Theoretical study of the third order optical nonlinearity of graphene

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Due to the gapless linear dispersion of its low energy electronic excitations, graphene exhibits unique and remarkable optical properties: The linear optical response is characterized by a universal conductivity $\sigma_0=\frac{e^2}{4\hbar}$ at wavelengths from the mid-infrared to the visible, with a monolayer absorbance of about ~2.3%; thus graphene can be thought of as either highly transparent or, considering its monolayer thickness, highly absorbing. With its strong optical coupling, broadband absorption, and other novel material and electronic properties, graphene is natural candidate for use in optically controlled devices in photonics and optoelectronics [1,2]. In moving towards any application an important step is understanding the optical nonlinearities of graphene. Third order nonlinearities in graphene have been experimentally investigated by four-wave mixing [3,4], third harmonic generation[5,6], two-photon absorption [7], Kerr effects [2,8], and coherent current control [9]; however, the extracted effective $\chi^{(3)}_{\text{eff}}$ values show discrepancies and strongly depend on measurement method, photon energy, and perhaps sample preparation. Although strong nonlinearities are found in all these experiments, the mechanism has not been well understood. Theoretically, besides the expression provided by Hendry *et al.* [3], Rioux *et al.* [10] investigated two-photon absorption and two-color coherent current injection by using Fermi Golden Rule for pristine graphene, Zhang et al.[11] used the density matrix method to study four wave mixing in the saturation regime in undoped graphene, and Jafari [12] calculated the nonlinear optical response in gapped graphene by adding a mass term. However, a general calculation of third order nonlinearities, even in the simplest linear dispersion approximation, is still missing.

In this paper, we perform a perturbative calculation of the third order optical susceptibilities of doped graphene, using approximations valid around the Dirac points and neglecting scattering and electron-electron interaction effects. In this limit analytic formulas can be constructed for the conductivities. We discuss in detail the results for third harmonic generation, Kerr effects and two-photon absorption, parametric frequency conversion, and two-color coherent current injection. We find a complicated dependence on the chemical potential and photon energies. The linear dispersion causes resonances over a wide photon energy window, and it is possible to obtain large optical nonlinearities by tuning the chemical potential.

For undoped graphene, our model leads to a simple expression for the fully symmetrized conductivity

$$\sigma^{(3);dabc}(\omega_1,\omega_2,\omega_3)=\frac{\sigma_0(\hbar v_F e)^2}{\hbar^4(\omega_1+\omega_2)(\omega_2+\omega_3)(\omega_3+\omega_1)(\omega_1+\omega_2+\omega_3)}$$

with v_F being the Fermi velocity. For doped graphene with a chemical potential $|\mu|$, however, our formulas can be written as

$$\sigma^{(3);dabc}(\omega_1,\omega_2,\omega_3)=\frac{\sigma_0(\hbar v_F e)^2}{\pi|\mu|^4}S^{dabc}\left(\frac{\hbar\omega_1}{|\mu|},\frac{\hbar\omega_2}{|\mu|},\frac{\hbar\omega_3}{|\mu|}\right).$$

We discuss the obtained third order conductivities for third harmonic generation, two photon absorption, Kerr effects, parametric frequency conversion, and two color coherent current injection. As an example, $S^{dabc}(-w,w,w)$ for two-photon absorption and Kerr effects with $w<2$ is plotted in Figure 1. This graph show divergences related to the resonances between the involved photon energy and the chemical potential gap $2|\mu|$. For other frequency combinations, results are quite different. Combined with the tunability of $|\mu|$ by an external gate voltage or chemical doping, this should lead to a novel approach for controlling the nonlinear optical properties of graphene, and indeed to the possibility of graphene-based nonlinear optics on demand.

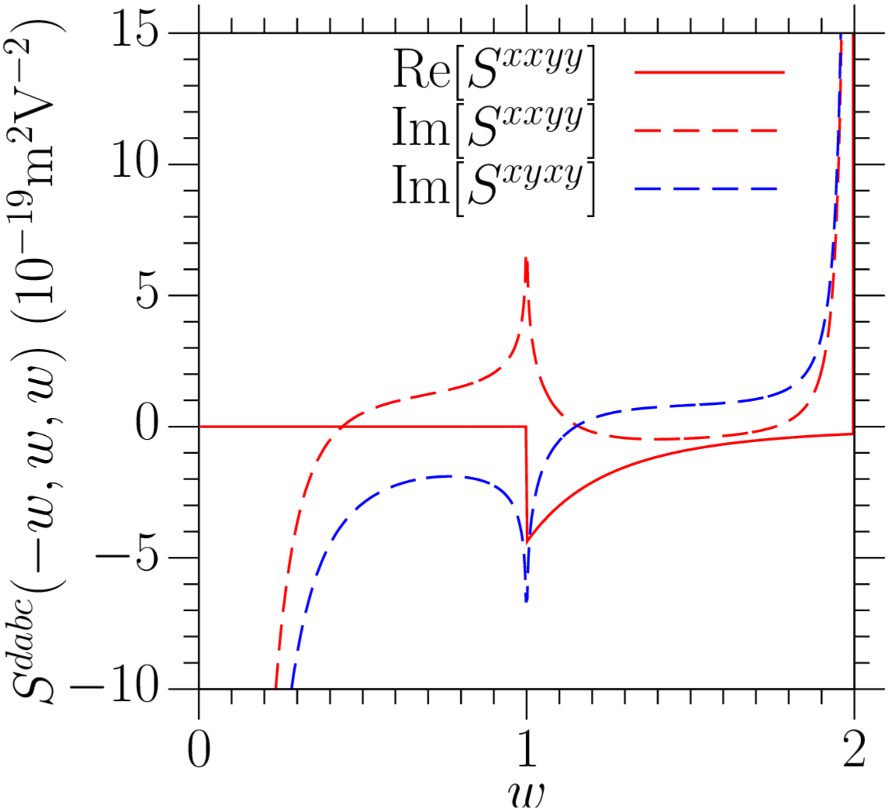

Figure 1: Two-photon absorption and Kerr effect coefficients $S^{dabc}(-w,w,w)$ for $w<2$.

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