Three-Step Model for High-Harmonic Generation in Many-Electron Systems

ROBIN SANTRA, Argonne National Laboratory, Argonne, IL 60439, ARIEL GORDON, Research Laboratory of Electronics, Massachusetts Institute of Technology, 77 Massachusetts Ave., Cambridge, MA 02139 — The three-step model (TSM) of high-harmonic generation (HHG) is generalized to atomic and molecular many-electron systems. Using many-body perturbation theory, corrections to the standard TSM due to exchange and electron–electron correlations are derived. It is shown that canonical Hartree-Fock orbitals represent the most appropriate set of one-electron states for calculating the HHG spectrum. To zeroth order in many-body perturbation theory, a HHG experiment allows direct access in general to a combination of occupied Hartree-Fock orbitals rather than to the highest occupied molecular orbital by itself.

1This work was supported in part by the Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy, under Contract No. W-31-109-ENG-38. Partial support by AFOSR under grant FA9550-04-1-0011 is gratefully acknowledged.