Applying Direct Quantum Dynamics to Photo-Excited Processes: Proton Transfer and Charge Migration

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In order to understand photo-excited processes it has been proven to be important to describe significant quantum effects such as tunnelling and conical intersections. A method of direct quantum dynamics for the study of such photo-excited processes has recently been implemented in the Quantics software package. The DD-vMCG (direct dynamics by the variational multi-configurational Gaussian) method promises to be fast, efficient, and with better convergence properties than previous methods based on classical trajectories [1]. As it is in the development phase, testing is required.

The underlying physical processes by which mutagenesis occurs in DNA/RNA, whether by UV radiation or spontaneous occurrence, has been the subject of extensive chemical and biological research. Prototypically the inter- and intra-molecular proton-transfers in excited states of 2-pyridone/2-hydroxypyridine and Formamide/Formamidic acid have been used as analogues to the description of mutagenesis. They present an interesting system to study with the DD-vMCG method as they may demonstrate competition between proton-transfer and non-adiabatic curve crossing [2,3].

Another photo-excited process of interest is the competition between charge-transfer and charge-migration. A new direct dynamics method, based on the Ehrenfest approach, has been used previously to study charge-migration in the Toluene cation [4]. A model Hamiltonian for the Allene (C_3H_4) radical cation has been used as a test in previous Quantics studies on charge-transfer [5], and presents an ideal system to test the Ehrenfest approach.

References

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