Temperature-dependent of Nonlinear Optical Conductance of Graphene-based Systems in High-intensity Terahertz Field

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Abstract: For multi-photon processed with the linear dispersion in the high-intensity terahertz (THz) field, we have systematically investigated the temperature-dependent nonlinear optical response of graphene-based systems, including single layer graphene, graphene superlattice and gapped graphene. In the intrinsic single layer graphene system, it demonstrates that, at low temperature, nonlinear optical conductivities of the third- and fifth-order are respectively five and ten orders of magnitude larger than the universal conductivity with high-intensity and low frequency THz wave.In the graphene superlattice and gapped graphene systems, the optical responses enhanced because of the anisotropic massless and massive Dirac fermions.

Keywords: Nonlinear; Graphene; Terahertz; Conductance; Transmittance

Introduction

Graphene is a single atomic layer of carbon atoms tightly packed into a highly symmetric two-dimensional honeycomb lattice [1]. The spectrum of graphene quasiparticles, electrons and holesare described by an effective Dirac equation with a vanishing effective mass [2]. The massless energy dispersion of graphene quasiparticles leads to many unique physical properties, such as the electron-hole symmetry and a half-integer quantum Hall effect [3-5], a finite conductivity of 4 e^2/h even when the carrier concentration tends to zero [3], the strong suppression of weak localization [6-8] and ultrahigh carrier mobility [1]. Therefore, graphene is considered as a promising material for electronic applications.

The optical properties of graphene have been studied both experimentally [9-17] and theoretically [18-30]. In the terahertz to the far-infrared (FIR) spectral regime, optical conductance of graphene-based systems attracts many researchers' interests due to the ongoing search on viable terahertz detectors and emitters. Graphene is traditionally a poor conductor in this part of the spectrum, with the universal conductivity $\sigma_0 = e^2/4\hbar$ leading to absorb only 2.3% at normal incidence per graphene layer [11]. As discussed by Miknailov et al. [21,22], they developed a quasi-classical kinetic theory to predict the stronger nonlinear electromagnetic response of graphene under electromagnetic radiation at different frequencies ranging from microwaves to infrared. These studies show that irradiation of graphene by an electromagnetic wave of frequency ω should lead to excitation of odd high-harmonics (i.e. 3ω, 5ω, 7ω, $\cdots$). This may make graphene as a simple and natural frequency multiplier and open up exciting opportunities for using graphene in terahertz electronics. Subsequently, a series of theoretical works were carried out to seek for the strong nonlinear terahertz

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responses of graphene-based systems, such as single layer graphene (SLG) [23-25], bilayer graphene (BLG) [26], graphene p-n junction [27], graphene nanoribbons (GNRs) [28], semihydrogenated graphene (SHG) [29] and Kronig-Penney type (KP) graphene superlattice [30]. As studied by Hendry et al. in 2010 [15], they first measured the coherent nonlinear optical response of single- and few-layer graphene using four-wave mixing. Their results demonstrate that graphene exhibits a very strong nonlinear optical response in the near-infrared spectral region. The large optical nonlinearity originates from the interband electron transitions and is eight orders of magnitude larger than the nonlinearities observed for dielectric materials without such transitions.Recent experimental progress presented the results on studying the carrier dynamics in epitaxially grown graphene at higher photon energies from 10 to 250 meV [16], as well as the nonlinear THz response of graphene excited at electric field strengths in the nonperturbative regime [17].

The theoretical progresses have been shown that the linear energy dispersion of quasiparticles, electrons and holes in graphene leads to the strongly nonlinear optical response of this system in the terahertz to the FIR regime [21]. As a consequence, graphene is a rather strong nonlinear material. Wright and Xu et al. [23,24] adopted a quantum-mechanical approach to study the linear, third-order and fifth-order nonlinear optical response of intrinsic SLG under an electric field of around 10^3 V/cm, which indicates these relative nonlinear terms make graphene a potential applications for photonic and optoelectronic device. Zhou et al. [25] also treated the same topic with more rigorous perturbation theory. Yee Sin Ang et al. [28, 29] investigated the optical conductivity of SHG and KP graphene superlattic. It has been shown that SHG possesses a unique potential as a two-color nonlinear material and the total optical response of the graphene superlattice was enhanced due to the formation of anisotropic Dirac fermions in the terahertz frequency region. It was also revealed that the band structure isotropy is not a prerequisite for the strong optical nonlinearity in graphene.

Previously letters only studied the third- and fifth-order nonlinear optical response to each graphene system with frequency respectively, and did not mention the temperature factors when calculating the relationship between the transmittance and the frequency in the terahertz regime. The main purpose of this paper is to theoretically investigate the temperature-dependent nonlinear optical conductance of different graphene-based systems in high-intensity terahertz field based on the simplified model for multiphoton process with linear dispersion. Firstly, we investigated the strong nonlinear optical response of intrinsic single layer graphene systematically, and obtained relationship between trans-

mittance versus temperature and intensity. Then, we studied the nonlinear optical response of KP graphene superlattice,considering the anisotropy massless Dirac fermions. Moreover, the strong optical nonlinearity existed in gapped graphene was revealed.

Theory and discussion

Massless Dirac fermions

We adopted the approach that couples the massless Dirac fermions to the time-dependent electric field quantum mechanically to calculate the nonlinear terms, both in high-order electrical field and in multiple frequencies.When a time-dependent external field $E(t) = E_0 e^{i\omega t}$ is along the x-axis, the low energy tight binding Hamiltonian of graphene-based systems can be written as,

$$H = v_F \begin{pmatrix} 0 & p_- + eA_- \\ p_+ + eA_+ & 0 \end{pmatrix} \quad (1)$$

where $P_{\pm} = P_x \pm \lambda i P_y$, $A_{\pm} = A = (E_0/i\omega)e^{i\omega t}$ and $v_F \approx c/300 = 10^6$ m/s is the Fermi velocity. λ is an anisotropy parameter, which modifies the y-direction group velocity by $v_y = \lambda v_F$, and can be continuously tuned in the range of $0 < \lambda \leq 1$. The massless Dirac fermions is perfectly isotropic in all directions in the SLG which is the isotropic case $\lambda = 1$, but travels anisotropically with v_F in the direction along the periodicity of the KP potential (x-direction) and reduced velocity of λv_F in the direction perpendicular periodicity (y-direction) in the KP graphene superlattice. The time-dependent two-component wave function can be expanded in the basis set,

$$\psi(\mathbf{p},t) = \sum_{n=0}^{\infty} \begin{bmatrix} \alpha_n(\mathbf{p}) \\ \beta_n(\mathbf{p}) \end{bmatrix} e^{i(n\omega - \varepsilon/\hbar)t} \quad (2)$$

where $\varepsilon = v_F|P|$, by substituting Eq. (1) and (2) into the Schrödinger equation $i\hbar(\partial\psi/\partial t) = H\psi$. Due to the orthonormal relation of $e^{in\omega t}$, we can write the coupled recursion relations for the spinor components,

$$\alpha_n = \frac{eE_0 v_F}{i\omega\hbar} \times \frac{v_F p_- \alpha_{n-1} + (v_F p - n\hbar\omega)\beta_{n-1}}{n\omega(n\hbar\omega - 2v_F p)}$$
$$\beta_n = \frac{eE_0 v_F}{i\omega\hbar} \times \frac{v_F p_+ \beta_{n-1} + (v_F p - n\hbar\omega)\alpha_{n-1}}{n\omega(n\hbar\omega - 2v_F p)} \quad (3)$$

The recursion relation couples the n photon processes to the $n - 1$ photon processes. From the solutions to Eq. (3) we can calculate the nth order total current,

$$J_n = \frac{1}{2\pi^2} \int N(\varepsilon)p\,dp\,d\theta \mathrm{Re}\Big(\sum_{n=0}^{\infty} \alpha_i^* \beta_{n-i} + \beta_i^* \alpha_{n-i}\Big) \quad (4)$$

where $N(\varepsilon) = n_F(-\varepsilon) - n_F(\varepsilon) = \tanh(\varepsilon/2k_B T)$ is the temperature factor.

The linear ($n = 1$) optical conductivity of graphene-based systems, which is a single photon process where an electron absorbs a photon making a transition from the valence band to the conduction band is described as,

$$\sigma_1 = \sigma_1(\omega) = \frac{\sigma_0}{\lambda} N\left(\frac{\hbar\omega}{2}\right) \tag{5}$$

Because of time-reversal symmetry, the second and fourth order solutions make no contributions to the total current.

The third-order nonlinear ($n = 3$) optical conductivity is made up of two components: (i) $\sigma_3(\omega)$, single-frequency term which corresponds to the absorption of two photons and the simultaneous emission of one photon; and (ii) $\sigma_3(3\omega)$, triple-frequency term which corresponds to the simultaneous absorption of three photons, is described as,

$$\begin{aligned} \sigma_3 &= \sigma_3(\omega) + \sigma_3(3\omega) \\ &= \frac{\sigma_0}{\lambda} \frac{2e^2 v_F^2 I}{\varepsilon_0 c\hbar^2 \omega^4}[N_{31}(\omega)e^{i\omega t} + N_{33}(\omega)e^{i3\omega t}] \end{aligned} \tag{6}$$

where $I(\text{W/cm}^2)$ is the intensity and ε_0 is vacuum permittivity. The temperature factor are given by $N_{33}(\omega) = 13/48 N(\hbar\omega/2) - 2/3 N(\hbar\omega) + 15/16 N(3\hbar\omega/2)$ and $N_{31}(\omega) = 2N(\hbar\omega)$.

The fifth-order nonlinear ($n = 5$) optical conductivity is made up of three components: (i) $\sigma_5(\omega)$, single-frequency term which corresponds to the absorption of three photons and the simultaneous emission of two photons; (ii) $\sigma_5(3\omega)$, triple-frequency term which corresponds to the absorption of four photons and the simultaneous emission of one photon; (iii) $\sigma_5(5\omega)$, five-frequency term which corresponds to simultaneous absorption of five photons, is described as

$$\begin{aligned} \sigma_5 &= \sigma_5(\omega) + \sigma_5(3\omega) + \sigma_5(5\omega) \\ &= \frac{\sigma_0}{\lambda} \frac{4e^4 v_F^4 I^2}{\varepsilon_0^2 c^2 \hbar^4 \omega^8}[N_{51}(\omega)e^{i\omega t} + \\ &\quad N_{53}(\omega)e^{i3\omega t} + N_{55}(\omega)e^{i5\omega t}] \end{aligned} \tag{7}$$

where

$$N_{51}(\omega) = \frac{159}{256} N\left(\frac{3\hbar\omega}{2}\right)$$

$$N_{53}(\omega) = \frac{2}{15} N(\hbar\omega) - \frac{867}{15360} N\left(\frac{3\hbar\omega}{2}\right) + \frac{16}{135} N(2\hbar\omega)$$

$$\begin{aligned} N_{55}(\omega) = &\frac{413}{23040} N\left(\frac{\hbar\omega}{2}\right) - \frac{2048}{23040} N(\hbar\omega) + \\ &\frac{6102}{23040} N\left(\frac{3\hbar\omega}{2}\right) - \frac{8192}{23040} N(2\hbar\omega) + \\ &\frac{4925}{23040} N\left(\frac{5\hbar\omega}{2}\right) \end{aligned} \tag{8}$$

Considering both the linear and nonlinear (the third- and fifth-order) terms, the total interband optical conductivity can be described by the formula,

$$\begin{aligned} \sigma^{\text{inter}}(\omega) &= \sigma_1 + \sigma_3 + \sigma_5 \\ &= \frac{\sigma_0}{\lambda} \Bigg\{ N\left(\frac{\hbar\omega}{2}\right) + \frac{2e^2 v_F^2 I}{\varepsilon_0 c\hbar^2 \omega^4}[N_{31}(\omega)e^{i\omega t} + \\ &\quad N_{33}(\omega)e^{i3\omega t}] + \frac{4e^4 v_F^4 I^2}{\varepsilon_0^2 c^2 \hbar^4 \omega^8}[N_{51}(\omega)e^{i\omega t} + \\ &\quad N_{53}(\omega)e^{i3\omega t} + N_{55}(\omega)e^{i5\omega t}] \Bigg\} \end{aligned} \tag{9}$$

For intrinsic single layer graphene ($\lambda = 1$), the temperature dependence of linear σ_1 and third-order nonlinear optical conductivities $\sigma_3(\omega)$ and $\sigma_3(3\omega)$ in unit of σ_0 is plotted in Fig. 1(a) at a frequency of 1 THz under the intensity of 53 W/cm². We can find that the two nonlinear terms of the third-order conductivity decrease with temperature. At low temperature, the nonlinear term $\sigma_3(3\omega)$ less than the linear conductance σ_1, and then stays as the same as the σ_1 even at room temperature. And the $\sigma_3(\omega)$ is greater than the linear conductance in the whole temperature regime. But when the frequencies below 0.7 THz, all the two third-order nonlinear terms are larger than the linear conductance in the whole temperature regime, and decrease rapidly with temperature than at 1 THz.

Figure 1(b) shows the intensity dependence of the third-order nonlinear optical conductance. It is demonstrated that when the intensity below 100 W/cm², the nonlinear term is so tiny that can be neglected. The ratio of third-order nonlinear optical conductivity and σ_0 increases with the intensity and the growth rate gradually slowed down. In the same intensity, the ratio decreases with increasing frequency. For high intensity and low frequencies, the nonlinear terms dominate; on the contrary, the linear terms dominate. The nonlinear conductance can be around an order of magnitude larger than the linear conductance when the intensity is 4000 W/cm².

Figure 2(a) shows the temperature dependence of linear σ_1 and fifth-order nonlinear optical conductivities $\sigma_5(\omega)$, $\sigma_5(3\omega)$ and $\sigma_5(5\omega)$ in unit of σ_0 at a frequency of 1 THz under the intensity of 120 W/cm². We can find that the three nonlinear terms decrease with increased temperature, and the $\sigma_5(\omega)$ decreases rapidly than the other two terms. At the low temperatures, the nonlinear term $\sigma_5(3\omega)$ is less than the linear conductance, and then stays as the same as the linear conductance even at room temperature. The $\sigma_5(\omega)$ is larger than the linear conductance in the whole temperature regime. But the $\sigma_5(5\omega)$ is less than the linear conductance and about one order magnitude lower than the $\sigma_5(\omega)$ in the low temperature regime. However, when the frequencies below 0.7 THz, all fifth-order nonlinear terms are larger than the linear conductance in the whole
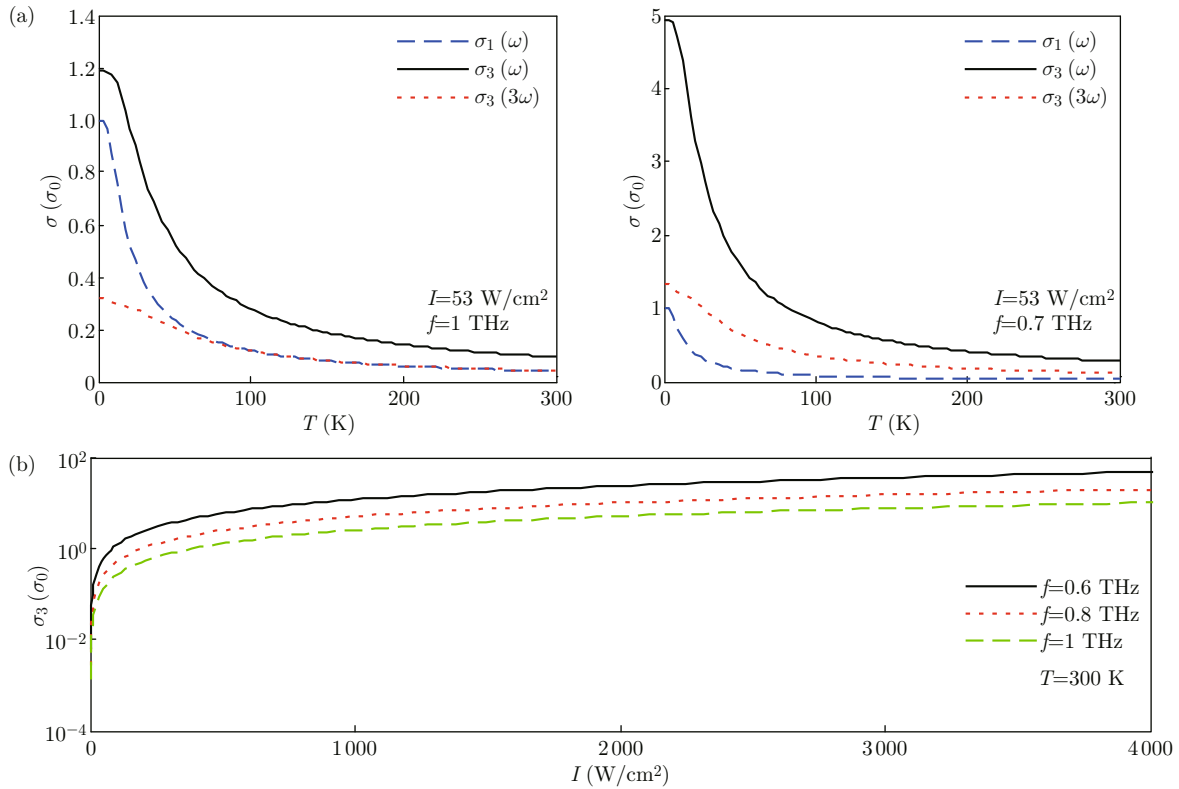
Fig. 1 For the SLG, (a) The linear and third-order nonlinear conductivity vs temperature for frequency of 1 THz and 0.7 THz; the intensity is 53 W/cm^2; (b) The third-order nonlinear conductivity vs intensity for frequency of 0.6, 0.8, 1 THz at room temperature.
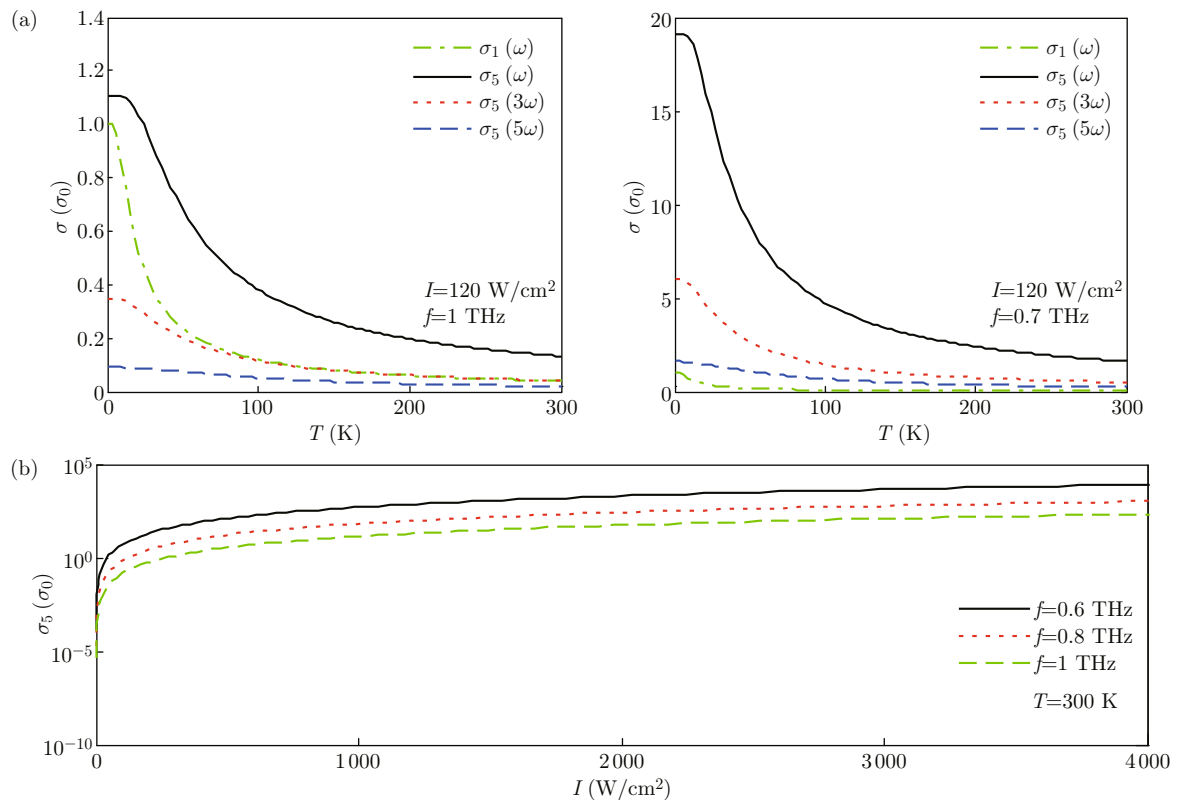

Fig. 2 For the SLG, (a) The linear and fifth-order nonlinear conductivity vs temperature for frequency of 1 THz and 0.7 THz; the intensity is 120 W/cm^2; (b) The fifth-order nonlinear conductivity vs intensity for frequency of 0.6, 0.8, 1 THz at room temperature.

temperature regime, and decrease rapidly with increased temperature than those behave at 1 THz. Figure 2(b) illustrates the intensity dependence of the fifth-order nonlinear optical conductance. It is shown that when the intensity is weak, the nonlinear term is so tiny that can be neglected. The ratio of fifth-order nonlinear optical conductivity and σ_0 increases rapidly with the intensity which is faster than the third-order terms. Contrasting with Fig. 1(b) we can discover that the fifth-order nonlinear conductance can be about three orders magnitude larger than the third-order nonlinear conductance when the intensity is 4000 W/cm^2.

In general, we find that the nth order conductivity term is proportional to the parameter $Z = (ev_F I^{0.5}/\hbar\omega^2)^{n-1}$. This means that at sufficiently high intensity and low frequency, nonlinear terms are relevant. To the present THz experiments, the value of intensity 1000 W/cm^2 is not so large for THz pulses, but it is very large for continuous wave (CW) sources. For example, if CW THz laser generated 1 THz with a spot diameter 300 micron, it would need around 1 W power to achieve this intensity. For lower frequency, it would require more power because the spot size should inevitably increase. However, with the development of THz technologies, this intensity value will eventually reach. Such a high-intensity THz wave yields non-negligible numbers of photo-generated electrons and holes. In the absence of fast recombination processes, the photo-generated electrons and holes should increase the distribution functions and, thus, decrease the nonlinear conductivities. As a result, the actual nonlinear conductance is not so large above the intensity 1000 W/cm^2.

In order to contrast the third- and fifth-order nonlinear optical response more effectively, we plot the frequency dependent on the third- and fifth-order nonlinear optical conductance of SLG in unit of σ_0 for two different temperatures in Fig. 3. The intensity is 1000 W/cm^2. All nonlinear terms decrease rapidly with increased frequency. When frequencies below 1 THz, the nonlinear term dominates and the fifth-order nonlinear terms exceed the third-order nonlinear terms, which is about two orders of magnitude larger than that behaves in 0.5 THz. The huge nonlinearities in the frequency below 1 THz are associated with the strong interaction of carriers with low-energy photons, which is characterized by the perturbation Hamiltonian inversely proportional to the photon energy. Within the frequency range 1.5~3 THz, the third-order nonlinear response is stronger than the fifth-order nonlinear response; however, they are so tiny that can be neglected completely. At the intensity around 1000 W/cm^2 and $f = 1$ THz, the value of $Z = 10$ for third-order conductance and $Z = 120$ for fifth-order conductance. However, for $f = 3$ THz, the resulting at about $Z = 10^{-1}$ for third-

order and $Z = 10^{-2}$ for fifth-order nonlinear conductance, which is so small in the higher frequency that can be totally neglected.
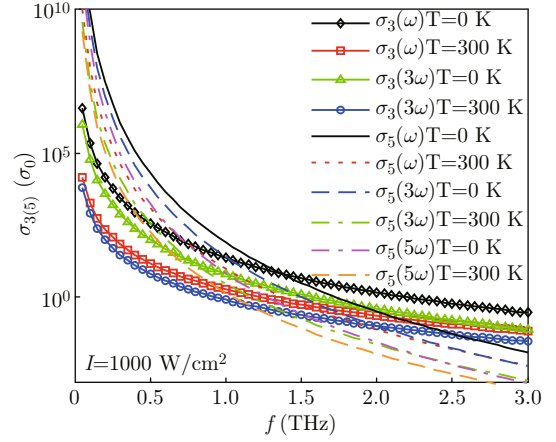

Fig. 3   For the SLG, the third-order ($\sigma_3(\omega)$ and $\sigma_3(3\omega)$) and fifth-order ($\sigma_5(\omega)$, $\sigma_5(3\omega)$ and $\sigma_5(5\omega)$) nonlinear conductivity vs frequency at zero and room temperatures; the intensity is 1000 W/cm^2.

In Fig. 4(a) we plotted the total interband optical conductivity of the frequency with $I = 1000$ W/cm^2 at $T = 0$ K and 300 K, respectively. For frequencies below 0.5 THz, because the third- and fifth-order nonlinear terms play an important role compared with the linear term; the total interband optical conductivity is strongly decrease with frequency, about $10^5\,\Omega^{-1}$ at 0.1 THz, which is more than five orders of magnitude larger than the universal conductivity. In the high-frequency part of the terahertz regime, the nonlinear terms play so small roles in the total interband optical conductivity that can be neglected completely. In the frequency range 3~10 THz, the total interband optical conductivity is almost unchanged with the frequency nearly equal to the universal conductivity $\sigma_0 \approx 6.08 \times 10^{-5}\Omega^{-1}$.

We plotted the temperature dependent the total interband optical conductance with $I = 1000$ W/cm^2 for three different frequencies in Fig. 4(b). In the same frequency, the total interband optical conductance decreases with increasing temperature. The total interband optical conductance at 0 K is around two orders of magnitude higher than the total interband optical conductivity at 300 K for a frequency of 0.1 THz, and is about an order magnitude larger than that behaves at 1 THz. The large optical nonlinearity originates from the interband electron transitions and the temperature-dependence of the nonlinear conductivities is due to the decrease in the electron and hole distribution functions at integer multiples of half the photon energy. In the high-frequency part of the terahertz regime, the total interband optical conductivity is nearly equal to σ_0 at zero temperature and decreases slightly at room temperature.
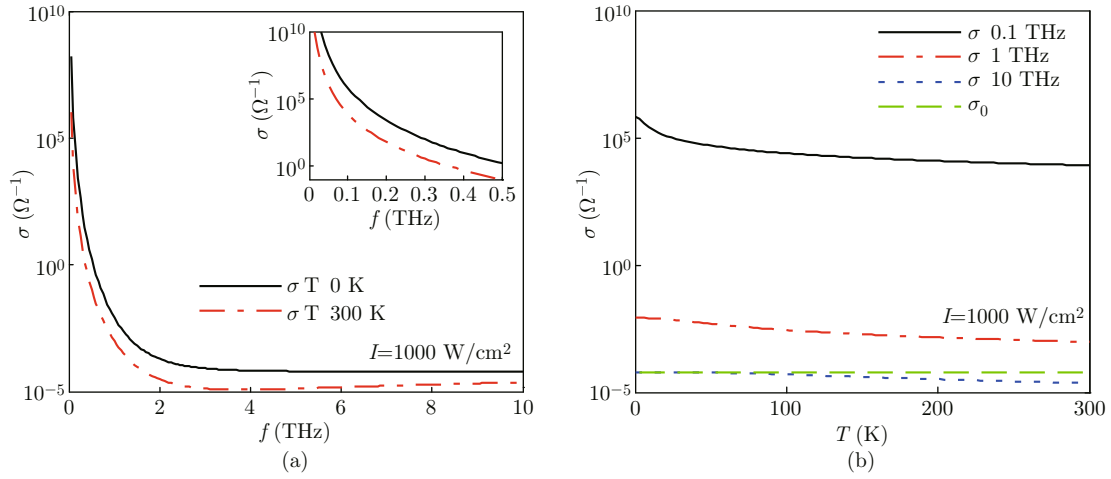
Fig. 4 (a) The total interband optical conductivity of SLG vs frequency at zero and room temperatures; the intensity is 1000 W/cm^2. The total optical conductivity in the low-frequency part of the terahertz regime (frequencies below 0.5 THz) is shown in the inset. (b) The total interband optical conductivity of SLG vs temperature for frequency of 0.1, 1 and 10 THz; the intensity is 1000 W/cm^2.

Optical transitions in graphene include interband and intraband transitions. Based on linearization of the tight binding Hamiltonian of graphene near the Dirac points of the first Brillouin zone, its total optical conductivity can be expressed as the sum of the two contributions,

$$\sigma(\omega) = \sigma^{\text{inter}}(\omega) + \sigma^{\text{intra}}(\omega) \tag{10}$$

The Drude model can estimate the intraband optical conductivity, which can be approximated as follows,

$$\sigma^{\text{intra}}(\omega) = \frac{\text{i}e^2 E_F}{\pi\hbar^2(\omega + \text{i}\tau^{-1})} \tag{11}$$

where τ is the relaxation time, the Fermi level E_F is determined by the carrier concentration $n_0 = (E_F/\hbar v_F)^2/\pi$. The linear intraband conductivity associated with intraband transitions can be described as follows,

$$\text{Re}(\sigma_1^{\text{intra}}) = \frac{e^2}{2\pi\hbar}\left(\frac{E_F}{\hbar}\right)\frac{\tau}{1 + (\omega\tau)^2} \tag{12}$$

At the intensity of 1000 W/cm^2, the generation rate of electrons and holes is about 10^{11} cm^{-2} in 1 ps. For the relaxation time of 1 ps, the real part of the linear intraband optical conductivity of SLG is around 0.13 Ω^{-1} at 0.1 THz and 0.0044 Ω^{-1} at 1 THz. Thus, the intraband conductivity is much weak than the interband conductivity that can be neglected completely.

According to the relation between the real part of optical conductivity and the transmittance [31], the transmittance of intrinsic SLG as follows,

$$T_r = \frac{4\varepsilon_0^2}{(2\varepsilon_0 + \sigma(\omega)/c)^2} \tag{13}$$

In Fig. 5, considering the third- and fifth-order nonlinear responses and the temperature factor, we plot the transmittance versus the temperature for a frequency of 0.1, 0.2, 0.3 and 1 THz respectively with $I = 1$ W/cm^2. At low frequencies (0-0.2 THz), the transmittance is strongly decreased in the entire temperature regime by the nonlinear terms effect, in which the fifth-order terms play more important roles than the third-order terms. The transmittance decreases with decreasing temperature, and almost all absorbed at zero temperature for 0.1 THz. While the frequency is greater than 0.3 THz, both the third- and fifth-order nonlinear optical responses play so small roles in the total conductivity, consequently the transmittance is nearly equal to a constant slightly larger than 97.7% at room temperature.

Figure 6 illustrates the transmittance versus the intensity for a frequency of 0.1, 0.5, 1 and 10 THz respectively at room temperature. Considering the fifth-order nonlinear responses, the transmittance decreases quickly with increased intensity. When the intensity is around 10^3 W/cm^2 and the frequency below 0.5 THz, the single layer intrinsic graphene is almost entirely absorption. However, in the high-frequency part of the terahertz regime, the transmittance is nearly equal to a constant slightly larger than 97.7%, which is the same as the situation in the visible regime, and almost not changed by the increase of the intensity.

In the KP graphene superlattice $(0 < \lambda < 1)$, the group velocity of quasiparticle no longer travels with uniform v_F in all directions. From the Eq. (6) and (7), the third- and fifth-order nonlinear optical conductances of graphene superlattice are all universally enhanced by a factor of $1/\lambda$. In Fig. 7(a), the third-order nonlinear optical conductivities $\sigma_3(\omega)$ and $\sigma_3(3\omega)$ at
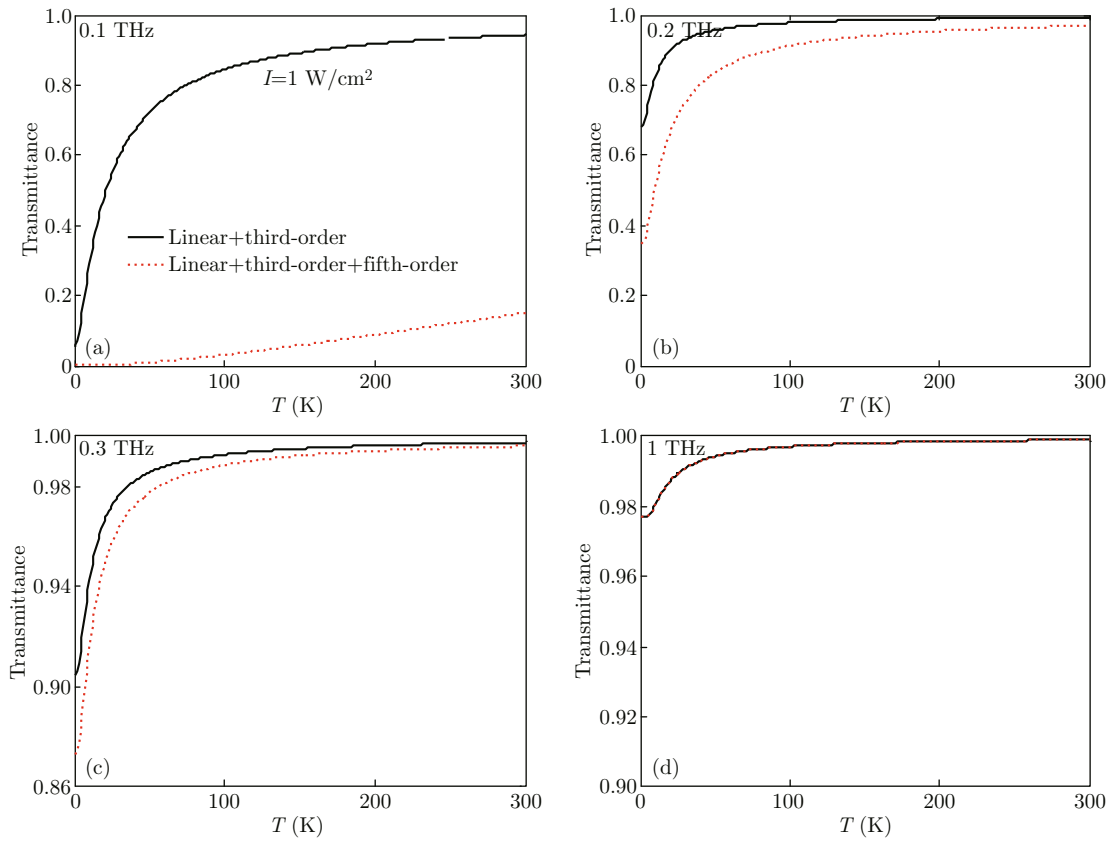
Fig. 5   The transmittance of intrinsic SLG vs temperature for frequency of 0.1, 0.2, 0.3 and 1 THz; the intensity is 1 W/cm².
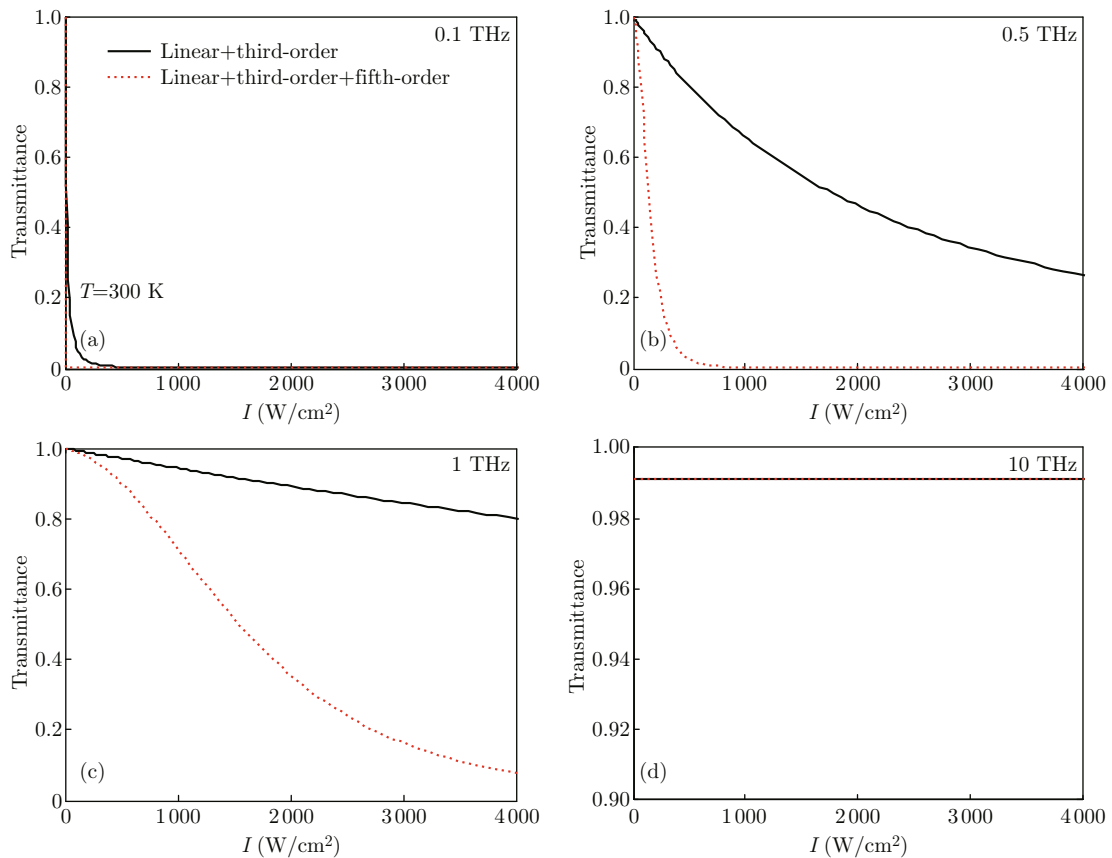

Fig. 6   The transmittance of intrinsic SLG vs electric field for frequency of 0.1, 0.5, 1 and 10 THz at room temperature.
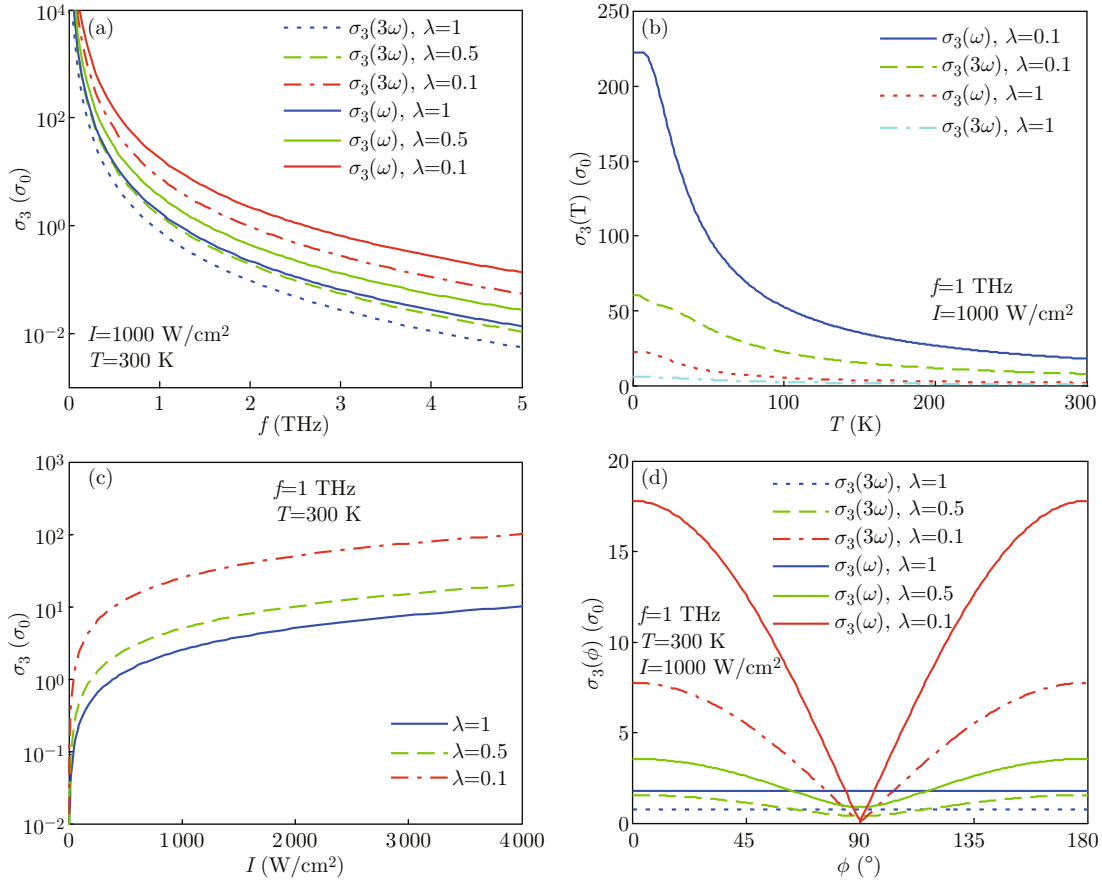
Fig. 7 For the SLG ($\lambda = 1$) and KP graphene superlattice ($0 < \lambda < 1$), (a) The third-order nonlinear conductivity vs frequency for the intensity is 1000 W/cm² at room temperature; (b) The third-order nonlinear conductivity vs temperature for frequency of 1 THz; the intensity is 1000 W/cm²; (c) The third-order nonlinear conductivity vs intensity for frequency at 1 THz; $T = 300$ K; (d) The third-order nonlinear conductivity vs angle ϕ for frequency of 1 THz at room temperature; the intensity is 1000 W/cm².

$T = 300$ K and $I = 1000$ W/cm² are plotted for different λ. In comparison to the isotropic case (SLG), the nonlinear responses of graphene superlattice are universally enhanced by the anisotropy factor when the external field aligns with the supperlattice periodicity. The temperature dependence of the third-order nonlinear optical conductivitiesis plotted in Fig. 7(b) at 1 THz and the intensity of 1000 W/cm². The two nonlinear terms decrease with increasing temperature, which is the same as the isotropic case but enhanced by a factor of $1/\lambda$. Figure 7(c) shows the intensity dependence of the third-order nonlinear optical conductance for different λ. For the KP graphene superlattice, the y-direction group velocity is reduced to $v_y = \lambda v_F$, the y-component momentum p_y in an equi-energy slice becomes larger, so the overall larger momentum of the charge carrier is the reason for the enhancement of the optical conductance. In a more general case, when E is in any angle ϕ as measured from the x-axis, the combine effect of the x-directional $\sigma(x)$ and y-directional $\sigma(y)$, the total angular dependence conductivity, measured along the ϕ-direction, as

follows, $\sigma(\phi)^2 = \sigma(x)^2 + \sigma(y)^2$, the ratio as follows, $\sigma_n(\phi, j\omega)/\sigma_n(j\omega) = (\cos^2(\phi)/\lambda^2 + \lambda^2 \sin^2(\phi))^{1/2}$. The ϕ-dependence of the total third-order nonlinear conductivity is demonstrated in Fig. 7(d). The maximum conductance occurs at $\phi = 0°$, $180°$ when the E is along the x-axis main enhanced by the factor of $1/\lambda$. When the ϕ in the vicinity of $90°$, $\sigma(y)$ becomes dominant and the influence of the factor λ over the factor of $1/\lambda$.

Massive Dirac fermions

The graphene with a finite band gap that could be created via breaking the symmetry of A and B sublattices in graphene-based system, the low-energy quasipaticles into massive Dirac fermions which are described by the finite-mass Dirac equation $H_\Delta = \sigma_x p_x + \lambda \sigma_y p_y + (\Delta/2)\sigma_z$. For the typical band gap value of $\Delta = 0.03$ eV, the linear and third-order nonlinear optical conductivities at low temperature are plotted in Fig. 8. It is shown that the gapped graphene also exists a strong nonlinear optical response by the three-photon

process $\sigma_3(3\omega)$ even when the energies lower than the band gap (sub-gap frequency regime) where the linear response is forbidden. And by changing anisotropy parameter, the nonlinear optical response is similarly tuned-up $1/\lambda$. So this kind of gapped graphene as two-color nonlinear material can be applied to nonlinear optics since the linear process is fully suppressed in the terahertz regime.
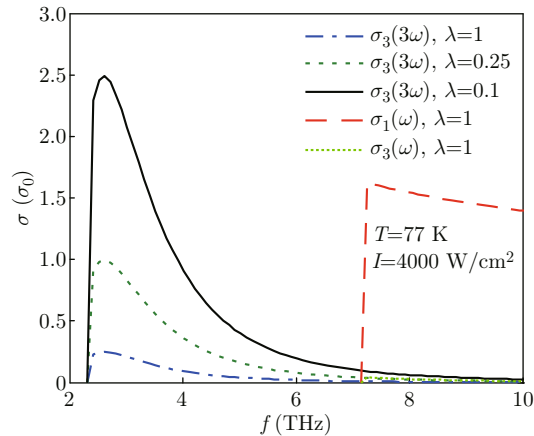

Fig. 8 The linear and third-order nonlinear optical conductivities of gapped graphene at low temperature 77 K. The intensity is 1000 W/cm².

Conclusion

Depends on a theoretical investigation of the linear and nonlinear (the third- and fifth-order) optical conductivity, we have shown that graphene-based systems exhibit strong nonlinear optical response in the terahertz regime. Our results demonstrate that at low temperature, the third- and fifth-order nonlinear optical conductivities in the intrinsic single layer graphene system are more than five and ten orders of magnitude respectively, larger than the universal conductivity at the intensity around 1000 W/cm² and the frequency below 1 THz. In addition, the strong temperature-dependent nonlinear optical responses make the transmittance of SLG decrease rapidly and induce strong absorption. In the KP graphene superlattice system, the anisotropic Dirac fermions enhanced the optical response by a factor of $1/\lambda$ and this kind of enhancement also occurs in gapped graphene system. These properties make graphene-based systems a potential candidate as a terahertz detector.

Acknowledgement

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