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Vibrational- and Laser-Driven Electronic Dynamics in the Molecules

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Electronic dynamics within molecules can be driven by both motions of the atoms, via non-Born-Oppenheimer coupling, and by applied laser fields, driving electron motions on sub-cycle time scales. The challenging but most general case of Molecular Dynamics is where electronic and vibrational motions are fully coupled, the making and breaking of chemical bonds being the most prominent example. Time-Resolved Coincidence Imaging Spectroscopy (TRCIS) is a ultrafast photoelectron probe of Molecular Frame dynamics in polyatomic molecules. It makes use of full 3D recoil momentum vector determination of coincident photoions and photoelectrons as a function of time, permitting observations of coupled electronic-vibrational dynamics from the Molecular Frame rather than the Lab Frame point of view. Methods in non-resonant quantum control, based on the dynamic Stark effect, have also emerged as important tools for enhancing molecular dynamics studies. In particular, molecular alignment can fix the Molecular Frame within the Lab Frame, avoiding loss of information due to orientational averaging. Provided that the molecular dynamics are fast compared to rotational dephasing, this method also permits time-resolved Molecular Frame observations. As laser fields get stronger, a sub-cycle (attosecond) physics emerges, leading to new probes of driven multi-electron dynamics in polyatomic molecules. Understanding driven multi-electron responses will be central to advancing attosecond science towards polyatomic molecules and complex systems.