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Interpreting Learning Control of Molecular Fragmentation in Terms of Enhanced Ionization MARK BAERTSCHY, University of Colorado at Denver, DAVID CARDOZA, Stony Brook University, THOMAS WEINACHT, Stony Brook University — We interpret a molecular learning control experiment aimed at optimizing the CF_3^+/CH_3^+ ratio during dissociative ionization of trifluoroacetone, CH_3COCF_3. The experiment makes use of a learning algorithm and shaped ultrafast laser pulses, and our interpretation of the control is in terms of enhanced molecular ionization during dissociation. Our understanding of the physical control mechanism is built upon necessary features in optimal pulse shapes discovered by the learning algorithm, *Ab initio* molecular structure calculations, pump-probe measurements of the molecular fragmentation, and quasi-static tunnel ionization calculations. This control mechanism is quite general, motivating continued studies of a range of molecules in the ketone family. It may also provide for new measurements of molecular relaxation and dissociation rates.

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