High-Harmonic Generation Enhancement with Graphene Heterostructures

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High-harmonic generation (HHG) in graphene heterostructures, consisting of metallic nanoribbons separated from a graphene sheet by either a few-nanometer layer of aluminum oxide or an atomic monolayer of hexagonal boron nitride, is investigated. The nanoribbons amplify the near-field at the graphene layer relative to the externally applied pumping, thus allowing the observation of third- and fifth-harmonic generation in the carbon monolayer at modest pump powers in the mid-infrared. The dependence of the nonlinear signals on the ribbon width and spacer thickness, as well as pump power and polarization are studied, and enhancement factors (EF) relative to bare graphene reaching 1600 and 4100 for third- and fifth-harmonic generation, respectively, are demonstrated. The work supports the use of graphene heterostructures to selectively enhance specific nonlinear processes of interest, an essential capability for the design of nanoscale nonlinear devices.

1. Introduction

High-harmonic generation (HHG) has been intensely investigated as a route toward the generation of coherent attosecond radiation in the extreme ultraviolet and X-ray spectral regions. Atomic gases have thus far been the most successful among demonstrated systems for HHG,[1,2] although they require high vacuum, making them impractical for the design of integrated devices. As a result, the development of solid-state HHG systems has become an important challenge, with promising results demonstrated for various crystalline materials,[3–5] in nonlinear metasurfaces[6–9] and proposals have been made to enhance HHG using layered nanostructures.[10] However, the mechanisms of HHG in solid state appear to be fundamentally different from that in atomic gases, and are highly sensitive to crystal and polarization orientations,[11,12] as well as to the fundamental optical properties of the materials. In particular, theoretical models proposed for solid-state HHG have identified both interband transitions[11] and intraband electron dynamics[12] as radically different sources for the required anharmonicity.

Graphene constitutes an appealing choice of material in this context because it features sizeable intraband and interband contributions to the linear[13–16] and nonlinear[17–19] conductivity in the infrared (IR). Moreover, as a 2D material, it simplifies the conditions needed to achieve phase matching of the signal generated at different spatial locations (e.g., this is automatically guaranteed for pumping at normally incidence), which commonly limits the strength of nonlinear processes in thick
crystals. Third,[13,14,20–24] and second-order[25] nonlinearities have already been demonstrated in graphene, while higher-order harmonic generation has also been observed.[23,26–29] These results have stimulated proposals for the exploitation of the nonlinear response of graphene to implement devices for quantum technology.[30–33]

The conical electronic band structure of graphene has been argued to boost the efficiency of intraband-mediated HHG,[37,38] However, most measurements of HHG in graphene have identified a thermal origin and focused on THz frequencies.[26–28] Recent work has been reported on the observation of mid-IR HHG up to the fifth harmonic in multilayer graphene,[23] as well as mid-IR HHG from monolayer graphene, though this has required extremely high peak intensities of ≈1TW cm⁻²,[29] at which ablation of the material is expected to take place.[16] An empirical demonstration of the sought-after efficient mid-IR HHG from monolayer graphene is still awaiting, to the best of our knowledge.

In this paper, we report on the observation of fifth-harmonic generation (FHG) from monolayer graphene in the mid-IR spectral range with modest pumping peak intensities of <1 GW cm⁻². This result is made possible by the fabrication of heterostructures in which graphene is accompanied by metallic components to boost the external near field actually acting on the carbon sheet. In this work, we refer to the entire structure, including the nanoribbons, as our heterostructure. More precisely, we fabricate heterostructures made of graphene and gold nanoribbons that are separated by an insulator layer of either a 5 nm-thick layer of aluminum oxide (Al₂O₃) or a monolayer of hexagonal boron nitride (h-BN). Similar heterostructures have also been used to launch acoustic graphene plasmons,[37–39] observe plasmon-mediated third-harmonic generation (THG),[40] and enhance HHG at THz frequencies.[41] Here, the nanoribbons act as mid-IR antennas, serving to enhance the incident pump field in the graphene layer and, consequently, boost the resulting harmonic generation. Importantly, we corroborate that ribbons exhibit no observable THG or FHG signals without the graphene layer. Upon investigation of the role of the insulator spacer thickness s and the ribbon width W, we find that different ribbon widths are obtained to optimize the THG and FHG efficiencies, a surprising result that we attribute to an increased absorption of the fifth harmonic field by plasmonic modes in the metal ribbons at the generated optical wavelengths. Our findings are pivotal for the design of high-efficiency nanoscale nonlinear frequency-conversion devices.

2. Results

A schematic of our heterostructure design is shown in Figure 1a. The samples consist of a SiO₂-Si-SiO₂ substrate, on which a monolayer of graphene is grown by chemical vapor deposition. This is then covered by a dielectric: either a monolayer of h-BN or a 5nm film of Al₂O₃. On top of that, arrays of gold nanoribbons with different widths W and fixed interribbon gaps of g = 50 nm are etched using electron beam lithography and lift off (see refs. [37, 40] for more details). Figure 1b shows a typical scanning electron microscopy (SEM) image of the resulting high-quality metal edges of a representative sample. In Figure 1e, we plot the simulated electric field in the graphene layer for our heterostructures using a monolayer of h-BN as a spacer for a variety of ribbon widths. The linear response of the system was calculated using a commercial finite-element method software (COMSOL Multiphysics). Due to the symmetry of the system, a 2D domain coupled with periodic boundary conditions was employed. All the geometrical and illumination parameters were taken to be the ones used in the experiment, and detailed in this work. The dielectric permittivities of Au, h-BN, Al₂O₃ and SiO₂ were respectively taken from ref. [42–45], and the optical conductivity of graphene was introduced as described in ref. [46]. In Figure 1e, one can observe the antenna-like effect of the ribbons: the field is primarily concentrated in gap at the edge of the ribbons. Simulations for our heterostructures with the 5 nm Al₂O₃ spacer are qualitatively similar. We will refer to the different structures as follows: the samples with the 5 nm Al₂O₃ spacer and ribbon width W are called AlO₂₀, while we refer to the samples with the monolayer h-BN spacer and ribbon width W as HBNW. For example, AlO₂₀₀ is the heterostructure with a 5 nm Al₂O₃ spacer, g = 50 nm and W = 200 nm.

To measure the nonlinear emission produced by our samples we use a modified z-scan setup, in which the nonlinear signal (either FHG or THG) is measured while the sample is moved along the z axis through the focus of the laser beam (see Figure 1f). Our pump beam is a linearly-polarized pulsed laser with a ~260fs pulse duration, a central wavelength of 3900 nm (0.225 eV), and a 76 MHz repetition rate. This beam is generated by an optical parametric oscillator (OPO), fed by a mode-locked femtosecond Ti:sapphire laser. We use a half-wave plate (HWP) to rotate the polarization of the incoming beam to that set by the polarizer (pol). A lens with a 5.95 mm focal length focuses the pump beam down to a waist of ~13.5 μm. When the sample is moved parallel to the pump beam (along the z axis), the nonlinear emission occurs most efficiently where the fluence is maximum (i.e., at the focal point). Afterward, a lens with an 11 mm focal length collimates the beam, which is then sent through a set of spectral filters that separate the nonlinear emission from the pump beam. The resulting nonlinear signal is coupled into a multimode fiber, which can be sent to one of two detectors. The THG signal (at ~1300 nm) is measured using a large-area SNSP detector from PhotonSpot with a ~60% detection efficiency at this wavelength. We combine this detector with a Gemini interferometer from the company NIREOS to measure the spectrum of the THG. To acquire the FHG signal, which is centered around 780 nm, we connect the fiber to a single-photon sensitive silicon Andor spectrometer with a resolution of 1 nm and a detection efficiency of ~10%.

To verify the origin of our nonlinear signals, we perform z-scans on various regions of our samples. As shown in Figure 1b, neither the substrate (green diamonds) nor the gold nanoribbons without the graphene layer (green circles) display any measureable nonlinear response. However, both graphene (red triangles) and graphene with nanoribbons (red squares) show a significant nonlinear signal at the focal point of the pump beam (see Figure 1c). These control measurements demonstrate that only the graphene layer contributes to any measurable nonlinearity in our setup. Moreover, they already
show the very large nonlinear enhancement provided by the gold nanoribbons.

To further verify that our observed nonlinear signals are indeed associated with HHG, we measure their spectra. As displayed in Figure 2a, we find a THG signal at ≈1300 nm and a FHG signal at ≈780 nm. Both of them are at the expected wavelengths for our 3900 nm pump beam. In addition to these two signals, we observe broadband white light generation due to thermal photoluminescence from the graphene layer. This has been previously reported in ref. [47], and, as we discuss in the Appendix, the model presented therein fits our observed spectrum.

Owing to the geometry of the gold nanoribbons, the pump field enhancement is strongly dependent on the polarization of the input light. As shown in Figure 2b, the enhancement and the resulting nonlinear signal are maximized when the light polarization is perpendicular to the direction of the nanoribbons. However, when the polarization is rotated away from such perpendicular direction, the nonlinear signal decreases as (cos θ)^6 and (cos θ)^10, for the THG and FHG signals, respectively, reaching a minimum when the polarization is oriented along the nanoribbons. At this point, the signal strength is even lower than that of planar graphene without gold nanoribbons because of the screening produced by currents induced in the metal. In Figure S1, Supporting Information shows similar measurements performed on graphene without the nanoribbons, which demonstrate only a small (few percent) polarization dependence of the nonlinear signals, attributed to birefringence of the silicon substrate.

To better understand the nature of the enhancement, we first study its dependence on the material and thickness s of the dielectric spacer between the graphene and gold nanoribbons, and...
then proceed to characterize the dependence of the third- and fifth-harmonic signals on the nanoribbon width. To quantify the effect of the spacer material and thickness, in Figure 3a we show the enhancement of the THG signals for both ALO_500 (dark green squares) and HBN_200 (bright green triangles). These are compared to the signal from bare graphene (red squares). We fit the nonlinear signals as a function of input power (straight lines) assuming a third order dependence and with the interception point in the logarithmic plot (which determines the magnitude of the third-order signal) as the only fitting parameter. The enhancement factor (EF) is then given by the difference between these interception points. We find an EF of 26\pm1 with the 5 nm Al_2O_3 spacer and an EF of 1600\pm500 with the monlayer h-BN spacer. In Figure 3b, we show a similar analysis for the FHG signals, assigning a fifth-order dependence of the signal. Here, we present measurements from ALO_500 (dark blue squares) and HBN_70 (bright blue squares). We find an EF of 260\pm30 for the 5 nm Al_2O_3 spacer and an EF of 4100\pm600 with the h-BN spacer. While it would in principle be possible to grow the gold ribbons directly on the graphene, this would not significantly enhance the field concentration. For example, ref. [37] noted that, when using a nonlocal description of the graphene conductivity, spacer thicknesses below one monolayer only marginally increase the field concentration. We discuss the width dependence in detail below.

Our power-scaling measurements indicate that we remain in the perturbative regime for both the THG and FHG measurements, in which the power of the nth-order harmonic scales with \( p_{\text{THG}} \). Deviation from the expected power dependencies is only observed for the highest incident powers used here. As pointed out in
ref. [40], this is caused by an increased electron temperature,\(^{[30]}\) which reduces the nonlinearity. Efficient HHG for higher-harmonics in gas media typically takes place in the nonperturbative regime, wherein the power of all harmonics scales as \(p^{2}\).\(^{[51]}\) This limit has also been reached in solid-state HHG,\(^{[4,52]}\) including in graphene.\(^{[29]}\) We were unable to reach this limit either in extended graphene or in our heterostructures without damaging the samples. In particular, we found that for average pump powers above \(\approx 30\) mW \((\approx 1\) GW cm\(^{-2}\)), the power of the THG and FHG signals began to permanently decrease after several seconds, presumably due to laser-induced structural modifications, as found in ref. [36].

To quantify the effect of the nanoribbon width on the THG and FHG signal enhancement, in Figure 4 we compile a series of measurements performed for devices with the monolayer h-BN spacer. For the Al\(_2\)O\(_3\) samples, we were only able to measure a FHG signal using ALO\(_{500}\). In our attempts to measure FHG in the Al\(_2\)O\(_3\) samples with other widths, we observed laser-induced damage of the nanoribbons before detecting FHG. As such, we will not discuss the width dependence in that sample. In panels (a) and (b), we show the third- and fifth-harmonic signals as a function of input power for nanoribbon widths of 20 to 500 nm with the monolayer h-BN spacer (yellow to blue) and bare graphene (red squares). Again, the points represent experimental data and the straight lines are fits, where only the interception points are free parameters (one per line) and the slopes are set to 3 and 5, respectively. Due to the long acquisition times and slow laser power fluctuations, we were unable to acquire reliable FHG power-scaling plots for HBN\(_{20}\), HBN\(_{50}\), and HBN\(_{500}\). Note also that it was necessary to carry out these measurements in planar graphene using higher powers, which resulted in visible damage to these samples for the highest powers used.

In panels (c) and (d), we calculate the effective third-order and fifth-order susceptibilities of the graphene heterostructures,\(^{[13,53]}\) and find the maxima to be \(\chi^{(3)} = 5.6 \times 10^{-6}\) and \(\chi^{(5)} = 1.8 \times 10^{-14}\) esu. These values are several orders of magnitude larger than for bare graphene, highlighting the benefit of such nanostructures in enhancing the nonlinear properties of 2D materials. Note that our extraction of these susceptibilities makes several assumptions about the material properties of the heterostructures (see Experimental Section). Hence, we also

Figure 4. Nanoribbon width dependence of nonlinear enhancement with h-BN spacer. a,b) The power dependence of the THG and FHG signals for bare graphene (red) and graphene with gold nanoribbons of different widths (yellow through blue). For all the data in this figure, the pump wavelength is 3900 nm, resulting in third- and fifth-harmonic signals at 1300 and 780 nm, respectively. The interception points found with the fits, whose slopes are set to 3 and 5, respectively, enable calculating the enhancement of the signals due to the field confinement. In THG, the maximum enhancement occurs at a nanoribbon width \(W = 200\) nm and for FHG it occurs at 70 nm. c) and d) summarize \(\chi^{(3)}\) and \(\chi^{(5)}\) for different nanoribbon widths and given powers \(P = 0.4\) mW and \(P = 10.6\) mW, respectively. Inset: Visible reflection spectra of the 100, 200, and 500 nm structures, colored light to dark blue as in the color bar.
report the direct conversion efficiencies of the different processes, which is simply defined as the ratio of the generated THG or FHG power to the input power. We find a maximum THG conversion efficiency of \( \approx 2 \times 10^{-9} \) with HBN_200 at a pump power of 2.5 mW, and an FHG conversion efficiency of \( \approx 4 \times 10^{-12} \) in HBN_70 with a pump power of 10 mW. While these conversion efficiencies are lower than those that have been achieved in the THz regime,\[^{[26-28]}\] they are significantly higher than in previous work within the mid-IR due to the enhancement produced by the heterostructures in our work. For example, ref.\[^{[23]}\] found THG and FHG conversion efficiencies of \( 3 \times 10^{-39} \) and \( 9 \times 10^{-34} \), respectively, in five-layer graphene.

The nonlinear enhancement in our structures is primarily driven by the concentration of the electric field in the graphene (as indicated by our field simulations shown in Figure 1e). Hence, as a rough approximation, the incoming field \( E_0 \) is increased by a constant factor, so the effective field becomes \( E_{\text{eff}} = \kappa E_0 \), where \( \kappa \) is a field concentration factor. Then, the THG produced by our heterostructures should be

\[
\chi^{(3)} E_{\text{eff}}^3 \rightarrow \kappa^3 \chi^{(3)} E_0^3 \]  

and the THG due to graphene should be

\[
\kappa \chi^{(1)} E_0^2 \]  

Hence, the THG/THG and THG/FHG conversion factors for the heterostructures can be expressed as

\[
\kappa = \frac{P_{\text{THG}}}{P_{\text{THG}}} = \frac{P_{\text{THG}}}{P_{\text{THG}}} \]  

and

\[
\kappa = \frac{P_{\text{THG}}}{P_{\text{THG}}} = \frac{P_{\text{THG}}}{P_{\text{THG}}} \]  

respectively, in five-layer graphene.

We observe that the peak of this plasmonic mode red shifts from about 675 nm for the 100 nm structure to 750 nm for the 500 nm structure. The encroaching plasmon resonance attenuates the FHG signal (generated at 780 nm) more for the wider ribbons, while the THG signal does not undergo this additional absorption. This plasmon-quenching effect accounts for the weaker-than-expected FHG for wider ribbons. Because of this additional absorption, the field concentration factors computed from FHG and THG no longer agree for the larger structures. For example, in HBN_200 we find \( \kappa = 1.8 \pm 0.2 \) using our FHG data and \( \kappa = 3.4 \pm 0.4 \) using our THG data.

3. Conclusion

In conclusion, we have demonstrated that graphene-based heterostructures can be used to enhance nonlinear conversion efficiencies in the mid-IR spectral range at relatively low pump powers. This has enabled us to observe FHG in the perturbative regime, wherein the generated nonlinear signal scales with the input pump power to the power of five. Such enhancement also allows us to observe third- and fifth-harmonic generation in graphene at modest pump powers in the mid-IR. We study the dependence of the nonlinear signals on the ribbon width and spacer thickness, as well as pump power and polarization, and demonstrate enhancement factors for third- and fifth-harmonic generation of up to 1600 and 4100, respectively, compared to bare graphene.

We have shown that the ribbon width is one of the key parameters to adjust in order to optimize nonlinear conversion. For THG, we observe the largest nonlinear signals for ribbon widths of \( \approx 200 \) to 500 nm, an effect stemming from the optimal enhancement of the pumping field acting on the graphene. For FHG, we observe a maximum nonlinear conversion for smaller ribbon widths of 70 nm. For larger ribbon widths, the FHG is likely quenched due to spectral overlap with the plasma resonance of the ribbons, which shifts toward the FHG emission wavelength of 780 nm for larger ribbons. This demonstrates the fine tuning required in order to maximize a structure for a specific generation wavelength. This information is crucial in the design of future plasmonic, nanoscale HHG devices, particularly those intended to generate visible and near-IR wavelengths.

4. Experimental Section

Extraction of the Third-Order and Fifth-Order Susceptibilities: Experimentally, \( \chi^{(3)} \) and \( \chi^{(5)} \) was estimated following the procedure reported in ref.\[^{[13]}\], which was similar to that discussed in ref.\[^{[54]}\]. The measured average power is proportional to the squared modulus of the electric field through the following relation

\[
P(\omega) = \frac{1}{\mathcal{P}} \left( \frac{E}{|E|} \right) \int W \conjugate{E}^2 d\mathcal{P} \]

where \( f \) is the laser repetition rate, \( \tau \) is the temporal pulse width, \( W \) is the Gaussian beam diameter, \( n \) is the effective refractive index of the medium, and \( \varepsilon_0 \) and \( c \) are the permittivity and speed of light in vacuum. In the experiment here reported, \( f = 76 \text{MHz}, \tau = 260\text{fs} \) (FWHM), \( W = 27 \mu\text{m} \), and the refractive index \( n = 2.4 \) was considered to be constant; note, however, that the additional absorption in the heterostructures could modify \( n \).

The generated THG and FHG electric fields in a graphene monolayer of thickness \( d_p = 0.33 \text{nm} \) were related to the electric field at the fundamental pump frequency \( E(\omega_0) \) through the following relations\[^{[53,55]}\]}
\[ E(3\omega_b) = \frac{i\omega_b}{8\pi} \chi^{(3)}(d_E E(\omega_b)) \]
\[ E(5\omega_b) = \frac{i\omega_b}{8\pi} \chi^{(3)}(d_E E(\omega_b)) \]

From Equation (2), it was thus possible to calculate the effective \( \chi^{(3)} \) and \( \chi^{(5)} \) susceptibilities. Note that all the quantities in Equations (1) and (2) were given in SI units, while the \( \chi^{(3)} \) and \( \chi^{(5)} \) values in Figure 3c,d are given in the electrostatic system of units (esu). The expressions

\[ \chi^{(3)}(\text{SI}) = \frac{4\pi}{3(\lambda^4)} \chi^{(3)}(\text{esu}) \]
\[ \chi^{(5)}(\text{SI}) = \frac{4\pi}{3(\lambda^4)} \chi^{(5)}(\text{esu}) \]

provided the relative conversion factors.

**Polarization Dependence of Bare Graphene:** In this section, the polarization dependence of THG from bare graphene is discussed. The upper panel of Figure S1, Supporting Information showed the transmission of the pump beam through the sample as a function of the orientation of its linear polarization. In this experiment, far-field light with a wavelength of 3900 nm was normally incident on the sample from the side of the substrate, composed of a stack of two layers of silica and one layer of silicon (see Figure 1). The silica layers had a thickness of 285 nm, while the silicon layer had a thickness of 500 μm (see ref. [37] for more detail of the sample fabrication). The measurements reported in Figure S1, Supporting Information were carried out on an area of the sample containing only graphene (i.e., there were no gold nanoribbons), just like in the violet triangles in Figure 1a of the main text. The red points in Figure S1, Supporting Information showed a periodic pump transmission through the sample as a function of the incoming linear polarization angle. The measured relative difference in transmission was ~5%. In the lower panel, the green points showed the polarization-dependent normalized THG power together with the normalized cubed transmitted power (blue points). The quantitative agreement between the two explained the polarization dependence of the THG signal.

The physical origin of the polarization effect could be explained by a slight birefringence in the silicon ref. [56]. Another possible explanation was the dependence of the Fresnel reflection coefficient on polarization for off-normal incidence.[57] Indeed, for an interface between air and silica, an incidence angle of ~7° relative to the interface normal was enough to induce a relative difference in reflectance by 5% between s and p polarizations.

**Thermal Radiation Background:** In this section, the physical origin of the background signal observed in the FHG spectrum of Figure 2 was identified as ultrafast photoluminescence from graphene. The significant broadband light emission from graphene under excitation by femtosecond laser pulses was studied in ref. [47] in great detail and was employed here to model the measurements. The ultrafast heating of the electron gas was modeled with a two-temperature model, comprising the electronic temperature \( T_e \) and the temperature of strongly coupled optical phonons \( T_{op} \).

\[ \frac{dT_e(t)}{dt} = \left( \frac{1}{\tau_e} \right) \left[ I(t) - \Gamma(T_e(t), T_{op}) \right] \]
\[ \frac{dT_{op}(t)}{dt} = \left( \frac{1}{\tau_{op}} \right) \left[ \Gamma(T_e(t), T_{op}) T_{op}(t) - T_0 \right] \]

with a phonon decay time \( \tau_{op} = 1 \text{ps} \) and a specific heat capacity of the phonons \( c_{op}(T_{op}) \) and electrons \( c_e(T_e) \) taken to be

\[ c_{op}(T_{op}) = 5.79 \times 10^9 + 1.82 \times 10^4 T_{op} + 1.34 \times 10^4 T_{op}^2 + 5.16 T_{op}^3 \]

and

\[ c_e(T_e) = \frac{18 \times 2.02}{1.38 \times 10^{-26} k_B T_e} \]

respectively, where \( v_F \) is the Fermi velocity and \( k_B \) the Boltzmann constant.

The electron–phonon energy exchange rate \( \Gamma \) was defined as

\[ \Gamma(T_e, T_{op}) = \alpha \left[ n(T_{op}) + 1 \right] \left[ 1 - f(E_{F}, T_e) \right] \frac{E}{E_{F}} \left( 1 - f(E_{F}, T_{op}) \right) \]

where \( E_{F} = 0.18 \text{ eV} \) is the phonon energy, \( n(T_{op}) = \exp(E_{F}/k_BT_{op}) - 1 \) represents the phonon population at temperature \( T_{op} \), and \( f(E, T_e) = \exp(2(E/k_BT_e)) + 1 \) is the Fermi–Dirac distribution for electrons at temperature \( T_e \). The phonon coupling constant \( \alpha \) corresponds to

\[ \alpha = \frac{3}{\pi^2} \frac{\hbar a}{\hbar c} \]

with \( \hbar = 6.6 \times 10^{-34} \text{erg s} \) and \( a = 1.05 \times 10^{-7} \text{ cm} \) as the density of graphene.[58,59]

The calculated electron temperature in graphene for 260 fs pulses and an incident peak intensity of 100 MW cm \(^{-2} \) was plotted in the inset of Figure S2, Supporting Information. To get the corresponding spectral radiant fluency, we use Planck’s law

\[ f(\lambda, T_e) = e^{\lambda} \frac{2\hbar c^2}{\lambda^5} \left[ \exp \left( \frac{\hbar c}{\lambda k_B T_e} \right) - 1 \right]^{-1} \]

The emissivity \( e(\lambda) \) of graphene was calculated via the absorption of light coming from air, taking into account the wavelength-dependent modulation introduced by the cavity formed by the 285 nm layer of glass between the graphene and Si. Assuming that the illuminated graphene area acts like a thermal Lambert emitter, the lens was estimated to collect up to an angle \( \theta = \arctan \left( \frac{D_{eff}/2}{f} \right) \) where \( f = 11 \text{ mm} \) is the focus length and the effective diameter is scaled to match the experimental signal via \( D_{eff} \approx 30% \times 4.5 \text{ mm} \). This was the only scaling parameter, which had been tweaked to match the thermal estimate with the background that was observed in the experiment, as shown in Figure S2, Supporting Information. It compensated alignment issues, as the apparatus was designed to measure HHG, but not white light as the apparatus was designed to measure HHG, but not white light.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or by contact with the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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