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New results for quantum control problems

Optimal control of light-induced processes in complex molecular systems

Jens Petersen

Department of Physical and Theoretical Chemistry, University of Würzburg
Am Hubland, 97074 Würzburg, Germany

jens.petersen@uni-wuerzburg.de

Optimally shaped electric fields can be used to control a large variety of dynamical phenomena in molecules, such as isomerization, internal conversion, electron and proton transfer, or fragmentation. The simulation of such processes requires an atomistic description of the coupled electron-nuclear dynamics including the interaction with the control field. For this purpose, we have developed the field-induced surface-hopping method (FISH) [1], which is based on a combination of classical nuclear dynamics with quantum mechanical electronic population dynamics. This allows for an efficient treatment of complex molecular systems including all degrees of freedom. The electric fields used for control are incorporated straightforwardly without further approximations, thus enabling the use of arbitrarily complex pulse shapes. We will present the validation of our approach against numerically exact quantum dynamics simulations for the strong-field control of electronic state populations in the K_2 molecule [2]. The broad scope of the FISH method for exploring control strategies in complex systems and unravelling the underlying mechanisms will then be illustrated on examples of control in the condensed phase. It will be shown how FISH simulations employing experimentally shaped fields reveal the mechanism responsible for optimal dynamic discrimination between two spectroscopically almost identical biomolecules. Moreover, the theoretical design of laser pulses capable to extend the excited state lifetime of the DNA base adenine will be presented [4]. Finally, in a digression into nanooptics it will be demonstrated how a combination of quantum chemistry with classical electrodynamics allows one to simulate the optimal control of spatio-temporal field localization in a molecular-sized nanostructure [5].

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