High-order harmonic generation of N_2 molecule in two-color circularly polarized laser fields*

Hui Du(杜慧), Jun Zhang(张军), Shuai Ben(贲帅), Hui-Ying Zhong(钟慧英), Tong-Tong Xu(徐彤彤), Jing Guo(郭静), and Xue-Shen Liu(刘学深)†

Institute of Atomic and Molecular Physics, Jilin University, Changchun 130012, *China*

(Received 17 November 2015; revised manuscript received 15 December 2015; published online 20 February 2016)

The generation of high-order harmonics and the attosecond pulse of the N_2 molecule in two-color circularly polarized laser fields are investigated by the strong-field Lewenstein model. We show that the plateau of spectra is dramatically extended and a continuous harmonic spectrum with the bandwidth of 113 eV is obtained. When a static field is added to the *x* direction, the quantum path control is realized and a supercontinuum spectrum can be obtained, which is beneficial to obtain a shorter attosecond pulse. The underlying physical mechanism is well explained by the time–frequency analysis and the semi-classical three-step model with a finite initial transverse velocity. By superposing several orders of harmonics in the combination of two-color circularly polarized laser fields and a static field, an isolated attosecond pulse with a duration of 30 as can be generated.

Keywords: high-order harmonic generation, two-color circularly polarized laser fields, attosecond pulse

PACS: 32.80.Rm, 42.65.Ky, 42.65.Re **DOI:** 10.1088/1674-1056/25/4/043202

1. Introduction

In the past two decades, high-order harmonic generation (HHG) has been extensively studied as a powerful tool for generating isolated attosecond pulses (IAP)[1–5] and a coherent soft x-rays source.[6] The physical mechanism of the HHG is well established by the three step model:[7–9] the electron is freed by tunneling through the barrier of the atomic potential, then the ionized electron is accelerated by the laser field and captures energy, finally it may be driven back to the parent ion, emitting high energy photons. Compared with HHG from atoms, the process of harmonic emission from the molecules is much more complicated because of the interferences of different cores and complex molecular structures.

The potential applications of HHG from molecules such as real-time observation, molecular orbital tomographic imaging and reconstruction have attracted a lot of attention.[10–15] The effect of intramolecular interference was investigated theoretically in H_2^+ and H_2 molecules by numerical solution of the Schrödinger equation or S-matrix method.[16–18] In 2005, Itatani *et al.*[19] realized to control the high order harmonic generation process by changing the alignment of molecules, they also accomplished the highest occupied molecular orbital (HOMO) of the N_2 molecule. With the development of science and technology, a lot of efficient methods have been proposed to provide information about the dynamics of orbital and nuclear.[20–22] Recently, Guo *et al.*[23] predicted that the linear combination of the HOMO and the lower-lying orbital below the HOMO (HOMO-1) can enhance the intensity of the HHG of the N_2 molecule. Pan *et al.*[24] generated an isolated 38 as pulse from the oriented molecule CO exposed to a combined field of a few-cycle chirped laser and a unipolar pulse. As we all know, when an atom is exposed to circularly polarized laser fields, the ionized electron can hardly return to the parent ion, which leads to low efficiency of HHG. Much effort has been made to further investigate this phenomenon.[25–27] It has been shown that a supercontinuum can be obtained by combining a left circularly pulse and a right circularly pulse with a certain delay.[28] Yuan *et al.*[29] achieved a single circularly polarized attosecond pulse by an intense few cycle elliptically polarized laser pulse combined with a terahertz field. Mancuso *et al.*[30] observed experimentally the three-dimensional (3D) photoelectron distributions by two-color circularly polarized laser fields, which validated the theoretical strong-field model of HHG.

In this paper, we theoretically study the high-order harmonic generation of the N_2 molecule in the two-color circularly polarized laser fields by the strong-field Lewenstein model. We show that a continuous harmonic spectrum is obtained by the two-color circularly polarized laser fields alone, and in the addition of a static field to the *x* direction, the quantum path control is realized. We perform the time–frequency analysis and the semi-classical three-step model to show the underlying physical mechanism. As a result, an isolated attosecond pulse with a duration of 30 as can be generated directly by the superposition of the supercontinuum harmonics.

*Project supported by the National Natural Science Foundation of China (Grant Nos. 61575077, 11271158, and 11574117).

†Corresponding author. E-mail: liuxs@jlu.edu.cn

© 2016 Chinese Physical Society and IOP Publishing Ltd

2. The model

The strong-field Lewenstein model has been widely used to investigate the HHG of atoms and molecules.[7,28,31,32] The transition amplitude for HHG can be calculated by the integral

$$\begin{aligned} r(t) &= \mathrm{i}\int_0^{\infty}\mathrm{d}\tau\left(\frac{\pi}{\varepsilon+\mathrm{i}\tau/2}\right)^{3/2} d^{*}_{\mathrm{rec}}[p_{\mathrm{st}}(t,\tau)-A(t)] \\ &\quad\times d_{\mathrm{ion}}[p_{\mathrm{st}}(t,\tau)-A(t-\tau)] \\ &\quad\times \exp[-\mathrm{i}S(p_{\mathrm{st}},t,\tau)]E(t-\tau)+\mathrm{c.c.}, \end{aligned} \tag{1}$$

$d_{\mathrm{ion}}[p_{\mathrm{st}}(t,\tau)-A(t)]$ and $d_{\mathrm{rec}}[p_{\mathrm{st}}(t,\tau)-A(t)]$ are the two transition dipoles of ionization and recombination. ε is a positive regularization constant, τ is the traveling time of the free electron between ionization and recombination. $E(t)$ is the electric field of the laser pulse and $A(t)$ is the associated vector potential. $p_{\mathrm{st}}(t,\tau)$ is the stationary momentum obtained by the stationary points integral algorithm. $S(p_{\mathrm{st}},t,\tau)$ is the quasi-classical action of the electron.

We investigate the HHG of the N_2 molecule in the two-color circularly polarized laser fields, in which the HOMO constructed by the GAMESS-UK package[33] is taken as the initial state. The transition amplitudes along the x and y direction can be expressed respectively as

$$\begin{aligned} r_x(t) &= \mathrm{i}\int_0^{\infty}\mathrm{d}\tau\left(\frac{\pi}{\varepsilon+\mathrm{i}\tau/2}\right)^{3/2} \\ &\quad\times d^{*}_{\mathrm{recx}}[p_{\mathrm{st}}(t,\tau)-A(t)] \\ &\quad\times\{d_{\mathrm{ionx}}[p_{\mathrm{st}}(t,\tau)-A(t-\tau)]E_x(t-\tau) \\ &\quad+d_{\mathrm{iony}}[p_{\mathrm{st}}(t,\tau)-A(t-\tau)]E_y(t-\tau)\} \\ &\quad\times\exp[-\mathrm{i}S(p_{\mathrm{st}},t,\tau)]+\mathrm{c.c.}, \end{aligned} \tag{2}$$

$$\begin{aligned} r_y(t) &= \mathrm{i}\int_0^{\infty}\mathrm{d}\tau\left(\frac{\pi}{\varepsilon+\mathrm{i}\tau/2}\right)^{3/2} \\ &\quad\times d^{*}_{\mathrm{recy}}[p_{\mathrm{st}}(t,\tau)-A(t)] \\ &\quad\times\{d_{\mathrm{ionx}}[p_{\mathrm{st}}(t,\tau)-A(t-\tau)]E_x(t-\tau) \\ &\quad+d_{\mathrm{iony}}[p_{\mathrm{st}}(t,\tau)-A(t-\tau)]E_y(t-\tau)\} \\ &\quad\times\exp[-\mathrm{i}S(p_{\mathrm{st}},t,\tau)]+\mathrm{c.c.}, \end{aligned} \tag{3}$$

where the quasi-classical action $S(p_{\mathrm{st}},t,\tau)$ at the stationary points can be calculated by

$$\begin{aligned} S(p_{\mathrm{st}},t,\tau) &= \tau I_p-\frac{1}{2}[p^2_{\mathrm{st},x}(t,\tau)+p^2_{\mathrm{st},y}(t,\tau)] \\ &\quad+\frac{1}{2}\int_{t-\tau}^{t}[A_x^2(t'')+A_y^2(t'')]\,\mathrm{d}t''. \end{aligned} \tag{4}$$

The harmonic spectrum intensity is proportional to the Fourier transformation of the time dependent dipole acceleration

$$P_a(\omega)\sim\left|\int[r_x(t)\boldsymbol{e}_x+r_y(t)\boldsymbol{e}_y]\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t\right|^2. \tag{5}$$

The temporal profile of an attosecond pulse can be obtained by superposing several harmonics in the x and y directions as follows:

$$I_x(t)=\left|\sum a_{qx}\,\mathrm{e}^{\mathrm{i}\omega qt}\right|^2, \tag{6}$$

$$I_y(t)=\left|\sum a_{qy}\,\mathrm{e}^{\mathrm{i}\omega qt}\right|^2, \tag{7}$$

where $a_{qx}=\int r_x(t)\,\mathrm{e}^{-\mathrm{i}q\omega t}\,\mathrm{d}t$ and $a_{qy}=\int r_y(t)\,\mathrm{e}^{-\mathrm{i}q\omega t}\,\mathrm{d}t$. q is the harmonic order.

As we know, the electron ionized without an initial transverse velocity in the elliptical polarized laser field will miss the parent ion. A semi-classical three-step model with a proper initial transverse velocity was introduced, and the transverse displacement caused by the external field is compensated by an initial transverse velocity.[34–36] Thus the semi-classical electron trajectories can be obtained as

$$x(t)=\int_{t_0}^{t}\left(\int_{t_0}^{t'}E_x(t'')\,\mathrm{d}t''+v_{x0}\right)\mathrm{d}t', \tag{8}$$

$$y(t)=\int_{t_0}^{t}\left(\int_{t_0}^{t'}E_y(t'')\,\mathrm{d}t''+v_{y0}\right)\mathrm{d}t'. \tag{9}$$

The initial velocities can be calculated as $v_{x0}=v_{\parallel}\cos(\alpha_0)+v_{\perp}\sin(\alpha_0)$ and $v_{y0}=-v_{\parallel}\sin(\alpha_0)+v_{\perp}\cos(\alpha_0)$, where $v_{\parallel}$ and $v_{\perp}$ are the initial parallel velocity and transverse velocity, respectively. Since the electron leaves the molecule by tunneling, the initial velocity parallel to the ionizing field can be set as $v_{\parallel}=0$. α_0 is the angle between the ionizing field vector and the coordinate axis at the ionization time t_0. The recombination time with a proper finite initial velocity will be obtained by calculating $x(t)=0$, $y(t)=0$.

3. Numerical results

We investigate the HHG process of the N_2 molecule in two-color circularly polarized laser fields and the HOMO is taken as the initial state as shown in Fig. 1. Atomic units are used throughout this paper unless otherwise stated. The electric fields of the left circularly polarized laser pulse $E_l(t)$ and right circularly polarized laser pulse $E_r(t)$ have the forms :

$$E_l(t)=E_0 f_1(t)\,(\boldsymbol{e}_x\cos\omega_1 t+\boldsymbol{e}_y\sin\omega_1 t), \tag{10}$$

$$E_r(t)=E_0 f_2(t)\,(\boldsymbol{e}_x\cos\omega_2 t-\boldsymbol{e}_y\sin\omega_2 t), \tag{11}$$

here, $f_1(t)=f_2(t)=\exp[-2\ln 2(t/\tau)^2]$ and $E_0=0.1688$ a.u. is the peak amplitude, which corresponds to 1×10^{15} W/cm^2. The carrier frequency $\omega_1=0.1156$ a.u. (The unit a.u. is short for atomic unit.) ($\lambda=394$ nm) and $\omega_2=0.0569$ a.u. ($\lambda=800$ nm). $\tau=3$ fs is the full-width at half-maximum of the laser fields.

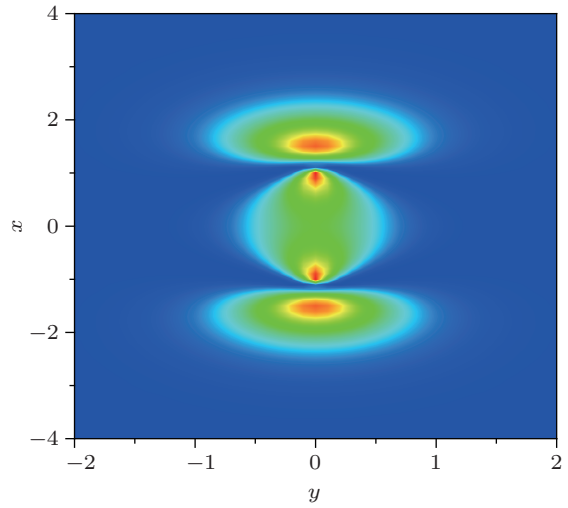


Fig. 1. (color online) The highest occupied molecular orbital (HOMO) of the N_2 molecule using the GAMESS-UK package.

Figure 2(a) shows the electric field of the left circularly polarized laser pulse (green solid line), which shows that the laser field spirals contra-clockwise along the time positive axis. The ionized electrons circularly move away and hardly recombine with the parent core since the electric field force is always tangent along the circumference. The blue dashed line is the projection of left circularly polarized laser pulse on the *x*–*y* plane. In this paper, the time (the unit on the *x* axis) is shown in optical cycle (O.C.) of the left circularly polarized laser pulse (394 nm in wavelength).

Figure 2(b) shows the electric field of the right circularly polarized laser pulse (green solid line) which circles clockwise spirally (800 nm in wavelength) along the time positive axis. The blue dashed line is the projection of right circularly polarized on the *x*–*y* plane.

Fig. 2(c) shows that the two-color circularly polarized laser field (the green solid line) changes are complicated and the projection on the *x*–*y* plane (the blue short dotted line) takes the shape of three-lobed distribution. It is clear to see that the electric field amplitude appears three peak values per laser cycle, which means that the ionized electrons can be driven back to the parent ion three times per laser cycle, each time at a different angle.

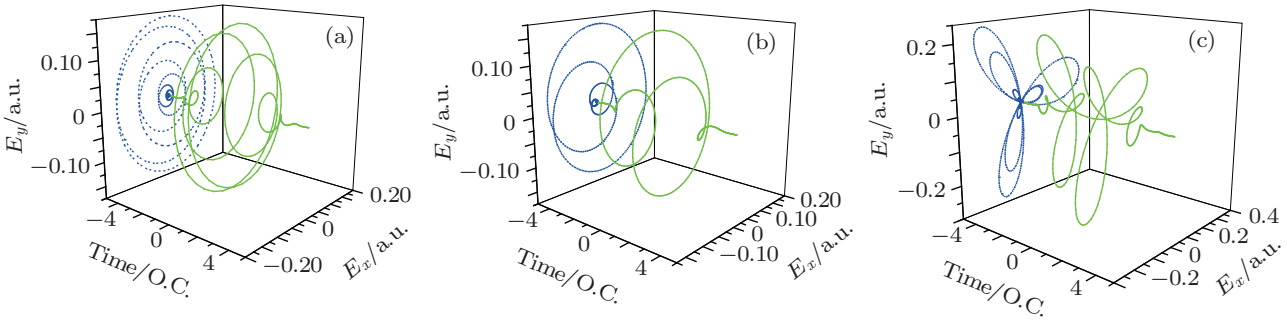


Fig. 2. (color online) (a) The electric field of the left circularly polarized laser pulse (green solid line) and the projection on the *x*–*y* plane (the blue dashed line); (b) The electric field of the right circularly polarized laser pulse (green solid line) and the projection on the *x*–*y* plane (the blue dashed line); (c) The electric field of the two-color circularly polarized laser fields (green solid line) and the projection on the *x*–*y* plane (the blue dashed line).

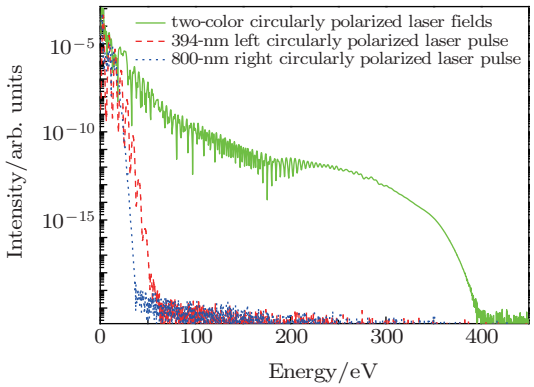


Fig. 3. (color online) Harmonic spectra of the N_2 molecule generated in the left circularly polarized laser pulse (red dashed line), the right circularly polarized laser pulse (blue dotted line) and two-color circularly polarized laser fields (green solid line).

Figure 3 shows the harmonic spectra of the N_2 molecule generated by the left circularly polarized laser pulse, the right circularly polarized laser pulse and two-color circularly polarized laser fields. It is clear to see that there is almost no harmonic generated by the left or right circularly polarized laser pulse. While in the case of the two-color circularly polarized laser fields, we can see that the plateau of the spectra is extended and a continuous spectrum is generated from 250 eV to 363 eV with a bandwidth of about 113 eV.

To explain the underlying mechanism of the extension of the HHG plateau, we demonstrate the emission time of the harmonics along the *x* and *y* directions in terms of the time–frequency analysis as shown in Figs. 4(a) and 4(b). It is shown that in both *x* and *y* directions there are five similar emission peaks, but the intensity of the emission peaks in the *x* direction is stronger than that in the *y* direction. Taking the emission time of the harmonics along the *x* direction for example, the maximum kinetic energy of the emission peak located around 0.24 O.C. reaches 363 eV, which corresponds to the cut-off energy of the spectrum. There is only one peak which makes a major contribution to the spectrum above 213 eV and the intensity of the short quantum path is much stronger than that of the long path. The destructive interference is weak,

which leads to a continuous spectrum. The other emission peaks make a comparable contribution to the harmonics below 213 eV, which results in modulation structures in the lower harmonic energy region. All the discussions above are in good agreement with the results shown in Fig. 3 (see the green solid line).

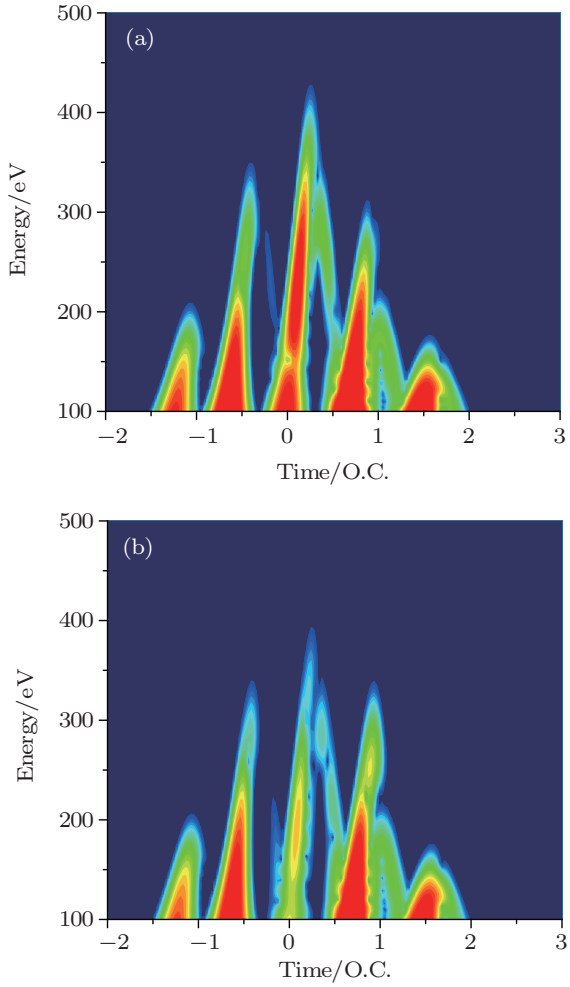


Fig. 4. (color online) The time–frequency analysis of the HHG spectrum along (a) x direction and (b) y direction in the two-color circularly polarized laser fields.

In order to extend the cut-off energy, we add a static electric field to the x direction and the expression is $E_{\text{static}} = \alpha E_0$, where α is the static parameter. We investigate the HHG by the two-color circularly polarized laser fields combined with the static electric field for $\alpha = 0.1$, $\alpha = 0.2$, and $\alpha = 0.3$. Figure 5 shows that in the lower energy region, the spectrums have little difference for three cases, but in the higher energy region, the plateau of the spectrum is dramatically extended with the increase of α. In the case of $\alpha = 0.3$, the spectrum becomes very smooth, in which a supercontinnum appears with a bandwidth of 350 eV from 250 eV–600 eV and finally it reaches the cut-off at 730 eV.

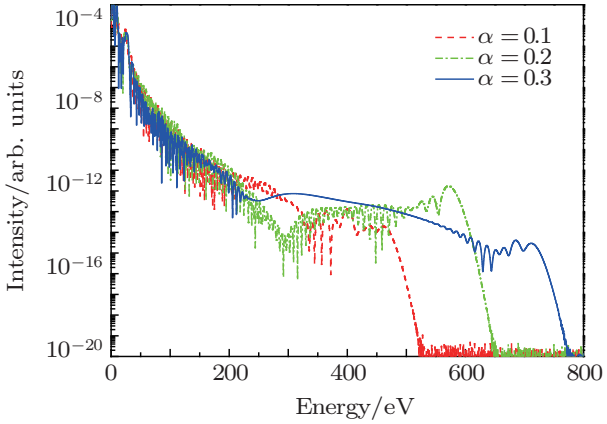


Fig. 5. (color online) Harmonic spectrums generated by the two-color circularly polarized laser fields combined with a static electric field for the static parameters $\alpha = 0.1$, $\alpha = 0.2$, and $\alpha = 0.3$, respectively.

As the intensity of the harmonics for the x component is stronger by adding a static field in the x direction, we focus on the HHG process along the x direction. We demonstrate the corresponding time–frequency analysis of the spectrums along the x direction for three cases in Fig. 6(a), 6(b), and 6(c). One can see that only one emission peak is generated at about 0.4 O.C. in the higher energy region for three cases. With the increase of α the maximum energy of the emission peak is increased. For $\alpha = 0.3$, the maximum energy of the emission peak reaches 730 eV and the long path is almost suppressed, which results in a supercontinumm spectrum in the higher energy region.

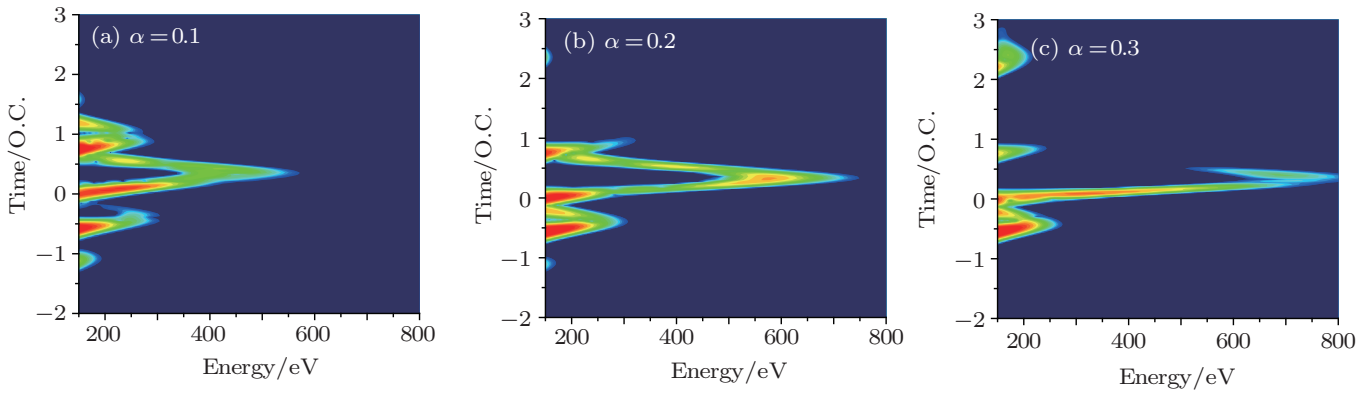


Fig. 6. (color online) The time–frequency analysis of harmonic spectrums along the x direction generated in two-color circularly polarized laser fields combined with a static field for (a) $\alpha = 0.1$, (b) $\alpha = 0.2$, and (c) $\alpha = 0.3$.

In order to further explain the mechanism of HHG in two-color circularly polarized laser fields combined with a static field for $\alpha = 0.3$, we demonstrate the semi-classical three-step model with a finite initial transverse velocity, which is shown in Fig. 7(a). Under 230 eV, there are many emission peak bursts, which may lead to destructive interference structures among the spectrum in the lower energy region. The electrons ionized around -1.38 O.C. returned at about 0.33 O.C. with the maximum energy of 730 eV and the short trajectory makes a major contribution to the harmonics above 230 eV. Thus a supercontinuum spectrum can be obtained. These results are in good agreement with the analysis in Fig. 5 and Fig. 6(c).

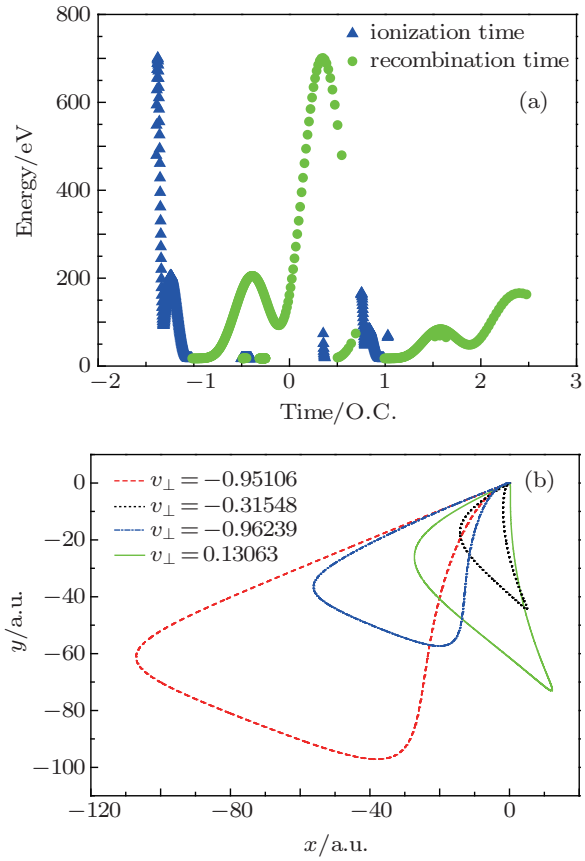


Fig. 7. (color online) (a) The dependence of the energy on the ionization (solid blue triangles) and emission times (solid green circles) in two-color circularly polarized laser fields combined with a static field for $\alpha = 0.3$; (b) The electron trajectories with finite initial transverse velocities.

The electron trajectories around -1.35 O.C. with proper initial transverse velocities are shown in Fig. 7(b). It is shown that the electron can return to the parent ion with proper transverse velocities and thereby contribute to the generation of harmonics. Figure 7(b) also shows that the freed electron travels in triangular paths. The electric field of the two-color circularly polarized laser fields appear three peak values per laser cycle as shown in Fig. 2(c), which makes the freed electron may move along the triangular trajectories.

Figure 8 shows the attosecond pulse generated by the two-color circularly polarized laser fields (the blue dashed line shown in Fig. 8(a)) and the two-color circularly polarized laser fields combined with a static electric field for $\alpha = 0.3$ (the green solid line shown in Fig. 8(b)). In the two-color circularly polarized laser fields, an IAP with the duration of 50 as is obtained by superposing several orders of the harmonics from 205 eV–283 eV, and with the addition of the static electric field, by superposing the harmonics from 245 eV–403 eV, the pulse width of the IAP's can be reduced to 30 as.

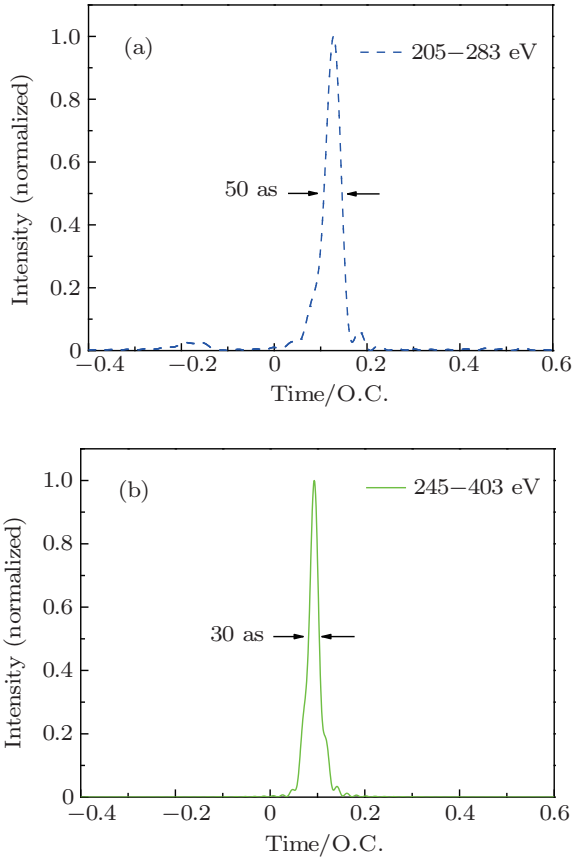


Fig. 8. (color online) The temporal profiles of the attosecond pulses generated by superposing several orders of the harmonics in (a) the two-color circularly polarized laser fields and (b) the combination of two-color circularly polarized laser fields and the static electric field for $\alpha = 0.3$.

4. Conclusions

We investigated the HHG and the attosecond pulse generation of the N_2 molecule in two-color circularly polarized laser fields by the strong-field Lewenstein model. We show that a continuous spectrum of 113 eV can be generated by the two-color circularly polarized laser fields. By adding a static electric field to the x direction, the plateau of harmonic spectra is extended and a supercontinuum spectrum is generated. Both the time–frequency analysis and the semi-classical three-step model show that a dominant short quantum path is selected to contribute the harmonics in the higher energy region. By superposing several orders of harmonics in the combination of two-color circularly polarized laser fields and a static electric field, an isolated attosecond pulse with a bandwidth of 30 as is

obtained directly.

References

[1] Cao X, Jiang S, Yu C, Wang Y, Bai L and Lu R 2014 *Opt. Express* **22** 26153
[2] Mohebbi M 2015 *Phys. Rev. A* **91** 023835
[3] Xue S, Du H C, Xia Y and Hu B T 2015 *Chin. Phys. B* **24** 054210
[4] Chou Y, Li P C, Ho T S and Chu S I 2015 *Phys. Rev. A* **91** 063408
[5] Xia C L and Liu X S 2013 *Phys. Rev. A* **87** 043406
[6] Bartels R A, Paul A, Green H, Kapteyn H C, Murnane M M, Backus S, *et al.* 2002 *Science* **297** 376
[7] Lewenstein M, Balcou P, Ivanov M Y, L'Huillier A and Corkum P B 1994 *Phys. Rev. A* **49** 2117
[8] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[9] Schafer K J, Yang B, DiMauro L F and Kulander K C 1993 *Phys. Rev. Lett.* **70** 1599
[10] Maurer J, Dimitrovski D, Christensen L, Madsen L B and Stapelfeldt H 2012 *Phys. Rev. Lett.* **109** 123001
[11] Goulielmakis E, Loh Z H, Wirth A, *et al.* 2010 *Nature* **466** 739
[12] Krausz F and Ivanov M 2009 *Rev. Mod. Phys.* **81** 163
[13] Zhang C P and Miao X Y 2015 *Chin. Phys. B* **24** 043302
[14] Zeng Z N, Li R X and Xu Z Z 2009 *Chin. Phys. Lett.* **26** 054202
[15] Zhu X, Zhang Q, Hong W, Lan P and Lu P 2011 *Opt. Express* **19** 438
[16] Kopold R, Becker W and Kleber M 1998 *Phys. Rev. A* **58** 4022
[17] Kamta G L and Bandrauk A D 2005 *Phys. Rev. A* **71** 053407
[18] Yuan K J, Bian X B, and Bandrauk A D 2014 *Phys. Rev. A* **90** 023407
[19] Itatanil J, Levesque J, Zeidler D, *et al.* 2004 *Nature* **432** 867
[20] Ge X L, Wang T, Guo J and Liu X S 2014 *Phys. Rev. A* **89** 023424
[21] Miao X Y and Du H N 2013 *Phys. Rev. A* **87** 053403
[22] Jin C, Bertrand J B, Lucchese R R, Wörner H J, Corkum P B, Villeneuve D M, Le A T and Lin C D 2012 *Phys. Rev. A* **85** 013405
[23] Guo J, Ge X L, Zhong H, Zhao X, Zhang M, Jiang Y and Liu X S 2014 *Phys. Rev. A* **90** 053410
[24] Pan Y, Zhao S F and Zhou X X 2013 *Phys. Rev. A* **87** 035805
[25] Milosevic D B, Becker W and Kopold R 2000 *Phys. Rev. A* **61** 063403
[26] Baer R, Neuhauser D, Ždánská P R and Moiseyev N 2003 *Phys. Rev. A* **68** 043406
[27] Mauger F, Bandrauk A D, Kamor A, Uzer T and Chandre C 2014 *J. Phys. B: At. Mol. Opt. Phys.* **47** 041001
[28] Chang Z 2004 *Phys. Rev. A* **70** 043802
[29] Yuan K J and Bandrauk A D 2013 *Phys. Rev. Lett.* **110** 023003
[30] Mancuso C A, Hickstein D D, Grychtol P, *et al.* 2015 *Phys. Rev. A* **91** 031402
[31] Zhao D and Li F L 2013 *Chin. Phys. B* **22** 064215
[32] Etches A and Madsen L B 2010 *J. Phys. B: At. Mol. Opt. Phys.* **43** 155602
[33] GAMESS-UK is a package of *ab initio* programs; Guest M F, Bush I J, Van Dam H J J and Sherwood P, *et al.* 2005 *Mol. Phys.* **103** 719
[34] Möller M, Cheng Y, Khan S D, Zhao B, *et al.* 2012 *Phys. Rev. A* **86** 011401
[35] Sansone G 2009 *Phys. Rev. A* **79** 053410
[36] Li Y, Zhu X, Hong W, Zhang Q, Qin M and Lu P 2013 *Opt. Express* **21** 4896