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Using high-harmonics for probing real-time dynamics of highly excited molecules

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Molecular processes such as inner-valence ionization and shake-up can lead to formation of highly excited states which are close to the double ionization threshold. In order to time resolve the dynamics of such states, one needs a source of light with high photon energy and femtosecond time resolution. Laser high harmonic generation provides such a source with photon energies up to 100 eV with time durations approaching the attosecond regime. We report our recent experiments where we use high-harmonics in conjunction with coincident 3-dimensional photo-fragment momentum imaging technique to time resolve the excited state molecular dynamics. High- harmonic source is used to pump the molecule to highly-excited molecular ion state. The ensuing dynamics is observed by using a time delayed infra-red pulse to further ionize the molecular ions to doubly charged state.