Impact of electron-electron Coulomb interaction on the high harmonic generation process in graphene

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Generation of high harmonics in a monolayer graphene initiated by strong coherent radiation field, taking into account electron-electron Coulomb interaction is investigated. A microscopic theory describing the nonlinear optical response of graphene is developed. The Coulomb interaction of electrons is treated in the scope of dynamic Hartree-Fock approximation. The closed set of integrodifferential equations for the single-particle density matrix of a graphene quantum structure is solved numerically. The obtained solutions show the significance of many-body Coulomb interaction on the high harmonic generation process in graphene.

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I. INTRODUCTION

In the last decade, graphene and its analogs have attracted enormous interest due to their unique electronic and optical properties. The potential of graphene as an effective nonlinear optical material has triggered many nonlinear optical studies. In particular, graphene-like nanostructures can serve as an active medium for nanolasers and frequency multipliers. In their original structure freestanding graphene is centrosymmetric and even-order nonlinear effects vanish within the dipole approximation. The latter is fully justified for perpendicular incidence of a pump wave to the graphene plane, and the symmetry-allowed odd-order nonlinear optical effects are very strong in graphene. This is confirmed by the experimental and theoretical investigations of the third harmonic generation process. With the increase of the pump wave intensity one can enter into the regime where multiphoton effects are essential and high-harmonics are generated, which until last decade have been the prerogative of atomic systems.

As was shown experimentally and theoretically in contrast to the single-particle picture, the real spectrum of graphene is nonlinear near the neutrality point, and Fermi velocity describing its slope, increases significantly. Even an excitonic condensate along with the opening of an energy gap has been predicted.

Electron-electron interaction also significantly modifies linear optical response of graphene. Hence, it is of interest to clear up the influence of electron-electron interaction on the nonlinear optical response of graphene, which is the subject of the present investigation.

In the present work, we develop a nonlinear microscopic theory of a monolayer graphene interaction with the coherent electromagnetic radiation taking into account the electron-electron Coulomb interaction using the self-consistent Hartree-Fock approximation that leads to a closed set of integrodifferential equations for the single-particle density matrix. We neglect the scattering processes which are described by the second-order terms in the carrier-carrier interaction. Thus, we consider nonlinear coherent interaction in the ultrafast excitation regime when relaxation processes are not relevant. Since we are interested in both inter and intraband transitions for the light-matter interaction Hamiltonian, we use a length gauge. As is well known, in this gauge it is straightforward to study quantum transitions via intermediate states and to obtain gauge-independent transition probabilities.

The derived equations are solved numerically for a graphene in the Dirac cone approximation. Then we consider high harmonic generation process for moderately strong pump waves and show that one can achieve considerable enhancement of the harmonic generation rate due to the many-body Coulomb interaction between the charge carriers.

The paper is organized as follows. In Sec. II the Hamiltonian with many-body Coulomb interaction in the scope of mean-field approximation and the set of equations for a single-particle density matrix are formulated. In Sec. III, we consider multiphoton excitation of Fermi-Dirac
sea and generation of harmonics in graphene. Finally, conclusions are given in Sec. IV.

II. EVOLUTIONARY EQUATION FOR SINGLE-PARTICLE DENSITY MATRIX

Let a graphene monolayer interact with plane quasi-monochromatic electromagnetic wave field. To exclude the effect of wave’s magnetic field we assume that the wave propagates in a perpendicular direction to the graphene plane (XY). Thus, this travelling wave for graphene electrons becomes a homogeneous quasiperiodic electric field of carrier frequency $\omega$ and slowly varying envelope $E_0(t)$. We assume linearly polarized (along the x-axis) wave:

$$E(t) = \hat{k} E_0(t) \cos \omega t. \quad (1)$$

The wave amplitude is described by the sin-squared envelope function $E_0(t) = E_0 f(t)$:

$$f(t) = \left\{ \begin{array}{ll} \sin^2(\pi t/T_p), & 0 \leq t \leq T_p, \\ 0, & t < 0, t > T_p, \end{array} \right. \quad (2)$$

where $T_p$ characterizes the pulse duration. Note that, the Gaussian and sin-squared envelopes lead to very similar results. The latter is more convenient for numerical and analytical calculations.

Low-energy excitations which are much smaller than the nearest neighbor hopping energy can be described by an effective Hamiltonian

$$H_0 = \hbar v_F \left( \begin{array}{cc} 0 & \hat{k}_x - i\hat{k}_y \\ \hat{k}_x + i\hat{k}_y & 0 \end{array} \right), \quad (3)$$

where $v_F \approx c/300$ is the Fermi velocity ($c$ is the light speed in vacuum), $\hbar \hat{k}$ is the electron momentum operator. The eigenstates of the effective Hamiltonian are the spinors,

$$\psi_{k,\lambda}(r) = \frac{1}{\sqrt{2A}} \left( \begin{array}{c} e^{i\theta(k)} \\ \lambda \end{array} \right) e^{ikr}, \quad (4)$$

corresponding to energies

$$\mathcal{E}_\lambda(k) = \hbar v_F k. \quad (5)$$

Here the band index $\lambda = \pm 1$, and $A$ is the quantization area,

$$\theta(k) = \arctan \left( \frac{k_y}{k_x} \right) \quad (6)$$

is the polar angle in the momentum space. We will work in the second quantization formalism, expanding the fermionic field operators on the basis of states given in (4), that is,

$$\hat{\Psi}(r) = \sum_{k,\lambda} \hat{c}_{\lambda,k} \psi_{k,\lambda}(r), \quad (6)$$

where $\hat{c}_{\lambda,k}$ ($\hat{c}_{\lambda,k}^\dagger$) is the annihilation (creation) operator for an electron with momentum $k$ and band $\lambda$ (for conduction ($\lambda = 1$) and valence ($\lambda = -1$) bands). In (6) we have omitted real spin and valley quantum numbers because of degeneracy.

The electrons interact through long-range Coulomb forces and the Hamiltonian for electron-electron interactions can be written in terms of the field operators, $\hat{\Psi}(r)$, as:

$$\hat{H}_c = \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \hat{\Psi}^\dagger(\mathbf{r}) \hat{\Psi}^\dagger(\mathbf{r}') V_c(\mathbf{r} - \mathbf{r}') \hat{\Psi}(\mathbf{r}) \hat{\Psi}(\mathbf{r}), \quad (7)$$

where $V_c(r) = e^2/(\varepsilon |\mathbf{r}|)$ is the bare Coulomb potential, $\varepsilon$ is the effective dielectric constant of the substrate on which graphene is deposited.

The light–matter interaction part is taken in the length gauge:

$$\hat{H}_{\text{int}} = e \int d\mathbf{r} \hat{r} \hat{\Psi}^\dagger(\mathbf{r}) \mathbf{E}(t) \hat{\Psi}(\mathbf{r}). \quad (7)$$

The latter is given in terms of the gauge-independent field $E(t)$.

Taking into account expansion (6), the total Hamiltonian can be represented as follow:

$$\hat{H} = \sum_{\lambda,k} \mathcal{E}_\lambda(k) \hat{c}_{\lambda,k}^\dagger \hat{c}_{\lambda,k} + H_{\text{Coul}}$$

$$+ ie \sum_{\lambda,k,k'} \left( \mathbf{E} \cdot \frac{\partial \delta(k - k')}{\partial k'} \right)$$

$$\times \left( \mathcal{D}_{k,k'}^{(+)} \hat{c}_{\lambda,k}^\dagger \hat{c}_{\lambda,k} + \mathcal{D}_{k,k'}^{(-)} \hat{c}_{\lambda,k}^\dagger \hat{c}_{-\lambda,k} \right). \quad (8)$$

The Dirac delta function $\delta(k - k')$ in the light-matter interaction part provides proper inclusion of inter and intraband transitions. The Coulomb interaction reads:

$$\hat{H}_{\text{Coul}} = \frac{1}{2A} \sum_{\lambda_1\lambda_2\lambda_3\lambda_4} \sum_{\mathbf{q},\mathbf{k},\mathbf{k}'} V_{2D}(\mathbf{q}) F_{\lambda_1\lambda_2\lambda_3\lambda_4}(\mathbf{q},\mathbf{k},\mathbf{k}')$$

$$\times \hat{c}_{\lambda_1,k+\mathbf{q}}^\dagger \hat{c}_{\lambda_2,k'}^\dagger \hat{c}_{\lambda_3,k} \hat{c}_{\lambda_4,k'}, \quad (9)$$

where

$$V_{2D}(\mathbf{q}) = \frac{2\pi e^2}{\varepsilon |\mathbf{q}|} F(\mathbf{q}) \quad (10)$$

is the 2D Coulomb potential in momentum space and

$$F_{\lambda_1\lambda_2\lambda_3\lambda_4}(\mathbf{q},\mathbf{k},\mathbf{k}') = \frac{1}{4} [\lambda_1 \lambda_2 \lambda_3 \lambda_4]$$

$$+ e^{i[\theta(k+\mathbf{q})+\theta(k'-\mathbf{q})-\theta(k)-\theta(\mathbf{k}')}}$$
\[ D^{(\pm)}_{k'k} = \frac{1}{2} \left( e^{i(\theta(k') - \theta(k))} \pm 1 \right). \]  

At that, the term proportional to \( D^{(+)}_{k'k} \) is responsible for intraband transitions, while the term proportional to \( D^{(-)}_{k'k} \) describes interband transitions.

The Coulomb interaction part \( \bar{V} \) contains products of four fermionic operators. We will treat Coulomb interaction in the scope of mean-field theory, reducing the Hamiltonian into so-called mean-field Hamiltonian which allows obtaining closed set of equations for the dynamic quantities. We need to choose the proper mean field parameters. Due to the homogeneity of the applied wave-field and initial system, as a mean field parameters are taken distribution functions for conduction \( N_c (k,t) = \langle \hat{c}^\dagger \hat{c} \rangle \) and for valence \( N_v (k,t) = \langle \hat{c}^\dagger \hat{c} \rangle \) bands carriers, and interband polarization \( \mathcal{P} (k,t) = \langle \hat{c}^\dagger \hat{c} \rangle \). For the Coulomb interaction, this is a Hartree-Fock approximation. For the mean-field Hamiltonian we will use the following decompositions:

\[ \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} = \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Fock}} = -\tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} = \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} \]

\[ \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} = \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Fock}} = -\tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} = \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} \]

with the condition

\[ \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle = \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle \delta_{\nu \mu}; \quad f, g = a, b, c, d. \]

Taking into account mean-field parameters, the second quantized Hamiltonian \( \bar{H} \) Eqs. \( 13 \), and \( 14 \) for the Coulomb part we have

\[ H_{\text{MFC}} = -\frac{1}{\lambda} \sum_{\lambda_1 \lambda_2 \lambda_3 \lambda_4} \sum_{k' \neq k} V_{2D} (k' - k) \bar{F}_{\lambda_1 \lambda_2 \lambda_3 \lambda_4} (k, k') \]

\[ \times \langle \bar{c}^\dagger_{\lambda_1,k} \bar{c}_{\lambda_4,k'} \rangle \bar{c}^\dagger_{\lambda_2,k} \bar{c}_{\lambda_3,k} \]

\[ + \frac{1}{2\lambda} \sum_{\lambda_1 \lambda_2 \lambda_3 \lambda_4} \sum_{k' \neq k} V_{2D} (k' - k) F_{\lambda_1 \lambda_2 \lambda_3 \lambda_4} (k, k') \]

\[ \times \langle \bar{c}^\dagger_{\lambda_1,k} \bar{c}_{\lambda_3,k'} \rangle \bar{c}^\dagger_{\lambda_2,k} \bar{c}_{\lambda_4,k} \]

\[ \bar{f}_{\mu} \delta_{\nu \mu}; \quad f, g = a, b, c, d. \]

\[ \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle = \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle \delta_{\nu \mu}; \quad f, g = a, b, c, d. \]

\[ \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} = \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} \]

\[ \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} = \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Fock}} = -\tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} = \tilde{\alpha}^\dagger \tilde{\alpha} \tilde{\beta}^\dagger \tilde{\beta} \tilde{\gamma} \tilde{\delta} \bigg|_{\text{Hartree}} \bigg|_{\text{Fock}} \]

\[ \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle = \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle \delta_{\nu \mu}; \quad f, g = a, b, c, d. \]

\[ \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle = \langle \tilde{f}_{\mu} \tilde{g}_{\nu} \rangle \delta_{\nu \mu}; \quad f, g = a, b, c, d. \]
\[
\frac{\partial N_e (\mathbf{k}, t)}{\partial t} - \frac{eE}{\hbar} \frac{\partial N_e (\mathbf{k}, t)}{\partial \mathbf{k}} = -i\Omega_{R-C} (\mathbf{k}, t) P^* (\mathbf{k}, t) + \text{c.c.}, \tag{22}
\]

\[
\frac{\partial P (\mathbf{k}, t)}{\partial t} - \frac{eE}{\hbar} \frac{\partial P (\mathbf{k}, t)}{\partial \mathbf{k}} = i\omega_{D-C} (\mathbf{k}, t) P (\mathbf{k}, t) - i\Omega_{R-C} (\mathbf{k}, t) (N_e (\mathbf{k}, t) - N_v (\mathbf{k}, t)), \tag{23}
\]

where

\[
\Omega_{R-C} (\mathbf{k}, t) = \frac{eE}{2\hbar} \left(\frac{\partial \theta (\mathbf{k})}{\partial \mathbf{k}} - i \frac{1}{2\hbar A} \sum_{\mathbf{k}' \neq \mathbf{k}} V_{2D} (\mathbf{k} - \mathbf{k}') \times \sin \left[\theta (\mathbf{k}) - \theta (\mathbf{k}')\right] (N_e (\mathbf{k}', t) - N_v (\mathbf{k}', t)) \right)
\]

\[- \frac{1}{\hbar A} \sum_{\mathbf{k}' \neq \mathbf{k}} V_{2D} (\mathbf{k} - \mathbf{k}') |P' (\mathbf{k}', t) + i \cos \left[\theta (\mathbf{k}) - \theta (\mathbf{k}')\right] P'' (\mathbf{k}', t)| \times \cos \left[\theta (\mathbf{k}) - \theta (\mathbf{k}')\right] (N_v (\mathbf{k}', t) - N_e (\mathbf{k}', t)) \]

\[+ \frac{2}{\hbar A} \sum_{\mathbf{k}' \neq \mathbf{k}} V_{2D} (\mathbf{k} - \mathbf{k}') \sin \left[\theta (\mathbf{k}) - \theta (\mathbf{k}')\right] |P'' (\mathbf{k}', t)|. \tag{25}
\]

As is seen from Eqs. (21)-(25) in the scope of mean-field approximation the Coulomb interaction leads to a renormalization of the light-matter coupling, which depends on \( P \) and \( N_{e,v} \). Also, the transition energies become renormalized due to the Coulomb interaction. Note that, in general, one should also include in Eqs. (21)-(25) the relaxation terms because of carrier-carrier and the carrier-phonon scatterings. Hence, our consideration is valid in the ultrafast excitation regime and it is correct only for the times \( t < \tau_{\text{min}} \), where \( \tau_{\text{min}} \) is the minimum of all relaxation times. Experiments and theory suggest that carrier-carrier scattering and the carrier-phonon coupling are the main relaxation channels for the radiation-excited current in graphene. At the excitation by 800nm laser, the carrier-carrier scattering results in a decrease of the current with a decay constant of 100 fs that is almost 40 wave periods. Electron-electron interactions in graphene give rise to a linear energy dependence of the inverse lifetime. Thus, one can extrapolate this scale to low energy excitations. The electron-phonon coupling in graphene is considerable for optical phonons. Therefore here we consider excitation with THz waves far below the energy threshold for the emission of optical phonons (0.2 eV being a characteristic optical phonon frequency). In considered case, the electron linewidth due to electron-phonon interaction is negligible, while it increases linearly beyond this threshold making relaxation times about 1 ps. This, hence the pulse duration \( T_p = 36T \) where \( T \) is the wave period, which allows us do not include relaxation processes.

The obtained equations are closed set of nonlinear integro-differential equations which should be solved with the proper initial conditions. In the scope of the mean-field theory one can define the ground state self-consistently. In this case, at vanishing temperatures the ground state is an excitonic condensate with a certain energy gap. The gap size is very sensitive to the 2D model of Coulomb potential and is very small when one takes into account Fermi velocity renormalization. According to the experimental result, the gap is smaller than \( \sim 0.1 \) meV. Note that tight-binding ground state is also solution of the stationary self-consistent mean-field equations. Therefore, the initial state one can assume Fermi-Dirac distribution with the temperature larger than the predicted excitonic gap:

\[
N_{c,v} (\mathbf{k}, 0) \simeq \frac{1}{1 + e^{\frac{\omega - \omega_{\text{F}}}{k_F}}}, \quad P (\mathbf{k}, 0) = 0. \tag{26}
\]

Here \( k_F \) is the Fermi wave number and it is assumed linear dispersion. The Fermi velocity renormalization is incorporated into the definition of scaled temperature \( \tilde{T} \). For the latter it is assumed \( T^* = 0.1\hbar\omega/v_F \).

For the initial functions \( \omega_{D-C} (\mathbf{k}, 0) = 0 \) and

\[
\omega_{D-C} (\mathbf{k}, 0) = 2\tilde{v}_F k, \tag{28}
\]

where

\[
\tilde{v}_F = v_F + \frac{e^2}{2\pi\varepsilon\hbar k} \times \int_0^{k_c} k_c \, dk' \int_0^{2\pi} \cos \theta \, d\theta \frac{(N_e (k', 0) - N_e (k', 0))}{\sqrt{k^2 + k'^2 - 2kk' \cos \theta}} \tag{29}
\]

is the renormalized Fermi velocity. We note that the integral of Eq. (29) has a ultraviolet high-momentum logarithmic divergence, which must be regularized through a high wave vector cutoff \( k_c \), of the order of the inverse lattice spacing. Conserving the total number of states in the Brillouin zone, we choose \( k_c = (4\pi/A_c)^{1/2} \), where \( A_c = 3\sqrt{3}a^2/2 \) is the area of the hexagonal unit cell, and \( a = 1.42 \times 10^{-8} \) cm is the carbon-carbon distance.

Thus the renormalized frequency can be represented as

\[
\omega_{D-C} (\mathbf{k}, t) = \omega_{D-C} (\mathbf{k}, 0) + \tilde{\omega}_{D-C} (\mathbf{k}, t) \tag{30}
\]
FIG. 1: (Color online) Electron distribution function $N_c(k,t_f)$ (in arbitrary units) after the interaction at the instant $t_f = 36\tau$, as a function of dimensionless momentum components. The Coulomb interaction parameter $\alpha_g = 0$. The wave-particle dimensionless interaction parameter is taken to be $\chi_0 = 0.3$.

where $\omega_{D-C}(k,0)$ is given by the regularized expression

Because of finite excitation of Brillouin zone near the Dirac points now $\tilde{\omega}_{D-C}(k,t)$ and $\Omega_{R-C}(k,t)$ are convergent and one can make integration only near Dirac points.

III. MULTIPHOTON EXCITATION AND GENERATION OF HARMONICS

As was mentioned above, equations (21), (22), and (23) are integrodifferential set of nonlinear equations, which can not be solved analytically. Before numerical solution, one can considerably simplify the problem. We can make a change of variables and transform the partial differential equations into ordinary ones. The new variables are $t$ and $k = k - k_E(t)$, where

$$\hbar k_E(t) = -e \int_0^t E(t') dt'$$

is the classical momentum given by the wave field. After these transformations, the integration of equations (21)-(23) is performed on a grid of $6000-20000$ $(k_0, \theta_0)$-points depending on the intensity of the pump wave. For the integration over polar angle, we use Gaussian quadrature with 60 points. For $k_0$ we take points homogeneously distributed between the points $k_0 = 0$ and $k_0 = \alpha \omega_0/\nu_F$, where $\alpha$ depends on the intensity of the pump wave. The time integration is performed with the standard fourth-order Runge-Kutta algorithm.

In graphene, because of the linear scaling of the kinetic energy with momentum, the ratio of Coulomb to kinetic energy is independent of the electronic density and equals to $\alpha_g = e^2/(\varepsilon \hbar \nu_F)$ depending only on material properties and environmental conditions. For freestanding graphene ($\varepsilon = 1$) $\alpha_g \approx 2.2$. In most of the experiments, graphene lies on top of some substrate. In particular, for substrate $\text{SiO}_2$ the Coulomb interaction is moderate. For graphene in contact with air and $\text{SiO}_2$, $\varepsilon \approx 2.75$ and for interaction parameter we have $\alpha_g \approx 0.8$. The background dielectric constant can be significantly enhanced in the presence of substrates in contact with strong dielectric liquids such as ethanol ($\varepsilon \approx 13$). Thus, in the experiment one can change $\varepsilon$ and, as a consequence, to tune Coulomb interaction. In graphene, the wave-particle interaction is
to the electron-electron interactions, and as a consequence, the velocity increases because of self-energy corrections due to transition frequency \( \omega_t \). In particular, effective Fermi is the mean value of the Coulomb and wave-fields dressed interaction parameter \( \alpha_g \). Here at \( \alpha_g = 0.5 \) four-photon transition is effective due to electron-electron Coulomb interaction.

We further examine the nonlinear response of graphene considering the generation of harmonics from the multiphoton excited states. At the multiphoton excitation, particle-hole annihilation will cause intense coherent radiation of the harmonics of the applied wave field. For the coherent part of the radiation spectrum, one needs the mean value of the current density operator. The optical excitation via a linearly polarized coherent radiation pulse induces transitions in the Fermi-Dirac sea which results in the surface current in the polarization direction of the pump wave

\[
J_x(t) = -\frac{e g_s g_v}{\pi^2} \int d\mathbf{k} \mathbf{E}_F \text{cos} \theta(\mathbf{k}) \left( \mathcal{N}_c(\mathbf{k}, t) - \mathcal{N}_v(\mathbf{k}, t) \right) + 2 \sin \Theta(\mathbf{p}) \mathcal{P}''(\mathbf{k}, t),
\]

where \( g_s = 2 \) and \( g_v = 2 \) are the spin and valley degeneracy factors, respectively. This current has a nonlinear dependence on the pump wave field. At that, due to the graphene symmetries, one can expect intense radiation of odd harmonics of the incoming wave field. The harmonics will be described by the additional generated fields \( E_x^{(g)} \). We assume that the generated fields are considerably smaller than the incoming field \( |E_x| \ll |E| \). In this case, we can solve Maxwell's wave equation in the propagation direction with the given source term:

\[
\frac{\partial^2 E_x^{(t)}}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E_x^{(t)}}{\partial t^2} = \frac{4\pi}{c} \frac{\partial J_x(t)}{\partial t} \delta(z).
\]

Here \( \delta(z) \) is the Dirac delta function (\( z = 0 \) is the graphene plane), \( E_x^{(t)} \) is the total field. The solution to equation \( (32) \) reads

\[
E_x^{(t)}(t, z) = E_x(t - z/c) - \frac{2\pi}{c} \left[ \theta(z) J_x(t - z/c) + \theta(-z) J_x(t + z/c) \right],
\]

where \( \theta(z) \) is the Heaviside step function. The first term in Eq. \( (33) \) is the pump wave. From Eq. \( (33) \), we see that after the encounter with the graphene sheet two propagating waves are generated. One traveling in the propagation direction of the incoming pulse and one traveling in the opposite direction. We assume that the spectrum is measured at a fixed observation point in the forward propagation direction. For the generated field at \( z > 0 \) we have

\[
E_x^{(g)}(t - z/c) = \frac{2\pi}{c} J_x(t - z/c).
\]

\[
\chi_0 = \frac{e E_0 V_F}{\omega_0} \frac{1}{\hbar \omega_0},
\]

which represents the work of the wave electric field \( E_0 \) on a period \( 1/\omega_0 \) in the units of photon energy \( \hbar \omega_0 \). Here we consider moderately strong pump waves \( \chi_0 \lesssim 1 \). Photoexcitations of the Fermi-Dirac sea are presented in Figs. 1–4. In Fig. 1 and 2 density plot of the particle distribution function \( N_c(\mathbf{k}, t_f) \) after the interaction at the instant \( t_f = 36 T \), as a function of dimensionless momentum components are shown. In both figures the wave-particle dimensionless interaction parameter \( \chi_0 \) is taken to be \( \chi_0 = 0.3 \). As is seen from these figures, depending on the strength of the Coulomb interaction, the photoexcitation picture changes significantly. Thus, for \( \alpha_g = 0 \) the main contribution is conditioned by the one and two-photon transitions. For \( \alpha_g = 2.2 \) it is clearly seen the three-photon transition. This is a consequence of Coulomb interaction on the quasienergy spectrum. Thus, the multiphoton probabilities of particle-hole pair production will have maximal values for the resonant transitions

\[
\varpi_0(\mathbf{k}_0) = \hbar \omega_c, \quad n = 1, 2, 3, \ldots,
\]

where

\[
\varpi_0(\mathbf{k}_0) = \frac{1}{T} \int_0^T \omega_{\text{D}-c}(\mathbf{k}_0 + \mathbf{k}_E(t), t) \, dt
\]

is the mean value of the Coulomb and wave-fields dressed transition frequency \( \omega_{\text{D}-c} \). In particular, effective Fermi velocity increases because of self-energy corrections due to the electron-electron interactions, and as a consequence resonant wave numbers are decreased. Besides, due to the enhancement of the effective interaction parameter, we see in Fig. 2 three-photon transitions. The same picture we see in Figs. 3 and 4 for slightly higher intensities but for moderate Coulomb interaction parameter \( \alpha_g = 0.8 \). Here at \( \alpha_g = 0.5 \) four-photon transition is effective due to electron-electron Coulomb interaction.

FIG. 4: (Color online) Same as Fig. 3 but for the Coulomb interaction parameter \( \alpha_g = 0.8 \).
Thus, solving Eqs. (21), (22), and (23) with the initial condition (26) and making integration in Eq. (31), one can reveal nonlinear response of the graphene.

Before proceeding to the high harmonic generation process we will analyze the impact of Coulomb interaction on the electromagnetic response of graphene when the perturbation theory is valid. We will consider linear and third order response of graphene assuming infinite perturbation theory of the linear response of graphene. The perturbation result of the linear conductivity depends on the interaction parameter \( \alpha_g \). As is seen from Fig. 5, for small \( \alpha_g \) the linear conductivity is close to the universal value \( \sigma_0 = e^2 / (4 \hbar) \). However, it increases for the large interaction parameter \( \alpha_g \). The latter is consistent with the perturbative result of the linear response of graphene. The third order conductivity strongly depends on the pump frequency \( \omega_0^4 \), so in Fig. 6 we plot \( |\sigma(3)| / \kappa \) versus Fermi wave number. The pump wavelength is taken to be \( \lambda_0 = 0.01 \, \text{cm} \).

Making Fourier transform of the generated field (34), one can calculate the strength of the harmonics. The emission strength of the th\( \text{h} \) harmonic will be characterized by the dimensionless parameter

\[
\chi_s = \frac{e |E_x^{(g)}(s)| v_F}{\hbar \omega_0^2} = \chi_0 \left| \frac{E_x^{(g)}(s)}{E_0} \right|,
\]

(36)

where

\[
E_x^{(g)}(s) = \frac{\omega_0}{2\pi} \int_0^{2\pi/\omega_0} E_x^{(g)}(t) e^{i\omega_0 t} \, dt.
\]

(37)

With the fast Fourier transform algorithm instead of discrete functions \( \chi_s \) we calculate smooth function \( \chi(\omega) \) and so \( \chi_s = \chi(s \omega_0) \).

Figures 7 and 8 show the radiation spectrum via logarithm of the normalized field strength \( \chi(\omega) \) for \( \alpha_g = 2.2 \) and \( \alpha_g = 0 \), respectively. Here we plot \( \chi(\omega) \) versus Fermi wave number. The latter is normalized to \( k_0 = \omega_0 / v_F \). The pump wavelength is taken to be \( \lambda_0 = 0.01 \, \text{cm} \). Comparing Figs. 7 and 8 we see the strong influence of the Coulomb interaction on the high harmonics radiation spectrum. In Fig. 7 with strong Coulomb interaction, 5th and 7th harmonics appear, while at \( \alpha_g = 0 \) only 3rd harmonic is feasible. We also see that due to Coulomb interaction the peaks are broadened.

We also have made calculations for moderate Coulomb interaction parameter \( \alpha_g = 0.8 \). The results are shown in Figures 9 and 10. In this case, we also see the enhancement of harmonics order due to Coulomb interaction.
In Fig. 11 we plot high harmonics generation rate depending on the Coulomb interaction parameter at the fixed values of the Fermi wave number ($k_F = 0$) and pump wave frequency. As is seen from Fig. 11, we have an increase of the harmonics emission rates at the large Coulomb interaction.

We also examine how the revealed picture behaves depending on the pump wave frequency at $k_F = 0$. The results of our calculations are shown in Fig. 12. Thus, at moderately strong pump waves for the broad range of frequencies we have intense radiation of harmonics due to Coulomb interaction.

For the stronger pump waves $\chi_0 > 1$ one should increase integration domain in Eqs. (24), (29), and decrease time step, which considerably enhance computation time and requires calculations on the supercomputer. However, taking into account above reported results, one can definitely state that at the consideration of high-harmonics generation in graphene one should take into account collective electron-electron interaction.

We have taken into account the electron-electron interaction in the Hartree-Fock approximation. The latter is justified if the characteristic Coulomb interaction energy is smaller than the kinetic energy of electrons. For the massless particles the ratio of interaction energy to the kinetic energy, as mentioned above, does not depend on the electronic density, and is $\alpha_g$. However, after the renormalization of Fermi velocity $\tilde{v}_F$ here the effective interaction parameter is $\tilde{\alpha}_g = e^2/(\varepsilon_0 \hbar \tilde{v}_F) < 1$. Hence,
FIG. 12: (Color online) The radiation spectrum via logarithm of the normalized field strength $\chi(\omega)$ (in arbitrary units) versus pump wavelength. The Fermi wave number is taken to be $k_F = 0$. The dimensionless interaction parameter is taken to be $\chi_0 = 0.5$. The Coulomb interaction parameter $\alpha_g = 0.8$.

We have presented the microscopic theory of nonlinear interaction of the monolayer graphene with strong coherent radiation field taking into account many-body electron-electron Coulomb interaction. For the Coulomb interaction, we have used the self-consistent Hartree-Fock approximation that leads to a closed set of integrodifferential equations for the single-particle density matrix. The latter is solved numerically for graphene in the Dirac cone approximation and ultrafast excitation regime. For the pump wave, THz frequency range has been taken. We have considered multiphoton excitation of Fermi-Dirac sea towards the high harmonics generation. It has been shown that the role of Coulomb interaction in the nonlinear optical response of graphene is quite considerable that persist for a wide range of the pump wave frequencies and intensities. Numerical calculations show that one can reach the efficient generation of high harmonics with radiation fields of moderate intensities due to Coulomb mediated enhancement of harmonics order. Because of limited computation resources, we have made calculations for $\chi_0 < 1$. However, our results show that at least in the THz range of pump wave frequencies and at the $\alpha_g > 0.5$ one should take into consideration many-body Coulomb interaction for investigation of the nonlinear optical response of graphene.

**IV. CONCLUSION**

We have presented the microscopic theory of nonlinear interaction of the monolayer graphene with strong coherent radiation field taking into account many-body electron-electron Coulomb interaction. For the Coulomb interaction, we have used the self-consistent Hartree-Fock approximation that leads to a closed set of integrodifferential equations for the single-particle density matrix. The latter is solved numerically for graphene in the Dirac cone approximation and ultrafast excitation regime. For the pump wave, THz frequency range has been taken. We have considered multiphoton excitation of Fermi-Dirac sea towards the high harmonics generation. It has been shown that the role of Coulomb interaction in the nonlinear optical response of graphene is quite considerable that persist for a wide range of the pump wave frequencies and intensities. Numerical calculations show that one can reach the efficient generation of high harmonics with radiation fields of moderate intensities due to Coulomb mediated enhancement of harmonics order. Because of limited computation resources, we have made calculations for $\chi_0 < 1$. However, our results show that at least in the THz range of pump wave frequencies and at the $\alpha_g > 0.5$ one should take into consideration many-body Coulomb interaction for investigation of the nonlinear optical response of graphene.

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