Molecular orientation effects in CO fragmentation induced by charge-changing collisions of 6 MeV O\(^{4+}\) ions

T Mizuno\(^1\), T Majima\(^2\), H Tsuchida\(^1\), Y Nakai\(^3\) and A Itoh\(^1\)

\(^1\)Department of Nuclear Engineering, Kyoto University, Kyoto 606-8501, Japan
\(^2\)Genesis Research Institute, Inc., Ichikawa 272-0001, Japan
\(^3\)RIKEN (The Institute of Physical and Chemical Research), Wako 351-0198, Japan

E-mail: mizuno@nucleng.kyoto-u.ac.jp

Abstract. Ionization and fragmentation of CO molecules have been investigated in charge-changing collisions of 6 MeV O\(^{4+}\) ions. Fragment ions from CO were measured in coincidence with outgoing projectile charge states by means of a momentum 3D imaging technique. Cross sections for the production of fragment ions in electron loss and capture collisions were obtained as a function of the angle between the molecular axis and the beam direction. It was found that double ionization (CO)\(^{2+}\) induces rather isotropic molecular fragmentation for both electron capture and loss collisions. On the other hand, triple ionization (CO)\(^{3+}\) was found to induce highly anisotropic fragmentation during electron loss collisions.

1. Introduction
For the last decade, collision-induced ionization and fragmentation of diatomic molecules such as N\(_2\), O\(_2\), CO and NO have been extensively studied by using various projectile ions [1-12]. It is reported that the molecular orientation effect appears distinctively when a target molecule is highly ionized [4-8]. This is evidently a direct reflection of anisotropic distribution of the electron density of the target molecule. So far, correlation between fragment ions and kinetic energy release (KER) of fragment ions have been extensively investigated by means of multiple coincidence techniques [1-8]. However, little is known about molecular fragmentation correlated with charge-changing reactions between collision partners [11, 12]. As collision-induced fragmentation of a diatomic molecule takes place faster than its rotation time and is assumed to occur along the molecular axis, data analysis is often carried out simply by utilizing an axial recoil approximation [1]. Theoretically, molecular orientation effects have been accounted for fairly well by a statistical energy deposition model [7-9].

In this work, we measured fragment ions from CO produced in electron loss and capture collisions of 6 MeV O\(^{4+}\). Measurements were performed by using a technique of momentum 3D imaging, enabling us to achieve a kinematically complete study of molecular fragmentation.

2. Experiment
Experiments were performed at the QSEC 1.7-MV tandem accelerator facility of Kyoto University. 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 CO. A base pressure of the collision chamber was kept below $5 \times 10^{-6}$ Pa. After collisions, final charge states of outgoing projectile particles were analyzed by an electrostatic beam deflector. Fragment ions produced in collisions were measured with a position-sensitive delay line detector [13]. 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 (Wavepro 7000, LeCroy) in coincidence with outgoing projectile ions. From 3D maps of fragment ions, initial momentum vectors of all fragment ions were determined as a function of the orientation angle $\theta$ of CO defined as the angle between the molecular axis and the incident beam axis. We studied the following single electron (1e) capture and loss collisions:

$$\text{O}^4+ + \text{CO} \rightarrow \text{O}^3+ + \text{CO}^{++} + (r - 1)e^- \quad (1\text{e-capture}) \quad (1)$$

$$\rightarrow \text{O}^5+ + \text{CO}^{+} + (r + 1)e^- \quad (1\text{e-loss}). \quad (2)$$

3. Results and discussion

Figure 1 shows a time-of-flight mass spectrum obtained for 1e-loss collisions of 6 MeV O$^{4+}$. One can see that peak widths of monoatomic fragment ions of C$^q$ and O$^q$ are substantially broader than intact CO$^+$ ions, indicating that fragment ions are produced with larger kinetic energies gained from coulomb explosion [10, 11]. Figure 2 is a typical example of time-of-flight map showing correlations between two fragment ions produced simultaneously from CO. Fragment ions arising from various ion pairs of (C$^+$, O$^+$), (C$^{2+}$, O$^+$), (C$^+$, O$^{2+}$) and (C$^{2+}$, O$^{2+}$) are well separated from one another. From peak intensities of ion pairs of (C$^{2+}$, O$^+$) and (C$^+$, O$^{2+}$), branching ratios from triply ionized (CO)$^{3+}$ ions into these pairs were obtained as follows:

$$\text{(CO)}^{3+} \rightarrow (\text{C}^{2+}, \text{O}^+) : 74\% (1\text{e-loss}), 78\% (1\text{e-capture})$$

$$\rightarrow (\text{C}^+, \text{O}^{2+}) : 26\% (1\text{e-loss}), 22\% (1\text{e-capture}).$$

Relative cross sections for the production of (C$^+$, O$^+$), (C$^{2+}$, O$^+$) and (C$^+$, O$^{2+}$) are presented in Figs. 3–5 as a function of the molecular orientation angle $\theta$. We analyzed these angular...
Figure 3. Production cross sections of the ion pair (C$^+$, O$^+$) as a function of the molecular orientation angle $\theta$ measured for 1e-loss (upper) and 1e-capture (lower) collisions. Solid lines are the fitting results with Eq. (3) using anisotropy parameters $\beta$ as denoted. Dashed lines show $\sin \theta$ for isotropic distributions.

Figure 4. The same as in Fig. 3 but for (C$^{2+}$, O$^+$).

Figure 5. The same as in Fig. 3 but for (C$^+$, O$^{2+}$).
distributions $N(\theta)$ by using the following formula applicable in fast collisions [4]:

$$N(\theta) = N_0(1 + \beta P_2(\cos \theta)) \sin \theta,$$

(3)

where $P_2(x) = (3x^2 - 1)/2$ the second term of Legendre polynomials, $\beta$ the anisotropy parameter and $N_0$ is a normalization constant. The term $\sin \theta$ corresponds to the number of orientation directions at a fixed $\theta$ viewed from an incident ion. Equation (3) reads that, for isotropic fragmentation with $\beta = 0$, $N(\theta)$ becomes a $\sin \theta$ distribution as depicted by dotted lines in the figures. Fitting curves calculated with the best values of $\beta$ are shown by solid lines. Fitting results are reasonably in good agreement with experimental curves. As a whole, fragmentation associated with 1e-capture collisions seems approximately isotropic both in doubly (CO) $^{2+}$ and triply (CO) $^{3+}$ ionization. By contrast, in 1e-loss collisions, molecular fragmentation is highly anisotropic, particularly for triply ionization. This implies that the triple ionization of CO occurs preferentially in parallel collisions ($\theta \simeq 0, 180^\circ$) compared to perpendicular collisions.

Present results of different behaviors in electron loss and capture collisions can be explained qualitatively as in the following way. Firstly, it should be noted that the degree of pure-ionization $n_i$ is different in capture and loss collisions in the production of (CO) $^{+}$ ions. Namely, $n_i$s are $r-1$ and $r$ in 1e-capture and loss collisions, respectively. Thus, it can be stated that the 1e-loss collision is more violent than the 1e-capture collision. Secondary, ionization (electron loss) of a multiply charged projectile ion O $^{4+}$ by a neutral molecule may be limited to only small impact-parameter collisions because a large amount of inelastic energy is required to remove a tightly bound projectile electron. Note that the ionization potential of O $^{4+}$ is 114 eV. In parallel collisions, a projectile ion can undergo close collisions two times with composite atoms, and consequently the electron loss probability is enhanced compared to perpendicular collisions in which the probability may be reduced by about half. On the contrary, electron capture by O $^{4+}$ is expected to occur at relatively larger impact parameters compared to loss collisions. Obviously such distant collisions may not depend greatly on the orientation angle, resulting in nearly isotropic distribution. Siegmann et al also investigated CO fragmentation by 100keV D $^+$ ions[4]. They measured fragment ion pairs without coincidence with projectile final charge states. Their results show that both double and triple ionization induce almost isotropic fragmentation but quadruple ionization induces anisotropic fragmentation due to close collisions. Furthermore, their highly charged ion experiment [6] shows anisotropic behavior to appear more distinctively in higher degrees of ionization. These results are qualitatively consistent with our results.

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