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Imaging Ultrafast Dynamics in the Molecular Frame

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Time-Resolved Coincidence Imaging Spectroscopy (TRCIS) is a femtosecond photoelectron probe of Molecular Frame ultrafast dynamics in polyatomic molecules. TRCIS makes use of 3D particle timing-imaging detectors for full 3D recoil momentum vector determination of coincident photoions and photoelectrons as a function of time. One vector correlation is particularly interesting as it permits Time-Angle-Energy resolved photoelectron studies from the Molecular Frame rather than the lab frame point of view. An alternate approach to Molecular Frame ultrafast dynamics is to make use non-resonant laser field pre-alignment. Provided that the molecular dynamics are fast compared to rotational dephasing, this method also permits time-resolved Molecular Frame observations. We experimentally demonstrate both these approaches, comparing and contrasting their relative merits.