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Effect of multiple rescattering processes on harmonic emission in spatially inhomogeneous field*

Cai-Ping Zhang(张彩萍)[1,2], Chang-Long Xia(夏昌龙)[1], Xiang-Fu Jia(贾祥富)[1,†], and Xiang-Yang Miao(苗向阳)[1,‡]

[1] College of Physics and Information Engineering, Shanxi Normal University, Linfen 041004, China
[2] College of Chemistry and Materials Science, Shanxi Normal University, Linfen 041004, China

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The effect of multiple rescattering processes on the harmonic emission from He atom in a spatially inhomogeneous field is discussed by solving the one-dimensional time-dependent Schrödinger equation and the classical equation of motion. By establishing the physical model of the harmonic emission in the inhomogeneous field, we discuss the related characters of the multiple rescatterings process in the harmonic generation process. It shows that the second rescattering rather than the first rescattering tends to determine the harmonic cutoff energy when the inhomogeneous parameter is larger than 0.0055. Additionally, with the classical simulation, the underlying physical mechanism of the continuum–continuum harmonics is also revealed. Moreover, this work may provide new physical insight into the harmonic generation in an inhomogeneous field, and is beneficial to further extract the harmonic emission from molecular systems.

Keywords: high-order harmonic generation, multiple rescatterings, spatially inhomogeneous field

PACS: 42.65.Re, 42.25.Hz, 33.80.Rv **DOI:** 10.1088/1674-1056/27/3/034206

1. Introduction

High-order harmonic generation (HHG) is a useful tool to generate the isolated attosecond (as) pulse, which can imagine the electron motion or the molecular orbital.[1] Numerous methods have been proposed to obtain an ultrashort pulse, such as two-color laser field,[2,3] chirped laser field,[4,5] and polarization gating technique.[6,7] Moreover, the maximum kinetic energy obtained in these traditional schemes is about $3.17U_p$ based on the three-step model,[8] here U_p is the ponderomotive potential. Recently, the harmonic emission in a spatially inhomogeneous field has been paid much attention to experimentally and theoretically.[9–13] Kim *et al.*[9] showed that the incident laser pulse can be enhanced up to three orders of magnitude between the vertices. With an extremely extended harmonic spectrum in a spatially inhomogeneous field,[10,11] an isolated attosecond pulse can be achieved by superposing proper harmonic orders.[12,13] Additionally, Zagoya *et al.*[14] performed a phase-space analysis caused by the field inhomogeneity. With more complicated physical mechanisms, the molecular HHG in the spatially inhomogeneous field has also attracted much attention. Wang *et al.*[15] showed that the ultrashort pulse with duration of 127 as can be generated from H_2^+ by reducing the harmonic chirp in a spatially inhomogeneous field. Feng[16] proposed that the harmonic efficiency of H_2^+ can be enhanced by five orders of magnitude in a chirped inhomogeneous field. Yavuz *et al.*[17] pointed out that the nuclear vibration does enhance the harmonic yield while the maximum kinetic energy of $4U_p$ is greatly maintained. Furthermore, the interference effects and the asymmetric recollision have also been discussed in the asymmetric inhomogeneous field.[18]

In these studies mentioned above, the harmonic emissions both in homogeneous and inhomogeneous fields are mainly focused on the first rescattering, which can provide intuitive and quantitative explanations on the existence and the value of the cutoff. However, the electron may revisit the parent ion more than once and experience longer traveling time in the continuum before emitting the harmonic photon. In other words, the multiple rescatterings could contribute to the HHG process. Moreover, Tate *et al.*[19] and He *et al.*[20] have discovered the trajectories of multiple rescatterings in atomic harmonic emission in homogenous field with long wavelength. Based on the quantum simulation and classical simulation, we have discussed the characters of the atomic multiple rescatterings in a homogeneous trapezoid field.[21] It is found that the maximum kinetic energy obtained in the homogeneous field via the $2n$-th rescattering is lower than that via the $(2n-1)$-th rescattering and the $(2n+1)$-th rescattering ($n = 1, 2, 3, \ldots$). Furthermore, it also applies to the homonuclear multiple rescattering in molecular harmonic emission.[22] In general, the cutoff energy in a homogenous field is determined by the first rescattering. With the same mechanism, the ionized electron may also miss the parent ion and be reaccelerated in an in-

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†Corresponding author. E-mail: jiaxf@dns.sxnu.edu.cn
‡Corresponding author. E-mail: sxxymiao@126.com

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homogeneous field. In other words, the multiple rescatterings probably contribute to the HHG process in an inhomogeneous field. Furthermore, due to the dependence of the laser intensity on the space position in an inhomogeneous field, the laser intensity could be extremely enhanced at larger space position with proper inhomogeneity.[10,11] And then the electron may obtain higher energy after longer acceleration time in high-order rescattering process, however, it is quite different from the case of homogeneous field that the laser intensity tends to decrease when the electron returns back to recombine with the parent ion after the laser field reversing its direction as shown in Fig. 2 in Ref. [21]. So it is worth to further discuss the multiple rescattering in an inhomogeneous field to discover more interesting physical information of the harmonic emission.

In this paper, we investigate the effect of the multiple rescatterings on atomic harmonic emission in an inhomogeneous field by solving the time-dependent Schrödinger equation (TDSE) and the classical equation of motion. The multiple rescatterings in the inhomogeneous field exhibit quite different characters from those in the homogenous field. With the time–frequency maps, the interference between the first and the second rescatterings contributes to the continuum–continuum harmonics. Furthermore, we also discuss the effect of the carrier-envelope phase (CEP) on the continuum–continuum channel.

2. Theoretical method

The quantum simulation of multiple rescatterings in a spatially inhomogeneous field is presented by numerically solving the TDSE.[23–26] In the one-dimensional case, the TDSE based on the single active electron approximation can be expressed as (atomic units (a.u.) are used throughout this paper unless otherwise indicated)

$$\mathrm{i}\frac{\partial}{\partial t}\psi(x,t)=\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2}+V_{\mathrm{C}}(x)+\int_0^x E(x',t)\,\mathrm{d}x'\right]\psi(x,t), \quad (1)$$

where $V_{\mathrm{C}}(x)=-1/\sqrt{a+x^2}$ is the "soft-core" potential and $\int_0^x E(x',t)\,\mathrm{d}x'$ is the external interaction between the laser field and the He atom. The parameter $a=0.484$ is chosen to mach the ionization energy of the ground state (i.e., 24.6 eV). Here, the driving inhomogeneous field $E(x,t)$ is in a similar way as that in Refs. [14] and [27], which can be viewed as a good approximation of the field generated near metal nanospheres for the case of small inhomogeneity.[28,29] The corresponding expression is

$$E(x,t)=E_0\sin^2(\pi t/t_{\mathrm{d}})\cos(\omega t+\phi)(1+\beta x), \quad (2)$$

where t_{d} lasting for four optical cycle (o.c.) is the total interaction time with 1 o.c. = 2.67 fs, and laser frequency ω equals to 0.057 a.u., corresponding to the wavelength of 800 nm. E_0 and ϕ are the peak amplitude of the electric field and CEP, respectively. β is the inhomogeneity parameter. The CEP is set to be 0 unless otherwise stated.

The time-dependent wave function is advanced using the standard second-order split-operator method.[30–32] According to the Ehrenfest theorem,[33] the dipole acceleration $d_{\mathrm{A}}(t)$ can be written as

$$d_{\mathrm{A}}(t)=\langle\psi(x,t)\mid-\frac{\partial V(x,t)}{\partial x}\mid\psi(x,t)\rangle, \quad (3)$$

with

$$V(x,t)=V_{\mathrm{C}}+\int_0^x E(x',t)\,\mathrm{d}x'. \quad (4)$$

The time-frequency distribution is given by means of the wavelet transform[34,35]

$$d_\omega(t)=\int d_{\mathrm{A}}(t')\sqrt{\omega_0}W(\omega_0(t'-t))\,\mathrm{d}t', \quad (5)$$

with

$$W(x)=\frac{1}{\sqrt{\tau}}\mathrm{e}^{\mathrm{i}x}\mathrm{e}^{-x^2/2\tau^2}. \quad (6)$$

The classical simulation is based on the Newtonian classical dynamics: $\ddot{x}(t)=-E_0\sin^2(\pi t/t_{\mathrm{d}})\cos(\omega t+\phi)(1+\beta x)$. The electron velocity $\dot{x}(t)$ and electron position $x(t)$ can be expressed as follows: $\dot{x}(t)=\int_{t_{\mathrm{i}}}^{t_{\mathrm{r}}}\ddot{x}(t)\,\mathrm{d}t$ and $x(t)=\int_{t_{\mathrm{i}}}^{t_{\mathrm{r}}}\dot{x}(t)\,\mathrm{d}t$, where t_{i} and t_{r} are the ionization time and the recombination time, respectively. The initial velocity and position are both set to be 0. And the kinetic energy is $E_{\mathrm{k}}=\dot{x}^2(t)/2$.

3. Results and discussion

In this section, we investigate the harmonic emission from He atom in a spatially inhomogeneous field as Eq. (2) shown, and the related laser intensity is 1.0×10^{15} W/cm^2. The harmonic spectra (solid curves) and the classical returning energy maps for the first rescattering (circles) in fields with different inhomogeneous parameters are presented in Figs. 1(a)–1(d). Here, the harmonic energy (horizontal axis) excludes the ionization energy. For $\beta=0$ (i.e., in a homogeneous field) as shown in Fig. 1(a), the maximum kinetic energy of the He atom is equal to $3.17U_{\mathrm{p}}$. Moreover, compared with the case of $\beta=0$, the corresponding maximum kinetic energy is extended extremely with the increase of β due to the reacceleration process in the inhomogeneous field around 2.2 o.c.[12–14] Additionally, the cutoff energy based on quantum calculation (as marked by the red arrow) is still well accord with the maximum energy of the first rescattering based on classical calculation (as denoted by the dash line) for the case of $\beta=0.005$. However, the energy difference between the two simulation methods tends to be larger with the increase of β from 0.0055 to 0.0065 as shown in Figs. 1(c) and 1(d).

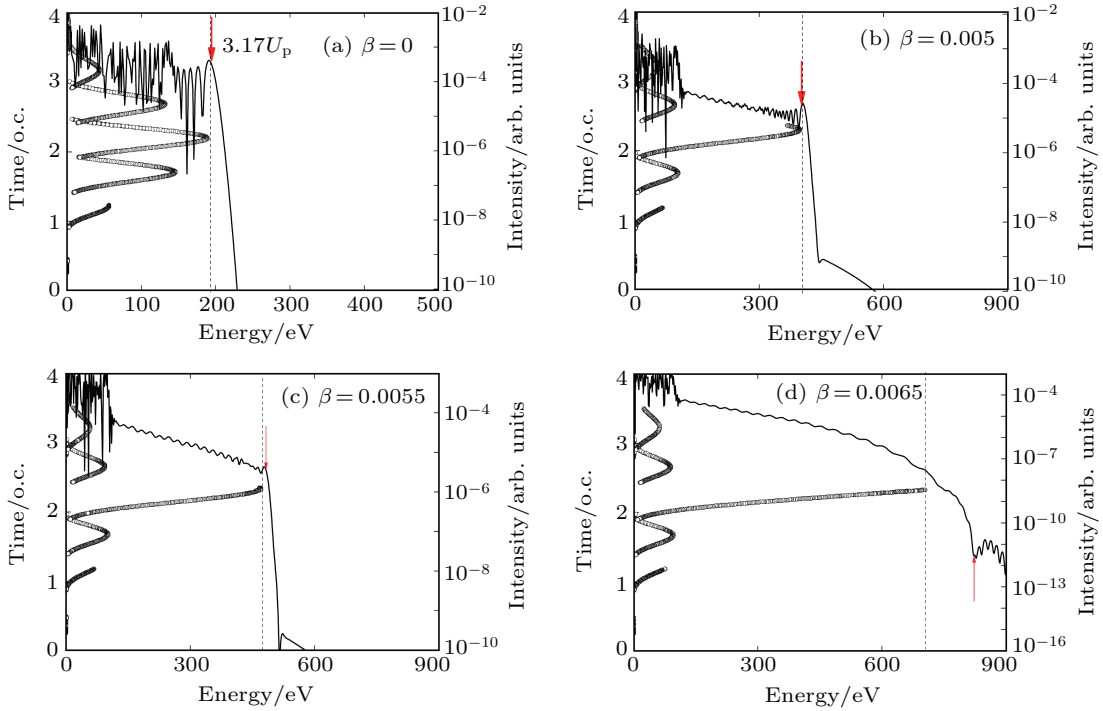


Fig. 1. (color online) Harmonic spectra of He atom (solid line) in a spatially inhomogeneous field with different inhomogeneous parameters: (a) $\beta = 0$, (b) $\beta = 0.005$, (c) $\beta = 0.0055$, (d) $\beta = 0.0065$. The harmonic energy excludes the ionization energy. The circles represent the dependence of the kinetic energy on the recombination time for the first rescattering. The red arrow and dash line show the cutoff position in the harmonic spectrum and the maximum returning energy from the first rescattering, respectively.

To seek the underlying mechanism of the energy difference, we establish the physical image of the harmonic emission in the inhomogeneous field. As seen from the electric field displayed in Fig. 2(a), five peaks (*A*, *B*, *C*, *D*, and *E*) will contribute to the harmonic generation. The electron (gray dot) can be ionized around *A*, and then is accelerated in the external field. Finally, when the laser field changes its direction after 0.5 o.c., the electron returns back to the parent ion around *B* and emits the harmonic photons (i.e., R_1 process). With a similar mechanism, the high-energy photons can also be emitted around *C*, *D*, and *E*, which correspond to the ionization around *B*, *C*, and *D*, respectively. In these processes, the ionized electron visits the parent ion only once before emitting the harmonic photon, they are named as the first rescattering as the blue arrow shown. Furthermore, the electron ionized around *A* may miss the parent ion around *B* during the recombination process, and be reaccelerated in the external field. Finally, it recombines with the parent ion around *C* as the red arrow shown (i.e., R_1' process). With twice visits to the parent ion, this process is named as the second rescattering process. Additionally, the second rescattering process can also take place around *D* and *E*, which relate to the ionization processes around *B* and *C*, respectively. According to the three-step model,[8] the recombination processes around different peaks correspond to different harmonic energies so that the harmonic emissions around *B*, *D*, and *E* contribute to the plateau region of the harmonic spectrum while the maximum peak *C* determines the cutoff position. In the following discussion, we mainly focus on the recombination around *C* to seek the physical mechanism of the energy difference shown in Fig. 1.

Based on the above physical image, both the first rescattering (i.e., ionization around *B* and recombination around *C*) and the second rescattering (i.e., ionization around *A* and recombination around *C*) would contribute to the harmonic emission around *C*. However, they tend to make different contributions to the harmonic generation for different values of β. For the case of $\beta = 0$ (i.e., the homogeneous field), the characters of the multiple rescatterings of He have been discussed in Ref. [21], which shows that the maximum energy obtained via the first rescattering is larger than that via the second rescattering. And the cutoff energy is determined by the first rescattering as depicted in Fig. 1(a). Nerveless, with the increase of β, the laser intensity is closely related to the electron position as shown in Eq. (2), and then the harmonic energies obtained via multiple rescatterings would exhibit different characters from those in the homogeneous field. To further understand the electron motion in the inhomogeneous field, the classical returning energy map for $\beta = 0.0065$ is presented in Fig. 2(b). The green circles and red circles correspond to the recombinations in the first rescattering and the second rescattering, respectively. As one can see, both the first rescattering and the second rescattering contribute to the harmonic emission around 2.2 o.c. (i.e., around peak *C*), which further verifies the discussion in Fig. 2(a). Furthermore, it is noteworthy that the maximum energy obtained from the

second rescattering (812 eV) as shown by the horizontal red arrow in Fig. 2(b) consists well with the cutoff energy shown as the vertical red arrow in Fig. 1(d), which is higher than that from the first rescattering (704 eV) as shown by the horizontal green arrow in Fig. 2(b). However, it is quite different from the result in the homogeneous field that the first rescattering determines the cutoff energy.[2–4] To have a clear insight into this phenomenon, we show the dynamics of the electron obtaining maximum kinetic energy in the first rescattering (blue line) ($t_i = 1.324$ o.c., $t_r = 2.325$ o.c.) and the second rescattering (red line) ($t_i = 1.225$ o.c., $t_r = 2.341$ o.c.) in Fig. 2(c). The enlargement of the time period from 1.22 o.c. to 1.32 o.c. is shown in Fig. 2(d). As the blue line depicted, the ionized electron can visit the parent ion only once around 2.325 o.c. in the first rescattering process in the inhomogeneous field. However, the electron can revisit the parent ion around 2.341 o.c. after the first encounter around 1.301 o.c. as the red line shown in Fig. 2(d). Moreover, the electron moves much farther away from the nucleus in $x > 0$ direction in the second rescattering than in the first rescattering. Based on Eq. (2), the laser intensity around C will be greatly enhanced in the inhomogeneous field so that the maximum energy obtained from the second rescattering will be higher than that from the first rescattering. As a result, the cutoff energy will be determined by the second rescattering in the inhomogeneous field for larger β.

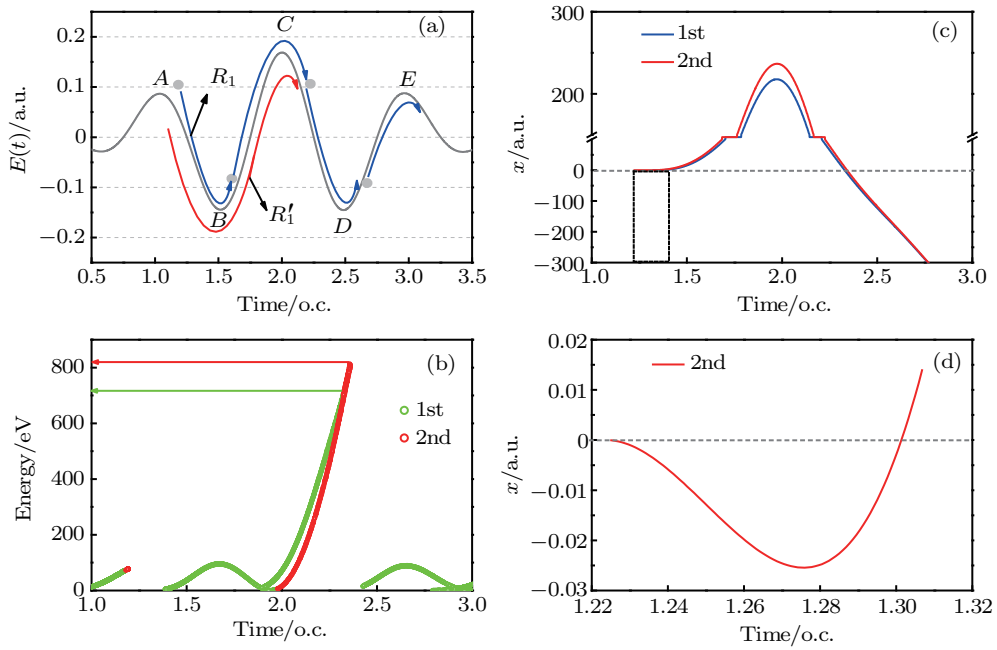


Fig. 2. (color online) (a) The electric field of a spatially inhomogeneous laser pulse (800 nm, 1.0×10^{15} W/cm^2) at $x = 0$ from 0.5 o.c. to 3.5 o.c., the gray dot represents the electron. (b) Dependence of the kinetic energy on the recombination time (green circles for the first rescattering and red circles for the second rescattering) for $\beta = 0.0065$. (c) Electron trajectories for the electrons obtaining maximum energy in the first rescattering (blue line) and the second rescattering (red line) processes. (d) The enlargement of panel (c) in the time period from 1.22 o.c. to 1.32 o.c.

To obtain a deeper insight into the atomic harmonic emission in the inhomogeneous field with larger inhomogeneity, the harmonic spectrum for $\beta = 0.0065$ in Fig. 1(d) is analyzed by the time–frequency distribution in Fig. 3(a). Moreover, the corresponding classical returning energies both from the first rescattering and the second rescattering are superimposed with the time–frequency distribution. The blue and magenta dots represent the dependence of energy on the recombination time for the first rescattering and the second rescattering, respectively. It can be seen that the quantum results accord well with those classical ones except for the arch structure in lower energy region around 2.2 o.c. as the red arrow pointed. Recall the harmonic emission illustrated in Fig. 2(a), part of electron wave packet reaccelerated in the continuum state via the first rescattering can gain extra kinetic energy E_1, while others may gain extra kinetic energy E_2 via the second rescattering. When they return back and recombine with the parent ion at the same time around peak C, the related electron wave packets will interference with each other and emit the energy $|E_1 - E_2|$, i.e., continuum–continuum harmonic emission.[36,37] To distinguish the additional channel, the energy difference between the first rescattering and the second rescattering at identical emission moment has been calculated based on the classical model. The classical energy difference as red stars described consists well with the arch structure, which indicates that the additional channel around 2.2 o.c. is the continuum–continuum harmonic channel. In other words, the multiple rescatterings could lead to the continuum–continuum harmonic emission in the inhomogeneous field. As can be seen from the sketch of inhomogeneous field with $\phi = 0$ for $\beta = 0.0065$ in Fig. 3(b), the intensity in $x > 0$ direction is more intense than that in $x < 0$

direction around 2 o.c., which indicates the strong dependence of the laser intensity on CEP in the inhomogeneous field. How does it affect the HHG process? By changing the CEP from $\phi = 0$ to $\phi = 0.4\pi$ for the case of $\beta = 0.0065$, we provide the corresponding time–frequency distribution from 1.5 o.c. to 3.5 o.c. in Fig. 3(c). Notably, the intensity of the first rescattering is similar with that of the second rescattering, which may result in the intense interference between two rescattering processes. And then the continuum–continuum harmonic around 2.75 o.c. shown in Fig. 3(c) is more intense than that for $\phi = 0$ in Fig. 3(a). Next, we will pay attention to the effect of the CEP on the continuum–continuum harmonics quantitatively. As seen from Fig. 3(c), different quantum paths contribute to different harmonic energies at different recombination time as the white dashed rectangle depicted. For example, the continuum–continuum channel contributes to 85 eV harmonic energy around 2.75 o.c. Accordingly, the intensity of the selected energy at determinate emission moment can be used to evaluate the contribution of the continuum–continuum channel.

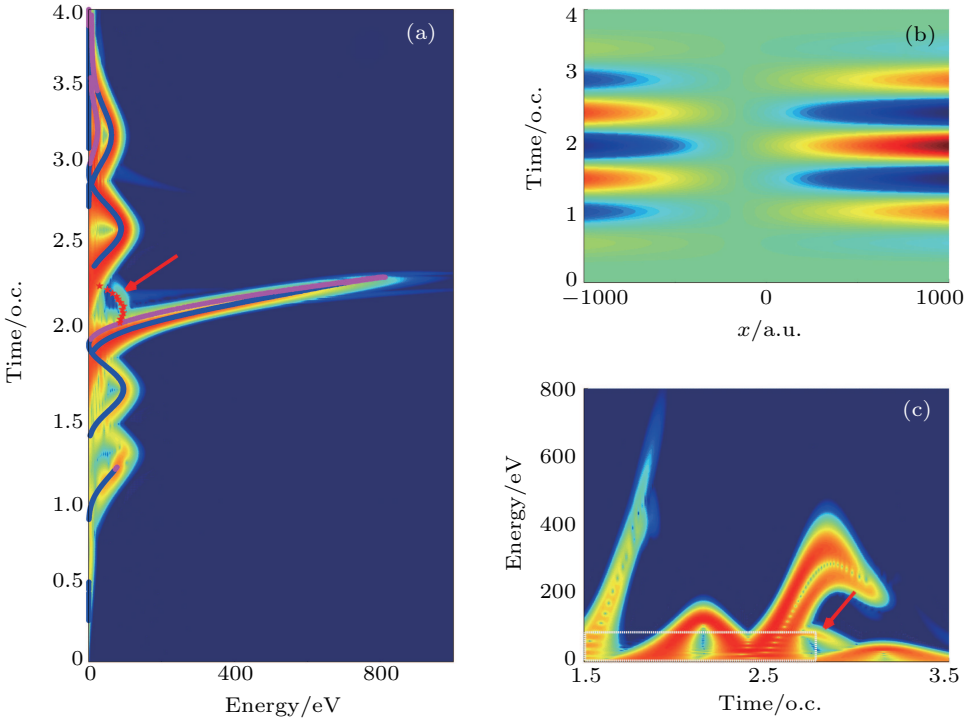


Fig. 3. (color online) (a) The time–frequency map is superimposed with the classical return energies for β = 0.0065: blue dots for the first rescattering, magenta dots for the second rescattering, and red dots for the continuum–continuum emission. (b) Sketch of the spatially inhomogeneous field with $\phi = 0$. (c) The related time–frequency map from 1.5 o.c. to 3.5 o.c. with $\phi = \pi$.

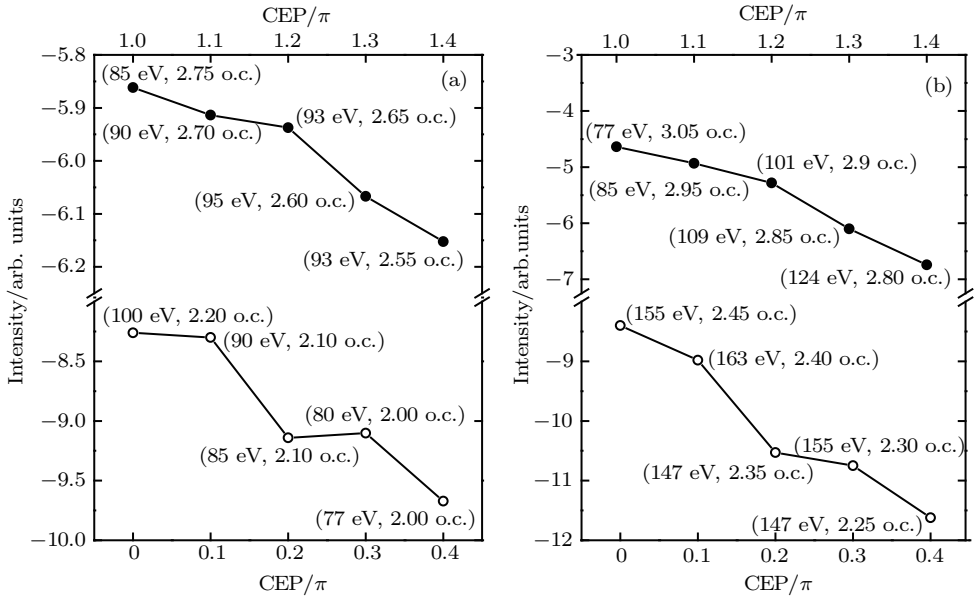


Fig. 4. The dependence of the continuum–continuum harmonic emission on CEP from 0 to 0.4π (black circles) and from π to 1.4π (black dots): (a) for the inhomogeneous field $E(x,t) = E_0 \sin^2(\pi t/t_d)\cos(\omega t + \phi)(1 + \beta x)$, (b) for the inhomogeneous field $E(x,t) = E_0 \sin^2(\pi t/t_d)\sin(\omega t + \phi)(1 + \beta x)$. Here β equals to 0.0065.

We provide the dependence of the harmonic intensity originating from the continuum–continuum channel on CEP from $\phi = 0$ to $\phi = 0.4\pi$ (black circles) and from $\phi = \pi$ to $\phi = 1.4\pi$ (black dots) in cosinoidal inhomogeneous field expressed as Eq. (2) for $\beta = 0.0065$ with the laser intensity of 1.0×10^{15} W/cm^2 in Fig. 4(a). The selected energy and recombination time correspond to the maximum intensity of the continuum–continuum harmonics. It shows that the intensity in the CEP region from π to 1.4π is three orders of magnitude more intensive than that from 0 to 0.4π. By simulating the variations of the cosinoidal and sinusoidal inhomogeneous fields with different CEP, it can be found that both inhomogeneous fields exhibit similar tendencies in space and time dimensions with the variation of CEP. So similar dependence of the continuum–continuum harmonics on CEP can be seen in sinusoidal inhomogeneous field as shown in Fig. 4(b). Additionally, the results from 0.5π to 0.9π (1.5π to 1.9π) based on the cosinoidal inhomogeneous field presented in Fig. 4(a) correspond to the results from π to 1.4π (0 to 0.4π) based on the sinusoidal inhomogeneous field shown in Fig. 4(b). Therefore, the contribution of the continuum–continuum channel to harmonic generation is sensitive to the CEP, and the contribution may reach the maximum for 0.5π (π) in cosinoidal (sinusoidal) inhomogeneous field. In general, both the multiple rescattering processes and the continuum–continuum channel can be manipulated by adjusting the CEP of the few-cycle inhomogeneous field.

4. Conclusion

We have investigated the effect of multiple rescatterings on the atomic harmonic emission in a spatially inhomogeneous field by numerically solving the time-dependent Schrödinger equation and the classical equation of motion. Both the quantum and the classical results show that with longer reacceleration process, the maximum kinetic energy of the second rescattering tends to beyond that of the first rescattering with the increase of the inhomogeneous parameter. In other words, the former determines the cutoff energy for a larger inhomogeneous parameter, which is distinct from the case in the homogeneous field. Moreover, it is found that the interference between the first rescattering and the second rescattering leads to the contribution of continuum–continuum harmonic emission in lower energy region. Additionally, it exhibits a similar dependence on the CEP in both the cosinoidal and the sinusoidal inhomogeneous fields that the intensity from π to 1.4π is more than three orders of magnitude higher than that from 0 to 0.4π. Furthermore, these results may lay the foundation for further discussion about the molecular harmonic emission in a spatially inhomogeneous field.

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