Supplementary Information

Thermoelectric graphene photodetectors with sub-nanosecond response times at Terahertz frequencies

Leonardo Viti,1 Alisson R. Cadore,2 Xinxin Yang,3 A. Vorobiev,3 Jakob E. Muench,2 Kenji Watanabe,4 Takashi Taniguchi,4 Jan Stake,3 Andrea C. Ferrari,2 Miriam S. Vitiello1

1. NEST, Istituto Nanoscienze - CNR and Scuola Normale Superiore, Piazza San Silvestro 12, 56127 Pisa, Italy
2. Cambridge Graphene Centre, University of Cambridge, 9, JJ Thomson Avenue, Cambridge CB3 0FA, UK
3. Department of Microtechnology and Nanoscience, Chalmers University of Technology, SE-41296 Gothenburg, Sweden
4. National Institute for Materials Science, 1-1 Namiki, Tsukuba, 305-0044, Japan

Simulations of the on-chip RF filter

Figure S1: Design of the on-chip transmission line. (a) low-pass filter geometry and electric field distribution on the filter for an input frequency of 3.5THz, calculated using CST Microwave Studio. P1 and P2 indicate the input and output ports of our simulation. (b) Electrostatic finite elements (FEM) simulation of the low-pass filter performed using COMSOL Multiphysics. The presence of the filter adds a capacitance of 500 aF to the transmission line. (c) S-parameters (S11, red line, S21, green line) of the transmission line not including the hammer-head filter in the frequency range from 1 GHz to 3.5 THz. (d) S-parameters of the transmission line with the hammer-head filter in the frequency range from 1 GHz to 3.5 THz.
Radio-frequency (RF) and electromagnetic simulations are performed using CST Microwave Studio. The substrate is modeled as a loss-free high-resistivity (> 10000 Ω cm) Si, with dielectric permittivity 11.9 and thickness 350 μm, and the strips are modelled as lossy Au layers with electrical conductivity $4.56 \times 10^7$ Sm$^{-1}$. We use a two-ports configuration, indicated in Fig. S1a as P1 (input) and P2 (output). This allows us to evaluate the transmission line S-parameters between the two ports in presence of the capacitive hammer-head filter (Fig. S1a,b). In particular, we are interested in the reflected (S11) and transmitted (S12) amplitudes as a function of the input frequency with (Figure S1d) and without (FigureS1c) the filter.

**Device layout, optical images**

![Optical images of the two samples](image)

Figure S2: Optical images of the two samples. Optical microscope images of samples A and B. Source (s), drain (d) and gates (g$_{TR}$, g$_{TL}$) are marked on the images.

**Intensity distribution of the Terahertz field**

The Airy pattern shown in Fig. 2b of the main text has four minima, radially spaced by $\delta r = 190$ μm. This is compatible with the expected [1] $\delta r \sim 1.01(\theta/\alpha) = 175$ μm, where $f = 30$ mm is the focal
length of the focusing lens and \( a = 15 \) mm is its clear aperture. The small discrepancy between predicted and measured \( \delta r \) is probably due to a sub-mm misalignment with respect to the focal plane along the optical axis. We fit the intensity distribution with a two dimensional-Gaussian \( g_{2d} \) (Figure S3), which, in Cartesian coordinates, reads:

\[
g_{2D}(x, y) = g_0 + A \exp \left[ -\frac{[(x-x_c) \cos \theta + (y-y_c) \sin \theta]^2}{2\sigma_x^2} - \frac{[(x-x_c) \sin \theta + (y-y_c) \cos \theta]^2}{2\sigma_y^2} \right] \tag{eS3}
\]

where \( g_0 \) is a global offset, \( A \) is the maximum photovoltage, \( x_c \) and \( y_c \) are the mean values along \( x \) and \( y \) directions, \( \theta \) is the in-plane rotation angle of the distribution and \( \sigma_x \) and \( \sigma_y \) are the standard deviations along \( x \) and \( y \). From fitting, we estimate standard deviations \( \sigma_x = 95 \pm 1 \) \( \mu \)m and \( \sigma_y = 87 \pm 1 \) \( \mu \)m and FWHM = \( 2 (2\ln2)^{1/4} \cdot (\sigma_x^2 + \sigma_y^2)^{1/4} \) = 303 \( \pm \) 2 \( \mu \)m. The other parameters are \( g_0 = 3.0 \pm 0.1 \) \( \mu \)V, \( A = 110 \pm 1 \) \( \mu \)V, \( x_c = 63 \pm 1 \) \( \mu \)m, \( y_c = -5 \pm 1 \) \( \mu \)m, \( \theta = 0^{\circ} \pm 0.1^{\circ} \).

Figure S3: Intensity distribution of the THz field at the focal plane. Blue surface: \( \Delta \nu \) map corresponding to the plot reported in Figure 2b (main text). Yellow surface: Gaussian surface fitting with the photovoltage map (equation eS3).

References


Photothermoelectric effect

The THz frequency-induced PTE in a single-gated GFET is based on the asymmetric absorption of electromagnetic energy by free carriers, at one side of the PTE device (between the two antenna halves, connected to source and gate). As a result, a temperature difference \( \Delta T_e \) arises, driving the
diffusion of carriers from the hot to the cold end of the LMH channel. As reported in Refs. [1, 2], when measured in open circuit configuration (drain contact connected to a voltmeter with high input impedance), this charge displacement is balanced by a photovoltage $\Delta u_{\text{PTE}}$ which is proportional to $\Delta T_e$ as $\Delta u_{\text{PTE}} = \Delta T_e (S_g - S_u)$. Here, $S_u$ is the Seebeck coefficient of the ungated region between the source and gate electrodes and $S_g$ is the Seebeck coefficient of the gated LMH channel. Therefore, $\Delta u_{\text{PTE}}$ has the same functional dependency on $V_g$ as $S_u$, thus it is non-monotonic and follows the Mott relation [2]: $S_g = -eL_0 T \cdot \sigma' (\partial \sigma / \partial V_g) \cdot (\partial V_g / \partial E_F)$, where $L_0 = (\pi k_B)^2 / (3e^2)$ is the Lorenz number, $k_B$ is the Boltzmann constant and $E_F$ is the Fermi energy. $\partial V_c / \partial E_F$ can be evaluated from $E_F = h v_F (\pi C_g \delta V_g / e)^{1/2}$ [3], where $h$ is the reduced Planck constant, $v_F = 1.1 \times 10^6$ ms$^{-1}$ is the Fermi velocity and $\delta V_g = |V_g - V_{\text{CNP}}|$.

In a doped LMH channel, this expression of $\Delta u_{\text{PTE}}$ gives rise to two different possible scenarios in terms of the dependence of $R_v$ on $V_g$, depending on whether the ungated LMH region is a $p$-type or an $n$-type semiconductor. These two configurations are schematically summarized in Figure S4. When the ungated LMH is $n$-type, $S_u$ is negative (Figure S4a). The photoresponse sign reverses twice as a function of $V_g$, and we can identify three ranges of $V_g$. For $V_g < \text{CNP}$ (range I) the gated area is $p$-type, $S_g$ is positive and the moduli of $S_g$ and $S_u$ add up giving rise to a positive $R_v$. The single-gated GFET behaves as a $p$-$n$ junction in this range. For CNP $< V_g < 0$ V (range II) the gated area is $n$-type, $S_g$ is negative and $|S_g| > |S_u|$. In this range both $(S_g - S_u)$ and $R_v$ are negative. For $V_g > 0$ V (range III) the gated area is $n$-type, but now $|S_g| < |S_u|$ and the responsivity is again positive, although typically small since the difference between $S_g$ and $S_u$ is smaller in this range. An opposite scenario holds when the ungated graphene is $p$-type and $S_u$ is positive (Figure S4b). Here the signal is negative in range I ($|S_g| < |S_u|$, both positive), positive in range II ($|S_g| > |S_u|$, both positive) and negative in range III ($S_g$ is negative and adds up to $S_u$ in the expression of $\Delta u_{\text{PTE}}$). For $V_g \sim 0$ V the PTE response is typically small, since $S_g - S_u \sim 0$. Since, at CNP, $S_g$ is zero, $\Delta u_{\text{PTE}}$ (CNP) = $- \Delta T \cdot S_u$.

Figure S4: Photothermoelectric effect. Qualitative representation of the photothermoelectric $R_v$ as a function of $V_g$ for a n-type (a) and p-type (b) GFET.
References


Responsivity evaluation

The responsivity ($R_v$) is the ratio between the signal (photovoltage or photocurrent) and the optical power ($P_a$) that reaches a detector and is one of the most important figures of merit for a THz detector. In general, there are two ways to evaluate the responsivity, which are distinguished by the calculation of the quantity $P_a$. The first is the internal responsivity, in which $P_a$ accounts for the power absorbed by the active element (e.g. the graphene flake), which is a fraction of the total power that is shed onto the active element itself. The second is the external responsivity, which assumes that all the power that is delivered to the active element is therein absorbed. In this paper, we evaluate the external responsivity and we assume that all the power coupled to the device is absorbed within the single layer graphene.

We calculate the optical power that reaches the detector through the relation: $P_a = P_t \cdot (A_\lambda/A_{\text{spot}})$, where $P_t$ is the total power that is focused on the detector and $A_\lambda/A_{\text{spot}}$ is the ratio between the diffraction limited area ($A_\lambda = \lambda^2/4$) and the beam spot size. The choice of this normalization procedure is the most common way to normalize the power in the evaluation of the external responsivity [1-6]. This takes into account the fact that both the antenna size and the antenna scattering cross-section are much smaller than the beam spot size, therefore only a fraction of $P_t$ is actually delivered to the graphene channel. However, the diffraction limited area is typically larger than the antenna size (as in our case); basically, this takes into account that, for a given wavelength, it would not be possible to squeeze a free-space photon into the sub-wavelength-sized antenna. When evaluated with this approach, the external $R_v$ reproduces the figures of merit that one can reach after combining the detector with an optimized focusing system.

When the detector size is smaller than the beam spot size there are other possibilities to evaluate the external responsivity. Here we list some notable examples:

1. The external responsivity can be calculated considering $P_a = P_t$ [7]. This method underestimates $R_v$ with respect to the one proposed in this manuscript, unless the antenna (or coupling element) is larger than the beam spot size. If the detector is much smaller than the spot size, the
responsivity value obtained with this method is strongly dependent on the optical setup (improving the focusing results in a larger $R_v$, and vice versa), thus it can give underestimated outcomes.

2. The active area can be considered equal to the detector physical size, which includes the size of the antenna, if present [8,9]. This method is typically used for multi-pixel architectures. Being the pixel size typically smaller than the wavelength, the physical area of the detector results to be smaller than the diffraction limited area, therefore this method overestimates $R_v$ with respect to the one proposed in this manuscript.

3. The active area can be considered to be equal to the area of the active element $P_a = P_t \cdot (A_e/A_{spot})$. Here $A_e$ is the area of the structure that absorbs the THz energy (e.g., a graphene flake). This method largely overestimates $R_v$ with respect to the one proposed in this manuscript. This is typically used in cases in which the active element is not connected to an antenna [10]. However, even in this case, the metallic structures connected to the active element (e.g., the electrodes) create a local field enhancement in close proximity of the metal and onto the active element itself, therefore increasing its effective scattering cross section.

References


**Noise equivalent power evaluation**

Here we describe and motivate the rationale of evaluating the noise equivalent power (NEP) under the assumption that the noise spectral density (NSD) of the detectors is dominated by thermal fluctuations (Johnson-Nyquist noise).

The NEP denotes the minimum required incident light power to achieve a signal to noise ratio of 1 at 1 Hz bandwidth and it is calculated as the ratio between the NSD and the detector responsivity $R_v$. In the present configuration, there are four main sources of noise that contribute to the total detector noise figure: the Johnson-Nyquist noise ($N_j$), the flicker noise ($1/f$, or telegraph noise) [1], the shot noise [2] and the generation-recombination noise [3]. In the following, we describe them individually.

The Johnson noise, in a conductor, is the electronic noise due to the thermal fluctuations of charge carriers, which results in a random voltage fluctuation ($V_{th}$) at the conductor ends. Its power spectral density is related to the conductor resistance $R$ and to its temperature $T$ via $N_j^2 = \langle V_{th}^2 \rangle = 4k_B T R$, where $k_B$ is the Boltzmann constant. In our case, the contribution of $N_j$ to NSD is $\sim 10 \, \text{nVHz}^{\frac{1}{2}}$ at 300 K.

The flicker noise is related to a direct current in a conductor. Since the detectors are unbiased (between source-drain) and the operating reference frequency of our lock-in experiments is 1.333 kHz (as described in the Methods section), we can safely neglect the $1/f$ contribution, which is only significant at low operating frequencies.

Unlike thermal noise, the shot noise is related to a current flux and depends weakly on the temperature at which the system is operating. The shot noise of a quantum conductor typically increases under THz illumination due to the possibility of photon-assisted shot noise (PASN) phenomena [2]. However, since our detectors are operating at room temperature and under zero applied bias, the contribution of the shot noise to the total noise figure is expected to be orders of magnitude lower than $N_j$ [2] and can be neglected even under illumination.

The generation-recombination noise, in a semiconductor, originates from the random nature of the carrier generation and recombination processes, which results in a randomly varying carrier density. Its contribution to the NSD increases with the current density that is flowing in the conductor. In our case, this source of noise can be neglected, since its amplitude drops below $N_j$ under zero-bias and for current densities $< 1 \, \text{μA/mm}$ [3].

Following these considerations the Johnson-Nyquist noise is a safe approximation for the detector NSD.

In order to provide a further proof to this statement, we tested the noise of the detectors without illumination. We turned off the pulsed electrical excitation to the QCL. We set the gate
biases for the two devices at the value for which we have the maximum THz signal. We record the signal with the lock-in amplifier, passing through a voltage pre-amplifier with gain 100 and bandwidth 200 MHz, input impedance 1 MΩ. For both sample A and sample B we obtain a noise floor <1 µV, which corresponds to a noise spectral density of 10 nVHz\(^{1/2}\). This value is in good agreement with the Johnson-Nyquist noise floor (10 nVHz\(^{1/2}\) for sample A; 8 nVHz\(^{1/2}\) for sample B).

References


Noise Equivalent Energy

The combination of low response time (τ) and high sensitivity (low NEP) can be described in terms of a figure of merit, which is given by the quantity NEP × τ, referred to noise equivalent energy (NEE). The NEE can be intuitively described as the energy resolution of a detector and it is commonly used in pulsed laser physics. If we compare our detectors with other room temperature THz direct detectors we obtain the chart below.

![Figure S5: Noise equivalent energy (NEE). Horizontal axis: response time. Vertical axis: noise equivalent power. Log-log scale representation of recent results obtained with direct THz detectors operating at room temperature at frequency > 1.5 THz. Black points represent results retrieved from literature or in commercially available devices. Blue points represent the results obtained in this work (sample A and sample B). The green shaded area represents the theoretical speed limit for graphene-based devices, as reported in [2]. Diagonal lines represent points where NEE is constant. References are given in the text.](image)
References


[3*] Same as ref.[3], but the NEP is calculated using the same normalization of the power employed in this manuscript (diffraction limited area normalization, see S5).


