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State-selective Pump-probe Studies on CO₂ with Extreme Ultraviolet (XUV) and Near-infrared (NIR) Pulses¹ ANBU VENKATACHA-LAM, KANAKA RAJU PANDIRI, JAN TRO, YUBARAJ MALAKAR, SEYYED JAVAD ROBATAJAZI, SHASHANK PATHAK, ITZIK BEN-ITZHAK, ARTEM RUDENKO, DANIEL ROLLES, Kansas State Univ — State-selective excitation to a single (or a small subset of) excited neutral or ionic state(s), versus excitation to many possible states, with a broadband pulse is a powerful tool for the study and control of ultrafast molecular dynamics. We use a single-harmonic extreme ultraviolet (XUV) pulse, produced as the 11th harmonic of an 800-nm near-infrared (NIR) laser, to ionize carbon dioxide (CO₂) to the vibrationally excited ground (X $^{2}\prod_{\sigma}$) state or to the first excited (A $^2 \prod_{u}$) state of the mono-cation (CO₂⁺). Using a delaycontrolled NIR probe pulse, the mono-cation is fragmented via different pathways to yield CO⁺ or O⁺ fragments. By comparing the results to a second measurement performed with the 13th harmonic and to a similar pump-probe experiment with a comb of harmonics, where the excited ionic state is determined by photoelectron and photo-ion coincidence, we can clearly separate the role played by each ionic state and confirm the role of molecular rotation in the time-dependent ion yields.

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