Multiphoton excitation and high-harmonics generation in topological insulator

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Abstract

Multiphoton interaction of coherent electromagnetic radiation with 2D metallic carriers confined on the surface of the 3D topological insulator is considered. A microscopic theory describing the nonlinear interaction of a strong wave and metallic carriers with many-body Coulomb interaction is developed. The set of integrodifferential equations for the interband polarization and carrier occupation distribution is solved numerically. Multiphoton excitation of Fermi–Dirac sea of 2D massless carriers is considered for a THz pump wave. It is shown that in the moderately strong pump wave field along with multiphoton interband/intraband transitions the intense radiation of high harmonics takes place.

Keywords: strong-field, topological insulator, high-harmonics, multiphoton, Hartree–Fock approximation

(Some figures may appear in colour only in the online journal)

1. Introduction

Along with graphene topological insulators (TI) recently emerged as a central theme in condensed matter physics [1, 2]. Three-dimensional TIs are bulk insulators endowed with a topological invariant that manifests itself through robust 2D metallic surface states. These states are helical with massless linear Dirac like energy dispersion, that is, each surface-momentum state possesses a unique spin direction and are protected against backscattering by time-reversal symmetry [3, 4]. The unique properties of the surface states are responsible for their exotic electromagnetic properties. Several theoretical works on light-TI interaction have illustrated interesting effects [5–9]. Experimentally, Kerr [10] and Faraday [11] effects, and second-harmonic generation [12] process in TIs have been studied. Metallic surface states being Dirac like are responsible for strong nonlinear terahertz response of TI [13–15], and like to graphene TIs have great potential as an effective nonlinear optical material [16]. In particular, in the strong pump field limit, one can realize the regime where multiphoton effects are essential [17–20] and high-harmonics are generated. The experiment [21] with the generation of ninth harmonic in graphene opens new avenue towards high-harmonic generation in 2D nanostructures. Hence it is of interest to investigate multiphoton excitation and subsequent high harmonic generation process in TIs.

In the present work, we develop a nonlinear microscopic quantum theory of interaction of 2D metallic carriers confined on the surface of the 3D TI (e.g. Bi2Se3) with coherent electromagnetic radiation. We also take into account electron–electron Coulomb interaction with induced many-body effects. We consider nonlinear coherent interaction in the ultrafast excitation regime when relaxation processes due to electron–phonon coupling via longitudinal surface phonons are not relevant. We use the self-consistent Hartree–Fock approximation that leads to a closed set of integrodifferential equations for the interband polarization and carrier occupation distribution. The latter is solved numerically. Then we consider high-harmonic generation process for strong pump waves and show that one can achieve efficient generation of high harmonics in TIs.

The paper is organized as follows. In section 2 the set of equations for the interband polarization and carrier occupation distribution is formulated. In section 3, we consider multiphoton excitation of Fermi–Dirac sea and generation of harmonics in TI. Finally, conclusions are given in section 4.
2. Evolutionary equation for single-particle density matrix

Low-energy excitations of 2D metallic surface states of TI which are much smaller than the bulk gap energy (0.3 eV for Bi$_2$Se$_3$) can be described by the effective Hamiltonian

$$H_0 = \hbar V_F \begin{pmatrix} k_x \sigma_y - k_y \sigma_x & i \hbar V_F \begin{pmatrix} 0 & -k_x + i k_y \\ k_x + i k_y & 0 \end{pmatrix} \end{pmatrix},$$

where $V_F \approx c/450$ is the Fermi velocity for the topological insulator ($c$ is the light speed in vacuum), $i \hbar$ is the 2D electron momentum operator, $\sigma_x$ and $\sigma_y$ are the Pauli matrices. The eigenstates of the effective Hamiltonian (1) are

$$|\psi_{\eta,k}(r)\rangle = \frac{1}{\sqrt{A}} |\varphi_{\eta,k}\rangle e^{i kr},$$

where the spinors

$$|\varphi_{\eta,k}\rangle = \frac{1}{\sqrt{2}} (e^{-i\omega(k)} |\eta\rangle + e^{i\omega(k)} |\eta\rangle)$$

correspond to energies

$$\mathcal{E}_\eta (k) = \eta \hbar V_F k.$$

Here for conduction band $\eta = 1$ and for valence band $\eta = -1$, $A$ is the surface area, and

$$\theta(k) = \arctan (\frac{k_y}{k_x})$$

is the polar angle in the momentum space.

Although equation (3) is similar to the dispersion of quasiparticles in graphene, however, there are several physical differences. In graphene, Pauli matrices act in the pseudospin space corresponding to two sublattices. As a result, in graphene, we have twofold degeneracy of electronic states by two spin projections. In addition, we have a degeneracy upon two nonequivalent valleys. In the case of TIs, the Pauli matrices $\sigma_x$ and $\sigma_y$ act in the real electron spin space and the mean value of an electron spin in the 2D surface states of TI is

$$\frac{\hbar}{2} \langle \psi_{\eta,k} | \sigma | \psi_{\eta,k} \rangle = \frac{\eta \hbar}{2} (\hat{z} \times \hat{k}),$$

where $\hat{z}$ and $\hat{k}$ are unit vectors directed along the $k$ and the $z$-axis (normal to the surface), respectively. As is seen from equation (5), in TI the spin of electron lies in the surface plane and is perpendicular to its momentum. That is, the spin of a quasiparticle is locked to its translational momentum. At that, for conduction band, it is directed in the counterclockwise direction and inversely for the valence band. These properties are the key to realizing fundamentally new phenomena [1, 2] in condensed matter physics which are not possible with conventional Dirac fermions of the graphene variety.

Let a plane linearly polarized (along the $x$-axis) quasi-monochromatic electromagnetic radiation of carrier frequency $\omega_0$ and slowly varying envelope $E_\omega(t)$ interacts with the 3D TI. We assume perpendicular to the metallic surface incidence. In general, for TIs along with surface states, one should also consider the contribution of bulk states to the electromagnetic response of the material since the surface signatures can be easily suppressed by bulk contributions to transport. This is unavoidable when one excites TI with above-bandgap photon energy. For such photons optical excitation populates high-lying bulk states, which rapidly decay to lower energy states, which in turn fill the surface states with a steady supply of carriers [22]. In the current paper, we are interested in the nonlinear electromagnetic response of 2D metallic carriers confined on the surface of the 3D topological insulator. Thus, for photon energies, we assume $h\omega_0 \ll \mathcal{E}_F$, where $\mathcal{E}_F$ is the TI’s bulk gap. Besides, one should restrict wave intensity to avoid transition within the bulk bands, since a sufficiently high field will induce tunneling across the band gap whenever the field energy is large enough. The latter is governed by the Keldysh parameter $\Gamma_K$ that expresses the ratio between energy gap and period-averaged field-energy [23]:

$$\Gamma_K = \frac{\mathcal{E}_F}{eE_0 V_F}.$$

Thus, we will consider the interaction regime $\Gamma_K \gg 1$. Under these circumstances, one can neglect bulk excitations and the nonlinear electromagnetic response of TI will be conditioned by the 2D surface states. Thus, the light–TI interaction Hamiltonian in the length gauge will be:

$$\hat{H}_{int} = e \int \frac{d\vec{r}}{\mathcal{V}} \hat{\Psi}^\dagger (\vec{r}) \mathcal{E}(t) \hat{\Psi}(\vec{r}),$$

where $\hat{\Psi}(\vec{r})$ is the fermionic field operator and

$$\mathcal{E}(t) = \hat{\mathbf{E}} E_0(t) \cos \omega_0 t.$$

We will work in the second quantization formalism using the Fermi–Dirac field operator

$$\hat{\Psi}(\vec{r}) = \sum_{k\eta} \hat{e}_{\eta,k} |\psi_{\eta,k}(\vec{r})\rangle,$$

where $\hat{e}_{\eta,k}$ ($\hat{e}_{\eta,k}^\dagger$) is the annihilation (creation) operator for an electron with momentum $k$ and band $\eta = \pm 1$. The electrons interact through the long-range Coulomb forces and the Hamiltonian for electron–electron interactions can be written in terms of the field operators $\hat{\Psi}(\vec{r})$, as:

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

where $V_c(\vec{r}) = e^2 / (e |\vec{r}|)$ is the bare Coulomb potential, $e$ is the effective dielectric constant of the TI.

Taking into account expansion (9), the total Hamiltonian can be represented as follow:

$$\hat{H} = \sum_{\eta,k} \mathcal{E}_\eta (k) \hat{e}_{\eta,k}^\dagger \hat{e}_{\eta,k} + H_{Coul} + \frac{e \mathcal{E}(t)}{\mathcal{A}} \int \sum_{\eta'k'} d\vec{r} d\vec{r}' \hat{\Psi}^\dagger (\vec{r},\vec{r}') \hat{\Psi}^\dagger (\vec{r}') \hat{\Psi}(\vec{r}') \hat{\Psi}(\vec{r})$$

(10)

The Coulomb interaction reads:
\[
H_{\text{Coul}} = \frac{1}{2\mathcal{A}} \sum_{\eta_1, \eta_2, \eta_3, \eta_4} \sum_{\mathbf{q}, \mathbf{k}, \mathbf{k}'} V_{2D}(\mathbf{q}) F_{\eta_1, \eta_2, \eta_3, \eta_4}(\mathbf{q}, \mathbf{k}, \mathbf{k'}) \\
\times \hat{c}_{\eta_1, \mathbf{k}+\mathbf{q}, \mathbf{k}'-\mathbf{q}}^\dagger \hat{c}_{\eta_1, \mathbf{k}} \hat{c}_{\eta_1, \mathbf{k}'},
\]

(11)

where

\[
V_{2D}(\mathbf{q}) = \frac{2\pi e^2}{\varepsilon |\mathbf{q}|}
\]

(12)
is the 2D Coulomb potential in the momentum space and

\[
F_{\eta_1, \eta_2, \eta_3, \eta_4}(\mathbf{q}, \mathbf{k}, \mathbf{k}') \equiv \langle \varphi_{\eta_1, \mathbf{k}+\mathbf{q}} | \varphi_{\eta_2, \mathbf{k}'} \rangle \langle \varphi_{\eta_3, \mathbf{k}'} | \varphi_{\eta_4, \mathbf{k}} \rangle.
\]

(13)

In the light–TI interaction part of the Hamiltonian (10) there are terms responsible for intraband transitions (\(\eta = \eta'\)), as well as terms that describe interband transitions (\(\eta = -\eta'\)).

In order to develop a microscopic theory of the multiphoton TI interaction we will treat under Hartree approximation the Coulomb interaction leads to a renormalization (23) in the scope of Hartree–Fock approximation, which is valid for short time scales. The Hartree contribution \(\sim V_{2D}(\mathbf{q} = 0)\) is zero, which is physically related to the neutrality of charge of the total system. For the Fock part we will use decomposition:

\[
\hat{c}_{\eta_1, \mathbf{k}+\mathbf{q}, \mathbf{k}'}^\dagger = \hat{c}_{\eta_1, \mathbf{k}+\mathbf{q}, \mathbf{k}'}^\dagger - \left\{ \hat{c}_{\eta_1, \mathbf{k}}^\dagger \hat{c}_{\eta_1, \mathbf{k}} (\hat{c}_{\eta_1, \mathbf{k}}^\dagger \hat{c}_{\eta_1, \mathbf{k}}) \delta_{\mathbf{q}, \mathbf{k}'-\mathbf{k}} \right\}
\]

(16)

Taking into account definition (14), the second quantized Hamiltonian (10), and equations (11) and (16), from equation (15) one can obtain the following equations for \(N_v(\mathbf{k}, t)\), \(N_c(\mathbf{k}, t)\) and \(P(\mathbf{k}, t)\):

\[
\frac{\partial N_v(\mathbf{k}, t)}{\partial t} - \frac{e\mathbf{E}}{\hbar} \frac{\partial N_c(\mathbf{k}, t)}{\partial \mathbf{k}}
= -i (\Omega_R(\mathbf{k}, t) + \Omega_{PN}(\mathbf{k}, t)) P^*(\mathbf{k}, t) + \text{c.c.},
\]

(18)

\[
\frac{\partial P(\mathbf{k}, t)}{\partial t} - \frac{e\mathbf{E}}{\hbar} \frac{\partial P(\mathbf{k}, t)}{\partial \mathbf{k}}
= i \left[ \omega(\mathbf{k}) + \omega_{PN}(\mathbf{k}, t) \right] P(\mathbf{k}, t)
\]

(19)

\[
= -i (\Omega_R(\mathbf{k}, t) + \Omega_{PN}(\mathbf{k}, t)) (N_v(\mathbf{k}, t) - N_c(\mathbf{k}, t)),
\]

\[
= \frac{e\mathbf{E}}{2\hbar} \frac{\partial \theta(\mathbf{k})}{\partial \mathbf{k}}
\]

(20)

is the Rabi frequency and

\[
\omega_{PN}(\mathbf{k}, t) = -\frac{1}{2\hbar \mathcal{A}} \sum_{\mathbf{k} \neq \mathbf{k}'} V_{2D}(\mathbf{k} - \mathbf{k}') \sin[\theta(\mathbf{k}) - \theta(\mathbf{k}')]\]

\[
\times (N_v(\mathbf{k}', t) - N_c(\mathbf{k}', t)) - \frac{1}{\hbar \mathcal{A}} \sum_{\mathbf{k} \neq \mathbf{k}'} V_{2D}(\mathbf{k} - \mathbf{k}')
\]

\[
\times [P'(\mathbf{k}', t) + i \cos[\theta(\mathbf{k}) - \theta(\mathbf{k}')] P^*(\mathbf{k}', t)]
\]

(21)

The transition frequency is defined by

\[
\omega(\mathbf{k}) = 2V_{2D} k
\]

(22)

and

\[
\omega_{PN}(\mathbf{k}, t) = \frac{1}{\hbar \mathcal{A}} \sum_{\mathbf{k} \neq \mathbf{k}'} V_{2D}(\mathbf{k} - \mathbf{k}')
\]

\[
\times \cos[\theta(\mathbf{k}) - \theta(\mathbf{k}')] (N_v(\mathbf{k}', t) - N_c(\mathbf{k}', t))
\]

\[
+ \frac{2}{\hbar \mathcal{A}} \sum_{\mathbf{k} \neq \mathbf{k}'} V_{2D}(\mathbf{k} - \mathbf{k}') \sin[\theta(\mathbf{k}) - \theta(\mathbf{k}')] P^*(\mathbf{k}', t).
\]

(23)

In equations (21) and (23) \(P'(\mathbf{k}, t)\) and \(P'^*(\mathbf{k}, t)\) are the real and imaginary parts of \(P(\mathbf{k}, t)\), respectively. As is seen from equations (17)–(23) in the scope of Hartree–Fock approximation the Coulomb interaction leads to a renormalization of the light-matter coupling and effective Rabi frequency becomes \(\Omega_R(\mathbf{k}, t) + \Omega_{PN}(\mathbf{k}, t)\). The last term is due to the internal fields and depends on \(P\) and \(N_c\). Also, the transition energies become renormalized due to the Coulomb interaction and we have additional term \(\omega_{PN}(\mathbf{k}, t)\). The obtained equations are closed set of nonlinear integrodifferential equations.

As an initial state we assume undoped TI and for temperature we assume \(T < \hbar \omega_0\). Hence, for the initial distribution function we take the limit \(T = 0\):

\[
N_v(\mathbf{k}, 0) = 1, \quad N_c(\mathbf{k}, 0) = 0, \quad P(\mathbf{k}, 0) = 0.
\]

(24)

For the initial density matrix (24) (for any isotropic distribution) \(\Omega_{PN}(\mathbf{k}, 0) = 0\) and

\[
\omega_{PN}(\mathbf{k}, 0) = \frac{1}{\hbar \mathcal{A}} \sum_{\mathbf{k} \neq \mathbf{k}'} V_{2D}(\mathbf{k} - \mathbf{k}') \cos[\theta(\mathbf{k}) - \theta(\mathbf{k}')].
\]

(25)
The latter is the difference of self-energy corrections due to the electron–electron interactions [24], and can be written as

$$\omega_{PN}(k,0) = \frac{e^2}{2\pi\hbar c} \int \frac{dk'}{k'} \int_0^{2\pi} \frac{\cos \theta}{\sqrt{k'^2 + k'^2 - 2kk'\cos \theta}}. \tag{26}$$

We note that the integral of equation (26) has an ultraviolet high-momentum logarithmic divergence, which must be regularized through a high wave vector cutoff $k_c$. As is usual in condensed matter physics, there is a natural cutoff in the momentum arising from the lattice structure and, therefore, we have taken $k_c = 2\pi/a$, where $a = 0.41$ nm is the lattice spacing.

Thus the renormalized frequency can be represented as

$$\omega_{PN}(k,t) = \omega_{PN}(k,0) + \tilde{\omega}_{PN}(k,t),$$

where $\omega_{PN}(k,0)$ is given by the regularized expression (26) and

$$\tilde{\omega}_{PN}(k,t) = \frac{2}{\hbar A} \sum_{k' \neq k} V_{2D}(k - k') \sin [\theta(k) - \theta(k')] P''(k',t)$$

$$- \frac{2}{\hbar A} \sum_{k' \neq k} V_{2D}(k - k') \cos [\theta(k) - \theta(k')] N_F(k',t). \tag{27}$$

Because of finite excitation of Brillouin zone around Dirac point now $\tilde{\omega}_{PN}(k,t)$ and $\Omega_{PN}(k,t)$ are convergent. The domain of integration and the nonlinearity of the light-TI coupling is defined by dimensionless parameter:

$$\chi_0 = \frac{eE_0V_F}{\hbar\omega_0^2}, \tag{28}$$

which is the ratio of the amplitude of the momentum given by the wave field to characteristic excitation momentum $\hbar\omega_0/V_F$. In the limit $\chi_0 \ll 1$ the multiphoton effects are suppressed. The multiphoton effects become essential at $\chi_0 \sim 1$. In terms of the dimensionless interaction parameter $\chi_0$, the Keldysh condition (6) which restricts transitions within the bulk bands can be written:

$$\chi_0 \ll \frac{\xi_e}{\hbar\omega_0^2}. \tag{29}$$

Note that for THz photons the condition (29) can be fulfilled with large $\chi_0 \lesssim 10$.

The terms with partial derivative $\partial/\partial k$ in the left-hand side of equations (17)–(19) describe intraband transitions. In these equations, we can make a change of variables and transform the partial differential equation into an ordinary one. The new variables are $t$ and $k_0 = k - k_F(t)$, where

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

is the classical momentum given by the wave field.

Equations (17) and (18) yield the conservation law for the particle number:

$$N_e(k_0, t) + N_f(k_0, t) = 1. \tag{30}$$

With the conservation law (30) one can exclude equation for $N_e(k_0, t)$.

Note that here we consider a coherent interaction of TI 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 considered case, at the excitation energies $\mathcal{E} \ll \xi_e = 0.3$ eV, typical for $\text{Bi}_2\text{Se}_3$, the dominant mechanism for relaxation will be electron–phonon coupling via longitudinal surface phonons [25, 26]. In the temperature domain $2v_1\mathcal{E}/\mathcal{V} \ll T \ll \mathcal{E}$, where $v_1 = 2.9 \times 10^5$ cm s$^{-1}$ is the velocity of the longitudinal acoustic phonon, the relaxation time for the energy level $\mathcal{E}$ can be estimated as

$$\tau(\mathcal{E}) = \left( \frac{D^2\mathcal{E}T}{2\rho_m\hbar^3\mathcal{V}^3} \right)^{-1}. \tag{31}$$

Here $D = 22$ eV is the deformation potential, and $\rho_m = 7.7 \times 10^{-7}$ g cm$^{-2}$ is the mass density. For the THz photon energies $\mathcal{E} = 0.004$ eV, at temperatures $T = 0.1\mathcal{E}$, from equation (31) we obtain $\tau(\mathcal{E}) = 140$ ps. Thus, in this energy range, one can coherently manipulate with interband multiphoton transitions in TI on time scales $t \lesssim 100$ ps. For this reason, we consider short pump wave pulses. The wave amplitude is described by the envelope function $E_0(t) = E_0|f(t)|$:

$$f(t) = \begin{cases} \sin^2(\pi t/T_p), & 0 \le t \le T_p, \\ 0, & t < 0, t > T_p, \end{cases} \tag{32}$$

where $T_p$ characterizes the pulse duration and is chosen to be $T_p = 32T$, where $T$ is the wave period.

3. Multimode excitations and generation of harmonics

The integration of equations (17)–(19) is performed on a grid of $10000$–$20000$ $(k, \theta)$-points depending on the intensity of the pump wave. For the integration over polar angle, we use Gaussian quadrature with 60 points. For the quantity $k$ we take points homogeneously distributed between $k = 0$ and $k = \omega_0/V_F$, where parameter $\alpha$ depends on the intensity of the pump wave. The time integration is performed with the standard fourth-order Runge–Kutta algorithm.

The strength of Coulomb interaction is characterized by the dimensionless parameter $\alpha$, defined as a ratio of characteristic Coulomb interaction energy to kinetic energy. For the massless particles, $\alpha$ does not depend on the electron density and equals to $\alpha = e^2/\varepsilon\hbar V_F$. The static dielectric constant of crystals such as $\text{Bi}_2\text{Se}_3$ is estimated to be greater than 50. We assume that the effective dielectric constant is the average of that in the TI and in the vacuum, and take a value of $\varepsilon = 20$ [24]. Thus, for all calculations, we set $\alpha = 0.164$.

Photoexcitations of the Fermi–Dirac sea are presented in figures 1–3. As a reference frequency, we have taken $\nu_0 = \omega_0/2\pi = 1$ THz. In figure 1 a density plot of the particle distribution function $N_e(k, \nu)$ after the interaction is shown. The wave dimensionless amplitude is taken to be $\chi_0 = 0.2$. For this intensity only one and two-photon transitions take place.
In figure 2 the creation of a particle-hole pair in TI is shown for stronger wave intensity $\chi_0 = 0.5$. With increasing pump wave intensity and approaching to the domain $\chi_0 \sim 1$, the multiphoton excitations takes place and the Rabi oscillations appear corresponding to multiphoton transitions. At that, one should take into account the intensity effect of the pump wave (Stark shift due to free–free intraband transitions) and Coulomb effect on the quasienergy spectrum. Thus, the multiphoton probabilities of particle-hole pair production have maximal values for the resonant transitions

$$\varpi (k_0) = n\omega_0, \quad n = 1, 2, 3, ...$$

(33)

where

$$\varpi (k_0) = \frac{1}{T} \int_0^T (\omega (k_0 + k_E (t)) + \omega_{\text{PN}} (k_0 + k_E (t), t)) \, dt$$

(34)

is the mean value of the Coulomb and wave-fields dressed transition frequency. For the effective high-harmonic generation multiphoton transitions (33) should have reasonable probabilities, that is, the generalized Rabi frequency and interaction time should be large enough for full Rabi flopping. As is seen from figure 3 at $\chi_0 = 1$, the probabilities of multiphoton transitions are considerable up to photon numbers $s = 5$. With the multiphoton excitation the total electronic density

$$n_e (t) = \int N_e (k, t) \, \frac{d^3 k}{(2\pi)^3}$$

(35)

is also varied, approaching to a maximal value, and then falling. The latter is plotted in figure 4. Here $n_0 = \omega_0^2 / (2\pi v_F^2)$ and for a THz photon $n_0 = 1.43 \times 10^9 \text{ cm}^{-2}$.

At the multiphoton excitation, the particle-hole annihilation and the intraband transitions will cause intense coherent radiation of the harmonics of the applied wave field. Here we consider the possibility of generation of harmonics at the multiphoton excitation depending on the pump field intensity and frequency. For the radiation spectrum, one needs the mean value of the current density operator

$$\hat{j} = -e\langle \hat{\Psi} | \hat{v} | \hat{\Psi} \rangle,$$

(36)

where $\hat{v} = h^{-1} \partial H_0 (k) / \partial k$ is the velocity operator. Here we need only the surface current in the polarization direction of the pump wave: $\mathcal{J}_z (t) = \langle \hat{j}_z \rangle / A$. For the effective Hamiltonian (1) the x-component of the velocity operator reads

$$\hat{v}_x = V_F \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}. $$

(37)
With the help of equations (9), (14), (36) and (37), the surface current can be written as

\[ J_x(t) = -eV_F F(2\pi)^2 \int dk \left[ \cos \theta(k) (N_c(k,t) - N_v(k,t)) + 2 \sin \theta(k) P''(k,t) \right]. \]

(38)

Thus, having solutions of equations (17)–(19), then making an integration in equation (38) one can calculate the harmonic radiation spectrum with the help of a Fourier transform of the function \( J_x(t) \). We assume that the spectrum is measured at a fixed observation point in the backward propagation direction (and pump wavelength is much larger than TI film thickness). For the generated field we have

\[ E^{(s)}_x(t + z/c) = -\frac{4\pi}{c} J_x(t + z/c). \]

(39)

The emission strength of the \( s \)th harmonic will be characterized by the dimensionless parameter

\[ \chi_s = \frac{eE^{(s)}_x(s)}{h\omega_0} = \chi_0 E^{(s)}_x(s) E_0, \]

(40)

where

\[ E^{(s)}_x(s) = \frac{\omega_0}{2\pi} \int_0^{2\pi/\omega_0} E^{(s)}_x(t) e^{i\omega_0 t} dt \]

(41)

is the Fourier component of the generated field. In figure 5 the density plot of the radiation spectrum via logarithm of the normalized field strength \( \chi(\omega) \) (in arbitrary units) versus the pump wave intensity is illustrated. Note that with the fast Fourier transform algorithm instead of discrete functions \( \chi_s \) we calculate smooth functions \( \chi(\omega) \). From this figure, we clearly notice maximums at the odd harmonics and with the increase of the wave intensity the emission strengths of the high harmonics become feasible and for \( \chi_0 \) = 2 harmonics up to 21th are sizable.

We further examine emission rates of the 3rd and 5th harmonics for various pump wave frequencies versus intensity, which are shown in figures 6 and 7. For the considered intensities the perturbation theory is not applicable and in figures 6 and 7 we have a strong deviation from power law for the emission rates of harmonics. In particular, the rate of the 3rd harmonic scales almost linearly on the pump wave strength.
\[ \chi_3 \sim \chi_0. \] Whereas it should show the \[ \chi_0^3 \] dependence in the perturbative limit. Besides, these figures show that the emission rates almost independent of the pump wave frequency.

As was mentioned above for the available TIs the screening is very large and the Coulomb interaction parameter is small \( \alpha_c \). Thus, one can not expect new qualitative effects in harmonics generation process due to Coulomb interaction. However quantitative effects are essential, which is more evident in the perturbative by the wave-field regime. In figure 8 we plot normalized third harmonic emission rate \( \chi_3/\chi_0^3 \) in TI versus pump wave intensity for independent \( (\alpha_c = 0) \) and for interacting particles. As is seen from figure 8 in both cases we have the \( \chi_0^3 \) dependence of \( \chi_3 \), but for interacting particles the rate \( \chi_3 \) is larger.

Thus, calculations show that at the multiphoton excitation of 2D metallic surface states of TI the generation of high harmonics is possible which takes place for the wide range of pump wave frequencies. The average intensity of the wave expressed by \( \chi_0 \), can be estimated as

\[ I_{\chi_0} = \chi_0^2 \times 2 \times 10^2 \text{ W cm}^{-2} \left( \frac{\nu_0}{\text{THz}} \right)^4. \] (42)

The intensity \( I_{\chi_0} \) strongly depends on the pump wave frequency. In particular for THz pump waves, high-harmonics can be generated at the intensities \( I_{\chi_0} \approx 1 \sim 2 \times 10^2 \text{ W cm}^{-2} \).

4. Conclusion

We have presented a nonlinear microscopic theory of the TI interaction with coherent electromagnetic radiation in the ultrafast excitation regime. Electron–electron Coulomb interaction has been taken into account with the self-consistent Hartree–Fock approximation that leads to a closed set of integrodifferential equations for the interband polarization and carrier occupation distribution. The dynamics of the multiphoton excitation of 2D metallic surface states of TI depending on the wave intensity has been considered and analyzed on the basis of numerical simulations. It has been shown that by THz radiation of moderate intensities, one can control interband multiphoton transitions in 2D metallic surface states of TI on time scales \( t \lesssim 100 \text{ ps} \). Furthermore, we have shown that along with multiphoton transitions there is an intense radiation of high harmonics at the interband (particle-hole annihilation) and intraband transitions induced by a pump wave. The obtained results certify that the process of high-harmonic generation for THz photons can be already observed for intensities \( \sim 0.2 \text{ kW cm}^{-2} \) and temperatures \( T \ll \hbar \omega_0 \).

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References

[1] Hasan M Z and Kane C L 2010 Rev. Mod. Phys. 82 3045
[2] Qi X L and Zhang S C 2011 Rev. Mod. Phys. 83 1057
[3] Hsieh D et al 2009 Phys. Rev. Lett. 103 146401
[4] Zhang T et al 2009 Phys. Rev. Lett. 103 266803
[5] Tse W K and Macdonald A H 2010 Phys. Rev. Lett. 105 074501
[6] Tse W K and Macdonald A H 2010 Phys. Rev. B 82 161104
[7] Hosur P 2011 Phys. Rev. B 83 035309
[8] Iurov A, Gumbs G, Roslyak O and Huang D 2013 J. Phys.: Condens. Matter 25 135502
[9] Rahim K, Ullah A, Tahir M and Sabeek K 2017 J. Phys.: Condens. Matter 29 425304
[10] Jenkins G S, Sushkov A B, Schmadel D C, Butch N P, Syers P, Paglione J and Drew H D 2010 Phys. Rev. B 82 125120
[11] Sushkov A B, Jenkins G S, Schmadel D C, Butch N P, Paglione J and Drew H D 2010 Phys. Rev. B 82 125110
[12] Hsieh D, McIver J W, Torchinsky D H, Gardner D R, Lee Y S and Gedik N 2011 Phys. Rev. Lett. 106 057401
[13] Peres N M R and Santos J E 2013 J. Phys.: Condens. Matter 25 105801
[14] Autore M, Di Pietro P, Di Gaspare A, D’Apuzzo F, Giorgianni F, Brahlek M, Koirala N, Oh S and Lupi S 2017 J. Phys.: Condens. Matter 29 183002
[15] Giorgianni F et al 2016 Nat. Commun. 7 11421
[16] Mikhailov S A and Ziegler K 2008 J. Phys.: Condens. Matter 20 384204
[17] Avetissian H K, Avetissian A K, Mkrtchian G F and Sledrakian Kh V 2012 Phys. Rev. B 85 115443
[18] Avetissian H K, Avetissian A K, Mkrtchian G F and Sledrakian Kh V 2012 J. Nanophoton. 6 061702
[19] Avetissian H K, Mkrtchian G F, Batrakov K G, Maksimenko S A and Hoffmann A 2013 Phys. Rev. B 88 165411
[20] Avetissian H K, Ghazaryan A G, Mkrtchian G F and Sledrakian Kh V 2017 J. Nanophoton. 11 016004
[21] Yoshikawa N, Tamaya T and Tanaka K 2017 Science 356 736
[22] Sobota J A, Yang S, Analytis J G, Chen Y L, Fisher I R, Kirchmann P S and Shen Z X 2012 Phys. Rev. Lett. 108 117403
[23] Keldysh V 1964 Sov. Phys.—JETP 20 1307 (www.jetp.ac.ru/cgi-bin/dn/e_020_05_1307.pdf)
[24] Abergel D S L and Das Sarma S 2013 Phys. Rev. B 87 041407
[25] Das Sarma S and Li Q 2013 Phys. Rev. B 88 081404
[26] Viljas J K and Heikinheimo T T 2010 Phys. Rev. B 81 245404