2D semiclassical model for high harmonic generation from gas

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Abstract The electron behavior in laser field is described in detail. Based on the 1D semiclassical model, a 2D semiclassical model is proposed analytically using 3D DC-tunneling ionization theory. Lots of harmonic features are explained by this model, including the analytical demonstration of the maximum electron energy 3.17 U_p. Finally, some experimental phenomena such as the increase of the cutoff harmonic energy with the decrease of pulse duration and the "anomalous" fluctuations in the cutoff region are explained by this model.

Keywords: **high harmonic generation, tunneling ionization, ultrashort laser pulse.**

High harmonics generation (HHG) is an intriguing and experimentally well-confirmed phenomenon which results from ionization of gas atom by laser field. HHG has been studied in the simple but illustrative models including classical models[1], semiclassical models[2] and quantum models[3,4]. The semiclassical model put forward by Corkum and Kulander et al. explains the emission of high harmonic radiation in terms of three discrete steps: First, the electron tunnels through the barrier formed by the atomic Coulomb potential and the laser field; the quasi-free electron subsequently acquires kinetic energy from the laser field; and half an optical cycle later, it returns to its parent ion and emits a photon. But the Corkum model has not considered the effect of magnetic field of the laser and the laser pulse duration which is essential in ultrashort laser-atom interactions[5,6]. So, it is necessary to adjust this model in ultrashort laser-atom interactions. This paper is the extended 2D model which can be used in the larger range.

1 Theoretical model

During the laser-atom interactions, the atom will ionize in the laser field. When the Keldysh parameter $\gamma > 1$, the ionization is multiphoton, whereas $\gamma < 1$ corresponds to the tunneling regime. The key difference between them is the comparison of electron barrier transition time and the laser optical cycle while the tunneling time is decreased with laser intensity.

For atom tunneling ionization rate, there are several equations. Augst[7] checked the experimental results and theoretical predictions, and found them fitted well with each other in the intensity of 10^{16} W/cm^2 except the Szöke's equations. So, we can use the three-dimensional DC-tunneling theory which is not time-averaged when we want to calculate the behavior of electron in laser field. That is

$$P(Z,t) = 4W_a\left(\frac{E_i^{5/2}}{|E_L|}\right)\exp\left[-\frac{2}{3}\frac{E_i^{3/2}}{|E_L|}\right], \tag{1}$$

where $W_a = 4\times10^{16}\ \mathrm{s}^{-1}$ is the atomic frequency unit; $E_i = E_a/E_H$ is the ratio of the ionization potential of the atom we studied and hydrogen. $E_L(Z,t) = E(\eta)/E_a = 1.16\times10^5(r_b/\lambda)a(\eta)$, E_a is the atomic unit of field. R_b is the Bohr radius. λ is the laser wavelength. $E(\eta)$ is the well-known solution of one-dimentional wave equation. Here the field is static, and no atomic shell dependence is included.

The evolution of electron density n is given by

$$\frac{\partial n}{\partial t} = (n_0 - n)P = Pn_0\exp\left(-\int p\,dt\right), \tag{2}$$

$$n = n_0\left[1 - \exp\left(-\int p\,dt\right)\right], \tag{3}$$

where n_0 is the initial gas density.

After ionization, the ion remains its position for its large mass. The motion of the ionized electron in a linearly polarized laser field $A = A(\eta)\hat{x}$ is governed by Lagrangian equations

$$\frac{d}{dt}\left(m\gamma u_x - \frac{e}{c}A_x\right) = 0, \tag{4}$$

$$\frac{d}{dt}(m\gamma u_z) = \frac{d}{dt}(m\gamma c) = -\frac{e}{c}\boldsymbol{u}\cdot\frac{\partial}{\partial n}\boldsymbol{A}. \tag{5}$$

In non-relativistic limit, we have

$$u_x/c - a = \alpha_1,\quad u_z/c - \frac{u^2}{2c^2} = \alpha_2, \tag{6}$$

where α_1 and α_2 are the constants to be determined by initial conditions. Since the electron has zero velocity when it is ionized, we get $\alpha_1 = -a_i$ and $\alpha_2 = 0$, where $a_i = a(z_i, t_i)$ is the laser strength at ionization, and then we get

$$u_x/c = a - a_i,\quad u_z/c = 1 - \sqrt{1-(a-a_i)^2}, \tag{7}$$

$$x = c\int_{t_i}^{t} a\,d\eta - ca_i(t - t_i), \tag{8}$$

$$z - z_i = c(t - t_i) - c\int_{t_i}^{t}\sqrt{1-(a-a_i)^2}\,d\eta. \tag{9}$$

The trajectory of the electron is then fully determined.

In general, a free electron quivering in laser field has very low probalility of collision with ions. For example, in an ionized gas with $Z = 50$, $n_0 = 10^{17}\ \mathrm{cm}^{-3}$, and temperature $T_e = 100$ eV, the electron-ion collision rate is merely $\gamma_{ei}/\omega \approx 4.8\times10^{-5}$. However, since the electrons are ionized at rest, their behavior in the initial stage is somewhat special. We will show that in the first half optical cycle after ionization an electron ionized by a linearly polarized light has rather good chance to be recombined. This interesting phonomenon is relevant to the high harmonic generation from rare gas.

Let us first consider a planar laser of infinite duration: $a(\eta) = a_0\sin(k\eta)$, where $k = \omega/c$. The trajectory is

$$\begin{cases} x = a_0[\cos t_i - \cos t - (t - t_i)\sin t_i], \\ z = \dfrac{1}{2}a_0^2\left[(t_i - t)\left(\dfrac{1}{2} + \sin^2 t_i\right) + \dfrac{3}{4}\sin(2t_i) - \cos t\left(2\sin t_i - \dfrac{1}{2}\sin t\right)\right]. \end{cases} \tag{10}$$

Fig. 1 presents the trajectories of electrons being ionized at different time: $t_i = -20°$ and $50°$, where $a_0 = 0.02$ and $z_i = 0$. In order to get a better view, an amplified scale is used for the axis of z. The electrons entering the area, like that being ionized at $t_i = 50°$, are considered to return to the vicinity of its parent ion (defined as Bohr radius $R_B = 3.3 \times 10^{-4}$). It is clear that these electrons will be recombined, with a photon of energy $E_\gamma = mu^2/2 + E_a$ emitted. Let us first consider the displacement in z direction after an optical cycle is

$$z_c = \pi a_0^2[0.5 + \sin^2 t_i] \geqslant \pi a_0^2/2. \tag{11}$$

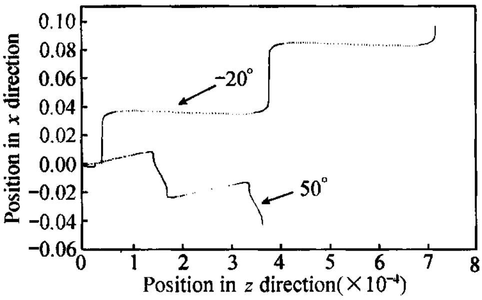

Fig. 1. The trajectory of electron driven by the same laser field with different ionization time.

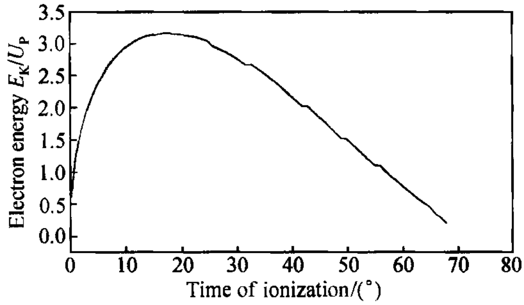

Fig. 2. The relation between electron energy and ionization time.

Electron will return to the vicinity of ion in the condition of $z_c < 2R_B$. We can get three important results:

(i) Electrons failed to return in the first half optical cycle have little chance to do so in their future flights. After the initial stage, an ionized electron behaves more and more like a quivering free electron. The probability of collision with ions is then described by γ_{ei}/ω.

(ii) Because the effect of magnetic field was considered, a maximum laser intensity should be put forward in order to ensure that some electrons return into Bohr radius in an optical cycle. When laser intensity exceeds this value, no harmonic will be generated because the displacement in z axis is greater than the Bohr radius and the electron escapes from the parent ion. For example, this intensity is 6.04×10^{14} W·cm^{-2}($a_0 = 0.021$) for hydrogen in 800-nm wavelength if condition was satisfied. This intensity together with saturation intensity decide the maximum value for HHG.

(iii) Not all the electrons will return to parent ion and emit photon in an optical cycle as fig. 1 shows. From eq. (10) as $x = 0$, we can get the relation between ionization time and the recombination time. Electrons will go back to ions if they are ionized at 0°—70°. Another feature is that the recombination time is inversly proportional to the ionization time, i.e. the earlier the electrons are ionized, the longer they stay in the laser field.

Fig. 2 presents the kinetic energy of returned electron $E_k = mu^2/2$ as a function of ionization time, where $z_i = 0$, $a_0 = 0.01$. Since the field is periodical, this figure represents the electron behavior in each opticle cycle. Depending on the phase of the field at ionization, some electrons will go back with different kinetic energy and some will not. The electrons ionized at $t = 17°$ will have the maximum kinetic energy on their return, and the maximum energy happens to be $E_k/U_p = 3.17$. These are in agreement with Corkum's prediction. It is well known that the maximum kinetic energy is the physical origin of the $\hbar\gamma_{max} = 3.17U_p + E_a$ law for the high harmonic radiation cutoff. This important cutoff law can be proved to be true for all laser strengths: when the recombination time $x(t) = 0$, from eq. (8) we can get

$$\cos(t-\tau) = \cos t + \tau\sin(t-\tau), \tag{12}$$

where $\tau = t - t_i$, and then

$$\cos^2(t-\tau/2) = a^2(\tau)/[a^2(\tau)+b^2(\tau)], \tag{13}$$

where $a(\tau) = \dfrac{2\sin(\tau/2)}{\tau} - \cos(\tau/2)$, $b(\tau) = \sin(\tau/2)$.

So, the generated photon energy is

$$E_{\text{kin}} = \frac{1}{2}v^2 = 8U_p\frac{a^2(\tau)b^2(\tau)}{a^2(\tau)+b^2(\tau)}. \tag{14}$$

For $\dfrac{dE_{\text{kin}}}{d\tau} = 0$ while the electrons have the maximum energy, one can get $\tau = 4.12$. So one can get $t = 4.42$ from eq. (9) and then $E_{\max} = 3.17\ U_p$. The corresponding ionized time is $t_i = t - \tau = 17°$.

When electrons are ionized by circularly polarized laser, the velocity and displacement of the electron in z axis are

$$u_z = a_0(\cos t_i - \cos t), \tag{15}$$

$$z = a_0[(t-t_i)\cos t_i - \sin t + \sin t_i]. \tag{16}$$

The displacement in the z axis is the same as that in the x axis. One can then get the expression of the displacement of electron from parent ion:

$$L^2 = a_0^2\{(t-t_i)^2 + 2 - 2\cos(t-t_i) + 2(t-t_i)[\sin(2t_i) - \sin(t+t_i)]\}. \tag{17}$$

It is always greater than the Bohr radius. One can also get the recombination electron energy: $E_k = 8\ U_p\sin^2[(t-t_i)/2]$. It is greater than the electrons heated by linearly polarized lasers.

So, one can conclude that the electrons heated by circularly polarized laser never return to the parent ion and emit photons. That is fit for experimental results.

Then we consider a Gaussian pulse of duration τ:

$$a(\eta) = a_0\sin(k\eta)\exp\left[-\frac{(n-L/2)^2}{L^2}\right], \tag{18}$$

where $L = c\tau$. The motion of ionized electrons can still be described by eqs. (4)—(9). The recombination of electrons is associated with photon radiation. And the power of radiation is given by

$$P = \left(\frac{1}{2}mu^2 + Ea\right)\frac{dn}{dt}. \tag{19}$$

Note that time delay must be accounted: the radiation power is of recombination time. Using Larmor formula we get the power radiated per unit solid angle

$$\frac{dP}{d\Omega} = \frac{3}{8\pi}\sin^2\theta P, \tag{20}$$

where θ is measured from the direction of polarization. At $\theta = \pi/2$,

$$\frac{dP}{d\Omega} = \frac{3}{8\pi}P = |G(t)|^2. \tag{21}$$

The Fourier transform $G(\omega)$ of $G(t)$ gives the frequency spectrum

$$\frac{d^2I}{d\Omega} = 2|G(w)|^2. \tag{22}$$

2 Results and discussion

We can use this 2D semiclassical model to explain the effect of high harmonic with pulse duration. Fig. 3 shows the emitted photon frequency spectrum produced by 50 fs laser pulse interacted with He gas atom, where $a = 0.01$. One can get from the figure that it is odd times wave frequencies, and the maximum order is 53 (choosing the magnitude drop to 4 orders as the cutoff).

In order to increase the cutoff energy of harmonics, higher laser intensity must be used to increase the electrons energy and then increase the emitted photon energy under the same conditions. But the laser intensity cannot increase to the saturation intensity; i.e. there is a saturation value for a certain gas density, a certain laser pulse duration and a certain species gas atom. Beyond this value, most of the gas atoms ionized and the intensity of harmonics will minimize to zero. From eq. (3), one can get the time-dependence ionization rate curve (as fig. 4 shows). One can conclude that the saturate intensity increases with the decrease of laser duration. We repeat the calculation and get the time spectrum and frequency spectrum of He with 5 fs duration and $a = 0.02$, as fig. 5 shows. One can get that the harmonics order increases rapidly over 100, the cutoff order is beyond 250.

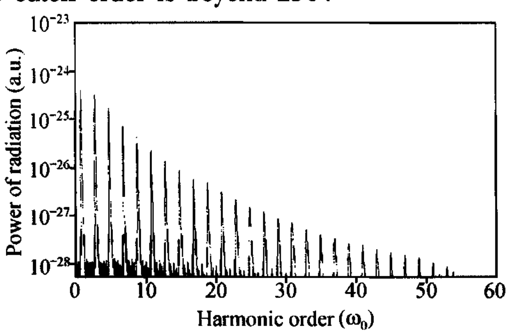

Fig. 3. The frequency spectrum of harmonics produced by laser with 50 fs pulse duration. The highest harmonic order is 53 ($a_0 = 0.01$).

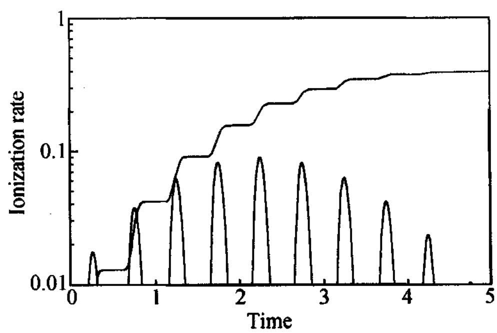

Fig. 4. The ionization rate of the atom generated by ultrashort intense pulse.

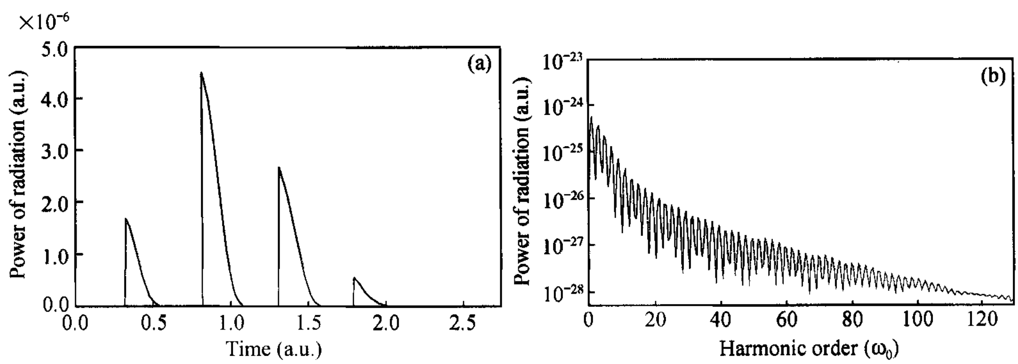

Fig. 5. The time spectrum (a) and frequency spectrum (b) of high harmonics with 5 fs pulse duration interacted with He. The harmonic order is over 100 ($a_0 = 0.02$).

For the harmonic cutoff energy is proportional to the laser saturate intensity and ionization potential, the cutoff energy will drop if we choose Xe noble gas for its less saturate intensity and ionization potential. Fig. 6 shows the frequency spectrum of Xe, where $a = 0.02$ and at 5 fs pulse duration just as fig. 5. Compared with the frequency spectrum of He gas, the harmonic orders of Xe gas decrease rapidly.

Finally, our calculation can explain the large fluctuations observed experimentally[8] in the

X-ray intensity at wavelength < 10 nm in spite of the relatively small energy fluctuation of the 5 fs pulse (5%). Fig. 7 shows the extreme ultraviolet (XUV) harmonic emission at the highest energies emitted for the same parameters as fig. 5 for initial laser phase of 0° and 90°. The strong phase dependence of the XUV yield near cutoff region translates into corresponding fluctuation of the XUV emission produced by non-phase-stabilized pulses. For the laser initial phase is not controlled and hence cannot be kept constant in our laser pulses currently, so this anomalous fluctuation in the cutoff region is unavoidable until the carrier phase can be controlled.

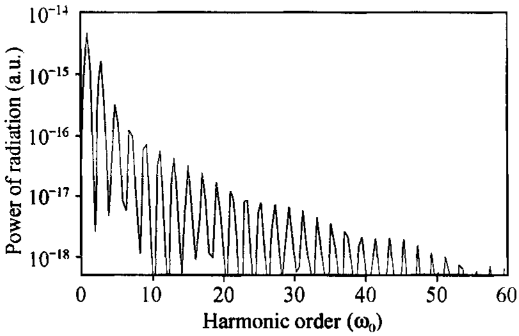

Fig. 6. The frequency spectrum of harmonics produced by 5 fs pulse interacted with Xe. The harmonics order is about 59.

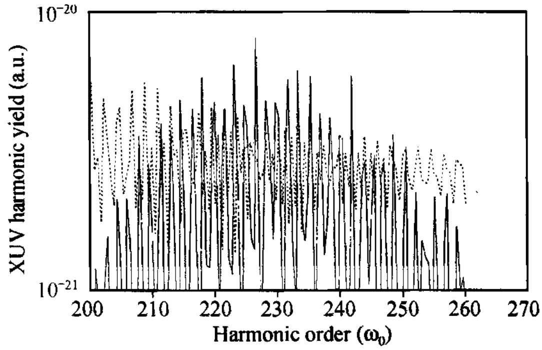

Fig. 7. XUV harmonic emission spectra at the highest photon energies in sine (dotted line) and cosine (solid line) carrier in the same laser intensity with 5 fs pulse duration.

3 Conclusions

The electron behavior in laser field is described in detail in this paper. Based on the 1D semiclassical model, a 2D semiclassical model is proposed analytically using 3D DC-tunneling ionization theory. Lots of harmonic features are explained by this model, including the analytical demonstration of the maximum electron energy 3.17 U_p. Finally, some experimental phenomena such as the increase of cutoff energy with the decrease of laser duration and the "anomalous" fluctuations in the cutoff region can be explained by this model.

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