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Coherent control of wavepacket launch and evolution in molecular cations in strong-field regime DMITRI ROMANOV(1,3), KATHARINE MOORE TIBBETTS(2,3), MARYAM TARAZKAR(2,3), TIMOTHY BOHIN-SKY(2,3), SPIRIDOULA MATSIKA(2,3), ROBERT LEVIS(2,3), (1) Department of Physics, (2) Department of Chemistry, and (3) Center for Advanced Photonics Research, Temple University — The time-resolved dissociative ionization dynamics for a family of acetophenone radical cations has been studied in pump-probe experiments. Modifications of the relative fragment yield have been measured as a function of the pump laser wavelength from 790 nm to 1500 nm. In the case of tunnel ionization (1150-1500 nm pump), the time-resolved transients of the parent and fragment ions probed with a weak 790 nm pulse reveal an order-of-magnitude enhancement of the peak-to-peak amplitude oscillations, ~ 100 fs longer coherence time, and an order-of-magnitude increase in the ratio of parent to fragment ions, as compared to the case of multiphoton ionization (790 nm pump). The results are quantitatively explained with a model of wavepacket evolution on the ground (D0) and excited (D1 and D2) ionic potential energy surfaces, with the probe-induced and conical-intersection-related transitions between the surfaces. The theory predicts the periods of fragment-ratio oscillations, thus confirming the ability to prepare and manipulate multiple wavepackets in the vicinity of a conical intersection for polyatomic molecules on the time scale of picoseconds.

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