THz Emission From Graphene Induced By Dynamical Photon Drag

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Abstract— We demonstrate broadband coherent THz emission at room temperature from epitaxial graphene under femtosecond optical excitation induced by photon drag effect. We interpret the emitted THz radiation characteristics with a model describing the electron and hole states beyond the usual massless relativistic scheme. This second-order nonlinear effect relies on the dynamical transfer of light momentum to the carriers by the ponderomotive electric and magnetic forces. Finally, our results indicate that optical rectification in graphene can provide emission up to 60 THz, opening new routes for the generation of ultra-broadband THz pulses at room temperature.

I. INTRODUCTION

C urrent terahertz (THz) technologies suffer from the lack of compact room temperature THz sources, limiting the proliferation of consumer applications. Owing to its specific linear dispersion and high electron mobility at room temperature, graphene is attractive for realizing strong nonlinear effects, in particular generation of terahertz radiation. THz generation relying on third-order nonlinearity has been recently demonstrated in graphene excited by optical pulses at normal incidence [1]. Most important in views of applications, second-order nonlinear effects are forbidden by symmetry as graphene is a centrosymmetric material. However, photo-excitation at oblique incidence can itself break the symmetry of the material system, related to the inplane photon momentum [2-3].

II. RESULTS

The investigated multilayer graphene sample is grown on 4H-SiC(0001) and contains typically 35-40 layers: the first four layers near the substrate are heavily doped, whereas the upper remaining layers are quasi-neutral. We use a conventional emission spectroscopy experiment based on a mode-locked Ti:Sa laser delivering 110 fs with an optical fluence ranging up to $35 \,\mu J/cm^2$. All measurements are performed at room temperature. The THz waveform generated by exciting graphene with an incidence angle of 37° is reported in Fig. 1a. and the corresponding amplitude spectrum that consists of a single broad peak centered at 1.25 THz is shown in Fig. 1b. Figure 1d shows that the amplitude of the electric field scales linearly with the excitation fluence, indicating a second-order nonlinear process. Whereas, in usual second order nonlinear processes, the emission would be insensitive to the incidence angle, we observe critical changes: for opposite incidence angles, the time-oscillations show reverse polarity, as shown in Fig. 1e and 1f for f=±14°. We demonstrate that the relative field amplitude is proportional to the in-plane component of the photon momentum, which is a distinctive feature of the dynamical photon drag effect [4].

In order to describe quantitatively the experimental findings and interpret the temporal waveform of the THz signal, we calculated the time-dependent average current generated in one graphene layer excited by femtosecond optical pulses, up to the second order in the exciting electric field (Fig. 1c). We demonstrate that, in contrast to most optical processes in graphene, the next-nearest-neighbour couplings as well as the distinct electron-hole dynamics are of paramount importance in this dynamical photon drag effect.



Fig. 1. a) Electric field waveform emitted by the multilayer graphene under an incidence angle of 37° and its associated spectrum (b). c) Transient nonthermal electron and hole distributions for oblique illumination. d) THz electric field amplitude as a function of the optical fluence. f-g) Time resolved electric field profiles measured for two opposite angles of incidence (f=±14°).

Our results also indicate that dynamical photon drag effect can provide emission up to 60 THz opening new routes for the generation of ultra-broadband THz pulses at room temperature.

References

[1]. D. Sun, *et al.* "Coherent Control of Ballistic Photocurrents in Multilayer Epitaxial Graphene Using Quantum Interference," *Nano Lett.* vol. 10, pp 1293 2010.

[3]Young-Mi Bahk et al., "Plasmon Enhanced Terahertz Emission from Single Layer Graphene," ACS Nano, vol. 8, pp 9089–9096, 2014.

[4]. J. Maysonnave, *et al.* "Terahertz Generation by Dynamical Photon Drag Effect in Graphene Excited by Femtosecond Optical Pulses," *Nano Letters* vol. 14, pp. 5797, 2014.

^[2] Glazov, M. M., Ganichev, S. D. "High frequency electric field induced nonlinear effects in graphene," *Phys. Rep.* vol. 535, pp 101-138, 2014.
[3]Young-Mi Bahk *et al.*, "Plasmon Enhanced Terahertz Emission from