Classical calculation of high-order harmonic generation of atomic and molecular gases in intense laser fields

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Abstract

Based upon our previous works (Eur.Phys.J.D 6, 319(1999); Chin.Phys.Lett. 18, 236(2001)), we develop a classical approach to calculate the high-order harmonic generation of the laser driven atoms and molecules. The Coulomb singularities in the system have been removed by a regularization procedure. Action-angle variables have been used to generate the initial microcanonical distribution which satisfies the inversion symmetry of the system. The numerical simulation show, within a proper laser intensity, a harmonic plateau with only odd harmonics appears. At higher intensities, the spectra become noisier because of the existence of chaos. With further increase in laser intensity, ionization takes place, and the high-order harmonics disappear. Thus chaos introduces noise in the spectra, and ionization suppresses the harmonic

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generation, with the onset of the ionization follows the onset of chaos.

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I. INTRODUCTION

The development of the high-power femtosecond laser has stimulated the investigation of the multi-photon processes of atoms and molecules interacting with intense laser fields [1-9]. Recently, there are many theoretical and experimental references about these multiphoton process. Within a proper intensity region, lots of odd harmonics of laser are generated by atomic and molecular gases [1-5,14]. The harmonic structure distributes as a plateau which is cut off at a special high-order harmonic. When the intensity increases, the ionization channel is opened and the high-order harmonics disappear. The above threshold ionization (ATI) occurs when the laser intensity is sufficient power (above $10^{13} \text{W/cm}^2$). Various structures (plateau, angular distribution, etc.) in ATI spectra have been detailed in Ref.[6-8]. The above threshold dissociation (ATD) and dissociation-ionization of molecular systems are also reported [9-10]. Studying the laser-matter interaction deeply, not only can obtain new knowledge of the interacting mechanism, but also can provide widely application in the generation of high-order coherent harmonics, X-rays laser and γ-rays laser.

The classical dynamics of most laser-driven systems is generally chaotic, due to the existence of nonlinearity. Chaos usually manifests itself as some control parameters (initial energy, laser intensity, laser frequency, etc.) are varied. The microscopic systems, in particular those involving atoms and molecules, are governed by a Hamiltonian. To study these systems are of great importance in the context of quantum-classical correspondence. At the investigated high power laser intensity ($10^{13}-10^{15} \text{W/cm}^2$), the electric field of the laser is equal in strength to the Coulomb field of the nuclei [8], and the laser field can not be looked as a perturbation to the field-free system, for the states themselves are no longer independent of the laser. Generally, the microscopic systems (atoms or molecules) are intrinsically quantum mechanical systems, thus they must be described by quantum mechanics. However, due to the presence of intense laser, the exact calculation even numerical simulation based upon quantum mechanics tend to be very difficult to perform. Fortunately, classical approach to these similar problems is useful for providing physical insight into dynamics

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processes [10-13]. Classical chaos associating with the microwave ionization of atomic hydrogen reveals that the detailed mechanism of atomic ionization in terms of transport in phase space [11-12]. Classical prediction for scaled frequencies has been verified by quantum calculations [12-13], and, in turn, has confirmed the dynamical significance of classical chaos. The harmonic generation (HG) of laser driven hydrogen atoms is simulated with classical Monte-Carlo method [14], the numerical results are qualitatively consistent with the quantum mechanic results and the experimental observation. For the molecular systems, the classical calculation is a good first step, since length and energy scales are often large enough for classical mechanics to be at least approximately valid. Using softened potential model, the classical dynamics of the one-dimensional hydrogen molecular ion $H_2^+$ interacting with an intense laser pulse are detailed [15-16].

There are two important and realistic examples of laser driven systems, where nonlinear dynamics has played a major role, which have been traditionally studied. One is the laser driven hydrogen atom that is the fundamental system in atomic physics, the other is the laser driven hydrogen molecular ion $H_2^+$, which is the fundamental diatomic molecular system in molecular physics. Bellow, we shall show our classical calculation of the high-order harmonic generation of these two fundamental systems. The outline of this paper is as follows. The regularized model and the corresponding initial microcanonical distribution are presented in the next section. In section III, we show the numerical results in details. A briefly summary and discussion is given out in the last section.

II. MODEL AND INITIAL MICROCANONICAL DISTRIBUTION

A. Regularized model of laser driven hydrogen atom

In this article, we consider a classical hydrogen atom, with an infinite mass nucleus fixed at the origin of coordinates, interacting with a high intense laser field which is linearly polarized along the $z$ – axis, and with the electric field component $\varepsilon(t)$. Thus the Hamiltonian
in atomic units in Cartesian coordinates is

\[ H = H_0 + H_i, \]
\[ H_0 = \frac{1}{2} p^2 - \frac{1}{r}, \quad H_i = -z \varepsilon(t). \]

where, \( r = \sqrt{x^2 + y^2 + z^2} \) and \( p^2 = p_x^2 + p_y^2 + p_z^2 \). Apparently, the above Hamiltonian has a Coulomb singularity corresponding to electron-nucleus collision. To remedy the singularity, we introduce the parabolic coordinates \((u, v, \phi)\)

\[ x = uv \cos \phi, \quad y = uv \sin \phi, \quad z = (v^2 - u^2)/2. \]

and a new fictive time scale \( \tau \)

\[ dt/d\tau = G(u, v) = v^2 + u^2. \]

In order to implement regularization, following the notation of Szebehely [17], regarding the motion time \( t \) and the negative total energy \(-E\) as generalized coordinate and generalized momentum respectively, then the Hamiltonian function in extended space becomes into

\[ H^* = H - E \equiv 0 = H^*(x, y, z, t; p_x, p_y, p_z, -E) \]

For the sake of obtaining the regularized Hamiltonian, we introduce the third category generating function \( F_3(p_x, p_y, p_z, -E; u, v, \phi, t) \), then obtain

\[ x(u, v, \phi) = -\frac{\partial F_3}{\partial p_x}, \quad y(u, v, \phi) = -\frac{\partial F_3}{\partial p_y}, \quad z(u, v, \phi) = -\frac{\partial F_3}{\partial p_z}, \quad t(u, v, \phi) = -\frac{\partial F_3}{\partial E}. \]

So the third category generating function \( F_3(p_x, p_y, p_z, -E; u, v, \phi, t) \) is in the form of

\[ F_3 = -xp_x - yp_y - zp_z + Et, \]
\[ = -uv \cos \phi p_x - uv \sin \phi p_y - \frac{1}{2}(v^2 - u^2)p_z + Et. \]

The new momenta \((p_u, p_v, p_\phi, -E)\) are related with old momenta \((p_x, p_y, p_z, -E)\) by

\[ p_u = -\frac{\partial F_3}{\partial u} = v \cos \phi p_x + v \sin \phi p_y - up_z, \]
\[ p_v = -\frac{\partial F_3}{\partial v} = u \cos \phi p_x + u \sin \phi p_y - vp_z, \]
\[ p_\phi = -\frac{\partial F_3}{\partial \phi} = -uv \sin \phi p_x + uv \cos \phi p_y, \]
\[ E = \frac{\partial F_3}{\partial t} = E. \]
Then the regularized Hamiltonian can be expressed as

\[ K = \frac{dt}{d\tau} H^* = G(u, v)(H - E) \equiv 0, \]

\[ = \frac{1}{2}[p_u^2 + p_v^2 + (u^{-2} + v^{-2})p_{\phi}^2] - 2 - E(u^2 + v^2) - \frac{1}{2}(v^4 - u^4)\varepsilon(t). \quad (8) \]

If we define

\[ K_u = \frac{1}{2}[p_u^2 + u^{-2}p_{\phi}^2] - 1 - Eu^2 + \frac{1}{2}u^4\varepsilon(t), \]

\[ K_v = \frac{1}{2}[p_v^2 + v^{-2}p_{\phi}^2] - 1 - Ev^2 - \frac{1}{2}v^4\varepsilon(t). \quad (9) \]

In the field-free case, \( \varepsilon(t) = 0 \), \( K_u = -K_v \) is the component of the Laplace-Runge-Lenz vector along \( z \)-axis, i.e., \( K_u = -K_v = R_\varepsilon \). The equations of motion can be derived from the above regularized Hamiltonian, as the following:

\[ \frac{d\phi}{d\tau} = \frac{\partial K}{\partial p_\phi} = p_\phi(u^{-2} + v^{-2}), \quad \frac{dp_\phi}{d\tau} = -\frac{\partial K}{\partial \phi} = 0, \quad (10) \]

\[ \frac{du}{d\tau} = \frac{\partial K}{\partial p_u} = p_u, \quad \frac{dp_u}{d\tau} = -\frac{\partial K}{\partial u} = 2Eu + p_{\phi}^2u^{-3} - 2u^3\varepsilon(t), \quad (11) \]

\[ \frac{dv}{d\tau} = \frac{\partial K}{\partial p_v} = p_v, \quad \frac{dp_v}{d\tau} = -\frac{\partial K}{\partial v} = 2Ev + p_{\phi}^2v^{-3} + 2v^3\varepsilon(t), \quad (12) \]

and

\[ \frac{dE}{d\tau} = \frac{\partial K}{\partial t} = \frac{1}{2}(u^4 - v^4)\frac{d\varepsilon(t)}{dt}, \quad \frac{dt}{d\tau} = -\frac{\partial K}{\partial E} = G(u, v). \quad (13) \]

Apparently, \( p_\phi \) is a constant which corresponding to the component of angular momentum along \( z \)-axis.

**B. Regularized model of laser driven hydrogen molecular ion \( H_2^+ \)**

Within Born-Oppenheimer approximation, we can assume that two protons are fixed at the positions \( A \) and \( B \), with a distance \( R \) away from each other, in the locations \((0, 0, -R/2)\) and \((0, 0, R/2)\) of the Cartesian coordinates system. A single electron at \((x, y, z)\) is subject
to the Coulomb attraction of both protons and the interaction of the laser. Let \( r_A \) be its
distance from \( A \), and \( r_B \) be its distance from \( B \). Within the dipole approximation, the
Hamiltonian in atomic units is

\[
H = H_0 + H_i,
\]

\[
H_0 = \frac{1}{2}p^2 - \frac{1}{r_A} - \frac{1}{r_B},
\]

\[
H_i = -ze(t).
\]

(14)

where, \( \varepsilon(t) \) is the electric field of the laser pulse, \( H_i \) is the interacting Hamiltonian. Ap-
parently, the above Hamiltonian has singularities at points \( r_A = 0 \) and \( r_B = 0 \), which
 corresponds to electron-proton collision. To overcome this barrier, regularization has to be
performed. Define the new coordinates \((u, v, \phi)\) as

\[
x = -\frac{R}{2} \sin u \sinh v \cos \phi, \\
y = -\frac{R}{2} \sin u \sinh v \sin \phi, \\
z = \frac{R}{2} \cos u \cosh v.
\]

(15)

and introduce the new fictive time scale \( \tau \) satisfying

\[
dt/d\tau = G(u, v) = r_A r_B = R^2 (\cosh^2 v - \cos^2 u)/4.
\]

(16)

Similar to the previous subsection, regarding the motion time \( t \) and the negative total energy
\( -E \) as generalized coordinate and generalized momentum respectively, then we can write
the Hamiltonian function in extended space as

\[
H^* = H - E \equiv 0 = H^*(x, y, z, t; p_x, p_y, p_z, -E)
\]

(17)

Introducing the third category generating function \( F_3(p_x, p_y, p_z, -E; u, v, \phi, t) \), which satisfies

\[
F_3 = -\frac{R}{2} \sin u \sinh v \cos \phi p_x - \frac{R}{2} \sin u \sinh v \sin \phi p_y - \frac{R}{2} \cos u \cosh v p_z + Et.
\]

(18)

Thus momenta \((p_u, p_v, p_\phi, -E)\) are related with old momenta \((p_x, p_y, p_z, -E)\) by

\[
p_u = -\frac{\partial F_3}{\partial u} = \frac{R}{2} \cos u \sinh v \cos \phi p_x + \frac{R}{2} \cos u \sinh v \sin \phi p_y - \frac{R}{2} \sin u \sinh v p_z,
\]

\[
p_v = -\frac{\partial F_3}{\partial v} = \frac{R}{2} \sin u \cosh v \cos \phi p_x + \frac{R}{2} \sin u \cosh v \sin \phi p_y - \frac{R}{2} \sin u \cosh v p_z,
\]

\[
p_\phi = -\frac{\partial F_3}{\partial \phi} = -\frac{R}{2} \sin u \sinh v \sin \phi p_x + \frac{R}{2} \sin u \sinh v \cos \phi p_y,
\]

\[
E = \frac{\partial F_3}{\partial t} = E.
\]

(19)
And the regularized Hamiltonian is
\[
K = \frac{dt}{d\tau} H^* = G(u, v)(H - E) \equiv 0, \\
= \frac{1}{2} [p_u^2 + p_v^2 + (\csc^2 u + \csc h^2 v)p_\phi^2] - R \cosh v - \frac{(E - H_i)}{4} R^2 (\cosh^2 v - \cos^2 u). \tag{20}
\]
where, \( H_i = -z \varepsilon(t) = (-R/2) \cos u \cosh v \varepsilon(t) \). Obviously, \( p_\phi \) is a constant, which corresponds to the component of the angular momentum which along the \( z-axis \). If \( p_\phi \) is equal to zero, the electron is constrained in a plane which can be chosen as \( y = 0 \), corresponding to a two-dimensional motion. In the field-free case, \( \varepsilon(t) = 0 \), this two-dimensional model corresponds to the classical hydrogen molecular ion in ground-state. Equations of the motion for the ground-state hydrogen molecular ion interacting with laser field can be easily obtained from the regularized Hamiltonian, i.e.,
\[
\frac{du}{d\tau} = \frac{\partial K}{\partial p_u} = p_u, \\
\frac{dp_u}{d\tau} = -\frac{\partial K}{\partial u} = \frac{ER^2}{4} \sin(2u) + \varepsilon(t)(z \frac{\partial G}{\partial u} + G \frac{\partial z}{\partial u}), \tag{21}
\]
\[
\frac{dv}{d\tau} = \frac{\partial K}{\partial p_v} = p_v, \\
\frac{dp_v}{d\tau} = -\frac{\partial K}{\partial v} = R \sinh v + \frac{ER^2}{4} \sinh(2v) + \varepsilon(t)(z \frac{\partial G}{\partial v} + G \frac{\partial z}{\partial v}), \tag{22}
\]
and
\[
\frac{dE}{d\tau} = \frac{\partial K}{\partial t} = zG \frac{d\varepsilon(t)}{dt}, \frac{dt}{d\tau} = -\frac{\partial K}{\partial E} = G(u, v). \tag{23}
\]
In the field-free case, defining
\[
K_u = \frac{1}{2} (p_u^2 + p_\phi^2 \csc^2 u) + \frac{ER^2}{4} \cos^2 u, \\
K_v = \frac{1}{2} (p_v^2 + p_\phi^2 \csc h^2 v) - R \cosh v - \frac{ER^2}{4} \cosh^2 v. \tag{24}
\]
thus \( K_u(= -K_v) \) is a constant of the field-free motion, they are related to \( \gamma \) and \( \Omega \) by
\[
K_u = -K_v = -\gamma R^2/4 = ER^2/4 + \Omega/2. \tag{25}
\]
Constants $\Omega$ and $\gamma$ are first introduced by Erickson [18] and Strand [19] respectively, which have the following forms

$$
\gamma = -E - 2\Omega/(mR^2), \nonumber
$$

$$
\Omega = \mathbf{L}_A \cdot \mathbf{L}_B + emR^2(\cos \theta_A - \cos \theta_B). \tag{26}
$$

Here, $\mathbf{L}_A$ and $\mathbf{L}_B$ are the angular momentum vector of the motion around nucleus $A$ and $B$ respectively, $\theta_A$ and $\theta_B$ are the angle from the vector $\mathbf{r}_A$ and $\mathbf{r}_B$ to positive $z$-axis respectively, $m$ and $e$ are the mass and the charge of the electron respectively.

C. Action-angle variables and initial distributions

The action-angle variables for separated systems are defined as

$$
I_i = \frac{1}{2\pi} \oint p_i dq_i, \quad \theta_i = \frac{\partial}{\partial I_i} \int p_i dq_i = \int \frac{\partial p_i}{\partial H} \frac{\partial H}{\partial I_i} dq_i. \tag{27}
$$

The motion of the classical field-free hydrogen atom is periodic, then it need only a pair of conjugated action-angle variables [20]

$$
H_0 = E_0 = -\frac{1}{2I^2}. \tag{28}
$$

The corresponding angle is given by the Kepler’s equation

$$
\theta = u - e \sin u. \tag{29}
$$

This means $\theta$ is the mean anomaly of the free orbits. The eccentric anomaly $u$ can be obtained from $r = a(1 - e \cos u)$. Here, $r$ is the distance from the electron to the origin, $a$ is the instantaneous semimajor axis which satisfies $a = -1/(2E_0)$, $e$ is the eccentricity of the free orbits satisfying $e = \sqrt{2E_0L^2 + 1}$, $L$ is the total angular momentum.

The motion of the ground-state hydrogen molecular ion $H_2^+$ is quasi-periodic, it need two pairs of action-angle variables. With elliptic integrals, action $I_u$ can be expressed as follows.
\[
I_u = \begin{cases} 
\sqrt{8K_u - 2E_0R^2} F_1\left(\frac{\pi}{2}, \sqrt{1 + \frac{4K_u}{E_0R^2-4K_u}}\right), & \text{for } K_u > 0, \\
\sqrt{-E_0R^2/\pi}, & \text{for } K_u = 0, \\
\sqrt{-2E_0R^2/\pi} \left[F_1\left(\frac{\pi}{2}, \sqrt{1 - \frac{4K_u}{E_0R^2}}\right) - \frac{4K_u}{E_0R^2} F_2\left(\frac{\pi}{2}, \sqrt{1 - \frac{4K_u}{E_0R^2}}\right)\right], & \text{for } K_u < 0.
\end{cases}
\]

where, the first category elliptic integral \( F_1(\varphi, k) \) and the second category elliptic integral \( F_2(\varphi, k) \) are in the form of

\[
F_1(\varphi, k) = \int_0^{\varphi} \sqrt{1 - k^2 \sin^2 x} \, dx,
\]

\[
F_2(\varphi, k) = \int_0^{\varphi} \frac{1}{\sqrt{1 - k^2 \sin^2 x}} \, dx.
\]

Generally, a single trajectory lacks the inversion symmetry of the real physical systems. So the spectrum obtained from a single trajectory exhibits unphysical even harmonics. A natural way to remedy the unphysical even harmonics is to consider an ensemble of trajectories, evolving from an initial microcanonical distribution with inversion symmetry. For a chaotic system, the initial distribution can be generated with Monte-Carlo method. However, for an integrable system, the distribution generated by Monte-Carlo method does not possess of ergodicity. We find that the points on the same equienergy surface generated by action-angle variables with regular steps possess of good ergodicity. To reconstruct the inversion symmetry, there must exist pairs of \((x_0, y_0, z_0; p_{x_0}, p_{y_0}, p_{z_0})\) and \((-x_0, -y_0, -z_0; -p_{x_0}, -p_{y_0}, -p_{z_0})\) in the initial distribution. For the regularized model for hydrogen atom, it corresponds to pairs of \((u_0, v_0, \phi_0; p_{u_0}, p_{v_0}, p_{\phi_0})\) and \((u_0, v_0, \phi_0 + \pi; p_{u_0}, p_{v_0}, p_{\phi_0})\). And for the regularized hydrogen molecular ion \(H_2^+\), it corresponds to pairs of \((u_0, v_0, \phi_0; p_{u_0}, p_{v_0}, p_{\phi_0})\) and \((u_0 + \pi, v_0, \phi_0; p_{u_0}, p_{v_0}, p_{\phi_0})\).

**III. NUMERICAL SIMULATION**

The numerical computational procedure is based upon the classical trajectory Monte Carlo (CTMC) method [20-21]. CTMC simulation procedure involves three stages, (i) choice of initial conditions, (ii) numerical integration of equation of motion, and (iii) categorization
of each trajectory as excitation, charge transfer or ionization. In the process of numerical integrating, numerical accuracy and computing time are two primary aspects that must be considered. We use the fourth-order Runge-Kutta method with variable steps to perform the numerical calculation. Note also that computer can not deal with singularity, which corresponds to electron-nucleus collision, to overcome this difficulty, we have implemented regularization. With complete regularization, numerical simulation can be established with required precision before, at, and, after collision successfully.

To obtain the harmonic spectra, our procedure also calculate the averaged dipole moment of the excited trajectories with pairs of inversion symmetric initial conditions in the same distribution. Having determined the actual trajectories of the electron, one can easily obtain the component of the averaged dipole moment, which along the laser polarization direction. Then the harmonic spectra of the driven dipole is straightforwardly obtained from it’s power spectra

$$D(\omega) = \lim_{t \to +\infty} \frac{1}{t} \left| \frac{1}{t} \int_0^t \langle \mu(\tau) \rangle e^{i\omega\tau} d\tau \right|^2.$$  

(31)

where, \( \langle \mu(\tau) \rangle \) is the component of the averaged dipole moment along the laser polarization direction. In our calculation, the electric fields of the ultrashort laser pulses are chosen as

$$\varepsilon(t) = \begin{cases} 
E_M \sin^2(\omega_L t/40) \sin(\omega_L t), & \text{for } 0 \leq t \leq 40\pi/\omega_L, \\
0, & \text{otherwise}.
\end{cases} \quad (32)$$

The maximum electric field strength \( E_M \) is related to the laser intensity \( I = \sqrt{\varepsilon_0/\mu_0} E_M^2/2 \), and \( \omega_L \) is the angular frequency, and period \( T_L \) is equal to \( 2\pi/\omega_L \).

The time-dependent dipole moment \( \mu_z(t) \) of a single trajectory along the laser polarization direction sensitively depends on the laser intensity. Fig.1 shows the time evolution of the dipole of the hydrogen molecular ion with initial energy \( E_0 = -1.1034 \) hartree, internuclear distance \( R = 2.00 \) bohr, laser wavelength \( \lambda = 600 \) nm and different laser intensity. And some dipoles of the hydrogen atom and their power spectra are presented in Fig.2, with initial energy \( E_0 = -0.5 \) hartree, laser wavelength \( \lambda = 532 \) nm and different laser
intensity. With the increasing of the laser intensity, the oscillations of the dipole moments are modulated gradually, and it follows a regular pattern with both high and low frequency components. Such patterns are independent of initial conditions. When the laser intensity is large enough, ionization takes place, which strongly modifies the subsequent time evolution of the dipole moment.

The power spectra obtained from a single trajectory of the laser driven hydrogen molecular ion, which with initial energy \( E_0 = -1.1034 \text{ hartree} \), internuclear distance \( R = 2.00 \text{ bohr} \), laser wavelength \( \lambda = 600 \text{ nm} \) and different laser intensity, are presented in Fig.3. The first one corresponds to the field-free case. For the hydrogen atom, it possesses of only regularly decreasing peaks locating at multiples (harmonics) of Kepler’s frequency, that is, \( \omega_n = n\omega_0 \), for \( n = 1, 2, 3 \cdots \), which manifests that the free motion is periodic. While for the hydrogen molecular ion, the free motion is quasi-periodic, it appears regularly decreasing peaks locating at two different characteristic frequencies and their combinations, i.e., at \( n\omega_{01} + m\omega_{02} \), for \( n = 0, 1, 2, 3 \cdots, m = 0, 1, 2, 3 \cdots \) and \( n + m > 0 \). For the laser driven systems, the peaks of harmonic spectra depend strongly on the laser intensity. In addition to the origin peaks, a dominant line at the laser frequency \( \omega_L \) appears, which is the Rayleigh component in the light scattered by the atoms and molecular ions. High-order harmonics, which consist of both odd and even components, do appear in the spectra, and their orders and strengths increase with the increasing of the laser intensity. For a low laser intensity, i.e., in the perturbative regime, the characteristic peaks dominate the power spectrum. For a proper laser intensity, the plateau structure containing both odd and even harmonics appears. For a larger laser intensity, because of the presence of chaos, the spectra become noisier even the motion is bound. For a strong enough laser intensity, the motion becomes unbound and the corresponding spectrum is dominated by a very noisy background, which is generated by the ionized electrons, the only line surviving being the one at the laser frequency, which corresponds to the light scattered by the asymptotically free electron Thomson scattering.

To eliminate the unphysical even harmonics, averaging an ensemble of trajectories evolving from an inversion symmetric distribution is necessary. The time evolution of the averaged
dipole, which evolve from a microcanonical ensemble of 5000 trajectories, are presented in Fig.4, the first one corresponds to the hydrogen atom with initial energy $E_0 = -0.5 \text{ hartree}$, laser wavelength $\lambda = 532 \text{ nm}$ and laser intensity $I = 5.0 \times 10^{14} \text{ W/cm}^2$, the second one corresponds to the hydrogen molecular ion with initial energy $E_0 = -1.1034 \text{ hartree}$, internuclear distance $R = 2.00 \text{ bohr}$, laser wavelength $\lambda = 600 \text{ nm}$ and laser intensity $I = 1.0 \times 10^{14} \text{ W/cm}^2$. Comparing with the evolution of the dipole of a single trajectory, one can easily find that the oscillation of the averaged dipole is smooth and it globally follows the laser oscillation.

The spectra obtained from an ensemble of trajectories are showed in Fig.5 and Fig.6. Fig.5 is the harmonic spectra of the ground-state hydrogen atoms interacting with the laser pulses with $\lambda = 532 \text{ nm}$ and different laser intensity. Fig.6 is harmonic spectra of the hydrogen molecular ion with initial energy $E_0 = -1.1034 \text{ hartree}$, internuclear distance $R = 2.00 \text{ bohr}$, laser wavelength $\lambda = 600 \text{ nm}$ and different laser intensity. As a consequence of averaging process, the unphysical even harmonics are remedied really. Within a proper laser intensity range, the plateau structure that only possesses of odd harmonics appears. As pointed out previously, at a higher intensity, the spectra become noisier even the ionization does not happen because of the effects of chaos. This indicates that the chaos cause the noise of the harmonic spectra. When the laser intensity is high enough, the ionization takes place, thus the noise background conceals the high-order harmonics. This means that the onset of ionization follows the onset of chaos and the ionization suppresses the harmonic generation.

IV. SUMMARY AND DISCUSSION

In summary, within the Born-Oppenheimer approximation and using the classical trajectory method, we have calculated the high-order harmonic generation spectra of the hydrogen atom and the hydrogen molecular ion interacting with ultrashort intense laser pulses. The other multi-photon phenomena, such as multi-photon ionization and above threshold disso-
ciation, can also be simulated. In our subsequent calculations, the dynamics of the electron is investigated by numerical integrating the equations of motion using regularized coordinates. To eliminate the unphysical even harmonics of a single trajectory, averaging over an ensemble of trajectories evolving from an initial microcanonical distribution with inversion symmetry is necessary. Such distribution is constructed using action-angle variables. A plateau structure in the spectra with only odd harmonics is observed within a proper laser intensity range of about $10^{14}$ $W/cm^2$. From our numerical results, we observe that the high order harmonics are cut off at a special order harmonic. At higher laser intensities, chaos introduces noise into the spectra even though the motion is still bound. Finally as the intensity is further increased, ionization takes place, and the harmonics disappear.

These results are qualitatively consistent with recent quantum calculations [16] and experimental observations [1-5], but the cutoff order $N_m$ of the plateau structure is not precisely consistent with formula $N_m = (I_p + 3.17U_p)/\hbar\omega_L$ [27], here, $I_p$ is the ionization potential and $U_p = e^2E_M^2/4m_e\omega^2_L$ denotes the quiver energy or the ponderomotive energy of an electron. As an example, when the laser intensity $I = 10^{14}$ $W/cm^2$ and wavelength $\lambda = 600$ nm, the ionization potential $I_p$ of the ground-state hydrogen molecular ion is 1.1034 $\text{hartree}$ ($29.77 eV = 14.50 \hbar\omega_L$), the quiver energy $U_p = 3.36 eV$ ($= 1.63 \hbar\omega_L$), then $N_m = 19$, and when $I = 7.5 \times 10^{13}$ $W/cm^2$, $N_m = 18$; however, from our simulation, the harmonic plateau are both cut off at 17, and for very large intensity (above $10^{15}$ $W/cm^2$) the high-order harmonics are concealed by the noises. To obtain quantitative results, we have to integrate the time-dependent Schrödinger equation, this can be realized with the split-operator method [22]. For the hydrogen molecular ion, within BOA, a proper internuclear distance $R$ will enhance the high-order harmonic generation[16], and it will be interesting to go beyond the Born-Oppenheimer approximation to investigate what further interesting insights can be obtained when the nuclear motion is taken into account [23-24].

In our model, we only consider the non-relativistic case with dipole approximation. When the laser is sufficiently intense, photoelectrons of relativistic energies can be produced, necessitating a fully relativistic treatment [25]. The dipole approximation is no longer valid,
and the magnetic field is not only present, but acquires an importance similar to that of the electric field. And even before the appearance of the relativistic photoelectrons, the effects of the magnetic field may be very important too [26]. Due to the wiggly motion and the acceleration of the electron near the outermost turning points induced by the magnetic field, the cut-off order of the harmonic plateau maybe higher.

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FIGURES

FIG. 1. Temporal variation of the dipole moment of a single electronic trajectory of the hydrogen molecular ion for different laser parameters with initial energy $E_0 = -1.1034$ hatree and internuclear distance $R = 2.00$ bohr.

FIG. 2. The dipole and their power spectra of a single electronic trajectory of the hydrogen atom for different laser parameters with initial energy $E_0 = -0.5$ hatree.

FIG. 3. Power spectra of the dipole of a single electronic trajectory of the hydrogen molecular ion for different laser parameters with initial energy $E_0 = -1.1034$ hatree and internuclear distance $R = 2.00$ bohr.

FIG. 4. Temporal variation of the averaged dipole moment for different laser parameters. This first one corresponds to the hydrogen atom with initial energy $E_0 = -0.5$ hatree, the other one corresponds to the hydrogen molecular ion with initial energy $E_0 = -1.1034$ hatree and internuclear distance $R = 2.00$ bohr.

FIG. 5. Harmonic spectra of the ground-state hydrogen atoms interacting with different laser pulses, the last two are magnifications of the first one.

FIG. 6. Harmonic spectra of the ground-state hydrogen molecular ion interacting with different laser pulses, with initial energy $E_0 = -1.1034$ hatree and nuclear internuclear distance $R = 2.00$ bohr.
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Fig. 1
I=0.0 W/cm², λ=532 nm

I=10⁻¹³ W/cm², λ=532 nm

I=3.0*10⁻¹⁴ W/cm², λ=532 nm

Fig. 2
Fig. 3
I = 5.0 \times 10^{14} \text{ W/cm}^2, \lambda = 532 \text{ nm}

I = 10^{14} \text{ W/cm}^2, \lambda = 600 \text{ nm}

Fig. 4
Fig. 5
Fig. 6