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Adaptive Control Goal Selection for Strong-Field Dissociative Ionization of Polyatomic Molecules DMITRI ROMANOV (1,3), HUYEN TRAN (2,3), ROBERT LEVIS (2,3), (1) Department of Physics, (2) Department of Chemistry, and (3) Center for Advanced Photonics Research, Temple University, Philadelphia PA — In many settings (for instance, in strong-field mass-spectral sensing technologies) improving control efficiency is more important than achieving specific control goals. In this case, control goals may be adaptively formulated in the process of a strong-field experiment. To determine the pairs of fragment ions in a mass spectrum that are most susceptible to control by adaptive optimization of the laser pulse shapes in the strong-field regime, a statistical method is proposed that is based on covariance analysis of the mass spectral fragmentation patterns generated by a set of random shaped pulses. As a test, the method was applied to fragmentation of a large organic molecule dimethylmethylphosphonate, (CH_3O)-PO-(OCH_3)-(CH_3). All possible pairs of the ionized fragments in *tof* mass spectrum were ranked by the value of their correlation coefficients ranging from +1 to –1. A genetic-algorithm based adaptive control was then used to optimize the ion peak ratios in these pairs. Convincingly, the pairs of fragment ions that have higher negative covariances possess a correspondingly higher degree of controllability, while the pairs that have higher positive covariances possess correspondingly lower controllability.

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