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Classical dynamics of 3D Hydrogen molecular ion in intense laser fields

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The classical trajectory method is used to study the dynamics of 3D Hydrogen molecular ion H_2^+ interacting with intense laser fields. In the 3D classical model, a three-body Hamiltonian with one-dimensional nuclear motion restricted to the direction of the laser field is considered. The motion of electron and nucleus is described by the classical Hamiltonian canonical equations. The probabilities of ionization, dissociation and Coulomb explosion as functions of time are calculated and the average distances from electron to the mass-center for various laser parameters are implemented by symplectic method. The dynamics of H_2^+ in two-color laser fields are also investigated. We compare our results with the corresponding quantum-mechanical calculations and find they produce similar qualitative features in many cases.

KEY WORDS: Hydrogen molecular ion, classical trajectory method, symplectic method, intense laser field

1. Introduction

The studies of interaction of intense laser pulses (The intensity is more than 10^{14} W/cm^2) with atoms and molecules have been an active area during the past two decades [1–3]. With the development of femtosecond lasers, the femtochemistry becomes possible where chemical reaction dynamics can be probed at the atomic scale [4]. The Hydrogen molecular ion H_2^+ was always chosen for theoretical and experimental study because it is the simplest molecular system, which consists of one electron and two protons. During the recent years, some numerical calculations about population effects in harmonic generation by H_2^+ are considered. Because the perturbation theory is not appropriate for the intense laser field, people usually attempt to study this problem by solving numerically the 3-dimensional time-dependent Schrödinger equation (TDSE) [5]. A clear structure in the kinetic energy spectra and three separate Coulomb

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explosion velocity groups corresponding to critical distances of about 8, 11, and 15 a.u. of H_2^+ and D_2^+ system were observed in experiment [6]; laser-induced dissociation and ionization of H_2^+ were studied using coincidence 3D momentum imaging [7].

While quantum methods can provide exact solutions to the multi-photon dynamics, the classical theories can also be used to study the same problem and the corresponding results are in good agreement with quantum calculations, such as strong-field ionization [8], the stabilization of a molecule in super-intense laser fields [9], etc. Recently, classical simulations of the dissociation, ionization and Coulomb explosion of H_2^+ employing a 1D model have also been studied [10–12].

In this paper, we study the classical dynamics of Hydrogen molecular ion H_2^+ in an intense laser field using a 3D classical model by the symplectic method. We calculate the probabilities of the dissociation, ionization and Coulomb explosion and the average distance from electron to the mass-center for various laser parameters, and discuss the classical dynamics of Hydrogen molecular ion H_2^+ caused by the relative phase in two-color laser field.

The 3D classical model of H_2^+ is presented in section 2. The classical dynamics of H_2^+ in intense laser fields are investigated in section 3. Finally, the conclusions are given in section 4.

2. The classical approximation of 3D Hydrogen molecular ion H_2^+

The classical three-body Hamiltonian with one-dimensional nuclear motion restricted to the direction of the laser field and the electron is allowed to move in three dimensions is given in this section. For the classical approximation of 3D Hydrogen molecular ion H_2^+ in an intense laser field, the molecular rotation is neglected by restricting the nuclei to move only along the direction of the electric field. Thus the classical Hamiltonian of H_2^+ in intense laser field can be given by (in atomic units):

$$H\left(x, p_x; y, p_y; z, p_z; R, p_R; t\right) = \frac{p_x^2}{2\mu_e} + \frac{p_y^2}{2\mu_e} + \frac{p_z^2}{2\mu_e} + \frac{p_R^2}{2\mu_p} + V_c\left(x, y, z, R\right) + V_{ex}\left(x, t\right). \quad (1)$$

Where R is the separation between the protons and p_R is the corresponding conjugate momentum, x, y, z are the coordinates in 3-dimensional Cartesian system of the electron from the center-of-mass of the protons with their corresponding conjugate momentum p_x, p_y, p_z, respectively. The reduced masses are $\mu_p = m_p/2$ and $\mu_e = 2m_e m_p/(2m_p + m_e)$, where m_e, m_p represent the masses of electron and nucleus, respectively.

$$V_c(x, y, z, R) = \frac{1}{R} - \frac{1}{\sqrt{\left(x - \frac{R}{2}\right)^2 + y^2 + z^2}} - \frac{1}{\sqrt{\left(x + \frac{R}{2}\right)^2 + y^2 + z^2}} \tag{2}$$

is the Coulomb interaction between electron and protons, and

$$V_{ex}(x, t) = -\sigma x E(t). \tag{3}$$

is the interaction potential with the intense laser field $E(t)$, where

$$\sigma = 2\left(m_p + m_e\right)/\left(2m_p + m_e\right).$$

Because of the singularities of the bare Coulomb potential $V_c(x, y, z, R)$, numerical integration becomes unstable. Such singularities can be removed by using the regularized coordinates [13]. The usual remedy is to use a screened Coulomb potential [11]

$$V_{sc}(x, y, z, R) = \frac{1}{R} - \frac{1}{\sqrt{\left(x - \frac{R}{2}\right)^2 + y^2 + z^2 + q_e}} - \frac{1}{\sqrt{\left(x + \frac{R}{2}\right)^2 + y^2 + z^2 + q_e}}. \tag{4}$$

In equation (4), we introduce a screened parameter q_e. In figure 1 we show the screened Coulomb potential $V_{sc}(x, y, z, R)$ between the electron and the protons of H_2^+ for the fixed $R = 2.0$ a.u. in x and z directions with $q_e = 1.0, 0.1, 0.06$. 0.0. We choose screened parameter $q_e = 0.06$ in our computation because the shape of the screened potential with $q_e = 0.06$ is closer to the bare potential curve.

We assume that, $p = (p_x, p_y, p_z, p_R)^T$, $q = (x, y, z, R)^T$, then the Hamiltonian function (1) can be written in the form

$$H\left(x, p_x; y, p_y; z, p_z; R, p_R; t\right) = H(p, q, t) = T(p) + V(q, t), \tag{5}$$

and $T(p) = \frac{p_x^2}{2\mu_e} + \frac{p_y^2}{2\mu_e} + \frac{p_z^2}{2\mu_e} + \frac{p_R^2}{2\mu_p}$, $V(q, t) = V_{sc}(x, y, z, R) + V_{ex}(x, t)$. It is a separable Hamiltonian system whose Hamiltonian function contains the time variable [14]. The corresponding Hamiltonian canonical equations of the system is

$$\begin{aligned} \frac{dq}{dt} &= \frac{\partial T(p)}{\partial p} \\ \frac{dp}{dt} &= -\frac{\partial V(q, t)}{\partial q} \end{aligned} \tag{6}$$

We choose a set of initial stable states $\left(x(0), p_x(0), y(0), p_y(0), z(0), p_z(0), R(0), p_R(0)\right)$ and solve the equation (6) numerically in order to get the corresponding time evolution of the electronic position $(x(t), y(t), z(t))$ and the internuclear separation $R(t)$.

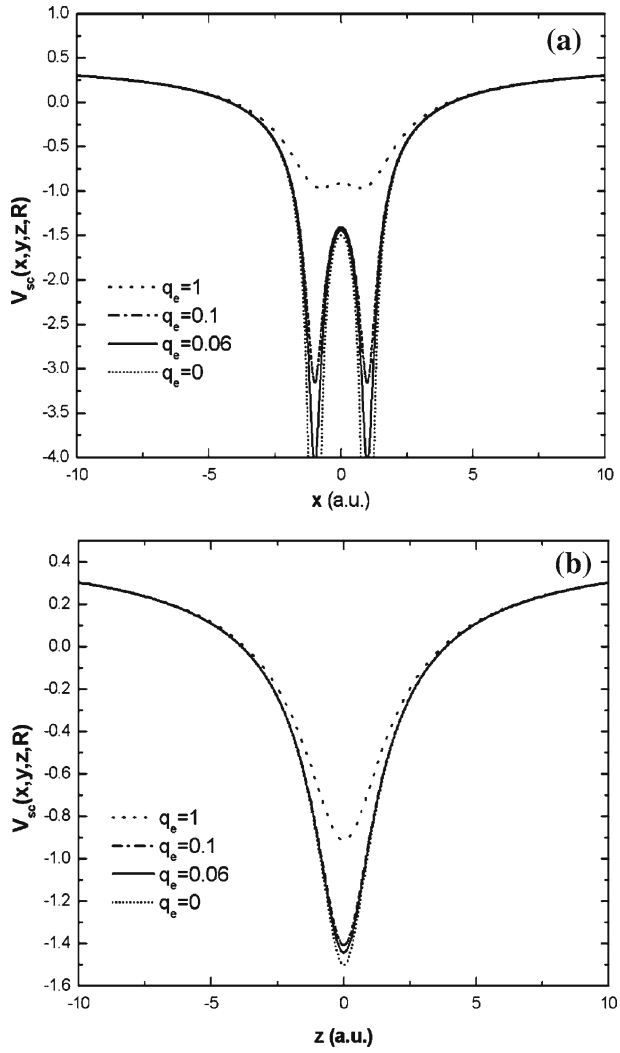

Figure 1. (a) The interaction potential between the electron and the protons of H_2^+ for the fixed R = 2.0 a.u. and y = 0,, z = 0 in x direction. (b) The interaction potential between the electron and the protons of H_2^+ for the fixed R = 2.0 a.u. and x = 0, y = 0 in z direction.

2.1. *Symplectic method*

As we know, the symplectic method is the difference method that preserves the symplectic structure, and is a better method in the long-time many-step calculations. Since the Hamiltonian system (6) is a separable Hamiltonian system whose Hamiltonian function contains the time variable, we may use an explicit symplectic scheme to solve it so that we can calculate the classical trajectories of

Hydrogen molecular ion H_2^+ in the intense laser field [14, 15]. For example, the following four-stage fourth-order explicit symplectic scheme can be adopted to solve Hamiltonian canonical equation (6) [16]:

$$
\begin{aligned}
q^1 &= q_n + \tau d_1 \left(\frac{\partial T}{\partial p}\right)_{p_n}, \quad p^1 = p_n - \tau c_1 \left(\frac{\partial V}{\partial q}\right)_{q^1,t_n}, \quad t^1 = t_n + \tau d_1 \\
q^2 &= q^1 + \tau d_2 \left(\frac{\partial T}{\partial p}\right)_{p^1}, \quad p^2 = p^1 - \tau c_2 \left(\frac{\partial V}{\partial q}\right)_{q^2,t^1}, \quad t^2 = t^1 + \tau d_2 \\
q^3 &= q^2 + \tau d_3 \left(\frac{\partial T}{\partial p}\right)_{p^2}, \quad p^3 = p^2 - \tau c_3 \left(\frac{\partial V}{\partial q}\right)_{q^3,t^2}, \quad t^3 = t^2 + \tau d_3 \\
q_{n+1} &= q^3 + \tau d_4 \left(\frac{\partial T}{\partial p}\right)_{p^3}, \quad p_{n+1} = p^3 - \tau c_4 \left(\frac{\partial V}{\partial q}\right)_{q_{n+1},t^3}, \quad t_{n+1} = t^3 + \tau d_4
\end{aligned}
$$

where $p^j, q^j, t^j, j = 1, 2, 3$ are intermediate stages, and $c_1 = 0$, $c_2 = c_4 = \alpha$, $c_3 = \beta$, $d_2 = d_3 = (\alpha + \beta)/2$, $d_1 = d_4 = \alpha/2$, $\alpha = (2 - 2^{1/3})^{-1}$, $\beta = 1 - 2\alpha$.

2.2. The initial states

In order to solve numerically canonical equation (6), the initial conditions can be chosen by using a single trajectory in the field-free case at random in time intervals [17]. The stable initial state $x(0)$, $p_x(0)$, $y(0)$, $p_y(0)$, $z(0)$, $p_z(0)$, $R(0)$, $p_R(0)$) is somewhat arbitrary, and we could use many methods to choose the initial values, such as the Monte Carlo method, the ensemble method etc. The justification is based on the ergodicity of the system [11]. First, we give the initial energy -0.60132 a.u. and the internuclear separation $R = 2.0$ a.u., which are equal to those of the ground state of the corresponding quantum system. Next we choose a set of (N) initial coordinates of electron x_i, y_i, z_i $(i = 1, 2, \ldots, N)$ at random and set $p_{R_i} = 0$, $p_{x_i} : p_{y_i} : p_{z_i} = \sqrt{2} : 1 : 1$ for convenience. Then we could get the values of p_{x_i}, p_{y_i}, p_{z_i} by the following Hamiltonian of H_2^+ in field-free

$$
H\left(x, p_x; y, p_y; z, p_z; R, p_R\right) = T(p) + V_{sc}(x, y, z, R) = -0.60132,
$$

which can guarantee that the energy is equal for N initial values chosen in such way. Finally we solve numerically the initial value problem of the canonical equations of H_2^+ system in the field-free by symplectic method for every initial condition $(x_i, p_{x_i}, y_i, p_{y_i}, z_i, p_{z_i}, R_i, p_{R_i})$ and obtain the classical trajectories in phase space. In such way, we can obtain a set of (N) classical trajectories corresponding to the N initial conditions. We select M points on every curve at random and obtain the interactive NM = N × M initial conditions. We solve numerically the canonical equation (6) of H_2^+ system in intense laser fields with

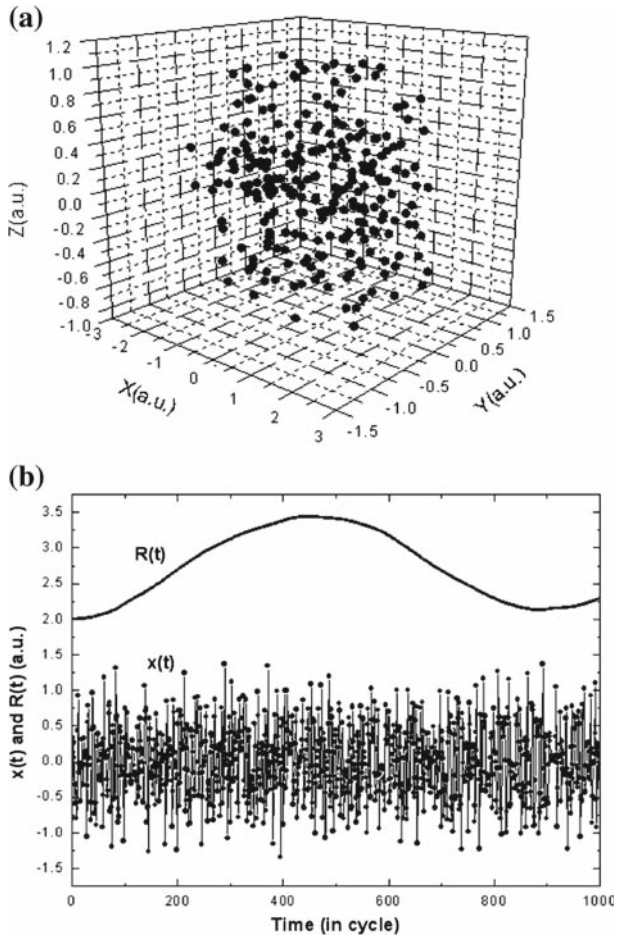

Figure 2. The time evolution of a typical trajectory (electronic position and the internuclear separation) in field-free. The time step is $\tau = 0.01$ a.u. (a) The distribution of the electronic position; (b) The nuclear position and electronic position in x direction.

these NM initial conditions and can obtain NM classical trajectories of H_2^+ in the laser pulse.

The time evolution of a typical trajectory (electronic position and the internuclear separation) of the 3D Hydrogen molecular ion in field-free is displayed in figure 2. One can see that the electron and the nuclei oscillate near the equilibrium position in field-free.

Figure 3 shows the time evolution of the energy of the Hydrogen molecular ion H_2^+ for a typical trajectory in field-free by symplectic method and Runge-Kutta method, respectively. One can see that the evolution of the energy in field-free by using symplectic method can be preserved for long-time computation, but the energy in field-free by using Runge-Kutta method decreases

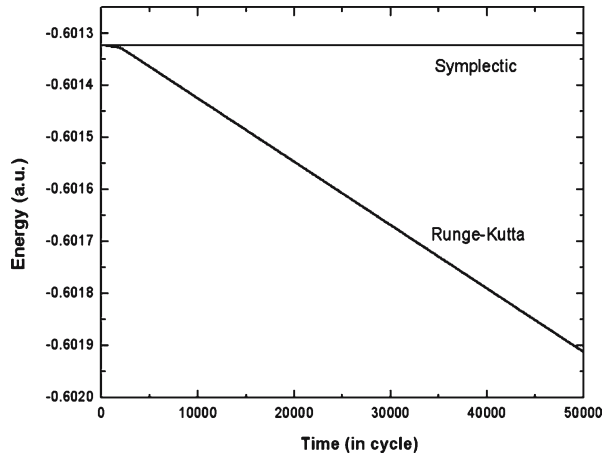

Figure 3. The time evolution of the energy of the Hydrogen molecular ion H_2^+ in field-free by fourth-order symplectic scheme and fourth-order Runge-Kutta scheme, respectively.

rapidly for long-time computation. Thus by using symplectic method, all NM initial conditions chosen for solving numerically the canonical equation (6) of H_2^+ system in intense laser fields can guarantee the energy is equal. It is shown that the initial conditions calculated by symplectic method are reasonable and effective.

2.3. *Probabilities of survival, ionization, dissociation and Coulomb explosion*

We solve numerically NM initial value problems of canonical equation (6) of H_2^+ system in intense laser fields and obtain NM classical trajectories of H_2^+ in the laser pulse. In the computation we choose N = 1000, M = 100. If we choose more points, the results are almost the same.

From NM classical trajectories, we can get the compensated energy of the electron [17] and the total energy of the electron

$$E_x(t) = \frac{p_x^2 + p_y^2 + p_z^2}{2\mu_e} - \frac{1}{\sqrt{\left(x - \frac{R}{2}\right)^2 + y^2 + z^2 + q_e}} - \frac{1}{\sqrt{\left(x + \frac{R}{2}\right)^2 + y^2 + z^2 + q_e}}. \tag{7}$$

Ionization will occur when the total energy of the electron is greater than zero. Some authors use the compensated energy of the electron $E_c(t)$ as the criteria of ionization. But in our study, the compensated energy $E_c(t)$ oscillates quickly with time. In order to eliminate this oscillation, we use $E_x(t)$ as the criterion of

ionization and determine the time at which the ionization occurs. Whereas H_2^+ is thought to be dissociated when the internuclear separation $R(t) > R_d = 9.5$ a.u. [17]. One of the following four dynamic processes must occur at any time interval during the interaction of H_2^+ in an intense laser field:

$$\begin{array}{ll} H_2^+ + [I] \to H_2^+ & (E_x(t) \leqslant 0, R(t) \leqslant R_d) \\ H_2^+ + [I] \to H_2^{2+} + e & (E_x(t) > 0, R(t) \leqslant R_d) \\ H_2^+ + [I] \to H + H^+ & (E_x(t) \leqslant 0, R(t) > R_d) \\ H_2^+ + [I] \to 2H^+ + e & (E_x(t) > 0, R(t) > R_d) \end{array} . \tag{8}$$

These four processes are defined as survival, ionization, dissociation and Coulomb explosion or dissociative ionization [17], respectively.

Suppose T is the cycle of laser pulse and take a sufficiently large positive integer Z. Let the time step be $\tau = T/Z$ and denote $t_k = k\tau$, $k = 0, 1, 2, \ldots, Z$. Using the symplectic method, we can gain NM classical trajectories of H_2^+ system in the intense laser pulse. For every time t_k, we compute the number of classical trajectories of survival, ionization, dissociation and Coulomb explosion according to the criteria (8). For every time t_k, suppose S_{sur}, S_{ion}, S_{dis}, S_{exp} are the numbers of the classical trajectories of survival, ionization, dissociation and Coulomb explosion, respectively, then we obtain the probabilities of survival, ionization, dissociation and Coulomb explosion at every time t_k which are

$$P_{\mathrm{s}} = S_{sur}/NM, \quad P_{\mathrm{i}} = S_{ion}/NM, \quad P_{\mathrm{d}} = S_{dis}/NM, \quad P_{\mathrm{C}} = S_{\mathrm{exp}}/NM.$$

3. Results and discussions

We give results of the classical trajectory calculations of the interaction between 3D H_2^+ and the intense laser field and discuss the dynamic processes of survival, ionization, dissociation and Coulomb explosion in single-color and two-color laser fields.

3.1. *Single-color laser field*

We first consider the dynamic processes of survival, ionization, dissociation and Coulomb explosion of H_2^+ system in the intense laser field. The electric field was described by the formula

$$E = E_0 f(t) \sin n\omega_0 t, \tag{9}$$

which is assumed to be along the x axis, where E_0 is the peak amplitude of the laser field, $n\omega_0$ is the laser frequency, where n is a parameter. $f(t)$ is a laser

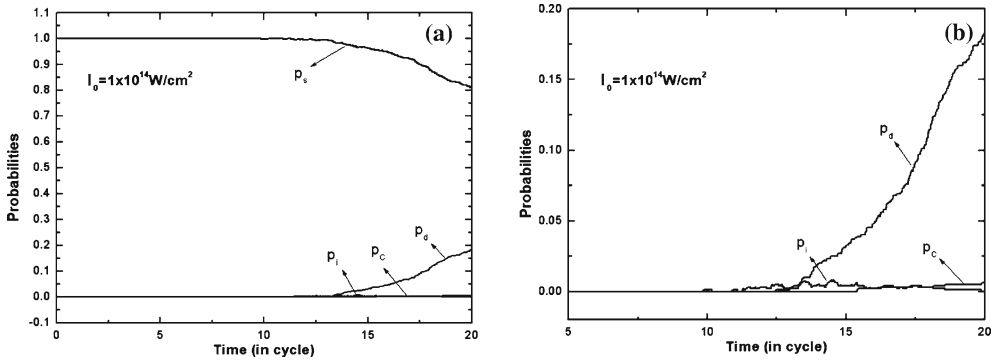

Figure 4. The time evolution of the probabilities of survival (p_s), ionization (p_i), dissociation (p_d) and Coulomb explosion (p_C) in a single-color intense laser field at intensity $I_0 = 1.0 \times 10^{14}$ W/cm^2. (a) The time evolution of the probabilities, (b) The enlarged picture of part of (a).

envelope, here we use the following pulse shape

$$f(t) = \begin{cases} \sin^2 \frac{\pi t}{20T_0} & 0 < t < T_d \\ 0 & otherwise \end{cases}, \tag{10}$$

where $T_d = 20T_0$, and $T_0 = \frac{2\pi}{n\omega_0}$ is the period of laser pulse. For all the calculations reported below, we assumed $\omega_0 = 0.08565$ a.u. When $n = 1$, its corresponding wavelength is 532 nm.

In order to illustrate the time-evolution of the classical dynamic processes, we first calculate the classical trajectories of the survival, ionization, dissociation, and Coulomb explosion in the single-color intense laser field, and then obtain the probabilities of survival, ionization, dissociation, and Coulomb explosion. The time-evolution of probabilities is displayed in figure 4 when the intensity is $I_0 = 1.0 \times 10^{14}$ W/cm^2 ($E_0 = 0.053$a.u.) and the parameter $n = 1$.

We can observe from figure 4 that in the weak laser field ($I_0 = 1.0 \times 10^{14}$ W/cm^2) the ionization channel is turned on earliest and then the dissociation channel and Coulomb explosion channel are turned on. As time increases, the dissociation probability is larger than ionization and Coulomb explosion probability. This phenomenon might be explained in the following way: No matter how weak the field is, the slight electron could easily get the energy from the laser pulse for ionization, however, in such a short time, the heavy nuclei could not be separated apart. Then the electron may move close to one of nucleus gradually and finally be captured, so they form the neutral atom H, the Hydrogen molecular ion dissociates, meanwhile, as a result of interacting with the laser field, the other nucleus having a unit of positive charge becomes far away from the neutral atom H. When the direction of laser field is changed, the same circumference occurs on another nuclear. Two approaches of the mechanism of dissociation may exist:

(1) The dissociation from H_2^+ directly. With Coulomb interaction between two nuclei of H_2^+, the nucleus may be separated apart rapidly while the electron still attaches to one of them. Thus the H and H^+ are formed, i.e., the dissociation occurs.
(2) After being ionized from H_2^+, the detached electron may gradually move and close to one of the nuclei and then be captured again, and then the dissociation happens.

As a result, in the weak laser field, the dissociation process is predominant.

In Ref. [18–21], the time evolution of the dissociation and the ionization probability was calculated by solving numerically the 1+3D time-dependent Schrödinger equation, Their results (Figure 1(a) in Ref. [18]) showed the same tendency with the classical result that when the laser intensity is weak ($3.5 \times 10^{13}\,\mathrm{W/cm^2}$), the ionization channel is turned on earlier than the dissociation channel of the initial $v = 6$ state; the dissociation probability is larger than ionization probability. All these illustrate that the dissociation progress is predominant in the weak laser field.

In figure 5, we show the time-evolution of survival, dissociation, ionization and Coulomb explosion of H_2^+ for different high intensities of laser fields with the wavelength $\lambda = 532\,\mathrm{nm}$. From figure 5, we can observe that the ionization channel is turned on earliest; the dissociation channel and the Coulomb explosion channel are turned on almost at the same time when the ionization probability reaches its peak value; then the ionization probability reduces rapidly, meanwhile, the dissociation probability goes up slightly, and the Coulomb explosion probability goes up rapidly and reaches its peak value, then almost keeps unchanged. This phenomenon could be explained as follows: The electron gets the energy from the laser pulse rapidly until it is ionized, the heavier nuclei is not detached at that time, so the ionization channel is turned on first and the H_2^{2+} is formed, and the probability of ionization rises with time. Subsequently, two nuclei of H_2^{2+} are separated rapidly under the Coulomb force of each other, thus the Coulomb explosion appears. The probability of Coulomb explosion arrives at the same peak value of the ionization probability till H_2^{2+} disappears, and then keeps unchanged. There is no evident change for the probability of dissociation with the increasing of laser intensity. This is because that the electron is ionized rapidly under ultrashort intense laser, and it is too late for the nuclei with big mass to respond when the pulse ends. We can also observe that the larger the laser pulse intensity is, the earlier the three channels are turned on. We also find that the peak value of Coulomb explosion is a little larger than that of ionization. Some H atom originated from dissociation progress is ionized into H^+ ion, which will enhance the Coulomb explosion probability. During the process of the laser-matter interaction, ionization, dissociation and Coulomb explosion occur in turns:

$$H_2^+ \rightarrow H_2^{2+} + e \rightarrow 2H^+ + e \rightarrow H + H^+$$

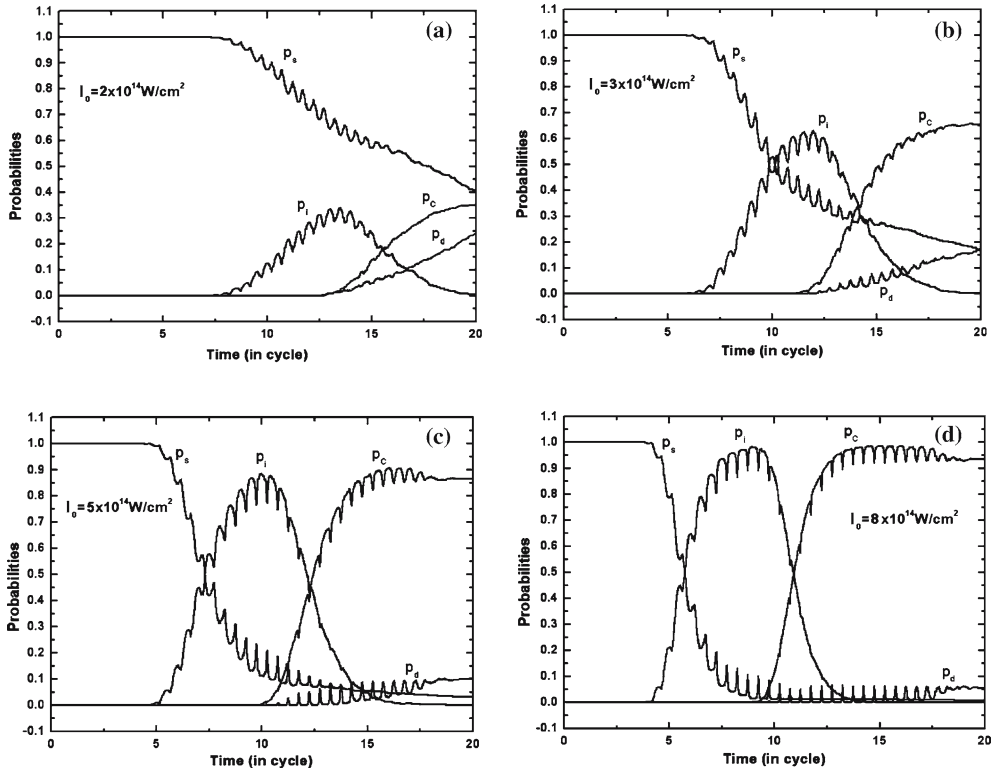

Figure 5. The time evolution of the probabilities of survival (p_s), ionization (p_i), dissociation (p_d) and Coulomb explosion (p_C) in the single-color intense laser field at different laser intensities. (a) $I_0 = 2.0 \times 10^{14}$ W/cm^2, (b) $I_0 = 3.0 \times 10^{14}$ W/cm^2, (c) $I_0 = 5.0 \times 10^{14}$ W/cm^2, (d) $I_0 = 8.0 \times 10^{14}$ W/cm^2.

It should be mentioned that the dissociation probability becomes smaller as the laser intensity becomes larger in the single-color laser field. This is attribute to the fact that the probabilities of the two nuclei are separated but the electron is still bounded around one of the nuclei are very seldom when such an intense ultrashort pulse is used. However, the fact that the electron does not obtain the energy from laser pulse but the nuclear do during the interacting will seldom occur. In fact, when the laser intensity becomes larger, the probability that the two nuclei are separated but the electron is still bounded around one of the nuclei is very small. Furthermore, in our 3D classical model, the electron moves along different directions which will reduce the dissociation probabilities.

In Ref. [18, 20], the ionization and dissociation probabilities of the initial $v = 6$ vibrational state were calculated at $I = 1.0 \times 10^{14}$ W/cm^2 and $I = 1.0 \times 10^{15}$ W/cm^2 by using the quantum method. Under the interaction with the photons the Hydrogen molecular ion is ionized first and then dissociated. In weak field, the dissociation probability is much larger than ionization probability, whereas in the strong field this is not the case, i.e., the latter is much larger than

the former; when the ionization probability is close to the peak value the dissociation channel is turned on and the dissociation probability is small all the time. Despite the initial states are different from those we used, the quantum-mechanical calculations are in agreement with our classical results qualitatively with the explanations stated above.

We also display the time evolution of the probabilities of survival, ionization, dissociation, and Coulomb explosion in the single-color intense laser field for different laser frequencies in figure 6. Here we choose the laser intensity $I_0 = 4.0 \times 10^{14}\,\mathrm{W/cm^2}$. We could see from figure 6 that the larger the laser frequency is, the earlier the three channels turned on. The oscillations of the probabilities are stronger and the amplitude of the oscillations becomes larger as the frequency of the laser field increases. We can also observe from figure 6 that the dissociation probability become larger as the laser frequency increases, which can be interpreted as follows: The energy of one-photon becomes larger with the increasing of the laser frequency, the ionized electron in Hydrogen molecular ion moves in the field and then the probability of electrons bounded by one of the nucleus increases, which result in the enhancement of dissociation probability. When the laser frequency is added to some extent, the dissociation probability is not increasing any more.

3.2. *Two-color laser field*

We consider the following two-color intense laser pulse:

$$E(t) = E_0 f(t)\left[\cos(\omega_0 t) + \frac{1}{2}\cos(2\omega_0 t + \phi)\right],$$

where E_0 is the laser peak intensity, ω_0 is the fundamental laser frequency, ϕ is the relative phase. The pulse shape $f(t)$ is the same as equation (10). We discuss the time evolution of probabilities for different laser intensities and the classical motion of the electron and the nuclei for different parameters in two-color laser fields, and also illustrate the classical dynamics of Hydrogen molecular ion H_2^+ in two-color laser fields.

(a) The time evolution of probabilities for different intensities

In figure 7, we show the time evolution of the probabilities of survival (p_s), ionization (p_i), dissociation (p_d), and Coulomb explosion (p_C) in the two-color intense laser field for different laser intensities, the relative phase ϕ is assumed to be zero.

For weak laser intensities, compared to the singe-color field (see figure 5(a,b)), the ionization, dissociation and Coulomb explosion channels are turned on a little earlier, and the peak values of the probabilities of ionization and Coulomb explosion are larger in two-color laser field (see figure 7(a,b)). These indicate that using the lower two-color laser intensities instead of the higher single-

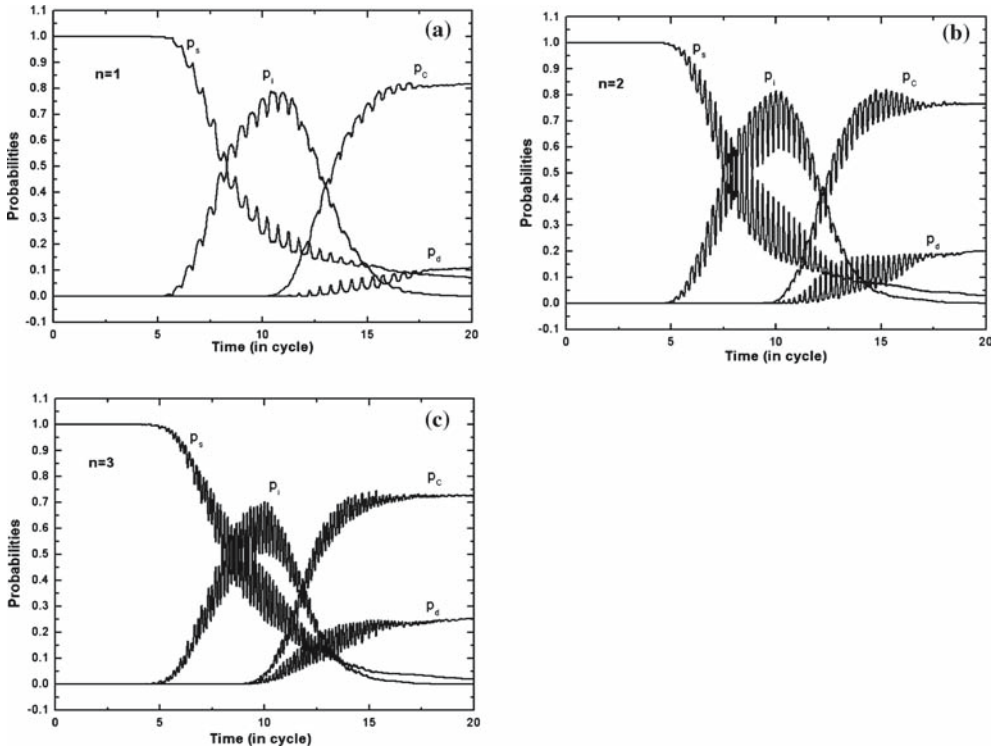

Figure 6. The time evolution of the probabilities of survival (p_S), ionization (p_i), dissociation (p_d), and Coulomb explosion (p_C) in single-color intense laser field with $E_0 = 4.0 \times 10^{14}$ W/cm^2 for different frequencies. (a) $n = 1$, (b) $n = 2$, (c) $n = 3$.

color intensities could obtain the same production in a sense. But for stronger laser intensities (see figure 5(d) and 7(d)), the classical dynamics are almost the same in one-color laser field and two-color laser field. From figure 5(b) and figure 7(a) we know that the total laser intensity $I_0 = 3.0 \times 10^{14}$ W/cm^2. To our surprise, the survival, dissociation, ionization, and Coulomb explosion probability in one-color laser field is larger than those in two-color laser field while the total laser intensity is equal.

The dissociation probability also becomes smaller with the increasing laser intensity in two-color laser field. This phenomenon is the same as that in single-color laser field.

(b) The motion of the electron and the nuclei for different laser parameters

Figure 8 shows the average distance from electron to the mass-center for various relative phases. The overall response of the electron is different for various relative phases. We observe the symmetric oscillation of the quick response of electron, which is reflecting the shape of the laser amplitude. We found that the distribution of the electron is quite biased to one side for both cases

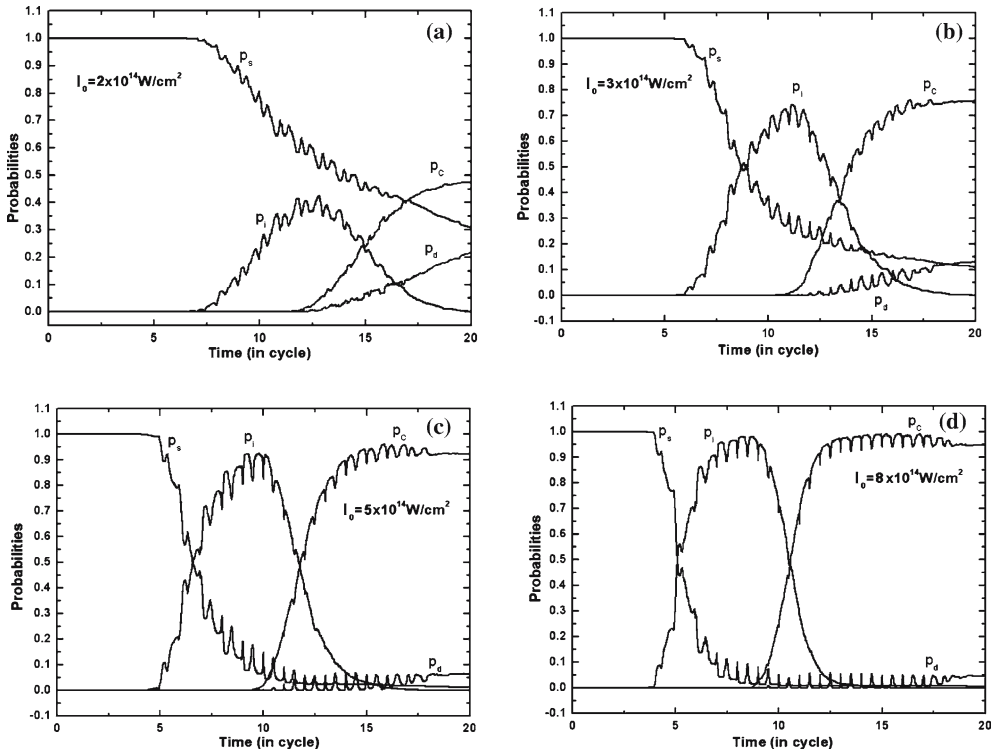

Figure 7. The time evolution of the probabilities of survival (p_s), ionization (p_i), dissociation (p_d) and Coulomb explosion (p_C) in two-color intense laser field with $\phi = 0$ for different intensities. (a) $I_0 = 2.0 \times 10^{14}$ W/cm^2, (b) $I_0 = 3.0 \times 10^{14}$ W/cm^2, (c) $I_0 = 5.0 \times 10^{14}$ W/cm^2, (d) $I_0 = 8.0 \times 10^{14}$ W/cm^2.

of $\phi = 0$, $\phi = \frac{3\pi}{2}$ and $\phi = \frac{\pi}{2}$, $\phi = \pi$, which indicates that the direction of electronic movement changes as the phase changes. We could get the conclusion from figure 8 that the average distance of electric position changes greatly not only in directions but also in values for various phase-differences. The quantum-mechanical calculation also refers to the asymmetric electron-nuclear dynamics of photo-fragments in H_2^+ through the laser phase directional control, and the phenomenon we observed is similar to the quantum calculation [22], which has explicit explanations.

Figure 9 shows the change of the average internuclear separation $\overline{R(t)}$ with the time in two-color laser field. One can see the response of nucleus with the increasing of laser intensity. The average internuclear separation $\overline{R(t)}$ increases slowly until the electron is ionized, the internuclear distance increases rapidly due to the Coulomb explosion after the electron is ionized. As the laser intensity increases, internuclear separation $\overline{R(t)}$ grows more quickly. We can also use quantum theory to explain this phenomena: the nuclei move as free particles

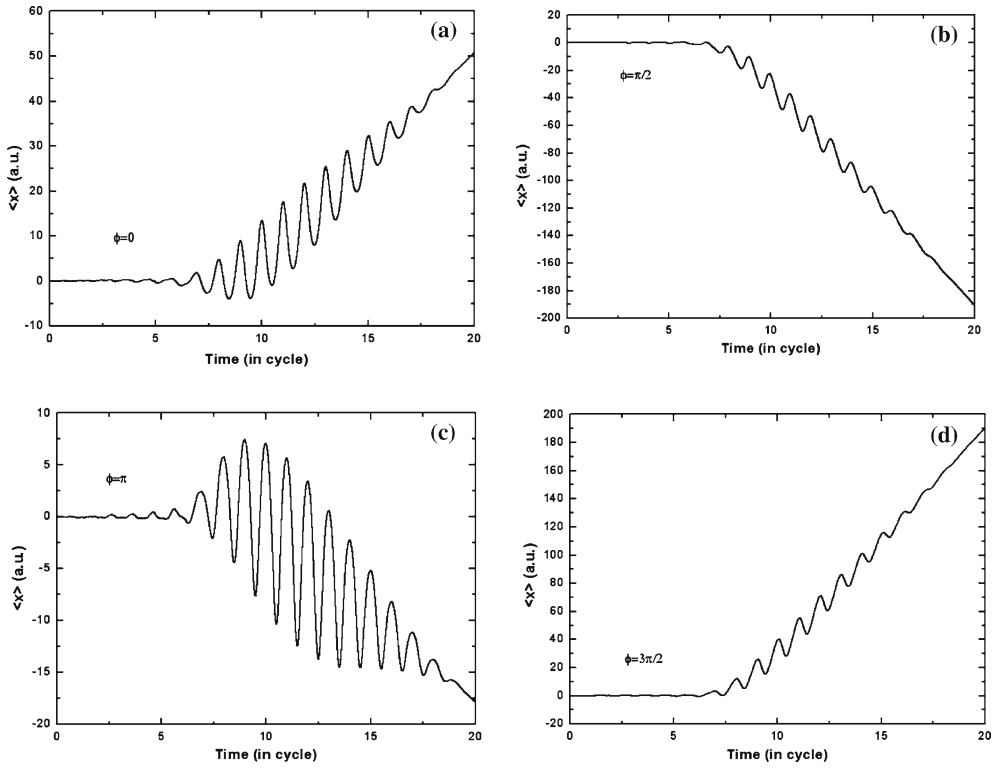

Figure 8. The average distance from electron to the mass-center for $I_0 = 3.0 \times 10^{14}$ W/cm^2 and various relative phases. (a) $\phi = 0$, (b) $\phi = \frac{\pi}{2}$, (c) $\phi = \pi$, (d) $\phi = \frac{3\pi}{2}$.

when they are separated from each other, and the energy of the nuclei is close to the vibrational energy. Such a quasi-static picture is expected to be valid for low laser intensities, in which the electron cloud in the laser field is shifted to the nuclear and consequently the bond is very weak; and another nuclear is accelerated in the laser field. One can see that the average separation velocity of nuclei increases with the increasing laser intensity. Therefore, the stronger laser pulse would enhance the ratio of high-energy parts of the kinetic energy spectrum of the nuclei. The result that we have got is in agreement with those obtained by 1D model [17] (Figure 3 in Ref. [17]) and similar to those calculated by quantum method [20] (Figure 7 in Ref. [20]).

4. Conclusions

The classical dynamics of Hydrogen molecular ion H_2^+ in intense fields are studied by the classical theory and the symplectic method. In this paper, 3D model of H_2^+ is used, with the bare Coulomb potential replaced by a

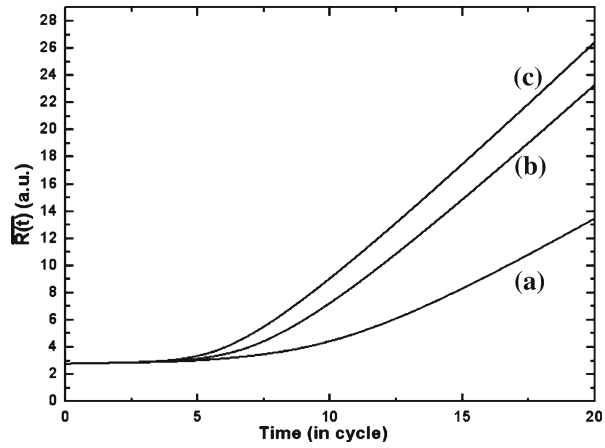

Figure 9. The time dependence of the average internuclear separation $\overline{R(t)}$ for different laser intensities. The curves (a), (b), and (c) correspond to the laser pulse intensities 2.0×10^{14} W/cm^2, 5.0×10^{14} W/cm^2 and 8.0×10^{14} W/cm^2, respectively.

screened potential to avoid the singularities at close electron–proton encounters. The initial conditions are chosen at random in the field-free case, and then the Hamiltonian canonical equations of H_2^+ system in the intense laser field are solved numerically by means of the symplectic method under these initial conditions. We discuss the probabilities of ionization, dissociation, and Coulomb explosion of the H_2^+ system in the one-color and two-color laser field. As expected, the ionization and Coulomb explosion probabilities increase and the dissociation probability decreases with the increasing of intensity. The peak of the ionization process occurs sooner for higher laser intensity. Once ionized, the Coulomb energy between the nuclei will be released quickly via the Coulomb explosion. This leads to the rapid increasing of internuclear separation. So the dissociative ionization channel follows ionization directly. We could also observe that the dissociation probability is much larger than ionization probability in weak laser field; this phenomenon could also be seen in the quantum calculations using the exact 1+3D model.

In the 3D model the dissociation probability is smaller in the intense laser field as the electron departs in different directions. In addition, we also calculate the average distance from electron to the mass-center for various relative phases and the change of the average internuclear separation $\overline{R(t)}$ with the time, which illustrate that the stronger laser pulse would enhance the ratio of high-energy parts of the kinetic energy spectrum of the nuclei. Although the classical trajectory method can't describe some quantum effects, it is in agreement with quantum-mechanical calculations and may be extended to larger molecular system.

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References

[1] P. Dietrich and P.B Corkum, J. Chem. Phys. 97 (1992) 3187.
[2] P.H. Buksbaum, A. Zavriyev, H.G. Muller and D.W. Schumacher, Phys. Rev. Lett. 64 (1990) 1883.
[3] X. Urbain, B. Fabre, E.M. Staicu-Casagrande, N. de Ruette, V.M. Andrianarijaona, J. Jureta, J.H. Posthumus, A. Saenz, E. Baldit and C. Cornaggia, Phys. Rev. Lett. 92 (2004) 163004.
[4] X.M. Tong, Z.X. Zhao and C.D. Lin, Phys. Rev. Lett. 91 (2003) 233203.
[5] A.D. Bandrauk and H.Z. Lu, Phys. Rev. A 62 (2000) 053406.
[6] D. Pavicic, A. Kiess, T.W. Hansch, H. Figger, Phys. Rev. Lett. 94 (2005) 163002.
[7] I. Ben-Itzhak, P.Q. Wang, J.F. Xia, A.M. Sayler, M.A. Smith, K.D. Carnes, B.D. Esry, Phys. Rev. Lett. 95 (2005) 073002.
[8] D.A. Wasson and S.E. Koonin, Phys. Rev. A 39 (1989) 5676.
[9] R. Grobe and C.K. Law, Phys. Rev. A 44 (1991) R4114.
[10] D.M. Villeneuve, M.Y. Ivanov and P.B. Corkum, Phys. Rev. A 54 (1996) 736.
[11] Y.W. Duan, W.K. Liu and J.M. Yuan, Phys. Rev. A 61 (2000) 053403.
[12] S.X. Liu, Y.Q. Lang, X.S. Liu, P.Z. Ding and Y.Y. Qi, J. Math. Chem. 41 (2006) 113.
[13] K. Richter, G. Tanner and D. Wintgen, Phys. Rev. A 48 (1993) 4182.
[14] Th. Monovasilis, T.E. Simos, Chem. Phys. 313 (2005) 293.
[15] K. Tselios, T.E. Simos, J. Math. Chem. 35 (2004) 55.
[16] X.S. Liu, L.W. Su, P.Z. Ding, Intern. J. Quantum Chem. 87 (2002) 1.
[17] W.X. Qu, S.X. Hu and Z.Z. Xu, Phys. Rev. A 57 (1998) 2219.
[18] S. Chelkowski, T. Zuo, O. Atbek and A.D. Bandrauk, Phys. Rev. A. 52 (1995) 2977.
[19] T. Zuo and A.D. Bandrauk, Phys. Rev. A 52 (1995) R2511.
[20] S. Chelkowski, A. Conjusteau, T. Zuo and A.D. Bandrauk, Phys. Rev. A 54 (1996) 3235.
[21] S. Chelkowski, T. Zuo and A.D. Bandrauk, Phys. Rev. A 46 (1992) R5342.
[22] Y. Ohta, J. Maki, H. Nagao, K. Nishikawa, Inter. J. Quantum Chem. 97 (2004) 891.