

Abstract Submitted
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High-order harmonic generation from a coherent superposition of atomic and molecular states SZCZEPAN CHELKOWSKI, Département de chimie, Université de Sherbrooke, Sherbrooke, Qc, J1K 2R1, Canada, TIMM BREDTMANN, Institut für Chemie und Biochemie, Physikalische und Theoretische Chemie, Freie Universität Berlin, Takustraße 3, 14195 Berlin, Germany, ANDRÉ BANDRAUK, Département de chimie, Université de Sherbrooke, Sherbrooke, Qc, J1K 2R1, Canada — In our previous work [1] we studied numerically pump-probe schemes for monitoring electron motion in molecules and in atoms using a mid-infrared intense few femtosecond probe laser pulse which generate high-order harmonics (HHG) from a coherent superposition of electronic states prepared by a weak femtosecond UV pump pulse from an initial bound state. We showed that contour graphs of HHG signal as function of time delay and harmonic order show striking regularities. In this work, by studying HHG in simpler systems we identify even more clear interference patterns in HHG as function of the pump-probe delay and explain these regularities using an extended three-step model based on quantum trajectories. In particular, by varying the time delay between two pulses one can shift (up or down) the harmonic frequency in a continuous way. We explain this continuous frequency red-shift as a result of interference between a long trajectory originating from the lower state (and returning to the same state) and a short trajectory originating from the upper state (and returning to the lower state).

[1] T. Bredtmann, S. Chelkowski, A.D. Bandrauk, *J. Phys. Chem.* **116**, 11398 (2012).

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