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Long-lived Electronic Coherence of Rydberg States in the Strong-Field Ionization of a Polyatomic Molecule ARKAPRABHA KONAR, YINAN SHU, BENJAMIN LEVINE, VADIM LOZOVY, MARCOS DANTUS, Michigan State University — Here, we report on quantum coherent control of a large (>20 atoms) polyatomic molecule. In particular, we explore the time resolved dynamics of dicyclopentadiene when excited by a pair of phase-locked intense 800nm femtosecond pulses by monitoring changes in ion yield of the parent and fragments. Long-lived oscillations are observed for ~ 500 fs in the parent ion yield indicating the presence of long lived-electronic states. We take advantage of the long-lived electronic coherence to control the yield of different fragment ions. The presence of Rydberg states is further supported by *ab initio* calculations at the EOM-CCSD/6-31+G** level of theory which identified five low-lying electronic states of neutral DCPD in the regions between 6.4 and 7.0 eV in vertical excitation energy. States of both pure Rydberg and mixed $\pi \rightarrow \pi^*$ /Rydberg character are observed in this low energy region and are known to originate from ethylene. The multiphoton excitation of two or more Rydberg states, separated by the photon energy is the key to the observed long-lived electronic coherence in DCPD with a quantum beat at the difference frequency. Rydberg states are expected to have very similar potential energy surfaces and the Rydberg electron is relatively uncoupled to the nuclear dynamics, therefore supporting long electronic coherence time.

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