Electronic Supplementary Information

Electrically tunable, plasmon resonance enhanced, terahertz third harmonic generation via graphene

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Graphene Conductivity

Within the random phase approximation and in the absences of magneto-static bias field and spatial dispersion (i.e. the dependency of graphene conductivity on the propagation constant of plasmons), the graphene isotropic complex surface conductivity can be expressed as (assuming $exp(j\omega t)$ time harmonic variation): [1]

$$\sigma(\omega,\mu_c,\Gamma,T) = -j\frac{e^2K_BT}{\pi\hbar^2(\omega - j\tau^{-1})}\left(\mu_c/K_BT + 2\,ln\left(exp\left(-\mu_c/K_BT\right)+1\right)\right) - \frac{je^2}{4\pi\hbar}\,ln\left[\frac{2|\mu_c| - (\omega - j\tau^{-1})\hbar}{2|\mu_c| + (\omega - j\tau^{-1})\hbar}\right] \quad (1)$$

where the first and second terms imply to the intraband and interband conductivities, originated from intraband and interband transitions of electrons. In this relation *-e* is the electron charge, ω is the angular frequency, $\hbar$ is the reduced Planck's constant, *T* is the temperature, τ is the electron-phonon relaxation time, μ_c is the chemical potential representing the graphene Fermi level, K_B is the Boltzmann's constant, and ϵ is the energy. The interband term is approximately calculated by considering the constraint of $K_BT \ll |\mu_c|, \hbar\omega$. For frequencies $\hbar\omega \gtrsim 2\mu_c$ the graphene conductivity is dominated by the interband transition of electrons which is a major mechanism of plasmon loss (via excitation of electron-hole pairs). [1,2] Furthermore in this frequency range the sensitivity of graphene conductivity to the Fermi level and frequency is negligible and posses a universal value of $\sigma_0 \approx e^2/4\hbar$ at $\hbar\omega \gg 2\mu_c$ corresponding to a constant absorption of 2.3% per graphene layer. [3,4] On the other hand, for frequencies $\hbar\omega < 2\mu_c$ (corresponding to THz frequencies for moderately doped graphene samples), Pauli principle will block interband transitions (in n-doped graphene the electron states in resonance in the conduction band are occupied and in the p – doped graphene there are no electron available for the interband transition) and intrabnd transition of electrons (free carrier absorption) play significant role. In this frequency range which is considered as a low loss regime (the real part of conductivity is much smaller in comparison with the imaginary part) for graphene operation, the graphene conductivity shows great tunability with Fermi level. [1, 5] This feature would be more interesting by considering the fact that the current continuous development of terahertz technology with myriad applications in sensing, security, quality control in the pharmaceutics industry and so on requires the materials with proper response to the terahertz radiation (such as graphene) which cannot be found in the most of standards materials. The advent of metamaterials has partially reduced this dilemma, however, the complexity in design and difficulties in the fabrication process have remained as obstacles. [6-8]

By defining a volume conductivity for a Δ - thick graphene layer as $\sigma_v \equiv \sigma/\Delta$, the volume current density and so the Ampere's law can be written as $J = \sigma_v E$ and $\nabla \times H = J + j\omega\varepsilon_0 E = (\sigma_v + j\omega\varepsilon_0)E$ respectively. By this way, we can ascribe an equivalent complex permittivity of $\varepsilon_{eq} \equiv \varepsilon_0 + \sigma_i/\omega\Delta - j\sigma_r/\omega\Delta$ and thus an equivalent effective linear susceptibility of $\chi^{(1)}_{eff,g} = -j\sigma/\omega\varepsilon_0\Delta$ to the Δ - thick graphene layer where σ_r and σ_i denote to the real and imaginary parts of graphene conductivity. [9] By the same way, an equivalent nonlinear susceptibility can be designated to graphene defined as $\chi^{(3)}_{eff,g} = -j\sigma_3/\omega\varepsilon_0\Delta$. [10,11]

Although, it seems that more appropriate implementation of graphene as a two dimensional material in the FDTD code would be by its linear and nonlinear conductivities (and so linear and nonlinear currents), the bulk representation of graphene (certainly of finite thickness) by its linear and nonlinear susceptibilities (and so linear and nonlinear polarizations) in this study is only due to the presence of Kerr-nonlinear medium in the graphene environment which must be modelled by polarization.

References

1 G. W. Hanson, *IEEE Trans. Antennas Propag.,* 2008, **56**, 747-757.

2 M. Jablan, H. Buljan, and M. Soljacic, *Phys. Rev. B.*, 2009, **80**, 245435.

3 A. B. Kuzmenko, E. Heumen, F. Carbone, and D. Marel, *Phys. Rev. Lett*., 2008, **100**, 117401.

4 P.R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres, A. K. Geim, *Science,* 2008, **320**, 1308-1308.

5 Q. Bao, and K. P. Loh, *ACS Nano,* 2012, **6**, 3677-3694.

6 C. Rizza, A. Ciattoni, E. Spinozzi, and L. Columbo, *Opt. Lett.,* 2012, **37**, 3345-3347.

7 A.J. Gallant, M.A. Kaliteevski, D. Wood, M. C. Petty, R. A. Abram, S. Brand, G.P. Swift, D. A. Zeze, and J. M. Chanberlain, *Appl. Phys. Lett.,* 2010, **91**, 161115.

8 P. Weis, J.L.G. Pomar, M. Hoh, B. Reinhard, A. Brodyanski, and M. Rahm, *ACS. Nano,* 2012*,* **6***,* 9118-9124.