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COLTRIMS Imaging of Molecular Fragmentation Dynamics of CO Molecules Induced by Slow He^{2+} Ions

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Abstract: We study the multiple ionization and fragmentation of CO in collisions with slow He^{2+} ions. Correlated measurements of all fragment ions and the projectile final charge state were performed using our coincidence COLTRIMS imaging technique coupled to projectile charge state separation. Complete fragmentations of CO, following double electron capture, into $C^{m+}+O^{n+}$ with a total charge of 4 were investigated. The kinetic energy release (KER) distributions and their dependence on the molecule-axis orientation and on the projectile ion velocity were studied. A strong dependence of the kinetic energy release on the velocity of the He ions is observed. Furthermore, we find that the KER values are shifted toward higher energies when the molecule is aligned with the beam direction.

Keywords: Ion-molecule collisions; Molecular fragmentation; Kinetic energy release; Recoil-ion spectroscopy; Molecular orientation; Multi-hit COLTRIMS imaging.

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Introduction

The multiple ionization and fragmentation of molecules have recently been an active area of research [1-4]. In a collision with an energetic ion, a target molecule receives a considerable amount of electronic energy, leading to molecular excitation, multiple ionization and fragmentation. Furthermore, the amount of electronic energy deposition into a target is supposed to change considerably depending on the impact parameter between collision partners and on the orientation of the target molecule. Recent and rapid development of multicoincidence and momentum imaging techniques allows to measure important parameters such as molecular orientation before collisions, kinetic energy of fragment ions, initial charge state of molecules prior to dissociation, … and so forth [2,3]. To date, however, collision induced molecular fragmentation using slow ions is not fully understood yet.

In this work, we study the fragmentation processes of CO molecules induced by slow ions. The dependence of kinetic energy release (KER) on the molecular orientation is investigated. Measurements were performed by using a technique of momentum 3D imaging, enabling us to achieve a kinematically complete study of molecular fragmentation. The technique is known as COLTRIMS imaging technique (COLTRIMS stands for cold target recoil ion momentum spectroscopy). It is one of the most powerful imaging techniques. It has a high multi-fragment detection efficiency and an excellent momentum resolution in fragmentation processes. The COLTRIMS technique has been reviewed by Dörner et al. [2].

Experiment

A schematic diagram of our COLTRIMS imaging setup is shown in FIG. 1. Within the COLTRIMS spectrometer, a supersonic gas jet

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intersects with the ion beam. All charged particles formed during the interaction are projected on time and position sensitive detectors by a well-designed electric field configuration. This way, the measurement covers the full 4π solid angle. The fragment ions produced in a single collision were measured in coincidence with the scattered projectile. The final charge state of the projectile was determined using a two-dimensional position sensitive MCP detector located 1.5 m downstream from the target. The 1.5 m flight length for the projectiles after leaving the target region was made for good separation of charge states on the projectile detector ~ 1.5 cm, which has a resolution of 0.2 mm. By measuring the impact position for each fragment ion on the detector and its time-of-flight, the fragment ion trajectory, and thus the ion momentum gained in the fragmentation, can be determined. To improve the momentum resolution, electrostatic lenses can be incorporated into the spectrometer system. Thus, the influence of the extended target region, from where the fragments originate, can be strongly reduced [3, 4]. In this experiment, particle detectors with fast delay-line position read-out were used to enable us to simultaneously detect two particles impacting on the detector at the 'same' instant ($\Delta t<1$ ns) – as long as they are separated in space by at least 10 mm. The data are acquired and stored in list mode event by event. The rate of fragmentation processes that can be processed per second can approach 100 kHz. To ensure good momentum resolution for the ionic fragments, the target must have a well-defined momentum or be at rest in the laboratory frame. Thus, the CO gas jet is made up of a modified Leybold coldhead with a 0.01 mm nozzle mounted to its tip. The gas passes through a copper tube and is precooled in the coldhead before it is allowed to expand through the nozzle. In the resulting directed gas stream, the so-called "zone of silence" is formed. In this region, the internal temperature of the CO gas in the zone of silence is only a few Kelvin. To realize a cold well localized CO target from the expanding gas, the CO jet is clipped out of the zone of silence by a cone-like aperture with a sharp edge (skimmer). For this work, a differential pumping stage with a second skimmer, which is pumped separately, is located between the expansion and the reaction chamber. Thereby, it is guaranteed that the vacuum in the reaction chamber does not become perturbed by the operation of the jet [2,3].

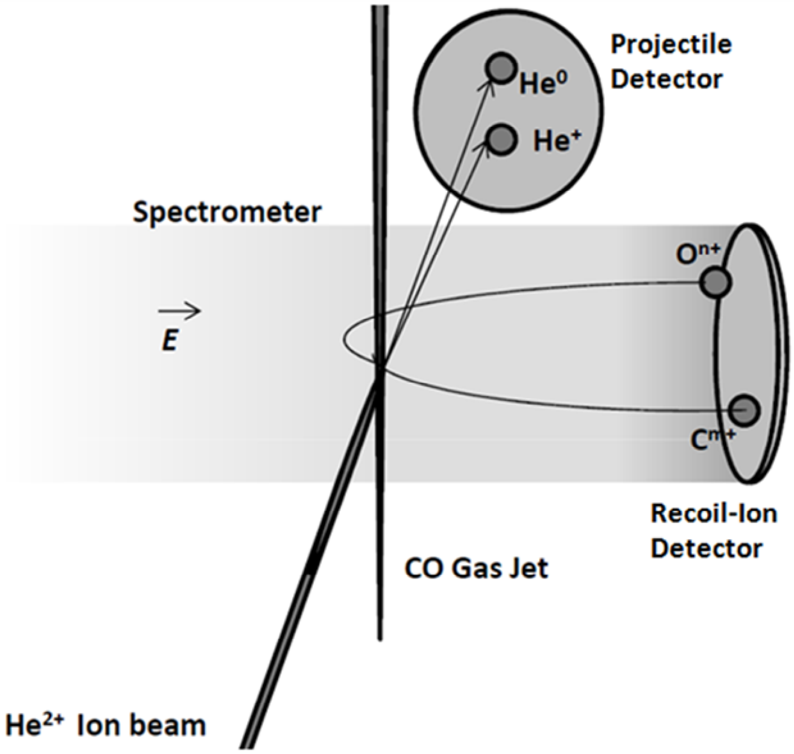

FIG. 1. A schematic diagram of COLTRIMS imaging setup.

Data and Discussion

The fragmentation of CO molecule induced by He^{2+} ion has been investigated at two different impact velocities of 0.7 au and 0.84 au. The fragment ions which resulted from the fragmentation of CO have been measured in coincidence with the scattered projectile. It is well known from the studies of ion–atom collisions at low energy that the capture process dominates the target electron removal and mainly valence target electrons are involved in the capture process [5, 6]. It is thus expected that this process should produce molecular ions in their lowest electronic energy levels. In the current study, we focused on the fragmentation channels which resulted from the capture processes.

It has been suggested that the molecular fragmentation process can be considered a two-step process [7]. In the first step, the neutral CO target is collisionally excited to a dicationic electronic state,

$He^{2+} + CO \rightarrow He^{0} + (CO^{2+})^{*}$

followed by fragmentation,

$(CO^{2+})^{*} \rightarrow C^{+} + O^{+}$.

As mentioned above, the other fragmentation channels: $(C^{2+}+O^{+})$, $(C^{+}+O^{2+})$ and $(C^{2+}+O^{2+})$ have been also detected. For each fragmentation channel, the time-of-flights of the two fragments which resulted from the fragmentation were measured in coincidence with the scattered projectile. FIG. 2 shows the time of flight spectrum (integrated overall impact positions) of the first arriving fragment ion measured relative to the projectile. We observe sharp peaks of CO^{+} and CO^{2+} ions having thermal energies. These ionic peaks arise from direct ionization of the CO molecule. Additionally, four ion fragments (O^{+}, C^{+}, O^{2+}, and C^{2+}) are also formed which arise from the fragmentation of the multiply charged $CO^{q}+$ ($q \leq 4$) ions. One can see that peak widths of monoatomic fragment ions of $C^{m}+$ and $O^{n}+$ are substantially broader than intact CO^{+} ions, indicating that fragment ions are produced with larger kinetic energies gained from Coulomb explosion. To identify the different fragmentation channels, it is convenient to display the data in a Projectile-ion1-Projectile-ion2-Coincidence Spectrum (PIPICO), where the time of flight of the first hit is plotted against the time of flight of the second hit. Correlations in the time of flight of the first and second hit might indicate momentum conservation and thus represent fragmentation channels. In a PIPICO spectrum, these correlations can be recognized as "stripe-like" structures as presented in FIG. 3. From this coincidence map, four fragmentation channels ($C^{+} + O^{+}$, $C^{2+} + O^{+}$, $O^{2+} + C^{+}$, and $C^{2+} + O^{2+}$) originating from the complete Coulomb fragmentation of CO^{q+} ($q = 2$–4) are observed. For all observed four channels shown in FIG. 3, wherein the complete Coulomb fragmentation processes occur, we consider only those counts which obey the law of momentum conservation.

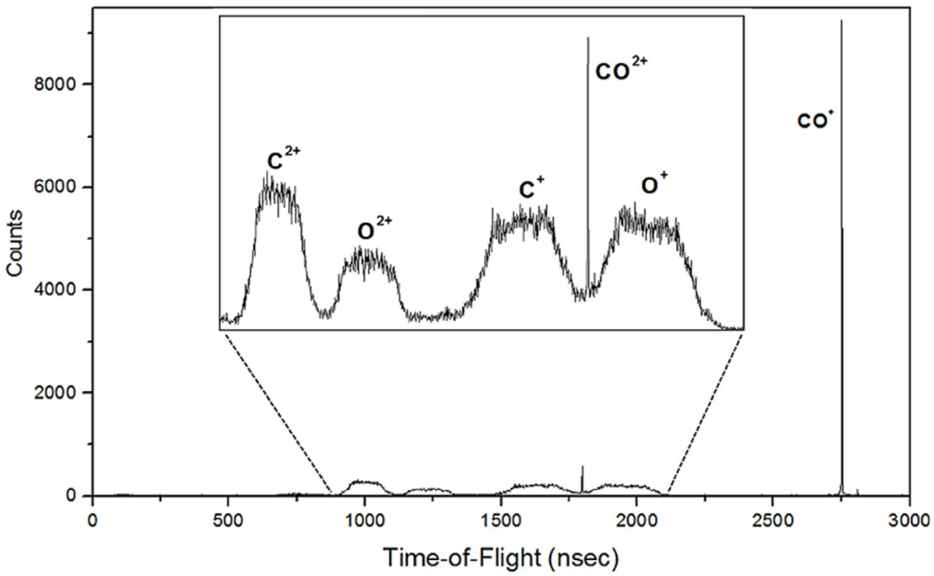

FIG. 2. Time-of-flight (TOF) spectrum of fragment ions from CO fragmentation produced in "double-electron capture" collisions of 0.84 a.u. He^{2+} ions.

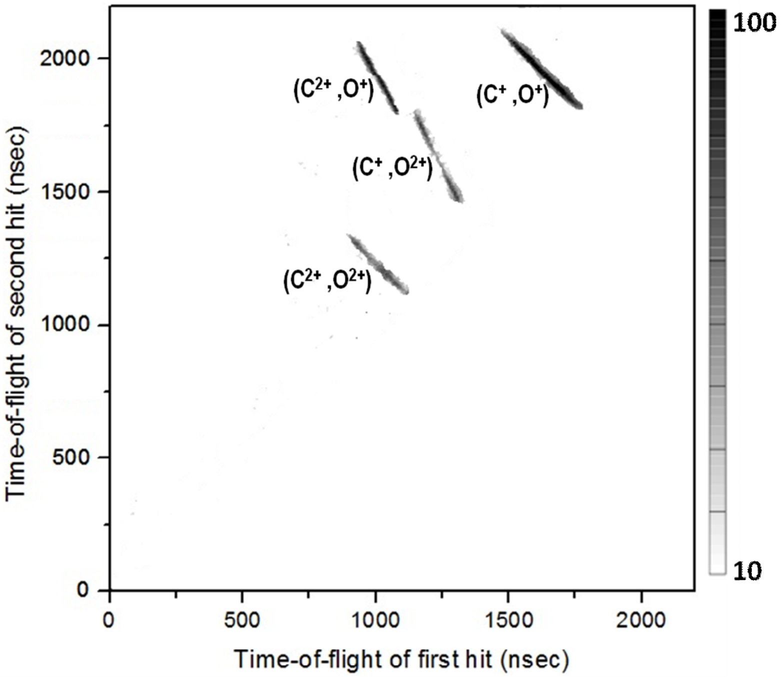

FIG. 3. PIPICO spectrum for CO fragmentation induced by He^{2+} ion impact showing TOF for first fragment *versus* TOF for second fragment. Four fragmentation channels can be observed.

From the experimental results presented in FIG. 3, two main features can be deduced. First, the (C^{+} + O^{+}) and (C^{2+} + O^{2+}) fragmentation channels are stronger than the (C^{+} + O^{2+}) and (C^{2+} + O^{+}) channels. This means that when multiple electrons are removed from the target, charge-symmetric dissociation is the favored dissociation pathway. This phenomenon has been observed in other collision systems [8, 9] and is understood to be a consequence of the ability of the electron cloud to rearrange quickly compared to the much slower nuclear motion in the dissociation process. Second, for the charge asymmetric channels (C^{+} + O^{2+}) and (C^{2+} + O^{+}), the production of multiply ionized carbon ions is generally favored over the production of the corresponding oxygen ions, most likely because oxygen has a larger binding energy. In other words, CO^{3+} fragmentation into the channel (C^{2+}, O^{+}) is significantly larger than the channel (C^{+}, O^{2+}) for all the incident ions. This is attributed, as mentioned above, to the smaller total ionization energy required to produce these states due to the smaller ionization potentials of the C atom compared to the O atom; the ionization potentials required for these ionized states are 49.3 and 59.9 eV for (C^{2+}, O^{+}) and (C^{+}, O^{2+}), respectively.

The dependence of the kinetic energy release (KER) distribution on the projectile ion velocity was investigated for different fragmentation channels. FIG. 4 shows the KER distributions for the C^{+} – O^{+} fragmentation channel for two different impact velocities of 0.7 au and 0.84 au. It is observed that with decreasing the impact velocities, the width of the KER distribution increases and high energy components start to show up in the KER distribution. This indicates that new highly excited states are populated. To understand this behavior, we should investigate the sources of the KER. It is known that the kinetic energy of the two fragments in the final state has two sources: the energy transferred from the projectile during the collision and the energy from the Coulomb repulsion between fragment ions. When the impact velocity decreases the collision time; i.e., the interaction time between the projectile and the molecule will be longer. Therefore, the momentum transfer to the molecule increases and consequently, more energy is collisionally transferred to the CO molecule. This gives the possibility that the resulting transient CO^{2+} ions populate higher excited states. This leads to an energy shift of the KER lines toward higher values. The result supports the picture of the dissociation process as a two-step process [10, 11]. It also confirms that the KER distributions are governed by the fragmentation dynamics; i.e., the potential energy curves of the molecular ion, rather than by the scattering itself.

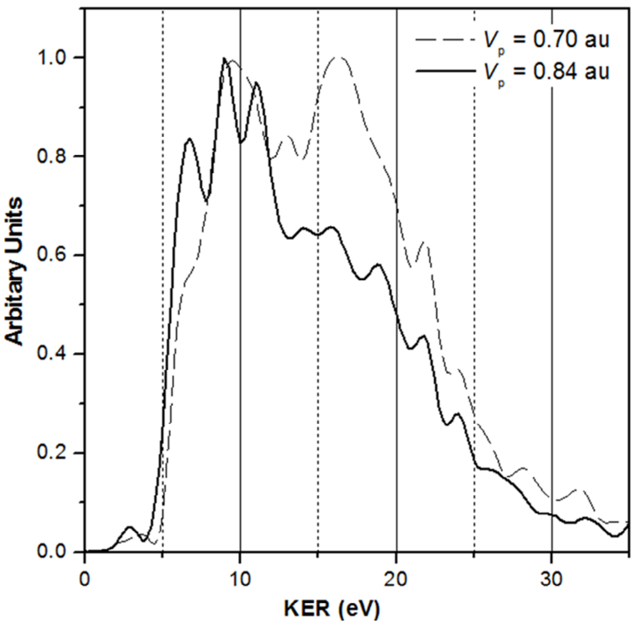

FIG. 4. Kinetic energy release distributions for (C^+ - O^+) fragmentation channel at two different ion velocities (a) 0.7 a.u. (dashed curve) and (b) 0.84 a.u. (solid curve).

Indeed, it is also possible by COLTRIMS imaging technique used in this study to measure, in coincidence with the KER, the angle between the molecular axis and the beam during the collision. This enables us to investigate the molecular orientation effect on the KER distributions. FIG. 5 shows the KER distributions for the C^+ + O^+ fragmentation channel at different molecular orientations. We find that the KER values are shifted toward higher energies when the molecule is aligned with the beam direction. This effect was predicted for slow highly charged projectile and light fragments by Classical Trajectory Monte Carlo (CTMC) calculations [12]. They found that fragmentation in the electric field of slow projectiles leads to energy shifts of the KER lines toward higher values when the molecule is aligned with the projectile beam direction.

Conclusions

The multiple ionization and fragmentation of CO induced by collisions with slow He^{2+} ions were studied. Correlated measurements of all fragment ions and the projectile final charge state were performed using our coincidence COLTRIMS imaging technique coupled to projectile charge state separation. Complete fragmentations of CO into C^{m+}+O^{n+} with a total charge of 4 were investigated. The dependence of kinetic energy release (KER) distributions on the molecule-axis orientation and on the projectile ion velocity was studied. A strong dependence of the kinetic energy release on the velocity of the He ions is observed. It is even noted that the KER values are shifted toward higher energies when the molecule is aligned with the beam direction.

Acknowledgments

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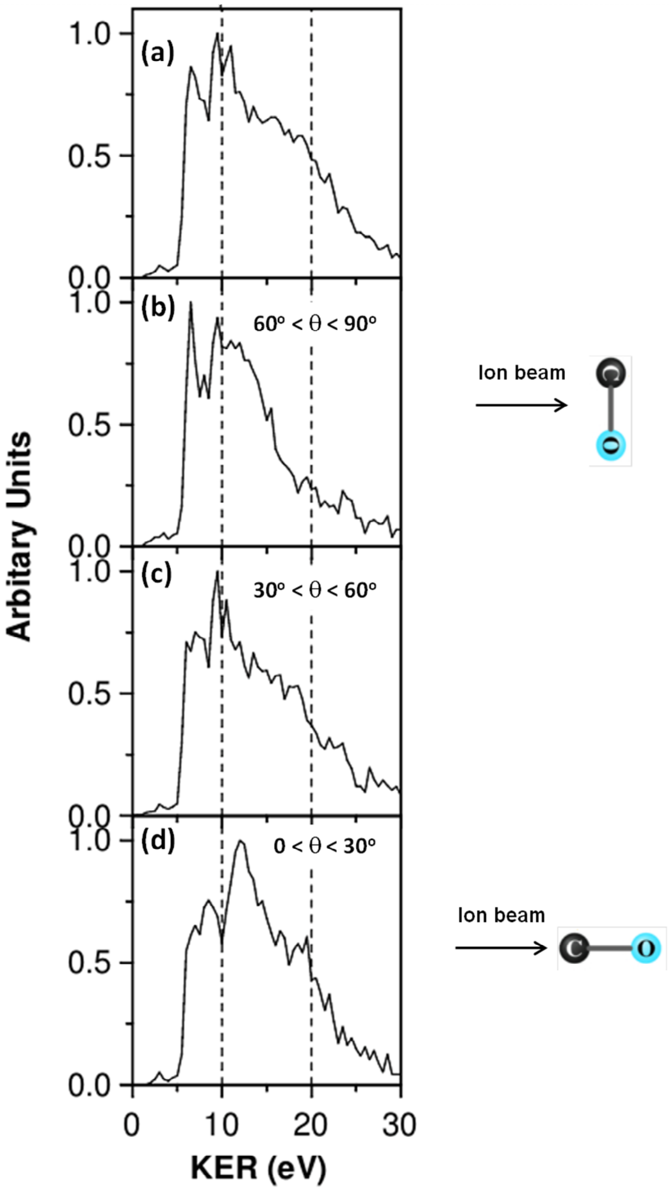

FIG. 5. Kinetic energy release distributions for different molecular orientations.

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