Delayed fragmentation of propyne and allene in collisions with highly charged ions

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Delayed deprotonation dissociation of dications of two C_3H_4 isomers, propyne and allene, induced by 50 keV/u Ne^{8+} ions is investigated employing the reaction microscope technique. The survival time of the precursor $C_3H_4{}^{2+}$ is retrieved according to the time-of-flight difference between the two ionic fragments recorded in coincidence. The survival time spectra of the two isomers exhibit very similar distributions, demonstrating that the initial structure of the isomer does not play a key role in the delayed fragmentation process. In addition, we found that the delayed fragmentation of both isomers could be well described by the power-law decay.

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I. INTRODUCTION

The ionization and dissociation of molecules are of fundamental interest in physics and chemistry, and play important role in many application fields, such as radiation damage of biological tissues [1,2], plasma physics [3], and chemistry of planetary atmosphere and interstellar media [4,5]. Once two electrons are removed, the formed molecular dication is usually unstable and may dissociate into pieces immediately. Nevertheless, it is also possible that the dication keeps stable once the internal energy is low [6–8]. A particular process in between is the delayed dissociation with the dication surviving for a long time before dissociation (e.g., with the timescale ranging from nanosecond to microsecond). Such delayed dissociation has been observed in the fragmentation of many molecules, such as the diatomic dications $N_2{}^{2+}$ [9], CO^{2+} [9–12], HCl^{2+} [13], and SH^{2+} [14], triatomic diactions $CO_2{}^{2+}$ and $CS_2{}^{2+}$ [9], and dication of hydrocarbon molecules $C_2H_2{}^{2+}$ [15,16] and $C_2H_4{}^{2+}$ [16–18]. Besides the molecules listed above, delayed ionization or dissociation is found to be a common phenomenon in the decay of the large ensembles [19], for example, fullerenes [20–26], metal clusters [21], biosystems, such as amino acid [27], adenine [28], and bromouracil [29].

For simple systems, such as diatomic molecules, the lifetime of the intermediate dication is usually determined by the specific states involved. The delayed decay of such systems is well interpreted by the exponential form. For a complex system with many degrees of freedom, it becomes impossible to prepare the system in one or a few specific states due to the high level density. In contrary, the deposited energy may distribute over all the available levels and reach an equilibrium state. If the energy exchange between different internal degrees of freedom is rapidly enough, the decay rate is a function of energy only and is proportional to the ratio of the level densities in the system ensembles of final and initial states. It has been demonstrated in Ref. [21] that the time dependence of the emission rate is close to a power-law distribution generally having an exponent of −1.0. Nevertheless, the exponent of the power law could be higher or lower than −1.0 [19–26]. Taking C_{60} as an example, the ionization with a time delay shorter than 10 μs could be well interpreted by the power law with the exponent of around −0.7 [20,23,24]. The obvious deviation of the exponent from −1.0 observed for C_{60} is attributed to the competition between delayed ionization and delayed evaporation of C_2 [20,23,24,26]. As the systems smaller than C_{60}, the power-law decay has been found to be valid for biomolecules, such as amino acid cations [27] and bromouracil cations [29]. The anions of metal clusters and fullerenes is another category of ensembles that can be interpreted by power-law decay [21,22]. The power-law dependence is found to be valid even for the ensemble as small as $Ag_5{}^-$. The measured decay rate for $Ag_5{}^-$ exhibits an exponent of −1.1 with small deviation from −1.0. Such deviation is well explained by a correction factor considering the degrees of freedom of the system [21].

The delayed deprotonation of small hydrocarbon dications, especially the ethylene dication $C_2H_4{}^{2+}$, has drawn great interest in recent years [16–18]. Larimian *et al.* investigated the delayed fragmentation of $C_2H_4{}^{2+}$ induced by strong laser field and obtained the lifetime of ∼498 ns by using the single exponential fitting [16]. The authors discussed possible vibrational states responsible for the delayed fragmentation process according to the calculated potential energy surfaces. In a later study by Jochim *et al.* [17], it was found that two lifetimes of ∼202 and ∼916 ns associates with the delayed fragmentation of $C_2H_4{}^{2+}$ by fitting the experimental data with a double exponential curve. The possibility of an isotopic effect is also discussed in this paper by comparing the results of $C_2H_4{}^{2+}$ and $C_2D_4{}^{2+}$ [17]. Recently, Takahashi *et al.* [18]

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investigated the delayed fragmentation of C_2H_4 in collision with different species of highly charged ions. It was found that the delayed fragmentation processes are commonly well described by power-law decay rather than the exponential decay. The exponents of power-law fitting are determined to be -1.2 and -1.3 [18]. Besides the different methods employed for ionization procedure, we would underline another difference between Refs. [16,17] and Ref. [18] that the former measurements cover an efficient range of survival time from 300 ns to around 1500 ns, whereas the later one covers an efficient range from 30 ns to around 300 ns. For survival time longer than 300 ns the experimental points in Ref. [18] disperse due to the limited statistics. In this paper, we choose two isomers of C_3H_4, propyne (CH_3CCH) and allene (CH_2CCH_2), as the targets and investigate the delayed dissociation induced by 50-keV/u Ne^{8+} employing the reaction microscope technique.

These two molecules are the smallest isomers that stably exist in the nature. They have been chosen as the model system to investigate the isomer effects in the ionization and dissociation processes [8,30–34]. Such a choice provides a direct comparison between the two isomers to investigate whether the specific structure of the molecule plays an important role in the delayed dissociation process or not. We will demonstrate that the delayed fragmentation of both CH_3CCH and CH_2CCH_2 could be interpreted statistically by the power-law decay with similar values of exponent.

II. EXPERIMENTS

The experiment was performed using the reaction microscope (also called cold target recoil-ion-momentum spectroscopy) [35–37] mounted at the 320-kV platform for multidisciplinary research with highly charged ions locating at the Institute of Modern Physics, Chinese Academy of Science. Since details of the present measurement have been presented in Refs. [8,37], only a brief introduction is given here. The Ne^{8+} beam with the incident energy of 50 keV/u intersects with the supersonic gas jets of propyne and allene. After collision, the scattered projectiles with different charge states are separated in position by an electrostatic deflector and are recorded by a time- and position-sensitive detector. The ionic fragments are extracted toward another time- and position-sensitive detector by uniform electric field. The spectrometer for measuring ionic fragments includes the extracting region with the length of 107.5 mm and the field-free drift region with the length of 215 mm. These lengths are chosen to reach the Wiley-McLaren time focusing configuration to get a better resolution for time-of-flight (TOF) measurement [38]. The uniform electric field used to extract ionic fragments is set to 180 V/cm. This value is much higher than in Refs. [16,17]. The residual Ne^{8+} ion beam without scattering is dumped into a Faraday cup. The scattered projectile and the ionic fragments are recorded in coincidence. Arriving time of the projectile acts as the trigger signal for the determination of the absolute TOF of the fragments. TOF information is used to identify the species of each ionic fragment and to retrieve the survival time of the metastable $C_3H_4^{2+}$.

Two isomers of C_3H_4, CH_3CCH and CH_2CCH_2, are measured successively with the same experimental conditions including the projectile energy, the beam current, the electric field for extracting ionic fragments, and the electric field for deflecting the scattered projectiles. In the present paper, we focus on the delayed deprotonation dissociation channel of $C_3H_4^{2+}$. The data presented in this paper are the triple coincidence events of the H^+, $C_3H_3{}^+$ and the scattered Ne^{7+} ion, i.e.,

$$Ne^{8+} + C_3H_4 \rightarrow Ne^{7+} + C_3H_4{}^{2+} + e^-, \tag{1}$$

$$C_3H_4{}^{2+} \rightarrow H^+ + C_3H_3{}^+. \tag{2}$$

III. RESULTS

Figures 1(a) and 1(b) display the TOF correlation spectrum between the two fragments detected in coincidence for propyne and allene, respectively. The oblique line structure locating inside the red oval in Fig. 1(a) mainly arises from the prompt fragmentation of the $C_3H_4^{2+}$ dication to $H^+ + C_3H_3{}^+$ without delay. Following the oblique line structure, there is a long tail extending toward the point with $TOF_1 = TOF_2 = TOF_0$ in the TOF correlation spectrum. Here TOF_i with $i = 0$–2 denotes the TOF of the nondissociative $C_3H_4^{2+}$, H^+, and $C_3H_3{}^+$, respectively. Such long tail structure is the typical feature of the delayed dissociation process. It originates from the events with the metastable dication surviving for different times before dissociation [11,13,15–18].

In Figs. 1(c) and 1(d), the data are displayed in the correlation plots as a function of the TOF difference ($\Delta TOF = TOF_1 - TOF_2$) and the sum TOF of the two fragments ($\Sigma TOF = TOF_1 + TOF_2$). As shown in these plots, the fragmentation events distribute broadly along ΔTOF. In contrast, ΣTOF changes slightly as ΔTOF increases. ΔTOF information is employed to retrieve the survival time of $C_3H_4^{2+}$. By neglecting the kinetic energies of both fragments obtained during fragmentation, the TOF_i ($i = 1, 2$) is written as

$$TOF_i(t) = \frac{\sqrt{a_0{}^2t^2 - a_i(a_0t^2 - 2L)} - a_0t}{a_i} + \frac{2L}{\sqrt{a_0{}^2t^2 - a_i(a_0t^2 - 2L)}}. \tag{3}$$

Here t is the survival time of the parent $C_3H_4^{2+}$ dication. L is the length of the extracting region of the spectrometer, whereas the length of the drift region is $2L$. a_0, a_1 and a_2 are the accelerations of the parent $C_3H_4^{2+}$, the H^+ and the $C_3H_3{}^+$ in the extracting region, respectively. The survival time for each fragmentation event is obtained by solve the equation,

$$\Delta TOF = TOF_1(t) - TOF_2(t), \tag{4}$$

and displayed in Fig. 2(a). This figure exhibits enhanced intensity with survival time shorter than 20 ns. Events in this region arise mainly from the prompt fragmentation marked by the red oval in Fig. 1(a), but not the true delayed fragmentation events. Following this enhanced area there is a broad distribution extending to around 660 ns. Such events correspond to the delayed fragmentation process.

For delayed fragmentation events, the momentum component along the extracting electric field direction which is deduced from the absolute TOF is missing. An Abel transform is employed to retrieve the three-dimensional momentum dis-

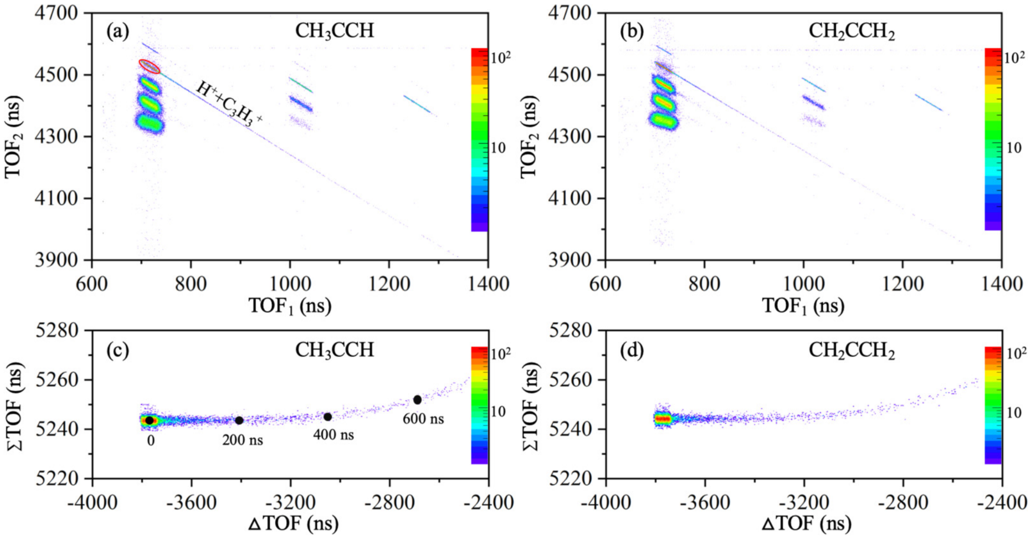

FIG. 1. (a) TOF coincidence spectrum for CH_3CCH, and (b) for CH_2CCH_2. Events inside the red oval in (a) arise from prompt dissociation process. (c) Correlation plot as a function of ΔTOF and ΣTOF for CH_3CCH, and (d) for CH_2CCH_2. Black dots in (c) display selected samples of the time delay between ionization and fragmentation retrieved from ΔTOF.

tributions from the two-dimensional momentum components on the detector plane [16,39]. The kinetic energy release (KER) is then obtained from the retrieved momentum. The blue squares in Figs. 2(c) and 2(d) display retrieved KER for prompt events employing Abel transform. These distributions are normalized to the KER distributions obtained directly from TOF and position information without Abel transform (black dots). The same parameters of Abel transform are then applied to retrieve the KERs of delayed events (red circles). As shown in this figure, the KER distributions of delayed fragmentation are almost the same as prompt fragmentation with small derivation.

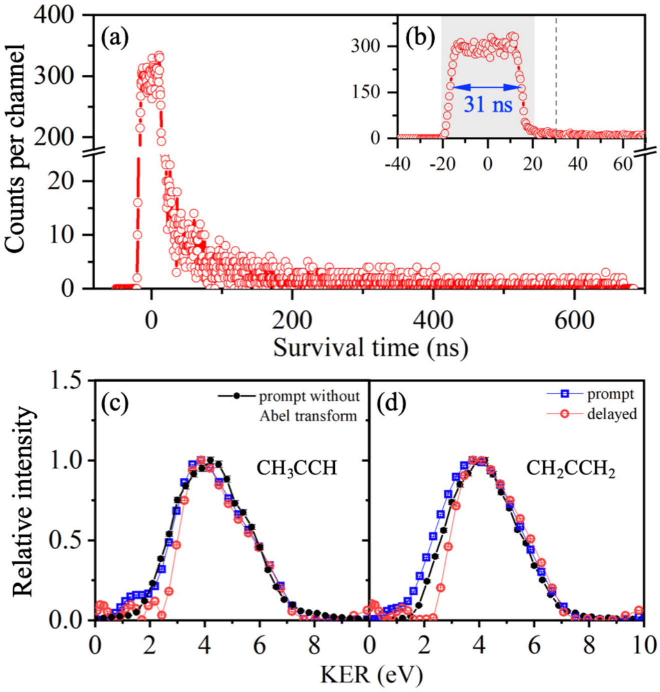

FIG. 2. (a) Retrieved survival time distribution of CH_3CCH. Prompt fragmentation events are included. (b) Zoomed-in view of the contribution from prompt fragmentation. Gray dashed line shows the cutoff for the data presented in Fig. 3. (c) KER distributions of the prompt and delayed fragmentation processes for CH_3CCH, and (d) for CH_2CCH_2.

The resolution of survival time t is mainly determined by the kinetic energies of the two fragments. As shown in Fig. 2(b) survival time distribution for prompt events ($t = 0$) distributes between -20 and 20 ns. It exhibits a plateau structure with the full width at half maximum (FWHM) of 31 ns. The resolution of t for delayed fragmentation is thus estimated to be 31 ns since the KER of delayed fragmentation is almost the same as prompt process. In the following part of this paper, the contribution of prompt dissociation is eliminated by setting the cutoff of the survival time to 30 ns [gray dashed line in Fig. 2(b)].

IV. DISCUSSION

Figure 3 presents the survival time (t) distributions ranging from 40 ± 10 to 660 ± 10 ns. The data are presented in semilogarithmic scale in Figs. 3(a) and 3(c). Also shown in these figures are the fitting results with exponential function,

$$y = Ae^{-t/\tau}. \tag{5}$$

The exponential fitting leading to component τ of 94 ± 6 and 93 ± 7 ns for propyne and allene, respectively. Taking account of the errors for τ caused by the fitting procedure, the fitting results of two isomers are almost the same with negligible difference.

Figure 3 exhibits obvious discrepancy between exponential fitting and experiment for both isomers. The parameter τ of the exponential fitting reflects the lifetime of the metastable state. To be strictly, this stands only if only one state contributes to the delayed fragmentation process. However, in collision with highly charged particles, a broad range of en-

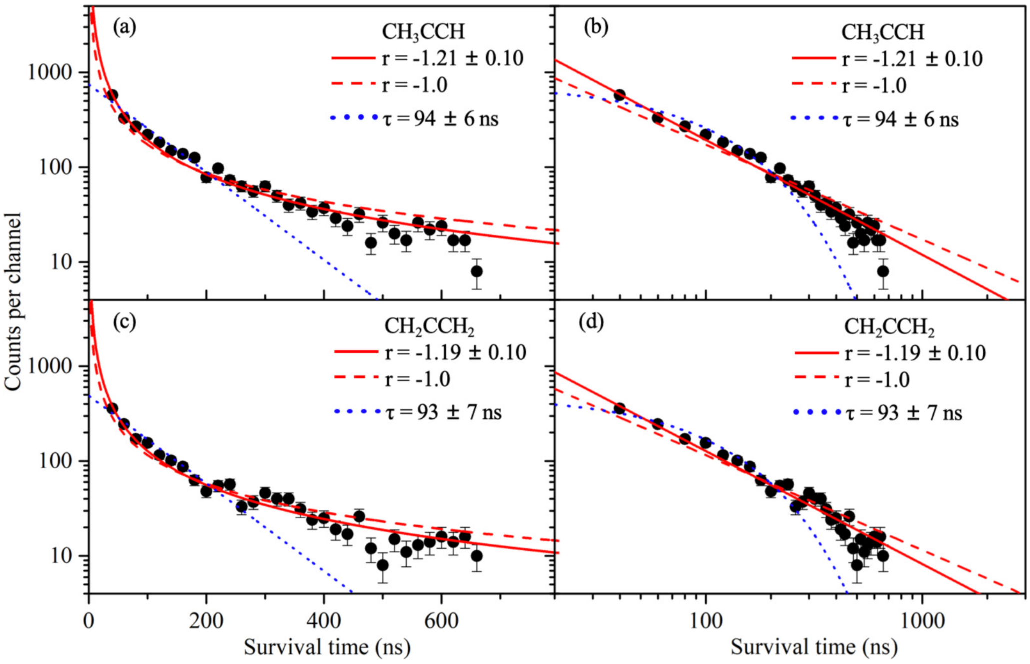

FIG. 3. $C_3H_4^{2+}$ survival time distributions against the deprotonation dissociation channel $H^+ + C_3H_3{}^+$. (a) Semilogarithmic plot and (b) log-log plot for propyne, (c) semilogarithmic plot, and (d) log-log plot for allene. The blue dot curves present the fits by exponential function given by Eq. (5). The red solid curves present the fits by function of power-law decay given by Eq. (6), whereas the red dashed curves present $1/t$ distributions. The experimental data range from 40 to 660 ns with the bin size of 20 ns for all plots.

ergy can be deposited into the C_3H_4 targets leading to the population of different states of the dications. A series of vibrational states with different modes can be populated on each electronic state [16]. The lifetimes corresponding to these different vibrational states are usually quite different from each other, and the lifetime of each vibrational state can be disturbed by rotational excitation [16]. Thus, the lifetime τ obtained by exponential fitting should be considered as the average value of various contributions. In addition, due to the complexity of the C_3H_4 molecules, the vibronic couplings [26] between different states (i.e., intramolecular vibrational redistribution [18]) become of primary importance, leading to the statistical population of various vibrational states. From this point of view, a statistical description of power-law form is necessary for the delayed decay process.

Thus, we switch the semilogarithmic plots shown in Figs. 3(a) and 3(c) to the log-log plots in Figs. 3(b) and 3(d). The log-log plots for both molecules present very similar linear distributions. Red solid curves in Fig. 3 present the fitting results by power law,

$$y = Bt^r, \tag{6}$$

which is equal to

$$\log_{10}(y) = \log_{10}(B) + r\log_{10}(t/1\text{ ns}). \tag{7}$$

The power-law fitting leads to oblique lines with slope of r in the log-log plots. Power-law fittings for propyne and allene exhibit almost the same r values, -1.21 and -1.19, respectively. The errors of r are conservatively estimated to be ± 0.10 for both isomers by combining consideration of the fitting uncertainty and the resolution of the survival time t. In general, the power-law fitting agrees much better with experimental data compared to the exponential fitting.

The exponent values r of -1.21 ± 0.10 and -1.19 ± 0.10 obtained in the present paper are close to the value of -1.2 and -1.3 for ethylene dication in Ref. [18]. The deviation from $1/t$ decay law with $r = -1.0$ toward a smaller r value was rationalized by taking account the finite capacity of the ensemble in previous studies [18,21]. For a system with small size ($s \leqslant 20$ [21]) the exponent r is corrected to

$$r = -\left(1 + \frac{1}{s-1}\right). \tag{8}$$

Here s is the effective number of degree of freedom of the molecular vibrations. The number of degree of freedom for a cluster or molecule with N atoms is given by $s = 3N - 6$. This well explains $r = -1.1$ observed for neutral emission from $Ag_5{}^-$ [21]. However, such a correction factor seems underestimate the deviation for C_2H_4 in Ref. [18] as well as C_3H_4 in our paper. It is suggested in Ref. [18] that such deviation may due to the fact that the internal temperature of the dication is not very high, and s can be smaller than $3 \times 6 - 6 = 12$. During the vibration of a hydrocarbon molecule, a C nucleus takes much lower energy and moves much slower than a proton since it is much heavier. Thus, the motion of proton plays the major role in the redistribution of internal energy among different degrees of freedom. In this respect, it might be reasonable to treat the C atoms as frozen. Following such an approximation, the effective value of s is modified to $3 \times 4 - 6 = 6$ and $r = -1.20$. This r value is very close to the values for C_2H_4 in Ref. [18] and C_3H_4 in our paper.

The two isomers exhibit very similar survival time spectra. Consequently, the parameters τ of exponential fitting as well as the exponent r of power-law fitting are almost the same for the two isomers with negligible difference considering the errors. Such similarities indicate that the isomer effect is negligible, i.e., the definite structure of the isomer does not dramatically influence the delayed fragmentation process. As discussed in Ref. [34], the dication of both isomers will deform to various structures before fragmentation. Due to the complexity of C_3H_4, the couplings between its various degrees of freedom become of primary importance, leading to the statistical population of various states [26]. Following this scenario, the decay process does not depend on the initial structures, and the theoretical description in the state-to-state approach is untreatable. Instead, the decay process can be described statistically by very general intrinsic properties, such as degree of freedom [26]. Thus, the absence of isomer effect is consistent with the fact that the decay processes of both isomers are well described by the power-law form with almost the same exponents.

V. CONCLUSION

The delayed deprotonation dissociations of C_3H_4 isomers, propyne and allene, induced by 50 keV/u Ne^{8+} are investigated employing the reaction microscope. Survival time of the metastable $C_3H_4^{2+}$ is determined according to the TOF difference between the two ionic fragments recorded in coincidence. Both exponential fitting and power-law fitting result in very similar parameters for both isomers, indicating that isomer effects does not play an important role in the delayed fragmentation process. Alternative to the exponential fitting which is found to agree with the delayed fragmentation of ${C_2H_4}^{2+}$ for the survival time longer than 300 ns [16,17], our results demonstrate that the deprotonation fragmentation with time delay between 40 and 660 ns are well described by the power-law decay with the fitted exponents of -1.21 and -1.19 for propyne and allene, respectively. The deviation of the exponents from -1.0 can be rationalized by treating the C atoms as frozen.

The present paper indicates that the power-law decay which has been extensively investigated in the decay of larger ensembles, such as biomolecules [27,29], metal clusters [21], and fullerenes [20–26] is probably a general behavior of the delayed deprotonation dissociation of dications of hydrocarbon molecules with the size bigger than C_2H_4 [18] and C_3H_4.

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