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DYNAMICS of H_2^+ in TWO-COLOR ULTRASHORT LASER PULSE *

CHEN ZHAO-YANG(陈朝阳), QU WEI-XING(屈卫星), XU ZHI-ZHAN (徐至展),
Laboratory for High Intensity Optics, Shanghai Institute of Optics and Fine Mechanics,
Chinese Academy of Sciences, Shanghai 201800, *China*

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Numerical simulations of ionization and dissociation processes of hydrogen molecular ion H_2^+ interacting with two-color intense (10^{14}W/cm^2–10^{15}W/cm^2) ultrashort (the duration $\approx$ 22fs) laser pulse are made. The result shows that the ionization and dissociation processes are strongly dependent upon the relative phase between the two color fields. It means that, in the case of ultrashort pulse, the phase coherence control of ionization and dissociation processes can be realized.

Keywords: hydrogen molecular ion, ionization, dissociation, two-color field
PACC: 3380

I. INTRODUCTION

The interaction of intense laser pulses (intensity $> 10^{13}$W/cm^2) with atoms and molecules has been an area of active studies during the last two decades.[1−3] It is known that molecules exposed to intense laser fields can show more interesting features due to the additional degree of freedom. Many nonlinear phenomena such as dissociation through bond softening,[4] above-threshold dissociation,[5] dissociation through Coulomb explosion,[6] and charge-enhanced ionization[7] have been observed in intense laser-molecule interaction experiments. Hydrogen molecular ion, the simplest molecule, has recently attracted both theoretical and experimental attention. But if the freedom degrees of electron and nuclei of H_2^+ are not separated, it is too difficult for a personal computer to exactly approach the dynamical process.[8,9] Fortunately, the collinear model for H_2^+ established by Kulander *et al.*[1] is very suitable to investigations of ionization and dissociation of H_2^+ interacting with intense fields. Recently, hydrogen molecular ion in intense two-color laser pulse has been investigated.[10,11] For example, Zuo *et al.*[11] examined the phase control of molecular ionization in intense two-color laser fields and found that ionization is strongly dependent on the relative phase between the two colors.

In this paper, we will use the collinear model and numerically solve the time-dependent Schrödinger equation (TDSE) to calculate the ionization and photodissociation of H_2^+ in intense two-color ultrashort laser pulses and compare them with the results for the case of one-color field. It is found that changing the coherence of the two-color field can achieve a remarkable control of the ionization and dissociation processes.

II. NUMERICAL METHOD

The linearly polarized two-color laser pulse field is

$$E(t) = f(t)[E_f \sin(\omega_f t) + E_h \sin(\omega_h t + \phi)], \qquad (1)$$

where E_f (E_h) and ω_f (ω_h) are the amplitude and frequency of the fundamental (third-harmonic) field respectively; ϕ is the relative phase between the fundamental and the third-harmonic fields, $f(t)$ is the pulse envelope function.

The time-dependent Hamiltonian of H_2^+ in the laser field $E(t)$ is given in atomic units as

$$H_I = -\frac{1}{2\mu}\frac{\partial^2}{\partial R^2} - \frac{1}{2}\frac{\partial^2}{\partial z^2} + V(R,z) + zE(t), \qquad (2)$$

where μ is the reduced mass of two protons. R denotes the distance between the two protons and z is the electron's coodinate. The potential $V(R,z)$ reads

$$V(R,z) = 1/\sqrt{R^2+q_n^2} - 1/\sqrt{(z-R/2)^2+q_e^2} - 1/\sqrt{(z+R/2)^2+q_e^2}, \qquad (3)$$

where $q_n = 0.01$ ($q_e = 1.0$) is a screening parameter of one-dimensional Coulomb potential. Therefore, the dynamics of hydrogen molecular ions in a two-color

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field can be obtained by numerically solving the time-dependent Schrödinger equation (TDSE)

$$i\frac{\partial}{\partial t}\Psi(R,z,t) = H_I\,\Psi(R,z,t). \tag{4}$$

In the paper we use the usual two-order split-operator algorithm to directly integrate Eq.(4) in the spatial range of $|z| \le z_{\max} = 102.4$ bohr with $\Delta z = 0.2$ bohr and $0 \le R \le 25.6$ bohr with the step size $\Delta R = 0.1$ bohr. The ground state of this system, which is determined by propagating a wave function on the two-dimensional potential in imaginary time, is chosen as the initial condition when the laser pulse is imposed on.[8] In order to avoid the reflection of the wave function on the grid edge, we use the usual masking function to absorb the wave function flux approaching to the grid edge for R and z dimensions. However, no absorption is introduced at $R = 0$ boundary since the two nuclei of H_2^+ cannot coincide and therefore the wave function $\Psi(R,z,t)$ automatically tends to zero at $R = 0$ boundary during the interaction.

The ionization probability is calculated through integrating the absorbed electron flux in large $|z|$ regions. The dissociation probability is determined by summing the probability density in the range of $R > R_D = 9$ bohr but also $|z| < Z_I + R/2$, where Z_I is equal to 32 bohr which defines the motion range of the bound electron; The time-dependent average separtion between the two nuclei $\langle R(t)\rangle$ is defined as

$$\langle R(t)\rangle = \frac{\int\int R\,|\Psi(R,z,t)|^2\,\mathrm{d}R\mathrm{d}z}{\int\int |\Psi(R,z,t)|^2\,\mathrm{d}R\mathrm{d}z}. \tag{5}$$

In our calculations, for simplicity, we consider the case that the strength of the third-harmonic laser equals that of the fundamental laser, i.e., $E_h = E_f$. In order to compare the result for two-color field with that for one-color field, the effective intensity is introduced. The effective intensity I_{eff} for the field described by Eq.(1) is defined in this paper as

$$I_{\rm eff}(\mathrm{W/cm^2}) = 3.52\times 10^{16}(E_h + E_f)^2, \tag{6}$$

where both E_h and E_f are in atomic units. The wavelength of the fundamental field λ_f is equal to 532nm. The total duration of the two-color laser pulse is equal to $12T$ ($\approx$ 22fs), where $T = 2\pi/\omega_f$ is the period of the fundamental field. The pulse envelope function is given by

$$f(t) = \begin{cases} \sin^2(\dfrac{\pi t}{10\mathrm{T}}), & 0 \le t < 5\mathrm{T}, \\ 1 & 5\mathrm{T} \le t < 7\mathrm{T}, \\ \cos^2(\dfrac{\pi(t-7\mathrm{T})}{10\mathrm{T}}, & 7\mathrm{T} \le t \le 12\mathrm{T}. \end{cases} \tag{7}$$

Ⅲ. RESULT AND DISCUSSION

We carefully examined the influence of the two-color field on the dynamics of H_2^+ by calculating the time-dependent ionization and dissociation probabilities for various effective intensities varying from $10^{14}\mathrm{W/cm^2}$ to $10^{15}\mathrm{W/cm^2}$ and different relative phases. It should be emphasized that, in the following, we shall give all figures at the same intensity, since we have calculated the results for different intensities and found that, in our calculated intensity range, the basic characteristics of ionization and dissociation processes are not strongly related to a special value of intensity. We first show a comparison of the ionization probabilities as a function of time for the pulses with the same duration and the same effective intensity but different relative phases ϕ. From Fig.1 one can clearly see that, although the relative phases are different, the ionizations begin at almost the same time. But the ionization probabilities for different phases do not reach the maximum at the same time and their maximum values vary with the relative phases. For example, at about 7 cycles the ionization probability is as high as 90 percent for $\phi = 0$ but only 70 percent for $\phi = \pi$.

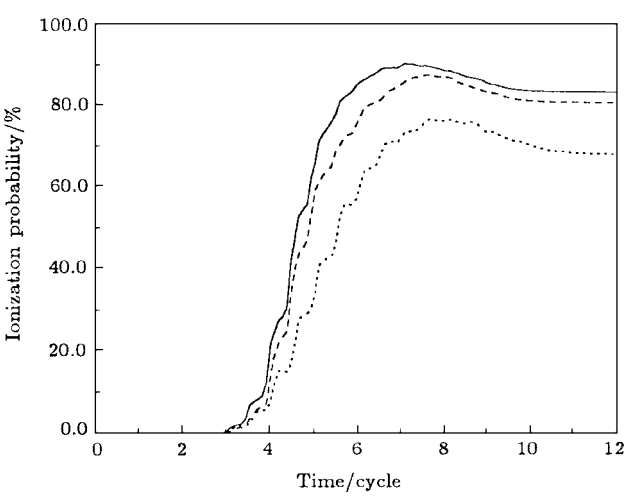


Fig.1. Ionization probability of 1D H_2^+ under a two-color laser pulse with the duration of 12T and effective intensity $7.5\times 10^{14}\mathrm{W/cm^2}$ as a function of time for different relative phases ϕ: $\phi = 0$ (solid line), $\phi = \pi/2$ (dashed line) and $\phi = \pi$ (dotted line).

The dissociation probability as a function of time for the pulses with the same duration and the same

effective intensity but different relative phases ϕ are plotted in Fig.2. It is obviously seen that the dissociation probability is strongly dependent on the relative phase. The dissociation for the relative phase $\phi = \pi$ is much stronger than for any other relative phases. Fig.2 shows that, at the end of the pulse the dissociation probability may be very different when changing the relative phase, for example, the maximum value can be twice the minimum. One clearly sees that the attainable degree of control of the dissociation process is considerable.

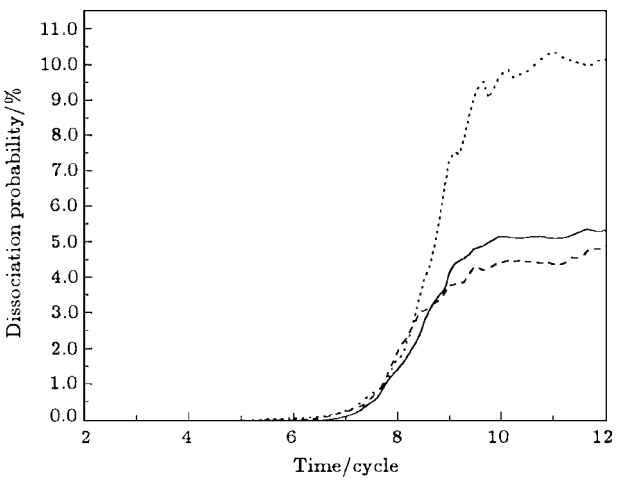


Fig.2. Dissociation probability of 1D H_2^+ as a function of time for different relative phases ϕ: $\phi = 0$ (solid line), $\phi = \pi/2$ (dashed line) and $\phi = \pi$ (dotted line). The other parameters are the same as in Fig.1.

Now, to further investigate the effect of two-color field on the dynamics of H_2^+, we plot in Fig.3 the ionization and dissociation probabilities when H_2^+ is exposed to the one-color field with intensity of $7.5 \times 10^{14}\mathrm{W/cm}^2$ and wavelength of 532nm. The pulse duration and the pulse envelope function are respectively 12T and $f(t)$. The results obtained from solving TDSE agree very well with that from the classical simulations.[9] Comparing Fig.1 with Fig.3 one can find that, ionization process occurs earlier in the case of two-color field than in the case of one-color field. For the case of one-color field, the ionization process begins at about 4 cycles and reaches its maximum at 9 cycles; but for the case of two-color field the ionization process begins at 3 cycles. Similarly, comparing Fig.2 with Fig.3 one can find that the dissociation process occurs earlier for two-color field than for one-color field, and at the same time, the dissociation probability for the case of two-color field and $\phi = \pi$ is much larger than for the case of one-color field. As will be discussed later, this just exhibits the fact that the nonadiabatic effect is enhanced for the case of two-color fields.

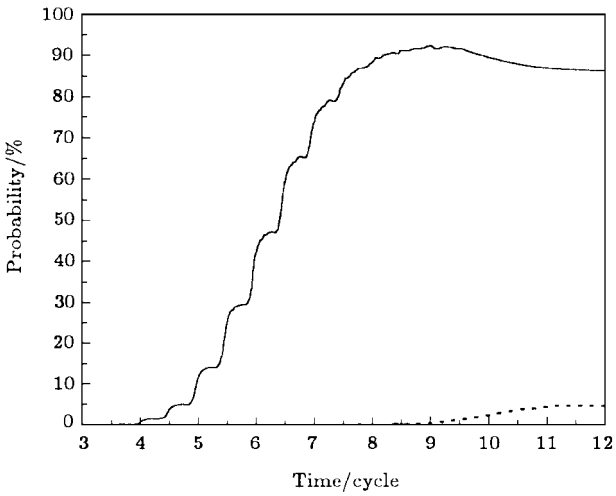


Fig.3. Ionization probability (solid line) and dissociation probability (dotted line) of 1D H_2^+ under a one-color laser pulse with the duration of 12T and peak intensity $7.5 \times 10^{14}\mathrm{W/cm}^2$ as a function of time.

Due to the repulsion of Coulomb potential between two nuclei, the internuclear separation will increase with time when the laser pulse is imposed on the H_2^+. The separation as a function of time is plotted in Fig.4. One can observe that the separation between two nuclei is also strongly dependent on the relative phase. Especially, when $\phi = \pi$ the separation increases rapidly with time after six cycles. This is in agreement with the dissociation process shown in Fig.2.

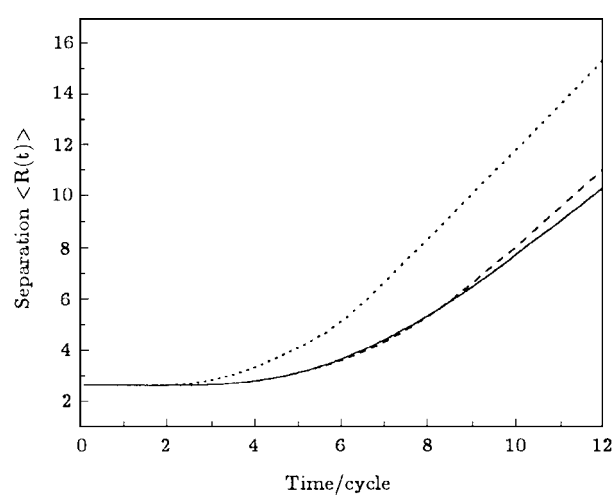


Fig.4. The separation between two nuclei of 1D H_2^+ under a two-color laser pulse with the duration 12T and effective intensity $7.5 \times 10^{14}\mathrm{W/cm}^2$ as a function of time for different relative phases ϕ: $\phi = 0$ (solid line), $\phi = \pi/2$ (dashed line) and $\phi = \pi$ (dotted line).

From Figs.1 and 2 one can see that, the ionization and dissociation processes can be controlled by varying the relative phase between two fields. One can easily find that the tendencies of the dependences of the ionization and dissociation processes on the relative phase are fundamentally opposite. There may be two reasons for this. One is that the movements of the electron and the nuclei are not on the same time scale. The change of the laser pulse can be considered as an adiabatic process for the movement of the elelctron, but must be conisdered as a non-adiabatic process for the movement of the nuclei. When H_2^+ interacts with two-color ultrashort laser pulses, the nonadiabatic effect is further enhanced. Another is that, although for the case of ultrashort laser pulses the ionization process always dominates, to some degree there is still a competition between the processes of ionization and dissociation. Moreover, the fact that when $\phi = \pi$ the ionization probability is minimum but the dissociation is maximum can be easily understood as due to the competition between the processes.

In addition, the influences of the laser pulse envelope (especially during the turn-off period) on the ionization and dissociation are different. When the turn-off period of the laser begins, the ionization probability begins to decrease but the dissociation probability continues to increase. The former is due to that the strength of laser pulse is decreased, the electron will have more probability to return to the vicinity of the ion and recombine with its parent ion. The latter is because that, the total Hamiltonian of H_2^+ + laser field is time-dependent and belongs to a non-autonomous one, so the energy of H_2^+ is not conservative and the nuclear separation must gradually increase with time after the pulse is imposed on.

IV.SUMMARY

In this paper we have numerically simulated the dynamics of hydrogen molecular ion in two-color ultrashort laser pulse by solving the time-dependent Schrödinger equation. Using the collinear model we can observe both the ionization and dissociation processes. Our results show that two-color intense (10^{14}W/cm^2–10^{15}W/cm^2) ultrashort ($\approx$ 22fs) laser pulse can significantly change the ionization and dissociation probabilities by tuning the relative phase between the two color fields. This means that the coherent control of the ionization and dissociation processes is available. The ionization process is stronger for the relative phase equal to 0 than for any other relative phases, but the dissoication process is stronger for the relative phase equal to π than for other phases. Particularly, for the dissociation process, the two-color ultrashort field exhibits more nonadiabitic effect than one-color ultrashort pulse.

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