X-ray induced Coulomb explosion imaging of complex molecules

Content

Recording images of individual molecules with ultrashort ”exposure times” has been a long-standing dream in molecular physics, chemistry, and biology, because this would allow one to follow the motion of atoms on their inherent timescale. While X-ray and electron diffraction have been successfully used to retrieve the structure of large biomolecules, both are very challenging to apply to small molecules in the gas phase. Moreover, these techniques are insensitive to hydrogen atoms, which are key to many chemical reactions.

We could recently demonstrate that snapshot images of the complete structure of a molecule with eleven atoms, including all hydrogens, can be recorded by Coulomb explosion imaging (CEI) when using very intense, femtosecond soft X-ray pulses from the European XFEL [1]. Up to now, it was generally expected that, in order to image polyatomic molecules via CEI, it would be necessary to record essentially all charged fragments in coincidence – a requirement that is currently impossible to fulfill for more than a few atoms within a reasonable measurement time. However, we show that, while it was possible to record up to six-fold ion coincidences in the experiment, even three-fold ion coincidences can be sufficient to image the full structure of a molecule. The X-ray intensity in the reaction microscope at the Small Quantum Systems (SQS) instrument is high enough (up to 10^{13} photons/μm^2) to produce extreme charge states in heavy atoms (e.g. up to 42+ in xenon atoms), and to Coulomb-explode molecules into individual atoms very quickly, such that the initial molecular structure is well preserved in the recorded momenta of all ions. The intriguingly clear momentum images allow us to identify each atom’s position in the molecule unambiguously.

The sensitivity of the fragmentation pattern to the molecular structure at the instant of ionization, in combination with the femtosecond pulse duration opens the door to monitoring the temporal evolution of the molecular structure with this method in a time-resolved experiment [2]. In two recent follow-up beamtimes, we could obtain first delay-dependent, three-dimensional ion momenta. Together with the prospect of even higher repetition rates at upcoming FELs, this brings the dream of recording molecular movies of photochemical reactions very close to realization.

[1] R. Boll et al., “X-ray multiphoton-induced Coulomb explosion images complex single molecules”, Nature Physics 18, 423 (2022).

[2] X. Li et al., Coulomb explosion imaging of small polyatomic molecules with ultrashort x-ray pulses, Physical Review Research 4, 013029 (2022).

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Contribution Type: Hot topic contribution