Induced by coherent THz-radiation high harmonics generation in bilayer graphene at high Fermi energies

A.G. Ghazaryan a, H.H. Matevosyan b, and Kh.V. Sedrakian a

a Centre of Strong Fields Physics, Yerevan State University,
1 A. Manukian, Yerevan 0025, Armenia
b Institute Radiophysics and Electronics NAS RA,
1 Alikhanian brs., Ashtarak 0203, Armenia

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The higher order harmonic generation process in the nonperturbative regime at the interaction of coherent electromagnetic radiation with the AB-stacked bilayer graphene at high Fermi energies is considered. The applied coherent low-frequency radiation field in the high Fermi energy zone of electrons excludes the interband transitions enhancing high harmonic rates. The developed microscopic nonlinear quantum theory for charged carriers interaction with a strong pump wave is valid near the Dirac points of the Brillouin zone. The Liouville-von Neumann equation for the density matrix in the multiphoton excitation regime is solved both analytically and numerically. Based on the numerical solutions, we examine the rates of higher-order harmonics of the pump wave of arbitrary polarization. Obtained results show that bilayer graphene can serve as an effective material for the generation of higher order harmonics from THz to the mid-IR domain of frequencies at the pump wave moderate intensities.

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I. INTRODUCTION

The nonperturbative regime of emission of harmonics - high harmonic generation (HHG) [1], [2] is one of the main nonlinear phenomena at the interaction of strong coherent electromagnetic (EM) radiation with the matter [3], which has been widely studied both theoretically and experimentally since the appearance of high intensity and ultrashort pulse-lasers [4]. Up to the past decade, HHG phenomenon has been extensively investigated in atomic and molecular gases [5]. The intensity of the gaseous harmonics is relatively weak because of low gas density, so that in the recent years became actual the nonperturbative study of optical phenomena such as HHG in the condensed matter-solids. Currently, the investigations of HHG and related nonlinear processes have been successfully extended to bulk crystals [6–16] and two-dimensional (2D) nanostructures, such as graphene and graphene-like materials [17–35], monolayer transition metal dichalcogenides [36, 37], hexagonal boron nitride [38], topological insulators [39], buckled 2D hexagonal nanostructures [40], and also solids [41]. Under some conditions, the process of HHG in bulk solids is similar to atomic HHG [13] and can be well described within a semiclassical three-step model [42], [43]. According to this model the electron-hole is created, then accelerated, and in the last step re-collided. At that, the substantial contribution to the HHG process gives the interband current. The experiments [10, 12, 44–46] evidence this fact in solids. The experimental verification of HHG phenomenon in a single layer transition metal dichalcogenides [15] and graphene [28] open up a wide way to the higher harmonics radiation in 2D nanostructures. The appearance of a new nano-optical materials-graphene-like nanostructures-with a very high carrier mobility and extraordinary properties, such as graphene [47], [48] contributes using 2D nanostructures for nonlinear optical applications.

The experiments related to HHG process via THz pump wave pulses reported about weak signals of harmonics [21] or lack [49] of nonlinear response in graphene. This is connected with the enormous fast relaxation of electrons in graphene [50] that prevents nonlinear optical processes in the THz frequency region. So, the HHG experiment is considered with the mid-IR light, where the generation of up to ninth harmonic in graphene has been reported [28]. Theoretically, HHG has been investigated with even mid-IR light in buckled graphene-like 2D nanostructures [40], where it has been shown that with the static electric field applied perpendicular to the nanostructure sheet one can control the topology of the bands and, as a result, one can generate even and odd high harmonics of arbitrary polarization excited by a single-color infrared pump field. In [51], the multicolor harmonic generation and wave-mixing nonlinear processes in the efficient 2D nanostructures is studied. The presence of the second laser field provides an

*Electronic address: amarkos@ysu.am
additional flexibility for the implementation of multiphoton excitation \[52, 53\] expanding the spectrum of possible combinations.

Among the mentioned materials, the bilayer graphene (AB-stacked) \[48, 54, 55\] possessing many interesting properties of monolayer graphene \[56–59\], provides a richer band structure. The interlayer coupling between the two graphene sheets changes the monolayer’s Dirac cone, inducing trigonal warping on the band dispersion and changing the topology of the Fermi surface. This significantly enhances the rates of HHG \[20\] in the THz region compared to monolayer graphene. Studies of the nonlinear coherent response in AB-stacked bilayer graphene under the influence of intense EM radiation also includes modification of quasi-energy spectrum, the induction of valley polarized currents \[60, 61\], as well as second- and third-order nonlinear-optical effects \[62, 65\]. The important advantage of bilayer graphene over the monolayer one is the possibility to induce large tunable band gaps under the application of a symmetry-lowering perpendicular electric field \[56, 66, 68\].

In the present paper, we develop a nonlinear theory of the AB-stacked bilayer graphene interaction with the coherent EM radiation, apart from the advantages of the induced gap. For the second-order nonlinearity, one should also take into account the trigonal warping of the bands. We consider a multiphoton interaction in the nonperturbative regime. To exclude the interband transitions, we have considered high Fermi energies, and an external EM wave is taken to be from THz to the mid-IR domain of frequencies. We show that there is an intense emission of harmonics at the moderate intensities of the pump wave.

The paper is organized as follows. In Sec. II, the set of equations for a single-particle density matrix is formulated and solved both analytically and numerically in the multiphoton interaction regime. In Sec. III, we consider the problem of harmonic generation at the multiphoton excitation of AB-stacked bilayer graphene at high Fermi energies in the low-frequency strong coherent radiation field. Finally, conclusions are given in Sec. IV.

II. BASIC MODEL

In this paper we consider the multiphoton regime of harmonics generation in a bilayer graphene by coherent radiation with frequency \(\omega\) from THz to the mid-IR and electric field amplitude \(E_0\), at high Fermi energies of graphene electrons. The wave-particle interaction at photon energies \(\hbar\omega > E_L\) for intraband transitions can be characterized by the dimensionless parameter \(\chi = eE_0/(\omega\sqrt{n\hbar\omega})\). Here \(E_L = mv_F^2/2 \approx 1\) meV - is the Lifshitz transition energy, \(m = \gamma_1/(2v_F^2)\) is the effective mass, \(v_3 = \sqrt{3}\gamma_3/(2\hbar) \approx v_F/8\) is the effective velocity, \(a \approx 0.246\) nm is the distance between the nearest \(A\) sites, \(\gamma_1 \approx 0.39\) eV, \(\gamma_3 \approx 0.32\) eV is the interlayer hopping for the AB-stacked bilayer graphene; and \(v_F\) is the Fermi velocity in a monolayer graphene. In considering case, corresponding interaction parameter \(\chi \geq 1\).

The intensity of the wave can be estimated as

\[I_\chi = \chi^2 \times 6 \times 10^{10} \text{Wcm}^{-2}(\hbar\omega/eV)^3,\] \hspace{1cm} (1)

so the required intensity \(I_\chi\) for the nonlinear regime strongly depends on the parameter \(\chi\) and photon energy \(\hbar\omega\). Particularly, in the region from THz to the mid-IR photons with wavelengths from \(30\) \(\mu\)m to \(3\) mm, the multiphoton interaction regime can be achieved already at the values of the parameter \(\chi \sim 0.5\) corresponding to pump wave intensities \(I_\chi \sim 10\) kWcm\(^{-2}\). In the opposite limit \(\chi \ll 1\), the multiphoton effects are suppressed.

Let us consider the nonlinear microscopic theory of the multiphoton interaction of bilayer graphene interaction with a coherent EM radiation. We will discuss the ansatz of the evolutionary equation for a single-particle density matrix. For the AB-stacked bilayer graphene, the low-energy excitations \(|\varepsilon| \ll \gamma_1 \approx 0.39\) eV in the vicinity of the Dirac points \(\zeta K\) (valley quantum number \(\zeta = \pm 1\)) can be described by an effective single-particle Hamiltonian \[51, 59\]:

\[\tilde{H}_\zeta = \begin{pmatrix} 0 & g_\zeta(p) \\ g_\zeta^*(p) & 0 \end{pmatrix}.\] \hspace{1cm} (2)

Here the operator \(g_\zeta(p)\) is defined by the relation

\[g_\zeta(p) = -\frac{1}{2m}(\zeta\hat{p}_x + i\hat{p}_y)^2 + v_3(\zeta\hat{p}_x - i\hat{p}_y),\] \hspace{1cm} (3)

where \(\hat{p} = \{\hat{p}_x, \hat{p}_y\}\) is the electron momentum operator. The first term in Eq. \[43\] corresponds to a pair of parabolic bands \(\varepsilon = \pm p^2/(2m)\), and the second term coming from \(\gamma_3\) causes trigonal warping in the band dispersion. In the low-energy region \(\hbar\omega < E_L\) the two touching parabolas are transformed into the four separate “pockets” \[50\]. The spin and the valley quantum numbers are conserved. There is no degeneracy upon the valley quantum number \(\zeta\) for the issue considered. However, since there are no intervalley transitions, the valley index \(\zeta\) can be considered as a characteristic parameter.
FIG. 1: (Color online) Particle distribution function $N_c(p, t_f)$ (in arbitrary units) as a function of scaled dimensionless momentum components is shown. The wave is assumed to be linearly polarized along the $y$ axis. Multiphoton excitation with the trigonal warping effect for the photon energy $\hbar\omega = 50$ meV $\simeq 50\mathcal{E}_L$, the temperature $T/\hbar\omega = 0.5$ are demonstrated at dimensionless intensity parameter $\chi = 1$ for valley $\zeta = 1$. (a)–(d) correspond to the density plot of the distribution function after the interaction at the instant $t_f = 0.25T$, $t_f = 0.5T$, $t_f = 0.75T$ and $t_f = T$, respectively.

The eigenstate functions of the effective Hamiltonian (2) are the spinors,

$$\Psi_\sigma(r) = \frac{1}{\sqrt{S}} |\sigma, p\rangle e^{i\hat{p}r},$$

where

$$|\sigma, p\rangle = \frac{1}{\sqrt{2S}} \left( \frac{1}{\zeta} \Upsilon(p) \right),$$

with the eigenenergies:

$$\mathcal{E}_\sigma(p) = \sigma \sqrt{(v_3p)^2 - \frac{3}{\zeta} \frac{v_3^3p^3}{m} \cos 3\vartheta + \left( \frac{p^2}{2m} \right)^2}.$$ (6)

Here, $\sigma$ is the band index: $\sigma = 1$ and $\sigma = -1$ for conduction and valence bands, respectively; $\vartheta$ is the angle $\vartheta = \arctan(p_y/p_x)$, $S$ is the quantization area, and

$$\Upsilon(p) = -\frac{p^2}{2m} e^{i2\zeta \vartheta} + \zeta v_3 p e^{-i\zeta \vartheta}. $$ (7)

We will investigate the case when the bilayer graphene interacts with a plane quasimonochromatic EM radiation of carrier frequency $\omega$ and slowly varying envelope. To exclude the effect of the magnetic field, the wave is taken propagating in the perpendicular direction to the graphene sheets ($XY$). To investigate the dependence on the wave polarization, the pump wave is taken to be elliptically polarized with $E(t)$:

$$E(t) = f(t) E_0 \left( \hat{x} \sin \phi \cos \omega t + \hat{y} \cos \phi \sin \omega t \right).$$ (8)

The slowly varying envelope of the wave describes by the $\sin^2$-squared envelope function:

$$f(t) = \begin{cases} 
\sin^2(\pi t/T), & 0 \leq t \leq T \\
0, & t < 0, t > T 
\end{cases}.$$ (9)
FIG. 2: Harmonic emission rate in bilayer graphene at multiphoton excitation via \( \log_{10}(n^2|J_n|^2) \) (in arbitrary units), as a function of the photon energy (in units of \( \hbar \omega \)), is shown for various intensities. The temperature is taken to be \( T / \hbar \omega = 0.5 \). The wave is assumed to be linearly polarized (\( \phi = 0 \)) with frequency \( \omega = 50 \) meV/\( \hbar \). The results are for the intensity parameter (a) \( \chi = 0.5 \), (b) \( \chi = 1 \), (c) \( \chi = 1.5 \), and (d) \( \chi = 2 \).

where \( T \) characterizes the pulse duration and is taken to be ten wave cycles: \( T = 10T_0 \); \( \phi \) is the pump wave polarization parameter, \( T_0 = 2\pi / \omega \) is the wave period.

To follow the second quantization formalism, expanding the fermionic field operators on the basis of the states \( \Psi_\sigma(r) \) \( \{5\} \),

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

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

\[
\hat{H} = \hat{H}_\zeta + \begin{pmatrix} \varepsilon(r) E(t) & 0 \\ 0 & \varepsilon(r) E(t) \end{pmatrix},
\]

where the light-matter interaction Hamiltonian is taken in the length gauge \([42, 43]\) that provides proper inclusion of the inter- and intraband transitions \([51]\). Taking into account an expansion \([10]\), the second quantized total Hamiltonian can be expressed as:

\[
\hat{H} = \sum_{\sigma, p} \mathcal{E}_\sigma(p) \hat{a}^+_{\sigma p} \hat{a}_{\sigma p} + \hat{H}_s,
\]

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

\[
\hat{H}_s = ie \sum_{p, p', \sigma} \delta_{p', p} \partial_t \mathbf{E}(t) \hat{a}^+_{p, \sigma} \hat{a}_{p', \sigma'}
\]
\[ + \sum_{p,\sigma} \mathbf{E}(t) \left( \mathbf{D}_t(\sigma, p) \hat{a}^\dagger_{p,\sigma} \hat{a}_{p,-\sigma} + \mathbf{D}_m(\sigma, p) \hat{a}^\dagger_{p,\sigma} \hat{a}_{p,\sigma} \right). \]  

Here the quantity \( \mathbf{D}_t(\sigma, p) \),

\[ \mathbf{D}_t(\sigma, p) = \hbar e \langle \sigma, p |i\hbar \partial_p | - \sigma, p \rangle \]

is the transition dipole moment and \( \mathbf{D}_m(\sigma, p) \)

\[ \mathbf{D}_m(\sigma, p) = \hbar e \langle \sigma, p |i\hbar \partial_p | \sigma, p \rangle \]

is the Berry connection or the mean dipole moment. Note that the matrix elements (14), (15) are actually gauge-dependent.

We will represent the multiphoton interaction of a bilayer graphene with a coherent EM wave field by the Liouville–von Neumann equation for a single-particle density matrix

\[ \dot{\rho}_{\alpha,\beta}(p, t) = \langle \hat{a}_{p,\beta}^\dagger(t) \hat{a}_{p,\alpha}(t) \rangle. \]

Here \( \hat{a}_{p,\alpha}(t) \) obeys the Heisenberg equation

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

The evolutionary equation for the single-particle density matrix will write in the form, using Eqs. (12)-(17):

\[ i\hbar \frac{\partial \rho_{\alpha,\beta}(p, t)}{\partial t} = -\frac{1}{\hbar} \mathbf{E}(t) \frac{\partial \rho_{\alpha,\beta}(p, t)}{\partial p} - \frac{e}{m} \mathbf{E}(t) \mathbf{D}_m(\alpha, p) \rho_{\alpha,\beta}(p, t) - \frac{e}{m} \mathbf{D}_m(\beta, p) \rho_{\alpha,\beta}(p, t) + \mathbf{E}(t) \left[ \mathbf{D}_t(\alpha, p) \rho_{-\alpha,\beta}(p, t) - \mathbf{D}_t(-\beta, p) \rho_{\alpha,-\beta}(p, t) \right]. \]

Here \( \Gamma \) is the damping rate. Note that due to the homogeneity of the problem we only need the \( p \)-diagonal elements of the density matrix. We will also incorporate relaxation processes into Liouville–von Neumann equation with inhomogeneous phenomenological damping term, since homogeneous relaxation processes are slow compared with inhomogeneous one.

In Eq. (18) the diagonal elements represent particle distribution functions for conduction \( N_c(p, t) = \rho_{1,1}(p, t) \) and valence \( N_v(p, t) = \rho_{-1,-1}(p, t) \) bands, and the nondiagonal elements are interband polarization \( \rho_{1,-1}(p, t) = P(p, t) \) and its complex conjugate \( \rho_{-1,1}(p, t) = P^*(p, t) \). As an initial state \( (t = 0) \) we assume an ideal Fermi gas in equilibrium with particle distribution function

\[ \rho_{\sigma,\sigma'}(0, 0) = \frac{1}{1 + e^{(\varepsilon_{\sigma'} - \mu)/T}} \delta_{\sigma,\sigma'}, \]

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

It is clear that a strong field will induce multiphoton transitions from the valence to the conduction band. Thus, because of Pauli blocking and low frequency of driving wave the interband transitions can take place via multiphoton channels: \( n_0 \hbar \omega > 2\varepsilon_F \). We will consider the case when \( 2\varepsilon_F \gg \hbar \omega \) and consequently \( n_0 \gg 1 \). This is a quasiclassical regime when the wave-particle interaction can be characterized by the work of the wave electric field during the wave period \( eE_0v_\chi/\omega \). To neglect the interband transitions the latter should be smaller than \( 2\varepsilon_F : eE_0v_\chi/\omega < 2\varepsilon_F \). This means that the wavefield can not provide sufficient energy for the creation of an electron-hole pair.

Thus, in particular case of a pump EM pulse with THz up to mid-IR frequencies and for high Fermi energies the interband transitions are excluded, one can simplify the closed set of the differential equations Eq. (18). In addition, we will assume that the system relaxates at a rate \( \gamma \) to the equilibrium \( N_c^{(0)}(p, t) \) distribution. Thus, from (18) we obtain the Boltzmann equation for the electrons distribution function \( N_c(p, t) \):

\[ i\hbar \frac{\partial N_c(p, t)}{\partial t} - \hbar e \mathbf{E}(t) \frac{\partial N_c(p, t)}{\partial p} = -\gamma \left( N_c(p, t) - N_c^{(0)}(p, t) \right). \]
FIG. 3: High harmonic spectra for bilayer graphene at multiphoton excitation is shown for various wave frequencies in logarithmic scale. The temperature is taken to be $T/\hbar \omega = 0.5$. The wave is assumed to be linearly polarized ($\phi = 0$) with the intensity $\chi = 1.5$. The results are for frequency (a) $\omega = 40 \text{ meV}/\hbar$, (b) $\omega = 50 \text{ meV}/\hbar$, (c) $\omega = 60 \text{ meV}/\hbar$, and (d) $\omega = 70 \text{ meV}/\hbar$.

We construct $N_c(0) (\mathbf{p}, t)$ from the filling of electron states according to the Fermi–Dirac distribution Eq. (19):

$$N_c(0) (\mathbf{p}, t) = \frac{1}{1 + e^{E_1(\mathbf{p}) - \mu} / T}.$$  

(21)

Note that this relaxation approximation provides an accurate description for the optical field components oscillating at frequencies $\omega \gg \gamma$ (in accordance with the relaxation rate $\Gamma \ll T^{-1}$).

The Eq. (20) can be solved analytically by the method of characteristics, then the solution has the form:

$$N_c(\mathbf{p}, t) = \gamma \int d\tau e^{-\gamma(t-\tau)} \left( N_c(0) (\mathbf{p} + \mathbf{p}_E (t, \tau)) \right),$$

(22)

where

$$\mathbf{p}_E (t, \tau) = -e \int_\tau^t \mathbf{E} (\tilde{t}) d\tilde{t}.$$  

(23)

is the momentum given by the wave field.

For the coherent part of the radiation spectrum, one needs the mean value of the current density operator,

$$j_\zeta = -2e \left\langle \hat{\Psi}(\mathbf{r}, t) | \hat{\mathbf{v}}_\zeta | \hat{\Psi}(\mathbf{r}, t) \right\rangle,$$

(24)

where $\hat{\mathbf{v}}_\zeta = \partial \hat{H} / \partial \hat{\mathbf{p}}$ is the velocity operator and we have taken into account the spin degeneracy factor 2. For the effective $2 \times 2$ Hamiltonian (2) the velocity operator in components reads:

$$\hat{\mathbf{v}}_\zeta = \zeta \left( \begin{array}{cc} 0 & -\frac{1}{m} (\zeta \hat{p}_x - i \hat{p}_y) + v_3 \\ -\frac{1}{m} (\zeta \hat{p}_x + i \hat{p}_y) + v_3 & 0 \end{array} \right),$$

(25)
\[ \hat{\nu}_{\zeta} = i \left( -\frac{1}{m} (\hat{p}_{\zeta x} + i\hat{p}_{\zeta y}) - v_3 \frac{1}{m} (\hat{p}_{\zeta x} - i\hat{p}_{\zeta y}) + v_3 \right). \]  \hspace{1cm} (26)

Using the Eqs. (24)–(26) and (16), the expectation value of the current for the valley \( \zeta \) can be written in the form:

\[ \hat{j}_z(t) = -\frac{e}{2\pi^2\hbar^2} \int dp V(p) N_z(p, t) \]  \hspace{1cm} (27)

where

\[ V(p) = \frac{v_3 p - 3\zeta v_{3\beta} p \cos 3\theta + 3\zeta v_{3\beta}^3 \sin 3\theta \frac{\partial \nu}{\partial p} + 2\frac{p^2}{(2m)^2}}{\varepsilon_1(p)} \]  \hspace{1cm} (28)

is the intraband velocity. As is seen from Eq. (27), in considering case the surface current provides a source for the generation of harmonic radiation. It is the intraband current \( N_z(p, t) \). The intraband high harmonics are generated as a result of the independent motion of carriers in their respective bands. The interband high harmonics (given by the term \( N_v(p, t) \)) which are generated as a result of the recombination of accelerated electron-hole pairs \([68]\), are unessential in the considering case.

Let us now consider a particular case of coherent interaction of a bilayer graphene with a pump EM 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 \( \varepsilon \ll \gamma_1 \approx 0.39 \text{ eV} \), the dominant mechanism for relaxation will be electron-phonon coupling via longitudinal acoustic phonons \([69]\), \([70]\). For the low-temperature limit, if \( T \ll 2(c_{\text{ph}}/v_F) \sqrt{\varepsilon \gamma_1} \), where \( c_{\text{ph}} \approx 2 \times 10^6 \text{ cm/s} \) is the velocity of longitudinal acoustic phonons, the relaxation time for the energy level \( \varepsilon \) can be estimated as \([70]\):

\[ \tau(\varepsilon) \approx \left( \frac{\pi D^2 T^2}{8 \rho_m \hbar^3 c_{\text{ph}}^2 V_F} \sqrt{\frac{\gamma_1}{\varepsilon}} \right)^{-1}. \]  \hspace{1cm} (29)

For the high-temperature limit \( T \gg 2(c_{\text{ph}}/v_F) \sqrt{\varepsilon \gamma_1} (\varepsilon \approx \mu = 10\hbar \omega, T \text{ is the room temperature}) \) we can use the relation:

\[ \tau \approx \left( \frac{D^2 T \gamma_1}{4 \rho_m \hbar^3 c_{\text{ph}}^2 V_F^2} \right)^{-1}. \]  \hspace{1cm} (30)

Here \( D \approx 20 \text{ eV} \) is the electron-phonon coupling constant, and \( \rho_m \approx 15 \times 10^{-8} \text{ g/cm}^2 \) is the mass density of a bilayer graphene. For \( \hbar \omega \approx 0.05 \text{ eV} \) and \( \varepsilon \approx \mu = 10\hbar \omega \) at the temperatures \( T = 0.025 \text{ eV} (T \gg 2(c_{\text{ph}}/v_F) \sqrt{\varepsilon \gamma_1}) \), from Eq. (30) we obtain \( \tau \approx 1.1 \text{ ps} \). Thus, in this energy range one can coherently manipulate with the multiphoton transitions in a bilayer graphene on time scales \( t \lesssim 1 \text{ ps} \) neglecting the particle-particle collisions.

### III. HHG AT THE ELECTRONS INTRABAND MULTIPHOTON EXCITATIONS

In this section, we will investigate the nonlinear response of a bilayer graphene in the process of higher harmonics generation under the influence of a pump EM wave with the frequencies in the domain from THz to mid-IR ones: \( \omega = 0.04 \pm 0.07 \text{ eV}/\hbar \).

Taking into account Eqs. (27) and (22), the total intraband current can be represented as:

\[ \hat{j}_z(t) = -\frac{\gamma e}{2\pi^2\hbar^2} \int_{-\infty}^{t} d\tau e^{-\gamma(t-\tau)} \int dp V(p) \left( N_z^{(0)}(p + p_E(t, \tau)) \right). \]  \hspace{1cm} (31)

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

\[ j_x = j_{1,x} + j_{-1,x}; \]  \hspace{1cm} (32)

\[ j_y = j_{1,y} + j_{-1,y}. \]  \hspace{1cm} (33)

From Eq. (31) we see that

\[ \frac{j_{x,y}}{j_0} = J_{x,y} \left( \omega t, \chi, \frac{\gamma}{\omega}, \frac{\varepsilon_L}{\hbar \omega}, \frac{\mu}{\hbar \omega}, \frac{T}{\hbar \omega} \right), \]  \hspace{1cm} (34)
where
\[ j_0 = \frac{e\omega}{\pi} \sqrt{\frac{m\omega}{\hbar}}, \]

and \( J_x \) and \( J_y \) are the dimensionless periodic (for monochromatic wave) functions which parametrically depend on the interaction parameter \( \chi \), scaled Lifshitz energy, and macroscopic parameters of the system. Thus, having solutions of \( \text{Eq. (20)} \) and making integration in \( \text{Eq. (31)} \), one can calculate the harmonic radiation spectra with the help of a Fourier transform of the function \( J_{x,y}(t) \). The emission rate of the \( n \)th harmonic is proportional to \( n^2|j_n|^2 \), where \( |j_n|^2 = |j_{xn}|^2 + |j_{yn}|^2 \), with \( j_{xn} \) and \( j_{yn} \) being the \( n \)th Fourier components of the field-induced total current. To find \( j_n \), the fast Fourier transform algorithm has been used. We have used the normalized current density \( 34 \) for the plots.

For the following investigations, we made a change of variables and transform the equations with partial derivatives into ordinary ones. The new variables are \( t \) and \( \tilde{p} = p - p_F(t) \). After the simple transformations, the integration of \( \text{equation (20)} \) is performed on a homogeneous grid of \( 10^4 \) \((\tilde{p}_x, \tilde{p}_y)\)-points. For the maximal momentum, we take \( p_{\text{max}}/\sqrt{m\hbar\omega} = 5.8 \). The time integration is performed numerically with the standard fourth-order Runge-Kutta algorithm. For numerical analysis of HHG rates in bilayer graphene, we assume high Fermi energy \( \varepsilon_F \simeq \mu = 10\hbar\omega \) \((\varepsilon_F \gg \hbar\omega)\). The damping rate \( \gamma/\omega = 0.5 \) will be assumed in all plots below.

In Fig. 4-5 the temperature is taken \( T = 0.025 \) eV, and the pump wave is assumed to be linearly polarized along the \( y \) axis. In Fig. 1, the wave pulse duration for \( \omega = 10\varepsilon_F/\hbar = 50 \) meV is \( T = 10\tau_0 \simeq 0.82 \) ps. Photoexcitations of the Fermi-Dirac sea are presented in Fig.1, where the density plot of the particle distribution function \( N_p(p_f) \) is shown as a function of scaled dimensionless momentum components after the interaction at the different instances of the pump wave pulse duration. The picture partially changes with the on and off the pulse of the wave. This figure shows the electrons’ intraband transition only. The nonlinear trigonal warping effect describing the deviation of the excited iso-energy contours from circles is seen clearly.

In Fig. 2 the high harmonic spectra for bilayer graphene at multiphoton excitation is shown for various wave intensities. As it is seen, the contribution of higher-order harmonics in emission rate is more significant with the intensity increase. The analysis also shows the linear dependence of the harmonics number cutoff on the amplitude of a pump electric field \( n_{\text{eff}} \sim \chi \). Note that in the considering case of an intrinsic gapless bilayer, the system possesses in-plane inversion symmetry, and at the normal incidence of the pump wave on the bilayer graphene only odd harmonics are generated [18].

For clarification of the harmonics generation regime, we examine the emission rate of the higher harmonics versus pump wave strength \( \chi \) at the same wave intensity \( I_x \simeq 1.7 \times 10^7 \text{ Wcm}^{-2} \) for various frequencies, which is shown in Fig. 3. Third \( (J_3/\chi^3) \) and fifth \( (J_5/\chi^5) \) harmonics scaled emission rate for bilayer graphene versus \( \chi \) is demonstrated in Fig. 4. As is seen from this figure, up to the field strengths \( \chi < 1 \) we almost have power-law for the emission rate in accordance with the perturbation theory. For large \( \chi \) we have a strong deviation from power law for the emission rate of high harmonics. The temperature dependence is demonstrated in Fig. 5. This investigation shows that in intrinsic bilayer graphene [20] the harmonics are suppressed at high temperatures. As show the plots of Fig. 2-5 at

\[ \text{FIG. 4: Third (} J_3/\chi^3 \text{) and fifth (} J_5/\chi^5 \text{) harmonics scaled emission rate (in arbitrary units) for bilayer graphene versus } \chi. \text{ The temperature is taken to be } T/\hbar\omega = 0.5. \text{ The wave is assumed to be linearly polarized along the } y \text{ axis with the frequency } \omega = 50 \text{ meV/} \hbar. \]
FIG. 5: High harmonic spectra for bilayer graphene at multiphoton excitation for a linearly polarized wave ($\phi = 0$) is shown for temperatures $T/\hbar \omega = 0.1$ and $T/\hbar \omega = 0.5$. The wave intensity $\chi = 1.5$ and frequency $\omega = 50$ meV/$\hbar$.

FIG. 6: High harmonic spectra in logarithmic scale for a elliptically polarized wave is shown at temperature $T/\hbar \omega = 0.5$ at the wave intensity $\chi = 1.5$ and frequency $\omega = 50$ meV/$\hbar$. The results are for (a) $\phi = 0$, (b) $\phi = \pi/6$, (c) $\phi = \pi/4$, and (d) $\phi = \pi/2$, respectively.

high Fermi energies the HHG rates are larger compare with the intrinsic bilayer graphene with taking into account the interband transitions with intraband ones.

Finally in Fig. 6, we show the dependence of HHG on the polarization of the pump wave. The results are for linearly polarized wave along the $x$ ($\phi = \pi/2$) and $y$ ($\phi = 0$) axes, for circular polarization ($\phi = \pi/4$) and for elliptic polarization ($\phi = \pi/6$). As is seen, orienting the linearly polarized pump wave along with these axes results in different harmonics spectra. This is because we have strongly anisotropic excitation near the Dirac points. For elliptic and
circular polarizations the rates for the middle harmonics increase, while high order harmonics are suppressed.

As was mentioned in [20], the current amplitude $I_n$ compare to intrinsic graphene $j_0$ [18], [19], for bilayer graphene is larger by a factor $(\gamma_1/2\hbar \omega)^{1/2}$. Besides, the cutoff harmonic is larger than in the case of monolayer graphene [18], which is a result of strong nonlinearity caused by trigonal warping. Hence, for considered setups $\hbar \omega \ll \gamma_1$ the harmonics’ radiation intensity is at least one order of magnitude larger than in the monolayer graphene.

Here we will estimate the conversion efficiency for harmonics $\eta_n = I_n/I$ as in [20]

$$\eta_n \sim 10^{-3} \chi^2 (d/\lambda)^2 (nJ_n)^2,$$

where $\lambda = 2\pi c/\omega$ and $d$ is the characteristic size of the bilayer graphene sheet. For the setup of Fig. 2 at the intensity parameter $\chi \simeq 1$, depending on the ratio, even for the $d \sim \lambda$, one can achieve conversion efficiencies $\eta_n \sim 10^{-2}$ for up to the ninth harmonic. Note that these quite large conversion efficiencies are obtained for high order harmonics in the case of the single bilayer graphene sheet. For the low order harmonics, we have $\eta_3 \sim 10^{-7}$, and for the experimental realization one can use multilayer $N_l \gg 1$ sheet [71] up to experimentally achievable values $N_l \sim 1$ monolayers [72] with the thickness $\sim 20$ nm. Since film thickness is much smaller than the considered wavelengths, the harmonics’ signal from all layers will sum up constructively. Thus, for the average conversion efficiencies we will have the same magnitude $\sim 0.02$. In an experiment, one can use many layers, which are comparable to what one expects to achieve with resonant two-level systems in nonlinear optics [72].

IV. CONCLUSION

On the base of the microscopic quantum theory of nonlinear interaction of a bilayer graphene with a coherent EM radiation at high Fermi energies of electrons towards the high harmonics generation has been investigated. The differential equations for the single-particle density matrix is solved both analytically and numerically in the vicinity of $\zeta K$ points in the Brillouin zone. We have considered the practically interesting regimes of multiphoton excitation of the Fermi-Dirac sea for effective generation of harmonics via the pump wave pulses from THz to the mid-IR domain of frequencies. The considered domains of frequencies and high Fermi energies exclude the valence band excitations and interband transitions. The cutoff of harmonics in these regimes increases with the pump wave intensity enhancement and harmonics emission processes become robust against the temperature increase. As a result of the strong nonlinearity caused by the trigonal warping, the current amplitude for bilayer graphene is at one order of the magnitude larger than in the intrinsic graphene. Moreover, it has been shown strict growing of HHG rates in considering case compare to the case of HHG in bilayer graphene with the intra- and interband multiphoton transitions [68]. The obtained results show that bilayer graphene may serve as an effective medium for generation of higher harmonics at room temperatures by the pump waves from THz to mid-IR frequencies and intensits $\sim 10$ kWcm$^{-2}$. In this context, the requirement of high intensity in the THz regime does not preclude the use of standard THz lasers, which are available [74]. Moreover, if in the THz domain of pump wave frequencies reported about weak signals at HHG [21] concerning the generation of high harmonics up to the mid-IR range, note that this has been demonstrated in the paper [68] where Quantum Cascade lasers [73] are readily available and can provide higher powers.

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