Quantum dynamics with the Multi-Configuration Time-Dependent Hartree (MCTDH) method: applications to the Infrared spectroscopy of protonated water clusters and to biological systems

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Many molecular processes, ranging from fundamental to applied problems, are known today to be impacted by strong nuclear quantum mechanical effects, including phenomena like tunneling, zero point energy effects, or non-adiabatic transitions. Recent success in helping to understand experimental observations in fields like heterogeneous catalysis, photochemistry, reactive scattering, optical spectroscopy, or femto- and attosecond chemistry and spectroscopy underlines that nuclear quantum mechanical effects affect many areas of chemical and physical research. The correct theory to describe the corresponding dynamics is Molecular Quantum Dynamics [1,2]. In contrast to standard quantum chemistry calculations, where the nuclei are treated classically, molecular quantum dynamics can cover quantum mechanical effects in their motion. New strategies have been developed to extend the studies to systems of increasing size. In particular, we present here several applications of the Multi-Configuration Time-Dependent Hartree method (MCTDH) [3,4,5] to the understanding and the control of molecular processes involving quantum effects. MCTDH can be seen as a time-dependent MCSCF approach for the nuclei where wavepackets are propagated on one or several potential energy surfaces. Several examples will be presented highlighting the presence and the exploitation of quantum effects in molecular processes. Special emphasis will be placed on the spectroscopy in full dimensionality of protonated water : Zundel cation (15 D) and Eigen cation (33 D). We will sketch possibilities of future studies of water clusters such as water dimer and trimer. We will also illustrate the possibility to treat very large systems with the Multi-Layer MCTDH approach with one biological system.

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