nu)$ shown by the red curve in Fig. 3(a), obtained by subtracting the direct signal from the experimental one. For the employed lock-in integration time of 100 ms, the signal is
above the noise floor from 60 GHz to 700 GHz. The magnified, low-frequency range in Fig. 3(b) allows a clearer comparison of the direct and photomixer signals (in terms of the amplitude envelope $\Delta I_{pm,e}$ for the latter) at low frequencies. That the signal is of photomixing origin, is demonstrated clearly by the oscillations in $\Delta I_{pm}$ vs. frequency (which varies with the relative phase $\varphi = \omega \tau$, $\tau$ being the delay between THz and optical beat-note at the detector) – a magnified range is shown in Fig. 3(c) (where the direct signal has already dropped to the noise floor). Note that the slower modulation on the envelope of the interference signal is due to reflections in the THz and optical beam paths (as also seen using a commercial photomixer for detection). In Fig. 3(d), we plot $\Delta I_{pm,e}(\nu)$ extracted from the $\Delta I_{pm}(\nu)$ in (a), as well as the corresponding data for a second device S2 (which showed somewhat lower performance). One sees the spectra roll-off with increasing frequency, with a rate close to $1/\omega^2$ above 200 GHz, which is partly due to the roll-off of the THz field spectrum. In order to calculate the inherent photomixing sensitivity $R_E(\nu) = \Delta I_{pm,e}(\nu)/E_{THz}(\nu)$ of the device ($E_{THz}$ being the amplitude of the THz radiation field in the photomixer gap), we combined both the experimentally measured THz power spectrum and simulations of the THz radiation coupling efficiency, to estimate $E_{THz}$. These simulations take into the coupling efficiency of the Si substrate lens and antenna response, (see Methods). The resultant spectrum $E_{THz}(\nu)$ is shown in Fig. 3(d) (blue curve, right scale), which exhibits roughly a $1/\omega$ roll-off which results from the frequency characteristics of the photoconductive-emitter source. Note that by defining $R_E(\nu)$ in terms of the estimated THz field in the gap also allows us to compare directly with the simulation results below.

We then arrive at the photomixer sensitivity $R_E(\nu)$ shown in Fig. 3(e) for the two devices. The highest sensitivity is reached at 79 GHz, with a photodetector sensitivity of 2.2 nA/(V/cm). Having compensated for the roll-off of the THz field, one sees that $R_E$ roughly follows a $1/\omega$-dependence above 200 GHz. As we will discuss on the basis of simulations below, this dependence is a consequence of the finite energy relaxation time of the charge carriers of graphene. Note that the enhanced photomixer contribution below 200 GHz is likely to arise from the wiring of the bow-tie antenna [14], and not the bow-tie antenna coupling efficiency (which simulations show is relatively flat over the full spectral range here, i.e. only varying by 1.5 dB – see Methods).

In Fig. 3(f) we plot the dynamic-range (DR) spectrum calculated from the photomixing signal spectrum in (d) for device S1 (red curve), calculated as $DR = 20 \log_{10}(\Delta I_{pm,e}(\nu)/\Delta I_{pm0})$ (i.e. referenced to THz power), where $\Delta I_{pm0}$ is the noise level calculated from the standard deviation of the signal in the absence of THz/optical power on the device. The peak value is 52 dB measured at 86 GHz. To put this performance into perspective, we also plot the DR spectrum for detection with the InGaAs photomixer of the Topica measurement unit (black curve). It exhibits a 32-dB higher current response at 86 GHz, growing to 50 dB at 200 GHz due to the roll-off of the graphene sensitivity. These and additional measurements with other graphene photomixers show that the graphene devices achieve an appreciable THz photomixing response, but still with a sensitivity and DR substantially lower than that of the InGaAs device. However, this comparison has to be taken in context, as one puts a highly optimized photomixer against an un-optimized one, e.g. the Topica device uses optical-fiber-coupling of the two-color near-IR radiation instead of the less-efficient free-space coupling in the case of the graphene photomixer. In the next section, we simulate the performance of the graphene device in order to understand the experimental results. We then identify substantial potential for improved performance, in particular, by optimization of the doping level.
FIG. 3. Experimental THz photomixer response vs. THz-wave frequency (lock-in integration time: 100 ms). (a) Detected photomixer current $\Delta I_{pm}$ (red) after subtraction of direct-THz photocurrent (green, measured in the absence of the optical beat on the receiver) for device S1. (b,c) Magnified frequency ranges of signals (linear vertical scales; in (b) plotting the amplitude envelope $\Delta I_{pm,e}$). (d) Photomixer amplitude spectra (left scale) for both devices, S1 and S2 (solid and dashed curves, respectively), and estimated THz field amplitude in the photomixer gap (right scale) calculated using experimental THz power spectrum and antenna response (see text). Roll-off curves following $1/\omega$ and $1/\omega^2$ included for comparison. (e) Photomixer sensitivity $R_E(\nu) = \Delta I_{pm,e}(\nu)/E_{THz}$ for each device, based on the results in (d). (f) Comparison of the dynamic range (DR) achieved with the graphene photomixer (red curve, device S1) and a commercial InGaAs photomixer (Toptica spectrometer, black curve).

Simulated photocurrent and photomixing response

In order to theoretically describe and quantify the photomixing process, we apply a modified version of the time-dependent conductivity model described in Ref.s [8] and [9], where here the modulation in the carrier heating from the two-color optical pump yields a periodic change in the conductivity which is then driven by (i.e. mixes with) the THz field to yield the detected DC current signal. A detailed description of the band structure and doping levels for such a multi-layer graphene system is non-trivial, as the adjacent layers may form a composite, gapped band structure [15], while even for weakly interacting layers one has to consider how substrate- and contact-induced-doping will distribute across the layers, including the effects of screening. For simplicity, we model the multi-layer graphene sample as a set of parallel layers (sheet conductances), each represented by the linear dispersion of single-layer graphene, with the same nominal doping.

It is well-established that for moderate excitation levels, single-layer graphene exhibits positive photocconductivity for low doping levels, which crosses over to negative photocconductivity (i.e. photoresistance) at higher doping, the latter as seen in our graphene devices (Fig. 2(a)). While this was first attributed to contributions from both photovoltaic effects (by optically excited e-h pairs) and bolometric effects (by a change in carrier distribution due to heating) [1], respectively, it was shown subsequently [4] that both regimes can be attributed to a bolometric response when accounting for the change in both the carrier temperature $T_{el}$ and chemical potential $\mu$ of the hot carriers. Underlying this line of argument is that the charge carriers rapidly relax after photoexcitation on a timescale $\lesssim 100$ fs and remain close to single hot Fermi-Dirac distribution $f(E, T_{el})$ (across both valence and conduction bands) due to rapid electron-electron (e-e) scattering, with a heat density $Q$ given by the absorbed optical intensity [4]. It is hence assumed in the following that the charge carriers are thermalized and that at any time, one can assign a temperature $T_{el}$ to the carrier ensemble.

The expression for the conductivity of each layer, based on the generalized Drude model with a hot-thermal carrier distribution is given in the Methods section (Eq. 1), with the $T_{el}$ and $\mu$ determined by $Q$ with fixed doping concentration (net carrier density) $N$. For the calculation of the sheet (2D) electric conductivity $\sigma(\omega)$ and the mobility $\mu(\omega)$, one requires the carrier-energy-dependent momentum scattering rate $\Gamma(E)$. Here, we include acoustic- and optical-phonon scattering, in addition to charged-impurity scattering $\Gamma_{c}(E)$ [8, 16] (see Methods). Results from this treatment are shown in Fig. 4 for $T = 300$ K vs. $Q$ for a set of doping levels $N$ (note that the experimental optical intensity levels above
correspond to a maximum of $Q < 0.5 \text{nJ cm}^{-2}$, see below). In Fig. 4(a,b), one observes: (i) the dependence of $T_{cl}(Q)$, which first becomes steeper and then less steep for small $N$ and hence is non-monotonic in $N$ due to a local maximum in the electronic heat capacity $C_{el}(N)$ at finite $T_{cl}$ [5], and (ii) the reduction in chemical potential $E_F(Q)$ with increasing $Q$, as required to conserve the total charge density.

Turning now to the sheet conductivity $\sigma = \text{Re}\{\tilde{\sigma}\}$, we first plot the spectral weight $W = \int_{-\infty}^{\infty} \sigma(\omega) \, d\omega$ in Fig. 4(c), which one can show depends only on the Fermi-Dirac distribution $f(E, T_{cl})$ of the carriers (and not $\Gamma(E)$), and hence allows more universal, initial statements about the changes of the conductivity vs. parameters such as $Q$ [4]. In Fig. 4(c), one observes that the slope of $W(Q)$, which is a measure for the photocconductivity, changes from positive to negative at $N \sim 0.5 \cdot 10^{12} \text{cm}^{-2}$. This behavior results from the detailed dependence of $\partial_{\tilde{E}} f$ for the carrier distribution, i.e. the density of neighboring vacant states for intraband scattering. At much higher $Q$ (outside of the plotted range and corresponding to $T_{cl} \gtrsim 1000 \text{K}$), $W(Q)$ becomes an increasing function of $Q$ for all $N$ [4].

The absolute DC conductivity $\sigma_0 = \sigma(\omega = 0)$ and DC photocconductivity (given by the differential $\Delta \sigma_0 = \sigma_0(Q) - \sigma_0(Q = 0)$), are shown in Figs. 4(d,e), respectively. Here, one also sees the crossover behavior vs. $N$, albeit at a slightly larger $N > 1 \cdot 10^{12} \text{cm}^{-2}$. One also finds that the photocconductivity has a larger gradient $d\Delta \sigma_0/dQ$ for small $N$, as per $W(Q)$. A cross-over is also seen in the slope of the mobility $\mu$, which is included for completeness in Fig. 4(f).

As mentioned above, we represent the multi-layer device as composed of 6 identical graphene layers. A doping level of $N = 1 \cdot 10^{12} \text{cm}^{-2}$ in each layer was chosen, which in combination with the assumed impurity scattering rate and cooling rate yields magnitudes of the DC resistance and photocconductivity close to those in the experiment. Using these results, we can calculate the time-dependent photocconductivity response to the THz wave due to heating by the absorbed optical pump intensity. Based on the experimental beam diameter and power, we calculate the average intensity $I_{1,2}$ in the electrode gap for each component of the two-color optical

FIG. 4. (a-f) Dependence of parameters for the carrier distribution and conductivity on heat density $Q$ for a set of doping levels $N$ in a single graphene layer, as used for the photocconductivity/photomixer simulations. (a) Temperature $T_{cl}$, (b) chemical potential $E_F$, (c) Drude spectral weight $W$, (d) DC conductivity (2D) $\sigma_0$, (e) differential conductivity $\Delta \sigma_0$ (i.e., photocconductivity) in (d), and (f) effective mobility $\mu$ (based on doping density $N$). Dashed horizontal lines for respective values for $Q = 0$. Arrows and multiplication factors in (d,e) in going from $N = 10^{12} \text{cm}^{-2}$ to $N = 0.1 \cdot 10^{12} \text{cm}^{-2}$ (for $Q = 0.5 \text{nJ cm}^{-2}$) – see Discussion. (g-l) Simulated time-dependences for the highest experimental power in Fig. 2 and a laser beat frequency of $\Delta \omega = 200 \text{GHz}$. (g) Optically deposited heat density (average per layer), (h) total and (i) differential sheet conductivity of the modeled 6-layer graphene device (j) Corresponding simulated result as in (i) but for single-layer graphene with near-intrinsic doping. Complementary results for a LT-GaAs photomixer with the same optical pump intensity and beat frequency (see text): (k) volume density of excited e-h pairs in the surface region and (l) time-dependent sheet conductivity. Vertical dashed lines in (g,k) to demonstrate the phase shift relative to the optical beat signal maxima at $t = 0$. 

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
pump (amounting to $I_{1,2} = 74\, \text{W mm}^{-2}$ for the maximum total pump power $P = 25.2\, \text{mW}$ in Fig. 2(a)). The time-dependent heat density $Q_n(t)$ in the $n$th layer is calculated by solving Eq. 2 (Methods) for the absorption of optical intensity and cooling due to energy relaxation by electron-phonon (e-ph) scattering, for which we assume a time constant of $\tau_c = 1\, \text{ps}$, based on literature reports [4, 5, 17]. Here we assume that the absorbed optical energy is initially preserved in the electronic system during rapid elastic e-e scattering, with e-ph scattering contributing only to the subsequent cooling (energy relaxation), which is a reasonable approximation for the relatively weakly doped graphene layers here [18]. As the lattice heat capacity is 2-4 orders larger than the electronic one [5], we treat the lattice as an ideal reservoir and neglect any further cooling bottlenecks (with the substrate ultimately extracting the lattice heat with only a small increase in lattice temperature [5]).

The resultant time-dependent heat density $Q(t)$ and conductivity $\sigma(t)$ (again, for the maximum experimental total power $P = 25.2\, \text{mW}$ in Fig. 2(a)) is shown in Figs. 4(g-i) for a laser beat frequency of $\Delta \nu = 200\, \text{GHz}$. Here one sees that the cooling is sufficiently fast that the heat density modulation depth is high $(1 - Q_{\text{min}}/Q_{\text{max}} = 0.81)$, resulting in a peak-valley variation in the total multi-layer conductivity of $\Delta \sigma_{pp} = 0.9 \cdot 10^{-8} \, \Omega^{-1}$ (Fig. 4(i)). The relative conductivity modulation amounts to $\Delta \sigma_{pp}/\sigma = 6.9 \cdot 10^{-4}$. For comparison, in Fig. 4(j) we plot the corresponding result for a single graphene layer, but for a ten times lower doping density $N = 0.1 \cdot 10^{12} \, \text{cm}^{-2}$ closer to intrinsic. As expected from Fig. 4(e), the polarity now corresponds to positive photoconductivity, and $\Delta \sigma_{pp}$ is a factor 4.5 times higher, despite only having a single layer (see discussion below).

To put the magnitude of this conductivity modulation into perspective, we performed complementary simulations of photoexcited low-temperature-grown LT-GaAs, where the photoconductivity modulation arises due to interband e-h generation and subsequent carrier trapping, using the same optical intensity and laser beat frequency (assuming an excitation wavelength close to the bandgap). We assumed a momentum relaxation rate of $1/\tau$, with $\tau = 160\, \text{fs}$, and a carrier trapping time (mobility lifetime) of $\tau_t = 300\, \text{fs}$ – see Methods. The results are shown in Figs. 4(k,l). One sees that the surface excitation densities for such a CW photomixer are only moderate (reaching values of $N_{e,h} \sim 10^{15} \, \text{cm}^{-3}$, significantly lower than reached with femtosecond pulsed excitation [19]), and when one integrates the total sheet conductivity, this amounts to $\Delta \sigma_{pp} \sim 50 \cdot 10^{-8} \, \Omega^{-1}$ (Fig. 4(l)), i.e. a factor 50 larger than we calculate for our present multi-layer graphene device, but only a factor $\sim 10$ larger than that predicted for a single-layer device tuned to near-intrinsic doping. Hence, given that only 2.3% of the optical power is absorbed in a single graphene layer (compared to complete absorption in the LT-GaAs surface layer), one sees that a comparable sheet photoconductivity per unit absorbed power is predicted.

On the basis of these results, we can calculate the predicted DC photoconductivity of the graphene device, by taking the cycle-average value of $\sigma_{pp} = \langle \Delta \sigma(t) \rangle$ (as we employ a nearly square-shaped device gap, the sheet conductivity also corresponds to the device conductance). This is shown in Fig. 5(a) along with the experimental results from Fig. 2(c) for the same conditions. Note that our choice of the doping level was guided by the magnitude of the experimental photocurrent, and hence the agreement seen is to be expected, although the result is also based on choosing reasonable values for the impurity scattering rate and cooling rate and reproducing the dark DC resistance of the device. Moreover, although there is a deviation between experiment and simulation at low power, the saturation behavior is present in both.

Finally, we calculated the predicted THz photomixer sensitivity $R_E(v)$ of the device on the basis of the simulation results above, for comparison with the experimental results in Fig. 3(e). If one considers a homogeneous THz electric field coupled into the electrode gap (replacing the DC bias used to measure the DC photocurrent above), $E(t) = E_0 \cos(\omega t)$, then the current density is given by a convolution of the driving field $E(t)$ with a Drude response whose parameters are modulated as per $Q(t)$ due to the two-color optical excitation. In general, this would allow for mixing terms with the modulation of both the plasma frequency and Drude scattering rate of the electron-hole gas. However, given the short momentum scattering time ($\ll 100\, \text{fs}$), the leading term can be expressed in terms of a time-dependent (parametric) conductivity, i.e. the quantity $\sigma(t)$ presented above (Fig. 4(h)). One can then show that the rectified (DC) current density amplitude is given by $j \sim \frac{1}{4} E_0 \Delta \sigma_{pp}$ (achieved for the optimal relative phase between the THz and optical-beat by scanning their relative delay at the detector), with a resulting device current $i = w \cdot j$ (with the electrode width $w = 13.6\, \mu\text{m}$). The simulated sensitivity spectrum is shown in Fig. 5(b), as well as the experimental one from Fig. 3(e). Although the simulation parameters were chosen to correspond to the experimental photocurrent in Fig. 5(a), no further extensions/modifications were employed to predict the THz sensitivity. In this sense, the agreement in Fig. 5(b) provides good independent support for the foregoing model, despite its simplifications. Good agreement is reached regarding the roll-off of the signal. The simulated sensitivity spectrum possesses a first-order ($\propto 1/\omega$) roll-off with a 3-dB bandwidth of $\sim 300\, \text{GHz}$, due to the electron cooling model used with $\tau_c = 1\, \text{ps}$. The experimental spectrum shows the predicted $1/\omega$-roll-off at high frequencies. The higher-than-predicted (and frequency-modulated) sensitivity below $\sim 150\, \text{GHz}$ is attributed mainly to the antenna effect of the wiring of the detector, cp. discussion of the photomixer sensitivity $R_E(v)$ as shown in Fig. 3(e).
scales with noise floor as low as possible [20]. The noise current 
100% as possible (cp. Figs. 4(k,l) for L T-GaAs) and the 
tically induced conductance modulation is as close to 
a high dark resistance of the devices, such that the op-
ductive nature of the graphene photomixer needs to be 
discussed. With photomixers, one commonly strives for 
a high dark resistance of the devices, such that the opti-
cally induced conductance modulation is as close to 
a high dark resistance of the devices, such that the op-
ductive nature of the graphene photomixer needs to be 
addressing this potential, we put this performance into 

Discussion
The predictions here indicate a path to significantly en- 
hance the graphene photomixing performance. Before 
addressing this potential, we put this performance into 
perspective with regard to conventional semiconductor-

By how much can the performance as expressed by 
the NEP be improved in the 200-GHz range and be- 
low? The key direction here would be to develop a 
photomixer with a single (hBN-encapsulated) layer of 
graphene and with control of the density of the charge 
carriers by a (back)gate voltage to approach the intrin-

Fig. 5. (a) Simulated and experimental photoconductivity vs. optical power for the multi-layer graphene device with experimental conditions as in Fig. 2. (b) Simulated and experimental (Fig. 3(e)) photomixer field sensitivity vs. THz frequency. Lines corresponding to a first-order response roll-off (∝ 1/ω) added as visual guides.
off than the detection with (optimally mobility-lifetime-engineered) semiconductor-based devices because of the longer relaxation time constant \( \tau_c \), (note that \( \tau_c \) further increases at lower temperature [5]). The graphene photomixer is hence mainly of interest for applications with sub-500-GHz radiation. There it can play out its advantages, of which the most important are that it is easy to fabricate (especially not requiring the expensive growth of dedicated materials by atomic-layer growth techniques such as MBE or CVD) and that it can be operated in combination with laser radiation over a broad wavelength range (from mid-IR to UV), which is also used for the generation of THz waves by photomixing.

As a final remark, we note that the ability here to enhance the photomixing response by approaching the intrinsic regime contrasts with the case of the self-driven THz nonlinearity, as the sensitivity to heating but also the absolute magnitude of the intraband conductivity which dictates the heat absorption and drops significantly as one approaches the intrinsic regime (Fig. 4(c)).

### Conclusion

We have explored a novel photomixing mechanism for the detection of continuous-wave THz radiation. Unlike classical semiconductor-based photomixers which rely on interband generation of mobile charge carriers, we employ the hot-carrier bolometric effect in graphene. Its ultrafast response gives rise to a “relaxational” nonlinearity which is used for the rectification of the THz radiation. This mechanism is related to mixing in conventional hot-electron bolometers (HEBs) [23]; while these require cryogenic cooling, the photomixing with graphene works at room temperature. Good agreement is found between the experimental data and simulations based on an energy-relaxation model of the charge carriers in graphene. Variation of model parameters has allowed us to make suggestions for the improvement of the photomixer detector. From a practical point of view, such devices have the advantages of their ease of fabrication and of their capability to be operated with (local oscillator) laser radiation at arbitrary wavelengths.

### Materials and methods

#### Detector fabrication

The devices were fabricated on sapphire substrates (thickness 300 \( \mu m \)). First, after cleaning of the substrate in an acetone and isopropanol bath, a 6–8-layer graphene sheet (tradename: Trivial Transfer Graphene), purchased from ACS Material LLC, was deposited, followed by photolithographic definition of the bow-tie antenna (300-nm-thick Au film on a 2-nm-thick Ti adhesion layer, antenna arm length: 493 \( \mu m \), bow-tie flare angle: 90.8\(^{\circ} \), gap area: 13.6 \( \times 11 \mu m^2 \). As the final step, the graphene was etched away in an oxygen plasma treatment everywhere except in the antenna gap. A photore sist layer protected the gap region during plasma treatment and was removed after the etch step.

#### Photocurrent and THz measurement setup

All optical measurements were performed with a CW THz spectrometer Terascan 1550 (vendor: Toptica Photonics AG, Gräfelfing/München). It uses two diode lasers operating at wavelengths in the telecom wavelength range (\( \lambda = 1550 \) nm). The fiber-coupled InGaAs photomixer serving as THz detector in the system was replaced by an optical bench for free-space illumination of our graphene photomixer device as illustrated in Fig. 1(d). The bench (a four-steel-rod cage) contained a collimation lens (AR-coated for 1550 nm, producing a collimated beam with a 1/e\(^2\)-diameter of 3.6-mm, a mechanical light chopper, an aperture, filters for beam attenuation and a AR-coated lens (\( f = 30 \) mm) to focus the laser radiation into the gap of the photomixer antenna. The lens was mounted on a xyz-translation stage for beam centering and optical prealignment. This is decisive for subsequent optimization of the photomixer signal, without affecting the pre-alignment of the photocurrent. The detector (held at room temperature and exposed to air, no purging of the beam path to remove water vapor) was mounted on a second xyz-translation stage (a 3-axis, high-precision flexure stage, Thorlabs NanoMax) for the alignment of the laser beam’s focus into the antenna gap.

For the photocurrent measurements, the laser beam was chopped at 400 Hz by the mechanical chopper. The maximum beam power before the chopper was determined with an integrating sphere photodiode power sensor (Thorlabs Inc, S145C). The power was then varied by inserting filters with different optical density (OD). The photocurrent \( I_{pc} \) was measured with one antenna leaf grounded, the other connected to the internal bias source (which provided the bias voltage \( U_b \)) of a low-noise current preamplifier (model SR570 from Stanford Research Systems). The optimization of the alignment of the optical beam was performed at a fixed bias of 0.6 V. The current amplifier was operated in high-pass (HP) mode (filter corner frequency: 100 Hz, filter slope: 6 dB, sensitivity: 500 \( \mu A/V \), gain mode: low-noise (LN)). Its output signal was fed to a lock-in amplifier (Ametek DSP 7265). The lock-in integration time constant was set to integration time 100 ms. The peak-to-peak photocurrent was determined as \( I_{lock-in} = (\pi/\sqrt{2}) \cdot I_{lock-in} \), with \( I_{lock-in} \) being the current reading of the lock-in amplifier.

For the THz sensitivity characterization, a hyperhemispherical Si-substrate lens with a diameter of 12 mm was mounted in a freely positionable way (not glued) against the back-side of the detector [24]. The lens was mounted on another \( xy \)-stage used for the alignment of the lens in the THz beam. Special care was taken to center the THz beam on the gap region of the antenna prior to the fine tuning of the substrate lens (in order to ensure a good
focussing at all THz frequencies). The rectified current amplitude due to the THz field was measured with a lock-in amplifier at the modulation frequency (7.6 kHz) of the InGaAs photomixer emitter bias (Toptica) with an integration time of 100 ms. The current amplitude was determined from $\Delta I = \sqrt{2} \cdot I_{lock-in}$ [24].

**Estimation of THz field amplitude in photomixer gap**

The amplitude of the THz electric field in the antenna gap is calculated from the voltage $U_{THz}$ of the THz signal across the gap as $E_{THz} = U_{THz}/d_{gap}$, the gap in the bow-tie antenna having the length $d_{gap} = 13.6 \mu m$. $U_{THz}$ is determined from the power of the incident THz radiation via $U_{THz} = \sqrt{P_{THz}} \cdot R_0$. Here, $R_0 = 1040 \Omega$ is the ohmic resistance of the graphene stripe in the gap, and $P_{THz}$ is the power of the THz beam delivered to the gap. $P_{THz}$ is calculated via $P_{THz} = P_D \cdot (4 \cdot \text{Re}(Z_{ant}(v))) \cdot \text{Re}(Z_C)/(Z_{ant}(v) + Z_C^2)^{1/2}$, where $P_D$ is the frequency-dependent antenna impedance $Z_{ant}(v)$ and $Z_C$ denotes the impedances of the antenna and the graphene channel, respectively [24]. We assume the latter to be real-valued and constant in frequency, $Z_C = R_0$. The frequency-dependent antenna impedance $Z_{ant}(v)$ is obtained from EM simulations (Keysight Advanced Design System (ADS)). $P_D$ is the fraction of the total free-space beam power $P_{TPHs}$, which is collected by the antenna and can be determined from $P_D = 1/2 : \eta_{op} \cdot \eta_{gauss} \cdot \eta_{ant}(v) \cdot P_{THz}$ [24, 26], such that the total power delivered to the multilayer graphene is obtained from $P_{THz} = \eta_{op}(v) \cdot P_{TPHs}$, where the total coupling efficiency is given by $\eta_{op}(v) = 1/2 : \eta_{op} \cdot \eta_{gauss} \cdot \eta_{ant}(v) \cdot \eta_{m}(v)$ accounts for all losses towards the antenna gap. The optical THz power $P_{TPHs}$ present at the air-to-silicon-lens interface, was measured with a calibrated Golay cell. The Toptica emitter delivers a power of 47, 5.2 and 0.4 $\mu W$ at 0.1, 0.3 and 0.7 THz respectively. The frequency-dependent antenna efficiency $\eta_{ant}(v)$ of the bow-tie antenna is also derived from EM simulations (ADS). $\eta_{op} = 0.7$ is the optical power loss factor accounting for reflection losses at the air-to-silicon-lens interface [24], $\eta_{gauss} = 0.9$ denotes the gaussian-beam coupling efficiency [24, 27], also known as Gaussicity, the factor of 1/2 accounts residual scattering of incident power in an receiving antenna element [25, 28]. Example values at the frequencies 0.1, 0.3 and 0.7 THz for the total coupling efficiency are $\eta_{tot}(v)$ of 2.1, 2.3 and 2.4% respectively.

**Theoretical approach**

The frequency-dependent, complex conductivity of each graphene layer is calculated via the generalized Drude response for each respective Fermi-Dirac carrier distribution [8]:

$$\sigma(\omega) = (e^2 v_F^2/2) \int_{-\infty}^{\infty} D(E)s(E, \omega) (-\partial_E f(E)) dE$$  \hspace{0.5cm} (1)

where $s(E, \omega) = (\Gamma(E) + i \omega)^{-1}$ represents the differential Drude response at each energy $E$, and $\Gamma$ is the energy-dependent momentum scattering rate. The Fermi-Dirac distribution $f(E) = [1 + e^{\beta(E-E_F)}]^{-1}$ with $\beta = (k_B T_{\text{eff}})^{-1}$ is calculated by imposing the conditions for the total energy $U = U_0 + Q$ and constant doping density $N$ [4, 29] to determine $T_{\text{eff}}$ and $E_F$, where $Q$ is the instantaneous heat density absorbed in each respective layer. For the impurity scattering rate, we take $\Gamma_i = (\gamma(E)|E|^{-1}$, where $\gamma(E)$ is a slowly varying function of $E$ [16], and is found to vary for different graphene samples. Here we tuned the value of $\gamma$ to yield the internal dark resistance of our experimental multi-layer device ($R_0 - 2R_c = 635 \Omega$, assuming a series contact resistance $R_c$ of 200 $\Omega$ at each electrode), corresponding to $\gamma(E = 0) = 0.12$ $fs$ $m/eV^{-1}$, which results in a magnitude for $\Gamma(E)$ intermediate between the values found in Refs. [8] and [16].

For calculating the time-dependent heat density $Q_n(t)$ in the nth graphene layer, we employ a simple single-component cooling rate with $\tau_c = 1$ ps, and solve the heat equation:

$$\partial_t Q_n = a_n (t) - Q_n/\tau_c$$  \hspace{0.5cm} (2)

where $a_n$ is the absorption fraction in the nth layer, including the weak depletion of the light for each subsequent layer approaching the substrate ($a_6 = 2.29\%$ for the uppermost layer). As we assume the cooling rate is independent of $Q$, one can solve for $Q(t)$ analytically with the incident two-color optical pump, $I(t) = I_{00} + I_{01} (1 + \cos(\omega t))$, where $\omega = 2\pi\Delta v$ and $\Delta v = v_2 - v_1$ is the frequency difference between the two lasers.

For the comparative calculations for a conventional-semiconductor (LT-GaAs) photomixer, we take a carrier trapping time of $\tau_c = 300$ fs, a Drude scattering time of $\tau = 160$ fs and the literature effective masses [30] (yielding a reduced plasma mass of $m_{\text{eff}} = (1/m_e + 1/m_h)^{-1} = 0.056 \cdot m_0$). We assume carrier heating to be negligible, and also that the bulk temperature remains close to room temperature (although there will be some lattice heating because a considerable part of the photon energy will be transferred to lattice vibrations by nonradiative energy relaxation processes).

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**Author Declarations**

The authors declare that there are no conflicts of interest related to this article.

**Author contributions**

FL and SA prepared the samples, FL, JH and RA performed the measurements, MDT contributed all simulations and analyzed the data in detail together with FL. The manuscript was written by MDT, FL and HGR, and reviewed by all authors. HGR conceived the idea and supervised the project.
Data Availability
Both the experimental and simulation data, and details concerning calibration/analyses can be obtained from the authors upon reasonable request.


