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Spatiotemporal Imaging of Chemical Reactions: Making Molecular Movies with Femtosecond X-Ray Scattering J.M. BUDARZ, Brown Univ., SLAC National Accelerator Laboratory, M.P. MINITTI, SLAC National Accelerator Laboratory, A. KIRRANDER, Univ. of Edinburgh, J.B. HASTINGS, SLAC National Accelerator Laboratory, P.M. WEBER, Brown Univ. — The study of ultrafast reaction dynamics of molecular systems has benefited from the rapid development of spectroscopic and imaging techniques that follow their temporal evolution on a sub-picosecond time scale. More complete understanding of molecular behavior, however, is expected to arise from a full observation of electronic and nuclear motions during reactions. Our recent experiments at the Linac Coherent Light Source (LCLS) have allowed us to develop and implement a method wherein the ultrafast reaction dynamics of molecules in dilute gases (4 Torr) are captured by time-resolved X-ray scattering. Using a pump-probe scheme with 267 nm excitation laser and 8.3 keV X-ray probe pulses, we performed a series of measurements on the interatomic positions at variable delay times to produce a 'molecular movie.' In our experiments, 1,3-cyclohexadiene (CHD) is prepared on an excited state surface, causing the molecule to accelerate down several potential energy surfaces coupled by conical intersections, to open into 1,3,5-hexatriene within 80 fs. The resulting 'movie' has been supplemented with molecular trajectory calculations to separate the multiple pathways the excited molecule takes toward the open ring. In this talk, the experimental methods and designs that made these experiments possible will be presented together with the first results describing the photochemical reaction dynamics of CHD.

James Budarz
Brown Univ

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