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Optimizing High Harmonic Generation for Studying Excited State Dynamics[1] JAMES CRYAN, ELIO CHAMPENOIS, RAFAL RAKOWSKI, ROGER FALCONE, ALI BELKACEM, Lawrence Berkeley National Laboratory — We demonstrate the generation of ultraviolet (UV) radiation in the range of 10–70 eV via high harmonic generation (HHG) using high energy (30 mJ/pulse), high average power (30 W) drive lasers. The HHG process driven by a high energy, many cycle laser pulse (27 fs at 780 nm) produces a UV spectrum with ∼80 nJ of energy in a single harmonic. Such high photon fluence leads to saturation of single photon transitions and allows us to access non-linear dynamics in atomic and molecular systems, for instance driving two photon transitions using two different harmonic photons. Additionally we have developed experimental apparatus to isolate two different harmonic frequencies and vary the relative time delay between them to perform time-resolved pump/probe spectroscopy of excited state dynamics. Following UV excitation most molecules will undergo radiationless decay through a non-Born-Oppenheimer processes and our pump/probe technique will allow us to follow these complicated dynamics. The femtosecond duration pulses intrinsically produced by HHG allow for the necessary temporal resolution of these dynamics. I will discuss our current progress in optimizing high pulse energy HHG and show our experimental results for studying molecular dynamics using UV pulse pairs.

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