Dynamics of Molecular Fragmentation Mediated by Charge-Transfer States ROBERT LEVIS(1,*), DMITRI ROMANOVOV(2,*), ALEXEI MARKEVITCH(1,*), (1) Department of Chemistry, (2) Department of Physics, (*) Center for Advanced Photonic Research, Temple University, STANLEY SMITH, Department of Chemistry, Wayne State University, DUSAN LORENCZ, DUSAN VELIC, Comenius University, Bratislava, Slovakia — Dissociative ionization of large organic molecules caused by ultrashort strong-field laser pulses is largely predeter-
mined by nonadiabatic electron dynamics during the pulse. The key element of the nonadiabatic process is the bottleneck transition from the systems ground state to the charge-transfer doorway state of the excited-state manifold. The induced charge transfer across large distances in polyatomic molecules and ions evolves into a complicated dynamics that can include prolong charge localization and sequential ion-
ization. This electron-charge dynamics affects essentially the ensuing nuclear motion and thus determines the fragmentation pattern and charge distribution among the fragments. We observed manifestations of nonadiabatic electron-nuclear dynamics mediated by charge-transfer states in a series of experiments on related polyaromatic molecules, including study of the fragmentation threshold as a function of the laser intensity, the proton kinetic energy distributions, and the relative yield of ionized fragments as a function of the carrier wavelength. In all these cases our model calculations agree quantitatively with the experimental data.

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