THE DEPENDENCE OF IONIZATION PROBABILITY AND HIGH-ORDER HARMONIC GENERATION OF H_2^+ ON INTERNUCLEAR SEPARATION

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ABSTRACT

We present Time Dependent Schrödinger Equation approach for numerical calculations of ionization probability and high-order harmonic generation (HHG) intensity of hydrogen molecular ions H_2^+ *with large internuclear distance, exposed to an intense laser field. We find that the ionization probability and HHG are strongly dependent on the internuclear distances. The ionization probability is significantly enhanced in a certain range of nuclear separations. However, the HHG rapidly increases and then reaches to a saturation value with increasing internuclear distances.*

Keywords: high-order harmonic generation, ionization probability, dissociation, internuclear separation, ultrashort laser.

TÓM TẮT

Sự phụ thuộc của xác suất ion hóa và cường độ sóng điều hòa bậc cao của H_2^+ vào khoảng cách liên hạt nhân

Chúng tôi trình bày phương pháp giải số phương trình Schrodinger phụ thuộc thời gian nhằm tính toán xác suất ion hóa và tín hiệu sóng điều hòa bậc cao (HHG) của ion phân tử H_2^+ *có khoảng cách liên hạt nhân lớn trong trường laser mạnh. Chúng tôi chỉ ra rằng xác suất ion hóa và cường độ HHG phụ thuộc mạnh vào khoảng cách liên hạt nhân. Xác suất ion hóa được tăng cường trong một vùng nhất định của khoảng cách liên hạt nhân. Tuy nhiên, khi tăng khoảng cách liên hạt nhân, cường độ HHG tăng mạnh, sau đó đạt tới giá trị bão hòa.*

Từ khóa: sóng điều hòa bậc cao, xác suất ion hóa, phân ly, khoảng cách liên hạt nhân, laser xung cực ngắn.

1. Introduction

Atoms and molecules exposed to ultrashort intense laser pulses reveal many nonlinear optical phenomena, for example high harmonic generation (HHG) [5], above threshold dissociation (ATD), and above threshold ionization (ATI) [1,7]. The

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investigations regarding these phenomena have many broad applications such as the control of the molecular processes, generation of few cycles of femtosecond and attoseconds pulses, and extraction of atomic and molecular structure information.

Due to the greater degree of freedom, the calculations of ionization probabilities and HHG from molecules are more complicated than that from atoms. Using the approximately semi-classical three-step model, the amount of calculation is greatly reduced. On the other hand, the numerically solving the time – dependent Schrödinger equation (TDSE) approach can give exact results but requires a large computer memory, so now only be applied to the simple molecules such as H_2, H_2^+, H_3^{2+}.

Ionization probabilities and HHG from atoms and molecules in an intense laser field has theoretically and experimentally studied for more than haft-century. Recently, in many works [2,6,7], researchers calculated ionization rates of molecules as a function of internuclear distances. In [7], by using three-dimensional TDSE method authors pointed out that the ionization rate of H_2^+ is found to exhibit maxima in a certain range of large internuclear separations, exceeding the atom limit by an order of magnitude, and this effect is called charge-resonance-enhanced ionization (CREI). Using semi-classical model, the maxima and minima in HHG spectra from diatomic molecules with large internuclear distances are sensitive to the laser intensity [2]. Moreover, in [6], the multiphoton ionization and HHG of H_2^+ in laser pulses with intensities of $5x10^{13}W/cm^2$ at large internuclear distances are calculated. At some large internuclear separations, the HHG intensities are strongly enhanced as a consequence of enhanced multiphoton ionization. However, calculation the HHG at large internuclear distances of H_2^+ exposed to laser with higher intensities as a result of tunnel ionization has not considered yet.

In this article, using TDSE method we calculate the ionization probability and high-order HHG intensity of hydrogen molecular ions H_2^+ exposed to intense laser fields with intensities higher than $10^{14}W/cm^2$. The molecule is supposed at states with various internuclear separations in the range between 0 and 30 a.u. Additionally, the numerical method for solving TDSE equation of H atom is also introduced in order to obtain the ionization probability and HHG spectra of hydrogen atom. In the second section, we start with TDSE approach for solving the molecular wave function of H_2^+ and H. In the next section, we discuss our results for HHG spectra and ionization probabilities of H_2^+ and H in the laser fields. The last section contains concluding remarks.

2. Methods

When a molecular ion H_2^+ interacts with a linearly polarized laser field, after being ionized, electron moves along the laser polarization direction. On the other hand, as an electric dipole in an electric field, the molecular axis quickly aligns along the polarization vector of intense laser pulse. Thus we restrict our study to the one-dimensional model (1D) to reduce the amount of calculations.

In this section, we introduce the way to solve the molecular wave function of H_2^+ with various internuclear separations and of atom H. Besides, calculation method of the ionization probabilities of molecular ion H_2^+ and atom H interacting with laser are also mentioned. We use the atomic units ($\hbar = e = m_e = 1$) throughout the paper unless stated.

The TDSE of molecular ion H_2^+ interacting with intense pulse laser has the following form

$$i\frac{\partial}{\partial t}\Psi_{MO}(z,R,t) = \left(-\frac{\partial^2}{2\partial z^2} + V_{C\_MO}(z,R) + V_L(z,t)\right)\Psi_{MO}(z,R,t), \tag{1}$$

where z is the electronic coordinate with respect to the center of mass of two nuclei for H_2^+, R is the internuclear distance. The Coulomb potential of molecular ion H_2^+ is written as

$$V_{C\_MO}(z,R) = \frac{1}{R} - \frac{1}{\sqrt{\left(z-\frac{R}{2}\right)^2+\varepsilon}} - \frac{1}{\sqrt{\left(z+\frac{R}{2}\right)^2+\varepsilon}}, \tag{2}$$

For atom H the TDSE equation can be written

$$i\frac{\partial}{\partial t}\Psi_{AT}(z,t) = \left(-\frac{\partial^2}{2\partial z^2} + V_{C\_AT}(z) + V_L(z,t)\right)\Psi_{AT}(z,t). \tag{3}$$

Its Coulomb potential has following form

$$V_{C\_AT}(z) = -\frac{1}{\sqrt{z^2+\varepsilon}}. \tag{4}$$

The constant $\varepsilon = 1$, called the softening parameter, is added to Coulomb potential expression to avoid the singularities and mimic energy curve of a real molecular. The equilibrium internuclear distance of molecular ion H_2^+ becomes R = 2.609 a.u.

The laser-molecular interaction is formulated in the dipole approximation in length gauge

$$V_L\left(z,t\right) = zE(t) = zE_0 f\left(t\right)\sin\left(\omega_0 t\right), \tag{5}$$

where E_0, ω_0 are the electronic amplitude and its angular frequency of laser pulse, respectively. $f(t)$, called the envelope of the laser pulse, has a sine-squared shape.

The equation (1) for H_2^+ can be numerically solved by the split operator method [3]. Accordingly, the wave function $\Psi\left(z,R,t\right)$ at any moment t is calculated by applying the time evolution operator to the initial wave function $\Psi\left(z,R,t=0\right)$. The initial wave function $\Psi\left(z,R,t=0\right)$ can be solved by imaginary-time method [4] at different values of internuclear distance. The same way is also applied to calculate the time dependent wave function of H atom.

The acceleration of induced dipole moment can be written as follows

$$\vec{a}(t) = -\vec{E}(t) - \left\langle \Psi \middle| \nabla V_c \middle| \Psi \right\rangle, \tag{6}$$

and high-order harmonics is obtained by Fourier transforming of $\vec{a}(t)$.

After calculating the wave function of molecular ion, we continue to calculate the ionization probability. For H_2^+ the ionization region S_i is defined as

$$S_{i\_MO} = \left\{ z \middle| Abs(z) > z_i + R/2 \right\}, \tag{7}$$

where $z_i = 20$ a.u., and $Abs(z)$ is the absolute value of z.

The time-dependent ionization probabilities of ion H_2^+ can be calculated as

$$P_{i\_MO}(t) = \iint_{S_{i\_MO}} \left|\Psi_{MO}(z,R,t)\right|^2 dzdR. \tag{8}$$

For H atom the ionization region is limitted

$$S_{i\_AT} = \left\{ z \middle| Abs(z) > z_i \right\}, \tag{9}$$

The ionization probability of atom H has following form

$$P_{i\_\mathrm{AT}}(t) = \int_{S_{i\_\mathrm{AT}}} \left|\Psi_{AT}(z,t)\right|^2 dz. \tag{10}$$

In this investigation, the numerical grid points were chosen over $0.5\text{a.u.} < R < 30\text{a.u.}$ and $-800\text{a.u.} < z < 800\text{a.u.}$ at interval of $\Delta z = 0.39$ a.u., $\Delta R = 0.04$ a.u., and the time step of the wave function propagation was set at $\Delta t = 0.18$ a.u.

3. Results

Firstly, we present our caculations of ionization probability of H_2^+ as functions of laser-molecular time interation and of internuclear separations. The time-dependent ionization probabilities for various internuclear separations are calculated (Fig. 1). The results, which are shown in Fig. 1 for laser with intensity of 2x10^{14}W/cm^2, wave length of 800 nm and time duration of 26.67 fs (10 optical periods), indicate that the onset time of ionization occurs after three cycles. The probabilities rapidly take off in next few laser periods, and then almost unchanged in last cycles. Moreover, the shapes of probability are repeated for all cases of internuclear separations.

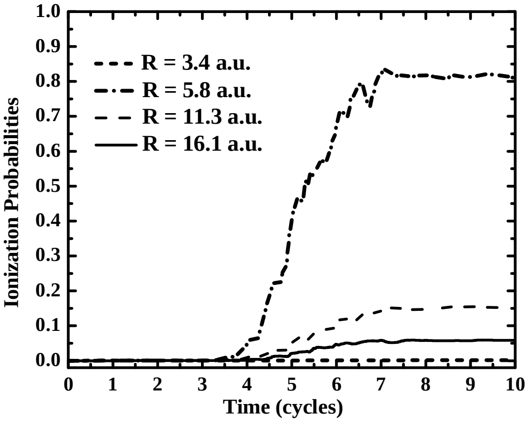

Fig. 1. *The time-dependent ionization probabilities of* H_2^+ *for different internuclear separations. The laser with peak intensity of 2x10*^{14} *W/cm*^{2}*, wave length of 800 nm and 10-cycle pulse is used.*

In order to find out the dependence of ionization probabilities of H_2^+ on internuclear distances, the probabilities at the end of the pulse are plotted in Fig. 2 as a function of separations. The lasers with different peak intensities are used. In Fig. 2, one can see for small internuclear separation, the ionization probability is negligible due to the fact that the electron is tightly confined in the potential well that lead to its difficult release into continuous region. After R=2.5 a.u., the ionization quickly increase to maximum values. However, as shown in Fig.2, it is clear that ionization probabilities are strongly enhanced just in a certain range of internuclear distances. The width of internuclear separations that enhanced ionization occurs is wider with increasing peak intensity of laser. By Gaussian peak fitting of the curves of ionization probabilities, we obtain the FHWM (full width of half maximum) is 0.38 a.u. for laser with intensity of 1x10^{14}W/cm^2, 3.4 a.u. for 2x10^{14}W/cm^2, 4.4 a.u. for 3x10^{14}W/cm^2 and 4.7 a.u. for 4x10^{14}W/cm^2. This phenomenon can be understood by considering the existence of charge-resonance states of odd-charge molecular ions in intense laser field as predicted in [7]. In this state, the electron population of the excited energy level $1\sigma^+$ is more sufficient than that in ground state $1\sigma^-$, thus the dominance of above-barrier ionization leads to the enhanced ionization. However, if the internuclear separation is greater than the critical values, the above-barrier ionization of excited level does not occur due to decreasing of inner potential well, that the ionization probability decreases. For large internuclear distances, the ionization probability is almost unchanged and approximately equals 0.6% for laser of 1x10^{14}W/cm^2, 6% for 2x10^{14}W/cm^2, 26% for 3x10^{14}W/cm^2 and 49% for 4x10^{14}W/cm^2.

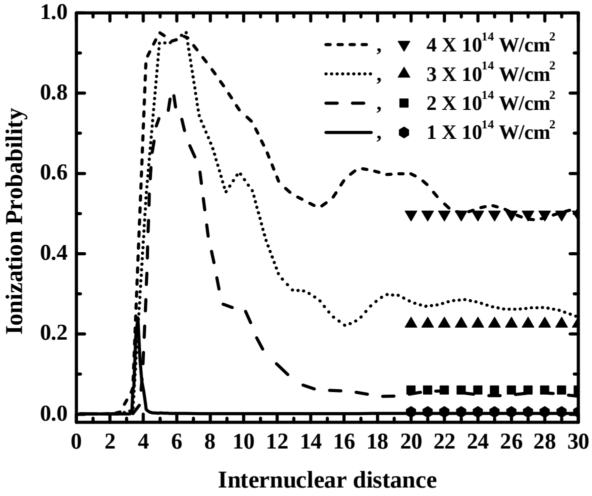

Fig. 2. *The ionization probabilities of* H_2^+ *(line) and H (scatter) in respect to different internuclear separations. The laser pulses with wavelength of 800 nm, time duration of 10 cycles, peak intensities of 1x10*^{14}*, 2x10*^{14}*, 3x10*^{14} *and 4x10*^{14} *W/cm*^{2}*.*

Secondly, we calculate the HHG intensities of H_2^+ molecule in states with some various internuclear separations exposed to intense laser. In Fig.3 we show the HHG intensities for R = 3.2; 3.4; 3.9 a.u. As one can see, the shapes of HHG spectra are the same for different separations. However, for the same harmonic order the difference between the HHG intensities at different distances R can be as large as several orders of magnitude. The more internuclear distance is, the strong HHG spectra are emitted.

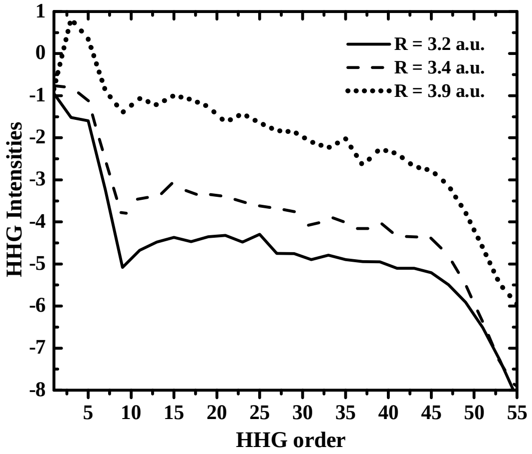

Fig. 3. *The HHG spectra of* H_2^+ *for different internuclear separations. The laser with peak intensity of 2x10*^{14} *W/cm*^{2}*, wave length of 800 nm and 10-cycle pulse is used. The laser parameters are the same in Fig 1.*

We continue to calculate the HHG spectra emitted from H_2^+ for internuclear distances in range of 2.5 to 30 a.u. Fig. 4. is the illustration for the calculation of the harmonics intensity as a function of internuclear separations for some invidual harmonics in plateau region. The HHG demonstrate strong dependence on the internuclear distance *R*. For distances from 2.5 a.u to 4.2 a.u. HHG intensities increase like-linearly and then from the separation 4.2 a.u they are nearly unchanged. We call that they reach the saturation point. This behavior of the HHG spectra is continuously confirmed with different laser pulses for all harmonics orders in plateau region and the similar results are obtained.

The explanation of HHG of H_2^+ base on the obtained dependence of ionization probability on internuclear distances. The like-linearly growing up of HHG intensity in the interval of internuclear separation from 2.5 a.u to 4.2 a.u is the consequence of increasing ionization probability of H_2^+ in this range (see in Fig. 2). But as one can see in Fig.2, the ionization probability is enhanced just in a certain range of nuclear separations, then it significantly decrease. However, the HHG intensity rapidly reach a saturation value and then keep almost unchanged in despite of decreasing of ionization probability.

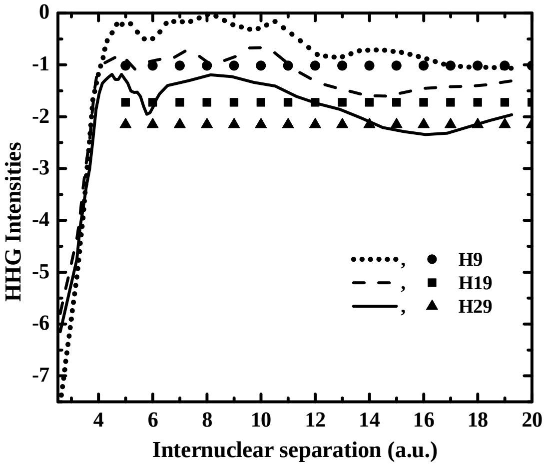

Fig. 4. *The HHG intensities emitted from* H_2^+ *(solide line) and H (dash line) in respect to different internuclear separations of 9th, 19th and 29th harmonics. The laser parameters are the same in Fig 1.*

To explain this contrast we carry out the calculation of ionization probability and HHG intensity of hydrogen atom H. This results are presented as scatters in Fig. 2 and Fig. 4. It is clear that for large internuclear separations the ionization probability and HHG signal of H_2^+ almost coincide with that of H for every harmonics order in plateau region in HHG spectra. Thus, we conclude that for large separations ion H_2^+ tends to dissociate to H atom and H^+ ion.

4. Conclusions

By numerically solving the TDSE we obtain the ionization probability and the HHG spectra of a H_2^+ molecule with various internuclear separations exposed to ultrashort intense laser fields. The ionization probability and HHG demonstrate strong dependence on the internuclear distance. For small internuclear distance the HHG intensity and ionization probability quickly increase with increasing internuclear separation. After that the ionization probability is enhanced in a certain range of internuclear separations, then rapidly decrease while the HHG signal reach a saturation value then almost unchanged. This results can be explain by the dissociation of ion H_2^+ to H atom and H^+ ion.

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