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EFFECTS OF NUCLEAR VELOCITY ON HIGH-ORDER HARMONIC GENERATION

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Abstract. *We solve numerically the time-dependent Schrodinger equation for the high-order harmonic generation (HHG) from hydrogen molecular ion exposed to the intense ultra-short pulsed laser light. We analyze the HHG spectra and find that the nuclear vibration affects significantly the intensity of harmonics. The dependence of high-order harmonic on time delay reveals the intensity of emitted harmonics is strongly influenced not only by the molecular configuration but also by the direction of initial nuclear velocity. The results show that the intensity of high-order harmonic generation is increased nearly when the inter-nuclear separation takes the equilibrium value and the nuclei are moving closer together. In contrast, with the same initial inter-nuclear separation but with the opposite nuclear velocity, the intensity of emitted light is reduced noticeably.*

I. INTRODUCTION

In the last two decades, high-order harmonic generation (HHG) has become one of the most interesting studied topics which attracts much attention due to its promising applications [1-2]. High-order harmonics are emitted when the ionized electron returns and recombines with its ion parent so it is rich in molecular structural information. Itatani *et al* [3] successfully reproduced highest occupied molecular orbital (HOMO) of N_2 in gaseous phase from the experimental HHG data using the ultra-short intense laser with duration of 30 fs. That achievement is followed by abundance of works in the direction of investigating dynamic imaging of molecules [4-6]. Next, in papers [7, 8], authors proposed the iterative method to retrieve the inter-nuclear separation from laser-induced high-order harmonic spectra using ultra-short laser. With the similar aim of extracting molecular dynamic information, in works [9, 10], scientists took advantage of interference in HHG spectra to obtain the inter-nuclear separation in femtosecond scale. In addition, HHG is an abundant source to probe nuclear dynamic. By analyzing the fine structure of HHG, in [11] authors obtained nuclear vibrational frequency of neutral molecule H_2 and its ion H_2^+. Baker et al. also demonstrated a technique that uses HHG to trace the nuclear dynamic and structural rearrangement in a subfemtosecond time scale [12]. Monitoring attosecond (as) dynamics of coherent electron-nuclear wave packets using HHG is also reported in [13] by Bandrauk et al.

The study HHG including vibrational motion has carried out by adding a nuclear correlation into the single active electron model [14] or solving numerically the time-dependent Schrodinger equation (TDSE). With the later approach, the practically solvable systems hitherto have been limited for those with one or two electrons such as H_2, H_2^+, H_3^{2+} because of the restriction of computer resources. The sensitivity of HHG to vibrational

states of molecular ion H_2^+ and D_2^+ was reported in [15]. In that work authors claimed that harmonics emitted from a higher vibrational level are more intense than those from lower one. In another paper, by numerically solving TDSE for neutral molecule H_2 in one dimension, authors showed that nuclear motion would cause considerable changes in time profile of HHG [16].

In sense of studying effects of nuclear vibration on HHG, the preparation of the initial nuclear-electron wave packet, in our point of view, needs to be investigated thoroughly. The initial nuclear condition should be understood to be the inter-nuclear separation and initial nuclear velocity. In papers, like [15, 16] the initially total nuclear-electron wave packet contains only one single vibrational state so the effect of nuclear velocity cannot be seen. Recently, by considering the molecule interacting with the intense laser from the initial wave function whose nuclear motion is described as superposition of single vibrational states, authors showed effects of initial molecular configuration on HHG intensity [17]. The influence of the initial velocity of nuclei was not analyzed yet.

In present work, we study the effects of initial condition, say averaged inter-nuclear separation and the direction of nuclear velocity, on the intensity of high-order harmonic emitted from H_2^+. The two dimensional model of H_2^+ is employed to solve numerically TDSE for HHG with different initial conditions of molecule. The initial nuclear wave function is prepared as superposition of single vibrational states on the lowest system potential curve. The molecule will freely oscillate before the pulsed laser is turn on at time t_0. By changing the turn on time of the intense pulsed laser, we wish to see the effect of initial conditions, especially the direction of nuclear velocity, on the intensity of emitted harmonics. The rest of the paper is arranged as follows. In section II we introduce formalism and physical model used to calculate HHG. In section III we show results of our calculation and discuss the effects of the initial conditions on the intensity of HHG. Section IV is the conclusion where we summarize what we study.

II. DETAIL CALCULATION

Numerical solution for TDSE to investigate molecular dynamic can be found in many papers [18-20]. Bandrauk et al. [18] used one-dimensional (1D) model for H_2^+ to study dynamic of nuclear-electron wave packet in intense laser field. Marangos et al. [19] also showed the alignment dependence of HHG from H_2^+ using 2D model with fixed nuclei. 2D model for H_2^+ was also employed by Becker et al. to carry a research into charge-resonance-enhanced ionization [20].

In this paper, we employ TDSE for H_2^+ with a single 2D electron and 1D for protons with dipole approximation and length gauge. The time dependent Hamiltonian can be written as

$$H(t) = -\frac{1}{2\mu}\frac{\partial^2}{\partial R^2} - \frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{2}\frac{\partial^2}{\partial y^2} + V(x,y,R) + (xcos\theta + ysin\theta)E(t), \qquad (1)$$

where x, y are electrons coordinate with respect to nuclear center of mass; R is the inter-nuclear separation; μ is reduced mass of two nuclei; θ is called alignment angle between laser polarization vector and molecular axis.
$V(x,y,R) = -\frac{1}{\sqrt{(x-R/2)^2+y^2+a}} - \frac{1}{\sqrt{(x+R/2)^2+y^2+a}} + \frac{1}{R}$ is soft-core Coulomb potential. The

constant a=0.5 is added to avoid the singularity of Coulomb potential and to mimic the real potential energy curve (PEC) of H_2^+. The electric field of laser is $E(t) = E_0 f(t) sin(\omega t)$ with sine-square envelope function consisting of 10 optical cycles. The laser intensity of $2.0\times10^{14} W cm^{-2}$ and wave length of 800 nm is used. Atomic units are used throughout the paper unless stated. We assume the molecule is prepared in the state as a superposition of single vibrational states which freely oscillate before starting to interact with laser field at time t_0,

$$\Psi(x, y, R, t_0) = \sum_\nu C_\nu \chi_\nu(R) \psi(x, y, R) e^{-iE_\nu t_0}. \tag{2}$$

E_ν, χ_ν are vibrational eigenvalues and eigenstates of nuclear motion on the lowest PEC of the system. The electronic wave function $\psi(x, y, R)$ is obtained by solving time-independent Schrodinger equation with each fixed inter-nuclear separation by means of an imaginary time propagation technique using Hamiltonian (1) without laser-molecule coupling factor. During the process of interacting with laser field, the total wave function is written as follow

$$\Psi(x, y, R, t, t_0) = \sum_\nu C_\nu \Phi_\nu(x, y, R, t) e^{-iE_\nu t_0}, \tag{3}$$

where $\Phi_\nu(x, y, R, t)$ is time-propagating wave function of ν^{th} state found by solving TDSE with initial wave function $\psi(x, y, R)$ and Hamiltonian (1) with the help of split operator method. In our calculation, a grid 400 a.u. x 400 a.u is used for electronic motion and inter-nuclear separation may change from 0.5 a.u. to 10.5 a.u. The acceleration of induced dipole moment defined as $\overrightarrow{a}(t, t_0) = -\overrightarrow{E}(t) - \langle\Psi|gradV|\Psi\rangle$ depends parametrically on t_0. Harmonic signals are obtained by transforming Fourier of the acceleration $\overrightarrow{a}(t, t_0)$,

$$I(\omega, t_0) = \left| \int \overrightarrow{a}(t, t_0) \overrightarrow{n} e^{i\omega t} dt \right|^2, \tag{4}$$

where $\overrightarrow{n}$ is the unit vector on an interesting direction.

III. RESULTS

In this part of the paper, we show results of the calculation with the assumption that the initial wave function is prepared as a superposition of two lowest vibrational states, (ν=0, 1) with the same probabilities. Two-level model is enough to check the influence of the direction of nuclear velocity on HHG in the present work but full model is necessary for further research. The figure 1 shows the intensity of harmonics released from H_2^+ aligned parallel to the intense linearly polarized laser varies as a function of time delay t_0. In Fig. 1, we plot the intensity of 21^{st} and 23^{rd} order, the averaged inter-nuclear separation and nuclear velocity changing by time delay t_0.

First of all, Fig.1 indicates that the intensity of HHG modulates with the same period of nuclear vibration ($\sim$ 18 fs). That is easily understood because the initial condition changes periodically. This result also confirm the oscillation of HHG emitted from 1D H_2^+ [17]. The most important point in Fig. 1 we would like to discuss is the correlation between the intensity of harmonics, the inter-nuclear separation and the direction of initial nuclear velocity. In Fig. 1, the HHG intensity reveals maxima nearly when inter-nuclear

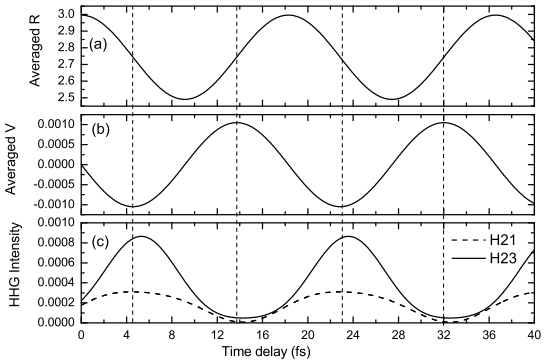

Fig. 1. The averaged inter-nuclear separation R (a) and nuclear velocity V (b) and the intensity of 21^{st} (dash line) and 23^{rd} harmonic (solid line) as functions of time delay t_0. The laser intensity of $2.0 \times 10^{14} Wcm^{-2}$, wave length of 800 nm and pulse duration of 10 cycles is used.

separation has equilibrium value and nuclei are moving closer together. However, with the same value of averaged inter-nuclear separation but with the opposite direction of nuclear velocity, HHG intensity demonstrates an obvious decrease around 8 times from the maximum value. This characteristic is also found with others harmonic orders in plateau region H15-H35. It means that the direction of initial velocity of nuclei plays an important role in the process of generating harmonics from molecule exposed to the intense laser.

Continuously, we check the influence of initial nuclear velocity on the intensity of HHG with the different alignment angles. In Fig. 2 we plot the averaged inter-nuclear separation, initial nuclear velocity and the intensity of 21^{st} and 23^{rd} harmonic when the alignment angle is 90 degrees.

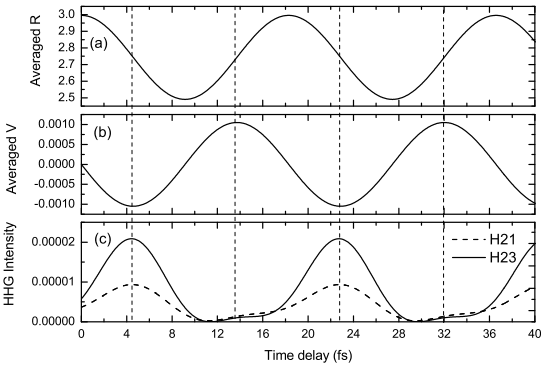

Fig. 2. The same Fig.1 with the alignment angle of 90^0.

In Fig. 2, one can see although the intensity of HHG from perpendicularly aligned molecules is around 40 times weaker than that with parallel alignment, it also exhibits the properties as mentioned in Fig.1. It means, once again, the intensity of HHG oscillates with the same period of the nuclear vibration and shows peaks when nuclei are passing through the equilibrium position and nuclear velocity is negative. Hence, we conclude that the intensity of HHG is decided by not only the molecular configuration but also nuclear velocity. To have an insight into the relation between the direction of nuclear velocity and the intensity of HHG to interpreter the above phenomenon, in our point of view, is worth investigating. We use the time-frequency analysis technique whose formula is

$$I(\omega, t, t_0) = \left| \int \overrightarrow{a}(t', t_0) \overrightarrow{n} e^{i\omega t'} e^{-(t'-t)^2/2\sigma^2} dt' \right|^2 . \tag{5}$$

The window function width σ is chosen as one-tenth of the laser optical period. This analysis technique gives us a time profile of harmonics which is dependent on orders ω and the emitting time t. In Fig. 3a, we plot the time profiles of 23^{rd} order in the case of parallel alignment with two values of time delay 4.58 fs and 13.70 fs when the iner-nuclear separations have the same value but opposite direction of nuclear velocity. We also add averaged inter-nuclear separations changing from initial values as functions of emitting time into Fig. 3b.

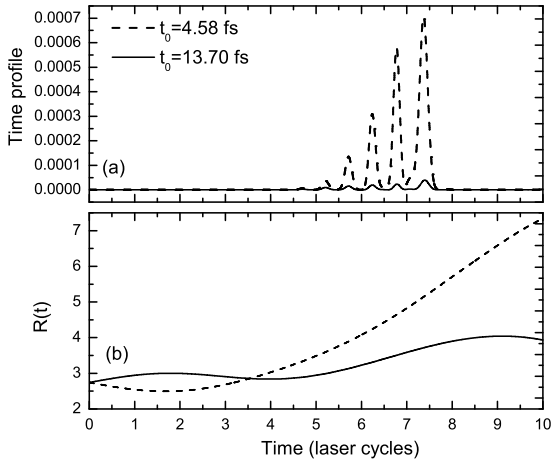

Fig. 3. Time profile of 23^{rd} harmonic (a) and inter-nuclear separation (b) changes as a function of emitting time with two cases of time delay 4.58 fs (dash lines) and 13.70 fs (solid lines)

In Fig. 3a, one can see the intensity of $23^r d$ changes as a function of the emitting time and depends parametrically on time delay t_0. It is obvious that how the releasing light is intense relies clearly on the time when the laser is turn on. This phenomenon may be explained by basing on how the inter-nuclear separation varies while the molecular ion

is exposed to the intense laser field. Fig. 3b shows that with the same initial value, the inter-nuclear separation can change in different ways due to the opposite sign of the initial nuclear velocity. At the end of the pulse, the inter-nuclear separation corresponding with time delay 4.58 fs can reach to value as 7 a.u. while it only gets to 4 a.u. if the intense laser is turn on at 13.70 fs. The growing up of inter-nuclear separation will lower the ionization potential of the molecular ion that makes the electron tunnel easier and lead to the fact that the HHG is more intense. Thus, we can conclude the direction of initial velocity that creates a taking off of inter-nuclear separation during the interacting time with laser pulse may lead to an increase in HHG intensity.

IV. CONCLUSION

In the present paper, we study the effects of the initial condition including the initial averaged inter-nuclear separation and nuclear velocity on the intensity of harmonic signal. The simulation indicates that not only molecular configuration but nuclear velocity also influences noticeably on the intensity of HHG. That property is well checked for two cases parallel and perpendicular alignment. The simulation shows that with the same initial inter-nuclear separation, the molecule whose nuclei are passing equilibrium position and inter-nuclear separation is decreasing can emit more intense laser light than that from a molecule with nuclear velocity having the opposite direction. This phenomenon is explained due to the increasing of the inter-nuclear separation during the emitting time using time-frequency analysis technique.

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