Vibrational Modulation of High Harmonic Generation in SF_6

Zachary B. Walters, Stefano Tonzani, and Chris H. Greene

Department of Physics and JILA, University of Colorado, Boulder, Colorado 80309-0440, USA

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A recent experiment performed at JILA (N. Wagner et al, unpublished) has demonstrated that the vibrational (nuclear) degrees of freedom of a molecule play a role in the electronic process of high-order harmonic generation. In the experiment, the molecule is hit by two pulses: the first stimulates Raman-active vibrations while the second generates high harmonics. The intensity of the generated harmonics oscillates as a function of delay time between the two pulses, with oscillation frequencies equal to those of the Raman-active modes.

We propose a novel mechanism which explains this oscillatory behavior as a form of quantum interference between neighboring vibrational states of the molecule.

In the conventional three-step model of high harmonic generation, an electron first tunnels free from an atom or molecule, then propagates in the ponderomotive potential of the applied laser field and finally recombines with the original atom or molecule.

In a molecule, the amplitudes for ionization and recombination can vary strongly with respect to distortions of the nuclear coordinates away from the equilibrium configuration. These derivatives with respect to nuclear coordinates mean that the molecule has some amplitude to switch from a state with vibrational quantum number ν in some normal mode to a state with vibrational quantum number $\nu \pm 1$ during the ionization and recombination processes. The resulting interference between neighboring vibrational states induces a modulation of the high harmonic signal.

This poster presents the theory of high harmonic modulation by means of Raman-excited quantum beats and compares the resulting theoretical prediction to the experimental results of Wagner *et al.*

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