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Intensity-dependent study of strong-field Coulomb explosion of H_2

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Abstract: We demonstrate experimentally that in pump-probe experiment of H_2 fragmentation by intense laser fields, the Coulomb explosion (CE) paths induced by the second ionization of dissociating H_2^+ show varied dependencies on the laser intensity. While the charge resonance enhanced ionization (CREI) channel is intensity dependent, the probe induced CE (PICE) channel is intensity independent at certain delay time. By using a classical model, we calculated the dissociation trajectories which agree well with the experimental data.

OCIS codes: (020.2649) Strong field laser physics; (020.4180) Multiphoton processes; (300.6350) Spectroscopy, ionization.

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1. Introduction

The fragmentation dynamics of simple molecules in strong laser fields is an important topic of molecular physics. A wealth of phenomena including bond softening (BS) which is correspond to one-photon excitation (1ω) [1], bond hardening (BH) [2], above threshold dissociation (ATD) which is correspond to net-two-photon excitation (net-2ω) [3], directional dissociative ionization (DDI) [4,5], and CREI [6,7] has been discovered over the past decades. As the simplest molecular system, the hydrogen molecule has been extensively studied both theoretically and experimentally [1–4,8–14]. Among them, femtosecond (fs) pump-probe experiment and the CE imaging technique together are able to probe the temporal evolution of the nuclear wave packets [12–14].

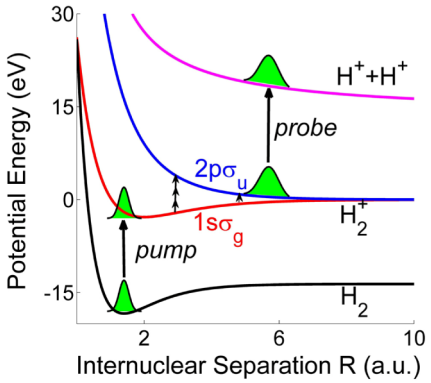

Fig. 1. Schematic view of the relevant potential curves of H_2 and H_2^+ in pump-probe experiments. Three-photon transition and one-photon transition are indicated by three arrows and one arrow, respectively.

As shown in Fig. 1, the pump pulse removes one electron from a neutral H_2, initiating a dissociating vibrational motion of the H_2^+. The second, delayed, probe pulse is then introduced at a fixed time delay to ionize the dissociating H_2^+, projecting the nuclear wave packet to the CE potential. And the protons from the following CE process can then be measured with the kinetic energy (KE) release dictated by the internuclear separation (R) at the moment when the second ionization takes place. This method has been applied to measure the motion of two different dissociative wave packets in I_2 [15], and was then widely used in the measurements of R-dependent ionization probability of diatomic molecules [12–14]. Few-cycle pulses with duration less than 10 fs has also been used to observe the vibrational motion of bound H_2 molecules [13,14].

It has been shown that the KE distribution of protons from CE can be modulated by the laser intensity [16–20]. For CE induced by CREI, the KE spectrum broadens and shifts to higher energy as the laser intensity increases, because the CREI occurs at smaller R as the

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laser intensity is increased [18–20]. In pump-probe experiments, the varying pump and probe laser intensities both can introduce excitation mechanisms to enrich the KE spectrum. However, the effect of field intensity in such the experimental scheme has not been elucidated systematically. In this work, we investigate the intensity dependence of the pump-probe study of the dissociating H_2^+ molecules. The results reveal that the PICE channel is intensity independent at a certain delay time while the CREI channel is intensity dependent, as previously reported [18–20].

2. Experiment and results

The experiments were performed using a Ti:sapphire chirped pulse amplifier laser system (Coherent Elite-HP) with the output pulses of 5 mJ, 40 fs, 1 kHz at the center wavelength of 800 nm. The pump and probe beams were produced by passing a portion of the laser output beam (1.2 mJ) through a Mach-Zehnder type interferometer. A motor stage with a resolution of 0.2 fs is used in one arm of the interferometer to adjust the time delay of the two pulses in the accuracy of 0.8 fs. The dual laser beams were then focused by an f = 75 mm concave mirror inside the ultrahigh vacuum chamber (2×10^{-10} mbar). The product ions from the fragmentation processes are guided by a weak electrical field of 8 V·cm^{-1} to the position-sensitive detector (Roentdek Handels GmbH). Three dimensional momentum vectors of the ions were calculated by its time-of-flight and position on the detector, and were used for determining the coincident events. For H_2 molecule, the coincident events mean two protons [H^+(A), H^+(B)] were detected simultaneously and fulfill the momentum conservation condition, $P_{sum} = |P[H^+(A)] + P[H^+(B)]| < 5$ a. u. (atomic unit).

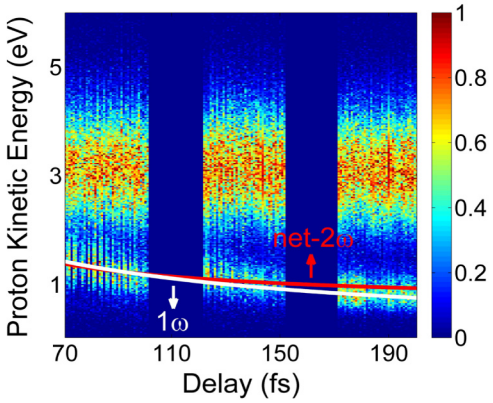

Fig. 2. The measured coincident proton KE spectrum versus the pump-probe delay. Two solid lines: classically simulated dissociation trajectories (KE at different time delay) of 1ω and net-2ω channels.

We first carried out the pump-probe experiments on H_2 molecules with the delay time beyond 70 fs, when the main part of pump and probe pulses were separated enough to avoid the strong interference. The intensity of each pulse was estimated to be 3.5×10^{14} W/cm^2. We choose three time intervals during 70 fs - 200 fs to measure the fragmentation signal for saving the acquisition time, the KE spectra of protons for the coincident events are plotted in Fig. 2. The spectra consist of two parts, the upper stripe around the energy of 3 eV and the lower stripe whose KE is delay dependent. The former is seen to be independent of the time delay, and represents the CREI of the H_2^+ molecular ion at R = 6 a. u [12,13,18–20]. The lower stripe is shown to be dependent on the delay time. As the delay of probe pulses increases, it shifts to the smaller kinetic energy. This is from the PICE at different R of the dissociating H_2^+ ions. This observation is consistent with the previous pump-probe experiments [12,13].We reconstructed the dissociation trajectories by assuming a classical

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motion of the protons, and the simulated kinetic energy shows good agreement with the experimental data as shown by the solid lines in Fig. 2.

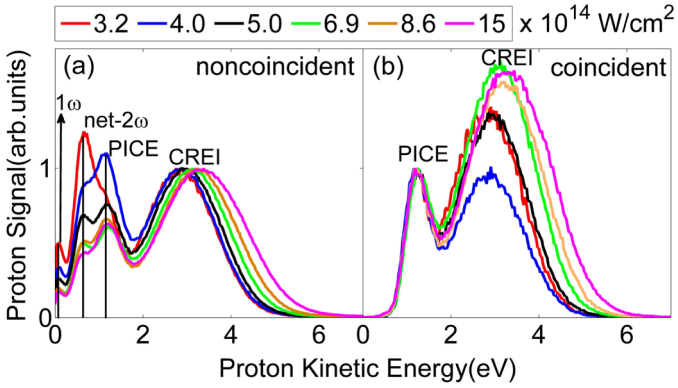

Fig. 3. (a) Noncoincident KE spectra of protons from the pump-probe experiments at the different intensities; (b) Same as (a) but only coincident events.

We changed the intensities of the pump and probe pulses simultaneously while fixing the delay time at 90 fs. Both the noncoincident and coincident proton KE spectra for different intensities are plotted in Fig. 3. The noncoincident spectra shown in Fig. 3(a) include both dissociation and CE events, whilst the coincident spectra shown in Fig. 3(b) include only CE events where two protons [H^+(A), H^+(B)] were detected simultaneously and fulfill the momentum conservation condition, $P_{sum} = |P[H^+(A)] + P[H^+(B)]| < 5$ a. u.. By applying these selection criteria to the measured data, we can separate CE events from dissociation fragments (one H^+ ion and one H atom were released and only one H^+ can be detected).

Figure 3(a) shows there are four peaks in the noncoincident spectrum. Protons with KE near 0.09 eV and 0.63 eV are due to the 1ω path and net-2ω path, respectively, whilst the protons with KE near 1.2 eV and 3 eV represent the PICE and CREI, respectively. The positions of the 1ω peak and the net-2ω peak do not change on varying the laser intensity, which agrees with previous studies [19]. The CREI peak shifts toward higher energy because CREI occurs at smaller *R* as the intensity is increased. By using a single pulse, we also observed similar shift effect for CREI peak, which is consistent with the previous studies [12,13,18–20]. For the PICE peak, no observable shift can be seen following the intensity change because the time delay between the two ionization steps driven by the pump and probe pulses stays relatively unchanged as both the pump and probe intensity are increased in tandem, which means the PICE almost occurs at the same *R* as the intensity is increased. This can be seen more clearly in Fig. 3(b) where only coincident events were shown and the spectra were normalized with proton yield at 1.2 eV.

The relative ratios between different fragmentation channels show complex intensity dependence in our experiment. Roughly, Fig. 3(a) shows the proportions of CE channels (PICE and CREI) increase when compared with dissociation channels (1ω and net-2ω) as the intensity is increased, because more H_2^+ was ionized at higher intensity. Figure 3(b) shows the relative ratio between PICE and CREI changes complexly. As we used two 40 fs laser pulses, the CREI is mainly induced by the second ionization of the pump pulse and the PICE is induced by the second ionization of the probe pulse. Both the second ionization probabilities for pump and probe pulse increased simultaneously as we increased the pump and probe intensities in tandem, which induced the complex intensity dependence.

3. Discussion

We reconstructed the dissociation trajectories by calculating the classical motion of the protons during the fragmentation process. First we estimate the internulcear separation *R* at different delay times. The final velocity *v* of the H^+ ion relative to the H atom is given by

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$$v = \sqrt{2E_d / \mu}, \tag{1}$$

in which μ is the reduced mass of the H_2^+ and E_d is the total KE release of H_2^+ dissociation [21]. For the 1ω and net-2ω channels, E_d (1ω) = 0.18 eV, E_d (net-2ω) = 1.26 eV from our measurement shown in Fig. 3(a).

The internuclear separation R (a. u.) at the delay time τ (fs) can be calculated by

$$R = R_0 + \bar{v} \times \tau, \tag{2}$$

where R_0 denotes the initial internuclear separation at the beginning of the dissociation, and for 800nm pulses, R_0 (1ω) = 4.8 a. u., R_0 (net-2ω) = 3.3 a. u.. Approximately it takes about 3 fs for the wave packet to propagate from 3.3 a.u. to 4.8 a.u [14]. We assume $\bar{v} = v / 2$ because the dissociation is an acceleration motion. The dissociation trajectories of 1ω and net-2ω can be calculated by [15]

$$R(1\omega) = 4.8 + 0.0785 \times (\tau - 3), \tag{3}$$

$$R(net - 2\omega) = 3.3 + 0.2075 \times \tau. \tag{4}$$

The KE of the proton produced by second ionization of dissociating H_2^+ at R can then be given by [21]

$$E_{CE} = \frac{1}{2}(E_d + \frac{e^2}{R}), \tag{5}$$

where e^2/R is the Coulomb repulsion energy of two protons at the internuclear separation R.

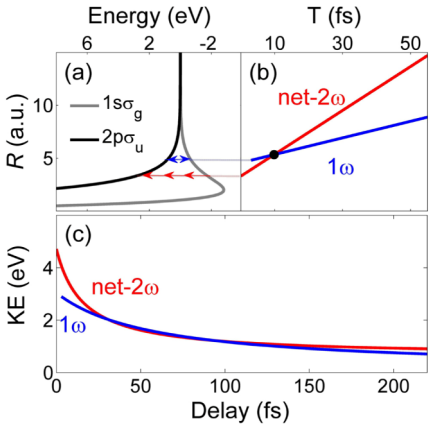

Fig. 4. (a) H_2^+ potential energy curves for the $1s\sigma_g$ electronic ground state and $2p\sigma_u$ first excited state. (b) Simulated dissociation trajectories for 1ω and net-2ω channels. (c) Simulated KE of the protons produced by second ionization of the dissociating H_2^+ at different delay times.

The simulation results are plotted in Fig. 4. Figure 4(a) shows the H_2^+ potential energy curves for the $1s\sigma_g$ electronic ground state and $2p\sigma_u$ first excited state. Figure 4(b) shows the simulated dissociation trajectories for 1ω and net-2ω channels, there is an intersection (9.8 fs, 5.3 a. u.) which was induced by the smaller R_0 and larger $\bar{v}$ for net-2ω channel when compared with the 1ω channel. Figure 4(c) shows the simulated KE of the protons produced by second ionization of the dissociating H_2^+ at different delay times. For the delay times after 70 fs, both curves in Fig. 4(c) present small slopes, which lead to the slow shift of PICE peak versus delay time in the delay-dependent experiment, as shown in Fig. 2.

#241260 Received 20 May 2015; revised 26 Jun 2015; accepted 8 Jul 2015; published 10 Jul 2015

4. Conclusion

In summary, we have observed different dependencies of laser field intensity for different CE channels in H_2 pump-probe experiment. The results reveal that the CREI channel is intensity dependent because CREI occurs at smaller R as the intensity is increased, while the PICE channel is intensity independent at a certain delay time because PICE almost occurs at the same R as both the pump and probe intensity are increased in tandem. We reconstructed the dissociation trajectories by calculating the classical motion of the protons during the fragmentation process, the calculation result agrees well with the experimental data. By choosing different laser pulse intensity and time delay in pump-probe experiment, different KE spectra can be obtained. Our studies complement earlier pump-probe works carried out at single laser pulse intensity. The observed different dependencies for different CE channels can be used to tune the proton KE spectra in H_2 spectroscopy studies.

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