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Vibrational Quantum Beats and High Harmonic Generation in SF_6[1] ZACHARY B. WALTERS, Department of Physics and JILA, University of Colorado, Boulder CO 80309, STEFANO TONZANI, Northwestern University Chemistry Dept, Evanston IL 60208, CHRIS H. GREENE, Department of Physics and JILA, University of Colorado, Boulder CO 80309 — Although HHG is commonly understood as an electronic process, vibrational degrees of freedom in molecules allow for phenomena which have no analogue in atomic systems. This was recently demonstrated in experiments performed with SF_6 (Wagner et al, PNAS **103** 13279, 2006). If a HHG laser pulse is preceded by a weaker pulse which stimulates Raman-active vibrations, the harmonic intensity oscillates with the interpulse delay time at the frequencies of the stimulated modes. We explain this modulation as quantum interference between adjacent vibrational states of the molecule, which are mixed during the high harmonic process. We present an improved version of the three-step model, which uses nonperturbative electron-ion scattering wavefunctions to find the recombination dipole, and which tracks the vibrational wavefunction of the molecule throughout the high harmonic process.

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