Structure deformation dynamics of acetylene molecules following electron loss and capture collisions of 6 MeV O$^{4+}$ ions

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Structure deformation dynamics of acetylene molecules following electron loss and capture collisions of 6 MeV O$^{4+}$ ions

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Abstract. We investigated structure deformation dynamics of acetylene molecules in charge-changing collisions of 6 MeV O$^{4+}$ ions by means of a momentum 3D imaging technique. We obtained an angular correlation between fragment ions of H$^+$ and C$^+$ produced in (H$^+$+C$^+$+C$^2+$+H), (H$^+$+C$^+$+C$^+$+H) and (H$^+$+C$^+$+CH$^+$) measured for single electron loss and capture collisions. It was found that in fragmentation channel of (H$^+$+C$^+$+CH$^+$) bending motion of C$_2$H$_2$ occurs strongly compared with (H$^+$+C$^+$+C$^2+$+H) and (H$^+$+C$^+$+C$^+$+H). We concluded that the structure deformation is caused by electronic and vibrational excitation.

1. Introduction
In the last decade, structure deformation and bond rearrangement dynamics of molecules have been extensively studied by using a 3D momentum imaging technique [1-10]. This method enables us to obtain information about molecular structure deformation prior to its dissociation since the momentum vectors of product ions are sensitive to the structure. Hishikawa et. al. demonstrated visualization of ultrafast migration of hydrogen in deuterated double charged acetylene ion using intense ultrashort laser pulses [7]. They used a pump-probe method in combination with a 3D momentum imaging technique. They found the deuterium atom to move at first from one carbon site to the other in a short time scale (90 fs) and then to migrate back to the original carbon site by 280 fs, in competition with the molecular fragmentation. Osipov et. al. studied hydrogen migration of C$_2$H$_2$ by inner-core ionization of carbon K shell [8]. They also found the hydrogen atom to move to the other carbon site within 60 fs. By contrast, little is known about structure and bond arrangements of multiply charged molecules for which the deformation process may be different greatly because the lifetime of the molecule prior to dissociation is expected to be shorter than neutral or lowly charged molecules.

In order to understand the deformation process of multiply charged molecules more clearly, we performed collision experiments of 6 MeV O$^{4+}$ ions in a C$_2$H$_2$ molecular target. This is because the swift heavy ion impact can easily produce highly charged molecules [4, 10] and serves as a useful tool to probe the structure deformation of multiply charged molecules. We used a momentum 3D imaging method to obtain information about the structure before its fragmentation for individual fragmentation channel. As the charge-changing collision of
projectiles plays an essential role in ionization and fragmentation of target molecules [10], we also measured the charge state of a outgoing particle.

2. Experiment

Experiments were performed at the QSEC 1.7-MV tandem accelerator facility of Kyoto University. A schematic diagram of the experimental setup is shown in Figure 1. A beam of 6 MeV O\(^{4+}\) ions was carefully collimated (<1 mm in diameter) and was crossed with an effusive molecular beam target of C\(_2\)H\(_2\). A base pressure of the collision chamber was kept below 1 × 10\(^{-5}\) Pa. After collisions, final charge states of outgoing projectile particles were analyzed by an electrostatic beam deflector and detected by a movable solid state detector (SSD). Fragment ions as well as intact ions produced in collisions were measured with a position-sensitive delay line detector (DLD) [11]. Mass distributions of fragment ions were obtained by a time-of-flight (TOF) technique under a Wiley-McLaren spatial-focusing condition. Signals of fragment ions were recorded event-by-event in a digital oscilloscope (Wavepro7000, LeCroy) in coincidence with outgoing projectile ions. In this way, we studied the following single electron (1e) capture and loss collisions.

\[
\text{O}^{4+} + \text{C}_2\text{H}_2 \rightarrow \text{O}^{3+} + \text{C}_2\text{H}_2^{++} + (r - 1)e^- \quad (1\text{-e-capture})
\]
\[
\rightarrow \text{O}^{5+} + \text{C}_2\text{H}_2^{++} + (r + 1)e^- \quad (1\text{-e-loss})
\]

The momentum vectors of fragment ions were deduced from positions \((x, y)\) and flight times \(t\) of these products.

3. Results and discussion

Figure 2 shows a time-of-flight spectrum obtained in 1e-capture collisions of 6 MeV O\(^{4+}\) ions. Peaks are mostly fragment ions like H\(^+\), C\(^+\), C\(^{2+}\) and C\(_2\)H\(_m^+\) (\(m=0,1,2\)) ions, and no other ions such as CH\(_m^+\) and H\(^{2+}\) produced from bond rearrangement reactions are observed.

The angular correlations between H\(^+\) and C\(^+\) measured for 1e-capture collisions are shown in Figure 3 (a)-(c), and those for 1e-loss collisions are in (d)-(f). The abscissa \(\theta\) is the angle between the momenta of C\(^+\) and H\(^+\), defined as, \(P(C^+) \cdot P(H^+) = |P(C^+)||P(H^+)| \cos \theta\). The top figures
In the latter case, C moves in the opposite direction, giving rise to the peak of 160 degrees. As for the case of fragmentation of \((C_2H_2)^{4+}\) and \((H^+)^{4+}\), there are two dissociation schemes of \((HC^{2+}+C^++H^+)^{4+}\) prior to dissociation. In the former case, both C and H move in the same direction after dissociation and separate from each other afterwards as depicted in the figure, giving rise to the correlation angle of 20 degrees. In the latter case, C and H move in the opposite direction, giving rise to the peak of 160 degrees. As for the case of fragmentation of \((C_2H_2)^{3+}\), there is only one dissociation scheme of \((H^+C^++C^+H)^{3+}\) resulting in the final form of \((H^++C^++C^++H)^{3+}\) as drawn in the figures. Hence, the bimodal pattern appears also in this case due to the same reason described above. On the other hand, the angular correlation exhibits single and broader peak structure in case of three-body fragmentation of \((H^++C^++CH^+)^{3+}\), as shown in the bottom figures. This is due to the reason that both C and H move in the same direction after dissociation.

The broader θ-distribution in three-body fragmentation indicates evidently that the bending motion of \(C_2H_2\) is enhanced more strongly in comparison with four-body fragmentation. This can be explained as in the following way. The coulomb repulsion force as well as excitation energy in four-body fragmentation is expected to be larger than three-body fragmentation. Thus, the lifetime of intact parent ions leading to four-body fragmentation may be shorter than three-body fragmentation, because highly charged and/or excited states are more unstable. As a result, the four-body fragmentation channels dissociate immediately, leading to near ground state structures. By contrast, the three-body fragmentation with relatively longer lifetimes has enough time for relaxation to bending structure states.

Figure 3. Angular correlations between two momenta of \(P(C^+)\) and \(P(H^+)\) obtained in single electron capture ((a)-(c)) and loss ((d)-(f)) collisions of 6 MeV \(O^{4+}\) ions.
Molecular deformation may be caused by the following three mechanisms. First is the Renner-Teller effect following $C 1s \rightarrow \pi^*$ transition. Saito et al. showed that this transition causes a bending motion more easily compared with C 1s ionization [9]. In the present collision system, however, the energy deposition to C 1s-electrons is smaller than those of valence electrons, as calculated from the Casp code [12]. Therefore, the Renner-Teller effect may be neglected in the present work. Second is the effect of valence electron excitation. For example, Zyubina et al. [13] reported that, in the first excited triplet state ($1^3\Pi$) of $(C_2H_2)^{2+}$, the linear structure $(HCCH)^{2+}$ is unstable but the vinylidene configuration $(CCH_2)^{2+}$ is stable, and they predicted the structure deformation to the vinylidene configuration from $(C_2H_2)^{2+}$. As fast ion collision is known to produce a large number of electronic excited states, the structure deformation due to this effect is possibly expected to play a role in our collision system. Third is the effect of vibrational excitation as reported by Rajgara et al. who investigated the molecular deformation of $CS_2$ by 120 keV $Ar^{8+}$ impacts [6]. Although the vibrational excitation would hardly occur in our fast collisions, relaxation from electronic to vibrational excitation may certainly take place because the three-body fragmentation has a long lifetime.

We found that the $\theta$-distribution in the four-body fragmentation is essentially the same for both electron loss and capture collisions. On the other hand, there is somewhat large difference in case of the three-body fragmentation, showing broader distribution in loss collisions compared to capture collisions. These results seem also support our remarks described above. Namely, the four-body fragmentation channels dissociate more rapidly without accompanying molecular deformation and do not depend greatly on the electron loss and capture collisions. On the other hand, the three-body fragmentation with relatively longer lifetimes can lead to bending structure more easily. Consequently, the degree of its deformation depends on the excited state population which is probably different for electron loss and capture collisions.

4. Summary
Fragmentation dynamics of $C_2H_2$ molecules by 6 MeV $O^{4+}$ impacts is studied by means of a 3D momentum imaging technique. The decay channel associated with $(C_2H_2)^{4+}$ was identified for the first time to $(HC^2+\cdot C^+\cdot H^+)$ and $(H^+\cdot C^2+\cdot C^+\cdot H)$ prior to its dissociation. We also found that in the three-body fragmentation the bending motion is enhanced strongly in comparison with four-body fragmentation. It is concluded that the structure deformation is certainly induced by valence electronic excitation and vibrational excitation but not by inner shell excitation.

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