

Abstract Submitted
for the DAMOP06 Meeting of
The American Physical Society

Observation of Intra-molecular Vibrational Dynamics using High-Harmonic Generation as a Probe NICK WAGNER, ANDREA WÜEST, MARGARET MURNANE, HENRY KAPTEYN, University of Colorado — Here we report one of the first observations of intramolecular dynamics using electrons rescattered during the process of high-order harmonic generation. We excite coherent vibrations in SF₆ using impulsive Raman scattering with a short laser pulse. A second, more-intense laser pulse generates high-order harmonics of the fundamental laser, at wavelengths of $\sim 20\text{-}50$ nm. The high-order harmonic yield is observed to oscillate, at frequencies corresponding to all the Raman-active modes of SF₆, with an asymmetric breathing mode most visible. This is in contrast to conventional impulsive stimulated Raman spectroscopy where only the symmetric breathing mode of the molecule is easily observed. The data also show evidence of relaxation dynamics following impulsive excitation of the molecule. Our results indicate that high harmonic generation is a sensitive probe of vibrational dynamics and may yield more information simultaneously than conventional ultrafast spectroscopic techniques. Since the de Broglie wavelength of the recolliding electron is on the order of interatomic distances, i.e. ~ 1.5 Å, small changes in the shape of the molecule lead to large changes in the high harmonic yield.

Nick Wagner
University of Colorado

Date submitted: 02 Feb 2006

Electronic form version 1.4