Gate- and Optically Controlled Nonlinear Optical Response in Graphene via Non-Perturbative Ultrafast Carrier Dynamics

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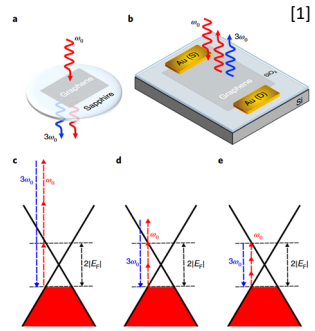
Abstract

We report the ultrafast spectral modulation of nonlinear optical signals in graphene under intense excitation. Utilizing a robust suspended-graphene platform that enables wide-range electrostatic gating, we observe dramatic frequency shifts (up to 8 THz) in third-harmonic (THG) and sum-frequency generation (SFG) driven by pump-induced nonequilibrium carrier dynamics. The magnitude and direction of these shifts are reversibly controlled via the Fermi level and optical excitation. Supported by a quasi-equilibrium hot-carrier theoretical framework, our findings elucidate the interplay between carrier heating and the Fermi level, offering a practical route toward high-speed, gate-tunable nonlinear photonic architectures.

Introduction



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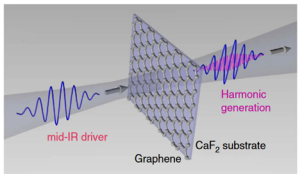

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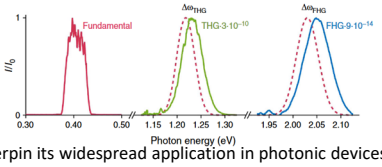
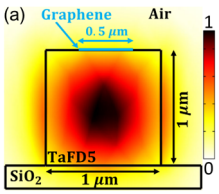
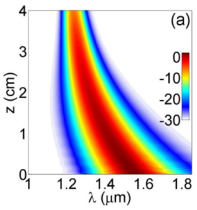


- Graphene's strong nonlinearities underpin its widespread application in photonic devices.
- New mechanisms remain to be discovered in the **non-perturbative** regime.





[3]

- Free-carrier-induced spectral blueshift

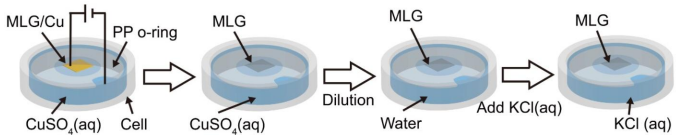
➢ How to obtain a **gate-tunable, high damage threshold** MLG sample ?

➢ How to explain ultrafast carrier induced spectral modulation?

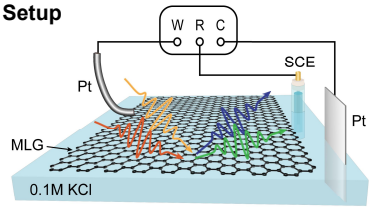
Methods

- **Sample preparation**

[4]

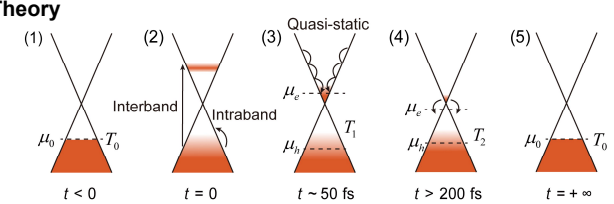


- **Setup**

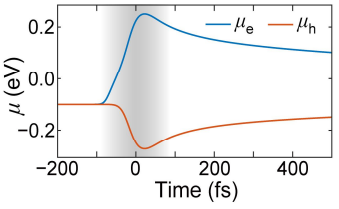


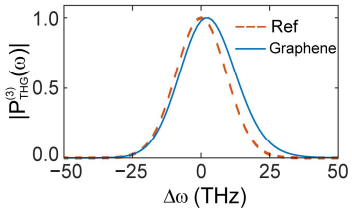
- Gate tunable (Dirac point→-0.6 eV)
- High damage threshold (≥20GW/cm^2 , 50fs IR)

- **Theory**



$$\chi^{(n)}(t) = \chi^{(n)}\left(\mu_e(t), \mu_h(t), T(t)\right); P^{(3)} = \chi^{(3)}A^3(\omega)e^{-3i\omega t} = |\chi^{(3)}(t)|e^{i\varphi(t)}A^3(\omega)e^{-3i\omega t}$$

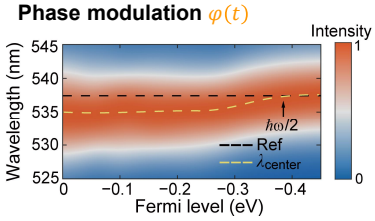


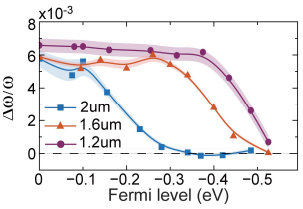


Results

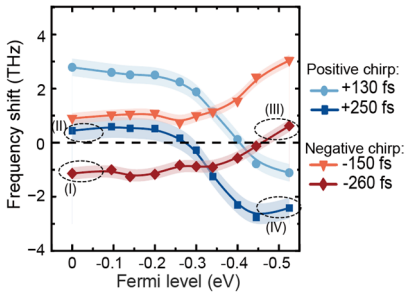
Third-harmonic generation(THG)

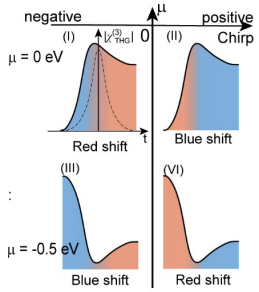
- **Phase modulation** $\varphi(t)$





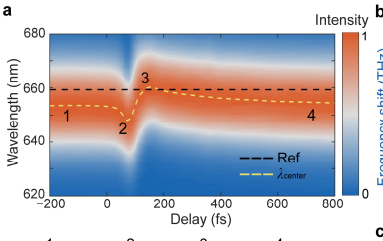
- **Amplitude modulation** $|\chi^{(3)}(t)|$

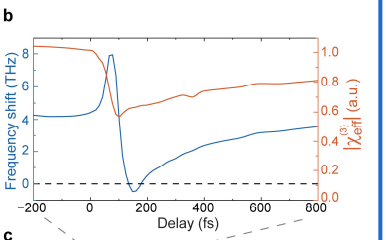


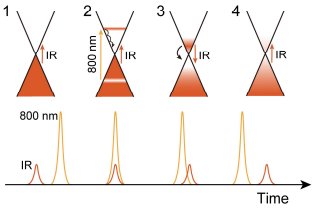


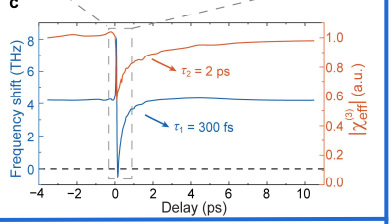
Fs800 pump-THG probe

- **Ultrafast all-optical modulation**



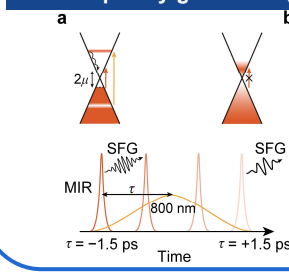


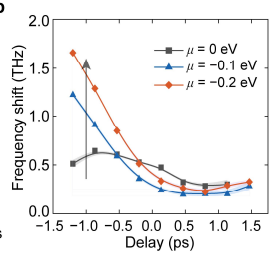




Sum-frequency generation(SFG)

- **Mechanism is universal**





$$\left|\chi^{(2)}_{SFG}(t)\right| e^{i\varphi(t)}$$

Conclusion

Observation: massive **(~8 THz)** and ultrafast **(tens of fs)** spectral modulation of THG and SFG in graphene.

Universal Control: Carrier-mediated mechanism validated across arbitrary nonlinear processes. Proven robust control via Fermi level and excitation pulse.

Impact: Establishes a foundation for dynamically reconfigurable nonlinear optics and explores the frontiers of time-varying photonics.

Reference

[1] G. Soavi et al., Nat. Nanotechnology. 13, 583–588 (2018).

[2] M. Baudisch et al., Nat. Commun. 9, 1018 (2018).

[3] Sahoo, A et al., Phys. Rev. A 104, 063501 (2021).

[4] Y. Xu et al., Nature 621, 1–5 (2023)