Control of electron localization in the dissociation of H_2^+ and its isotopes with a THz pulse*

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The molecular dissociation with a two-laser-pulse scheme is theoretically investigated for the hydrogen molecular ion (H_2^+) and its isotopes (HD^+ and HT^+). The terahertz pulse is used to steer the electron motion after it has been excited by an ultrashort ultraviolet laser pulse and an unprecedented electron localization ratio can be achieved. With the coupled equations, the mass effect of the nuclei on the effective time of the electron localization control is discussed.

Keywords: dissociation probability, hydrogen molecular ion, terahertz pulse, time-dependent Schrödinger equation

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1. Introduction

Diatomic molecules and molecular ions in intense laser fields continue to attract the attention of the atomic and molecular physics community despite several years of study. One of the main goals in this research direction is to find a way to selectively break and form molecular bonds in photochemical reactions.[1] With the advent of new laser technologies, in particular the carrier envelope phase (CEP) stabilized few-cycle pulses and the isolated subfemtosecond pulses, several control strategies have been proposed, taking advantage of controlling the motion of the electron wave packet in atoms and molecules.[2–9] The hydrogen molecular ion is of special interest since it is the simplest molecule, consisting of three particles only. It plays an important role in the theoretical and experimental studies of electron localization in the dissociation of molecules. In the previous work,[10–12] coherent control with a two-color laser field to investigate the asymmetry dissociation of HD^+ and H_2^+ was reported. Then the shaped laser pulse was proposed to control the dissociation pathway of H_2^+. Compared with a single pulse, two sequential pulses were proven to locate more electrons on the selected pathway.[13–16] The combination of an ultraviolet (UV) pulse and a time-delayed near-infrared pulse was used to realize an electron localization probability as high as 85% on one of the two nuclei.[13] More recently, a similar electron localization probability was obtained by using two-color mid- and near-infrared laser pulses.[16]

In our previous work,[17,18] a time-delayed terahertz (THz) pulse was utilized to steer the electron motion of H_2^+ between the two protons after an ultrashort UV laser pulse was used to excite the electron wave packet onto the dissociative $2p\sigma_u$ state.[19–24] Both the THz and the UV pulses were linearly polarized. By adjusting the time delay of the two pulses and the peak intensity of the THz pulse, an unprecedentedly high control ratio (the probability to localize electrons on one of the two nuclei) of 99.3% could be obtained, with almost zero ionization and 6.14% dissociation probability. The electron localization ratio could still be 96.3% even when the total dissociation probability reached 25.6%. For the dissociation of a diatomic molecule, a bound electron is driven back and forth between the potential of the dissociating molecule until the point where it can no longer overcome the barrier between the two nuclei, which is dependent on the distance between the two nuclei.[13] The efficient dissociation control is only possible within a brief period of time, which is called the effective time for electron localization, and the final state is sensitive to the direction and the field intensity of the electric field of the control pulse.[17,25] In this paper, we investigate the dissociation control of H_2^+ and its isotopes with the combination of UV and THz pulses in Sections 2 and 3. In Section 4, the relationship between the effective time of the electron localization and the nuclear mass of the molecule is discussed.

2. Simulation model and parameters of the electric fields

We use a reduced-dimensional model for the H_2^+ molecular ion and its isotopes in the calculation. The molecular axis is assumed to be parallel to the polarization direction of the

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two linearly polarized laser fields. Then we can use the one-dimensional (1D) non-Born–Oppenheimer time-dependent Schrödinger equation (TDSE) for the simulation.[26] The corresponding TDSE can be written as ($e = \hbar = m_e = 1$ in atomic units (a.u.), which are used throughout the paper unless otherwise stated)[17,27–30]

$$i\frac{\partial}{\partial t}\phi(z,R;t) = [T + V_0 + W(t)]\phi(z,R;t), \tag{1}$$

where T is the free Hamiltonian of the system, V_0 stands for the Coulomb interactions, and $W(t)$ describes the interaction with the external laser field. For our model, the kinetic energy T reads

$$T = -\frac{1}{2\mu}\frac{\partial^2}{\partial R^2} - \frac{1}{2\mu_e}\frac{\partial^2}{\partial z^2}, \tag{2}$$

and the Coulomb potential is

$$\begin{aligned} V_0 = & -[1+(z-Rm_n/(m_p+m_n))^2]^{-1/2} \\ & -[1+(z+Rm_p/(m_p+m_n))^2]^{-1/2} + 1/R, \end{aligned} \tag{3}$$

where R is the relative internuclear distance, z is the electronic coordinate with respect to the center of mass of the two nuclei, and $\mu = (1/m_p + 1/m_n)^{-1}$ and $\mu_e = (m_p + m_n)/(m_p + m_n + 1)$ are the reduced masses, with m_e, m_p, and m_n being the masses of the electron, the proton, and the second nucleus ($m_e = 1$; $m_p = 1837$; $m_n = 1837$, 3674, 5511 for H_2^+, HD^+, HT^+, respectively).

The interaction with the laser field is written in the dipole approximation (in the length gauge) as

$$\begin{aligned} W(t) = & -\left(\frac{m_p - m_n}{m_p + m_n}R + \frac{m_p + m_n + 2}{m_p + m_n + 1}z\right) \\ & \times [E_1(t) + E_2(t - \Delta t)]. \end{aligned} \tag{4}$$

The UV pulse is defined as $E_2(t) = E_{20}\sin(\pi t/T_2)^2 \sin(\omega_2 t)$, where T_2 is the total length of the pulse, i.e., 7.9 fs. The THz pulse is defined with the vector potential $A(t) = -E_{10}/\omega_1 \sin(\pi t/T_1)^2 \cos(\omega_1 t)$, where T_1 is the total length of the THz pulse; then $E_1(t) = -\partial A(t)/\partial t$. The total simulation time is defined by t_{end}, and Δt is the time delay between the two pulses, which is defined as the difference between the envelope peaks of the two electric fields. When the envelope peak of the THz pulse is ahead, Δt is positive, otherwise, Δt is negative. The two channels of the dissociation of these three molecule ions are defined as[9,13,16,31]

$$\begin{aligned} P_+ &= \int_{5.7}^{R_{max}} dR \int_{Rm_n/(m_p+m_n)-5.0}^{Rm_n/(m_p+m_n)+5.0} dz\,|\phi(z,R;t_{end})|^2, \\ P_- &= \int_{5.7}^{R_{max}} dR \int_{-Rm_p/(m_p+m_n)-5.0}^{-Rm_p/(m_p+m_n)+5.0} dz\,|\phi(z,R;t_{end})|^2, \end{aligned} \tag{5}$$

where R_{max} corresponds to the boundary of the R axis, and $\varphi(z, R; t_{end})$ is the final wave function of our system. In the simulation, $t_{end} = 106.8$ fs when $P_\pm$, the probabilities of the electron localized on one of the protons (left – or right +), are stable. The electron localization starts from about $R = 5.7$ a.u. So, we use $R_0 = 5.7$ a.u. in the definition of $P_\pm$.[17]

In this simulation, the linearly polarized 228-nm UV pulse with an intensity of 5.0×10^{14} W/cm^2 and a pulse length T_2 of 7.9 fs is used to resonantly excite the electron wave packet onto the dissociative state $2p\sigma_u$. Then the 25.6-μm (11.7 THz in frequency) THz pulse with an intensity of 3.8×10^{12} W/cm^2 and a pulse length T_1 of 85.3 fs is used to steer the electron motion.[32] This THz pulse does not induce any further ionization. In the calculation, we set the dissociation asymmetry parameter as $A = (P_+ - P_-)/(P_+ + P_-)$.

3. Simulation results and discussion

To reveal the details of the electron localization control of H_2^+, we define the time-dependent electron probabilities of different parts: $PD_\pm$ for the electrons localized on the right (or left) proton, PG for the electrons of the high vibrational bound states, $PGI_\pm$ and $PI_\pm$ for the ionized electrons of the six sections, respectively. They are given by

$$\begin{aligned} PG &= \int_0^{5.7} dR \int_{-5.0}^{5.0} dz\,|\phi(z,R;t_{end})|^2, \\ PGI_- &= \int_0^{5.7} dR \int_{-z_{max}}^{-5.0} dz\,|\phi(z,R;t_{end})|^2, \\ PGI_+ &= \int_0^{5.7} dR \int_{5.0}^{z_{max}} dz\,|\phi(z,R;t_{end})|^2, \\ PI_- &= \int_{5.7}^{R_{max}} dR \int_{-z_{max}}^{-R/2.0-5.0} dz\,|\phi(z,R;t_{end})|^2, \\ PI_+ &= \int_{5.7}^{R_{max}} dR \int_{R/2.0+5.0}^{R_{max}} dz\,|\phi(z,R;t_{end})|^2, \\ PD_\pm &= \int_{5.7}^{R_{max}} dR \int_{\pm R/2.0-5.0}^{\pm R/2.0+5.0} dz\,|\phi(z,R;t_{end})|^2. \end{aligned} \tag{6}$$

In the definition of $PD_\pm$, we also choose $R = 5.7$ a.u., as depicted in Fig. 1(a). Figure 1(b) shows the temporal evolution probabilities of different parts of H_2^+ with the UV and THz pulses. The delay time between these two pulses is $\Delta t = -9.4$ fs. One can find that for H_2^+, the effective time for electron localization lasts about 16.0 fs. For HD^+ and HT^+, when the same way is applied, the effective time can also be obtained only with the time windows changed to 16.2 fs and 16.3 fs, respectively.

In Fig. 2, we present the probabilities of electrons localized at the right (proton side, cycles) and the left (second nucleus side, stars) z axis, for the interaction of H_2^+, HD^+, and HT^+ with a single ultrashort UV pulse (the gray) or with a two-pulse scheme of the UV and THz spectral regimes with delay $\Delta t = 14.6$ fs (the red). When only the UV pulse is on, for H_2^+, are the electron localization probabilities of the two protons the same. While for the hetero-nuclear molecules, with

the increase of the quality of the second nucleus, more electrons gather along the proton side for the unsymmetirc ground state distribution. When with the two-pulse regime, the dissociation asymmetry parameter A varies as a function of Δt. In the simulation, one can find that for H_2^+, HD^+, and HT^+, the variations of A are almost the same. When $\Delta t = 14.6$ fs, A reaches the maximum, as shown in Fig. 3(a). For H_2^+, when $\Delta t = 14.6$ fs, $P_+ = 0.235$ and $P_- = 0.0097$, 96.0% of all the dissociation electrons can be controlled to the right potential well. For HD^+ and HT^+, when $\Delta t = 14.6$ fs, $P_+ = 0.199$, 0.171 and $P_- = 0.0083$, 0.0071, respectively. The localization ratios of the right nuclei are 96.08% and 96.0%, respectively, as shown in Fig. 2.

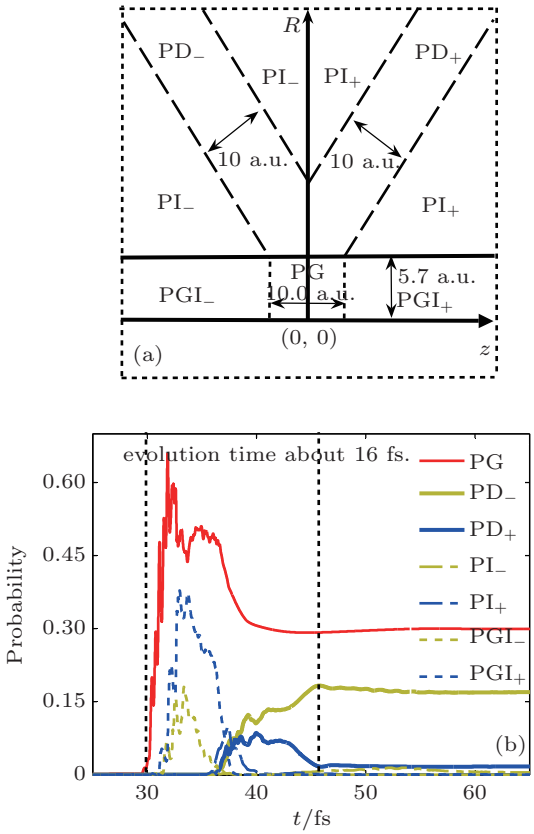


Fig. 1. (color online) (a) Time-dependent electron probabilities of different parts of H_2^+. (b) Time-dependent electron probabilities of H_2^+: $PD_\pm(t)$, $PI_\pm(t)$, $PG(t)$, and $PGI_\pm(t)$, with UV and THz pulses at $\lambda_2 = 228$ nm, $\tau_2 = 7.9$ fs, $I_2 = 5.0 \times 10^{14}$ W/cm^2 and $\lambda_1 = 25.6$ μm, $\tau_1 = 85.3$ fs, $I_1 = 3.8 \times 10^{12}$ W/cm^2, Δ respectively, and $\Delta t = -9.4$ fs.

Figure 3(b) depicts the dissociation asymmetry parameter A and the ionization probability I of H_2^+, HD^+, and HT^+ as functions of the UV pulse field strength E_{20} at $\Delta t = 14.6$ fs and $I_1 = 3.8 \times 10^{12}$ W/cm^2. When E_{20} is small, all the electrons of the dissociative states are excited onto the $2p\sigma_u$ state and a large localization ratio can be obtained with almost zero ionization and a small dissociation probability. For H_2^+, when $E_{20} = 0.0446$ a.u., $A = 0.986$, the electron localization ratio can reach 99.3% and the dissociation probability is 6.14%, with almost no ionization (less than 0.0013%). For HD^+ and HT^+, when $E_{20} = 0.0446$ a.u., the total dissociation probabilities and the electron localization ratios are 5.91%, 6.04% and 99.2%, 99.0%, respectively. The ionization ratio is also close to zero. With the increase of E_{20}, some electrons are excited onto the higher $3s\sigma$ state through the 3-photon resonance, which dramatically damages the electron localization control.[18]

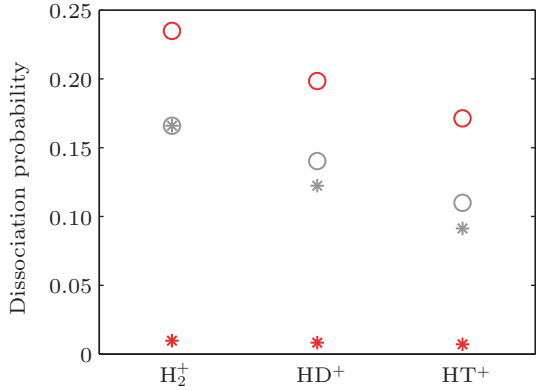


Fig. 2. (color online) Dissociation probabilities with electron localization along the right (proton side, cycles) and the left (second nucleus side, stars) z axis for the interaction of H_2^+, HD^+, and HT^+ with an ultrashort UV pulse at $\lambda_2 = 228$ nm, $\tau_2 = 7.9$ fs, and $I_2 = 5.0 \times 10^{14}$ W/cm^2 (the gray) or with the UV pulse and a THz pulse at $\lambda_1 = 25.6$ μm, $\tau_1 = 85.3$ fs, $I_1 = 3.8 \times 10^{12}$ W/cm^2, and $\Delta t = 14.6$ fs (the red).

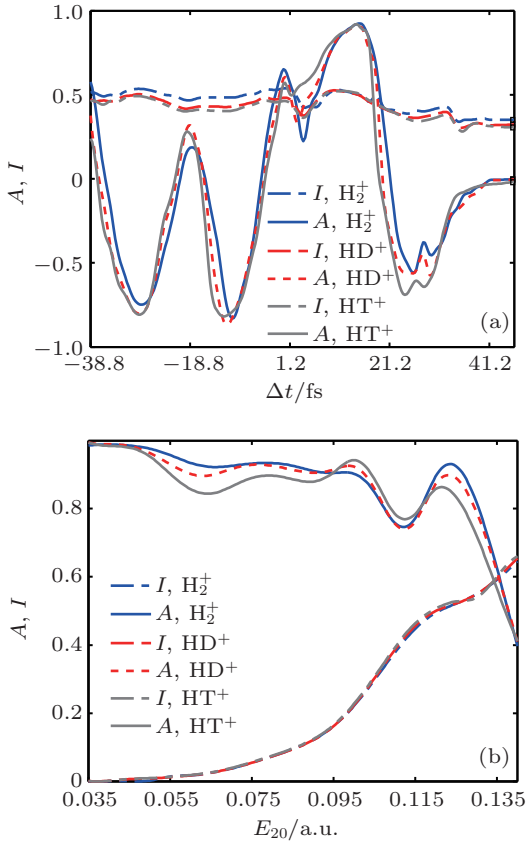


Fig. 3. (color online) (a) Asymmetry parameter A and ionization probability I as functions of Δt with $I_1 = 3.8 \times 10^{12}$ W/cm^2 and $I_2 = 5.0 \times 10^{14}$ W/cm^2 for H_2^+, HD^+, and HT^+. (b) The A and I as functions of the electric field strength of the UV pulse E_{20} with $I_1 = 3.8 \times 10^{12}$ W/cm^2 and $\Delta t = 14.6$ fs for H_2^+, HD^+, and HT^+.

4. Simulation with the coupled equations

But why is the THz pulse useful? As shown in Refs. [17] and [18], the electron localization by using two laser pulses of UV and THz spectral regimes can be much higher than that in the previous work[13] because there is an effective time for controlling the molecular dissociation. The half period of the control electric field needs to match the effective time of the molecule dissociation if a high control ratio is achieved. For H_2^+, this effective time is about 16.0 fs, and when all the electrons of the dissociative states are excited onto the $2p\sigma_u$ state, the dissociation control is implemented more easily. With the coupled equations, the same result can be obtained as with the TDSE[18] . So below, we use the coupled equations to investigate the effective time of homo-nuclear molecules with different masses. The coupled equations can be written as[17,18,33]

$$\begin{aligned} i\frac{\partial}{\partial t}\psi_l &= \left[-\frac{1}{m_p}\frac{\partial^2}{\partial R^2}+\frac{V_g(R)+V_u(R)}{2}+V_{gu}(R,t)\right]\psi_l \\ &\quad+\left[\frac{V_g(R)-V_u(R)}{2}\right]\psi_r, \\ i\frac{\partial}{\partial t}\psi_r &= \left[\frac{V_g(R)-V_u(R)}{2}\right]\psi_l \\ &\quad+\left[-\frac{1}{m_p}\frac{\partial^2}{\partial R^2}+\frac{V_g(R)+V_u(R)}{2}-V_{gu}(R,t)\right]\psi_r, \quad (7) \end{aligned}$$

where $V_g(R)$ and $V_u(R)$ are the binding and the dissociative potentials, respectively. $\psi_l(R,t)=\left(\psi_g(R,t)+\psi_u(R,t)\right)/\sqrt{2}$ and $\psi_r(R,t)=\left(\psi_g(R,t)-\psi_u(R,t)\right)/\sqrt{2}$ are the nuclear wave packets of the left and the right protons, respectively, $\psi_g(R,t)$ and $\psi_u(R,t)$ are the nuclear wave packets of states $1s\sigma_g$ and $2p\sigma_u$, respectively, and $V_{gu}(R,t)$ denotes the interaction with the external laser field. In Fig. 4(a), one can find the same effective time for effectively controlling the dissociation of H_2^+ as in Fig. 1(b). In this simulation, a Gaussian-type wave packet is pre-excited onto the dissociative state ψ_u with center position $R=3.3$ a.u. (resonant with the 228-nm pulse). The effective time increases with the increasing nuclear mass of the homo-nuclear molecule. When the nuclear mass $M=2.0m_p$, the time window lasts 16.0 fs; when $M=24.0m_p$, the time window lasts 53.8 fs, as shown in Fig. 4(b). So only the THz pulse is suitable for the dissociation control of a heavy molecule, no other laser pulse has such a long half period and also a high peak intensity (3.8×10^{12} W/cm^2) now.

Figure 5 shows the largest probability of electrons on the ψ_r proton (the right nucleus) by adjusting the time delay dt between the pre-excitation of the Gaussian-type wave packet and the envelope peak of the THz pulse (25.6-μm), with the nuclear mass of the molecule $M=2.0m_p$, $16.0m_p$, $32.0m_p$.[18] For H_2^+, when d$t=49.2$ fs, more than 99.7% of all the electrons can be controlled onto the ψ_r state, as shown in Fig. 5(a). For $M=16.0m_p$, more than 99.5% of all the electrons localize on the right nucleus when d$t=28.5$ fs, as shown in Fig. 5(b).

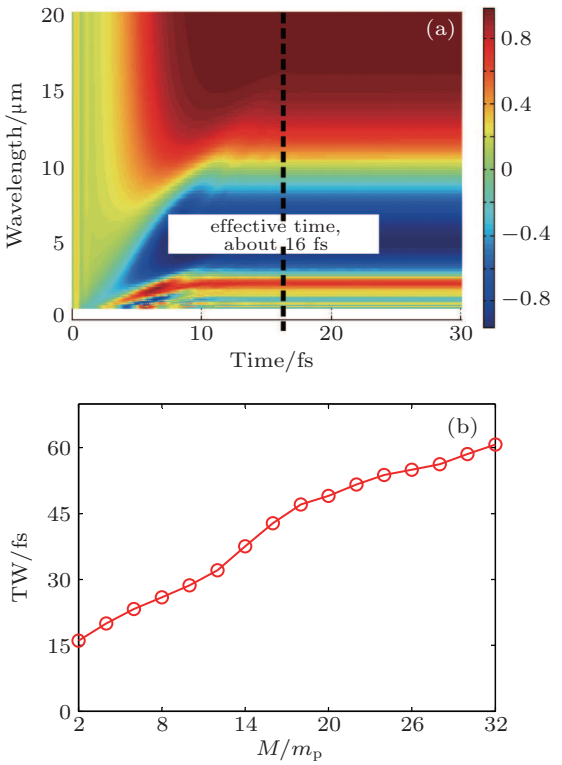


Fig. 4. (color online) (a) Calculated results with the coupled equations (7). Asymmetry parameter A of H_2^+ changes with different wavelengths of the control pulse. All the asymmetry parameters become stable after about 16.0 fs, even though the pulse is very long. (b) The effective time (TW) for dissociation control as a function of the molecule mass M.

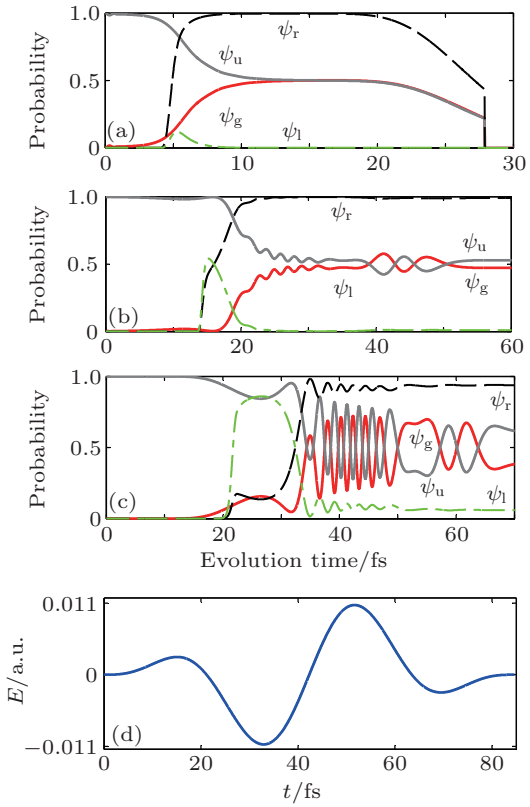


Fig. 5. (color online) Calculated results of the probabilities of electrons on states ψ_g, ψ_u, ψ_l, and ψ_r with the coupled equations (7). The mass of the molecule and the time delay between the pre-excited Gaussian-type wave packet and the THz pulse are (a) $M=2.0m_p$, d$t=49.2$ fs; (b) $M=16.0m_p$, d$t=28.5$ fs; and (c) $M=32.0m_p$, d$t=10.6$ fs. (d) The THz pulse utilized throughout this paper.

For $M = 32.0m_p$, the effective time is more than 60.8 fs, which is larger than the half period of the THz pulse that we chose, which lasted 42.7 fs, as shown in Fig. 5(d). The largest control ratio can only be achieved at some fixed time delay points. For example, when $dt = 10.6$ fs, one can obtain the largest localization ratio of the right nucleus, which can only reach 93.6%, as shown in Fig. 5(c). We have simulated that when the wavelength of the control pulse is doubled, more than 99.5% of all the electrons can be controlled to the right nucleus for $M = 32.0m_p$.

5. Summary

We have shown that the extremely high electron localization can also be achieved in the dissociation of HD^+ and HT^+ as that in H_2^+ by using two laser pulses of UV and THz spectral regimes. The curves of dissociation asymmetry parameter A and ionization probability I of these three homo- and hetero-nuclear molecule ions vary the same with the time delay of the two pulses Δt and the electric field strength of the UV pulse E_{20}. The electron localization effective time, which increases with the increasing nuclear mass of the homonuclear molecule, is almost the same for H_2^+ and its isotopes. By adjusting the time delay, more than 93.6% of all the dissociative events can be controlled to the pointed nucleus, though the molecule is 16.0 times heavier than H_2^+. If the laser wavelength is long enough, the requirement for the laser intensity can be lower, leading to much lower ionization probabilities. Thus, the proposed scheme paves a new approach to efficiently control the electron localization in molecular dissociation.

Appendix A

For the H_2^+ molecular ion and its isotopes, the Schrödinger equation can be written as

$$-\frac{\hbar^2}{2}\left(\frac{1}{m_p}\nabla_p^2+\frac{1}{m_n}\nabla_n^2+\frac{1}{m_e}\nabla_e^2\right)\phi+[V_0+W(t)]\phi=E\phi, \quad \text{(A1)}$$

where m_e, m_p, and m_n are the masses of the electron, the proton, and the second nucleus, respectively, V_0 stands for the Coulomb interactions, and $W(t)$ describes the interaction with the external laser field. Let $\boldsymbol{r}_p$, $\boldsymbol{r}_n$, and $\boldsymbol{r}_e$ represent the coordinates of the proton, the second nucleus, and the electron, respectively, and the polarization direction of the linearly polarized laser field is along the z direction. Then $W(t) = -e\varepsilon(z_p + z_n - z_e)$ and

$$V_0 = e^2\left(\frac{1}{|\boldsymbol{r}_p-\boldsymbol{r}_n|}-\frac{1}{|\boldsymbol{r}_p-\boldsymbol{r}_e|}-\frac{1}{|\boldsymbol{r}_n-\boldsymbol{r}_e|}\right).$$

In the center-of-mass coordinate, equation (A1) (without consideration of the partial equation of the center of mass, as well as the field-nuclei interaction) becomes

$$-\frac{\hbar^2}{2}\left(\frac{1}{\mu}\nabla_R^2+\frac{1}{\mu_e}\nabla_{R_e}^2\right)\phi+[V_0+W(t)]\phi=E\phi, \quad \text{(A2)}$$

where $\boldsymbol{R} = \boldsymbol{r}_p - \boldsymbol{r}_n$ is the relative internuclear distance, $\boldsymbol{R}_e$ is the electronic coordinate with respect to the center of mass of the two nuclei, and $\mu = (1/m_p + 1/m_n)^{-1}$ and $\mu_e = m_e(m_p + m_n)/(m_p + m_n + m_e)$ are the reduced masses.

When the molecular axis is assumed to be parallel to the polarization direction of the laser field and z is the electronic coordinate, which goes along with the molecular axis (R), then we have

$$-\frac{\hbar^2}{2}\left(\frac{1}{\mu}\frac{\partial^2}{\partial R^2}+\frac{1}{\mu_e}\frac{\partial^2}{\partial z^2}\right)\phi+[V_0+W(t)]\phi=E\phi. \quad \text{(A3)}$$

The Schrödinger equation of this system can also be written as

$$-\frac{\mathrm{i}}{\hbar}\frac{\partial}{\partial t}\phi=E\phi. \quad \text{(A4)}$$

With Eqs. (A3) and (A4), one can obtain

$$\frac{\mathrm{i}}{\hbar}\frac{\partial}{\partial t}\phi=\frac{\hbar^2}{2}\left(\frac{1}{\mu}\frac{\partial^2}{\partial R^2}+\frac{1}{\mu_e}\frac{\partial^2}{\partial z^2}\right)\phi+[V_0+W(t)]\phi. \quad \text{(A5)}$$

In atomic unit ($e = \hbar = m_e = 1$), the corresponding TDSE can be written as

$$\mathrm{i}\frac{\partial}{\partial t}\phi(z,R;t)=[T+V_0+W(t)]\phi(z,R;t), \quad \text{(A6)}$$

where

$$\begin{aligned}
T &= -\frac{1}{2\mu}\frac{\partial^2}{\partial R^2}-\frac{1}{2\mu_e}\frac{\partial^2}{\partial z^2},\\
V_0 &= -[1+(z-Rm_n/(m_p+m_n))^2]^{-1/2}\\
&\quad -[1+(z+Rm_p/(m_p+m_n))^2]^{-1/2}+1/R,\\
W(t) &= -\left(\frac{m_p-m_n}{m_p+m_n}R+\frac{m_p+m_n+2}{m_p+m_n+1}z\right)\varepsilon.
\end{aligned}$$

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