Electron-Nuclear Dynamics in time-dependent atomic and molecular processes: From charge transfer to molecular fragmentation

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The treatment of the dynamics of electrons and nuclei in time-dependent processes is of fundamental importance to the understanding of matter-interaction in quantum physics and chemistry. By using the time‑dependent variational principle and a parametrization of the wave function in a coherent-state manifold for electrons and nuclei, a system of coupled, first‑order, nonlinear differential equations is obtained. The equations resemble those of a Hamiltonian system within a generalized phase space that allows a systematic time‑dependent study of atomic and molecular processes from attosecond to picosecond time span. The approach is general and provides a computational framework for a variety of properties such as electronic transitions, excitation probabilities, and kinetic energy releases of interest in atomic and molecular physics. The simplest approximation we have implemented corresponds to the choice of a single determinantal wave function for the electrons moving in a classical nuclei description. Illustrative applications to atomic and molecular processes are presented with good to excellent agreement to other theoretical and experimental data. Specific examples are provided for charge transfer, total and differential cross sections, molecular fragmentation, with some examples towards femtosecond laser-assisted charge transfer, as well as for the energy loss and stopping cross section of interest in material damage.