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Terahertz photon mixing effect in graphene and topological insulator

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Abstract

We theoretically investigate the nonlinear terahertz photon-mixing effect in graphene and topological insulator. We show that the nonlinear intraband optical absorption in both materials dominate over the linear response under a very moderate external electric field strength. The nonlinear response is thermally enhanced and the optical nonlinearity is well-preserved even in high temperature regime.

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Terahertz Photon Mixing Effect in Graphene and Topological Insulator

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***Abstract*—We theoretically investigate the nonlinear terahertz photon-mixing effect in graphene and topological insulator. We show that the nonlinear intraband optical absorption in both materials dominate over the linear response under a very moderate external electric field strength. The nonlinear response is thermally enhanced and the optical nonlinearity is well-preserved even in high temperature regime.**

I. Introduction and Background

GRAPHENE is an atomically-thin film made up of entirely carbon atoms; three-dimensional topological insulators are materials consisted of bulk insulating bands and two-dimensional gapless surface states [1]. Surprisingly, the two seemingly unrelated materials share a common feature: their low energy electrons follow an unusual linear energy dispersion in analogue to a massless relativistic fermions as described by the relativistic Dirac's equation. These peculiar *quasiparticles* are highly capable of multiple-photon coupling, resulting in exceptionally strong nonlinear optical response. In graphene, the *interband* nonlinear optical response easily dominates over the linear response when a very moderate electric field of 10^3 V/cm is applied [2].

In this work, we theoretically investigate the nonlinear photon mixing effect in graphene and TI in THz regime. By decomposing the externally perturbed quasiparticle dynamics and population distribution function into linear and nonlinear components [4], we found that the *intraband* nonlinear optical response in graphene and TI is thermally enhanced for temperature up to 600 K. The nonlinear response easily dominates when an electric field well-within the experimentally achievable level of 10^4 V/cm is applied. Our results reveals the possibility of graphene and TI-based nonlinear THz optical device capable of room-temperature operation.

II. Theory

In tight-binding description, the Graphene and TI low energy Hamiltonian can be written as $\hat{H} = v_F \hat{\sigma} \cdot \hat{p}$ where $\hat{\sigma} = (\sigma_x, \sigma_y)$ is the Pauli spin matrix, $\hat{p} = (p_x, p_y)$, v_F is 10^6 m/s and 6.5×10^5 m/s [3] for graphene and TI, respectively. The energy dispersion is given as $\varepsilon_{s,p} = s v_F p$ with $s = \pm 1$. The velocity eigenvector is given as $\mathbf{v}_s^{(0)} = v_F \mathbf{p}/p$. We consider minimal coupling scheme where $p \to p - eA$ in the presence of an external field. When an external field is applied, the quasiparticles velocity and population distribution function are perturbed and can be Taylor-expanded in terms of the external field $\mathbf{E}$ to give $\tilde{\mathbf{v}}_s = \sum_{n=0}^{3} \mathbf{v}_s^{(n)}$ and $\tilde{f}(\tilde{\mathbf{v}}_s \cdot \mathbf{p}) = f(\varepsilon_{s,p}) + \sum_{n=1}^{3} f^{(n)}$, respectively, where superscript (n) signifies terms $\propto \mathbf{E}^n$ and $f(\varepsilon_{s,p})$ is the Fermi-Dirac function. The total current density is given by:

$$\mathbf{J}_{total} = -\frac{e}{(2\pi\hbar)^2} \sum_s \int \tilde{\mathbf{v}}_s \tilde{f}(\tilde{\mathbf{v}}_s \cdot \mathbf{p}) \mathrm{d}\mathbf{p} \tag{1}$$

The n-th order nonlinear optical current density can then be obtained by selecting all terms containing E^n.

III. Results

At $T = 0$ K, the nonlinear optical current density can be analytically solved as

$$\mathbf{J}_{T=0}^{(3)} = -is \frac{e^4 v_F^2}{8\pi\hbar^2 \mu} \sum_{\mu\nu\xi} \left(\frac{\mu}{\mu - \varepsilon_{\mu\nu\xi}} \right) \frac{\mathbf{E}_\mu \cdot \mathbf{E}_\nu \mathbf{E}_\xi}{\omega_\mu \omega_\nu \omega_\xi} \times \exp\left\{ i \left[(\mathbf{q}_\mu + \mathbf{q}_\nu + \mathbf{q}_\xi) \cdot \mathbf{r} - (\omega_\mu + \omega_\nu + \omega_\xi) t \right] \right\} \tag{2}$$

where the subscript $m = \mu, \nu, \xi$ indexes the three incoming photons with frequencies ω_ξ and wavevector q_m, and $\varepsilon_{\mu\nu\xi}$ is the energy sum of the three photons. The numerical results of the temperature dependence the third-order nonlinear current at $f = 1.5$ THz is shown in Fig. 1. The third-order nonlinear current is enhanced by (i) lower level of doping and (ii) by elevating the temperature. The underlying reason for (i) is that nonlinear velocity components of charge carriers with lower momentum is relatively larger and thus resulting in stronger non-linear optical response. For (ii), the electron states underneath the Fermi level are thermally evacuated, allowing deeper charge carrier to be optically excited. The current however does not grow indefinitely with increasing temperature. At very high temperature, the charge carrier residing in the $s = -1$ subband (i.e. the lower Dirac cone) are thermally excitable. These $s = -1$ carriers have opposite non-linear velocity in comparison to those residing in $s = +1$ subband. The inclusion of these carriers reduces the overall optical current. The nonlinear optical response of the surface states of TI and the low energy regime of graphene are similar since the quasiparticles in both system are massless Dirac fermions.

To quantify the optical nonlinearity, we define a critical field strength E_c at which the equality $J^{(3)} = J^{(1)}$ holds. The $T = 0$ K critical field is given as

$$E_c\,(T = 0) = \frac{2\omega}{v_F} \left[\frac{2\hbar\omega}{e^2} (\mu - 3\hbar\omega) \right] \tag{3}$$

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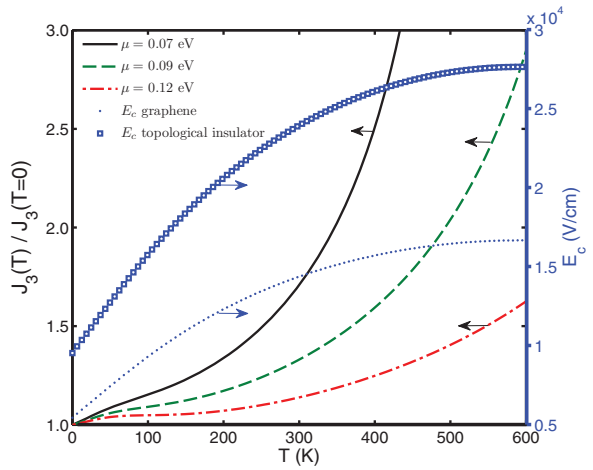

Fig. 1. Temperature spectrum of the third-order nonlinear optical conductivity of graphene and TI at 1.5 THz. The E_c's at $\mu = 0.12$ eV are also shown.

The critical field at finite temperature is numerically solved and is shown in Fig. 1. At 1.5 THz and up to $T = 600$ K, E_c is well within the moderate range of $\approx 10^4$ V/cm. The E_c of TI is slightly larger than graphene since its Fermi velocity v_F is smaller than that of the graphene. This indicates a strong *intraband* optical nonlinearity in graphene and TI which is a direct consequence of the non-parabolic energy dispersion of the 'massless' low energy quasiparticle [5]. The linear dispersion of the massless Dirac fermions results highly non-parabolic. The charge carriers velocity in the presence of an external perturbation is composed of many higher frequency harmonics, giving rise to strong nonlinear optical response even when the electric field strength is only moderate. The thermally enhanced third-order nonlinear optical current suggests that graphene and TI can be utilized as an efficient and practical room temperature THz four-wave mixer.

IV. Conclusion

We found that strong nonlinear intraband optical absorption of graphene and TI in THz frequency regime can be triggered with the application of an experimentally achievable electric field strength in the order of 10^4V/cm. The thermally enhanced third-order nonlinear response suggests that graphene and TI can be utilized as an efficient room-temperature THz photon-mixer.

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