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Observation of Intra-molecular Dynamics using High-Harmonic Generation as a Probe NICK WAGNER, XIBIN ZHOU, WEN LI, ROBYNNE HOOPER, MARGARET MURNANE, HENRY KAPTEYN, JILA/University of Colorado — We report two observations of intramolecular dynamics using electrons rescattered during the process of high-order harmonic generation. In the first experiment, we excite coherent vibrations in SF₆ using impulsive Raman scattering. A second, more-intense pulse generates high-order harmonics from the excited molecules, at wavelengths of 20-50nm. The 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 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 harmonic generation is a very sensitive probe of vibrational dynamics, yielding more information simultaneously than conventional ultrafast spectroscopies. In our second experiment, we dissociate CF₃I with a 266nm pulse, and monitor the dissociation by probing high harmonic emission from the intact and dissociated molecule. Since the de Broglie wavelength of the recolliding electron is on the order of interatomic distances ($\sim 1.5\text{\AA}$), small changes in the shape of the molecule lead to large changes in the high harmonic yield.

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