

Nuclear correlation in ionization and harmonic generation of H_2^+ in short intense laser pulses

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The dynamics of H_2^+ in short intense laser pulses with wavelengths of 532 nm and 800 nm is considered in two approaches, including and not including the motional degree of freedom of the two nuclei. In the situation with moving nuclei, referred to as the non-Born-Oppenheimer approach, the ionization is substantially increased with respect to the case with no motional degree of freedom of the nuclei, and consequently the nuclear separation also increases with rising laser intensity. This separation is accompanied by complex relative accelerations of the nuclei due to a strongly varying mutual shielding via the laser-driven electron. In the harmonic spectrum the irregularity of the electronic dynamics induced by the nuclear motion leads to a broadening of the harmonic peaks. In particular, the correlated nuclear motion is shown to lead to an earlier onset of the harmonic cutoff than for fixed nuclei, but also to a wider cutoff regime with significantly higher harmonics.

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I. INTRODUCTION

While the laser-atom interaction with one single active electron is now understood in quite some detail [1,2], increased interest has recently been devoted toward more complex systems like multielectron atoms [3] and molecules [4–13] in intense laser fields. Since the nuclei and electrons in the molecular system respond to the laser field on different time scales, the research into molecules in an intense laser pulse has mostly been limited to rather simple molecular systems, such as H_2^+ and H_2 [4–6]. However, it is also precisely these two time scales of the response of the molecular system to the laser field that exhibit abundant dynamical behavior, such as above-threshold dissociation [8], dissociation through bond softening [9], Coulomb explosion [10], high-order harmonic generation [11,12], two-color photodissociation [13], and charge-resonance-enhanced ionization [14].

The exact simulation of the dynamical behavior of molecules in intense laser fields via the time-dependent Schrödinger equation (TDSE) is rather demanding even with up-to-date computing facilities. Therefore many theoretical researchers have adopted the Born-Oppenheimer (BO) approximation to separate the electronic and nuclear degrees of freedom in describing the dynamical processes of a molecule in intense laser pulses. In the BO approach the electrons respond essentially instantaneously to the external field on the time scale of both the nuclear motion and the optical period of the laser field. However, when the laser pulse becomes short and intense, the BO approximation may become invalid. In high-intensity laser fields the potential structure of the molecular system is greatly modified by the applied laser field and substantially different from the situation when the BO approximation is applicable. Thus in such cases there may be a mixing of the nuclear and electron dynamics in molecules, imposing a simultaneous description of both dynamics [4,11,12].

In this article we consider this regime of mutual interplay

of the electron and nuclear dynamics of H_2^+ in short intense laser pulses. Particular emphasis is placed on the tunneling dynamics, with aspects investigated such as ionization, dissociation, and high-harmonic generation. We show that the broadening of the return times in the recollisions with the moving ions leads to an earlier onset of the cutoff regime as compared to the BO approach, but to a much wider regime with substantially higher harmonics. This is investigated in detail as a function of the applied laser frequency and intensity. This broadening also reduces the periodicity of the recollisions and thus increases the broadening of the spectral peaks.

The paper is organized as follows. We begin by discussing in the first section the collinear model of H_2^+ in intense laser fields. Then we introduce the two approaches to be compared, including and not including the motion of the nuclei. For both approaches we introduce the Hamiltonians governing the dynamics of H_2^+ to be studied for various intensities and wavelengths of short laser pulses. Through numerical solution of the TDSE, we investigate the time evolutions of the probabilities of ionization and dissociation, the spectra of high-order harmonic generation, and the nuclear accelerations, as well as the time-dependent average nuclear separation. Finally, in the main section we present and discuss the results from the two approaches before concluding.

II. COLLINEAR MODEL OF H_2^+

In intense linearly polarized laser fields, the major part of the laser energy is transferred into the electron motion along the polarization direction of the laser field. Thus most information about the dynamical behavior of the electron in the laser field is contained in the electron motion along the polarization direction of the laser field. In addition, the molecular axis rapidly aligns along the polarization direction of the laser field. Thus we here restrict ourselves to the one-dimensional collinear model of H_2^+ [15],

which was established on the basis of the two following assumptions: (a) The axis of the molecule in the intense laser field is aligned along the polarization direction of the laser field; (b) the motion of the electron in the molecule may be restricted to one dimension along the direction of polarization of the laser field. These two assumptions have been experimentally and theoretically confirmed [16]. The collinear model allows us to reduce the spatial variables of H_2^+ down to two, the distance from the electron to the center of mass of two nuclei, z , and the distance between the nuclei, r . A further significant advantage of this collinear model is that the wave function of H_2^+ can be easily obtained by exactly solving the TDSE numerically. In addition, the evolving wave packet of H_2^+ can be displayed within a three-dimensional (3D) plot, which is very helpful in studying the competition between ionization and dissociation in the molecule and in understanding the dynamical process of Coulomb explosions. We note, however, that Chelkowski *et al.* [5] achieved a solution of the corresponding 3D model for the first time in 1996, in particular to show the existence of charge-resonance-enhanced ionization.

In two-dimensional space, the field-free Hamiltonian of the collinear H_2^+ molecule has the following simple form (in atomic units):

$$H_0(z, r) = -\frac{1}{2} \frac{\partial^2}{\partial z^2} - \frac{1}{2\mu} \frac{\partial^2}{\partial r^2} + V(z, r), \quad (1)$$

where

$$V(z, r) = \frac{1}{\sqrt{r^2 + q_n}} - \frac{1}{\sqrt{(z - 0.5r)^2 + q_e}} - \frac{1}{\sqrt{(z + 0.5r)^2 + q_e}} \quad (2)$$

is the Coulomb potential of 1D H_2^+ . μ is the reduced mass of the two nuclei and we neglected terms weighted by the small ratio of the electron mass to that of the nuclei. q_e and q_n are the softening parameters that are used to eliminate the singularity of the one-dimensional Coulomb potential. This method of eliminating the singularity of the Coulomb potential has been frequently used in numerical calculations. In the dipole approximation [17], the coupling between the molecule and the laser field is written as

$$H_I(z, t) = zE(t). \quad (3)$$

The time-dependent Schrödinger equation has the following form:

$$i \frac{\partial \Psi(z, r, t)}{\partial t} = [H_0(z, r) + H_I(z, t)] \Psi(z, r, t). \quad (4)$$

Equation (4) can be numerically solved with the second-order split-operator method [18]. The formal expression for the wave function at the time $t + \delta t$ is

$$\Psi(z, r, t + \delta t) = e^{-i[H_0(z, r) + H_I(z, t)] \delta t} \Psi(z, r, t), \quad (5)$$

which can be approximately expressed as

$$\begin{aligned} & \Psi(z, r, t + \delta t) \\ &= \exp \left[i \frac{\delta t}{2} \left(\frac{1}{2} \frac{\partial^2}{\partial z^2} + \frac{1}{2\mu} \frac{\partial^2}{\partial r^2} \right) \right] \\ & \quad \times \exp \left[-i \frac{\delta t}{2} V(z, r) \right] \exp[-iH_I(z, t) \delta t] \\ & \quad \times \exp \left[-i \frac{\delta t}{2} V(z, r) \right] \exp \left[i \frac{\delta t}{2} \left(\frac{1}{2} \frac{\partial^2}{\partial z^2} + \frac{1}{2\mu} \frac{\partial^2}{\partial r^2} \right) \right] \\ & \quad \times \Psi(z, r, t) + O(\delta t^3). \end{aligned} \quad (6)$$

The wave function at any moment in time t , $\Psi(z, r, t)$, can be numerically solved by repeatedly applying the unitary operator in the right hand side of Eq. (6) on the initial wave function $\Psi_0(z, r) = \Psi(z, r, t=0)$. In order to evaluate the initial state, which is usually chosen to be the ground state of the molecular system, in the non-BO approach, we first evaluate the bound states $E_n < 0$ ($n=0, 1, 2, 3, \dots$) by using the autocorrelation function of any trial function in the field-free case, and then calculate the corresponding eigen wave functions, $\Phi_n(z, r)$ ($n=0, 1, 2, 3, \dots$), using the following formula:

$$\Phi_n(z, r) \sim \frac{1}{T_{tot}} \int_0^{T_{tot}} e^{iE_n t} W(t) \Psi(z, r, t) dt, \quad (7)$$

where $\Psi(z, r, t)$ is the wave function at time t that evolves in the field-free case from a trial wave function at $t=0$. T_{tot} is the total free-field evolution time, and $W(t)$ is the Hanning window function given by

$$W(t) = 1 - \cos \left(\frac{2\pi t}{T_{tot}} \right) \quad (0 \leq t \leq T_{tot}). \quad (8)$$

In order to avoid any reflections of the wave function from the boundaries of the calculation area, we also use an absorbing mask function to absorb the reflecting parts of the wave function after evolving for a time step δt [19]. However, since the two nuclei of H_2^+ cannot coincide, the wave function automatically tends to zero at the $r=0$ boundary. Thus no absorption is introduced at the $r=0$ boundary in the case of moving nuclei.

A. Case of moving nuclei

In the situation of moving nuclei (non-Born-Oppenheimer approximation) the wave function is a two-variable function, i.e., $\Psi(z, r, t)$ depends simultaneously on the electronic coordinates (z) and the nuclear coordinates (r), which have the same footing in the wave function. So the wave function $\Psi(z, r, t)$ includes dynamical information about the electrons and the nuclei. We discuss them separately. In all of our calculations, we choose the softening parameters to be $q_e = 1.0$ and $q_n = 0.03$. This choice renders the energies of the ground states equal to -0.778 a.u.

1. Dynamics of the electrons

When considering the motion of the electrons and nuclei, we define the total calculation region S , which is two dimensional and spanned by z with $|z| \leq z_{\max} = (n_z/2)dz$ and r with $0 \leq r \leq r_{\max} = n_r dr$. Here n_z (n_r) and dz (dr) are the number of grid points and the spatial step sizes along the z (r) axes. In this two-dimensional area we define the ionization region S_i as

$$S_i = \{z | \text{Abs}(z) > z_i + 0.5r\}, \quad (9)$$

where $z_i = 32.0$ a.u. and $\text{Abs}(z)$ designates the absolute value of z . The time-dependent probabilities of ionization can be evaluated with the following formula:

$$P_i(t) = \iint_{S_i} |\Psi(z, r, t)|^2 dz dr. \quad (10)$$

The expectation value of the dipole acceleration of H_2^+ is defined as

$$\langle a(t) \rangle = - \iint_S \Psi^*(z, r, t) \frac{\partial V(z, r)}{\partial z} \Psi(z, r, t) dz dr. \quad (11)$$

According to Eq. (11) we evaluate the spectrum of high-order harmonics, $A(\omega)$, by taking the modulus squared of the Fourier transform of the dipole acceleration:

$$A(\omega) = \left| \int_0^{T_d} \langle a(t) \rangle e^{-i\omega t} dt \right|^2, \quad (12)$$

where T_d is the full duration of the laser pulse.

2. Dynamics of the nuclei

Concerning the relative dynamics of the nuclei, using the wave function $\Psi(z, r, t)$, we are able to obtain the expectation value for the nuclear separation at any time,

$$R(t) = \iint_S r |\Psi(z, r, t)|^2 dz dr \bigg/ \iint_S |\Psi(z, r, t)|^2 dz dr, \quad (13)$$

and to calculate the time-dependent dissociation probability

$$P_d(t) = \iint_{S_d} |\Psi(z, r, t)|^2 dz dr \quad (14)$$

by integrating the probability density $|\Psi(z, r, t)|^2$ in the dissociation region S_d , which is defined as

$$S_d = \{r | r_d \leq r < r_{\max}\}, \quad (15)$$

where $r_d = 9.5$ a.u. In addition, we evaluate a time-dependent relative acceleration of nuclei from the expectation value of the nuclear distance:

$$a_n(t) = \ddot{R}(t) = \frac{d^2 R(t)}{dt^2}. \quad (16)$$

Using this, we may obtain the spectrum with respect to $a_n(t)$:

$$S(\omega) = \left| \int_0^{T_d} \ddot{R}(t) e^{-i\omega t} dt \right|^2. \quad (17)$$

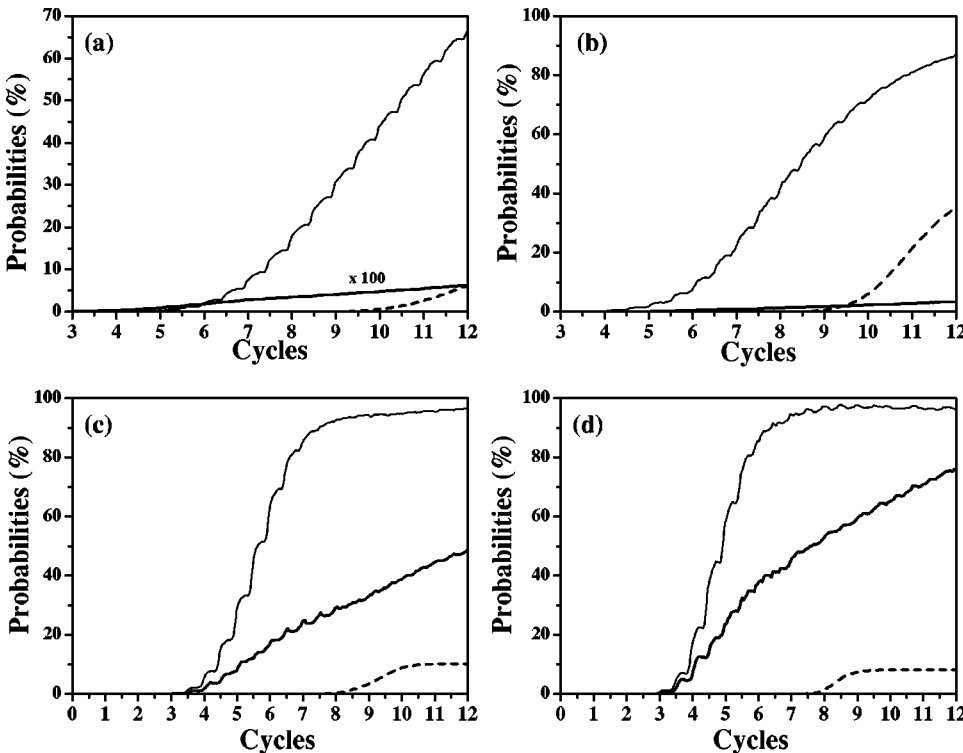


FIG. 1. The ionization probabilities of H_2^+ in fields of laser pulses with a wavelength of 532 nm and intensities of (a) 2.5×10^{14} W/cm², (b) 4.0×10^{14} W/cm², (c) 7.5×10^{14} W/cm², and (d) 1.0×10^{15} W/cm² for two cases: fixed (thick line) and moving (thin line) nuclei. The dissociation probabilities in the case of moving nuclei are shown with a dashed line. The initial state is assumed to be the ground state in all cases (1 cycle corresponds to 73.36 a.u. in time).

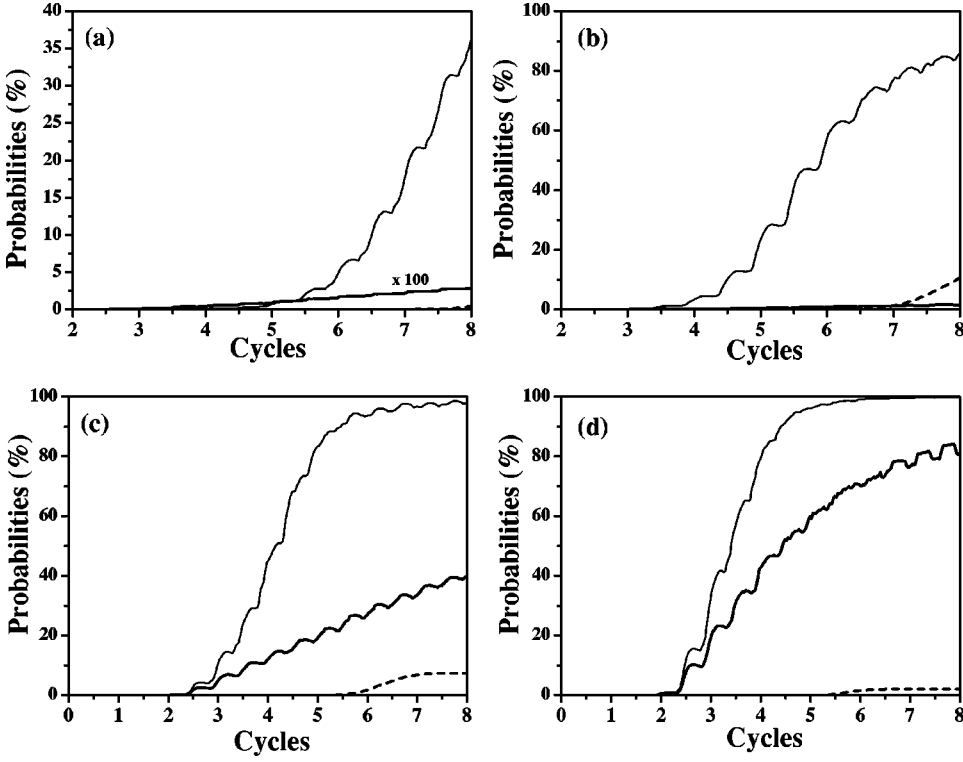


FIG. 2. Same as in Fig. 1 for a wavelength of 800 nm (1 cycle corresponds to 110.31 a.u. in time).

We note that this spectrum may be seen only as a frequency analysis of the relative motion of the nuclei but not as the isolated radiation spectrum of the nuclei. For this we would require the small center of mass acceleration of the ionic system via the electron, which is neglected in our treatment; see also [20].

B. Case of fixed nuclei

When the motional degree of freedom of the nuclei is not included, the calculation region reduces to a one-dimensional space. In order to compare the results here with those in the case of moving nuclei, we set the nuclear separation to be equal to the average distance between the nuclei in the ground state $|\Phi_0\rangle$ in the case of moving nuclei

$$r_g = \frac{\int \int_S r |\Phi_0(z, r)|^2 dz dr}{\int \int_S |\Phi_0(z, r)|^2 dz dr} = 2.63 \text{ a.u.}, \quad (18)$$

and calculate in this way the energy of the corresponding ground state to be -0.781 a.u. The methods for calculating the time-dependent probabilities and the spectrum of high-order harmonic generation are similar to those in the case of moving nuclei, except for the dissociation probability and the direct radiation from the nuclei, which obviously do not exist here.

III. RESULTS AND DISCUSSION

Throughout the whole discussion we use the same laser pulse shape identified by an electric field of the form

$$E(t) = E_0 f(t) \sin(\omega t), \quad (19)$$

where

$$f(t) = \begin{cases} \sin^2\left(\frac{\pi t}{6T_d}\right) & \text{when } 0 \leq t < T_d/3, \\ 1 & \text{when } T_d/3 \leq t < T_d. \end{cases} \quad (20)$$

Here $T_d = 880$ a.u. = 21.2 fs is the full duration of the laser field. Regarding other fixed parameters, we have employed $dz = 0.2$, $n_z = 1024$, $dr = 0.04$, $n_r = 512$, and $\delta t = 0.053$.

We calculate the time-dependent ionization and dissociation probabilities of H_2^+ interacting with ultrashort laser pulses with different intensities in two approaches and show the calculated results in Fig. 1 and Fig. 2 for 532 nm and 800

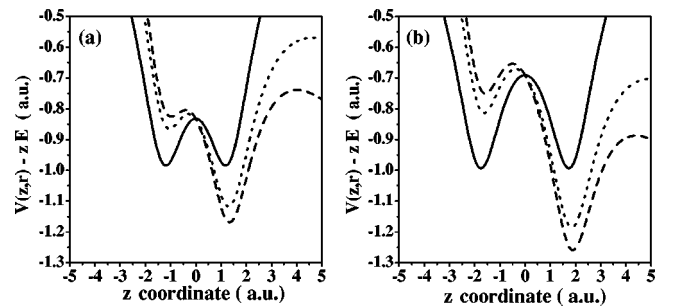


FIG. 3. The variation of the total potential $V(z, r) - zE$ as a function of the spatial coordinate z . The distance between the nuclei is assumed to be $r = 2.63$ a.u. in (a) and $r = 3.63$ a.u. in (b). The corresponding intensities of the laser pulse are $I = 0$ (solid line), $I = 4.0 \times 10^{14}$ W/cm² (dotted line), and $I = 7.5 \times 10^{14}$ W/cm² (dashed line).

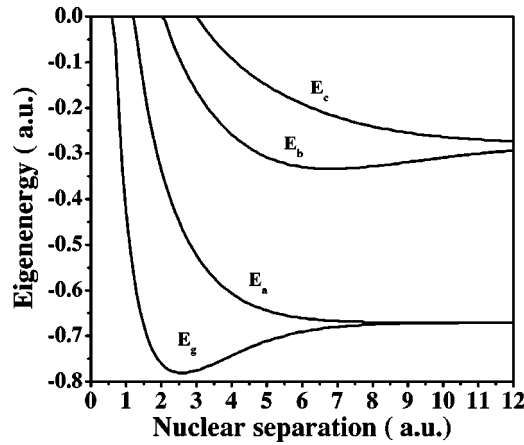


FIG. 4. The variation of the eigenlevels of the first four eigenstates with changing nuclear distance. E_g , E_a , E_b , and E_c stand for the potential curves of the ground and the first three excited states, respectively. The softening parameters are chosen to be $q_e = 1.0$ and $q_n = 0.03$.

nm pulses, respectively. Since the ionization in the case of fixed nuclei is almost vanishing (its maximum is smaller than 10^{-3}) at the intensity of 2.5×10^{14} W/cm², the fixed-nucleus ionization probabilities [thick lines shown in Fig. 1(a) and Fig. 2(a)] are enhanced by a factor of 100. We find from Fig. 1 and Fig. 2 that the onset of dissociation occurs about 6.5–7.0 fs later than that of ionization. The reason is that the nuclei are much heavier than the electrons and thus respond to the external field very slowly. Since the 800 nm laser pulse involves a greater ponderomotive energy than the 532 nm pulse and thus leads to a more rapid ionization of the electron in H_2^+ , most parts of the wave function flow out in the *ionization* direction ($|z| \rightarrow \infty$) but not in the *dissociation* direction ($r \rightarrow \infty$). Thus for our molecular system, the ionization process is dominant when an intense ultrashort laser pulse is used, especially for the case with a longer wavelength.

Comparing the time-dependent ionization probabilities calculated in the two approaches, we also find from comparing the pulses with the same intensity that the ionization probability is significantly smaller in the case of fixed nuclei. This phenomenon can be understood by considering the sys-

tem potential as formed by the combination of the Coulomb potential and the laser field. The potential well confines the electron and may release it if it is too shallow. When the nuclear separation is small, the system potential well is very deep and possesses quite high outer barriers, which can confine the electron tightly. However, when the nuclear separation increases not only do the outer barriers of the molecular potential well rapidly decrease but also the inside barrier of the well increases, as shown in Fig. 3. As a consequence the potential well becomes shallower with increasing nuclear separation and the ionization rate increases, as is properly accounted for only by the theory with moving nuclei. In addition we note from Fig. 4 that the eigenenergy of the ground state rises with increasing internuclear distance when $r > r_g$. This indicates also that H_2^+ is ionized more strongly due to the increase of the internuclear distance.

In Fig. 5(a) and Fig. 5(b), we show, respectively, the average nuclear separation as a function of time for the 532 nm and 800 nm pulses with different peak intensities. It is easily noted and not surprising that the more intense the pulse intensity, the faster the average relative nuclear velocity will be (i.e., the slopes of the curves in Fig. 5 are greater). The nuclear separation gradually increases with time before ionization occurs. Once the electron is ionized, due to the strong repulsion between the two nuclei, the nuclear separation rapidly increases with time, i.e., a Coulomb explosion takes place. Since a high-intensity laser pulse ionizes the electron earlier than a low-intensity pulse, the two nuclei begin to separate at smaller nuclear separations when high-intensity pulses are used. We note that the results obtained in this figure complement the results for a smaller laser intensity obtained via 3D calculations in Fig. 7 in [5].

In Figs. 6–9, we present the spectra of high-order harmonic generation of H_2^+ interacting with 532 nm and 800 nm laser pulses with different intensities in both moving and fixed nuclei approaches. The spectra calculated in both approaches have a general form similar to those produced by atoms in an intense laser field. Thus they have a rapid decrease in intensity for the first few harmonics, a plateau with harmonics at almost constant intensity, and a cutoff where the harmonics quickly decrease [21]. The more intense the laser pulse, the longer the plateau and consequently the higher the corresponding cutoff frequency. The characteris-

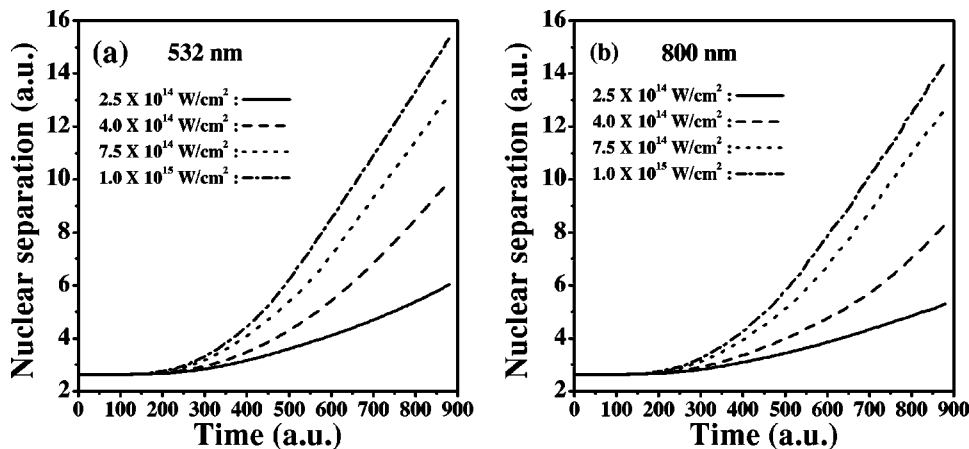


FIG. 5. The time-dependent nuclear separation of H_2^+ while interacting with an ultrashort laser pulse of shape given in Eq. (20) and various intensities as indicated in the figure. H_2^+ is initially assumed to be in the ground state.

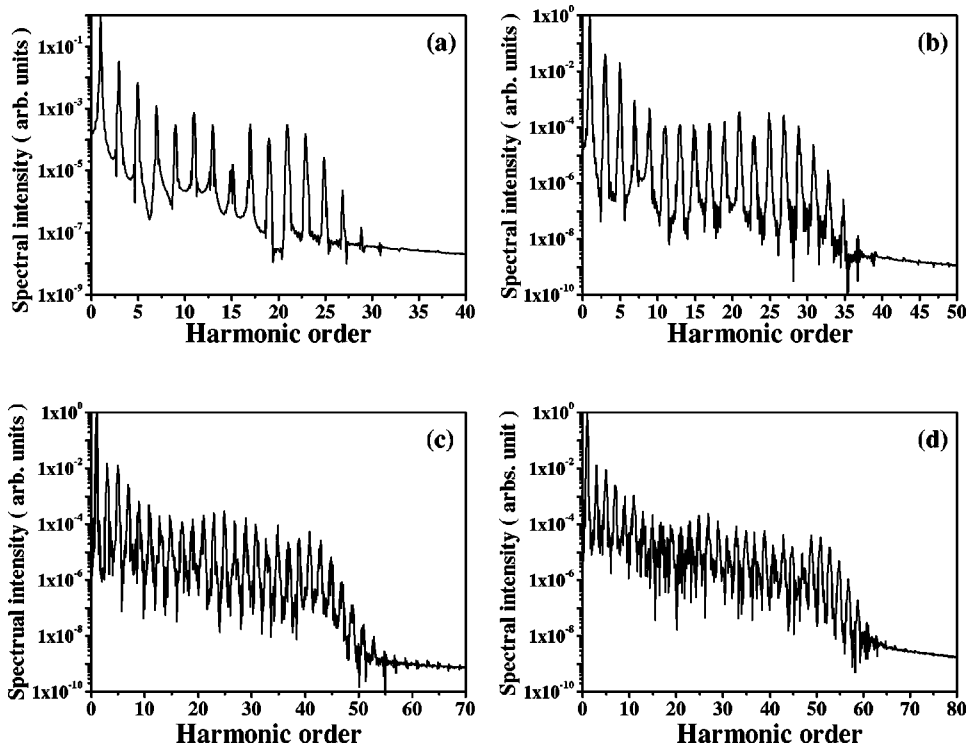


FIG. 6. The high-order harmonic spectra of H_2^+ arising from interaction with a laser pulse of shape given in Eq. (20), wavelength 532 nm, and various peak intensities: (a) 2.5×10^{14} W/cm², (b) 4.0×10^{14} W/cm², (c) 7.5×10^{14} W/cm², and (d) 1.0×10^{15} W/cm². H_2^+ is initially in the ground state. The nuclei are allowed to move.

tics are the same in the both approaches. However, the widths of the individual harmonic peaks in the spectra of H_2^+ are obviously increased with respect to atoms, especially for the case of moving nuclei. This peak-broadening effect is probably caused by both the adiabatic increase in the nuclear separation and the difference between the molecular and atomic potentials. Unlike the centric Coulomb potential of the atom, the Coulomb potential of H_2^+ is bicentric. We

find that when the nuclear separation is fixed the peak-broadening effect correspondingly decreases. A further reason is that, with varying positions of the nuclei in the recollision processes, the periodicity of the recollisions is reduced. This increases the width of the harmonics for moving nuclei.

It is easily seen from Figs. 6–9 that there are further striking differences in the envelopes of the harmonic spectra

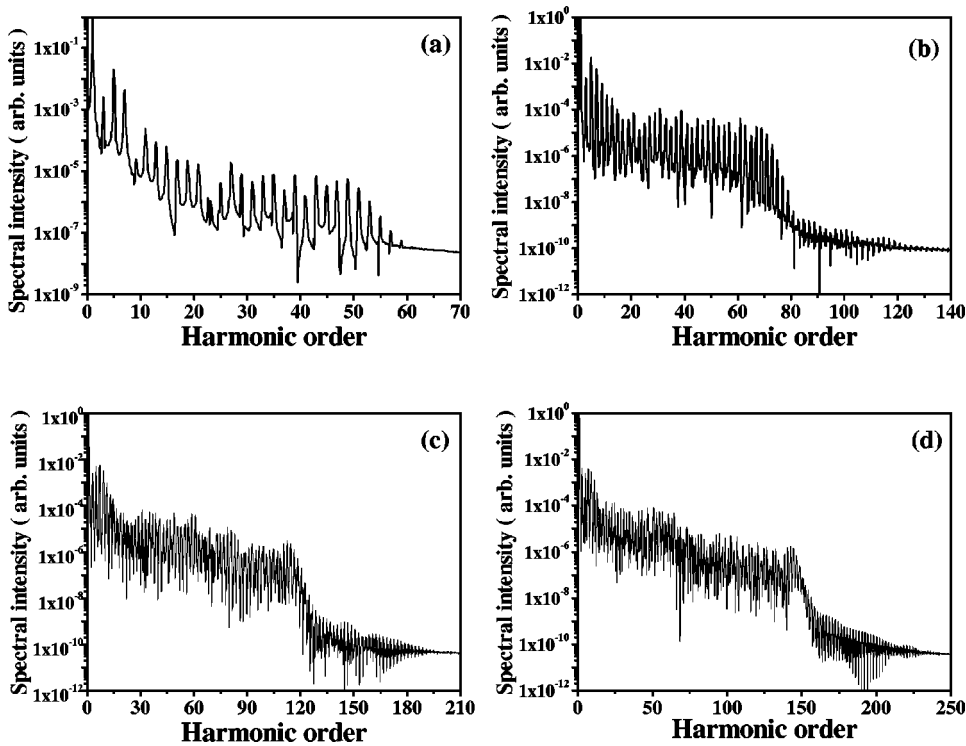


FIG. 7. Same as in Fig. 6 but for a laser light wavelength of 800 nm.

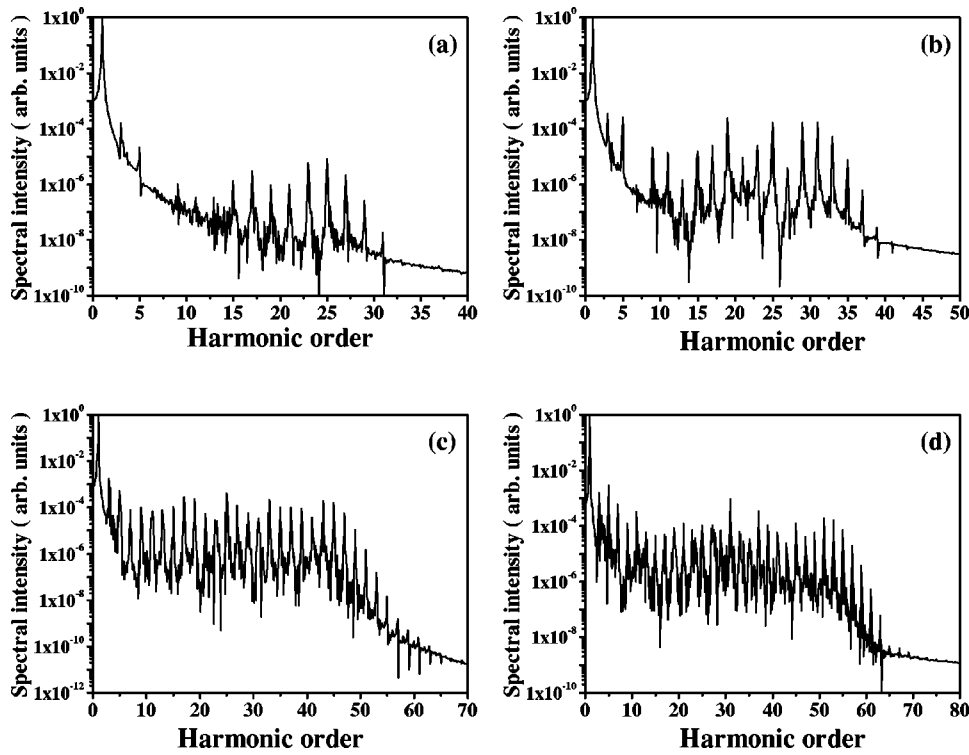


FIG. 8. The high-harmonic spectra of H_2^+ arising from interaction with a laser pulse of shape given in Eq. (20), wavelength 532 nm, and the various peak intensities: (a) 2.5×10^{14} W/cm², (b) 4.0×10^{14} W/cm², (c) 7.5×10^{14} W/cm², and (d) 1.0×10^{15} W/cm². H_2^+ is initially in the ground state. The nuclei are fixed in position [$r_g = 2.63$ as indicated in Eq. (18)].

calculated in the cases of moving and fixed nuclei. In the case of moving nuclei, the spectra have more intense low-order harmonics (compared to the situation of fixed nuclei), which rapidly decrease in intensity with increasing order (see Figs. 6 and 7). When pulses with high intensity and long wavelength are used, the harmonic spectra show cutoff regions with a long high-order tail but a less pronounced cutoff frequency [see Figs. 7(c) and 7(d)]. In the case of fixed nu-

clei, however, the low-order harmonics have no rapid decrease in intensity with increasing order, but have almost the same intensities as the high-order harmonics in the plateau [see Figs. 8(c and d) and 9(c and d)]. Some low-order harmonics even disappear for the low-intensity pulse [see Figs. 8(a and b) and 9(a and b)]. The differences in the cases of moving and fixed nuclei are greater for the 800 nm laser pulse than for the situation with 532 nm. Thus we see clearly

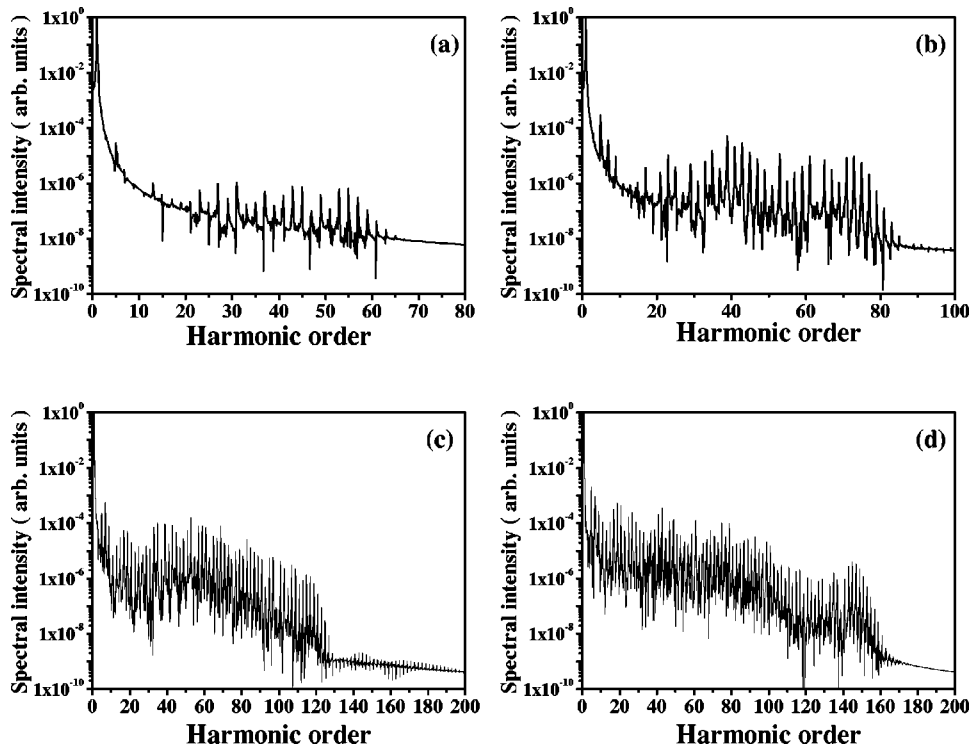


FIG. 9. Same as in Fig. 8 but for a laser light wavelength of 800 nm.

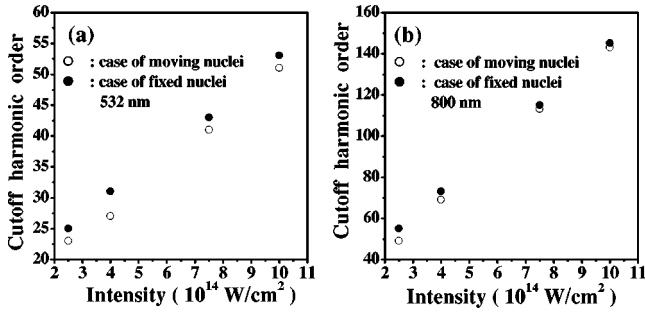


FIG. 10. The cutoff harmonic frequencies as a function of the maximal driving laser intensity. (a) and (b) relate to the situations with laser wavelengths of 532 nm and 800 nm, respectively.

that the adiabatic nuclear motion of H_2^+ significantly affects the structure of the harmonic spectrum in the interaction of (especially) the long wavelength laser pulse with a high intensity ($\sim 10^{15} \text{ W/cm}^2$) and a duration of about 20 fs. We add that the results obtained in Figs. 6–9 complement the results obtained via 3D calculations in Fig. 10 in [5]. In that paper no appreciable signature of the motion of the nuclei was noticeable in the harmonic spectra for smaller laser intensities than in our calculations. This can be understood because the motion of the nuclei is enhanced with increasing laser intensity. In addition, we find a somewhat smaller distance between the nuclei in the case of fixed nuclei [see Eq. (18)].

Using Figs. 6–9 we are now in a position to compare the cutoff frequencies ω_{max}^f and ω_{max}^m arising from the calculations with fixed and moving ions, respectively. This has been plotted in Fig. 10 where we see clearly that the onset and the maximum of the cutoff regime are at lower frequencies, but

from Figs. 6–9 that substantially higher harmonics are possible, with moving nuclei. From the data in Fig. 10 we can formulate the cutoff rule in the usual form as a function of the ionization energy plus a multiple of the ponderomotive energy $U_p = eE^2/4m\omega^2$. We obtain in atomic units

$$\begin{aligned}\omega_{\text{max}}^m &= 0.94 + 3.53U_p, \\ \omega_{\text{max}}^f &= 1.33 + 3.30U_p\end{aligned}\quad (21)$$

for a 532 nm pulse and

$$\begin{aligned}\omega_{\text{max}}^m &= 1.06 + 3.27U_p, \\ \omega_{\text{max}}^f &= 1.42 + 3.11U_p\end{aligned}\quad (22)$$

for an 800 nm pulse. Thus we obtain for the ionization energies 0.94 a.u. (25.58 eV) and 1.33 a.u. (36.19 eV) for the cases of moving and fixed nuclei for a 532 nm laser pulse, and 1.06 a.u. (28.84 eV) and 1.42 a.u. (38.64 eV) in the cases of moving and fixed nuclei for an 800 nm laser pulse. Thus H_2^+ has a higher ionization energy when exposed to an 800 nm laser pulse than to a 532 nm laser pulse. We explain this phenomenon as follows. The nuclei in H_2^+ respond differently to external fields with different wavelengths. Therefore the molecular potential that the electron senses is altered differently for 532 nm and 800 nm laser pulses. Thus in the molecular system the ionization energy is dependent on the wavelength (or frequency) of the external laser field. We note that the cutoff energy may be extended far further for longer laser pulses and for an initial excitation of the molecule in a two-step process [11].

In Fig. 11 we consider the relative velocity and accelera-

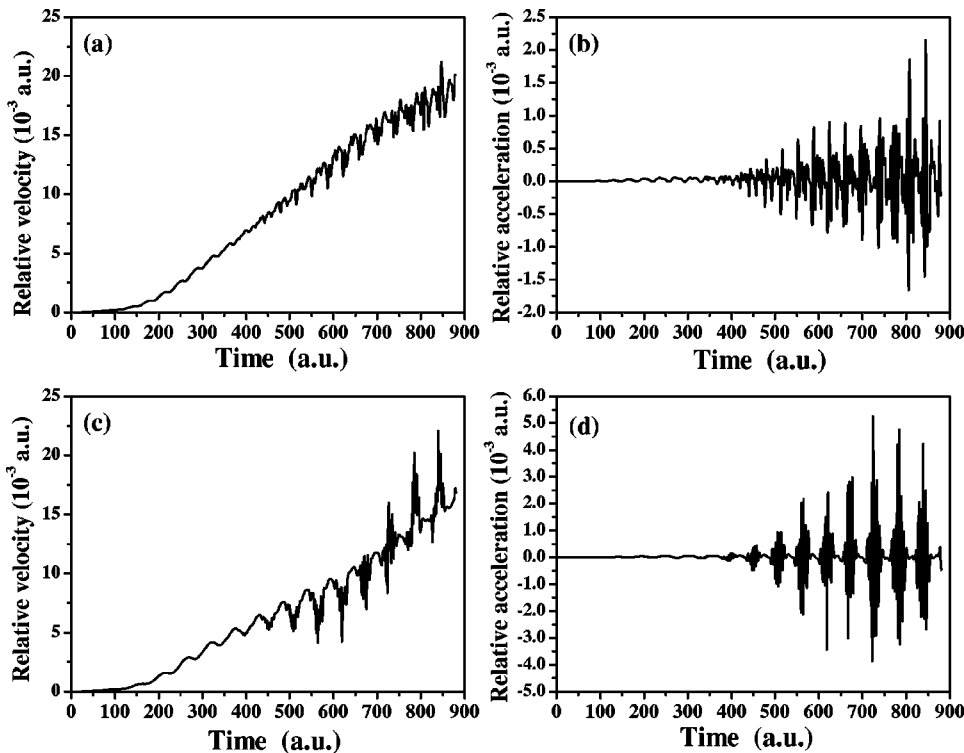


FIG. 11. The relative velocity \dot{R} and the relative acceleration \ddot{R} of the two nuclei of H_2^+ in a laser pulse of shape given in Eq. (20) and intensity $4.0 \times 10^{14} \text{ W/cm}^2$ and wavelength 532 nm [(a) and (b)] and 800 nm [(c) and (d)]. The initial states are given by the ground state.

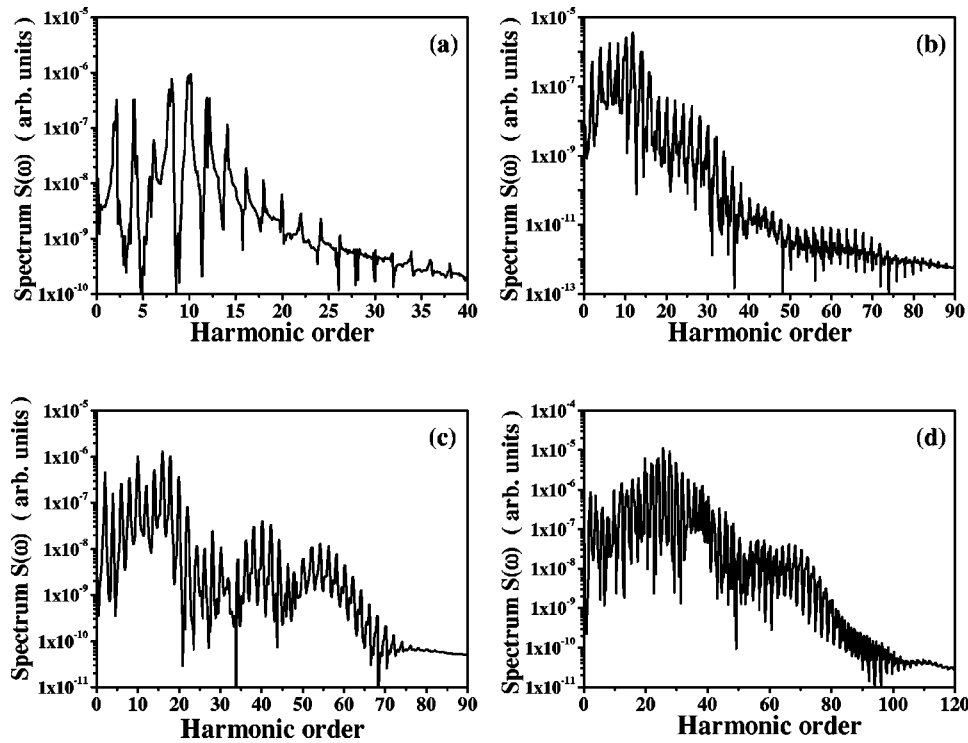


FIG. 12. The radiation spectra arising from the relative acceleration of the two nuclei in the H_2^+ molecule. The laser pulses with shapes following Eq. (20) are further characterized by the wavelengths 532 nm [(a) and (b)] and 800 nm [(c) and (d)] and the maximal intensities of 2.5×10^{14} W/cm² [(a) and (c)] and 4.0×10^{14} W/cm² [(b) and (d)]. H_2^+ is initially prepared in the ground state.

tion between the nuclei, $\dot{R}(t)$ and $\ddot{R}(t)$. We note that although the average distance between the nuclei increases monotonically with time (see Fig. 5), the relative acceleration of the nuclei does not. The reason is that the laser-driven electron [or the wave packet $|\Psi(z, r, t)|^2$] surrounds the nuclei and spreads outward. When the electron enters the area between the nuclei, however, the relative velocity of the nuclei decreases. In particular, when the electron moves to one side of the nuclei, the relative velocity increases. Thus the relative acceleration of the nuclei changes periodically as do the electrons in the field of the laser pulse. This is even clearer from Fig. 12, where the spectra of the relative accelerations of the nuclei for the 532 nm and 800 nm pulses with peak intensities of 2.5×10^{14} W/cm² and 4.0×10^{14} W/cm² are presented. High even multiples of the applied laser frequency are visible. Furthermore, the asynchronization in acceleration of the nuclei due to the electron leads to a split of the spectral peaks in Fig. 12, especially for the laser pulse with short wavelength (532 nm) and with highest intensity (4.0×10^{14} W/cm²). We note that the radiation $A(\omega)$ and $S(\omega)$ are employing the same scale, i.e., the relative accel-

eration of the nuclei is, not surprisingly, two to three orders of magnitude smaller than that of the electrons.

IV. CONCLUSION

From a dynamical investigation of H_2^+ in short laser pulses of various intensities and wavelengths with and without the BO approximation we were able to find the following. (a) The nuclear motion of H_2^+ in a field of intense ultrashort laser pulses induces an enhanced ionization with a dependence of the ionization energy on the applied laser wavelength. (b) A periodic shielding of the repulsion of the nuclei by the electron is shown to yield highly nonlinear accelerations between the nuclei. (c) In particular for long wavelengths, we find a broadening of the cutoff regime and a broadening of the harmonics themselves due to the nuclear motion. This is associated with the increased deviation of the recollision times and energies with moving nuclei.

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