

# Nonlinear optical spectrum of bilayer graphene in the terahertz regime

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We demonstrate that the nonlinear optical response in bilayer graphene is among the strongest, especially in the important frequency regime of terahertz to far-infrared. Furthermore, we show that both the single frequency and frequency tripled nonlinear response become comparable to the linear response at very moderate electric field. The field strength for the onset of nonlinear effect in bilayer graphene is well within the experimental achievable range in laboratories. Our result suggests that bilayers are preferred structures for developing graphene-based nonlinear photonics and optoelectronics devices. © 2010 American Institute of Physics. [doi:10.1063/1.3527934]

Since the isolation of single layers of graphite in 2003,<sup>1</sup> a lot of exciting work on single layer graphene (SLG) has been done;<sup>2</sup> for example, the prediction and observation of electron-hole symmetry and a half-integer quantum Hall effect,<sup>3–5</sup> finite conductivity at zero charge-carrier concentration,<sup>3</sup> the strong suppression of weak localization,<sup>6,7</sup> universal conductance<sup>8–11</sup> and magnetic enhancement of optical conductance in graphene nanoribbons,<sup>12</sup> and strong nonlinear response in the terahertz frequency regime.<sup>13,14</sup>

Bilayer graphene (BLG) exhibits additional new properties not seen in single layer graphene; chief among them are the trigonal warping,<sup>15,16</sup> a phenomenon solely due to the interlayer coupling. The quantum Hall plateaus in BLG are doubled and independent of the interlayer coupling strength.<sup>17</sup> In general, electrons in bilayers behave qualitatively differently than in single layers. Phenomena such as interlayer drags<sup>18</sup> and correlations<sup>19</sup> are unique in bilayer. Various models for low energy BLG exist in literature depending on the coupling terms included, and whether electronic bands beyond the lowest energy subbands are retained.<sup>16,20</sup> Many interesting results were obtained based on a model that includes only the most dominant of the interlayer coupling terms in BLG, as well as the usual nearest neighbor intralayer term.<sup>21</sup> By including the second most dominant interlayer coupling, some unusual properties such as a peculiar Landau-level spectrum have been derived,<sup>16</sup> as well as a new low energy peak in the optical conductance.<sup>22,23</sup> By further increasing the layer numbers, one has graphene multilayers whose energy dispersion near the *K*-point can be tuned by a gate voltage.<sup>24</sup>

Graphene exhibits strong optical response.<sup>8–11,25</sup> The universal conductance  $\sigma_0 = e^2/4h$  leads to an absorption of around 2.3%. This absorption is very high for one atomic layer, or around two orders of magnitude higher per atomic layer in semiconductors. However, if the aim is to develop photonics applications based on single or a few layer graphene, the optical conductance in the terahertz-FIR regime is essentially confined to  $\sigma_0$ .

The purpose of this letter is to demonstrate that if one goes beyond the linear response regime, the optical response of intrinsic BLG ( $\mu=0$ ) can be very strong due to the third order effect. The nonlinear effects are particularly strong in

the low frequency regime, which covers the technologically important frequency band of terahertz to far-infrared. More importantly, the field intensity required for the onset of nonlinear response is rather low, indicating that BGL is an excellent material for nonlinear optics and photonics application.

Here we shall adopt an approach that treats the coupling of the Dirac electron to the time-dependent electric field quantum mechanically to calculate the nonlinear terms, both in high order electrical field and in multiple frequencies. We determine the required field strength to induce non-negligible nonlinear effects, and investigate the temperature dependence of these terms as well.

Let us consider intrinsic BLG under an applied field  $\mathbf{E}(t) = \mathbf{E}e^{i\omega t}$  whose direction is along the *x*-axis. The tight binding Hamiltonian in the low energy regime under effective mass approximation is given by<sup>16,26,27</sup>

$$H = \alpha \begin{pmatrix} 0 & (p_- + eA)^2 \\ (p_+ + eA)^2 & 0 \end{pmatrix} - \beta \begin{pmatrix} 0 & p_+ + eA \\ p_- + eA & 0 \end{pmatrix}, \quad (1)$$

where  $\alpha = 1/(2m^*)$ ,  $m^* = 0.033m_e$ ,  $\beta = v_F \approx 10^5 \text{ ms}^{-1}$ ,  $p_{\pm} = p_x \pm ip_y$ , and  $A_{\pm} = A = (E/i\omega)e^{i\omega t}$ . The velocity operator  $\hat{v}_x = \partial H / \partial p_x$  can be split into a quadratic part  $\hat{v}_A$  and a linear part  $\hat{v}_B$  as follows:

$$\hat{v}_x = 2\alpha \begin{bmatrix} 0 & (p_- + eA) \\ (p_+ + eA) & 0 \end{bmatrix} - \beta \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} = \hat{v}_A + \hat{v}_B. \quad (2)$$

If  $\alpha \rightarrow 0$ ,  $v_A$  disappears, and so only  $v_B$  will contribute to *J*.

The wave function can be written in terms of two spinor components  $a_n(\vec{p})$  and  $b_n(\vec{p})$  as follows:

$$\psi(\vec{p}, n) = \sum_{n=0}^{\infty} \begin{bmatrix} a_n(\vec{p}) \\ b_n(\vec{p}) \end{bmatrix} e^{i(n\omega - \epsilon/\hbar)t}. \quad (3)$$

Substituting Eq. (3) in the Schrödinger equation  $i\hbar(\partial\psi/\partial t) = H\psi$ , we obtain

$$i\hbar \frac{\partial\psi}{\partial t}(\vec{p}, n) = - \sum_{n=0}^{\infty} (n\omega\hbar - \epsilon) \begin{bmatrix} a_n(\vec{p}) \\ b_n(\vec{p}) \end{bmatrix} e^{i(n\omega - \epsilon/\hbar)t}. \quad (4)$$

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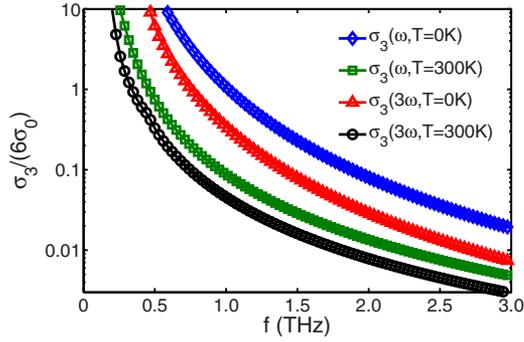


FIG. 1. (Color online) Nonlinear conductance vs frequency at zero and room temperatures; the electric field is 1000 V/cm.

Solve Eq. (4) and absorb the  $e^{i\omega t}$  and  $e^{i\omega 2t}$  terms into the spinor components to obtain  $a_{n-1}$ ,  $a_{n-2}$  and  $b_{n-1}$ ,  $b_{n-2}$ , respectively. The recursion formula is hence given by

$$\begin{aligned} (\varepsilon - n\omega\hbar)a_n &= Y_- b_n + \frac{eE}{i\omega} X_- b_{n-1} - \alpha \frac{e^2 E^2}{\omega^2} b_{n-2}, \\ (\varepsilon - n\omega\hbar)b_n &= Y_+ a_n + \frac{eE}{i\omega} X_+ a_{n-1} - \alpha \frac{e^2 E^2}{\omega^2} a_{n-2}, \end{aligned} \quad (5)$$

where  $X_{\pm} = 2\alpha p_{\pm} - \beta$  and  $Y_{\pm} = \alpha p_{\pm}^2 - \beta p_{\pm}$ . In the absence of the electric field, only  $n=0$  terms are nonzero and the solution of Eq. (5) can be written as  $a_0 = Y_- / \varepsilon \sqrt{2}$  and  $b_0 = 1 / \sqrt{2}$  where  $\varepsilon$  is the energy dispersion and is given by

$$\varepsilon = \sqrt{Y_+ Y_-} = \pm \sqrt{\alpha^2 p^4 - 2\alpha\beta p^3 \cos 3\theta + \beta^2 p^2}.$$

The above equation contains information of all multiple photon processes in intrinsic graphene. The recursion relation couples the  $n$  photon processes to the  $n-1$  photon processes. From the solutions to Eq. (5) we can calculate the  $n$ th order total current, which is given by

$$\mathbf{J}_n^{\nu} = \frac{1}{4\pi^2} \int d\mathbf{p} \mathbf{j}_n^{\nu} N(\varepsilon). \quad (6)$$

Here  $N(\varepsilon) = n_F(-\varepsilon) - n_F(\varepsilon) = \tanh(\varepsilon/2k_B T)$ , and  $\hat{v}_{\nu}$  is given by Eq. (2).  $\mathbf{j}^{\nu} = e\psi^{\dagger} \hat{v}_{\nu} \psi$  and  $\mathbf{j}_n^{\nu}$  is the part of  $\mathbf{j}^{\nu}$  that is proportional to the  $n$ th power of the electric field. The  $n=1$  terms can be obtained as

$$\begin{aligned} a_1 &= \frac{eE}{i\sqrt{2}\omega^2\hbar\varepsilon(\omega\hbar - 2\varepsilon)} [\varepsilon(\varepsilon - \omega\hbar)X_- + X_+ Y_-^2], \\ b_1 &= \frac{eE}{i\sqrt{2}\omega^2\hbar\varepsilon(\omega\hbar - 2\varepsilon)} [\varepsilon X_- Y_+ + (\varepsilon - \omega\hbar)X_+ Y_-]. \end{aligned}$$

The total current calculated with  $n=1$  terms is equivalent to the linear response result obtained from the Kubo formula. Upon converting to real units, it results in a low energy linear conductance of  $6\sigma_0$  where  $\sigma_0 = e^2/4\hbar$  is the universal conductance. The second order solution makes no contribution due to time-reversal symmetry. We now proceed to calculate the third order current. We first obtain the explicit form of  $a_2$ ,  $b_2$ ,  $a_3$ , and  $b_3$ . There are distinct third order currents: One oscillates with  $\omega$ ,  $\mathbf{j}_3(\omega)$  and the other oscillates with  $3\omega$ ,  $\mathbf{j}_3(3\omega)$ . The combination of  $a_1$ ,  $b_1$ ,  $a_2$ , and  $b_2$  contributes to  $\mathbf{j}_3(\omega)$ ; and the combination of  $a_0$ ,  $b_0$ ,  $a_3$ , and  $b_3$  contributes to  $\mathbf{j}_3(3\omega)$ .

In Fig. 1 we plot the nonlinear conductance versus fre-

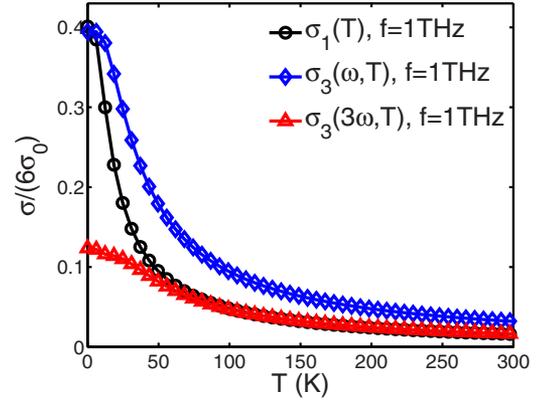


FIG. 2. (Color online) Nonlinear conductance vs temperature for frequency of 1 THz; the electric field is 600 V/cm.

quency in unit of  $6\sigma_0$  for two different temperatures. The electric field is 1000 V/cm. All nonlinear terms decrease rapidly with frequency. This is expected as linear response dominates at high frequencies in almost all systems. For BLG, the nonlinear response at single frequency is about five times stronger than frequency tripled terms.

Figure 2 shows the temperature dependent nonlinear conductance at a field of 600 V/cm and at a frequency of 1 THz. At low temperature, the nonlinear conductance exceeds the linear conductance. The  $\sigma_3(\omega)$  is greater than the linear conductance in the whole temperature regime. The all important  $\sigma_3(3\omega)$  stays as the same as the linear conductance even at room temperatures.

The quantity that characterizes nonlinearity of an electronic material is the field required for the nonlinear current to equal the linear current. We refer this field to be the critical field  $E_c$ . In the present case, there are two critical fields,  $E_c(\omega)$  and  $E_c(3\omega)$ . Figure 3 shows the frequency dependence of the critical fields at zero and room temperature. Within the frequency range 0–5 THz, the linear conductance is nearly a constant of  $6\sigma_0$ . In this frequency regime, the critical fields are well within the field strength achievable in a laboratory. At  $\omega=3$  THz,  $E_c(\omega)=400$  V/cm at zero temperature and 800 V/cm at room temperature, and  $E_c(3\omega)=700$  V/cm at zero temperature and 1100 V/cm at room temperature. This is comparable to the nonlinear effect in single layer graphene. This result suggests that interlayer coupling and doubling the carrier numbers in BLG do not reduce the nonlinear effect. If this trend is maintained up to a few layers, the potential for developing graphene-based nonlinear de-

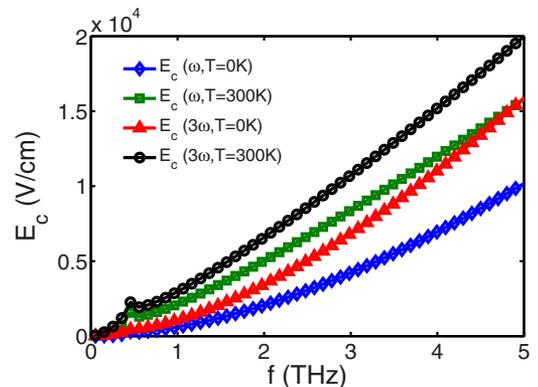


FIG. 3. (Color online) Frequency dependent critical fields at zero and room temperature.

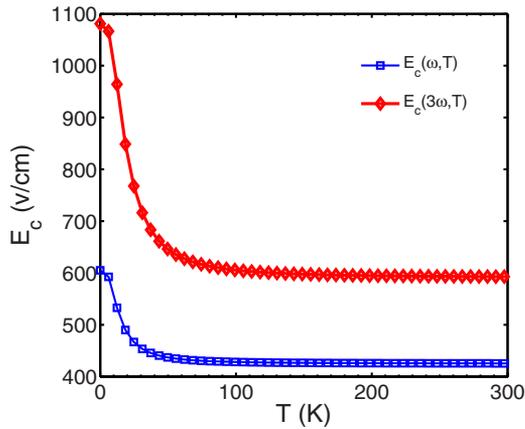


FIG. 4. (Color online) Temperature dependent critical fields for frequency of 1 THz.

vices can be significantly expanded. The small cusp at low frequency is due to a weak maximum<sup>27</sup> in the density of states, which gives rise to a large value of linear current.

In Fig. 4, we present the temperature dependence of the critical field. The rapid decrease in the critical field at low temperature is mainly due to the decrease in linear current. The sole contribution to the linear current is the direct transition from the valence band to the conduction band via one photon absorption. As temperature increases, the bottom part of the conduction band and the top part of the valence band are thermally occupied. This reduces the carriers available for the optical transition and thus reduces the linear current. Three photon processes are less affected by thermal effect at low temperatures due to the availability of the intermediate states.

To obtain further insight on the nonlinear effect in BGL, we estimate the contribution to the nonlinear current from regions around different Dirac points. The reason that BLG exhibits a strong nonlinear effect at low energy is due to a unique property of BLG, i.e., the trigonal warping. At low frequencies, the contribution to the total nonlinear current from the central Dirac points and the three satellite Dirac points can be separated. We found that for both  $\sigma_3(\omega)$  and  $\sigma_3(3\omega)$ , the contribution from the central Dirac point is less than 10% while each satellite Dirac point contributes around 30% of the total nonlinear current. This is a clear indication on the connection between the trigonal warping and nonlinear optical processes.

Our results demonstrate that BLG is a rather strong nonlinear material. This nonlinear effect is robust from low to room temperatures. The frequency tripling nonlinear term is comparable to the linear term in the terahertz frequency regime. This term's relative strength suggests that BLG has a potential in a terahertz emitter/detector at frequencies, which are traditionally difficult to obtain by using an existing emitter at one-third the frequency.

Finally we comment on the role of phonon excitation in BLG. In the temperature range of up to room temperature, the dominant electron-phonon coupling is via longitudinal acoustic (LA) phonons since either the couplings to other graphene lattice phonon modes are too weak or the energy scales of these optical phonon modes are far too high.<sup>28</sup> The velocity of the LA phonon is around  $2 \times 10^4$  m/s.<sup>28</sup> Under an electric field around 1000 V/cm with a frequency of 1

THz, the energy of the photoexcited electron is around 1 THz. These electrons are located very close to the Dirac point, and the electron velocity is around  $0.6 \times 10^6$  m/s. In the absence of other disorders and due to the energy conservation, the probability of single phonon emission is negligible. The multiple phonon excitation is possible but the probability is also very low due to the high order electron-phonon coupling. Therefore we do not expect that phonon excitation will play a significant role in altering the nonlinear electrical current in this energy regime.

In conclusion, we have shown that BLG exhibits a strong nonlinear effect in the terahertz to far-infrared regime under an electric field of around  $10^3$  V/cm. In particular, a moderate field can induce the frequency tripling term at room temperature. This suggests a potential for developing graphene-based optics and photonics applications.

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