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Attosecond coherent control of C2D4 dynamics PREDRAG RANITOVIC, CRAIG HOGLE, MARGARET MURNANE, HENRY KAPTEYN, University of Colorado - JILA — We employ ultrashort VUV pulses to initiate, and coherently control ultrafast dynamics and fragmentation of a C2D4 molecule by using an IR/VUV time-resolved attosecond COLTRIMS technique. A VUV frequency comb, containing the 5^{th}, 7^{th}, 9^{th},$11^{th,}$ and 13^{th} harmonic of the fundamental laser field (785 nm), was used to coherently populate several potential energy surfaces of a neutral C2D4 molecule, and a C2D4+ ion. By ionizing the neutral C2D4* molecule in a time-resolved fashion, using a strong ($3x10^{12}$ W/cm^2) laser field, we follow the fast relaxation of several excited states through conical intersections where the electronic excitation gets converted to vibrational motion. We investigate these dynamics on femtosecond and attosecond time scales. On the femtosecond time scale, we measure the decay constants of the fragments of interest (i.e. C2D4+, C2D3+, C2D2+, D+, and D2+), and find that the dynamics occur within the first 50 fs after the VUV pump pulse. On the attosecond time scale, we find that we can control the fragmentation through interference of electron wave packets and by changing the laser intensities. We find that the absolute phases of the fragmentation yields are sensitive to the VUV/IR delay, and change as the molecule relaxes through conical intersections. The relaxation through conical intersections is a complex and important mechanism that we studied using an electron/ion 3D momentum imaging COLTRIMS technique. The ability to demonstrate coherent control of this relaxation process on an ultrafast time scale, is an important step towards control of chemical reactions.

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