Abstract Submitted
for the MAR07 Meeting of
The American Physical Society

Using high-order harmonics with momentum imaging techniques to study atomic and molecular dynamics ARVINDER SANDHU, ETIENNE GAGNON, ARIEL PAUL, MARGARET MURNANE, HENRY KAPTEYN, JILA, University of Colorado and NIST, Boulder, Colorado 80309-0440 — 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 studies that employ high harmonics in conjunction with coincidence momentum imaging (COLTRIMS) techniques for studies of molecular dynamics. We generate pulse at ~ 43 eV photon energy by upconverting intense ($> 10^{14}\mathrm{Wcm}^{-2}$) 25 fs laser pulses in an argon filled waveguide. These photons illuminate a cold molecular beam of CO, CO_2 or N_2, with the ion and electron products from ionization/dissociation detected using time-and-position resolved detectors. We obtain count rates as high as 0.25 per harmonic pulse, sufficient for a variety of studies. By employing pump-probe techniques, we can launch molecules into highly excited states near the molecular double-ionization threshold, from where the dynamics unfold along different channels. We also employ field-free impulsive molecular alignment to demonstrate for the first time the use of single photon excitation to obtain ion and electron angular distributions in the lab frame.

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Date submitted: 20 Nov 2006 Electronic form version 1.4