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Interpreting Closed-Loop Learning Control of MolecularFragmentation in Members of the Ketone Family of Molecules MARK BAERTSCHY, University of Colorado at Denver, DAVID CARDOZA, Stony Brook University, THOMAS WEINACHT, Stony Brook University — We present the results of closed-loop learning control over molecular fragmentation in the ketone family, $CH_nX_mCOCY_3$, with X being Br or H; Y being H, D, F or Cl; and with n + m = 3. The experiments used shaped ultrafast laser pulses and a Genetic Algorithm controlling a pulse shaper. Pump-probe measurements were also carried out to supplement the learning control results. To interpret our learning control and pump-probe results, we used a combination of *ab initio* molecular structure calculations, modifications to the learning algorithm, and quasi-static molecular ionization calculations. For CH_3COCF_3 we discovered that the control mechanism is based on a subtle interplay between ionization enhancement and the details of the dissociation dynamics. We have since generalized these ideas to gain insight into the fragmentation control mechanisms for a broad class of molecules.

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