Role of the pre-acceleration in high harmonic generation from solids

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We analyze high harmonic generation (HHG) from solids in laser fields with different ellipticities but a constant maximum amplitude. It is shown that the cutoff of HHG is strongly extended in a circularly polarized field. More importantly, the harmonic yield in the circularly polarized field is comparable to or even higher than that in the linearly polarized field. To understand the physics underlying these results, we develop a reciprocal-space-trajectory method, which explains HHG in solids by a trajectory-ensemble from different ionization times and different initial states in the reciprocal space. We show that the cutoff extension and abnormal ellipticity dependence are related to an additional pre-acceleration step prior to ionization, which has been overlooked in solids. By analyzing the trajectories, the important role of initial states remote from the valence band top is demonstrated and the resulting four-step mechanism of HHG in solids is confirmed.

High harmonic generation (HHG) from gas-phase atoms and molecules has opened up a new frontier in ultrafast science, where the generation of attosecond pulses and the measurement with attosecond temporal and Angstrom spatial resolutions become accessible. In the very recent years, HHG has also been observed in solids, which makes it possible to extend the successfull attosecond metrology to solid systems.

HHG in two-dimensional (2D) nontrivially polarized laser fields in gases has attracted a great deal of attention in the past years, because it provides deeper insight into the mechanism of HHG and also promises unprecedented applications such as generation of circularly polarized high harmonics and uncovering the ultrafast dynamics in atoms. Accordingly, the HHG in solids with the circularly or elliptically polarized laser fields has attracted close attention very recently. However, because solid systems have much more complex structure and dynamical processes than gases, HHG in solids exhibits many distinct and unexplored features. Although several numerical models, such as numerically solving the time-dependent Schrödinger equation (TDSE), the semiconductor Bloch equation (SBE), and time-dependent density functional theory (TDDFT) provide good descriptions of HHG, the underlying mechanisms are buried in the wave functions. Following the principle of HHG in gases, a generalized three-step model has also been proposed for solids. In brief, near the peak of the laser field, the electron tunnels vertically from the valence band (VB) to the conduction band (CB). Then, the electron is accelerated and it recombines some time later with emission of a high harmonic photon. This model provides a useful and intuitive explanation of HHG in solids, but fails to give a satisfactory quantitative description. A quantitative model in terms of electron trajectories is still not established. Therefore, especially for 2D laser fields, a clear understanding has not been reached so far.

In this Letter, we investigate the HHG in solids driven by circularly and elliptically polarized laser fields. Our results indicate that the HHG spectra show a clear cutoff extension in a circularly polarized field. More importantly, by fixing the amplitude of electric field, the harmonic yield in the circularly polarized field is comparable to or even higher than that in the linearly polarized field. To explain these phenomena, a four-step picture of HHG in solids with an additional pre-acceleration prior to ionization is proposed. In addition, we develop a reciprocal-space-trajectory (RST) method based on accelerated Bloch states, in which the HHG can be described in terms of the quantum path integral of the trajectory-ensemble. This method fully includes quantum interference effect and allows us to reproduce the HHG spectrum. Based on the analysis of electron trajectories, the cutoff extension and the abnormal ellipticity dependence are explained, and the important role of the pre-acceleration as an intuitive interpretation is revealed.

Figure shows the high harmonic spectra of ZnO driven by laser pulses with different ellipticities. We use the same band structure as in Ref. The orientation of the reciprocal lattice of ZnO is chosen so that $\hat{x}\parallel\Gamma - M$, $\hat{y}\parallel\Gamma - K$ and $\hat{z}\parallel\Gamma - A$. The harmonic spectra are obtained by numerically solving the SBE. The dephasing effect does not change the structure of the harmonic spectra as shown in Ref. and is neglected in this work. For computational convenience, we perform two-dimensional (2D) calculations (i.e., $k_z = 0$ in the reciprocal space), which can well reproduce the features of HHG in 3D simulations (see the Supplemental Material). The frequency of the driving field is $\omega = 0.014$ atomic units (a.u.). The laser pulse is polarized in the $x$-plane, and the ellipticity is varied by keeping the laser amplitude in the major axis constant ($F_{x0} = 0.004$ a.u.) and changing the amplitude in the minor axis ($F_{y0}$) from...
where $m$ \(\hat{m}\) is the instantaneous momentum of the electron at the initial time \(t_0\). \(\phi_{m,k}(r)\) is the Bloch state and \(E_{m,k}\) is the energy of the band \(m\) and crystal momentum \(k\). The states \(\phi_{m,k}(t)\) are the accelerated Bloch states [37].

Based on the accelerated Bloch states, the evolution of the electron wave packet can be described as [37]

$$|\Psi_{k_0}(t)\rangle = \sum_m \alpha_{m,k_0}(t)e^{-i\int_{t_0}^{t} d\tau E_{m}(k(\tau))}|\phi_{m,k_0}(t)\rangle ,$$  \(3\)

where \(\alpha_{m,k_0}(t)\) is the complex amplitude for the contribution in band \(m\). The corresponding wave vector \(k(t)\) of the Bloch state constitutes a trajectory in reciprocal space. In the framework of RST, one can obtain a trajectory perspective on HHG in solids as illustrated in Fig. 2. For simplicity, here we consider only two bands i.e., \(m = c, v\) for the CB and VB, which has been shown to work well in previous works [21 32 38]. An electron initially located at \(k_0\) in the VB undergoes the following steps, where “ionization” is to be understood as excitation to the CB:

(0) Before ionization, the electron is accelerated and oscillates in the VB. This is different from the picture in gases where the electron is localized in the ground state prior to tunnel ionization. It is also in contrast to the usual assumption in the previous three-step model for HHG in solids, where the electron is initially located at the top of the VB and vertically promoted into the CB at the same crystal momentum [21 22]. We refer to this acceleration process prior to ionization as pre-acceleration.

(1) The electron can be excited from the VB to the CB at time \(t'\) with certain probability. When the depletion of the initial state can be neglected, one obtains the complex amplitude of the ionization rate \(\chi_{k_0}(t') = e^{i\Sigma_{c,v}(t',t_0)}\Omega_{c,v}(t')e^{-i\Sigma_{c,v}(t',t_0)}\Omega_{c,v}(t) = \xi_{c,v}(k(t))\).

\(F(t)\) involves the transition matrix elements \(\xi_{c,v}(k) = \langle c,k|\nabla_v|v,k\rangle_{cell} = \frac{1}{V} \int d^3r \xi^*(r)e_{c,v}(r){\nabla}_k u_{c,v}(r),\) where the integration is performed over the volume of a unit cell \(V\) and \(u_{m,k}\) is the lattice periodic state. \(\xi_{c,v}(k)\) can be obtained from the ab initio calculations [34] and can be simplified with appropriate approximations following [22].

The relative phase includes two parts: one is the phase \(S_{c,v,k_0}(t',t_0) = \int_{t_0}^{t'} d\tau E_{c}(k(\tau))\) accumulated in the VB; the other one is the phase \(S_{c,v,k_0}(t',t_0) = \int_{t_0}^{t'} d\tau E_{c}(k(\tau))\) accumulated in the CB. Note that the trajectories of different ionization times are coherent with each other and the ionization is dominant at the peak of the electric field.

(2) After ionization, the electron is accelerated by the external field and the trajectory in reciprocal space \(k(t)\) satisfies the acceleration theorem. During the acceleration process, the instantaneous electron energy is \(E_c(k(t))\) and \(E_v(k(t))\) in the CB and VB, respectively. Their energy difference is denoted as \(\Delta E(k(t)) = E_c(k(t)) - E_v(k(t))\).
During the oscillation, two kinds of induced currents are produced and harmonic photons are generated. An intraband current is induced by the charge transfer inside the VB and the CB, respectively. An interband current is induced by the polarization between the VB and the CB. The induced intraband and interband currents are produced and harmonic photons are generated. (3) During the oscillation, two kinds of induced currents are produced and harmonic photons are generated. An intraband current is induced by the charge transfer inside the VB and the CB, respectively. An interband current is induced by the polarization between the VB and the CB. The induced intraband and interband currents are produced and harmonic photons are generated.

The HHG spectrum obtained by the channel $C_0$ contains the trajectories from different initial locations satisfying $k_0 = -A(t)$ ($t_1$ is the ionization time). Due to the pre-acceleration step, these electrons are driven to reach the top of the VB at the instant of ionization. Since the energy gap at the top of the VB is smallest, these electrons contribute predominantly to the total HHG. The farther the electrons are initially located away from the top of VB, the larger max $\Delta E[-A(t)]$ can be gained. Therefore, a larger cutoff energy can be obtained by channel $C_0$, and the first plateau in the total spectrum is well reproduced. In contrast, the trajectories of $L_0$ are overlapped with each other and only gain the maximum energy of $\max\{\Delta E[-A(t_1) + A(t)]\}$, which is smaller than $\max\{\Delta E[-A(t_1)]\}$.

Next, we investigate the much weaker harmonic signal in the second plateau (from the 40th to 60th harmonic orders). We plot the HHG spectrum contributed by the electron trajectories ionized far away from the top of VB (denoted as $C_1$). As shown in Fig. 3(a) and (b), the electron trajectories of $C_1$ channel cover much higher energy region than that of $C_0$ and contribute to the harmonics in the second plateau. Since the ionization of $C_1$ occurs with a large band gap, the HHG yield is much lower than that of channel $C_0$.

The above results reveal an important role of the pre-acceleration, allowing the electrons located far away from the top of VB to be accelerated to the top of VB polarized field. For the RST method, we apply a 2D calculation by considering trajectories with initial states in the plane $k_z = 0$. By analyzing the HHG contributed by different trajectories, one can find the link between the high harmonic generation and the intuitive trajectories. We consider the electron trajectories ionized at the top of the VB (including 1% area of the BZ), denoted as channel $C_0$. According to Eqs. (1) and (4), the high harmonic contributed by the channel $C_0$ can be obtained: $Y_{C_0} =\text{FFT}\{\sum_{k_0} A_{c,k_0}(t) \int dt' \alpha_{c,k_0}(t,t')^2 [j_{c,v,k_0}(t) - j_{c,v,k_0}(t')] + (\int dt' \alpha_{c,k_0}(t,t')j_{c,v,k_0}(t) + c.c.)\}$. As a comparison, following previous works [20, 21], we also consider the electron trajectories initially located at the top of the VB (including 1% area of the BZ), which we denote as channel $L_0$. The harmonic spectrum with the full trajectory ensemble is also presented in Fig. 3(a). One can see that, although the spectrum obtained by the channel $L_0$ shows a plateau, the cutoff energy ($25\omega$) is smaller than the first plateau cutoff of the total spectrum ($29\omega$). In contrast, the spectrum obtained by the channel $C_0$ can well reproduce the main structure and the cutoff position of the total spectrum. To understand these results, we show the representative trajectories of different channels in Fig. 3(b). “1” and “2” mark two trajectories ionizing at different times. The difference between the highest and lowest reachable energies of each trajectory, which represents $\max \{\Delta E[k(t)]\} = \max \{E_c[k(t)] - E_v[k(t)]\}$, indicates the cutoff energy of HHG contributed by the trajectory $k(t) = k_0 + A(t)$. As shown in Fig. 3(b), the channel $C_0$ contains the trajectories from different initial locations satisfying $k_0 = -A(t)$ ($t_1$ is the ionization time). Due to the pre-acceleration step, these electrons are driven to reach the top of the VB at the instant of ionization. Since the energy gap at the top of the VB is smallest, these electrons contribute predominantly to the total HHG. The farther the electrons are initially located away from the top of VB, the larger $\max \{\Delta E[-A(t)]\}$ can be gained. Therefore, a larger cutoff energy can be obtained by channel $C_0$, and the first plateau in the total spectrum is well reproduced. In contrast, the trajectories of $L_0$ are overlapped with each other and only gain the maximum energy of $\max \{\Delta E[-A(t_1) + A(t)]\}$, which is smaller than $\max \{\Delta E[-A(t_1)]\}$.
and efficiently contribute to HHG. Hence, a higher cutoff energy than previous models \cite{20, 21} is obtained. Note that the vector potential (i.e. $|A(t_i)|$) is near zero when the electric field reaches its peaks in a linearly polarized field, therefore the difference between $\max(\Delta E[-A(t_i) + A(t)])$ and $\max(\Delta E[A(t)])$ is not so large. This is a possible reason why the importance of pre-acceleration has not been noticed before.

The effect of pre-acceleration will be more obvious in elliptically or circularly polarized fields. In Fig. 3(a), we discuss the results in a circularly polarized field. As above, we consider the spectra obtained by the channels $C_0$ and $L_0$ and compare them with the total spectrum. One finds that the spectrum obtained from the channel $C_0$ reproduces the total spectrum well. In contrast, the spectrum obtained from the channel $L_0$ shows a much lower cutoff energy. To obtain an intuitive understanding of this result, we analyze the trajectories and the harmonic yield contributed by the trajectories of electrons located at different initial momenta $k_0$. As a quantitative measure of the yield from initial momentum $k_0$, we calculate

$$\bar{Y}_{k_0} = \frac{\max(\Delta E[k_0 + A(t)])}{\min(\Delta E[k_0 + A(t)])} \cdot \frac{Y_{k_0}(\omega) \omega}{\max(\Delta E[k_0 + A(t)]) - \min(\Delta E[k_0 + A(t)])},$$

(6)

where $Y_{k_0}(\omega)$ is the HHG yield contributed by electron trajectories initially located at $k_0$. The resulting $k_0$-dependent harmonic yield is shown in Fig. 3(b). The dominant distribution of the $k_0$-dependent harmonic yield shows a donut structure at about $|k_0| = A_0 = F_0/\omega$, where $A_0$ and $F_0$ are the amplitude of the vector potential and electric field. This shape is familiar from the photonization of gases in circularly polarized fields \cite{39}, where electrons are born at non-zero vector potential. The exact shape of the dominant distribution will be related to the band structure of the system. In this work, the band structure is approximately rotational symmetric around the $k_z$ axis like those in atoms, so nearly round structure is revealed. The solid cyan line in Fig. 3(b) marks one representative trajectory of channel $C_0$, which is initially located at $|k_0| = F_0/\omega$. The trajectory is also presented in the $k_z$-energy space, see Fig. 3(c). The green line marks the pre-acceleration in VB. As shown in Figs. 3(b) and (c), due to the pre-acceleration, electrons initially located far away from the top of the VB are driven to the top of the VB at the instant of ionization near the peak of the electric field, and therefore efficiently contribute to the HHG yield. In contrast, the electrons initially located close to the top of VB are driven away from the top of VB. These electrons involve ionization at large band gaps and therefore give minor contribution to the HHG yield. Moreover, as already discussed in Fig. 3, electron trajectories initially located farther away from the top of VB contribute to a higher energy region of HHG than the trajectories initially located close to the top of the VB. Consequently, a harmonic spectrum with both broad spectral range and high intensity is obtained. Note that, the channel $C_0$ contains trajectories oscillating cover the energy range $\Delta E(-A_x(t_i) + A_x(t), -A_y(t_i) + A_y(t), -A_z(t))$, where $t_i$ is the ionization time. This is sensitive to the ellipticity $\epsilon$ of the laser field. With the decrease of $\epsilon$, the energy range contributed by $C_0$ decreases while the channel $C_1$ still contributes to the high energy region with relatively low efficiency. As a result, the two-plateau structure emerges when the driving field changes from circular to linear polarization.

In conclusion, we have investigated HHG from ZnO driven by laser fields with various ellipticities. We find that the harmonic cutoff is substantially extended in the circularly polarized field and the harmonic yield is comparable to or higher than that in the linearly polarized field. To reveal the underlying physics, we have developed a RST method, which builds a bridge between the HHG and the microscopic dynamics of electrons. It is shown that, different from atoms, HHG in solids is better understood in terms of a four-step picture with an additional pre-acceleration step prior to ionization. This pre-acceleration effect selects electron trajectories starting from locations other than the top of the VB to predominantly contribute to HHG and result in a broader spectral range of HHG. This effect is more obvious in the circularly polarized field, where the initial locations of the dominant electron trajectories exhibit a donut distribution and the cutoff is dramatically extended. The RST model will be a useful tool to understand the HHG in periodical systems, and it can be extended to more complicated forms of driving fields. This will shed light on
FIG. 4. (a) Harmonic spectra in a circularly polarized field obtained from different channels. For clarity, the spectra are shifted by constant factors $10^{-3}$ for $C_0$, and $10^{-10}$ for $L_0$. (b) Normalized $k_y$-dependent harmonic yield contributed by the trajectories of electrons initially located at different positions in reciprocal space. The green solid line and purple dashed line mark representative trajectories of $C_0$ and $L_0$, respectively. (c) The representative trajectory of the channel $C_0$ in $k_x - k_y$ - energy space.

the manipulation and optimization of the HHG in solids with well-designed driving fields.

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