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Time-Resolved Photoelectron Spectroscopy with a Tuneable Femtosecond VUV Source RUARIDH FORBES, Stanford University, USA; University of Ottawa, Canada, SIMON P. NEVILLE, University of Ottawa, Canada, MARTIN A. B. LARSEN, University of Copenhagen, Denmark, ANDREY E. BO-GUSLAVSKIY, University of Ottawa, Canada, RUNE LAUSTEN, MICHAEL S. SCHUURMAN, NRC, Canada, ALBERT STOLOW, University of Ottawa; NRC, Canada — Time-resolved photoelectron spectroscopy (TRPES) has emerged as the gold standard technique to interrogate non-adiabatic excited state dynamics in isolated molecules but, to date, has been mostly limited by a wavelength coverage limited the UV to IR. We report the construction of a tuneable vacuum-ultraviolet (VUV) femtosecond laser source based on four-wave mixing in rare gases, capable of producing pulses from 145-180 nm. The source has been utilized in a VUV/UV TRPES investigation into Rydberg-valence coupling in the polyatomic molecule acetone. The results highlight the complex interplay between internal conversion occurring within the Rydberg manifold and population of a $\pi\pi^*$ valence state, the latter of which has not been previously experimentally observed. The results were compared to Multi-Configurational Time-Dependent Hartree (MCDTH) calculations, which verified our spectroscopic assignments. As acetone is the simplest ketene, we expect these results will have implications in the excited state dynamics for a large set of molecular systems.

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