High Harmonic Generation in Triangular Graphene Quantum Dots

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Abstract—Higher harmonic generation in plane graphene quantum dots initiated by intense coherent radiation is investigated using dynamical Hartree–Fock mean-field theory. A microscopic theory describing the extreme nonlinear optical response of plane graphene quantum dots is developed. The closed set of differential equations for the single-particle density matrix at the multiphoton interaction of graphene quantum dots and strong laser field is solved numerically. The obtained solutions indicate the significance of the type of edge and lateral size and the band gap and laser field strength in the process of high harmonic generation in the triangular graphene quantum dot.

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1. INTRODUCTION

In the last decade, there has been growing interest to extend high harmonic generation (HHG) to two-dimensional (2D) crystals and nanostructures, such as semimetallic graphene [1] and semiconductor transition metal dichalcogenides [2]. The role of graphene as an effective nonlinear optical material has been discussed in many theoretical [3–22] and experimental [23, 24] studies that consider various extreme nonlinear optical effects, in particular, HHG, which take place in strong coherent radiation fields in the multiphoton regime at excitation of such nanostructures [25, 26]. On the other hand, apart from the remarkable and unique electronic and optical properties of graphene, the lack of an energy gap as a semimetal greatly limits their applicability, in contrast, for example, to bilayer graphene [27–32].

The problem of a zero energy gap has been solved by decreasing the lateral size of graphene [33]. As a result of dimensional quantization, an energy gap opens. Semimetallic graphene of finite dimensions becomes a semiconductor. Among carbon nanostructures, of particular interest as a nonlinear medium are graphene ribbons (nanoribbons) [33, 34], graphene-like quantum dots such as closed-convex fullerences of different basic symmetry [21, 22], and graphene quantum dots (GQDs) of various lateral sizes. The graphene nanostructure can be characterized by whether the sublattice symmetry is preserved. GQD has a gap that can be controlled by its lateral size, shape, and type of edge [33, 35]. For GQD, two types of ribs are possible: armchair and zigzag, and the presence or absence of sublattice symmetry plays an important role in determining the electronic properties of graphene nanostructures [33]. The behavior of GQDs is quantitatively different for structures with zigzag and armchair edges, which is related to the edge states present in systems with zigzag edges [36]. So, it is of interest to investigate the HHG process in GQDs with different edges. Such nanostructure exhibits optical properties that are fundamentally different from those of graphene [37–39]. At the same time, carriers in GQD have the same outstanding transport properties as in graphene [3].

The important advantage of GQDs over graphene nanoribbons [40] is the limitation of quasiparticles in space. The latter can be crucial for the efficiency of HHG, since the space limitation prevents the propagation of the electron wave packet deposited into also one additional dimension and, therefore, can enhance of the HHG yield [41].

In the present work, the HHG in triangular GQDs caused by intense coherent radiation is investigated. The closed set of differential equations for the single-particle density matrix at the GQD—strong laser field multiphoton interaction is solved numerically within the scope of the microscopic theory describing the extreme nonlinear optical response of GQD. The Coulomb electron-electron interaction (EEI) is described in the Hubbard approximation. The GQD energy gap is controlled by its lateral size, shape, and type of edge. The obtained solutions indicate the significance of the lateral size on the HHG process in triangular GQDs with either armchair or zigzag edge. Thus, we have investigated theoretically the effect of quantum confinement on HHG in GQDs by system-
attractively varying the lateral size of a model dot. Note
that the considered quantum dots are accessible experimen-

tally [33].

The paper is organized as follows. In Section 2, the
set of equations for the single-particle density matrix is
formulated. In Section 3, we consider multiphoton
excitation and generation of harmonics in triangular
GQD with different types of edges and lateral sizes.
Finally, conclusions are given in Section 4.

2. DYNAMICAL HARTREE–FOCK MEAN-
FIELD THEORY FOR THE HIGH-HARMONIC
GENERATION IN GQD

Let a plane GQD placed in the $xy$ plane bounded
along the $x$ and $y$ axes that interacts with a plane quasi-
monochromatic electromagnetic (EM) wave. We assume
that the EM wave propagates perpendicular to the
$xy$ plane. We assume neutral plane GQDs, which
interball hopping is much smaller than the on-ball
hopping. The TB Hamiltonian can describe finite-size
systems by restricting the tunneling matrix elements $t_{ij}$
to atoms within the quantum dot. The total Hamilton-
ian can be written as

$$
\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}.
$$

Here

$$
\hat{H}_0 = -\sum_{(i,j)\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_{\sigma} \left( c^\dagger_{i\sigma} c_{j\sigma} - \frac{n_i}{2} \right) \left( c^\dagger_{i\sigma} c_{j\sigma} - \frac{n_j}{2} \right)
$$

(2)

is the free Hamiltonian of the GQD for the TB model,
where $c^\dagger_{i\sigma}$ is the operator of creation of an electron with
spin polarization $\sigma$ at site $i$, and $(i,j)$ indicates a sum-

mation over nearest neighbor sites with the transfer
energy $t_{ij}$, $n_i$ is the total electron density for site $i$, and
$U$ is the EEI energy.

The TB Hamiltonian describes systems of finite
size by limiting the matrix elements $t_{ij}$ of tunneling
between atoms inside a quantum dot. The nonzero
matrix elements of the TB Hamiltonian given by the
first term in (2) correspond to the tunneling matrix
elements of the TB Hamiltonian given by the
between atoms inside a quantum dot. The total Hamilto-
nian can be written as

$$
\hat{H}_{\text{int}} = e \sum_{\sigma} \mathbf{r}_i \cdot \mathbf{E}(t) c^\dagger_{i\sigma} c_{i\sigma}
$$

(3)

with the elementary charge $e$, position vector $\mathbf{r}_i$, and
the electric field strength $\mathbf{E}(t)$. In the Hamiltonian, we
neglect the lattice vibrations. The hopping integral $t_{ij}$
between nearest-neighbor atoms of QGDs can be
determined experimentally and is usually taken to be
$t_{ij} = 2.7$ eV [3]. The wave is assumed:

$$
E_x(t) = f(t) E_0 \cos \omega t \cos \theta,
$$

$$
E_y(t) = f(t) E_0 \cos \omega t \sin \theta
$$

(3)

with the frequency $\omega$, pulse envelope $f(t) = \sin^2(\pi t/\tau)$,
the angle $\theta$ to the $x$ axis. The pulse duration $\tau$ is taken
to be 20 wave cycles: $\tau = 40\pi/\omega$. From the Heisenberg
equation $i \hbar \frac{\partial \hat{L}}{\partial t} = \{\hat{L}, \hat{H}\}$, one can obtain evolutionary
equations for the single-particle density matrix $\rho_{ij}^{(\sigma)} =
\langle c^\dagger_{i\sigma} c_{j\sigma}\rangle$. In addition, we will assume that the system
relaxes at a rate $\gamma$ to the equilibrium distribution $\rho_{ij}^{(\sigma)}$.

To obtain a closed set of equations for the single-par-
ticle density matrix $\rho_{ij}^{(\sigma)} = \langle c^\dagger_{i\sigma} c_{j\sigma}\rangle$, EEI will be consid-
ered under the Hartree–Fock approximation:

$$
\hat{H}_{\text{ee}}^{\text{HF}} = \frac{U}{2} \sum_{\sigma} \left( c^\dagger_{i\sigma} c_{i\sigma} - \frac{n_i}{2} \right) \left( c^\dagger_{i\sigma} c_{i\sigma} - \frac{n_i}{2} \right).
$$

(4)

Thus, we obtain the following equation for the den-
sity matrix:

$$
i \hbar \frac{\partial \rho_{ij}^{(\sigma)}}{\partial t} = \sum_{k} (t_{kj}^\dagger \rho_{ki}^{(\sigma)} - t_{jk} \rho_{ik}^{(\sigma)})
$$

$$
+ U (\rho_{ij}^{(\sigma)} - \rho_{ij}^{(\sigma)}) \rho_{ij}^{(\sigma)} + e \mathbf{E}(t) \cdot (\mathbf{r}_i - \mathbf{r}_j) \rho_{ij}^{(\sigma)}
$$

$$
- i \hbar \gamma (\rho_{ij}^{(\sigma)} - \rho_{ij}^{(\sigma)}).
$$

(5)

We numerically diagonalize the TB Hamiltonian
$\hat{H}_0$. For a half-filled system, the static Hartree–Fock
Hamiltonian vanishes, $\hat{H}_{\text{ee}}^{\text{HF}} = 0$. It should be men-
tioned that EEI in the Hartree–Fock limit is included
in empirical hopping integral $t_{ij}$ between nearest-
neighbor atoms which is chosen to be close to experi-
mental values. Thus, on-site EEI in the Hartree–Fock
approximation is relevant for the quantum dynamics
initiated by the pump laser field and, as we will see
below, considerably modifies the HHG spectrum.

With the numerical diagonalization, we find eigen-
states $\psi_\mu(t)$ and eigenenergies $\epsilon_\mu$ ($\mu = 0, 1, ..., N - 1,$
where \( N \) is the number of GQD atoms. The results of numerical diagonalization are shown in Figs. 3 and 4. Without tunneling, all energy levels were degenerate. So, the tunneling removed the degeneracy and led to the formation of the band of valence states below the Fermi level, a band of conduction states above the Fermi level (\( \epsilon_\mu = 0 \)), and a gap across the Fermi level (see also [33]). The quantum dynamics of the strong field of periodically driven GQD is governed by a closed set of differential equations (5), which should be solved with the proper initial conditions. We construct the initial density matrix \( \rho^{(0)}_{\mu i} \) via the filling of electron states in the valence band according to the Fermi–Dirac distribution. Since the energy gap is large enough, we assume the Fermi–Dirac distribution at zero temperature:

\[
\rho^{(0)}_{\mu i} = \sum_{\mu = N/2}^{N-1} \psi^*_\mu(j)\psi_\mu(i).
\]

### 3. NUMERICAL RESULTS FOR HHG EFFICIENCY IN TRIANGULAR GQD

The HHG spectrum is evaluated from the Fourier transformation \( \mathbf{a}(\Omega) \) of the dipole acceleration \( \mathbf{a}(t) = d^2 \mathbf{a}/dt^2 \). The dipole is defined as \( \mathbf{a}(t) = \langle \sum_{\mu} \mathbf{r}_{\mu}^\dagger \mathbf{c}_{\mu} \rangle \).

For precision, we normalize the dipole acceleration by the factor \( a_0 = \sigma_0 \mathcal{D} \), where \( \sigma_0 = 1 \) eV/\( \hbar \) and \( \mathcal{D} = 1 \) Å.

The power radiated at the given frequency is proportional to \( |\mathbf{a}(\Omega)|^2 \). In order to clarify the main aspects of HHG in triangular GQDs, we assume that the excitation frequency is \( \omega = 0.1 \) eV/\( \hbar \), which is much smaller than the typical gap \( U = 3 \) eV. The relaxation rate is taken to be \( \hbar \gamma = 50 \) meV. For most calculations, the wave (3) is assumed to be linearly polarized along the \( x \) axis (\( \theta = 0 \)). The \( x \) axis is in the plane of Figs. 1 and 2, and it is directed along \( x \) to the right. The wave–particle interaction will be characterized by the work of the electric field of the wave on the lattice spacing:

\[
W = eE_0a.
\]

Nonlinear and multiphoton effects take
place when $W$ becomes comparable to or larger than photon energy $\hbar \omega$. In our model, hexagonal two-dimensional nanostructures are formed from $\pi$ orbitals of carbon atoms. As a result, we neglect transitions in $\sigma$ and between $\pi$–$\sigma$ orbitals. These orbitals are separated from $\sigma$ orbitals by a large energy gap ($\sim 10$ eV) at several $t_{ij}$ [43]. Hence, we will consider moderately strong pump waves $\hbar \omega \ll W < t_{ij}$. For stronger fields, one should definitely take into account other bands.

Figures 1 and 2 illustrate schematically a graphene lattice and Figs. 3 and 4 show the TB energy spectrum in the vicinity of the Fermi level, $\varepsilon_F = 0$, for a triangular GQD with zigzag and armchair edges for the different numbers of carbon atoms. As is shown in Figs. 1–4, with an increase in the number of lattice atoms, the density of eigenstates increases. As will be seen later, this will enhance of the probabilities of multiphoton HHG.

To compare the HHG radiation spectra in the triangular GQDs with different edges at different numbers of lattice atoms, in further figures, we have plotted all results for the spectra normalized to the number of atoms $N$. In Fig. 5, we have plotted the components $|a_x(\Omega)|$ and $|a_y(\Omega)|$ in the strong EM wave with the amplitude $E_0 = 0.3$ V/Å and the EEI energy $U = 3$ eV. As shown in this figure, in a strong laser field, multiphoton harmonics are significant, and the HHG yields for GQDs are equally significant for both armchair and zigzag edges, especially for large $N$. In both cases, the HHG spectra have a multistep plateau structure, which is associated with the excitations of energy eigenstates between the unoccupied energy levels and the occupied level [33]. GQD with armchair-
like edges has axial symmetry (see Fig. 2), and, in particular, in this case, $|a_x| = 0$ (therefore, its graph is absent in Fig. 5), while in the case of zigzag edges both components $|a_x|$ and $|a_y|$ are visible (see Fig. 5). In addition, only odd harmonics are essential in the HHG spectrum in armchair-shaped GQDs, as in ordinary graphene [1]. However, for zigzag edges, owing to the absence of inversion symmetry, both odd and even harmonics are present in the HHG emission spectrum. To show this clearly, Fig. 6 separately shows the results for HHG for the first thirty harmonics with approximately the same number ($N \approx 60$) of atoms of a triangular GQD with different edges.

Next, we consider the HHG spectra as a function of pump wave intensity. Figure 7 presents the HHG spectra as a function of the EM field amplitude and the harmonic order for a fixed EEI energy $U \approx 3$ eV. For comparison, in Figs. 7a, 7c and Figs. 7b, 7d, we will investigate the GQDs with the same number of carbon atoms but with different edges. As shown in Fig. 7, the probability of HHG increases either with an increase in the number of atoms $N$ or with appearance of new energy states (see also Figs. 1–4). The number of the cutoff of harmonics linearly increases with increasing field strength. Then, upon reaching the harmonic $n_{\text{cut}}$ which corresponds to the transition of the lowest occupied energy state to the highest unoccupied one, the HHG rate is saturated (stepped yellow envelope). The cutoff of the harmonic numbers can be seen in Fig. 5: $n_{\text{cut}} = 160$. Note that the linear dependence of the cutoff of harmonics on the field strength is inherent in HHG via discrete levels or in crystals

Fig. 4. The same as for Fig. 3 but for triangular GQD with armchair edges. (a–d) $N = 60, 90, 126, 168$ atoms, respectively.
with linear energy dispersion. As in cases of atoms, for GQD with the growth of the pump wave electric strength at a fixed photon energy, the energy of cutoff of harmonic ($\omega_{\text{cut}}$) is increased.

In addition, as was shown in [29], the on-site EEI suppresses the charge fluctuation and reduces the absorbed energy. Suppression of HHG yields due to EEI is also expected. The latter is shown in Fig. 8, where the HHG spectra in the strong EM field regime versus harmonic number and EEI energy are shown for different border edges and carbon atom numbers.

As can be seen from Fig. 8, for a small number $N$ of GQD atoms with an increase in the EEI energy, the HHG rate is generally suppressed (therefore, Fig. 8 shows the dependence for mean harmonics with numbers <40). The latter is not the case for large $N$, when the density of energy states increases (Figs. 8c, 8d). The HHG spectrum ceases to depend on the Coulomb EEI. This property is inherent in ordinary graphene [1], unlimited in space.

We also investigated the dependence of the HHG spectra on the EM wave strength orientation. In Fig. 9,

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Fig. 5. The HHG emission rate expressed via Fourier transform of dipole acceleration $N^{-1}|a_i(\Omega)|/a_0$ on a logarithmic scale versus the harmonic number for triangular GQD. The components $|a_i|$ are given with (1) zigzag and (3) armchair edges; (2) the components $|a_j|$ are given with zigzag edges. (a–d) For cases 1 and 2, $N = 61, 97, 118, 141$ atoms, and (a–d) for case 3, $N = 60, 90, 126, 168$. EM wave is linearly polarized along the $x$ axis. The wave frequency is $\omega = 0.1$ eV/$h$ and the field strength is $E_0 = 0.3$ V/Å. The spectra are shown for moderate (typical) EEI energy $U = 3$ eV. The relaxation rate is $h\gamma = 50$ meV.
Fig. 6. The same as for Fig. 5 but without the logarithmic scale: shown are (a) harmonics (even and odd) in a triangular GQD with zigzag edges for $N = 60$ atoms and (b) harmonics (only odd) in a GQD with armchair edges for $N = 61$ atoms.

Fig. 7. The color bar represents the HHG emission rate via Fourier transform of dipole acceleration $N^{-1}|a_x(\Omega)|/a_0$ in the strong field regime on a logarithmic scale versus harmonic order and EM field amplitude for triangular GQD with different edges and number of carbon atoms $N = 61$ (a) and $N = 118$ (c) for GQD with zigzag edges and $N = 60$ (b) and $N = 126$ (d) for GQD with armchair edges. The wave is assumed to be linearly polarized along the $x$ axis. The wave frequency is $\omega = 0.1 \text{ eV}/\hbar$ and the EEI energy is $U = 3 \text{ eV}$. The relaxation rate is $\hbar \gamma = 50 \text{ meV}$.
we plot the HHG spectra versus the order of harmonics and pump wave strength orientation with respect to the x axis for a given strength ($E_0 = 0.3 \, \text{V/Å}$) and frequency and for moderate EEI energy $U = 3 \, \text{eV}$. As can be seen from Fig. 9, the orientation of the pump wave at different angles to the x axis leads to different harmonic spectra. This is due to the fact that the triangular GQD has no inversion symmetry (see Figs. 1–4). As shown in Figs. 9a and 9b for angles $0 < \theta < \pi/2$, the rate of middle harmonics (maxima for numbers $n = 10–40$) increases, and higher harmonics are suppressed. However, in Figs. 9c and 9d, it can be seen that, with an increase in the density of energy states, this regularity is violated, and the HHG spectrum ceases to depend on the orientation of the EM wave electric field strength. By this, the GQD becomes similar to graphene unbounded in space [1].

4. CONCLUSIONS

We have studied the influence of intense coherent radiation on GQDs. A microscopic theory has been developed to describe the extreme nonlinear optical response of triangular GQDs. A closed system of differential equations for a one-particle density matrix in the multiphoton interaction of a GQD with a strong laser field is solved numerically. The solutions obtained indicate the importance of the type of edge and lateral size, as well as the significance of the band gap and the magnitude of the laser field for the HHG process in plane GQDs. The harmonic cutoff number increases linearly with increasing electric field strength. As in the case of HHG on atoms, for a GQD, with an increase in the pump wave strength at fixed photon energy, the harmonic cutoff energy $\omega_{\text{cut}}$ increases linearly. We also investigated the dependence of the HHG spectra versus the pump wave strength orientation. Owing to the absence or presence of symmetry of the sublattice in a triangular GQD with zigzag edges, harmonics of both odd and even order appear during the generation in the field of an EM wave, but only odd harmonics are significant for armchair edges regardless of the laser field orientation. As the numerical results show, owing to the difference in the symmetry of the sublattice, the same angles give different partial yields in the HHG spectra for triangular GQDs with armchair and zigzag edges. In addition, the rate of middle harmonics for a small number of atoms increases, while higher harmonics are suppressed, which is not the case with an increase in the density of energy states. In addition, with a growth in the density of energy states, the rule is violated when,
in the case of a small number of GQD atoms, with a rise in the EEI energy, the HHG rate is generally suppressed. Thus, with an increase in the average density of eigenstates, the GQD behaves like graphene unbounded in space. So, we investigated the size and shape of the GQD using quasiparticle confinement in space. The obtained results show that GQDs can serve as an effective medium for the generation of even and odd high-order harmonics when interacting with a laser field of moderate intensity owing to the limitation of quasiparticles in the GQD. The probability of HHG increases with an increase in the number of GQD atoms or with the appearance of new energy states. This is a potential way to increase the quantum yield and photon energy during HHG in graphene-like quantum dots.

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Fig. 9. The HHG emission rate via Fourier transform of dipole acceleration $N^{-1} |a_\nu(\Omega)|/a_0$ versus harmonic order and angle $\theta$ of the intensity of the EM field with the x axis for $N = 61$ (a) and $N = 118$ (c) for a triangular GQD with zigzag edges and $N = 60$ (b) and $N = 126$ (d) for a triangular GQD with armchair edges. The wave frequency is $\omega = 0.1$ eV/ℏ, the field strength is $E_0 = 0.3$ V/Å, and the EEI energy is $U = 3$ eV. The relaxation rate is $\hbar \gamma = 50$ meV.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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