Wavelength dependence of high-order-harmonic yield in inhomogeneous fields

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(Received 29 July 2013; published 4 November 2013)

We theoretically investigate the wavelength dependence of the HHG yield in spatially inhomogeneous fields with few-cycle laser pulses. The results show that the HHG yield first decreases with a slower scaling $\lambda^{-4}$ as the increase of the laser wavelength compared with $\lambda^{-5}$ in homogeneous fields. However, with the laser wavelength further increased, an inflection of wavelength appears and then the HHG yield presents an upward trend. Based on the electron trajectories analysis, we demonstrate that the decrease in the scaling law $\lambda^{-4}$ mainly results from the cutoff extension in inhomogeneous fields. And the uprend in HHG yield is attributed to the increasing wavelength-dependent ionization rate of the additional quantum path that dominates the harmonic generation. In addition, the carrier-envelope phase dependence of the wavelength-dependent HHG yield is also investigated.

DOI: 10.1103/PhysRevA.88.053404 PACS number(s): 32.80.Qk, 33.80.Wz, 32.80.Wr, 42.50.Hz

I. INTRODUCTION

Currently, high-order harmonic generation (HHG) has been considered to be the most promising method to produce attosecond extreme ultraviolet pulses [1–4], which provides an important tool for probing the ultrafast electronic dynamics in atoms [5–7], attosecond time-resolved spectroscopy [8], and tomographic imaging of molecular orbitals [9–13], etc. The HHG process can be well depicted in terms of the semiclassical three-step model [14]: ionization, acceleration, and recombination of the electrons in the intense laser field. During the recombination, the maximum photon energy that corresponds to the harmonic cutoff energy obeys the cutoff law $\Omega_{\text{max}} = I_p + 3.17U_p$, where $I_p$ is the ionization potential and $U_p$ is the ponderomotive energy which is proportional to $I\lambda^2$. Here, $I$ and $\lambda$ represent the intensity and wavelength of the laser field, respectively.

In the past several years, the extension of the plateau in the harmonic spectrum has attracted a lot of attention [15,16], because a broader spectrum supports the generation of shorter attosecond pulses as well as a higher resolution for ultrafast measurement. According to the cutoff law, using a longer wavelength is advantageous to extend the cutoff to a higher photon energy compared with the commonly used Ti:sapphire laser (800 nm), since the harmonic cutoff scales as $\lambda^2$. This has motivated the investigations of HHG by using mid-infrared laser pulses [17–22]. However, in this regard, it is unfortunate that the harmonic yield drops dramatically as the laser wavelength increases. Lewenstein et al. [23] have first proposed a scaling of $\lambda^{-3}$ in harmonic yield due to the spreading of the returning wave packet, which has been partially supported by the experimental results [15]. However, recently, Tate et al. [24] have reported a faster decrease in the harmonic yield with $\lambda^{-5}$ based on their simulations. And this scaling law has been demonstrated by the further theoretical calculations [25,26] and experimental observations [27]. To slow down the decrease, Lan et al. [28] have investigated the HHG in a two-color scheme and predicted a power law as $\lambda^{-3(\text{--}4)}$. Besides, the HHG in preexcited medium [29–31] also provides a promising method. Nevertheless, the precise physical origin of the scaling law is not yet fully understood. Due to its potential applications, many efforts have been paid to study the wavelength scaling in the harmonic yield over the years.

The plasmonic field enhancement from metal nanostructures has currently attracted a lot of interest from investigators. As an alternative technique for HHG, the field enhancement based on surface plasmon resonances presents a potential application in improving the efficiency and duty cycle of XUV radiation without extra cavities or laser pumping to amplify the input laser power [32,33]. During the region where the HHG processes take place, the enhanced field is not spatially homogeneous, which is called an inhomogeneous field. By exploiting the enhanced field based on plasmonics, Kim et al. [32] have successfully produced the XUV wavelengths from the 7th (114 nm) to the 17th (47 nm) harmonics by inputting a seed femtosecond laser field, which is locally enhanced in the vicinity of the metal nanostructures. However, the experimental outcome of Kim et al. has been subject to an intense examination [34–36]. Fortunately, alternative approaches to realize plasmonic field enhancement were explored by employing different kinds of metallic nanostructures [33,37]. Husakou et al. [38] have performed a systematic theoretical investigation on the plasmonic field enhancement with a modified version of the Lewenstein model, in which the field inhomogeneity and electron absorptions from the metal surface are incorporated. By using the linearly spatial-dependent laser fields, researchers [39–46] have theoretically investigated the characteristics of the HHG in inhomogeneous fields, for instance, the generation of even harmonics, extension of the cutoff, and the selection of quantum paths, etc. Very recently, Ciappina et al. [47,48] have employed the electric fields obtained from 3D finite element simulations and demonstrated that the HHG process in inhomogeneous fields was sensitive to both the confinement of the electron motion and the inhomogeneity of the laser field. In addition, recently, the above-threshold ionization (ATI) process [49,50] driven by spatially inhomogeneous fields has also received special attention. However, in these...
previous studies, the wavelength-dependent harmonic yield in inhomogeneous fields has scarcely been reported.

In this paper, we theoretically investigate the dependence of the HHG yield on the wavelength of the driving laser in spatially inhomogeneous fields by using few-cycle laser pulses. Based on the numerical solutions of the time-dependent Schrödinger equation, we observe that, with the increase of the laser wavelength, the HHG yield in inhomogeneous fields first falls off with a scaling of $\lambda^{-4}$, which is somewhat slower than $\lambda^{-5}$ in homogeneous fields. However, when the laser wavelength is further increased, an inflection of wavelength appears and then the wavelength-dependent HHG yield complies with an uptrend. By calculating the classical electron trajectories, we find that the decrease in the scaling $\lambda^{-4}$ mainly originates from the extension of the cutoff in inhomogeneous fields. And the uptrend in HHG yield is due to an additional quantum path, which has a increasing wavelength-dependent ionization rate. It is worth mentioning that by using nonadiabatic turn-on (NAT) pulses, Pérez-Hernández et al. [51] have also discovered an uptrend in the HHG yield with increasing laser intensities. Nevertheless, in their work, the inflection is induced by a particular trajectory that appears during the pulse turn-on, which is different from that in this paper. In addition, the carrier-envelope phase (CEP) dependence of the HHG yield is also investigated in this work.

### II. THEORETICAL MODEL

In a linearly polarized laser pulse, the dynamics of an atomic electron is mainly along the direction of the laser field, which allows us to model the HHG process by performing the simulations of the time-dependent Schrödinger equation (TDSE) in one spatial dimension,

$$i \frac{\partial \Psi(x,t)}{\partial t} = H(t)\Psi(x,t) = \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} + V_{\text{atom}}(x) + V_{\text{laser}}(x,t) \right] \Psi(x,t), \quad (1)$$

where $V_{\text{atom}}(x)$ is the Coulomb potential of the atom, which can be described by

$$V_{\text{atom}}(x) = -\frac{1}{\sqrt{x^2 + \varepsilon}}. \quad (2)$$

Here, $\varepsilon$ is the soft core parameter. In our simulations, we chose argon as the target atom and the corresponding $\varepsilon$ equals 1.415. The potential $V_{\text{laser}}(x,t)$ represents the interaction of the atomic electron and the laser field. Due to the spatial dependence of the laser field, it is given by

$$V_{\text{laser}}(x,t) = E(x,t)x. \quad (3)$$

Here the spatially inhomogeneous field $E(x,t)$ which is linearly polarized along the $x$ axis is expressed as

$$E(x,t) = E_0 f(t) [1 + \beta x] \cos(\omega t + \phi), \quad (4)$$

where $E_0$ is the amplitude, $\omega$ is the frequency, and $\phi$ is the CEP of the incident laser pulse. $f(t)$ is the sin-squared envelope and $\beta$ is a small parameter that determines the order of inhomogeneity of the laser field. Specially, $\beta = 0$ corresponds to the homogeneous field in space. Equation (1) can be solved numerically by using the Crank-Nicholson method [52], and the generated harmonics can be calculated by the time-dependent dipole acceleration $a(t)$, which is given by

$$a(t) = \frac{d^2 \langle x \rangle}{dt^2} = -\langle \Psi(t) | [H(t), [H(t), x]] | \Psi(t) \rangle, \quad (5)$$

where $H(t)$ and $\Psi(x,t)$ are the Hamiltonian and the electron wave function, respectively. The harmonic spectrum is then obtained by Fourier transforming the dipole acceleration $a(t)$:

$$a_\omega = \left| \frac{1}{T} \int_0^T a(t) \exp(-i \omega t) dt \right|^2, \quad (6)$$

where $T$ and $\omega$ are the duration and frequency of the driving pulse, respectively. And $\omega$ corresponds to the harmonic order.

### III. RESULTS AND DISCUSSION

In our simulations, the intensity of the incident laser is taken to be $3 \times 10^{14}$ W/cm$^2$, and the duration of the laser pulse is five optical cycles. The corresponding carrier-envelope phase (CEP) is set to be 0. Then the wavelength scaling of the HHG yield is calculated by integrating the HHG yield from 20 to 50 eV with the laser wavelength changing from 0.8 $\mu$m to 2.0 $\mu$m. For comparison, we first evaluate the scaling law in the homogeneous fields. As shown in Fig. 1(a), the HHG yield scales as $\lambda^{-5.3}$ in the homogeneous fields at a constant laser intensity, which agrees qualitatively with the scaling $\lambda^{-5-6}$ in previous reports [24–26]. Figure 1(b) shows the results calculated in the spatially inhomogeneous fields. Here the inhomogeneity parameter $\beta$ is chosen to be 0.005. One can clearly see that the dependence of HHG yield on the wavelength falls off with a power law of $\lambda^{-4.3}$, which is somewhat lower than that in Fig. 1(a). This suggests a relatively

![Graph](image_url)

**FIG. 1.** (Color online) Wavelength scaling of HHG yield in (a) the spatially homogeneous fields ($\beta = 0$) and (b) inhomogeneous fields ($\beta = 0.005$). The intensity and the duration of the incident laser pulse are $3 \times 10^{14}$ W/cm$^2$ and five optical cycles, respectively. The harmonic yield is integrated over the energy range 20–50 eV, and the CEP is taken as 0 in the calculation.
slower decrease with a reduced factor of $\lambda^{-1}$ in HHG yield compared with $\lambda^{-5.3}$ in homogeneous fields.

To obtain a better understanding of the $\lambda^{-1}$ difference of the wavelength scaling laws between the homogeneous and inhomogeneous cases, we further calculate the classical electron trajectories by using the semiclassical three-step model [14]. Here, we first investigate the classical traveling time of the continuum electron trajectories, which is proportional to the wave-packet spread and directly affects the harmonic yield. Due to the degeneracy of the quantum paths contributing to the harmonic generation, we focus on the short path of the electron trajectory, that around the peak of the laser field, which provides the maximum contribution to the harmonic emission. The corresponding result is shown in Fig. 2(a). As can be seen in Fig. 2(a), for a fixed electron returning energy of 35 eV, the electron traveling time in homogeneous fields (red squares) is practically identical with that in inhomogeneous fields (green circles), only slight differences exist in the long-wavelength range (1600–2000 nm). This result implies that the spreading of the electron wave packet is not the critical factor that causes the difference in the scaling laws. We further consider the wavelength dependence of the harmonic cutoff in the two cases. As shown in Fig. 2(b), the cutoff energy $\Omega_{\text{max}} = I_p + 3.17U_p$ in homogeneous fields (red squares) is demonstrated to follow a scaling of $\lambda^{1.74}$, which is very close to the well-known $\lambda^2$. In the inhomogeneous fields (green circles), the harmonic cutoff is extremely extended and the cutoff energy increases with a slower scaling of $\lambda^{0.78}$, which consequently lowers the wavelength scaling law with a factor of $\lambda^{0.96}$, compared with $\lambda^{1.74}$ in homogeneous fields. This result provides a good explanation for the $\lambda^{-1}$ difference in Fig. 1. Besides, we also observe that the ionization rates of the electron trajectories for different laser wavelengths are sizable. Then we can conclude that the decrease in the wavelength scaling in inhomogeneous fields primarily originates from the extension of the cutoff energy, and the influence of the electron wave packet spread is negligible.

In Fig. 3 we investigate the dependence of HHG yields driven by inhomogeneous fields on the CEP, which is crucial for the harmonic generation with few-cycle laser pulses [53–55]. We have calculated the wavelength scalings $\lambda^{\beta}$ of harmonic yields in inhomogeneous fields with the CEP varying from 0 to $2\pi$. The result is shown as the green (lower) line in Fig. 3(a). For comparison, the result in the homogeneous fields is also presented [the red (upper) line]. It is obvious that the $x$ values of the power law in inhomogeneous fields are all within the range of 4–5, which is somewhat smaller than those (5–6) in homogeneous fields. With the same CEP value, it demonstrates a relatively slower decrease in HHG yield in the inhomogeneous case. Moreover, we can also find that, in inhomogeneous fields, the optimal value of CEP is $1.9\pi$ and the corresponding scaling is $\lambda^{−4.1}$, as shown in Fig. 3(b); the most negative one appears at $0.9\pi$ with a scaling of $\lambda^{−4.8}$, as shown in Fig. 3(c). The CEP difference gives rise to a $\lambda^{−0.7}$ factor to the wavelength scaling. On the other hand, for homogeneous fields, when the laser field is reversed (with a $\pi$ difference in CEP), the observed harmonic spectrum remains exactly the same. The CEP dependence of the scaling laws is modulo $\pi$ due to the inversion symmetry. In inhomogeneous fields, since the enhanced field is unevenly distributed in the space, the inversion symmetry is broken [39], which considerably affects the cutoff and efficiency of the generated harmonics. As a result, the dependence on CEP is modulo $2\pi$ in inhomogeneous fields.

Next we perform investigations about the influence of the inhomogeneity parameter on the wavelength-dependent HHG yield. Here, the CEP of the driving pulse is set to be 0. We have chosen different values for the inhomogeneity parameter $\beta$, and the corresponding calculated results are presented in Fig. 4. One can clearly see that for different inhomogeneity parameters, the HHG yield first decreases rapidly with a scaling of about $\lambda^{-4}$ as the laser wavelength increases. This

FIG. 2. (Color online) (a) Traveling time of the electron trajectories with the recombination energy of 35 eV in spatially homogeneous (red squares) and inhomogeneous (green circles) fields. (b) Wavelength dependence of the harmonic cutoff energy in the spatially homogeneous (red squares) and inhomogeneous (green circles) fields. Here, the parameter $\beta = 0.005$. Other parameters are the same as those in Fig. 1.

FIG. 3. (Color online) (a) Dependence of wavelength scalings on the CEP of the laser pulse in spatially homogeneous fields [the red (upper) line] and inhomogeneous fields [the green (lower) line]. (b, c) The wavelength scalings of the HHG yield for two different CEP values in the spatially inhomogeneous fields ($\beta = 0.005$). Other parameters are the same as in Fig. 1.
result agrees well with that in Fig. 1(b). However, when the wavelength is further increased, a turning point appears, which is called an inflection here. Beyond the inflection, the harmonic yield no longer declines as the $\lambda^{-4}$ scaling but presents an upward trend as the wavelength increases. We can also find that the wavelength of the inflection is gradually reduced as the inhomogeneity parameter $\beta$ increases. For the case of $\beta = 0.005$ of which the inhomogeneity of the driving laser is relatively low, the corresponding wavelength of the inflection may have gone out of the wavelength range in Fig. 4. For much longer wavelengths, the classical excursion of the ionized electrons becomes important and some effects that are not included in the 1D model, such as the probability of transversal ionization, should be taken into account. Then the 1D-TDSE model seems too simple to describe the harmonic generation at very long wavelengths. So here we no longer calculate the inflection of wavelength in the case of $\beta = 0.005$. To confirm the 1D simulations, we also calculate the wavelength-dependent HHG yield by resolving the 3D-TDSE. The calculation details can be found in Ref. [45]. Since the numerical simulations of 3D-TDSE require much longer computation times than those of the 1D model, especially in the mid-infrared band, here we calculate only the wavelength-dependent HHG yield with $\beta = 0.015$. The result is shown in Fig. 5. We can find that the upturn in the harmonic yield still exists in the long-wavelength range. Before the inflection, the decrease of harmonic yield follows a scaling law of $\lambda^{-4.25}$, which is very close to that of $\lambda^{-4}$ in Fig. 4. The only difference is the position of the inflection (2200 nm for the 3D model and 1900 nm for the 1D model). However, this difference will not influence the conclusion of this paper. To sum up, we find an upward trend of harmonic yield with the increase of laser wavelength in the long-wavelength range. Before the laser wavelength reaches the inflection, little dependence of the wavelength scaling law ($\sim \lambda^{-4}$) on the inhomogeneity $\beta$ is demonstrated.

To clarify the physical mechanism of the wavelength dependence of harmonic yield in spatially inhomogeneous fields, we further perform classical trajectory analysis based on the three-step model. Here, the inhomogeneity parameter $\beta$ is set to 0.015. We have calculated the classical electron trajectories with different driving laser wavelengths. The corresponding results are shown in Fig. 6. In Fig. 6(a), the laser wavelength is set to be 1500 nm, which is smaller than that of the inflection ($\sim 1900$ nm). The main electron trajectory that contributes to the harmonic emission is marked as $R_1$. This quantum path can be well used to explain the scaling $\sim \lambda^{-4}$ as shown in Fig. 2, since the scaling law is barely dependent on the inhomogeneity parameter $\beta$ in the range where the laser wavelengths are smaller than the inflection, while for the case of 2400 nm, which is longer than the inflection, the electron trajectories become more complex. Apart from the quantum path $R_1$, an additional quantum path marked as $R_2$ emerges as shown in Fig. 6(b). Furthermore, we performed the time-frequency analysis of the generated harmonic spectra driven by the laser fields mentioned in Fig. 6. The corresponding results are presented in Fig. 7. We find that in the case of 1500 nm [Fig. 7(a)], the major

![Figure 4](image1.png)

**FIG. 4.** (Color online) Wavelength dependence of the integrated harmonic yield for different inhomogeneity parameters $\beta$ in the spatially inhomogeneous fields. Other parameters are the same as in Fig. 1.

![Figure 5](image2.png)

**FIG. 5.** (Color online) Wavelength-dependent HHG yield based on the 3D-TDSE simulations with the inhomogeneity parameter $\beta = 0.015$.

![Figure 6](image3.png)

**FIG. 6.** (Color online) Rescattering energies of electrons as a function of the ionization time (red circles) and recombination time (green squares) for different laser wavelengths (a) 1500 nm and (b) 2400 nm. Here, the parameter $\beta$ is chosen as 0.015. Other laser parameters are the same as mentioned in Fig. 1.
harmonic emission bursts near the 2.4 optical cycle, which corresponds to the emission of quantum path $R_1$, while for 2400 nm [Fig. 7(b)], the harmonic emission from $R_1$ is deeply reduced. The intensity of harmonic emission from $R_2$ (the emission time is at 2.8 optical cycles) is much higher than that from $R_1$. This result implies that the harmonic generation is dominated by the quantum path $R_2$, and the contribution of path $R_1$ can even be ignored in the long-wavelength range.

A better understanding of the uptrend in HHG yield is obtained by performing the calculation of the electron trajectories for different wavelengths in the long-wavelength range (larger than the inflection). Figure 8(a) depicts the electron energies as a function of the ionization time for 2200 nm (red circles), 2400 nm (green diamonds), and 2600 nm (blue squares), respectively. Here, the abscissa is measured with the optical cycle of the laser field. One can clearly see that the longer the laser wavelength is, the later the ionization time of the quantum path $R_2$. Figure 8(b) shows the corresponding ionization rates for the three different wavelengths. It is obvious that the ionization rates calculated from the TDSE simulations with the three laser wavelengths are substantially the same. For the same electron rescattering energy, the quantum path $R_2$ for longer laser wavelength corresponds to a much higher ionization rate, which directly leads to a higher harmonic yield. Additionally, we also calculate the electron traveling times of quantum path $R_2$ for the three cases (not presented here). It is found that, for different laser wavelengths, the electrons from path $R_2$ recombine almost simultaneously, and the corresponding electron traveling time is barely changed, which will not affect the harmonic yield visibly as the laser wavelength increases. To sum up, we conclude that the upward trend of harmonic yield in the long-wavelength range mainly results from the increasing ionization rate of the additional quantum path $R_2$ as the wavelength increases.

In general, phase matching plays an important role in the efficiencies of macroscopic harmonic spectra, and the phase matching can be well satisfied by adjusting the experimental conditions. By using a thin gas jet with low gas pressure, the collective effects can be effectively avoided and the macroscopic harmonic emission can agree well with the result based on single-atom simulations. This has been demonstrated in several experiments [27,56]. Specifically, Shiner et al. [27], measured the harmonic efficiency versus wavelength by using a 0.5-mm gas jet where the phase matching is well achieved, and their result is very close to the theoretical predictions $\lambda^{-5-6}$ [24–26] based on single-atom calculations. In this paper, we aim to theoretically investigate the single-atom scaling of HHG yield and explore the physical origin of the scaling in inhomogeneous fields. We believe that under suitable experimental conditions as in Ref. [27], our results can also be demonstrated in experiment.

**IV. CONCLUSION**

In conclusion, we perform theoretical simulations of the wavelength-dependent HHG yield in spatially inhomogeneous fields with few-cycle laser pulses. Based on our numerical solutions of the time-dependent Schrödinger equation, we find that the HHG yield in inhomogeneous fields first falls off with a power law of $\lambda^{-4}$ with the increase of the laser wavelength, which is somewhat slower than for $\lambda^{-5}$ in homogeneous fields. With the CEP changing from 0 to $2\pi$, the $x$ values of the
scaling $\lambda^{-x}$ in inhomogeneous fields are maintained in the range of 4–5. By performing the classical electron trajectories analysis, we find that the decrease in the scaling law $\lambda^{-4}$ mainly results from the cutoff extension in inhomogeneous fields. However, with the laser wavelength further increased, the decrease of the HHG yield is interrupted by an inflection. Beyond the inflection, the HHG yield begins to rise as the laser wavelength increases, and the uptrend of the HHG yield is further demonstrated to originate from the additional quantum path ($R_2$), which corresponds to an increasing wavelength-dependent ionization rate.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China under Grant No. 91275126, No. 11234004, and No. 11204095, and the 973 Program of China under Grant No. 2011CB808103.