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Multidimensional electron-nuclear wavepacket dynamics via Time-, Energy- and Angle-resolved Photoelectron Spectroscopy. K. VEYRINAS, V. MAKHIJA, A. E. BOGUSLAVSKIY, University of Ottawa, R. FORBES, University of Ottawa, University College London, I. WILKINSON, Helmholtz-Zentrum Berlin, National Research Council, Ottawa, Canada, D. MOFFATT, R. LAUSTEN, National Research Council, Ottawa, Canada, A. STOLOW, University of Ottawa, National Research Council, Ottawa, Canada — Generating and probing a coherent superposition of coupled vibrational-electronic (vibronic) states – a multidimensional wavepacket – remains a challenging problem in molecular dynamics. Here, we present recent results using time-resolved photoelectron velocity map imaging (VMI) of complex vibronic wavepacket dynamics in the NO molecule following femtosecond single photon excitation in the vacuum ultraviolet (VUV) range ($\lambda_{\text{pump}} = 160$ nm, 80 fs). The induced ultrafast dynamics, involving highly excited valence and Rydberg states, is probed by single photon ionization ($\lambda_{\text{probe}} = 400$ nm, 40 fs). Varying the pump-probe time delay, the emitted photoelectrons are detected in a VMI spectrometer for time-, energy- and angle-resolved photoelectron spectroscopy. We observe that the different final vibrational states of the NO^+ ($X^1\Sigma^+$) cation, onto which this evolving vibronic wavepacket is projected, reveal different time dependences for the kinetic energy distribution and the laboratory frame photoelectron angular distribution (LFPAD). In particular, we observe unusually strong oscillations in the β_4 asymmetry parameter, indicating sensitivity to the higher angular momentum components of the electronic aspect of this complex vibronic wavepacket.

Kevin Veyrinas
University of Ottawa

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