

Graphene electrically tuneable third harmonic generation

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Layered materials have a strong nonlinear optical response [1]. Single Layer Graphene (SLG) can provide electrically tuneable nonlinearities over a broad bandwidth thanks to the linear dispersion of Dirac fermions [2]. We show that the THGE in SLG can be modulated by over one order of magnitude by controlling its Fermi Energy (E_F) [3]. We perform gate-dependent measurements on back-gated exfoliated SLG on Si/SiO₂. For the excitation of SLG we use the idler beam of an OPO (Coherent), focused by a 40X reflective objective to avoid chromatic aberrations. The THG signal is collected by the same objective and delivered to a spectrometer (Horiba iHR550) equipped with a nitrogen cooled Si CCD. The idler spot-size is $\sim 4.7\mu\text{m}$. This corresponds to an excitation fluence $\sim 70\mu\text{J}/\text{cm}^2$ for the average power (1mW) used in our experiments. The idler pulse duration is $\sim 300\text{fs}$. Fig.1 compares THGE experiments and theory for $\hbar\omega_0=0.59\text{eV}$ and different T_e over $-700\text{meV}<E_F<+150\text{meV}$ corresponding to $\approx 150\text{V}<V_G<+150\text{V}$. As a function of V_G , SLG displays a THG intensity enhancement by over a factor of 20, starting when $\hbar\omega_0<2|E_F|$. The best agreement between theory and experiments is reached when $\approx 1500\text{K}<T_e<2000\text{K}$. The observed gate-dependent enhancement of the THGE can

be qualitatively understood as follows. The linear optical response of SLG at $T_e=0\text{K}$ has a "resonance" for $\hbar\omega_0 = 2|E_F|$, the onset of intra- and inter-band transitions in SLG. In a similar way, for the SLG third-order nonlinear optical response, resonances occur at $T_e=0\text{K}$ for multi-photon transitions such that $m\hbar\omega=2|E_F|$ with $m=1,2,3$, which correspond to incident photon energies $\hbar\omega_0=2|E_F|$, $|E_F|$, $2/3|E_F|$ [3]. This result paves the way to novel SLG-based nonlinear photonic devices, in which the gate tunability of THG may be exploited to implement on-chip schemes for optical communications and signal processing, such as ultra-broadband frequency converters.

References

- [1] A. Säynätjoki, *et al.*, Nat. Comms. **8** (2017) 893.
- [2] H. Rostami and M. Polini, Phys. Rev. B **93** (2016) 161411.
- [3] G. Soavi *et al.*, arXiv:1710.03694 (2017).

Figures

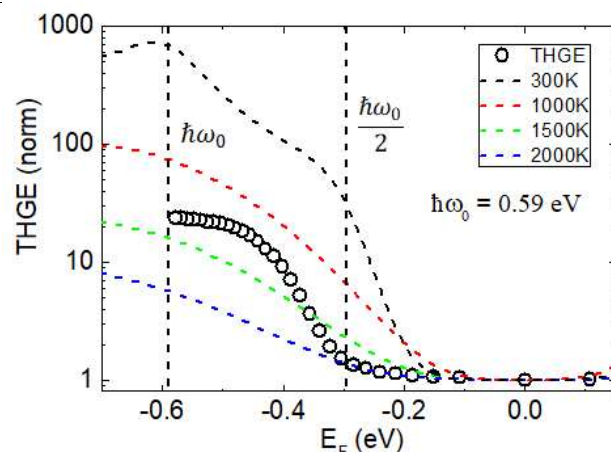


Figure 1: Experiments (circles) and theory (dotted lines) for THGE as a function of E_F and T_e for SLG on Si/SiO₂ at incident photon energies of 0.59eV.