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Electronic decay and fragmentation dynamics of iodomethane, multiply core-ionized by photoabsorption of intense XFEL pulses

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Synopsis We have studied charge migration and dissociation in iodine-substituted methane molecules using extremely intense and short 5.5 keV free electron laser pulses from the SACLA XFEL facility. Multiple core ionization down to I 2p subshells creates highly charged molecular states, the fragmentation of which was studied by ion momentum imaging multiparticle coincidence technique. We report experimental and modeling results on various dissociation pathways, fragment momentum correlations, and kinetic energy releases.

Organic and biomolecules containing substituted heavy elements become highly sensitive to absorption of tender and hard x-rays due to high cross-sections of inner-shell ionization of these elements. Such events deposit large, concentrated amounts of energy into single molecules consisting of predominantly light atoms. Deep core ionization is followed by Auger decay cascades creating multiply charged ionic states. Part of the initially absorbed energy is also spent inducing violent molecular dissociation as the electronic decay sequence progresses.

We have studied charge migration and dissociation in iodine-substituted I-methane and I-uracil molecules using very intense and short free electron laser pulses from SACLA XFEL facility in Japan. The extreme intensity of the XFEL radiation creates conditions under which the heavy element in the molecule absorbs a number of x-ray quanta before and during the electronic relaxation and dissociation process[1,2]. This multiphoton absorption thus results in highly charged molecular states, unique for studying the charge distribution and migration during the electronic relaxation and initial stages of the dissociation. Also, the dynamics of often violent multifragment Coulomb explosions can be observed.

Here we report the results of multiphoton ab-

sorption of 5.5 keV XFEL pulses by the CH₃I molecules, in which the shells as deep as 2p can be ionized. Iodomethane serves as a relatively simple system, a prototype for studying and theoretically modeling similar processes in larger biomolecules. The study was performed using an ion momentum imaging multicoincidence apparatus.

We report ion-ion coincidence measurements of iodine and carbon fragments up to the I¹⁴⁺, C⁴⁺ pairs that are created by 3-4 photon absorption. Ion momentum filtering was applied to extract the true coincidence events. The momentum filtering also allows us to extract additional information on the entire multiparticle fragmentation dynamics, since the momentum correlations reflect also the momenta of the undetected hydrogen/proton fragments. Momentum partitioning amongst the fragments reflects the dynamics of both the absorption sequences and the charge migration in the relaxation phase. Experimental results on ion kinetic energy distributions as well as energy releases in specific dissociation channels are reported as well. The experiment is compared with initial results of dissociation dynamics modeling.

References

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