# Atomic high-order harmonic generation from a circularly polarized laser pulse*

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We demonstrate theoretically that the high-order harmonic of an atom can be generated by a circularly polarized laser pulse. The harmonic spectrum shows a clear cutoff with an energy $I_p + 2U_p$. In particular, the high-order harmonic generation comes from the multiple recombination of the ionized electron with non-zero initial velocity. These results are verified by the classical model theory and the time-frequency analysis of a harmonic spectrum.

**Keywords:** high-order harmonic generation, circularly polarized intense laser

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With the rapid development of the ultra-intense laser technology, some high-order nonlinear effects including the high-order harmonic generation (HHG), the above threshold ionization, and the laser-induced electron diffraction, have attracted extensive research in the laser physics and photochemistry field.[1–7]

The high-order harmonic can be emitted when an atom is irradiated by a strong linearly polarized laser pulse. The harmonic spectrum takes on a typical plateau structure with the increase of the harmonic energy and ends up at a cutoff energy $I_p + 3.17U_p$ ($I_p$ is the ionization energy of the atom and $U_p = E_0^2/4\omega^2$, $E_0$ and $\omega$ are the peak amplitude and frequency of the laser field, respectively). Due to the existence of the plateau structure, HHG from rare gases is the most promising way to obtain the coherent attosecond pulse and the source of soft X-ray.[8–11] Recently it has been successfully applied to the research of atomic and molecular structural dynamic imaging and chemical dynamic imaging.[12–14]

The physical mechanism of HHG from the atom driven by a linearly polarized laser pulse is well explained by the well-known "three-step" model: the electron first tunnels through the potential barrier formed by the atomic potential and the laser electric field, then obtains energy from the field and is driven back to its ion, and finally comes back to its initial state and simultaneously emits a harmonic photon.[15,16] However, HHG from the circularly polarized driving laser pulse is still an open question. According to the semiclassical "three-step" model, when the atom is irradiated by the circularly polarized laser pulses, the ionized electrons with zero initial velocity have no chance to return to the vicinity of the parent ion, thus there is no high-order harmonic to emit. Non-zero initial velocity can emit a harmonic as shown in an elliptically polarized field. From the perspective of the quantum transition, for the pure circularly polarized laser, the angular momentum component of the photon in the direction of propagation is $+\hbar$ (the left-handed light) or $-\hbar$ (the left-handed light), and the atom in the ground state jumps to the continuous state after absorbing $k$ photons, whose angular momentum will be increased or decreased by $k\hbar$. According to the transition selection rule $\Delta L = \pm 1$, if $k > 1$, the transition from the continuous state to the ground state cannot be realized, that is to say, HHG from the atom driven by the circularly polarized light cannot be achieved. However, a circularly polarized field pulse is not a pure circularly polarized field, therefore its HHG is worth studying. Because the molecular gases possess an additional recombination center, they become the ideal media for investigating HHG from the circularly or elliptically polarized laser pulse. There are a few pieces of research on HHG from the gaseous molecules irradiated by the circularly polarized pulse, and the research on HHG from the atom irradiated by the circularly polarized pulses has not been reported yet.[17–24]

In this work, we report a new mechanism of HHG from the atom irradiated by the ultrashort circularly polarized pulse. At the rising edge of the laser pulse, the atom is ionized by absorbing many photons, which makes the ionized electron have a non-zero initial velocity. The initial velocity can be small compared with that ionized at the peak of the pulse. The velocity of the ionized electron is non-zero, even in the tunneling regime, and it has a certain probability distribution, and non-zero component. This ionized electron has a chance to

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return back to the vicinity of the parent ion periodically and recombine with it to generate the harmonic photon, as shown in Fig. 1.

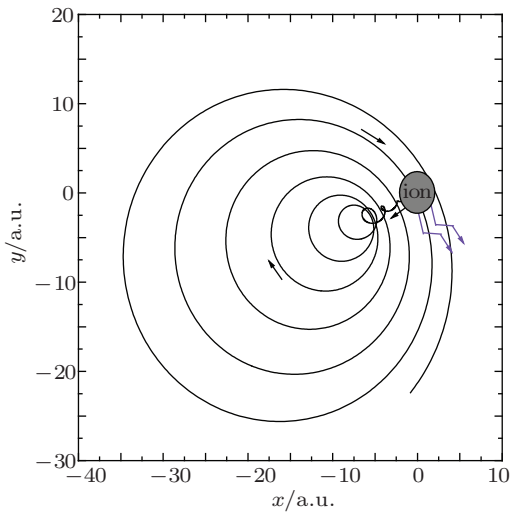



**Fig. 1.** Model for atomic HHG from a circularly polarized pulse (a.u. is the abbreviation to atomic unit).

In order to investigate HHG from the atom exposed to the circularly polarized laser pulse, we need to solve numerically the two-dimensional time-dependent Schrödinger equation (TDSE) under the dipole approximation (atomic units (a.u.) are used throughout the paper unless stated otherwise)

$$\begin{aligned} \mathrm{i}\frac{\partial}{\partial t}\psi(x,y,t) &= \left[-\frac{1}{2}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)+V_{\mathrm{a}}(x,y)\right. \\ &\quad +E_0 f(t)(x\cos(\omega t) \\ &\quad \left.+y\cos(\omega t+0.5\pi))\right]\psi(x,y,t), \end{aligned} \tag{1}$$

where the atomic potential is described by the soft-core Coulomb potential

$$V_{\mathrm{a}}(x,y) = -\frac{q}{\sqrt{x^2+y^2+a^2}}, \tag{2}$$

and the potential parameters are set as $q=1.5$, $a^2=0.6$, and thus the ground energy of the atom is 0.9. The envelope shape of the laser electric field is chosen as Gaussian form

$$f(t) = \mathrm{e}^{-4\ln(2)t^2/\tau^2},$$

in which $\tau$ is the full width at half maximum (FWHM) of the laser pulse and is set as 21 fs in this paper. $E_0$ and $\omega$ are the peak amplitude and angle frequency of the electric field, respectively. By using the Fourier split operator approach, one can obtain the time-dependent wave function $\psi(x,y,t)$ at instant $t$.[25]

Based on the time-dependent potential

$$\begin{aligned} V(x,y,t) &= V_{\mathrm{a}}(x,y)+xE_0 f(t)\cos(\omega t) \\ &\quad +yE_0 f(t)\cos(\omega t+0.5\pi), \end{aligned} \tag{3}$$

the power spectrum $P(\omega)$ of HHG is calculated by the Fourier transform of the expectation value of the dipole in acceleration form[26,27]

$$\begin{aligned} \langle a_x(t)\rangle &= \left\langle \frac{\mathrm{d}^2x(t)}{\mathrm{d}t^2}\right\rangle \\ &= \langle\psi(x,y,t)|-\nabla_x V(x,y,t)|\psi(x,y,t)\rangle, \\ \langle a_y(t)\rangle &= \left\langle \frac{\mathrm{d}^2y(t)}{\mathrm{d}t^2}\right\rangle \\ &= \langle\psi(x,y,t)|-\nabla_y V(x,y,t)|\psi(x,y,t)\rangle, \end{aligned} \tag{4}$$

$$P_x(\omega) = \left|\frac{1}{t_{\mathrm{f}}-t_{\mathrm{i}}}\frac{1}{\omega^2}\int_{t_{\mathrm{i}}}^{t_{\mathrm{f}}}\mathrm{e}^{-\mathrm{i}\omega t}\langle a_x(t)\rangle\,\mathrm{d}t\right|^2,$$

$$P_y(\omega) = \left|\frac{1}{t_{\mathrm{f}}-t_{\mathrm{i}}}\frac{1}{\omega^2}\int_{t_{\mathrm{i}}}^{t_{\mathrm{f}}}\mathrm{e}^{-\mathrm{i}\omega t}\langle a_y(t)\rangle\,\mathrm{d}t\right|^2. \tag{5}$$

In order to investigate the physical process of HHG, we perform a time-frequency analysis of the induced dipole moment by means of the wavelet transform:

$$A(t_0,\omega) = \int\langle \boldsymbol{a}(t)\rangle w_{t_0,\omega}(t)\,\mathrm{d}t, \tag{6}$$

with the wavelet kernel

$$w_{t_0,\omega} = \sqrt{\omega}W(\omega(t-t_0)).$$

For the harmonic emission, the mother wavelet is given by the Morlet wavelet

$$W(x) = \frac{1}{\sqrt{\sigma}}\mathrm{e}^{\mathrm{i}x}\mathrm{e}^{-x^2/2\sigma^2}, \tag{7}$$

where the characteristic width of the wavelet window is set as $\sigma=10$.

In Fig. 2, we present the HHG spectra (in $x$ and $y$ directions) generated from the model atom irradiated by a circularly polarized laser pulse in the length and acceleration forms, respectively. Here the peak amplitude and wavelength of the laser pulse are in turn set as 0.1 and 800 nm. It can be seen from Fig. 2 that even if the circularly polarized laser pulse is adopted, the harmonic spectrum still presents a similar structure as that from the atom irradiated by the linearly polarized laser pulse, i.e., there is an obvious plateau with a sharp cutoff position. The calculation results of the two different forms agree well in $x$ and $y$ directions. In addition, because the peak amplitude of the electric field in the $x$ and $y$ directions are the same, both the cutoff positions of harmonic spectra in the two different directions are the 43rd order. It is noticed that the 43rd harmonic of the cutoff position is smaller than the 58th harmonic generated from the linearly polarized laser pulse with the same peak amplitude. Using the strong field approximation (SFA) scheme,[16] we can obtain the same qualitatively consistent results as those calculated from the TDSE.

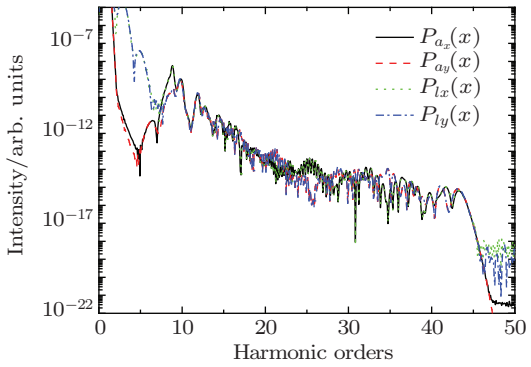



**Fig. 2.** (color online) HHG power spectra in $x$ and $y$ directions from an atom exposed to a circularly polarized laser pulse calculated by TDSE. The laser parameters are set as $E_0 = 0.1$ and $\omega = 0.057$.

In order to understand the difference of the high-order harmonic generated from the linearly polarized and circularly polarized laser pulses, we change the peak amplitude and frequency of the laser electric field. Our numerical simulation results demonstrate that the cutoff energy of HHG from the circularly polarized pulse also increases with the square of the peak amplitude of the electric field and decreases with the square of the central frequency of the laser field. As shown in Fig. 3, the harmonic spectra from different laser intensities are presented. When the peak amplitudes of the laser electric fields are 0.06, 0.08, 0.1, and 0.12, the corresponding cutoff energies of the HHG spectra are 24th, 33rd , 43rd, and 56th, respectively. It is found that the cutoff energies of the harmonic spectra at the different laser intensities follow the rule of $I_\mathrm{p} + 2U_\mathrm{p}$.

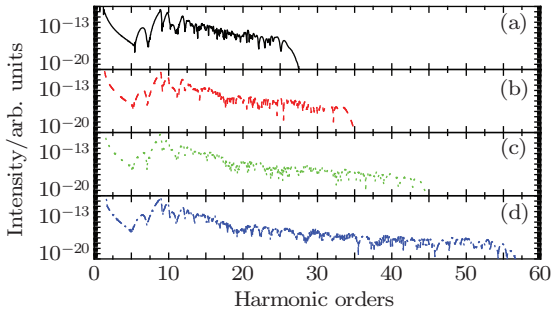



**Fig. 3.** (color online) The HHG in $x$ direction from the circularly polarized laser pulse at different peak amplitudes: (a) $E_0 = 0.06$; (b) $E_0 = 0.08$; (c) $E_0 = 0.1$; (d) $E_0 = 0.12$, respectively.

In view of the fact that the "three-step" model has successfully interpreted the high-order harmonic generation of the atom driven by the linearly polarized laser pulse, in this work, we will still utilize this model to interpret the mechanism of HHG of the atom irradiated by the circularly polarized laser pulse. When the electron is ionized, it will follow the classical Newton equation (neglecting the effect of the ion core):

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}x(t) = -E_x(t), \quad \frac{\mathrm{d}^2}{\mathrm{d}t^2}y(t) = -E_y(t), \tag{8}$$

$$v_x(t) = v_{x0} - \int_{t_0}^{t} E_x(t')\,\mathrm{d}t', \quad v_y(t) = v_{y0} - \int_{t_0}^{t} E_y(t')\,\mathrm{d}t', \tag{9}$$

$$x(t) = \int_{t_0}^{t} v_x(t')\,\mathrm{d}t', \quad y(t) = \int_{t_0}^{t} v_y(t')\,\mathrm{d}t'. \tag{10}$$

When the intensity of the laser pulse is high enough, the electron is not only ionized near the peak of the pulse, but also has the opportunity to be ionized at the leading part of the pulse. When the electron is ionized at the beginning of the pulse, the main ionization mechanism should be the multiphoton ionization. Thus, in the classical calculation, we consider the effect that the electron has non-zero initial velocity. As an example, in Fig. 4, we show the classical motion trajectory of the electron ionized at $t = 33$ a.u. in the circularly polarized laser field. The initial velocity of the ionized electron is chosen as $\boldsymbol{v}_0 = 0.02[\cos(1.1\pi)\hat{e}_x + \sin(1.1\pi)\hat{e}_y]$ in the calculation and other initial velocities present similar results. In the figure, one can see that the ionized electron circularly moves around the parent ion with the different radius. The radius is determined by the instant amplitude of the laser electric field. The ionized electron has many chances to come back to the parent ion and emit a harmonic in the process. When the electron recombines with the parent ion, it will emit a high energy photon, whose energy is the sum of the kinetic energy of the ionized electron and the ionization energy of the atom. In Fig. 5, we present the distance between the ionization electron and the ion core, and the kinetic energy of the ionized electron as a function of the recombination time $(R(t))$. In the figure there are six moments that the electron can return to the vicinity of the nucleus, and they are labeled as A, B, C, D, E, and F, respectively. The instant kinetic energy of the electron near the parent ion is changing and at the instant E the electron possesses the maximum kinetic energy $E_\mathrm{k} = 2U_\mathrm{p}$. According to the classical circumferential motion, the electric field force generates the centripetal force, i.e., $E = v\omega$, and the kinetic energy of the electron moving in the laser electric field is $E_\mathrm{k} = \frac{1}{2}v^2 = E^2/(2\omega^2)$. Using this formula, one can find that the maximum kinetic of the ionized electron circling around the parent core is $E_{\mathrm{k}\max} = 2U_\mathrm{p}$. It is obvious that

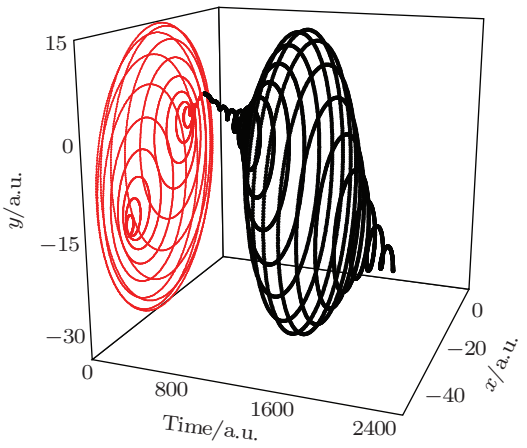



**Fig. 4.** (color online) Profile of electron trajectory as a function of time calculated from the classical calculation. The red line is the projection of the electron trajectory (black line) on the $x$–$y$ plane.

the photons with the maximal energy $I_p + 2U_p$ will be emitted when the ionized electron with the kinetic energy $2U_p$ recombines with the ion core.

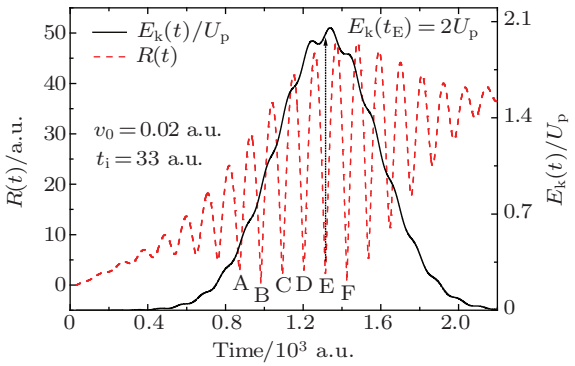



**Fig. 5.** (color online) Evolution of the kinetic energy (solid black line) and distance between the parent ion (dashed red line) as a function of recombination time for an electron ionized at instant 33 with initial velocity $v_0 = 0.02$.

In order to understand the dynamic process of the atomic HHG from the circularly polarized laser pulse, the time frequency analysis of HHG is preformed. As shown in Fig. 6, there is an obvious emission band for each harmonic at the harmonic emission profile, rather than two or more quantum paths (including the long and the short one) in the case of the linearly polarized condition. The emission profile can be understood by the model below: the ionized electron that possesses certain initial velocity will move periodically in the vicinity of the nucleus, and it has the chance to recombine with the nucleus and emit photons. The black curve in the figure denotes the energy of the harmonic photon calculated by the "three-step" model. It agrees well with the quantum calculation.

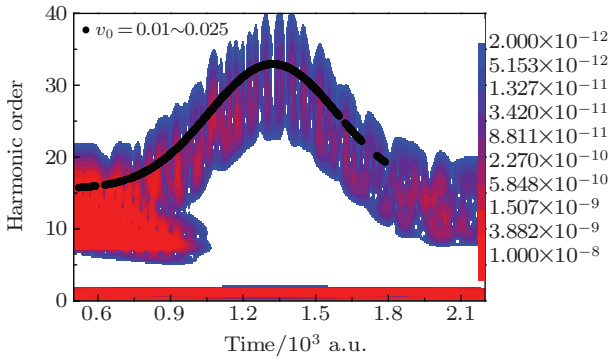



**Fig. 6.** (color online) Time-frequency profile of the time-dependent dipole (the laser parameter is the same as the HHG in Fig. 2). The black cycle is the result from the classical calculation with initial velocities $v_0 = 0.01 \sim 0.025$.

Additionally, taking the left-handed circularly polarized laser pulse as an example, we analyze the atomic HHG from the ultrashort circularly polarized laser pulse from the view of quantum transition. The angular momentum of the photon is not merely $+\hbar$ any more, and there exists a part of $-\hbar$ photons with a small probability. Hence, HHG can be generated from the ultrashort circularly polarized pulse.

One can notice that conversion efficiency of HHG from the circularly polarized pulse is smaller than that from the linearly polarized pulse. The reason is that the ionization rate at the rising edge of the pulse is low. To overcome this difficulty, we propose adding an ultrashort high frequency laser pulse at the beginning of the circularly polarized pulse. The additional pulse can largely enhance the probability of the ionized electron with the non-zero initial velocity. The high frequency laser pulse is chosen as $E_0 = 0.02$ a.u., $\omega_0 = 0.741$ a.u., and $\tau = 14$ a.u. The electric field of the synthesized pulse is shown in the inset of Fig. 7. From the figure, one can see that the harmonic emission intensity (dashed red line) from the synthesized pulse is higher than that from the monochrome pulse one (solid black line) by three orders of magnitude. It provides a new way to generate an isolated attosecond pulse using the multi-cycles laser pulse with controlling the parameters of the synthesized pulse. Due to the fact that the energy of HHG is determined by the initial velocity of the ionized electron, one can diagnose directly the initial momentum distribution of the ionized electron using HHG in the circularly polarized pulse.

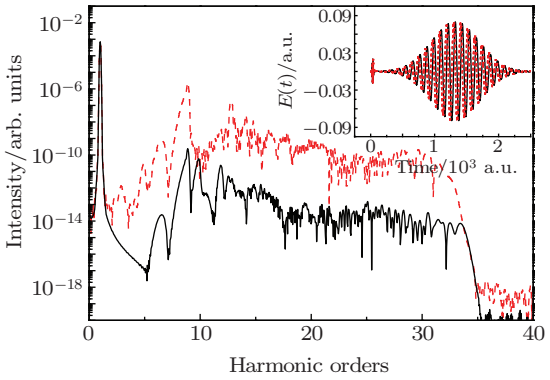



**Fig. 7.** (color online) High harmonic spectra from the fundamental frequency laser pulse (black solid line) and synthesized pulse (dashed red line). The synthesized laser field is shown in the inset.

In conclusion, we numerically calculated the HHG spectra from an atom irradiated by an ultrashort circularly polarized laser pulse. The spectra have a plateau structure that end at the cutoff energy $I_p + 2U_p$. The physical origin of HHG is the multiple recombination of the ionized electron with the non-zero velocity appearing at the beginning of the pulse. The harmonic intensity is largely enhanced by synthesizing a high frequency laser pulse with the low frequency circularly polarized pulse.

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