Quantum dynamics with wavepackets and density matrices: A novel computational tool with applications to biological enzymes.[1] SRINIVASAN IYENGAR, Indiana University — A recently developed computational approach for simultaneous dynamics of electrons and nuclei is discussed. The approach is based on a synergy between quantum wavepacket dynamics and ab initio molecular dynamics. The quantum dynamics is performed using an efficient banded, sparse and Toeplitz representation for the discretized free propagator that is formally exact. Ab initio molecular dynamics is achieved by using (a) an extended Lagrangian formalism, known as atom-centered density matrix propagation, that effects an adjustment of time-scales of the electronic motion, (b) Born-Oppenheimer dynamics. The quantum dynamics and ab initio dynamics schemes are coupled through a time-dependent self consistent field-like procedure. Higher order coupling between the subsystems is inherent when the Born-Oppenheimer procedure is used as opposed to atom-centered density-matrix propagation. A fundamental computational bottleneck associated with the computation of the interaction potential between the ab initio and quantum dynamical subsystem are overcome through a novel importance sampling approach and this aspect is also discussed. Further generalization for periodic quantum dynamical treatment in extended systems is outlined.

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Date submitted: 02 Dec 2005   Electronic form version 1.4