

## Channel-Resolved Above-Threshold Double Ionization of Acetylene

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(Received 17 November 2014; published 22 April 2015)

We experimentally investigate the channel-resolved above-threshold double ionization (ATDI) of acetylene in the multiphoton regime using an ultraviolet femtosecond laser pulse centered at 395 nm by measuring all the ejected electrons and ions in coincidence. As compared to the sequential process, diagonal lines in the electron-electron joint energy spectrum are observed for the nonsequential ATDI owing to the correlative sharing of the absorbed multiphoton energies. We demonstrate that the distinct channel-resolved sequential and nonsequential ATDI spectra can clearly reveal the photon-induced acetylene-vinylidene isomerization via proton migration on the cation or dication states.

DOI: 10.1103/PhysRevLett.114.163001

PACS numbers: 33.80.Rv, 34.80.Ht, 42.50.Hz, 42.65.Re

Electron-electron correlation plays an important role in the single-photon double ionization of atoms [1–3] with the underlying mechanisms of knockout; i.e., one electron absorbs a high-energy photon and knocks out a second electron via a binary collision, or shakeoff; i.e., the sudden departing of a fast electron changes the effective nuclear potential and leads to the release of a second electron. On the other hand, for the multiphoton double ionization driven by strong laser fields [4–12], two electrons are either successively released one after the other or ripped out in a coherent process due to the laser-induced rescattering of a preliberated electron. These processes are called sequential and nonsequential double ionization, respectively. As compared to the tunneling regime of ionization, where the electron escapes to the continuum through the potential barrier suppressed by laser field, above-threshold ionization (ATI) in the multiphoton regime [13] produces an electron of discretized energy by vertically absorbing many photons. Interestingly, the channel-resolved single-electron ATI photoelectron spectroscopy [14] allows one to probe the participation of different electronic continua in strong-field single ionization of molecules. Beyond single ionization, exchanging and sharing of the absorbed multiphoton energies by the freed electrons are expected in nonsequential above-threshold double ionization (ATDI) [15–19]. Recently, this was observed experimentally in argon atoms [20]. To date, however, the experimental demonstration of ATDI is still lacking for strong-field multiphoton ionization of molecules.

ATDI of molecules in the multiphoton regime is capable of resolving the rich and complicated multielectron dynamics of molecules into distinct channels. Among the manifold photon-induced molecular dynamics, isomerization is one of the most fundamental and interesting processes [21]. In particular, acetylene ( $C_2H_2$ ) has attracted extensive interest as a prototype to study the isomerization process [22–31]. For acetylene-vinylidene isomerization, the proton can migrate from one carbon atom to the other on either the

dication [22–28] or cation [29–31] states. Experimentally, the intermediate states for the proton migration are difficult to directly distinguish but are referred from the kinetic energy release (KER) of the ejected ionic fragments, as observed in previous experiments [22–25,29,31]. The molecule isomerizes from acetylene to vinylidene via proton migration on the dication or cation states, which eventually fragmentizes into the ( $C^+$ ,  $CH_2^+$ ) ion pair following double ionization and leads to the events at low and high KERs, respectively. An experimental verification of the intermediate cation or dication states for the intramolecular proton migration is highly desired.

In this Letter, we experimentally investigate the channel-resolved sequential and nonsequential ATDI of acetylene in the multiphoton ionization regime driven by an ultraviolet (UV) femtosecond laser pulse centered at 395 nm. Our results show correlated sharing of the multiphoton energies between the freed electrons in nonsequential ATDI of a polyatomic molecule. By resolving the multielectron dynamics into distinct channels, the observed molecular ATDI further allows us to distinguish the acetylene-vinylidene isomerization via proton migration on the cation or dication states.

The reason for experimental missing of the strong-field molecular channel-resolved ATDI measurement is mainly twofold. First, multiple particles including two electrons and one or two ions ejected from a doubly ionized molecule have to be detected in coincidence. Second, good energy resolution is essential to observe the discretized ATDI peaks in the electron spectrum. To circumvent these difficulties, we perform the measurement with cold-target recoil ion momentum spectroscopy (COLTRIMS) [32,33], where all the ejected electrons and ions are detected in coincidence by two time- and position-sensitive micro-channel plate detectors [34] at the opposite ends of the spectrometer. Meanwhile, an UV laser pulse centered at 395 nm is used, so that we are readily in the multiphoton regime of ionization and the ATI peaks of the electron spectrum are well spaced by the photon energy.

Experimentally, as illustrated in Fig. 1(a), a linearly polarized UV pulse is produced by frequency doubling a near-infrared (IR) femtosecond pulse from a Ti:sapphire laser system (25 fs, 790 nm, 10 kHz, femtolasers) in a 150- $\mu$ m-thick  $\beta$ -barium borate (BBO) crystal, whose polarization can be adjusted to be circular using a quarter-wave plate (QWP). The UV laser pulse is then focused onto a supersonic gas jet by using a concave silver mirror ( $f = 7.5$  cm) inside the vacuum chamber. The molecular jet is produced by coexpanding a mixture of 10%  $C_2H_2$  and 90% He through a 30- $\mu$ m nozzle under a driving pressure of 1.5 bar. As compared to the recently demonstrated two-photon absorption process [35], the temporal duration of the UV pulse in the interaction region is measured to be  $\sim 70 \pm 3$  fs by splitting the UV pulse into two parts and tracing the time-delay-dependent yield of the singly ionized molecule. Since the dispersion introduced by the QWP, the neutral filter, and the viewport of chamber is not compensated, the UV pulse in the chamber is relatively long compared to the driving near-IR pulse, which is checked using a homebuilt transient-grating-based frequency-resolved optical gating [36]. By tracing the field-intensity-dependent shift of the sum energy of electron and proton from the

multiphoton dissociative single ionization of  $H_2$  [37], the peak intensities of the linearly and circularly polarized UV pulses in the interaction region are measured to be  $3.6 \times 10^{13}$  and  $4.3 \times 10^{13}$  W/cm $^2$ , respectively. For the first ionization potential of  $C_2H_2$  ( $I_{p1} \sim 11.4$  eV), the Keldysh parameters [38] for the linear and circular UV pulses are calculated to be  $\gamma = 3.3$  and 4.3, respectively. We hence work in the multiphoton ionization regime.

The electron-to-ion count ratio in the reaction region is about 2:1 with a count rate of  $\sim 0.2$  electrons per laser shot on the electron detector. To identify electrons correlated to various channels and suppress the false coincidence, a momentum conservation gate of  $|p_{z,ion} + p_{z,e1} + p_{z,e2}| < 0.5$  a.u. ( $p_{z,ion} = p_{z,ion1} + p_{z,ion2}$  for the two-body fragmentation channel) is applied. In addition, one of the two detected electrons may originate from the ionization of a second molecule in the same laser pulse with an estimated probability of 30% in our experiment. We eliminate this kind of false coincidence by generating a spectrum using two electrons from different laser shots and subtracting it from the measured raw data [20]. This nicely suppresses the random background and enhances the intrinsic features of the ATDI spectra.

In the following, we will first use the nondissociative  $C_2H_2^{2+}$  to demonstrate the distinct sequential and non-sequential ATDI spectra, and then resolve the acetylene-vinylidene isomerization via proton migration on the cation and dication states by focusing on the two-body fragmentation channel of  $(C^+, CH_2^+)$ .

Figures 2(a) and 2(b) show the electron-electron joint energy spectra (JESs) of two electrons measured in coincidence with the  $C_2H_2^{2+}$  by circularly and linearly polarized UV pulses, respectively. By suppressing the electron recollision process, the  $C_2H_2^{2+}$  is mainly produced by successively removing two electrons through the intermediate  $C_2H_2^+$  in the circularly polarized pulse [5]. After having absorbed  $n_1$  and  $n_2$  photons, the energies of the two sequentially freed electrons approximately calculated as  $E_{ei} = n_i \hbar \omega - I_{pi} - U_{pi}$  ( $i = 1, 2$ ), where  $I_{p1}$  and  $I_{p2}$  ( $\sim 20.7$  eV) are the ionization potentials of the neutral and the singly charged molecule, and  $U_{p1}$  and  $U_{p2}$  are the ponderomotive energies of the first and second electrons, respectively. Correspondingly, the minimal photon numbers to free the first and second electrons are  $n_1 = 4$  and  $n_2 = 7$ , respectively. Due to the lack of energy sharing, as displayed in Fig. 2(a), two successively released electrons lead to discrete islands appearing at the crossing of straight lines corresponding to the absorption of different numbers of photons.

Such energy sharing is, however, expected for non-sequential ATDI [15–20], where two electrons are coherently released and is favored in a linearly polarized pulse. After absorbing  $m$  photons, these two electrons cannot be distinguished from each other with a sum energy of  $E_{e1} + E_{e2} = m \hbar \omega - I_p^{(2)} - U_p^{(2)}$ , where  $I_p^{(2)}$  and  $U_p^{(2)}$  are the effective double-ionization potential and ponderomotive energy of two electrons, respectively. As shown in Fig. 2(b), diagonal lines spaced by photon energy are

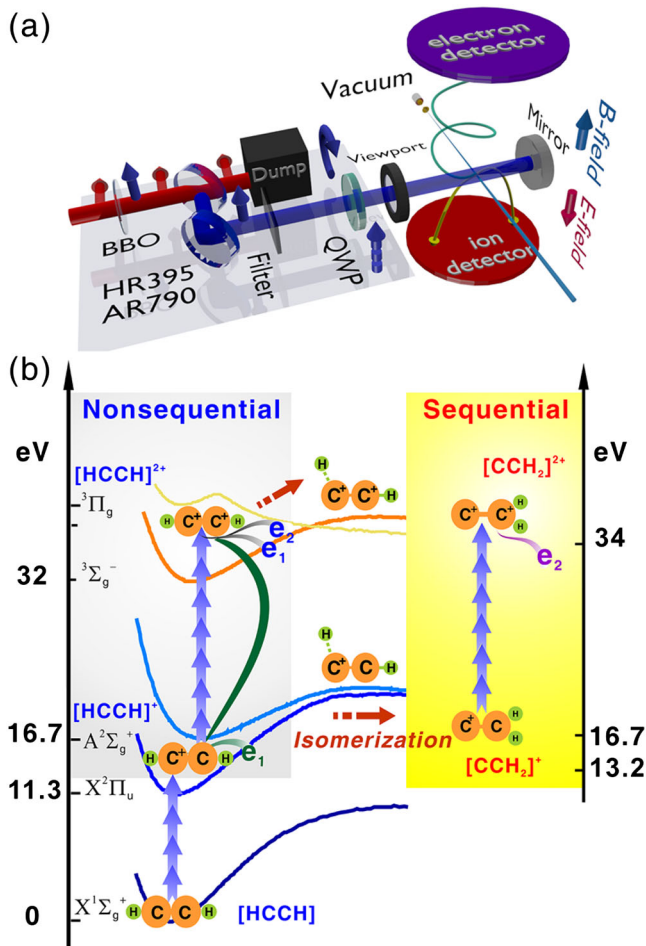


FIG. 1 (color online). Schematic diagrams of (a) the experiment setup and (b) the acetylene-vinylidene isomerization via proton migration on the dication (left) or cation (right) states. The potential curves in (b) are adopted from Ref. [27].

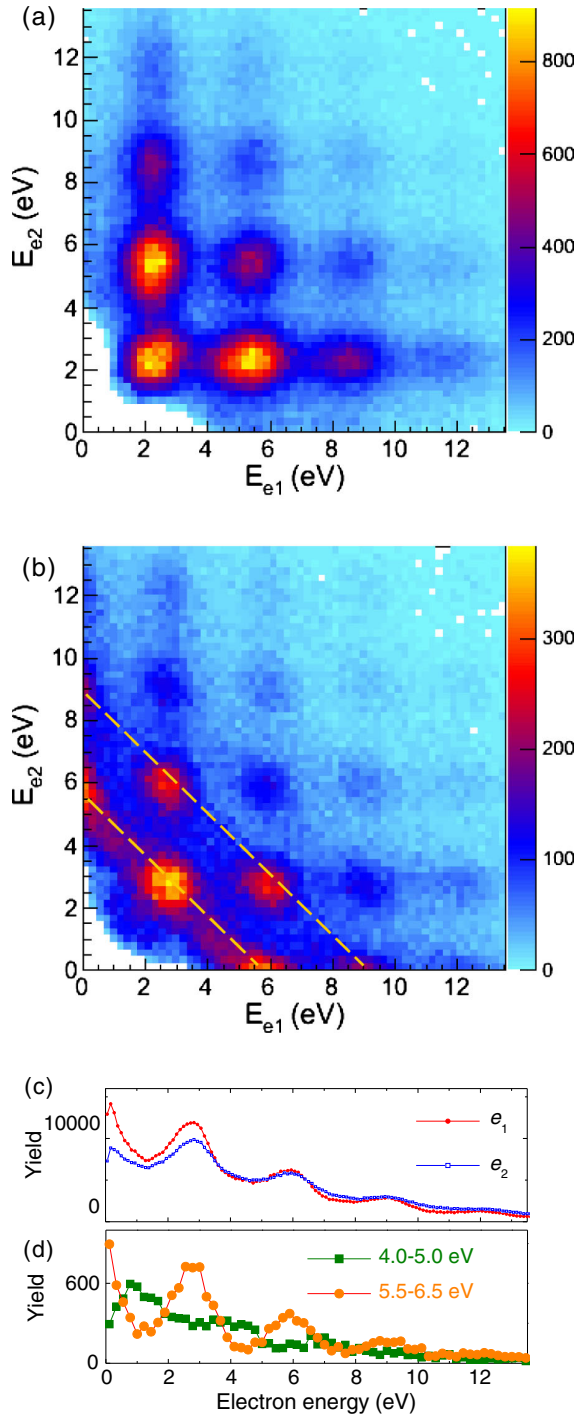


FIG. 2 (color online). (a),(b) Electron-electron JESs measured in coincidence with the  $C_2H_2^{2+}$  by (a) circularly and (b) linearly polarized UV laser pulses. The plots are symmetrized by switching the energies of two directly measured electrons. Tilted dashed lines are used to guide the diagonal structures in (b). (c) Energy of the electrons impacting on the detector as the first and second hits. (d) Projections of (b) to  $E_{e_2}$  for  $E_{e_1}$  in the ranges of 4.0–5.0 eV and 5.5–6.5 eV.

observed in the electron-electron JES for the linear polarization, illustrating the sharing of constant photon energies between two released electrons as an important feature of the nonsequential ATDI [15–20].

For the linearly polarized UV pulse used in our experiments, the maximal energy of the recolliding electron is about  $3.17 \times U_{p1} \approx 1.7$  eV, which is much smaller than the ionization potential of the  $C_2H_2^+$ . We hence exclude the mechanism of direct ( $e, 2e$ ) ionization [39] for the here-observed nonsequential double ionization. Alternatively, an intermediate doubly excited complex can be formed by multiple inelastic field-assisted recollisions [40–45], from which two coherently released electrons share a constant energy as observed in the nonsequential ATDI of argon atoms [20]. We expect a similar scenario of molecular nonsequential ATDI in our linearly polarized pulse, which is mostly suppressed in a circularly polarized pulse by steering the electron trajectory away from the parent ion. Although two released electrons have similar energies ( $E_e \sim 0.1 + m \times 3.1$  eV) when they are considered individually [see Fig. 2(c)], the second electron shows very different energy spectra when the energy of the first one is fixed at various energies, as shown in Fig. 2(d) [20]. This indicates the correlated sharing of the photon energies between two coherently released electrons when they are measured in coincidence.

We will now employ the channel-resolved ATDI electron spectra to reveal the acetylene-vinylidene isomerization via proton migration on the cation or dication states. We identify the acetylene-vinylidene isomerization by observing the coincident creation of a  $(C^+, CH_2^+)$  ion pair. As shown in Fig. 3(a), the  $(C^+, CH_2^+)$  channel clearly distinguishes from the  $(C^+, C^+)$  symmetric breakup channel in the photoion-photoion coincidence map with a yield ratio of about 2:5. As illustrated in Fig. 1(b), after removal of one electron from the next lower-lying highest-occupied molecular orbital, the acetylene can isomerize to vinylidene on the  $A^2\Sigma_g^+$  cation state on a time scale of  $\sim 50$  fs [29–31]. The singly charged vinylidene  $CCH_2^+$  is afterward ionized to the  $CCH_2^{2+}$ , which eventually dissociates into the  $(C^+, CH_2^+)$  ion pair. The stepwise and independent removals of two electrons separated by proton migration [28–31] will smear out any electron-electron correlation. For the  $(C^+, CH_2^+)$  channel, nonsequential ATDI electron spectra can be observed only when the proton migrates on the dication state where two electrons are released within a short time interval. Therefore, the observation of nonsequential ATDI electron spectra indicates the proton migration on the dication state, whereas the proton migration on the cation state leads to sequential ATDI electron spectra. As compared to previous experiments on the time-resolved dynamics of the proton migration [29,31], here we focus on the channel-resolved electron correlation in the photon-induced acetylene-vinylidene isomerization.

Figure 4(a) shows the two-dimensional map of the electron sum-energy versus the KER of the  $(C^+, CH_2^+)$  channel by a linearly polarized UV pulse. Discrete electron sum-energy structure appears at the low KERs ( $E_N < 5.3$  eV), while it becomes structureless at the high KERs ( $E_N > 5.3$  eV), indicating different dynamics. Figures 4(b) and 4(c) show the electron-electron JESs at low and high KERs, respectively. We note that no false coincidence spectrum is subtracted here



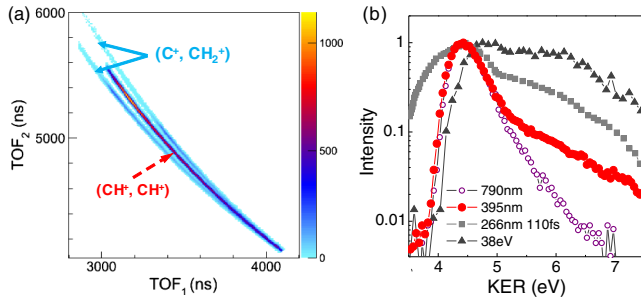


FIG. 3 (color online). (a) Photoion-photoion coincidence maps of the  $(\text{CH}^+, \text{CH}^+)$  and  $(\text{C}^+, \text{CH}_2^+)$  channels. (b) Normalized KER spectra of the  $(\text{C}^+, \text{CH}_2^+)$  channel in the 790-nm laser pulse, the 395-nm laser pulse, the 266-nm laser pulse (adopted from Ref. [31]), and the free electron laser at 38 eV (adopted from Ref. [29]).

for the limited counts attained in the quadruplet coincidence of two ejected electrons with the  $(\text{C}^+, \text{CH}_2^+)$  ion pair. Similar to the nonsequential ATDI spectrum shown in Fig. 2(b), discretized structures along the energy diagonals are observed for the low-KER region [Fig. 4(b)], indicating correlated energy sharing between two freed electrons [15–20]. No such electron energy diagonal is observed for the high-KER region [Fig. 4(c)].

Rather than the tilted lines in the electron-nuclear JES observed in the multiphoton dissociative single ionization of  $\text{H}_2$  [37], we note that the here-observed discretized electron sum-energy lines at low KERs are fairly straight versus the nuclear energy, indicating negligible energy correlation between the ejected electrons and ions. After nonsequential ejection of two electrons, the doubly charged molecular ion isomerizes from acetylene to vinylidene before it breaks into the  $(\text{C}^+, \text{CH}_2^+)$  ion pair. Due to the

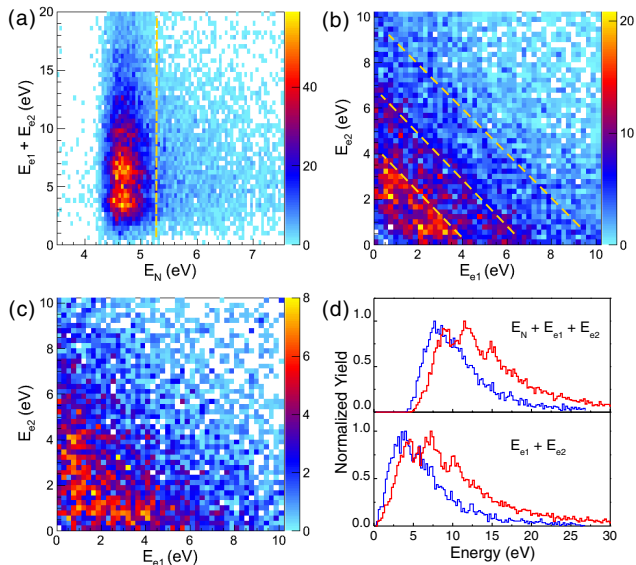


FIG. 4 (color online). (a) KER-dependent electron sum-energy distribution, and electron-electron JESs at (b) low and (c) high KERs measured in coincidence with the  $(\text{C}^+, \text{CH}_2^+)$  channel by a linearly polarized UV laser pulse. The corresponding sum-energy spectra of  $E_N + E_{e1} + E_{e2}$  and  $E_{e1} + E_{e2}$  at low KERs (red curve) and high KERs (blue curve) are shown in (d).

rich electronic structure of the polyatomic molecule, the coupling of various potential curves during the dissociation may smear out the energy correlation between the ejected electrons and the fragmented ions created later. This scenario is validated by comparing the energy spectra of  $E_N + E_{e1} + E_{e2}$  and  $E_{e1} + E_{e2}$ , which show almost the same structures but shift in energy [Fig. 4(d)]. The double ionization may populate several closely lying rovibrational states of the molecule [46] which are difficult to be resolved here for the broad bandwidth of the UV pulse ( $\sim 0.08$  eV) and the variation of the ponderomotive energy along the laser focal point ( $\sim 0.5$  eV). This may broaden the diagonal structures in the electron-electron JES. Nevertheless, as shown in Fig. 4(d), the correlated energy sharing can be clearly identified as the discretized electron sum energy, which corresponds to the energy diagonals in Fig. 4(b).

Differing from the low-KER region, both the energy spectra of  $E_N + E_{e1} + E_{e2}$  and  $E_{e1} + E_{e2}$  are fairly smooth for the high-KER region [Fig. 4(d)]. The structureless spectra indicate the absence of energy correlation between both the two ejected electrons and the electrons and ions, which is consistent with the scenario of stepwise releases of two electrons separated by the proton migration on the cation state. The distinct ATDI spectra at low and high KERs in linearly polarized light hence allow us to reveal the acetylene-vinylidene isomerization via proton migration on the dication or cation states, which are featured with nonsequential or sequential ATDI electron spectra, respectively. The here-identified KER-dependent intermediate states for proton migration are consistent with previous experiments in Refs. [22–25,29–31]. As shown in Fig. 3(b), the high-KER distribution is significantly enhanced in the 395-nm laser pulse as compared to the 790-nm laser pulse. The 395-nm laser pulse induced KER spectrum is consistent with the observations in the extreme UV light from free electron laser [29] or the 266-nm laser pulse [31], where the cation state is assigned to be the important intermediate for the proton migration.

In summary, by measuring all the ejected electrons and ions from a doubly ionized molecule in coincidence, we experimentally demonstrate the channel-resolved ATDI of a polyatomic molecule in the strong-field ionization of the multiphoton regime. The KER-dependent sequential and nonsequential ATDI electron spectra reveal the cation and dication states as the intermediates for the photon-induced acetylene-vinylidene isomerization. The molecular channel-resolved ATDI provides us with a powerful tool to reveal the rich multielectron dynamics of molecules in strong-field multiphoton ionization processes.

We thank R. Dörner, K. Henrichs, and F. He for helpful discussions. This work is supported by the National Natural Science Fund (Grants No. 11425416, No. 11374103, and No. 11434005), the Eastern Scholar program, the NCET in University, a project from the SSTC (Grant No. 13QH1401400), and the ShuGuang project (Grant No. 12SG25).

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