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Implementation of exact and approximate methods for nonadiabatic quantum molecular dynamics induced by the interaction with the electromagnetic field and their applications to local quantum control AURÉLIEN PATOZ, JIRI VANICEK, EPFL - Lausanne — we have implemented a general split-operator/Magnus integrator algorithm of arbitrary order in accuracy for exact nonadiabatic quantum dynamics of a molecule interacting with a time-dependent electromagnetic field. Then, we have derived and implemented analogous geometric integrators of arbitrary order of accuracy for several approximations of treating the molecule-field interaction: the time-dependent perturbation theory, separation of time scales, Condon, rotating-wave, and ultrashort, "extreme ultrashort," and "extremely extreme ultrashort" pulse approximations. Our general and efficient implementation permits every possible combination of these basic approximations, allowing testing the validity of each approximation under the experimental conditions independently. In addition, a local quantum control scheme has been implemented in the same formalism allowing using the exact method and several of our approximations. The algorithms are applied to the four-dimensional vibronic coupling model of pyrazine in order to compare the exact and approximate descriptions of the photoexcitation process with a single laser pulse of finite length as well as nonadiabatic quantum dynamics induced by pump and probe laser pulses.

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