The unending END: Expanding the END theory in honor of N. Yngve Öhrn

Ajith Perera[a] *and Jorge A. Morales*[b]

[a]*Department of Chemistry, Quantum Theory Project, University of Florida, Gainesville, FL 32611,* [b]*Department of Chemistry and Biochemistry, Texas Tech University, Lubbock, TX 79409.*

The electron nuclear dynamics (END)[1-2] theory is one of the best contributions by Prof. N. Yngve Öhrn in the field of theoretical chemistry. In this lecture, we would like to honor Prof. Öhrn on the occasion of his 80th birthday by presenting some of our recent developments in the END theory. END is a *time-dependent*, *variational*, *non-adiabatic* and *direct* method to simulate chemical reactions. END dynamical equations are obtained through the time-dependent variational principle applied to trial total wavefunctions in the coherent-state (CS) representation. Unlike Ehrenfest methods, END contains nucleus-electron non-adiabatic coupling terms. Unlike Born-Oppenheimer dynamics methods, END is not confined to the electronic ground state. Instead, it properly describes electron excitations and transfers. Simplest-level END (SLEND) describes nuclei classically and electrons with a single-determinantal wavefunction. Within SLEND, three interrelated novel developments will be presented[2-3]: **(1)** The development of different types of CSs[2] to generate quantum/classical connections for various degrees of freedom: rotational, vibrational (both harmonic and anharmonic), and electronic. Rotational and vibrational CSs reconstruct quantum excitation probabilities from the SLEND nuclear classical dynamics. The electronic Thouless CS provides an appropriate parameterization for the SLEND electronic wavefunction, while a suggested electronic valence-bond-theory CS generates a charge-equilibration model based on Sanderson's principle of electronegativity equalization. **(2)** The new SLEND/Kohn-Sham-density-functional-theory method[2-3]. And **(3)** our cutting-edge parallel code *PACE* (Python Accelerated Coherent-states Electron nuclear dynamics)[2] implementing the described models. These models successfully simulate various chemical reactions, including high-energy ion-molecule collisions, and S_N2 and Diels-Alder reactions. However, the more prominent applications of our models are to *proton cancer therapy* reactions, such as water radiolysis and damage processes on DNA components, *inter alia*.

[1] E. Deumens, A. Diz, R. Longo, and Y. Öhrn, *Reviews of Modern Physics* **66**, 917 (1994).
[2] C. Stopera, T. V. Grimes, P. M. McLaurin, A. Privett, and J. A. Morales, in *Advances in Quantum Chemistry*, Vol. **66**, Chapter 3, 113 (2013)
[3] S. A. Perera, P. M. McLaurin, T. V. Grimes, and J. A. Morales, *Chemical Physics Letters*, **496**, 188-195 (2010).