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Direct time-resolved observation of molecular dynamics induced by extreme-ultraviolet photoionization ARVINDER SANDHU, ETIENNE GAGNON, ARIEL PAUL, MARGARET MURNANE, HENRY KAPTEYN, JILA, University of Colorado and NIST, Boulder, Colorado 80309-0440, USA, PREDRAG RANITOVIC, C. LEWIS COCKE, J. R. Macdonald Laboratory, Kansas State University, Manhattan, Kansas 66506-2601, USA — Laser-generated high-order harmonics provide a source of extreme-ultraviolet radiation with unique capabilities for probing atomic and molecular dynamics. Here we present the first such studies by employing high harmonics in conjunction with coincident electron-ion 3D momentum imaging (COLTRIMS) technique. We generate femtosecond EUV pulses at ~ 42 eV photon energy by upconverting intense ($> 10^{14}$ Wcm $^{-2}$) 25 fs laser pulses in an argon filled waveguide. Using this ultrashort EUV pulse as a pump, we launch D₂, N₂ and CO into highly excited states near the molecular double-ionization threshold. The dynamics of these highly excited states unfold along different channels, which are identified by electron-ion correlation. By employing moderate intensity infrared fields, we show that we can influence and probe these dynamics on femtosecond timescales. We observe that the double ionization yield is significantly enhanced by the presence of an IR field in conjunction with the EUV pump. The kinetic energy release evolves as a function of delay time (internuclear distance), allowing us to map excited state dynamics.

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