

Impact of incident photon energy and pulse intensity on circularly polarized high harmonic generation in monolayer graphene

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Circularly polarized high harmonics provide important light sources for ultrafast detection of chiral molecules and solid materials. Although the circularly polarized high harmonic generation (HHG) has been reported in many solid materials, the comprehensive study about its dependence on the driving pulse parameters is still rare. Here, by using time-dependent density functional theory calculations, we have systematically simulated the generation of high harmonics from monolayer graphene driven by circularly polarized femtosecond pulses with different photon energies and peak intensities. Our calculation results show that when the driving pulses with high photon energy or low peak intensity are adopted, circularly polarized high harmonics can be generated, in which the harmonic radiation is dominated by intraband process and the harmonic order/polarization fulfill the selection rule. However, under driving pulses with low photon energy and high peak intensity, some of low-order harmonics exhibit noise spectra and their corresponding polarizations deviate significantly from the selection rule, which is because under this condition the number of incident photons is increased and the interband transitions between the empty and occupied bands with large density of states are enhanced. Our study clarifies the conditions for the generation of circularly polarized high harmonics in graphene and provides an important reference for selecting driving pulse parameters with the knowledge of electronic band structure. We also discover the correspondence between the HHG noise spectrum and the polarization, which can serve as a simple experimental criterion on whether the interband transitions become prominent in solid materials.

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I. INTRODUCTION

Circularly polarized high harmonic generation (HHG) has attracted great attention due to its applications in detecting and controlling the dynamics of chiral molecules [1,2] and solid materials [3–5]. However, circularly polarized HHG cannot be achieved in gas medium by using a single circularly polarized driving pulse. This is because the ionizing electrons will drift laterally in a circularly polarized driving field, preventing them from recollisions with the ions and emitting high harmonics. Although the scheme of two counter-rotating beams of circularly polarized laser pulses to drive a gas medium has been proposed and demonstrated to generate circularly polarized high harmonics [6–8], the low conversion efficiency as well as the complex and bulky facilities have limited its further development.

Unlike gaseous systems, the periodic and dense arrangement of atoms in solid-state systems makes them possible to generate circularly polarized high harmonics using single-color pulse. Since 2017, experiments on HHG using circularly polarized laser pulses in GaSe [9], Si [10], and quartz [11] crystals confirmed the existence of circularly polarized harmonic emission and the fulfillment of selection rule by measuring the helicity of the harmonics [12,13]. Up to now, researchers have devoted much effort to generating circularly polarized high harmonics in more solid-state systems. Graphene, as the first discovered two-dimensional (2D) material, has unique Dirac cone band structure and zero bandgap, making it an ideal system for realizing compact and advanced light sources on a desktop scale. However, it is unclear whether graphene can generate circularly polarized high harmonics under a single driving pulse. When a circularly polarized driving pulse with photon energy of 0.26 eV and peak intensity of 0.8 TW/cm² was used [14], no circularly polarized HHG has been observed, which is supported by the following theoretical studies [14–18]. In comparison, another theoretical work [19] has indicated that for the driving pulse with different parameters, such as photon energy of 1.55 eV and peak intensity of 3 TW/cm², the circularly polarized high harmonics can be generated. Thus, whether the circularly polarized high harmonics can be generated in

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monolayer graphene seems to be dependent on the parameters of driving pulses. Nevertheless, the relationship between the circularly polarized HHG and the driving pulse parameters is still not clear, and the underlying physical mechanism needs further elucidation.

In this paper, we have simulated the HHG of monolayer graphene driven by circularly polarized femtosecond pulses with different photon energies and peak intensities by means of time-dependent density functional theory (TDDFT) calculations. Our results show that monolayer graphene can generate circularly polarized high harmonics that comply with the selection rule under the driving pulse with high photon energy or low peak intensity. However, when the driving pulse with low photon energy and high peak intensity is adopted, some of low-order harmonics show noise spectra and the corresponding polarizations deviate from the selection rule. We then explore the origin of this phenomenon with in-depth analysis and provide suggestion for obtaining circularly polarized HHG with clear harmonic peaks.

II. COMPUTATIONAL METHOD

The calculations were performed based on the TDDFT [20] as implemented in the Octopus package [21–24]. The time-dependent Kohn-Sham equation under an external electromagnetic field is given by [25]

$$\left[-\frac{\hbar^2}{2m} \nabla^2 + V_{\text{eff}} + i \frac{e\hbar \mathbf{A} \cdot \nabla}{m} \right] \psi_{k,n} = i\hbar \frac{\partial}{\partial t} \psi_{k,n}, \quad (1)$$

where $V_{\text{eff}}(r)$ is the effective potential, $\mathbf{A}$ is the vector potential of laser field that can be expressed as [26]

$$\mathbf{A}(t) = \sqrt{\frac{2I}{\epsilon_0 \omega_0^2}} f(t) \left[\frac{1}{\sqrt{1+\epsilon^2}} \cos(\omega_0 t + \phi) \mathbf{e}_x + \frac{\epsilon}{\sqrt{1+\epsilon^2}} \sin(\omega_0 t + \phi) \mathbf{e}_y \right], \quad (2)$$

where I is the peak intensity, ϵ_0 is the vacuum permittivity, ω_0 is the central angular frequency, ϵ is the ellipticity, ϕ is the carrier envelope phase, $\mathbf{e}_x$ and $\mathbf{e}_y$ are unit vectors parallel to the two perpendicular in-plane directions of graphene, and $f(t)$ is the pulse envelope, which is described using a sin-squared function [19]

$$f(t) = \sin^2 \left(\frac{\pi t}{2\tau} \right). \quad (3)$$

Here τ is the pulse duration, the full width at half maximum (FWHM) on the time domain.

When the wave functions on all quasiwave vectors $\mathbf{k}$ and each electronic band n are calculated, the time-dependent current density can be obtained [25,27],

$$\mathbf{J}(t) = \int_{\text{BZ}} \sum_n \langle \psi_{k,n}(\mathbf{r}, t) | -\frac{e}{m} \mathbf{p} | \psi_{k,n}(\mathbf{r}, t) \rangle d\mathbf{k}, \quad (4)$$

where $\mathbf{p}$ is the gauge momentum operator. The high harmonic spectrum can then be obtained from the dipole approximation with the Fourier transform of the time-dependent current

density [25,27],

$$\text{HHG}(\omega) \propto \omega^2 |\mathcal{FT}(\mathbf{J}(t))|, \quad (5)$$

where $\mathcal{FT}$ denotes the Fourier transform. From the current density $\mathbf{J}(\omega)$ in the frequency domain, the current can be subjected to wavelet transform to obtain its time-domain waveform [26],

$$\mathbf{J}_{n\text{-th}}(t) = 2 \int_{(n-0.5)\omega_0}^{(n+0.5)\omega_0} \mathbf{J}(\omega) e^{i\omega t} d\omega, \quad (6)$$

where the factor 2 is a correction for negative frequencies. The radiation electric field is proportional to the derivative of the current [25–27],

$$\mathbf{E}_{n\text{-th}}(t) \propto \frac{\partial}{\partial t} \mathbf{J}_{n\text{-th}}(t). \quad (7)$$

For high harmonics, the absolute value of ellipticity ϵ at time t is calculated according to [26]

$$\epsilon(t) = \sqrt{\frac{\min[I_x(t), I_y(t)]}{\max[I_x(t), I_y(t)]}}, \quad (8)$$

and

$$I_i(t) = \frac{\int_{t-T/2}^{t+T/2} |E_i(t')|^2 dt'}{T} \quad (i = x, y), \quad (9)$$

where T is the time period of corresponding harmonic. The positive and negative signs of ellipticity ϵ are determined based on whether the electric field of harmonic at time t rotates clockwise (right-handed, +) or counterclockwise (left-handed, −) relative to that at time $t - \Delta t$, where Δt is the time step.

In graphene, the carbon atoms arrange in a 2D hexagonal lattice, for which the in-plane lattice constant of 4.65 bohr was adopted. The zigzag (ZZ) direction was defined as the x direction while the armchair (AC) direction was defined as the y direction. A vacuum space of 12.0 bohr plus an adsorbing layer of 3.0 bohr on each side of monolayer graphene was adopted, which accounts to a total computational length of 30.0 bohr along the z direction of the cell. The Hartwigsen-Goedecker-Hutter norm-conserving pseudopotentials and the local density approximation of exchange-correlation functional were used to describe the interactions between electrons and nuclei. A $60 \times 60 \times 1$ Monkhorst-Pack $\mathbf{k}$ -point mesh was utilized for the Brillouin zone sampling. The real-space spacing was set to 0.4 bohr and the time step was chosen to be 0.1 a.u. (2.4 as). The circularly polarized laser pulse with peak intensities I of 0.1 to 1 TW/cm² and photon energies $\hbar\omega$ of 0.6 to 1.2 eV, corresponding to wavelengths of 1033 to 2066 nm, were studied. The FWHM pulse duration τ of 35 fs and the carrier envelope phase of $\phi = \pi/2$ were adopted. We have examined our results with a denser $90 \times 90 \times 1$ $\mathbf{k}$ -point mesh, the Perdew-Burke-Ernzerhof functional, longer pulse durations (70 fs, 105 fs, 175 fs), and different carrier-envelope phases ($\phi = 0, \pi/2$), which have little influence on the HHG spectra (Figs. S1 to S4 in the Supplemental Material [28]) and thus validate the robustness of our conclusions.

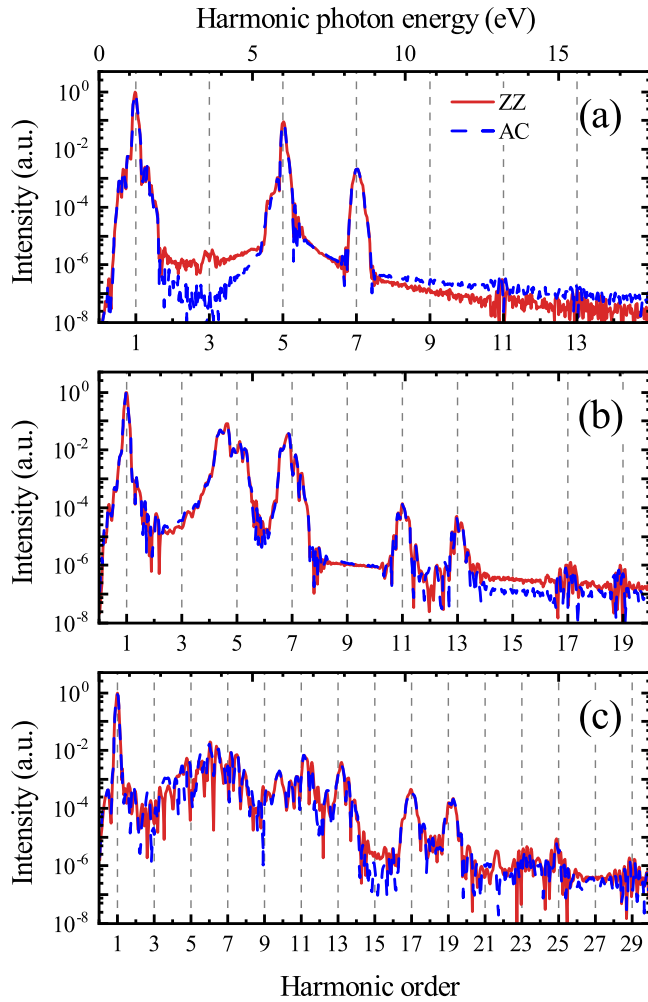


FIG. 1. Harmonic spectra of monolayer graphene driven by right circularly polarized laser pulses with peak intensity of 1 TW/cm^2 and photon energies of (a) 1.2 eV, (b) 0.9 eV, and (c) 0.6 eV, respectively. The red solid line and blue dashed line denote the respective harmonics along the zigzag (ZZ) and armchair (AC) directions.

III. RESULTS AND ANALYSES

We first investigated the high harmonic spectra of monolayer graphene driven by right circularly polarized pulses with peak intensity of 1 TW/cm^2 and photon energies of 1.2, 0.9, and 0.6 eV, respectively, as shown in Fig. 1. The circularly polarized pulse has an arbitrary rotational symmetry, and monolayer graphene has a sixfold rotational symmetry. Based on the symmetry consideration, only the harmonics with orders $6n \pm 1$ ($n = 1, 2, 3, \dots$) are allowed (selection rule) [12,13]. When the incident photon energy is relatively large, the generated harmonic orders are all in accordance with this selection rule. Specifically, when the incident photon energy is 1.2 eV, clear 5th- and 7th-order harmonic spectra can be observed [Fig. 1(a)]. As the incident photon energy decreases to 0.9 eV, the 5th-, 7th-, 11th-, and 13th-order harmonics display distinct spectra [Fig. 1(b)], albeit slight red-shift and broadening in the 5th-order harmonic peak. Once the incident photon energy reduces to 0.6 eV, the 5th- and 7th-order harmonics are no longer isolated peaks, while a few spectral

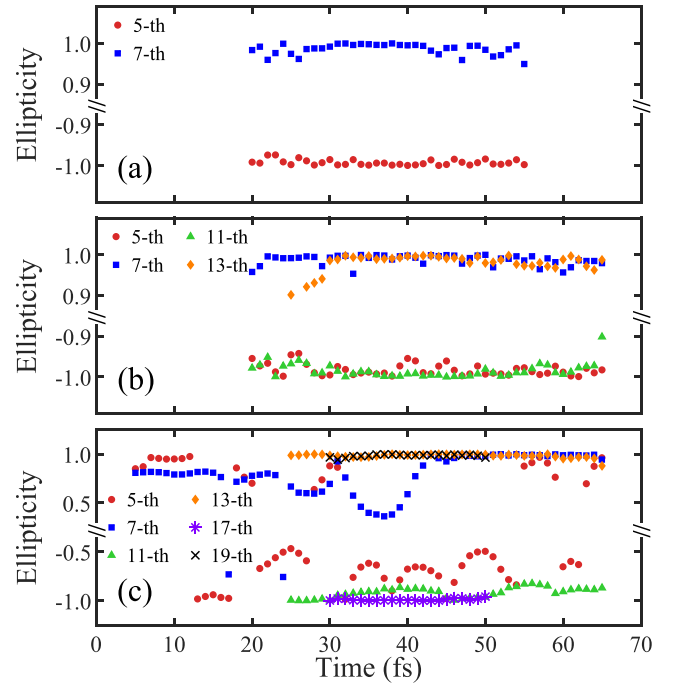


FIG. 2. Temporal evolution of ellipticity for each harmonic at driving pulse photon energies of (a) 1.2 eV, (b) 0.9 eV, and (c) 0.6 eV, respectively. The right-handed polarization is defined as positive and the left-handed polarization as negative.

peaks between 5th and 7th harmonics can be observed. These spectral peaks, which do not adhere the selection rule and exhibit intensities comparable to those of the 5th and 7th harmonics, are termed as noisy spectra. In comparison, the 11th- and 13th-order harmonics display distinct peaks with weak noise, while only the 17th- and 19th-order harmonics exist with clear peaks. From these results, it can be seen that as the incident photon energy decreases, irregular noisy spectra are observed near the low-order harmonics, while clean spectral peaks appear for all cut-off harmonics. We then studied the polarization of each harmonic generated by right circularly polarized pulses with different photon energies. According to the selection rule, the $(6n + 1)$ th harmonic rotates in the same direction as the driving pulse, while the $(6n - 1)$ th harmonic rotates in the opposite direction to the driving pulse [9,12], which is determined by the spatial rotational symmetry and the conservation of angular momentum. As shown in Fig. 2(a), for the incident photon energy of 1.2 eV, the 5th- and 7th-order harmonics are left and right circularly polarized, respectively, which are in excellent agreement with the selection rule. When the incident photon energy decreases to 0.9 eV [Fig. 2(b)], the polarizations of most harmonics are basically in accordance with the selection rule, but the 5th-order harmonic shows an obvious deviation from the expected left circular polarization. As the incident photon energy reduces to 0.6 eV [Fig. 2(c)], the deviations of polarizations for the low-order harmonics become more prominent. To be specific, the polarization of the 5th-order harmonic shows irregular evolution over time, mainly composed of right and left elliptical polarizations. For the 7th-order harmonic, it predominantly shows right elliptical polarization before 50 fs

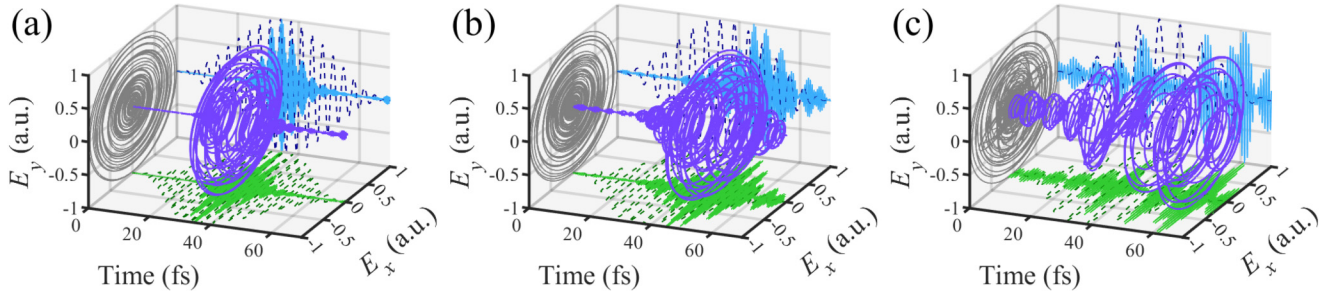


FIG. 3. Temporal evolution of the electric field vectors (purple lines) for the 7th-order harmonics of monolayer graphene driven by right circularly polarized laser pulses with photon energies of (a) 1.2 eV, (b) 0.9 eV, and (c) 0.6 eV, respectively. The green (blue) solid and dashed lines denote the E_x (E_y) components of the harmonic electric field and the driving electric field, respectively. The gray line represents the projection of the harmonic electric field vector onto the E_x - E_y plane over all times.

and subsequently turns to right circular polarization. As the harmonic order increases, the deviation from expected polarization becomes small, and all of them obey the selection rule. We thus find a correspondence between the polarization and the spectral peak for high harmonics: For the harmonics with clean spectral peaks (Fig. 1), their polarizations mainly retain circular and abide by the selection rule (Fig. 2), while the harmonics with noisy spectral peaks deviate from the circular polarization.

In order to explore the mechanism of circularly polarized HHG in monolayer graphene, we plot in Fig. 3 the temporal evolutions of the electric field vector (purple line) and its E_x (green line) and E_y (blue line) components for the 7th-order harmonics. For the driving pulses with photon energies of 1.2 eV [Fig. 3(a)] and 0.9 eV [Fig. 3(b)], the projections of the harmonic electric field vectors onto the E_x - E_y plane over all times (gray lines) clearly demonstrate that the 7th-order harmonics mainly keep the circular polarization, which is consistent with the ellipticity results in Fig. 2. We also note that the 7th-order harmonics in these two photon energies are mainly generated in the peaks and trailing edge parts of the driving pulses (dashed lines). Another prominent feature is that the envelopes of the harmonic components have the same periods as those of the driving pulses, which is a typical characteristic of the harmonic radiation induced by the intraband Bloch oscillation [19,29]. This characteristic of the harmonic is also in good accordance with the definition of intraband current in Otake's work [30], whose phase only depends on that of driving vector potential. In contrast, when the driving photon energy reduces to 0.6 eV [Fig. 3(c)], the projection of the harmonic electric field (gray line) shows obvious deviation from the circular polarization. Moreover, the period of the harmonic envelope does not coincide with that of the driving pulse field, suggesting that the HHG is contributed more by the interband transitions. Here the interband transition refers to the electron-hole recombination. Since the interband current not only depends on the phase of vector potential but also relates to the phases of eigenstates and projected coefficients of time-dependent wave function to the eigenstates [30], it leads to the asynchronous bursts of the harmonic with the driving field. From the results of the 7th-order harmonics at different incident photon energies, both the noisy spectral peaks of the harmonics [Fig. 1(c)] and the deviation from circular polarization [Figs. 2(c) and 3(c)] may be related to the interband transition process.

To further study the effect of interband transition on the harmonic generation, we plot the electronic band structure of monolayer graphene as in Fig. 4(a). At the K point of the Brillouin zone, the bands of the Dirac cone exhibit perfect linear dispersions. Around the M point, the bands at +1.7 eV above Fermi level (E_F) and -2.4 eV below E_F show saddle-point features. The corresponding electronic density of states (DOS) possesses high peaks at these two energies [black line in Fig. 4(b)], indicating that electrons have more probabilities to occupy these two bands. The joint density of states has a peak at around 4.1 eV in the low-energy region (see

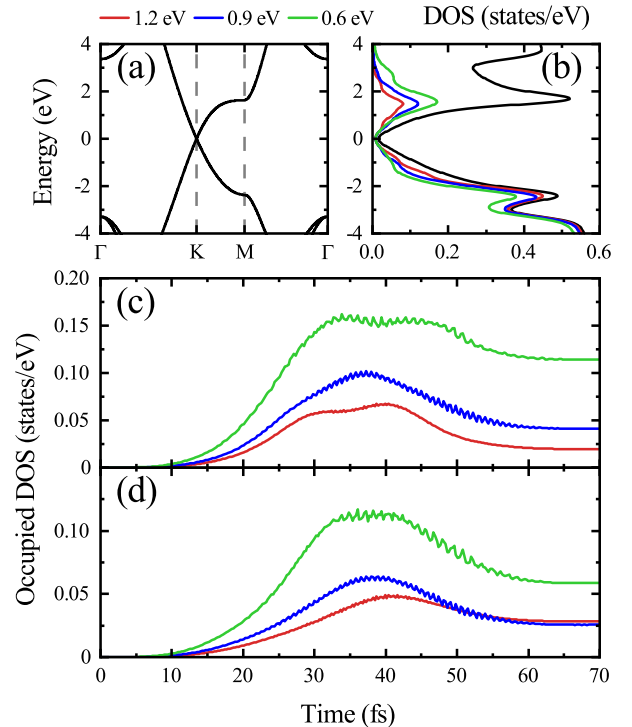


FIG. 4. (a) Electronic band structure of graphene. (b) Electronic DOS for the ground state of graphene (black line). The red, blue, and green lines correspond to the electron occupied DOSs at the pulse peak time ($t = 35$ fs) with the respective incident photon energies of 1.2, 0.9, and 0.6 eV. Temporal evolutions of (c) electron occupied DOSs at $E_F + 1.7$ eV and (d) hole occupied DOSs at $E_F - 2.4$ eV for the driving pulses with peak intensity of 1 TW/cm² and photon energies of 1.2 eV (red), 0.9 eV (blue), and 0.6 eV (green), respectively.

Fig. S5 in the Supplemental Material [28]), which equals to the energy difference between the aforementioned DOS peaks at $E_F + 1.7$ eV and $E_F - 2.4$ eV in Fig. 4(b), suggesting that possible interband transitions may happen between these two levels [27]. We use the definition of the electron occupied DOS under laser excitation, which is calculated from the projection of time-evolved wave functions to the initial eigenstates without driving field, to reflect the real-time information of electron fillings at different energy levels. By calculating the electron occupied DOS at the pulse peak time ($t = 35$ fs) under different incident photon energies, we can see that the largest changes in the occupied DOS appear near the peaks at $E_F + 1.7$ eV and $E_F - 2.4$ eV [colored lines in Fig. 4(b)]. For incident photon energies of 1.2 and 0.9 eV, the energies of the excited electrons and holes are concentrated in the range of -2.5 to $+3.0$ eV, suggesting that even if interband transitions occur, only harmonics with energy less than 5.5 eV can be generated, which is smaller than the photon energy of the 5th-order harmonic (6.0 eV) for the 1.2 eV driving pulse and that of the 7th-order harmonic (6.3 eV) for the 0.9 eV driving pulse. Therefore, harmonics of these and higher orders should be mainly contributed by the intraband oscillations, which present clear isolated peaks in the frequency domain (Fig. 1) and produce circular polarization in the time domain that complies with the selection rule (Figs. 2 and 3). In comparison, for the 5th-order harmonic (4.5 eV) of the 0.9-eV driving pulse, its energy is less than that of the interband transition range (5.5 eV). This harmonic may thus be partly attributed to the interband transition, leading to the slight distortion in spectral peak [Fig. 1(b)] and the deviation from circular polarization [Fig. 2(b)]. For the incident photon energy of 0.6 eV, the interband transition energy completely covers the 5th- and 7th-order harmonics [green line in Fig. 4(b)], allowing more interband transitions. As a result, the spectral peaks of the corresponding harmonics become unclear [Fig. 1(c)] and the polarizations deviate from the expected forms [Figs. 2(c) and 3(c)]. Over all, it can be speculated that the interband transitions are not favorable to the circularly polarized HHG in graphene.

For the two bands at $E_F + 1.7$ eV and $E_F - 2.4$ eV, we also calculated the time evolutions of the electron and hole occupied DOSs for different incident photon energies, as shown in Figs. 4(c) and 4(d). In contrast to Fig. 4(c) that adopts the raw data of color lines at $E_F + 1.7$ eV in Fig. 4(b), Fig. 4(d) utilizes the differences between the black line and the color lines at $E_F - 2.4$ eV in Fig. 4(b), i.e., translating the information of electron fillings to that of hole occupations. It can be seen that both the occupied DOSs of the electrons at $E_F + 1.7$ eV and holes at $E_F - 2.4$ eV increase with the decreasing incident photon energy. This is because, under the same peak intensity of driving pulses, the number of photons increases as the incident photon energy decreases, which can excite more electrons above the Fermi level and leave more holes below the Fermi level. The increase of electron excitation is more favorable for interband transitions, as evidenced by the small oscillations in the time evolution curves at low incident photon energies (blue and green lines). Through Fourier transformation, we find that the frequency of these small oscillations is six times the frequency of the driving pulse field. In order to better understand the source of these

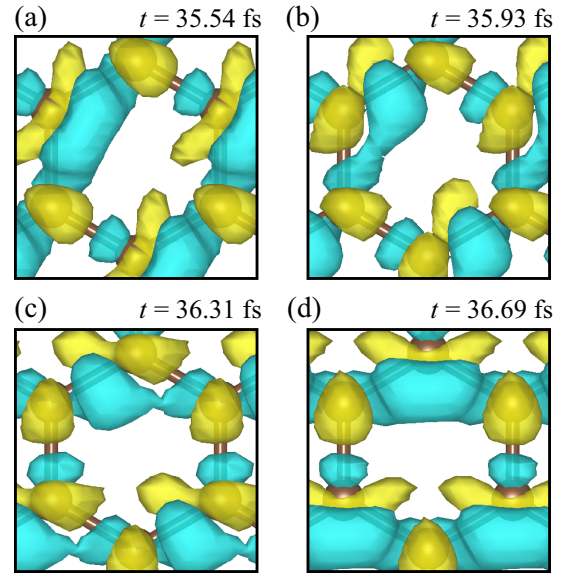


FIG. 5. Temporal evolution of differential charge densities within 1/6 period of the driving pulse field with photon energy of 0.6 eV. The yellow and blue isosurfaces denote the electron accumulation and depletion regions, respectively. The brown balls represent C atoms.

oscillations, we take the incident photon energy of 0.6 eV (corresponding to a driving period of 6.89 fs) as an example and plot the time evolutions of differential charge densities with respect to the ground state in a period of 1/6 of the driving pulse field (as shown in Fig. 5). It can be clearly seen that the electrons in real space rotate under the action of the driving pulse field, and within 1/6 period of the driving pulse field, the electrons and holes rotate $\pi/3$ around the hexagonal ring of graphene. Thus, within one driving pulse field period, the electrons will rotate 2π around the hexagonal ring of graphene and can recollide with C atoms six times. As a result, the oscillations of the electron occupied DOS exhibit sixfold frequency characteristics. The process of collision between rotating electrons and C nuclei is similar to that between free electrons and their parent ions in gas medium, which corresponds to the interband transition process. In Figs. 4(c) and 4(d), the amplitude of oscillations with the incident photon energy of 0.6 eV is greater than those of 0.9 and 1.2 eV, which means that there are more interband transitions under the former driving pulse.

Based on above results, for the same peak intensities, the driving pulse with low photon energy can excite more electrons above the Fermi level and cause more interband transitions, thereby affecting the spectra and polarizations of harmonics. Accordingly, if the peak intensity of driving pulse is reduced and the number of excited electrons is decreased, then there may be less interband transitions. We then calculated the HHG spectra and the ellipticity of each harmonic under the driving pulses with photon energy of 0.6 eV and reduced peak intensities of 0.5 TW/cm² and 0.1 TW/cm² (Fig. 6). Compared with the harmonic spectrum at a peak intensity of 1 TW/cm² [Fig. 1(c)], the spectral peak of the 7th-order harmonic is clearer at a peak intensity of 0.5 TW/cm² and the circular polarization is well maintained [Fig. 6(a)].

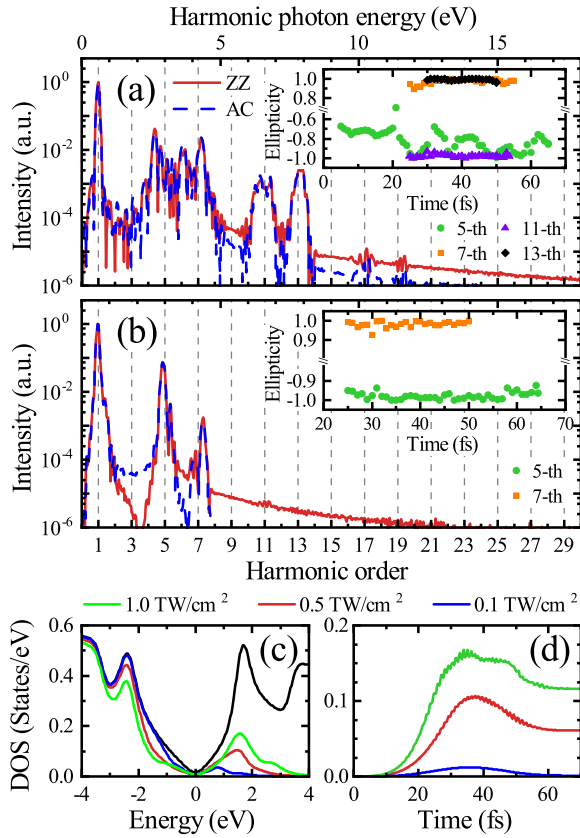


FIG. 6. Harmonic spectra of monolayer graphene driven by circularly polarized laser pulses with photon energy of 0.6 eV and peak intensities of (a) 0.5 TW/cm² and (b) 0.1 TW/cm², respectively. The red solid and blue dashed lines denote the harmonics along the zigzag and armchair directions of graphene, respectively. Insets show the temporal evolution of the ellipticity of each harmonic. (c) DOS for the ground state of monolayer graphene (black line). The green, red, and blue lines correspond to the electron occupied DOSs at the pulse peak moment when the peak intensities are 1, 0.5, and 0.1 TW/cm², respectively. (d) Temporal evolution of electron occupied DOSs of graphene at $E_F + 1.5$ eV for the incident photon energy of 0.6 eV and peak intensities of 1 TW/cm² (green), 0.5 TW/cm² (red), and 0.1 TW/cm² (blue), respectively.

Nevertheless, the 5th-order harmonic still has the noisy spectrum and is elliptically polarized most of the time [green dots in the inset of Fig. 6(a)]. When the peak intensity is further reduced to 0.1 TW/cm², both the 5th and 7th harmonic spectra are very clear and their circular polarizations are well maintained over time [Fig. 6(b)]. These phenomena originate from the reduced interband transitions under low peak intensities, as evidenced by decreased energy ranges and numbers of excited electrons and holes [Fig. 6(c)] as well as the subdued oscillations in the time evolution curves of occupied electron DOSs [Fig. 6(d)]. The usage of different pulse durations (35 fs, 70 fs, 105 fs, 175 fs) and different carrier envelope phases ($0, \pi/2$) do not change this conclusion (see Figs. S3 and S4 in the Supplemental Material [28]), indicating that the peak intensity, instead of the pulse duration or carrier envelope phase, is more critical to the spectral clarity and polarization purity of low-order harmonics. Similar

computations (see Fig. S6 in the Supplemental Material [28]) can also explain the observations in previous experiment [14]. These results confirm that the excessive proportion of interband transitions will affect the clarity of spectral peak and the circular polarization of harmonic.

IV. DISCUSSION AND CONCLUSION

We aim to find the driving pulse parameters that can generate clear circularly polarized high harmonics. When the driving pulses with high photon energies are used (Figs. 1 and 2), the circularly polarized harmonic radiations show clean spectra peaks in all harmonics, which is in line with other TDDFT calculation on graphene [19]. In contrast, when the driving pulses with low photon energies are adopted, the noisy peaks appear in the low-order harmonics. Regarding the reason of the noisy spectra peaks, previous studies have attributed them to impurity scattering [31], symmetry breaking [32], long decoherence time [33,34], or high joint density of states [26,27], the latter of which provides a condition that favors the interband transition. According to our calculation results [Figs. 1(c) and 3(c)], the harmonics with noisy spectra do not show apparent intraband Bloch oscillation feature with the harmonic envelopes changing periodically with the incident electric field. Once the incident peak intensity is large and the multiple of incident photon energy is close to the energy difference between the empty and occupied states with large DOSs, more electrons and holes in graphene can be excited (Fig. 4), which then leads to more interband transitions. The resulting harmonic radiation show spectrum broadening [Fig. 1(c)] and cannot maintain the circular polarization [Fig. 2(c)]. Therefore, selecting appropriate driving pulse parameters based on the band structures of solid materials to avoid excessive interband transitions will help improve the peak clarity and the circular polarization of the harmonic spectra (Fig. 6).

The appearance of noisy spectra [Fig. 1(c)] and deviation from circular polarization [Fig. 2(c)] seems not to confirm to the selection rule, in which the respective polarizations of $6n + 1$ and $6n - 1$ harmonics of graphene should be same as and opposite to that of the circular polarization of the driving pulse, respectively. This deviation may result from the symmetry breaking of the circular polarization of the driving pulse. In the derivation of selection rule, the circularly polarized driving pulse has an arbitrary rotational symmetry [12]. However, in our calculation, since the driving pulse has a few optical cycles, the rotational symmetry is broken to some extent by the ultrashort pulse duration and the significant intensity variation. For the intraband Bloch oscillation, the harmonics are emitted in phase with the driving electric field [Fig. 3(a)]. After several optical cycles, this in-phase signature of the intraband process weakens the influence of breaking symmetry of the few-cycle circularly polarized pulse, leading to the abidance of the selection rule. However, the harmonics generated by interband process are not in-phase with the driving electric field [Fig. 3(c)]. This signature enhances the influence of the symmetry breaking of the few-cycle circularly polarized pulse on the selection rule. Therefore, the harmonics generated by interband process show some prohibited

orders, and the corresponding polarizations deviate from the selection rule.

In summary, we have studied the HHG process of monolayer graphene driven by circularly polarized femtosecond pulses with different photon energies and peak intensities by means of TDDFT calculations. Our results show that when the photon energy of driving pulse is high or when both photon energy and peak intensity are low, the generated harmonics show clear isolated peaks, while both the harmonic order and the polarization state obey the selection rule. In these cases, the harmonic generation mainly comes from the intraband Bloch oscillation process. In comparison, when the photon energy of driving pulse is low but the peak intensity is high, some of low-order harmonics display noisy spectral peaks and deviate from the circular polarization. This is because at low photon energy and high peak intensity it is conducive to the interband transitions between the empty and occupied bands with large DOSs, which in turn affects the spectra and polarizations of the corresponding harmonics. Our study thus provides an important reference for the selection of driving pulse parameters and solid materials for the generation of circularly polarized high harmonics, which may promote future developments of nanoscale circularly polarized attosecond photonic devices on chips. Our theoretical findings on the

correspondence between clear harmonic peaks and circular polarizations can also serve as a simple experimental criterion for the occurrence of interband transitions in the HHG of solid materials.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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