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Phase control of molecular fragmentation with a pair of femtosecond-laser pulses[1] KARL-MICHAEL WEITZEL, GEORG BREUNIG, GUNTER URBASCH, Philipps Universitaet Marburg — We demonstrate the control of molecular fragmentation on a femtosecond-time scale in two-pulse measurements with a pair of femtosecond-laser pulses. The measurements were performed with o-xylene (C_8H_{10}). Parent and fragment-ion yields were recorded as a function of inter-pulse delays, i.e. different relative phases of the excitation pulses. The experiments revealed different fragmentation mechanisms in the temporal region of direct optical overlap and for separated pulses. For overlapping pulses all ion yields followed the excitation intensity which oscillated as a function of inter-pulse delay due to the change of constructive and destructive interference. For larger delays, in particular the oscillations of the C^+ and CH_3^+ fragment-ion yield showed a significant deviation from each other. This deviation vanished in measurements with chirped femtosecond-laser pulses where both parent and fragment-ion yields oscillated in phase for all investigated delays. The results are interpreted as a manifestation of optical phase-dependent electronic excitations mapped onto the nuclear fragmentation dynamics.

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Karl-Michael Weitzel
Philipps Universitaet Marburg

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