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Coulomb Explosion Imaging of Polyatomic Molecules: Towards Molecular Movies

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Recording images of individual molecules with ultrashort exposure times has been a long-standing dream in molecular physics, chemistry, and biology, and was one of the driving forces for the development of X-ray FELs. We have recently used femtosecond soft X-ray pulses from the European XFEL to record snapshot images of a molecule with 11 atoms, including all hydrogens, by Coulomb explosion imaging in a reaction microscope. Contrary to the general expectation that, to image polyatomic molecules, it is necessary to record all charged fragments in coincidence, we demonstrate that three-fold ion coincidences can be sufficient to image the full structure. The X-ray intensity of up to 10^{13}photons/μm^2 is high enough to produce extreme charge states in heavy atoms (e.g. up to 42+ in Xe atoms), and to Coulomb-explode molecules very quickly, such that the initial molecular structure is preserved in the recorded momenta. The intriguingly clear momentum images allow us to identify each atoms position in the molecule unambiguously, and to trace the rearrangement of charge within the molecule. By combining our experimental results with state-of-the-art molecular dynamics calculations, we can follow the charge-up dynamics of the molecule within the femtosecond X-ray pulse. These results can, in the next step, be employed to time-resolved imaging of molecular structure thus bringing the dream of recording molecular movies of photo-chemical reactions into reach.