Studying Molecular Structure and Dynamics via Coulomb Explosion Imaging with X-rays and Ultrafast Laser Pulses

U. Ablikim[*][†][1], C. Bomme[‡], E. Savelyev[‡], H. Xiong[§], R. Obaid[§], B. Kaderiya[*], S. Augustin[*], K. Schnorr[††], I. Dumitriu[¶], R. Bilodeau[†], D. Kilcoyne[†], V. Kumarappan[*], R. K. Kushawaha[*], F. Ziaee[*], T. Osipov[&], A. Rudenko[*], N. Berrah[§], and D. Rolles[*][2]

[*]James R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, KS 66506, USA
[†]Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94704, USA
[‡]Deutsches Elektronen-Synchrotron (DESY), Hamburg, 22607, Germany
[§]Department of Physics, University of Connecticut, Storrs, CT 06269, USA
[††]Department of Chemistry, University of California, Berkeley, CA 94720, USA
[¶]Hobart and William Smith Colleges, Geneva, NY 14456, USA
[&]SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA,

Synopsis Coincidence momentum imaging is a powerful method to study molecular structure and dynamics. Here, we used the Coulomb explosion imaging method together with synchrotron radiation and ultrafast laser pulses in order to distinguish between geometric isomers of $C_2H_2Br_2$, $C_2H_2Cl_2$, and difluoroiodobenzene and to investigate their fragmentation dynamics following both strong-field and inner-shell photoionization.

Isomers, i.e. molecules with the same chemical formula but different geometric structures, play an important role in many biological processes [1]. Despite containing the same atomic constituents, isomers can have very different physical, chemical, and biological properties. Therefore, it is of particular interest to experimentally distinguish isomers in order to investigate isomer-specific reactions and to study the interconversion between different isomers in time-resolved experiments.

We have conducted coincidence momentum imaging measurements on gas-phase molecular isomers, such as dibromoethane ($C_2H_2Br_2$) and dichloroethene ($C_2H_2Cl_2$), using a double-sided velocity map imaging (VMI) spectrometer at the Advanced Light Source as well as using a COLTRIMS setup and an ultrafast near-infrared laser at the J.R. Macdonald Laboratory at Kansas State University. The goal of the studies is to experimentally identify and separate *cis* and *trans* isomers using the Coulomb Explosion Imaging (CEI) method. Our results show that the geometric structure of the isomers can be distinguished by triply ionizing the molecule into the $C_2H_2^+ + Br^+/Cl^+ + Br^+/Cl^+$ fragmentation channels via inner-shell photoionization using X-ray synchrotron beams [2] or via strong-field ionization with ultrafast femtosecond laser pulses. Furthermore, numerical simulations using a classical Coulomb explosion (CE) model for both isomers match closely with the kinetic energies of the ionic fragments and the momentum correlation between their momentum vectors.

In order to further extend our CEI studies to other types of isomers and more complex molecules, we performed similar coincidence momentum imaging experiments on 2,6- and 3,5-difluoroiodobenzene (DFIB) [3]. Our experimental results and CE simulation indicate a fast charge migration along the phenyl ring after inner-shell photoionization of the iodine *4d* shell. Furthermore, we find that the majority of three-body fragmentation channels result from a sequential two-step fragmentation, where the iodine ion rapidly departs from the system after inner-shell ionization, leaving the remaining DFB dication in a rotationally excited metastable state, which subsequently fragments after a delay that is longer than its rotational period.
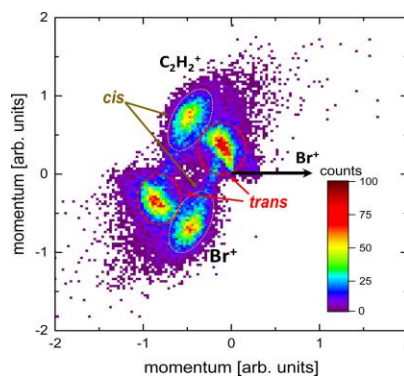
Figure 1. Newton plot of the three-body $C_2H_2^+ + Br^+ + Br^+$ coincidence channel created by inner-shell ionization of $C_2H_2Br_2$ (*cis/trans* mixture) at 140 eV photon energy.

References

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[1] E-mail: utuq@lbl.gov
[2] E-mail: rolles@phys.ksu.edu