

Momentum imaging of CO fragmentation products

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The breakup of CO molecular ions following multiple electron removal by 2.5 MeV/u Xe³⁴⁺ ions has been studied by means of recoil-ion momentum spectroscopy (RIMS). At 2.5 MeV/u the projectiles travel the approximate length of a CO molecule (3 Angstroms) in only 14 attoseconds, which is much less than the typical time period for molecular rotations and vibrations. Consequently, the molecule remains essentially frozen in space during the interaction with the projectile. Since the interaction time is also much shorter than the typical fragmentation times of the ionized molecules (around 10 fs), the combined final kinetic energy of the molecular fragments formed in a specific breakup channel follows a distribution that reflects the range and relaxation patterns of molecular states excited in the collisions. The relative abundance of the events in each fragmentation channel provides information about the electronic rearrangement during molecular breakup and the total cross sections for multiple ionization of the molecule. The differential cross sections can be obtained from the distributions of molecular orientations at the time of collision.

RIMS is a powerful tool that provides the means for measuring all the quantities described above. They can be derived from the velocity vector components of every charged fragment created in the molecular dissociation event. These velocity components are, in turn, determined from the times of flight of the fragments and the positions of the fragments at the time they hit the detector. The complete set of variables for each ion is measured directly using a multi-hit, two-dimensional, delay-line detector equipped with a hexanode.

Charged fragment pairs originating from parent molecular ions with charge up to 9+ were identified and analyzed. Compared to previous measurements, the current data are significantly improved in terms of accuracy, resolution and overall counting statistics. Some preliminary results are illustrated in Figures 1-5.

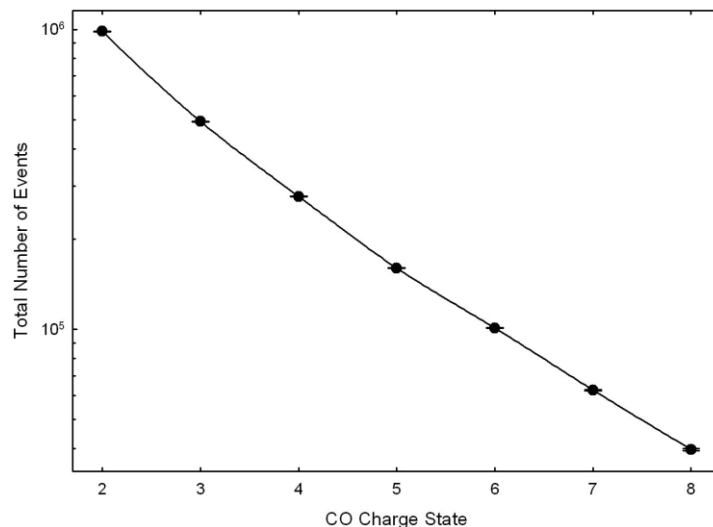


FIG. 1. Total measured number of CO molecular dissociation events as a function of the parent molecule's charge.

As shown in Fig. 1, the total measured number of CO molecular dissociation events is a monotonically decreasing function of the parent molecule's charge. A similar charge dependence was also found for the ionization of noble gas atoms, which indicates that collisions occur predominantly at large impact parameters, where the shape of the molecule and its charge distribution are not important. This is consistent with the direct ionization mechanism, which is expected (and experimentally verified) to be the dominant ionization process.

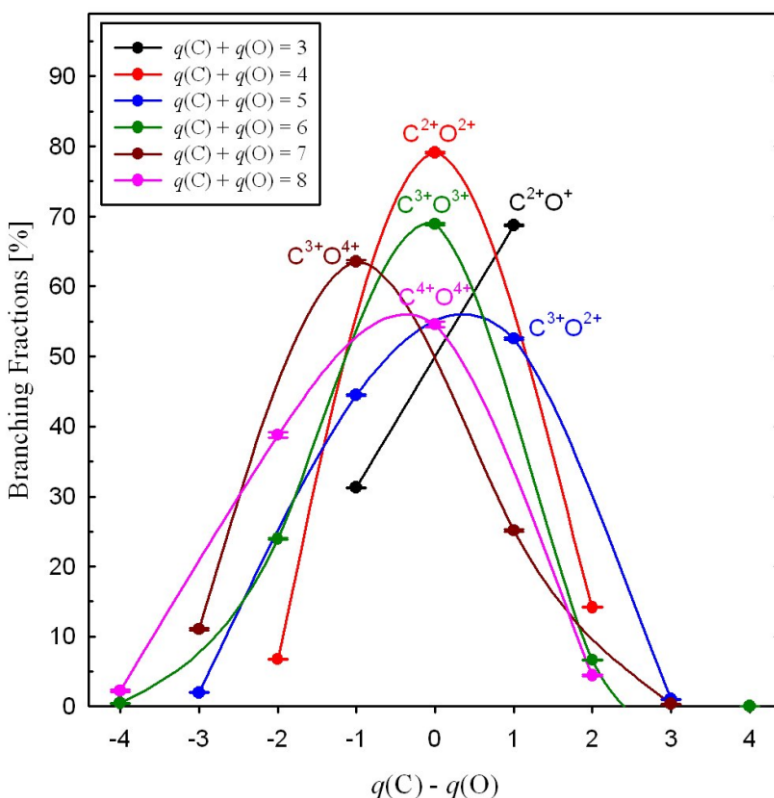


FIG. 2. Measured fractions of events in the dissociation channels originating from parent molecules having a given charge. The lines are drawn to guide the eye.

Fig. 2 shows the measured fractions of events in the dissociation channels originating from parent molecules having a given charge. It is evident that a symmetric division of charge between carbon and oxygen ions is preferred. On the other hand, for the closest dissociation channels with asymmetric charge division, it is found that carbon ions are more likely to have the higher charge when the parent molecule's charge is less than or equal to 5. The opposite is true when the parent molecule's charge is greater than 5. This effect is correlated with the total ionization energy of the molecular fragments in their final ground state. Namely, for the given combined charge of 4, 6, and 8, the total ionization energy is lowest when the fragments have equal charges. Also, the total ionization energy is lower for $C^{2+}O^+$, $C^{3+}O^+$, and $C^{3+}O^{2+}$, than for to C^+O^{2+} , C^+O^{3+} , and $C^{2+}O^{3+}$, respectively. For these combinations, the lower total ionization energy corresponds to the higher branching fraction. The same observation holds true for

some of the remaining dissociation channels ($C^{5+}O^+$ vs. C^+O^{5+} , $C^{5+}O^{2+}$ vs. $C^{2+}O^{5+}$, $C^{5+}O^{3+}$ vs. $C^{3+}O^{5+}$), but not for others ($C^{4+}O^+$ vs. C^+O^{4+} , $C^{4+}O^{2+}$ vs. $C^{2+}O^{4+}$, $C^{4+}O^{3+}$ vs. $C^{3+}O^{4+}$). The former group involves charge equal to 5 on the higher-charged fragment while the latter group involves charge 4. One possible explanation is that the latter group may be more affected by interatomic electron rearrangement in the early stages of dissociation.

The total kinetic energy release (KER) spectrum is shown in Figure 3 for the $CO^{2+} \rightarrow C^+ + O^+$ and $CO^{3+} \rightarrow C^{2+} + O^+$ dissociation channels. In the former case, the three well-resolved narrow peaks (the most prominent features in the spectrum) correspond to excitations of known metastable bound states of the CO^{2+} molecular ion [1]. The prominent narrow peak in the latter KER spectrum is likely to correspond to some metastable bound state in the CO^{3+} ion.

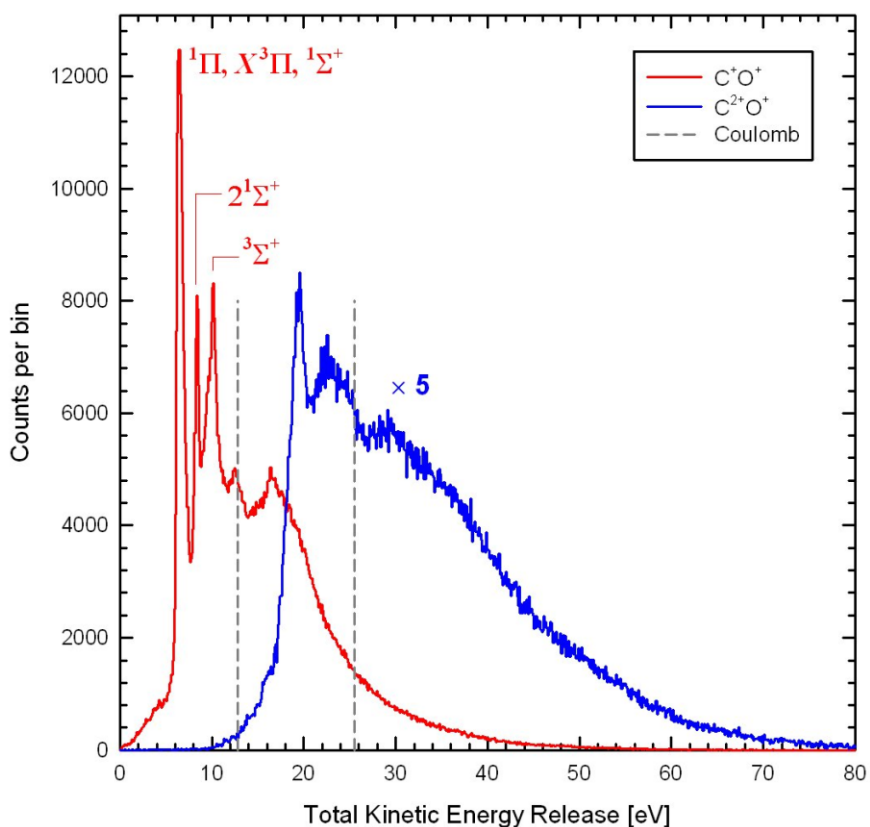


FIG. 3. Total kinetic energy release for C^+O^+ and $C^{2+}O^+$ pairs (red and blue lines, respectively). The vertical dashed gray lines indicate the KER's associated with pure Coulomb repulsion of the fragments initially separated by the CO equilibrium distance, assuming that they have non-overlapping spherical (or point-like) charge distributions.

The KER distributions for the breakup of the higher charged molecular ions, shown in Fig. 4, each display only a single broad asymmetric peak. A few of them also show additional partially

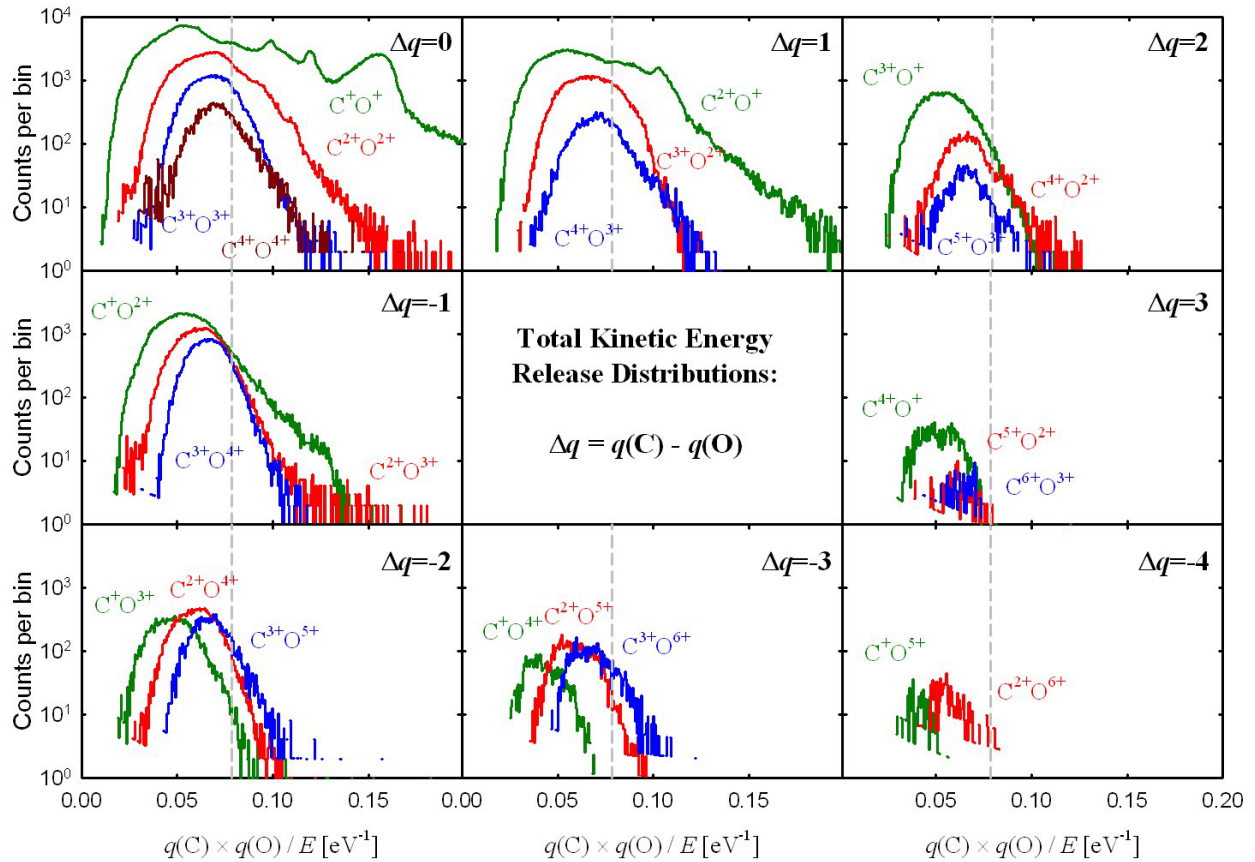


FIG. 4. Measured distributions of the total recoil-ion kinetic energy released (KER) in the breakup. The horizontal scale was chosen to condense the information to be conveyed and to provide a better match between the bin size and the counts per bin on one hand and the level of detail associated with the spectral features on the other hand. The vertical dashed gray line indicates the position of the point charge Coulomb explosion energy.

overlapping broader peaks. These broad peaks indicate the excitation of a wide range of repulsive states. It should be noted that all KER distributions were determined with a resolution of 1 eV (FWHM) or better.

The angular distributions, shown in Fig. 5, are essentially isotropic, except perhaps those for ion pairs originating from highly charged parent molecular ions, which tend to indicate a slight preference for emission perpendicular to the beam direction.

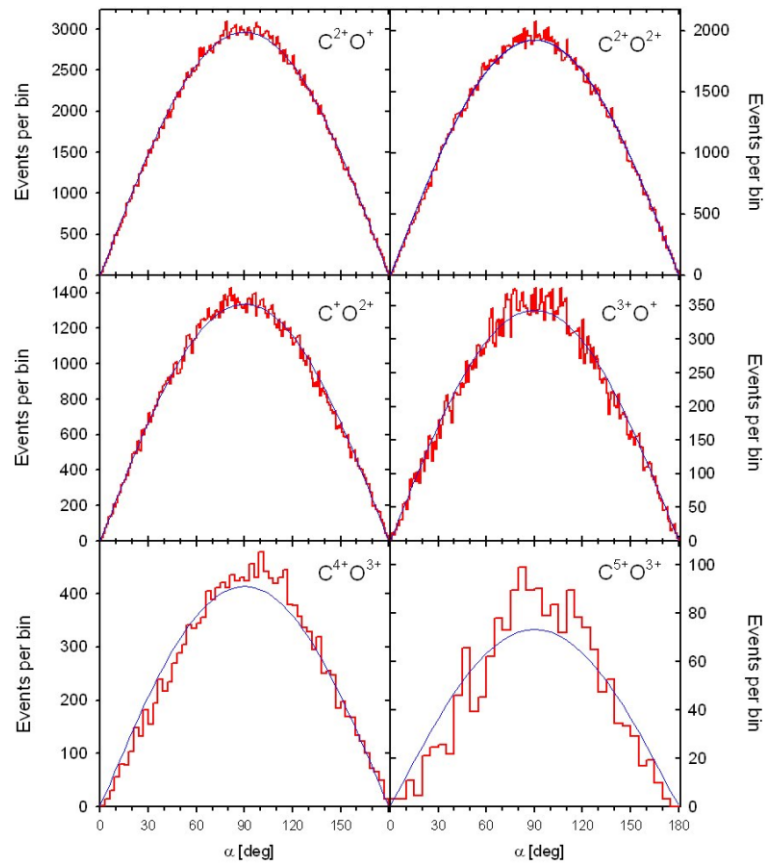


FIG. 5. Measured angular distributions for selected dissociation channels (red curve). α is the angle between the beam direction and the molecular axis at the time of collision. The thin blue line corresponds to an isotropic distribution.

- [1] M. Lundqvist, P. Baltzer, D. Edvardsson, L. Karlsson, and B. Wannberg, *Phys. Rev. Lett.* **75**, 1058 (1995).