

Two-particle one-dimensional model of the hydrogen molecular ion in an ultrashort laser pulse

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We consider a one-dimensional model of the simplest molecular system, H_2^+ , via direct numerical integration of the time-dependent Schrödinger equation. We consider its evolution in an ultrashort laser pulse of high intensity. The probabilities of excitation of different electronic and vibrational states, as well as the probabilities of dissociation and ionization are calculated as functions of the intensity of the laser field. © 1996 American Institute of Physics. [S1063-7761(96)00611-7]

1. INTRODUCTION

The dynamics of atomic and molecular systems in the field of intense laser radiation has been the subject of numerous recent experimental and theoretical papers.¹ Such effects as above-threshold ionization of atoms and molecules, stabilization of atoms in ultrastrong fields, the formation of multiply-charged ions, Coulomb explosions of molecules, and other phenomena have been experimentally observed or theoretically predicted.^{1–4} The direct numerical integration of the time-dependent Schrödinger equation for a quantum system in the field of an electromagnetic wave is one of the basic approaches to the theoretical study of these processes.^{5–7}

However, the capabilities of modern computers usually forces one to the study of one-electron systems in the one-dimensional approximation. The number of papers treating three-dimensional systems is not large;^{5,6,8–12} They have considered the dynamics of the ionization of the hydrogen atom, the negative hydrogen ion, as well as He and Xe treated as one-electron systems, i.e., systems in which all of the electrons except one are “frozen.” The process of ionization of the simplest two-electron system, the negative hydrogen ion, has also been considered in the one-dimensional approximation.¹³

The theoretical study of the dynamics of molecular systems in the field of a strong electromagnetic wave is a much more complicated problem. Even the simplest molecular system, the hydrogen molecular ion, is a two-particle system when the nuclear degrees of freedom are taken into account. Such a system can be treated analytically only by resorting to a number of approximations. The dynamics of molecular systems is usually described using the adiabatic approximation in which the motion of the electrons is assumed to be much faster than that of the nuclei. The justification of applying this approximation to molecules perturbed by ultrashort optical fields, where the energies can be larger than the energy of relative motion of the nuclei, has not been made clear. In addition, in the case of a numerical solution the adiabatic approximation does not lead, in our view, to a simplification of the problem, since it requires the solution of a system of many equations describing the amplitudes for oc-

cupying a large number of states of the discrete spectrum, as well as the electronic and vibrational continua. Hence direct numerical integration of the time-dependent Schrödinger equation with the total Hamiltonian taking into account the interaction with the electromagnetic field¹⁴ is a more reliable method. However there is an additional difficulty in the numerical integration of the Schrödinger equation for molecular systems because of the presence of the small parameter m/μ (m is the mass of the electron and μ is the reduced mass of the molecule), which, on the one hand, reduces the region of localization of the nuclear wave function in comparison to the electronic wave function, and hence imposes stringent requirements on the spatial computational grid in the nuclear coordinate, and on the other hand implies slow motion of the nuclei and hence increases the computational time required to study the molecular dissociation products.

In the present paper we consider a one-dimensional model of the hydrogen molecular ion taking into account the electronic and nuclear motions. The dynamics of the processes of ionization and dissociation of the system in a femtosecond laser pulse are studied by direct numerical integration of the time-dependent Schrödinger equation with the total electron–nucleus Hamiltonian. The probabilities of occupying the different electronic and vibrational states of the molecule and the photodisintegration probabilities are calculated as functions of the laser intensity. The possibility of observing stabilization of the system against ionization and dissociation is discussed.

2. HYDROGEN MOLECULAR ION H_2^+ IN THE ADIABATIC APPROXIMATION

The simplest molecular system is the hydrogen molecular ion, in which the chemical bond between the protons is accomplished by a single electron. The total Hamiltonian of the system is

$$H_0 = -\frac{\hbar^2}{2m} \nabla_x^2 - \frac{\hbar^2}{2M} \nabla_{R_1}^2 - \frac{\hbar^2}{2M} \nabla_{R_2}^2 + \frac{e^2}{|\mathbf{R}_1 - \mathbf{R}_2|} + V_e(\mathbf{x}, \mathbf{R}_1, \mathbf{R}_2). \quad (1)$$

Here m and M are the mass of the electron and the proton; \mathbf{x} , \mathbf{R}_1 , and \mathbf{R}_2 are the coordinates of the electron and the two

protons. The first three terms in (1) describe the kinetic energy of the electron and the first and second protons, the fourth term is the energy of the Coulomb repulsion of the protons, and the fifth term is the interaction energy of the electron with each of the protons.

In a coordinate system whose origin is fixed at the center of mass, and neglecting the rotational motion of the nuclei, it is not difficult to obtain from (1)

$$H_0 = -\frac{\hbar^2}{2m} \nabla_x^2 - \frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} + V_e(\mathbf{x}, R) + \frac{e^2}{R}. \quad (2)$$

where $\mu = M/2$ is the reduced mass of the molecule, and $R = |\mathbf{R}_1 - \mathbf{R}_2|$ is the internuclear distance.

In the one-dimensional model of H_2^+ it is assumed that the electron can move only along the axis of the molecule. Then the Laplacian operator in (2) can be replaced by the second derivative $\partial^2/\partial x^2$. The potential energy is chosen in the form

$$V_e(x, R) = -\frac{e^2}{\sqrt{(R/2-x)^2 + \alpha^2}} - \frac{e^2}{\sqrt{(R/2+x)^2 + \alpha^2}}, \quad (3)$$

which corresponds to a smoothed, double-well Coulomb potential similar to that considered earlier for the one-dimensional hydrogen atom.¹⁵

The parameter α in our calculations was taken to be $0.943a_0$, where a_0 is the Bohr radius.

The calculation of the stationary states of the Hamiltonian (2) with the potential (3) is a difficult problem. But because of the small parameter $m/M \ll 1$ we can use the adiabatic approximation and represent the total wave function of the molecule in the form

$$\Psi(x, R) = \Phi(R) \varphi(x, R), \quad (4)$$

where $\Phi(R)$ is the nuclear function, which depends on the nuclear coordinate as a parameter.

The electron and nuclear wave functions are solutions of the time-independent Schrödinger equation with the Hamiltonians H_e and H_N , respectively:

$$H_e = -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial x^2} + V_e(x, R), \quad (5)$$

$$H_N = -\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} + V_{\text{eff}}(R), \quad (6)$$

where

$$V_{\text{eff}}(R) = e^2/R + E_e(R) \quad (7)$$

is the effective interaction energy of the nuclei (the electronic term of the molecule), including the Coulomb repulsion of the nuclei and their effective attraction because of the presence of the electron. The quantity $E_e(R)$ is the eigenvalue of the electron Hamiltonian, and it also depends on R as a parameter.

The lowest few electronic terms of the molecule calculated with the help of (7) are shown in Fig. 1. The terms correspond to even and odd electronic states of the molecule, where the odd states do not necessarily guarantee a bound state of the molecule. Also note that the equilibrium internu-

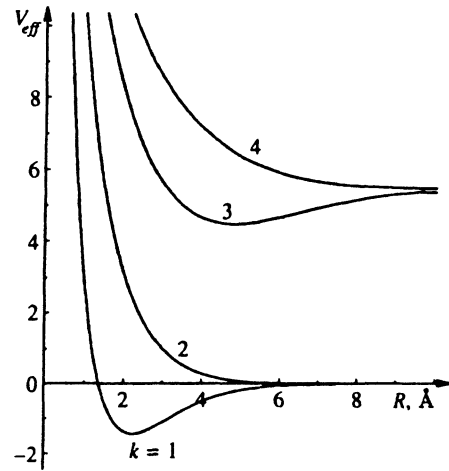


FIG. 1. Electronic terms of the one-dimensional hydrogen molecular ion for the states $k=1$ through 4.

clear distance, corresponding to the minimum of the curve V_{eff} , is significantly different for the different electronic states. The structure of the electronic terms of the one-dimensional H_2^+ molecule agree qualitatively with the terms of the real hydrogen molecular ion, although the quantitative agreement is poor. For example, in our model the potential well depth of the ground electronic term is ≈ 1.45 eV, while in the three-dimensional system it is ≈ 2.65 eV. The equilibrium distance between the nuclei is 2.2 and 1.06 Å in the one-dimensional and three-dimensional cases, respectively. Solution of the stationary problem for the nuclear subsystem with the Hamiltonian (6) gives the nuclear stationary states and the corresponding wave functions $\Phi_k^{(v)}(R)$ ($k=1,2,\dots$, $v=0,1,\dots$ are the electronic and vibrational quantum numbers of the molecule). The probability density in the ground state of the molecule is given by the expression

$$|\Psi(x, R)|^2 = |\Phi_1^{(0)}(R) \varphi_1(x, R)|^2. \quad (8)$$

Note that a wave function of the form (4) is not a solution of the time-independent Schrödinger equation with the total Hamiltonian (2),

$$H_0 \Psi(x, R) = E \Psi(x, R), \quad (9)$$

but it is quite close. To test the accuracy of the calculation of the wave function of the ground stationary state of H_2^+ in the adiabatic approximation, a wave function of the form (4) was substituted as the initial state into the nonstationary Schrödinger equation with the Hamiltonian (2)

$$i\hbar \frac{\partial \Psi(x, R, t)}{\partial t} = H_0 \Psi(x, R, t). \quad (10)$$

The time dependence of the square of the overlap integral of the wave function obtained from the solution (10) with the initial wave function (8) was calculated:

$$W_{10} = |C_{10}(t)|^2 = \left| \int \Psi^*(x, R, r) \Phi_1^{(0)} \times (R) \varphi_1(x, R) dx dR \right|^2. \quad (11)$$

The quantity $|C_{10}(t)|^2$, describing the probability of occupying the ground state of H_2^+ in the adiabatic approximation, oscillates in time with a period $T \approx 33$ fs and does not drop below the value 0.996. This means that the ground state of the system calculated in the adiabatic approximation is close to the actual ground state of H_2^+ and nonadiabatic transitions can be neglected. Our numerical calculations show that transitions inside the vibrational structure of levels belonging to a single electronic term are the most important. Therefore the adiabatic approximation is even more accurate in calculating the population densities of the different electronic states of the molecule.

3. INTERACTION OF H_2^+ WITH AN ELECTROMAGNETIC FIELD

The time evolution of the states of H_2^+ in the presence of an electromagnetic wave can be obtained from the solution of the time-independent Schrödinger equation

$$i\hbar \frac{\partial \Psi(x, R, t)}{\partial t} = (H_0 - e x \varepsilon(t) \cos \omega t) \Psi(x, R, t), \quad (12)$$

where H_0 is given by (1) and $\varepsilon(t)$ is the slowly-varying amplitude of the electromagnetic wave with frequency ω .

The laser pulse is assumed to have the gaussian form

$$\varepsilon(t) = \varepsilon_0 \exp\left[-\frac{1}{2} \left(\frac{t-t_0}{\tau}\right)^2\right],$$

where $\tau = 2$ fs is the half-width of the pulse and $t_0 = 6$ fs is the time of maximum intensity. The total computation time $t_p \approx 20$ fs was chosen in order that the dynamics of the system (especially the nuclei) after the passage of the laser disturbance could be studied. The calculations were performed for radiation intensities $P = 10^{12} - 10^{15}$ W/cm² and for the frequency of the laser field $\hbar\omega = 5$ eV.

The numerical integration of (12) with the initial condition

$$\Psi(x, R, t=0) = \Phi_1^{(0)}(R) \varphi_1(x, R)$$

was carried out in a 100 Å region in the electronic coordinates and in a 10 Å region in the nuclear coordinates with a total number of nodes equal to 675×210 . The spatial grid was chosen to be nonuniform in order to enhance the accuracy of the calculation in the region of localization of the wave functions of the states belonging to the discrete spectrum. An imaginary correction to the potential was introduced near the boundaries of the region in order to ensure the absorption of the wave functions and the absence of reflections from the boundary. The integration timestep was determined by the parameters of the electron potential $V_e(x, R)$ and was $\Delta t \approx 8.3 \cdot 10^{-3}$ fsec, i.e., approximately one one-hundredth of the period for $\hbar\omega = 5$ eV.

The general principles of the numerical solution of (12) are discussed in Ref. 10.

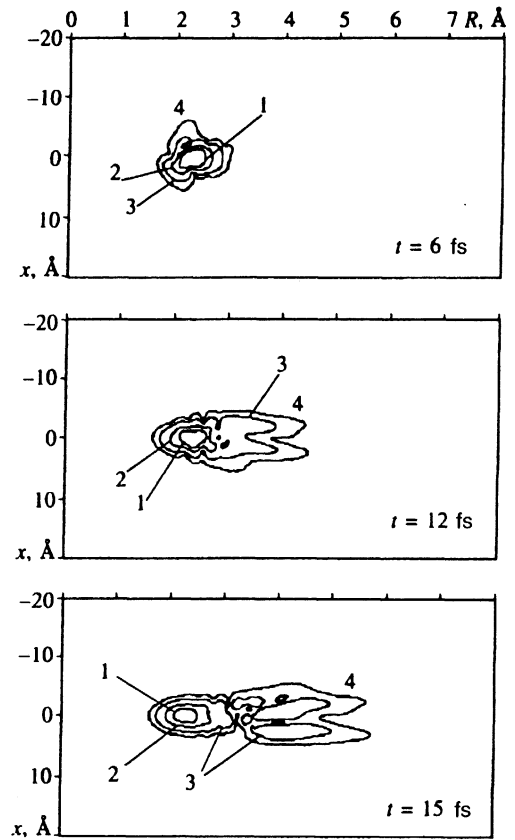


FIG. 2. Spatial distributions of the probability density $|\Psi(x, R)|^2$ for H_2^+ at different times for a laser pulse with intensity $P = 10^3$ W/cm². The curves correspond to the following values of the quantity $|\Psi(x, R)|^2$: 1) 0.3, 2) 0.1, 3) 0.01, 4) 0.001.

4. RESULTS AND DISCUSSION

The results of the numerical simulation of the dynamics of H_2^+ in a field of frequency $\hbar\omega = 5$ eV for different intensities are shown in Figs. 2 and 3. The spatial distributions of the square of the absolute value of the total wave function of the system shown in these figures correspond to the times $t = 6, 12, 15$ fs measured from the start of the laser excitation. We see that for comparatively low intensities $P \leq 10^{13}$ W/cm² (Fig. 2) mainly dissociation is observed in the system; the region of electron localization coincides with that of the nuclei during the entire time. For high intensities $P \geq 3 \cdot 10^{14}$ W/cm² (Fig. 3) ionization occurs for practically fixed nuclei and then, after the pulse has passed, motion of the nuclei due to their Coulomb repulsion is observed.

These features of the dynamics of ionization and dissociation of H_2^+ in an electromagnetic field are more obvious when the motions of the electron and the nuclei in space are considered separately. From the known wave function $\Psi(x, R, t)$ of the system the probability densities of the electronic $\rho_e(x, t)$ and nuclear $\rho_N(R, t)$ distributions can be calculated:

$$\rho_e(x, t) = \int |\Psi(x, R, t)|^2 dR, \quad (13)$$

$$\rho_N(R, t) = \int |\Psi(x, R, t)|^2 dx. \quad (14)$$

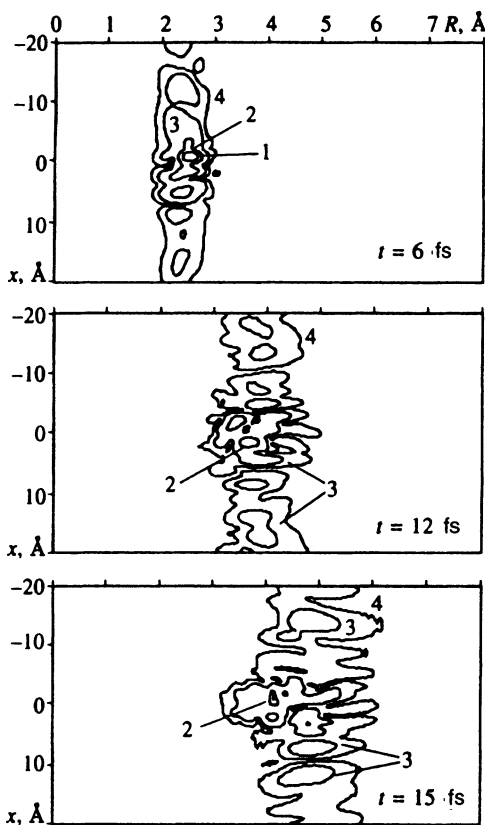


FIG. 3. The same as Fig. 2, but for the value $P=3 \cdot 10^{14}$ W/cm².

For $P=10^{13}$ W/cm² the electronic density distribution depends only weakly on time, while a wave packet moving away from the origin is formed on the distribution $\rho_N(R,t)$ after the end of the laser disturbance. A similar picture is observed in $\rho_N(R,t)$ for $P=10^{14}$ W/cm² (Fig. 4); however, in this case the probability of occupying the electron continuum is not small. With further increase in the intensity, the electron passes into the continuum at the leading edge of the laser pulse and hence the further evolution of the system is controlled mainly by Coulomb repulsive forces.

To interpret the results of the integration of (12), the exact wave function of the system was expanded in a series

of wave functions of the stationary states obtained in the adiabatic approximation

$$\Psi(x,R,t) = \sum_{k,v} C_{kv}(t) \Phi_k^{(v)}(R) \varphi_k(x,R) \exp\left(-\frac{i}{\hbar} E_{kv} t\right), \quad (15)$$

where E_{kv} is the energy of the v th stationary state belonging to the k th electronic term and C_{kv} is the amplitude of the probability of occupying the corresponding state as a function of time. The sum over k and v in (15) is to be understood as a summation over the states of the discrete spectrum and an integration over the states of the continuous spectrum.

As noted above, in the absence of the external electromagnetic field the quantities C_{kv} are practically independent of the time because of the high accuracy of the adiabatic approximation.

The probabilities of occupying the different states of the discrete spectrum were determined from the formula

$$W_{kv} = |C_{kv}(t)|^2 = \left| \int \Psi^*(x,R,t) \Phi_k^{(v)}(R) \times \varphi_k(x,R) dx dR \right|^2. \quad (16)$$

In our calculations the number of electronic states was limited to 20 and the number of vibrational states taken into account for the four lowest terms corresponding to an even electronic wave function was also equal to 20. Nonzero probabilities are obtained only for transitions between vibrational states inside the lowest electronic term. For all higher terms the probabilities of occupying the states of the discrete spectrum are negligibly small, since the corresponding Frank-Condon factors are small.

The probability of observing the molecule in a given electronic state can be written in the form

$$W_k = \sum_v |C_{kv}(t)|^2 = \int |\alpha_k(R,t)|^2 dR, \quad (17)$$

where

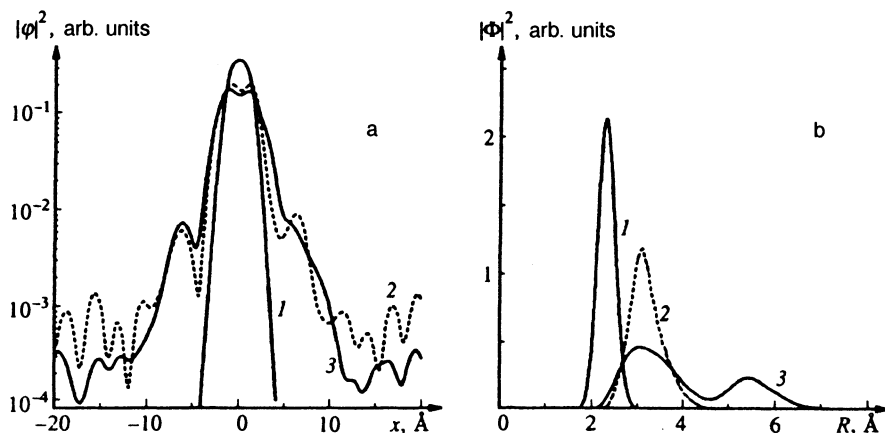


FIG. 4. Spatial distribution of the electronic (a) and nuclear (b) probability densities for H_2^+ at different times: 1) 0, 2) 6, 3) 12 fs. $P=10^{14}$ W/cm².

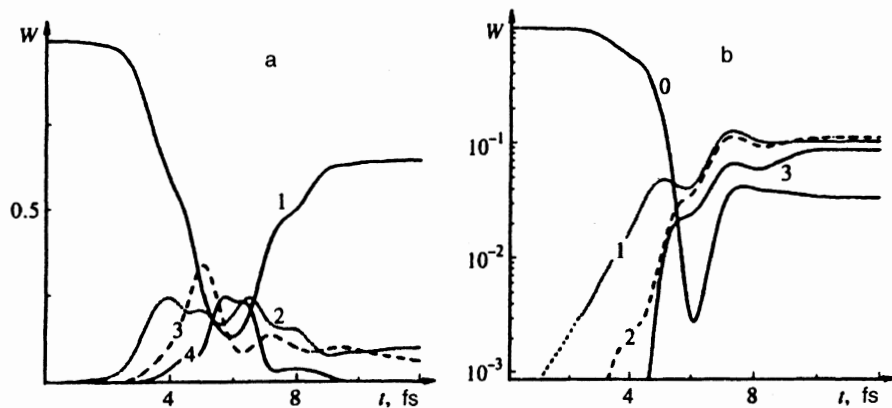


FIG. 5. Dynamics of the population densities of the four lowest electronic states (a) and the four lowest vibrational states belonging to the ground electronic term (b). The number of the curve corresponds to the number of the state.

$$\alpha_k(R, t) = \int \Psi^*(x, R, t) \varphi_k(x, R) dx.$$

As an example, the time dependence of the probabilities W_k , $k=1, \dots, 4$ and W_{1v} , $v=0, \dots, 3$, averaged over the high-frequency oscillations, is shown in Fig. 5 for $P=10^{14}$ W/cm² and $\hbar\omega=5$ eV. The curves show the typical increase in the probability of observing the molecule in the ground electronic state after the passing of the pulse. (Fig. 5a), the occupation of the excited states of the ground electronic term (Fig. 5b), and the formation of an oscillating wave packet on the nuclear wave function. In homonuclear molecules such as H₂⁺ direct transitions between vibrational levels belonging to the same electronic term are forbidden in the dipole approximation. Therefore the only possible channel for the occupation of the excited vibrational states of the ground electronic term is multiphoton transitions through intermediate states belonging to the higher electronic terms of the molecule. This nature of the process is illustrated by Fig. 5.

The distribution of probabilities of observing the molecule in different vibrationally excited states at the end of the laser disturbance is shown in Fig. 6 for different intensities. We see that as the intensity increases the relative probability of observing the molecule in states with large values of the vibrational quantum number becomes larger.

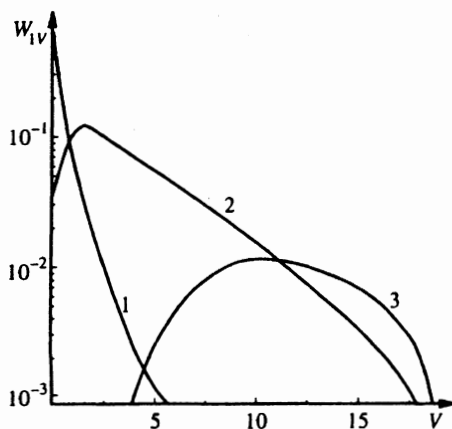


FIG. 6. Distribution of the population densities of the vibrational states of the ground term at the end of the laser pulse for different intensities: 1) 10^{13} , 2) 10^{14} , 3) $3 \cdot 10^{14}$ W/cm².

The probabilities of photoionization W_I and photodissociation W_D of the molecule as functions of time can be calculated using (16) and (17):

$$W_I(t) = 1 - \sum_k W_k, \tag{18}$$

$$W_D(t) = \sum_k W_k - \sum_v |C_{1v}|^2.$$

Unlike (15) and (17), here the summation goes only over the states of the discrete spectrum.

The quantity

$$W_F = W_I + W_D$$

is the total probability of photodisintegration of the molecule.

The intensity dependence of the probability $|C_{10}|^2$ of observing the molecule in the ground state and also the excitation probability $\sum_v |C_{1v}|^2 - |C_{10}|^2$, the ionization probability W_I , and the photodisintegration probability W_F are shown in Fig. 7. The nonmonotonic dependence of the excitation probability on the intensity is easily explained. In weak fields the molecule tends to remain in the ground state, while in strong fields the excitation probability is also small, since the prob-

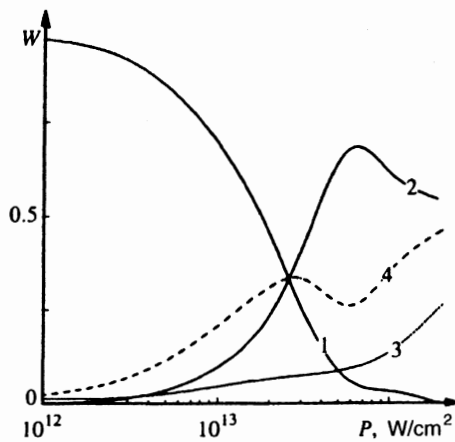


FIG. 7. Intensity dependence of the probability of observing H₂⁺ at the end of the laser pulse in the ground (1) and the excited (2) states, and also the probabilities of ionization (3) and photodisintegration (4).

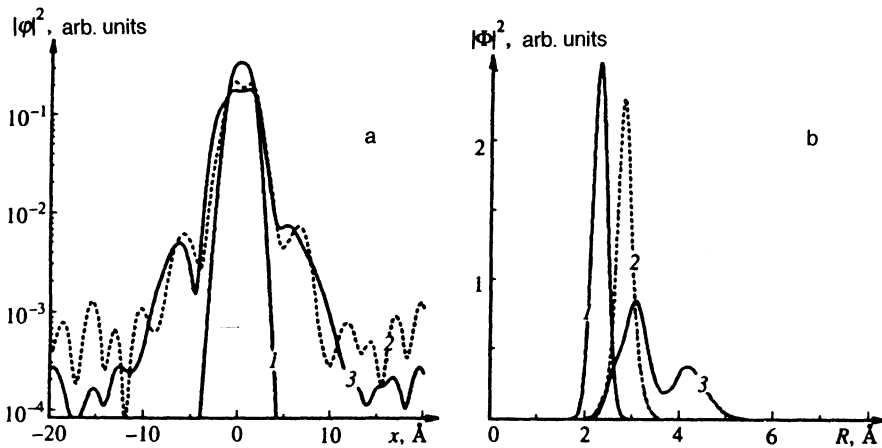


FIG. 8. The same as Fig. 4, but for the deuterium molecular ion.

abilities of photoionization and photodissociation increase rapidly. More surprising is the nonmonotonic nature of the dependence $W_F(P)$, since it implies that the molecule becomes more stable against photodisintegration at high intensities (3–6) 10^{13} W/cm². It is evident from Fig. 7 that this increase in stability is associated with a decrease in the probability of photodissociation at these intensities, whereas the quantity W_I monotonically increases.

This phenomenon can be interpreted as the stabilization of H_2^+ in a strong light field against dissociation. But an increase in the stability of molecular systems in strong light fields (i.e., against ionization of the molecule) is not expected for Kramers–Henneberger processes. It is known that stabilization in this case is associated with large-amplitude oscillations of the electronic wave packet in a superatomic field, such that the electron is practically free and the wave function is not seated on the atomic core. Therefore after the end of the laser disturbance the electron will be in a bound state with high probability.^{16,17} When a freely oscillating electron wave packet forms in a molecular system, the force of Coulomb repulsion between the nuclei dominates, which causes them to separate and prevents the electron from going back to the initial bound state.

Finally, we compare the processes of ionization and dissociation of molecular ions with different isotopic compositions, in particular, H_2^+ and D_2^+ . The D_2^+ molecule has twice the mass of H_2^+ , and therefore the frequency of the nuclear oscillations is lower and the functions $\Phi_k^{(v)}(R)$ are more localized in space. The spatial distributions of the electron and nuclear probability densities $\rho_e(x)$ and $\rho_N(R)$ are shown for the D_2^+ molecule in Fig. 8 at different times for $P=10^{14}$ W/cm². Comparison of Fig. 8 with Fig. 4 for H_2^+ shows that the only difference between the two cases is the slower separation of the deuterium nuclei from one another during and after the laser disturbance. Because the electronic densities $\rho_e(x)$ shown in Figs. 4 and 8 are practically the same, the dynamics of the ionization process can be considered assuming fixed nuclei, i.e., assuming that the process can be broken into two steps. In the first step (during the laser pulse) ionization and excitation of the different electronic states of the molecule occur, while in the second step (after the laser pulse passes) there is motion of the nuclei, which depends on

the form of the different electronic terms. In the limit of an ultrashort laser pulse the motion of the nuclei can be neglected, and one can treat the ionization and dissociation of the system assuming frozen nuclei (during the passage of the laser pulse). In this case the total wave function is represented as

$$\Psi(x, R, t) = \phi(x, R, t)\Phi(R), \quad (19)$$

where $\phi(x, R, t)$ is the electron wave function and $\Phi(R)$ is the nuclear wave function characterizing the position of the nuclei up to the start of the laser disturbance and remaining constant during the passage of the pulse. The electronic wave function satisfies the equation

$$i\hbar \frac{\partial \phi(x, R, t)}{\partial t} = \left(-\frac{\hbar^2}{2m} \frac{\partial^2}{\partial x^2} + V_e(x, R) - e x \varepsilon(t) \cos \omega t \right) \phi(x, R, t), \quad (20)$$

where $V_e(x, R)$ is given by (3).

At the end of the laser disturbance the system may be found with a certain probabilities in different electronic states of the discrete spectrum or the continuum, and these probabilities are determined by the appropriate coefficients in the expansion of the exact solution $\phi(x, R, t)$ in a series of eigenfunctions of the electron Hamiltonian:

$$\phi(x, R, t) = \sum_k C_k(R, t) \varphi_k(x, R) \exp\left(-\frac{i}{\hbar} E_k t\right). \quad (21)$$

The sum over k is to be understood as a summation over the states of the discrete spectrum and an integration over the states of the continuum.

The time dependence of the expansion coefficient $C_k(R, t)$ of (21) is obtained using the numerical solution of (20):

$$C_k(R, t) = \int \varphi^*(x, R, t) \varphi_k(x, R) dx. \quad (22)$$

Using the population densities of the different states, the ionization probability as a function of time for fixed R can be calculated from

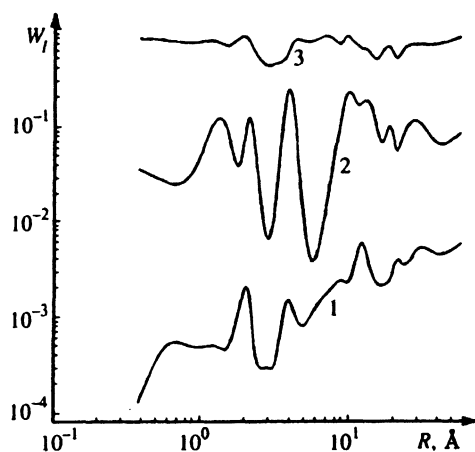


FIG. 9. Probability of ionization as a function of the inter-nuclear distance for $\hbar\omega=5$ eV and different intensities: 1) 10^{13} , 2) 10^{14} , 3) 10^{15} W/cm².

$$W_I(R,t) = 1 - \sum_{k=1} |C_k(R,t)|^2. \quad (23)$$

From the calculations we conclude that the probability of ionization depends sharply on the internuclear distance. This may be because ionization by a photon with $\hbar\omega=5$ eV occurs from several of the higher states, which are occupied to different degrees depending on R .

Figure 9 shows the dependence of the ionization probability on R for three different values of the laser intensity. The observed nonmonotonic dependence $W_I(R)$ may also be due to interference between the electronic wave packets formed in the process of ionization of the electron from the double-well potential. A similar nonmonotonic dependence of the probability of ionization on the internuclear distance was obtained in Ref. 18

Therefore the calculations performed in the approximation of frozen nuclei illustrate qualitatively the interesting features of the behavior of the system. The reliability of the results obtained in this approximation are demonstrated by the two-particle calculations.

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