Higher Harmonic Generation of Coherent Sub-THz Radiation at Lifshitz Transition in Gapped Bilayer Graphene

A. G. Ghazaryan
Centre of Strong Fields Physics, Yerevan State University, Yerevan, 0025 Armenia

Abstract—The microscopic quantum theory of nonlinear interaction of strong coherent electromagnetic radiation with gapped bilayer graphene is used for consideration of the high harmonic generation at low-energy photon excitation Lifshitz transition. The Liouville–von Neumann equation for the density matrix is solved numerically at the nonadiabatic multiphoton excitation regime. By numerical solutions, we examine the rates of the second and third harmonics generation at the particle-hole annihilation at Lifshitz transitions in the linearly polarized coherent electromagnetic wave. The obtained results show that the gapped bilayer graphene can serve as an effective medium for the generation of even and odd high harmonics in the sub-THz domain of frequencies.

DOI: 10.1134/S1063776121050034

1. INTRODUCTION

Many quantum electrodynamic nonlinear phenomena induced by strong laser radiation in condensed matter, specifically in graphene-like nanostructures, have significant contribution in low-energy physics and nano-opto-electronics and have been systematically investigated mainly in case of monolayer graphene [1] that has been conditioned by unique physical properties of such two-dimensional (2D) nanosystem of atomic thickness [1–3]. On the other hand, for induced electrodynamic phenomena in 2D atomic systems—nanostructures a bilayer graphene (AB-stacked) is of great interest too, since its electronic states are considerably richer than those for monolayer graphene. The multiphoton resonant excitation with high-harmonic generation (HHG) via nonlinear channels in bilayer graphene were also considered [4–6]. The pioneer studies of laser-induced HHG process were made generally in gaseous media. However, the study of high-order harmonics from bulk crystals [7–12] was reported much more in recent decade. It is known that in the linear approximation, the interaction of a large-amplitude electromagnetic wave with graphene can lead to a significant effective rearrangement of the energy spectrum of graphene [13–15]. As well, it is of interest the investigation of HHG and related processes in low-dimensional nanostructures, such as graphene and its derivatives [13–34], hexagonal boron nitride [35], monolayer transition metal dichalcogenides [36–38], topological insulator [39, 40] in monolayers of black phosphorus [41], buckled 2D hexagonal nanostructures [42], solids [43, 44], as well as in other 2D systems [45–48]. The nonlinear coherent response in AB-stacked bilayer graphene under the influence of intense electromagnetic radiation leads to the modification of quasi-energy spectrum, the induction of valley polarized currents [49, 50] as well as the second and third-order nonlinear-optical effects [51–54]. The graphene of bilayer system represents a unique system in which the topology of the band structure can be externally influenced and chosen. The bilayer graphene is a highly tunable material: not only one can tune the Fermi energy using standard gates, as in single-layer graphene. The band structure can also be modified by external perturbations such as transverse electric fields or strain [55–61]. Particularly, it is of interest to consider HHG process in the strong wave-bilayer graphene coupling regime with a bandgap induced by an external constant electric field [58, 62–64]. Moreover, with the current technology [64, 65] one can make such large gaps in AB-stacked bilayer graphene, which is sufficient to produce field-effect transistors not only at cryogenic temperatures but at room-temperature [66, 67].

It is known the graphene-based low energy photon-counting photodetector of different applications, in areas as diverse as medical and space sciences or security-applications. The tunable bandgap in bilayer graphene may enable sensitive photon-counting photodetectors to operate with a trade-off between resolution and operational temperatures, with resulting operational benefits. Note that the large bandgap can also make possible effective room temperature HHG [28] in bilayer graphene, which is suppressed in intrin-
sic bilayer graphene [16]. Unfortunately, the effective pump wave-induced Lifshitz transitions (with photon energy much smaller than Lifshitz energy) in \(AB\)-stacked gapped bilayer graphene are less investigated.

At the HHG process in gapped bilayer graphene in the coherent electromagnetic radiation field at Lifshitz transition with the energies much smaller than the so-called Lifshitz energy \(\mathcal{E}_L \sim 1\) meV have some peculiarities [68–75]. Two touching parabolas of the Fermi surface are broken into four separate “pockets”. As opposed to bulk graphite, the external perturbations like the strain [76, 77] or electric field [78] can modify the topology of the electronic dispersion and change the energy of the Lifshitz transition which connects regions of different Fermi contour topologies [79]. Due to the 2D nature of bilayer graphene, its chemical potential and its topology can be tuned with electrostatic gates [1], simplifying experimental studies of the Lifshitz transition. By the way, in unperturbed bilayer graphene, it is achieved at low energies \(\mathcal{E}_L = 1\) meV. To induce the asymmetry, a chemical doping can be used [55] or external gates [56] can be patterned. This induced asymmetry opens a bandgap in the energetic spectrum of the graphene [59, 71–75]. As is shown in [79], particularly for an induced asymmetry \(U = 100\) meV, the Lifshitz transition occurs at the higher energy \(\mathcal{E}_L \sim 1.6\) meV. One can conclude from the estimate given in [79] that the experimental observation of the Lifshitz transition should be facilitated by the layer-asymmetry induced bandgap and that, the wider the gap, the more enhanced the visibility of this effect.

At an intraband transitions the interaction of a particle with the wave at the THz or sub-THz photon low-energies \(\hbar \omega \ll \mathcal{E}_L\), characterizes by the effective interaction parameter \(\chi\) [16]:

\[
\chi = \frac{e E_0 v_3}{\hbar \omega},
\]

where \(E_0\) is the wave strength, \(\omega\) is the wave frequency, \(e\) is the electron charge; \(v_3 = \sqrt{3} a c / 2 h \approx v_F / 8\) is the effective velocity related to oblique interlayer hopping \(\gamma_3 = 0.32\) eV \((a = 0.246\) nm is the distance between the nearest \(A\) sites), \(v_F\) is the Fermi velocity in monolayer graphene. Due to the gap \(U\), the interband transitions are characterized by so-called Keldysh [80, 81] parameter expressed in the form:

\[
\gamma = \frac{\omega \sqrt{m U}}{e E_0} = \frac{v_3 \sqrt{m U}}{\hbar \omega}. \chi \omega.
\]

Here \(U\) is the bandgap energy; \(m = \gamma_1 / (2 v_F^2)\) is the effective mass, \(\gamma_1 = 0.39\) eV.

For the gapped materials, the Keldysh parameter gives the character of the ionization process which with the electron-hole pair creation is the first step of HHG. In the limit of \(\gamma \gg 1\), the multiphoton ionization dominates in the ionization process. In the so-called nonadiabatic regime \(\gamma \sim 1\), both multiphoton ionization and tunneling ionization can take place. In the limit of \(\gamma \ll 1\), the tunneling ionization dominates. For the considered case, the ionization process reduces to the transfer of the electron from the valence band into the conduction band that is the creation of an electron-hole pair. Since the interband transitions can be neglected when \(\gamma \gg 1\), then the wavefield cannot provide enough energy for the creation of an electron-hole pair, and the generation of harmonics is suppressed. So that, in the nonadiabatic regime due to the large ionization probabilities the intensity of harmonics can be significantly-enhanced compared with tunneling one [28, 33]. If \(\gamma \sim 1\) or \(\gamma \ll 1\), interband transitions take place. From this point of view, condensed matter materials with bilayer graphene are preferable due to the tunable bandgap with nontrivial topology.

In the following, we will consider the nonadiabatic regime for the generation of HHG at \(\gamma \sim 1\) and \(\gamma \ll 1\), when the multiphoton effects become essential. Our consideration is mainly focused on the low photon energies. Note that the average intensity of the wave expressed by \(\chi\) can be given as

\[
I_\chi = \chi^2 \times 1.96 \times 10^{13} \text{ W cm}^{-2} \left( \hbar \omega / eV \right)^4,
\]

so the required intensity \(I_\chi\) for the nonlinear regime strongly depends on the photon energy. Particularly, for the photons with the energies 0.4–0.9 meV, the multiphoton interaction regime can be achieved at the intensities \(I_\chi = 1–10^3\) W cm\(^{-2}\). The modern photonic-based THz and sub-THz (with energies 0.4–1.24 meV) courses include quantum cascade lasers and can achieve admirable output powers, mainly at cryogenic temperatures, whereas used in conjunction with nonlinear crystals can make microwatts of tunable continuous wave THz at room temperature [82]. Unfortunately, all of these sources offer impressive performance in their own ways, but none so far are easily integrated into larger digital electronic systems, which is arguably their biggest downfall for communication systems [82, 83].

In the present paper, with the help of the microscopic nonlinear quantum theory the interaction in \(AB\)-stacked gapped bilayer graphene with the strong laser radiation is investigated numerically. We find out the optimal values of the main parameters, in particular, for bandgap, pump wave intensity, graphene temperature for practically significant case in high-order harmonics coherent emission in the particle low-energy region of the induced Lifshitz transitions (the fragmentation of the singly-connected Fermi line into four separate pieces) [59, 68–70]. The Liouville-von Neumann equation is treated numerically for the generation of the higher (here -second, third) harmonics in the multiphoton excitation regime near the Dirac points of the Brillouin zone. We consider the harmonic generation process in the nonadiabatic regime
of interaction when the Keldysh parameter is of the order of unity. The picture of the multiphoton excitation of the Fermi–Dirac sea and the trigonal warping effect is also revealed. We examine the HHG rates at the particle-hole annihilation in the strong effective field of two linearly polarized plane electromagnetic wave for practically optimal parameters of the considering system. The obtained results show that for specially chosen values of the corresponding characteristic parameters of this process we can use gapped bilayer graphene as a convenient nonlinear medium to generate the higher harmonics of the pump wave with an effective yield in the sub-THz and THz domains of the spectrum, at the graphene temperatures of higher than the cryogenic temperatures.

The paper is organized as follows. In Section 2 the set of equations for a single-particle density matrix is formulated and numerically solved in the multiphoton interaction regime. In Section 3, we consider the problem of harmonics generation at the low-energy excitation of gapped bilayer graphene. Finally, conclusions are given in Section 4, and the complex formulas moved to Appendix.

2. BASIC THEORY

In the following we use the microscopic nonlinear quantum theory of interaction of coherent electromagnetic radiation with gapped bilayer graphene which was developed in [28, 33]. We propose linearly polarized plane electromagnetic wave in the plane of the graphene sheets (XY), with carrier frequency \( \omega \) and slowly varying amplitude of the electric field \( f(t)E_0 \).

\[
E(t) = f(t)E_0 e^{\cos\omega t},
\]

where \( e \) is the unit polarization vector. The pump wave slowly varying envelope is described by the function:

\[
f(t) = \begin{cases} \sin^2(\pi t/\tau) , & 0 \leq t \leq \tau, \\ 0 , & t < 0 , \quad t > \tau, \end{cases}
\]

where \( \tau \) characterizes the pulse duration and is taken to be \( \tau = 10\tau_0, \tau_0 = 2\pi/\omega \).

The effective single-particle Hamiltonian [58–60] for the low-energy excitations (\( \bar{\varepsilon}_{\sigma} < \gamma \approx 0.39 \text{ eV} \)) in AB-stacked gapped bilayer graphene in the vicinity of the Dirac points \( K \) have the form:

\[
\hat{H}_s = \begin{pmatrix} U/2 & q^*_\zeta(p) \\ q_{\zeta}(p) & -U/2 \end{pmatrix},
\]

where \( \zeta = \{ \zeta_x, \zeta_y \} \) is the electron momentum operator, \( \zeta = \pm 1 \) is the valley quantum number, \( U \) is the opened gap energy, and

\[
q_{\zeta}(p) = -\frac{1}{2m_s}(\zeta p_x + ip_y)^2 + v_s(\zeta p_x - ip_y).
\]

The first term in Eq. (4) are connecting with pair of parabolic bands \( \bar{\varepsilon} = \pm p^2/2m \).

The spin and the valley quantum numbers are conserved. There are no intervalley transitions, and the valley index \( \zeta \) can be considered as a parameter. The eigenstates of the effective Hamiltonian (3) are the spinors,

\[
\Psi_{\sigma}(r) = \frac{1}{\sqrt{S}}|\sigma,p\rangle \exp\left(\frac{i}{\hbar} p \cdot r \right),
\]

with

\[
|\sigma,p\rangle = \frac{1}{\sqrt{S}} \left[ \begin{array} \varepsilon_{\sigma} + U/2 \\ 1 \end{array} \right] \left[ \begin{array} \gamma(p) \\ 1 \end{array} \right],
\]

\[
\gamma(p) = -\frac{p^2}{2m} e^{2\zeta \phi} + \zeta v_s p e^{-2\zeta \phi},
\]

\( \phi = \arctan(p_x/p_y) \), \( \sigma \) is the band index (\( \sigma = 1 \) for the conduction band and \( \sigma = -1 \) for the valence band), and \( S \) is the quantization area;

\[
\bar{\varepsilon}_{\sigma}(p) = \sigma \sqrt{\frac{U^2}{4} + (v_s p)^2 - \zeta \gamma^2 v_s^2 p^2 m \cos 3\phi} + \left(\frac{p^2}{2m}\right)^2
\]

—corresponding eigenenergies.

The Fermi–Dirac field operator in the form of an expansion in the free states, given in (5), can be described using the second quantized technique, that is:

\[
\hat{\Psi}(r,t) = \sum_{p,\sigma} \hat{a}_{p,\sigma}(t) \Psi_{\sigma}(r),
\]

where \( \hat{a}_{p,\sigma}(t) \) is the annihilation (creation) operator for an electron with momentum \( p \) which satisfy the usual fermionic anticommutation rules. The single-particle Hamiltonian in the presence of a uniform time-dependent electric field \( E(t) \) can be expressed in the form:

\[
\hat{H}_int = \hat{H}_s + \begin{pmatrix} \epsilon r \cdot E(t) & 0 \\ 0 & \epsilon r \cdot E(t) \end{pmatrix},
\]

where for the interaction Hamiltonian we have used a length gauge, describing the interaction by the potential energy [84, 85]. Using the expansion (9), the second quantized total Hamiltonian can be written as:

\[
\hat{H} = \sum_{\sigma,p} \bar{\varepsilon}_{\sigma}(p) \hat{a}_{p,\sigma}^\dagger \hat{a}_{p,\sigma} + \hat{H}_int,
\]

where the light-matter interaction part is given in terms of the gauge-independent field \( E(t) \) as follow:

\[
\hat{H}_int = i e \sum_{p,p',\sigma} \delta_{p,p} \hat{a}_{p,\sigma}^\dagger E(t) \hat{a}_{p',\sigma} + E(t) \hat{a}_{p',\sigma}^\dagger \hat{a}_{p,\sigma}.
\]

\[
+ \sum_{p,\sigma} E(t)(\mathbf{D}_s(\sigma,p) \hat{a}_{p,\sigma}^\dagger \hat{a}_{p,-\sigma} + \mathbf{D}_m(\sigma,p) \hat{a}_{p,\sigma}^\dagger \hat{a}_{p,-\sigma}).
\]
Here

\[ D_m(\sigma,p) = \hbar e(\sigma,p)|\partial_\sigma|\sigma,p) \]  \hspace{1cm} (13)

is the mean dipole moment or Berry connection,

\[ D_t(\sigma,p) = \hbar e(\sigma,p)|\partial_\sigma|\sigma,p) \]  \hspace{1cm} (14)

is the transition dipole moment (the both are given in Appendix, see also in [28]).

Multiphoton interaction of a bilayer graphene with a strong radiation field will be described by the Liouville–von Neumann equation with inhomogeneous phenomenological damping rate \( \Gamma \), since homogeneous relaxation processes are slow compared with inhomogeneous. We will solve the set of Eqs. (27), and followed from the last closed set of differential Eqs. (28), (29) given in the Appendix, for the functions \( N_c(p, t), N_s(p, t) \), \( P(p, t) \) (definitions of these functions see in Appendix), taking into account the initial conditions (\( P(p, 0) = 0 \)):

\[ N_c(p, 0) = \frac{1}{1 + \exp[(\mathcal{E} - \mu)/T]}, \]  \hspace{1cm} (15)

\[ N_s(p, 0) = 1 - N_c(p, 0). \]  \hspace{1cm} (16)

Here \( T \) and \( \mu \) are the temperature and chemical potential, respectively, in energy units.

The set of Eqs. (28)–(30) can not be solved analytically for the general case. For the numerical solution we made a change of variables and transform the equations with partial derivatives into ordinary ones. The new variables are \( t \) and \( \tilde{p} = p - p_E(t) \), where

\[ p_E(t) = -\frac{1}{\epsilon} \int_0^t \mathbf{E}(t')dt' \]  \hspace{1cm} (17)

is the classical momentum given by the wave field.

Photoexcitations of the Fermi-Dirac sea–induced Lifshitz transitions, are presented in Figs. 1, 2. The effective wave is assumed to be linearly polarized along the \( y \) axis. After the corresponding transformations, the integration of Eqs. (28)–(30) is performed on a homogeneous grid of \( 10^4 \) \( (\tilde{p}_x, \tilde{p}_y) \)-points. For the maximal momentum we take \( \tilde{p}_{\text{max}}/\sqrt{\hbar m \omega} = 5 \). The time integration is performed with the standard fourth-order Runge–Kutta algorithm. For the relaxation rate we take \( \Gamma = t_0(\mu, T)\bar{\gamma}_p^{-1} \), where \( t_0(\mu, T) \) is the relaxation time.

We will estimate the relaxation time \( t_0(\mu, T) \). We study the coherent interaction of bilayer graphene with a pump wave in the ultrafast excitation regime, which is correct only for the times \( t < \tau_{\text{min}} \), where \( \tau_{\text{min}} \) is the minimum of all relaxation times. For the excitations of energies \( \mu < \gamma_p = 0.39 \text{ eV} \), the dominant mechanism for relaxation will be electron-phonon coupling via longitudinal acoustic phonons [86, 87]. For the low-temperature limit

\[ p_E(t) = -\frac{1}{\epsilon} \int_0^t \mathbf{E}(t')dt' \]  \hspace{1cm} (17)
\[ T \ll 2 \frac{c_{ph}}{v_F} \sqrt{\mu \tau}, \]

where \( c_{ph} = 2 \times 10^6 \text{ cm/s} \) is the velocity of the longitudinal acoustic phonon, the relaxation time for \( \mu \) be estimated as \([87]\):

\[ t_0(\mu, T) = \left( \frac{\pi D^2 T^2}{8 \alpha m^4 c_{ph} v_F} \frac{\gamma}{\mu} \right)^{-1}. \]  

(18)

Here \( D \approx 20 \text{ eV} \) is the electron-phonon coupling constant and \( \rho_m = 15 \times 10^{-8} \text{ g/cm}^2 \) is the mass density of the bilayer graphene. For \( \mu = 0.8 \text{ meV} \) at the temperatures \( T = 0.4\hbar \omega \), from Eq. (18) we obtain \( \tau = 60 \text{ ps} \). Thus, in this energy range one can coherently manipulate with multiphoton transitions in bilayer graphene on the time scales \( t \approx 60 \text{ ps} \), not taking into account the particle-particle collisions.

In Fig. 1 density plot of the particle distribution function \( N(p, t) \) is shown function of scaled dimensionless momentum components after the interaction at the different energy gaps. The pump wave pulse duration is \( T = 10T_0 = 50 \text{ ps} \). It is clearly seen the trigonal warping effect describing the deviation of the excited iso-energy contours from circles, which is smeared with the increase of the gap magnitude \( U \). In all considering cases, the two touching parabolas are transformed into the four separate “pockets.” Note that trigonal warping is crucial for even-order nonlinearity. As is seen with the increasing of \( U \) we approach to perturbative regime \( \gamma > 1 \) and only weak excitation of Fermi–Dirac sea.

In Fig. 2, we show the photoexcitation depending on the pump wave intensity at fixed sub-THz frequency. For the large values of \( \gamma \) when \( \gamma = 1 \) we clearly see multiphoton excitations. With the increasing wave intensity, the states with the absorption of more photons appear in the Fermi–Dirac sea. At the parameters \( \gamma \approx 1 \) when \( \gamma = 1 \), the multiphoton excitation of the Fermi–Dirac sea takes place along the trigonally warped isofrequency contours of the quasienergy spectrum modified by the wave field. Thus, the multiphoton probabilities of particle-hole pair production will have maximal values for the iso-energy contours defined by the resonant condition

\[ \mp \int_{0}^{r} 2\epsilon(t) dt = n\hbar \omega, \quad n = 1, 2, 3,... \]

These contours are also seen in Fig. 1. The investigations of the temperature dependence of the excitation of the Fermi–Dirac sea are cleared that for considering cases it exhibits a tenuous dependence on the optimal temperatures: the excited isofrequencies are slightly smeared out with temperature increase. This effect is small since \( U \gg T \) and one can expect that harmonic spectra will be robust against temperature change in contrast to the gap \( U = 0 \) case where harmonics radiation is suppressed with the increase of temperature in \([28, 33]\). So, the temperature dependence was missed.

In the following section, we will investigate the nonlinear response of the bilayer graphene in the process of the second and third-order harmonics generation under the influence of the laser field in the nonadiabatic regime \( \gamma \approx 1 \) with the frequencies in sub-THz domain: \( \omega = 0.4–0.9 \text{ meV/} \hbar \).

3. HARMONICS GENERATION AT INDUCED LOW-ENERGY TRANSITIONS IN GAPPED BILAYER GRAPHENE

In this section, we examine the nonlinear response of a bilayer graphene to harmonic generation process considering the nonadiabatic regime of induced Lifshitz transitions, when the Keldysh parameter is of the order of unity. For the coherent part of the radiation spectrum one needs the mean value of the current density operator,

\[ j_{\xi}(t) = -2e(\bar{\Psi}(r,t)) \bar{\xi}(\bar{\Psi}(r,t)). \]  

(19)

The velocity operator \( \bar{v}_{\xi} = \partial \bar{H}/\partial \bar{p} \) is given in Appendix (see Eq. (35), (36), and [30]). Using the Eqs. (19)–(36), the expectation value of the current for the valley \( \xi \) can be written in the form:

\[ j_{\xi}(t) \approx -\frac{2e}{(2\pi \hbar)^3} \int d\bar{p} \left[ \tilde{V}(\bar{p}) (N_\xi(p,t) - N_0(p,t)) + \frac{2i}{\hbar} \tilde{\eta}_\xi(p) \right]. \]  

(20)

where \( \tilde{V}(\bar{p}) \)—the intraband velocity (37). In Eq. (20) the first term—the intraband current which conditioned by intraband high harmonics and is generated as a result of the independent motion of carriers in their respective bands. The second term in Eq. (20) describes high harmonics which are generated as a result of the recombination of accelerated electron-hole pairs. Since we study the nonadiabatic regime, the contribution of both mechanisms is essential.

There is no degeneracy upon valley quantum number \( \xi \), so the total current can be obtained by a summation over \( \xi \):

\[ j_x = j_{\xi, x} + j_{-\xi, x}, \]  

(21)

\[ j_y = j_{\xi, y} + j_{-\xi, y}. \]  

(22)

The current density components \( j_{x, y} \) are defined as

\[ j_{x,y} = G_{s,y} \left( \omega, \chi, \frac{\epsilon_F}{\hbar \omega}, \frac{T}{\hbar \omega}, \frac{U}{\hbar \omega} \right). \]  

(23)

Here

\[ j_{\xi} = \frac{e \omega}{\pi} \sqrt{\frac{\omega}{\hbar}}, \]  

(24)
and $G_1$ and $G_2$—the dimensionless periodic (for monochromatic wave) functions which parametrically depend on the interaction parameters $\chi$, $\gamma$, scaled Lifshitz energy, and temperature. Thus, using solutions of Eqs. (28)–(30), and making an integration in Eq. (20), one can calculate the harmonic radiation spectra with the help of a Fourier transform of the function $G_{1,2}(\omega)$. The emission rate of the $n$th harmonic is proportional to $\sum_{\alpha=1}^{\infty} \left| j_{\alpha} \right|^2$, where $j_{\alpha} = j_{\alpha 1} + j_{\alpha 2}$, with $j_{\alpha 1}$ and $j_{\alpha 2}$ being the $\alpha$th Fourier components of the field–induced total current. To find $j_{\alpha}$, the fast Fourier transform algorithm has been used. We use the normalized current density (23) for the plots.

Note that the transition currents with comparing the intrinsic graphene $j_0$, [14, 15], for a bilayer graphene is larger by a factor $\sqrt{\gamma} / 2\hbar$. Besides, the cutoff harmonic is larger than in case of a monolayer graphene [14], which is a result of strong nonlinearity caused by trigonal warping. Hence, for considered setups $\hbar \omega \ll \gamma$, the harmonics’ radiation intensity is at least one order of magnitude larger than in the monolayer graphene.

For clarification of harmonics generation due to the multiphoton resonant excitation and particle–hole annihilation, from the coherent superposition states at $\gamma = 1$ initially, we examine the emission rate of the second and third harmonics. The emission rate versus the pump wave strength defined by the parameter $\chi$ at the same wave frequency is demonstrated in Fig. 3 for various gap energies.

In Fig. 3, plots for $U = 4$ meV and $U = 5$ mV coincided. As is seen from this figure, for the field intensities $\chi \approx 1$ at the considering values of the gap energy $U$ we have a strong deviation from power law for the emission rate of the second or third harmonic (in accordance with the perturbation theory $\chi^2$ and $\chi^3$, respectively). In Fig. 4, the second and third harmonics emission rate, expectantly, is assumed as a function of the energy gap at various intensities defined by the parameter $\chi$ at the same wave frequency. As shown in Fig. 4, all plots have the maximum values at $U \approx 2$ meV. As a result, we find the optimal parameters when the harmonic emission rate is significant at the larger intensity for the considered wave frequency.

Thus, in accordance with the results of Figs. 3 and 4, the intense radiation of the second and third harmonics at the pump-wave-induced particle or hole acceleration and annihilation in gapped graphene can be obtained with the pump wave frequency in the sub-THz domain. Then, as in case of the similar calculations for the intense pump wave or large gap energy $U$ ($U \gg T$) has shown that the emission rate exhibits a tenuous dependence on the temperature.

Figure 5 demonstrates the emission rate dependence on the pump frequency for gapped bilayer graphene. We plot the second and third harmonic emission rates for a gapped bilayer graphene versus the pump frequency for the gap $U = 2$ meV and various intensity parameters. As was expected and shown from this figure, the emission rate has different maxima at various photon energies and intensities. The maximal values are reached at frequency $\omega = 0.8$ meV/$h$. Concerning the generation of high harmonics up to the far-infrared range, note that this has been demonstrated in [33] where quantum cascade lasers are readily available and can provide higher powers.

JOURNAL OF EXPERIMENTAL AND THEORETICAL PHYSICS  Vol. 132  No. 5 2021
4. CONCLUSIONS

We have presented the microscopic nonlinear theory of interaction of the gapped bilayer graphene with strong coherent radiation field at low-energy photon Lifshitz transitions. The closed set of differential equations for the single-particle density matrix is solved numerically for a bilayer graphene in the Dirac cone approximation in the sub-THz frequency linearly polarized electromagnetic wave. We have considered nonadiabatic wave Lifshitz transitions of Fermi–Dirac sea towards the HHG. It has been shown that the role of the gap in the nonlinear optical response of bilayer graphene is quite considerable. In particular, even-order nonlinear processes are present, the cutoff of harmonics increases, and harmonic emission processes become robust against the temperature increase. The obtained results show that the gapped bilayer graphene can serve as an effective medium for the generation of even and odd high harmonics in the THz and sub-THz domain of frequencies, which is sufficient for a new high-speed wireless communication systems development [82, 83]. The obtained results certify that the process of high-harmonic generation for sub-THz photons (wavelengths from 0.3 to 1 mm) can already be observed for intensities $I_p = 1–10^3 \text{ W cm}^{-2}$ at the temperature of the sample $T < \hbar \omega$.

APPENDIX

The Liouville–von Neumann equation for a single-particle density matrix can be presented by the form

$$\rho_{\alpha \beta}(p, t) = \langle \hat{a}_p^{\dagger}(t) \hat{a}_{p \alpha}(t) \rangle,$$

where $\hat{a}_{p \alpha}(t)$ obeys the Heisenberg equation

$$i\hbar \frac{\partial \hat{a}_{p \alpha}(t)}{\partial t} = [\hat{a}_{p \alpha}(t), \hat{H}].$$

Due to the homogeneity of the problem we only need the $p$-diagonal elements of the density matrix. So, taking into account Eqs. (11)–(26), the evolutionary equation will be

$$i\hbar \frac{\partial \rho_{\alpha \beta}(p, t)}{\partial t} = -i\hbar \epsilon \mathbf{E}(t) \frac{\partial \rho_{\alpha \beta}(p, t)}{\partial p} \frac{\partial \rho_{\alpha \beta}(p, t)}{\partial p} = \begin{bmatrix} \mathcal{E}_\alpha(p) - \mathcal{E}_\beta(p) - i\hbar \Gamma(1 - \delta_{\alpha \beta}) & \rho_{\alpha \beta}(p, t) \\ \rho_{\alpha \beta}(p, t) & \rho_{\alpha \beta}(p, t) \end{bmatrix}$$

+nondiagonal $\Gamma$—the damping rate. In Eq. (27) the nondiagonal elements are interband polarization $\rho_{\perp, \perp}(p, t) = \rho_{22}(p, t)$ and its complex conjugate $\rho_{-\perp, \perp}(p, t) = \rho_{22}(p, t)$, and the diagonal elements represent particle distribution functions for conduction $\rho_{c, c}(p, t) = \rho_{11}(p, t)$ and valence $\rho_{v, v}(p, t) = \rho_{-1, -1}(p, t)$ bands. We will solve the set of differential equations for these functions:

$$i\hbar \frac{\partial N_c(p, t)}{\partial t} - i\hbar \epsilon \mathbf{E}(t) \frac{\partial N_c(p, t)}{\partial p}$$

$$= \mathbf{E}(t) \mathbf{D}_c(p) P^c(p, t) - \mathbf{E}(t) \mathbf{D}_c^*(p) P^c(p, t),$$

$$i\hbar \frac{\partial N_v(p, t)}{\partial t} - i\hbar \epsilon \mathbf{E}(t) \frac{\partial N_v(p, t)}{\partial p}$$

$$= -\mathbf{E}(t) \mathbf{D}_v(p) P^v(p, t) + \mathbf{E}(t) \mathbf{D}_c^*(p) P^c(p, t),$$

$$i\hbar \frac{\partial P(p, t)}{\partial t} - i\hbar \epsilon \mathbf{E}(t) \frac{\partial P(p, t)}{\partial p}$$

$$= \begin{bmatrix} 2\mathcal{E}_c(p) + \mathbf{E}(t) \mathbf{D}_c(p) - i\hbar \Gamma \mathbf{P}(p, t) \\ \mathbf{E}(t) \mathbf{D}_c(p) [N_c(p, t) - N_v(p, t)] \end{bmatrix}.$$
\begin{equation}
D_{\alpha}(\mathbf{p}) = \frac{\varepsilon h}{2|z|^2} \left[ \begin{array}{c}
v_3 p_y + v_1 p_x p_y \\
-v_1 (p_x^2 - p_y^2)
\end{array} \right],
\end{equation}

\begin{equation}
D_{\gamma}(\mathbf{p}) = \frac{\varepsilon h}{2|z|^2} \left[ \begin{array}{c}
v_3 p_y - v_1 p_x p_y \\
-v_1 (p_x^2 - p_y^2)
\end{array} \right],
\end{equation}

The components of the velocity operator given by the relation \( \hat{v}_x = \partial \hat{H} / \partial \mathbf{p} \) for the effective 2 \times 2 Hamiltonian (3), can be presented by the expressions:

\begin{equation}
\hat{v}_x = \frac{1}{m} \left( \begin{array}{cc}
1 & v_3 \\
-v_1 & 1
\end{array} \right),
\end{equation}

\begin{equation}
\hat{v}_y = \frac{1}{m} \left( \begin{array}{cc}
1 & v_3 \\
-v_1 & 1
\end{array} \right).
\end{equation}

The intraband velocity \( \mathbf{V}(\mathbf{p}) \) at HHG in bilayer AB-stacked graphene is given by the formula:

\begin{equation}
\mathbf{V}(\mathbf{p}) = \left[ v_3 \mathbf{p} - \frac{3v_3}{2m} \mathbf{p} \cos 3\theta + \frac{3v_3}{2m} \sin 3\theta \frac{\partial \mathbf{p}}{\partial \mathbf{p}} + \frac{2}{(2m)^2} \right] \zeta(\mathbf{p}).
\end{equation}

The author is deeply grateful to prof. H.K. Avetissian for permanent discussions and valuable recommendations. This work was supported by the Science Committee of Ministry of Education, Science, Culture and Sport of RA.

REFERENCES

1. K. S. Novoselov, A. K. Geim, S. V. Morozov, et al., Science (Washington, DC, U. S.) 306, 666 (2004).
2. A. K. Geim, Science (Washington, DC, U. S.) 324, 1530 (2009).
3. A. V. Rozhkov, A. O. Sboychakov, A. L. Rakhmanov, and F. Nori, Phys. Rep. 648, 1 (2016).
4. A. H. Castro Neto, F. Guinea, N. M. R. Peres, et al., Rev. Mod. Phys. 81, 109 (2009).
5. T. Brabec and F. Krausz, Rev. Mod. Phys. 72, 545 (2000).
6. H. K. Avetissian, Relativistic Nonlinear Electrodynamics, The QED Vacuum and Matter in Super-Strong Radiation Fields (Springer, Berlin, 2016).
7. Sh. Ghimire, A. D. DiChiara, E. Sistrunk, et al., Nature (London, U.K.) 7, 138 (2011).
8. O. Schubert, M. Hohenleutner, F. Langer, et al., Nat. Photon. 8, 119 (2014).
9. G. Vampa, T. J. Hammond, N. Thirat, et al., Nature (London, U.K.) 522, 462 (2015).
10. G. Ndbashimiye, S. Ghimire, M. Wu, et al., Nature (London, U.K.) 534, 520 (2016).
11. Y. S. You, D. A. Reis, and S. Ghimire, Nat. Phys. 13, 345 (2017).
12. H. Liu, C. Guo, G. Vampa, et al., Nat. Phys. 14, 1006 (2018).
13. S. A. Mikhailov and K. Ziegler, J. Phys.: Condens. Matter 20, 384204 (2008).
14. S. V. Syzranov, Ya. I. Rodionov, K. I. Kugel, and F. Nori, Phys. Rev. B 88, 241112(R) (2013).
15. Ya. I. Rodionov, K. I. Kugel, and F. Nori, Phys. Rev. B 94, 195108 (2016).
16. H. K. Avetissian, G. F. Mkrtchian, K. G. Batrakov, et al., Phys. Rev. B 88, 165411 (2013).
17. P. Bowlan, E. Martinez-Moreno, K. Reimann, et al., Phys. Rev. B 89, 041408 (2014).
18. I. Al-Naib, J. E. Sipe, and M. M. Dignam, New J. Phys. 17, 113018 (2015).
19. L. A. Chizhova, F. Libisch, and J. Burgdorfer, Phys. Rev. B 94, 075412 (2016).
20. H. K. Avetissian and G. F. Mkrtchian, Phys. Rev. B 94, 045419 (2016).
21. H. K. Avetissian, A. G. Ghazaryan, G. F. Mkrtchian, et al., J. Nanophoton. 11, 016004 (2017).
22. H. K. Avetissian, B. R Avchyan, G. F. Mkrtchian, et al., J. Nanophoton. 14, 026018 (2020).
23. L. A. Chizhova, F. Libisch, and J. Burgdorfer, Phys. Rev. B 95, 085436 (2017).
24. D. Dimitrovskii, L. B. Madsen, and T. G. Pedersen, Phys. Rev. B 95, 035405 (2017).
25. N. Yoshikawa, T. Tamaya, and K. Tanaka, Science (Washington, DC, U.S.) 356, 736 (2017).
26. A. Golub, R. Egger, C. Muller et al., Phys. Rev. Lett. 124, 110403 (2020).
27. H. K. Avetissian and G. F. Mkrtchian, Phys. Rev. B 97, 115454 (2018).
28. A. K. Avetissian, A. G. Ghazaryan, and Kh. V. Sedrakian, J. Nanophoton. 13, 036010 (2019).
29. A. G. Ghazaryan and Kh. V. Sedrakian, J. Nanophoton. 13, 046004 (2019).
30. A. G. Ghazaryan and Kh. V. Sedrakian, J. Nanophoton. 13, 046008 (2019).
31. A. K. Avetissian, A. G. Ghazaryan, K. V. Sedrakian, et al., J. Nanophoton. 11, 036004 (2017).
32. A. K. Avetissian, A. G. Ghazaryan, K. V. Sedrakian, et al., J. Nanophoton. 12, 016006 (2018).
33. H. K. Avetissian, A. K. Avetissian, A. G. Ghazaryan, et al., J. Nanophoton. 14, 026004 (2020).
34. Yu. Bludov, N. Peres, and M. Vasilevskiy, Phys. Rev. B 101, 075415 (2020).
35. G. L. Breton, A. Rubio, and N. Tancogne-Dejean, Phys. Rev. B 98, 165308 (2018).
36. H. Liu, Y. Li, Y. S. You et al., Nat. Phys. 13, 262 (2017).
37. G. F. Mkrtchian, A. Knorr, and M. Selig, Phys. Rev. B 100, 125401 (2020).
38. H. K. Avetissian, G. F. Mkrtchian, and K. Z. Hatsagortsyan, Phys. Rev. Res. 2, 023072 (2020).
39. H. K. Avetissian, A. K. Avetissian, B. R. Avchyan, et al., J. Phys.: Condens. Matter 30, 185302 (2018).
40. H. K. Avetissian, A. K. Avetissian, B. R. Avchyan, et al., Phys. Rev. B 100, 035434 (2019).
41. T. G. Pedersen, Phys. Rev. B 95, 235419 (2017).
42. H. K. Avetissian and G. F. Mkrtchian, Phys. Rev. B 99, 085432 (2019).
43. S. Almalki, A. M. Parks, G. Bart, et al., Phys. Rev. B 98, 144307 (2018).
44. B. Cheng, N. Kanda, T. N. Ikeda, et al., Rev. Lett. 124, 117402 (2020).
45. T. Cao, Z. Li, and S. G. Louie, Phys. Rev. Lett. 114, 236602 (2015).
46. L. Seixas, A. S. Rodin, A. Carvalho, et al., Phys. Rev. Lett. 116, 206803 (2016).
47. H. Sevincli, Nano Lett. 17, 2589 (2017).
48. J. Faist, F. Capasso, D. L. Sivco, et al., Science (Washington, DC, U.S.) 264, 533 (1994).
49. D. S. L. Abergel and T. Chakraborty, Appl. Phys. Lett. 95, 062107 (2009).
50. E. Suarez Morell and L. E. F. Foa Torres, Phys. Rev. B 86, 125449 (2010).
51. J. J. Dean and H. M. van Driel, Phys. Rev. B 82, 125411 (2010).
52. S. Wu, L. Mao, A. M. Jones et al., Nano Lett. 12, 2032 (2012).
53. Y. S. Ang, S. Sultan, and C. Zhang, Appl. Phys. Lett. 97, 243110 (2010).
54. N. Kumar, J. Kumar, C. Gerstenkornet, et al., Phys. Rev. B 87, 121406 (2013).
55. E. V. Castro, K. S. Novoselov, S. V. Morozov, et al., Phys. Rev. Lett. 99, 216802 (2007).
56. J. B. Oostinga, H. B. Heersche, X. Liu, et al., Nat. Mater. 7, 151 (2008).
57. Y. B. Zhang, T.-T. Tang, C. Girit, et al., Nature (London, U.K.) 459, 820 (2009).
58. F. Guinea, A. H. C. Neto, and N. M. R. Peres, Phys. Rev. B 73, 245426 (2006).
59. E. McCann and V. I. Falko, Phys. Rev. Lett. 96, 086805 (2006).
60. M. Koshino and T. Ando, Phys. Rev. B 73, 245403 (2006).
61. A. Varleta, M. Mucha-Kruczynski, D. Bischof, et al., Synth. Met. 210, 19 (2015).
62. M. Aoki and H. Amawashi, Solid State Commun. 142, 123 (2007).
63. L. A. Falkovsky, J. Exp. Theor. Phys. 110, 319 (2010).
64. K. Tang, R. Qin, J. Zhou, et al., J. Phys. Chem. C 115, 9458 (2011).
65. D. Xiao, M. C. Chang, and Q. Niu, Rev. Mod. Phys. 82, 1959 (2010).
66. L. Vicarelli, M. S Vitiello, D. Coquillat, et al., Nat. Mater. 11, 865 (2012).
67. K. Wang, M. M. Elahi, L. Wang et al., Proc. Natl. Acad. Sci. U. S. A. 116, 201816119 (2019).
68. I. M. Lifshitz, Sov. Phys. JETP 11, 1130 (1960).
69. J. L. Manes, F. Guinea, and M. A. H. Vozmediano, Phys. Rev. B 75, 155424 (2007).
70. G. P. Mikitik and Yu. V. Sharlai, Phys. Rev. B 77, 113407 (2008).
71. E. McCann, Phys. Rev. B 74, 161403 (2006).
72. H. Min, B. Sahu, S. Banerjee, and A. H. MacDonald, Phys. Rev. B 75, 155115 (2007).
73. E. McCann, D. Abergel, and V. Falko, Solid State Commun. 143, 110 (2007).
74. M. Mucha-Kruczynski, E. McCann, and V. I. Falko, Solid State Commun. 149, 1111 (2009).
75. D. Suszalski, G. Rut, and A. Rycerz, Phys. Rev. B 97, 125403 (2018).
76. M. Mucha-Kruczynski, I. L. Aleiner, and V. I. Falko, Phys. Rev. B 84, 041404 (2011).
77. M. Mucha-Kruczynski, I. L. Aleiner, and V. I. Falko, Solid State Commun. 151, 1088 (2011).
78. A. Varlet, D. Bischo, P. Simonet, et al., Phys. Rev. Lett. 113, 116602 (2014).
79. A. Varlet, M. Mucha-Kruczynski, D. Bischof, et al., Synth. Met. 210, 19 (2015).
80. L. V. Keldysh, Sov. Phys. JETP 7, 788 (1958).
81. L. V. Keldysh, Sov. Phys. JETP 20, 1307 (1965).
82. I. F. Akyildiz, J. M. Jornet, and C. Han, Phys. Commun. 12, 16 (2014).
83. H. Vettikalladi, W. T. Sethi, A. F. Bin Abas, et al., Int. J. Anten. Propagat. 2019, 9573647 (2019).
84. M. Lewenstein, Ph. Balcous, M. Yu. Ivanov, et al., Phys. Rev. A 49, 2117 (1994).
85. C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, Photons and Atoms. Introduction to Quantum Electrodynamics (Wiley, New York, 1989).
86. E. H. Hwang and S. Das Sarma, Phys. Rev. B 77, 115449 (2008).
87. J. K. Viljas and T. T. Heikkila, Phys. Rev. B 81, 245404 (2010).