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Monitoring Molecular Dynamics using Coherent Electrons from High-Harmonic Generation NICK WAGNER, ANDREA WÜEST, IVAN CHRISTOV, TENIO POPMINTCHEV, XIBIN ZHOU, MARGARET MURNANE, HENRY KAPTEYN, University of Colorado and National Institute of Standards and Technology — In this talk, we will discuss the first observation of intramolecular vibrational dynamics using electrons rescattered during the process of high-order harmonic generation. We excite coherent vibrations in SF_6 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 $\sim$ 20-50 nm. The high-order harmonic yield is observed to oscillate, at frequencies corresponding to all the Raman-active modes of SF_6, with an asymmetric 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. $\sim$ 1.5 Å, small changes in the shape of the molecule lead to large changes in the high harmonic yield. This work therefore demonstrates a new spectroscopic technique for probing ultrafast internal dynamics in molecules that uniquely combines ultrafast time resolution with atomic-scale structural information.

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