

Abstract Submitted  
for the DAMOP07 Meeting of  
The American Physical Society

**Intramolecular Dynamics Probed using High Harmonic Generation** ROBYNNE HOOPER, XIBIN ZHOU, WEN LI, NICK WAGNER, HENRY KAPTEYN, MARGARET MURNANE, JILA — We observed intramolecular dynamics as a modulation in high harmonic emission. We excite coherent vibrations in  $\text{CF}_3\text{Cl}$  using impulsive Raman scattering with a short laser pulse. A second laser pulse generates high harmonics. The harmonic yield is observed to oscillate at frequencies corresponding to three vibrational modes of  $\text{CF}_3\text{Cl}$ . In a second experiment, we used UV light to excite and dissociate  $\text{CF}_3\text{I}$ , and follow the dynamic evolution by monitoring the harmonic yield. We observe a large modulation of the harmonic yield, likely due to resonance excitation and subsequent dissociation of the molecule. We speculate that the less-than full baseline recovery after the UV pulse is due to ionization, and that the harmonic yield is sensitive to the bond length during dissociation. By these two experiments, we confirm that high harmonic generation is a sensitive probe of intramolecular dynamics and may yield more information simultaneously than conventional ultrafast spectroscopic techniques.

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Date submitted: 02 Feb 2007

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