Towards in Silico Photochemical Experiments using Ab Initio Nonadiabatic Molecular Dynamics

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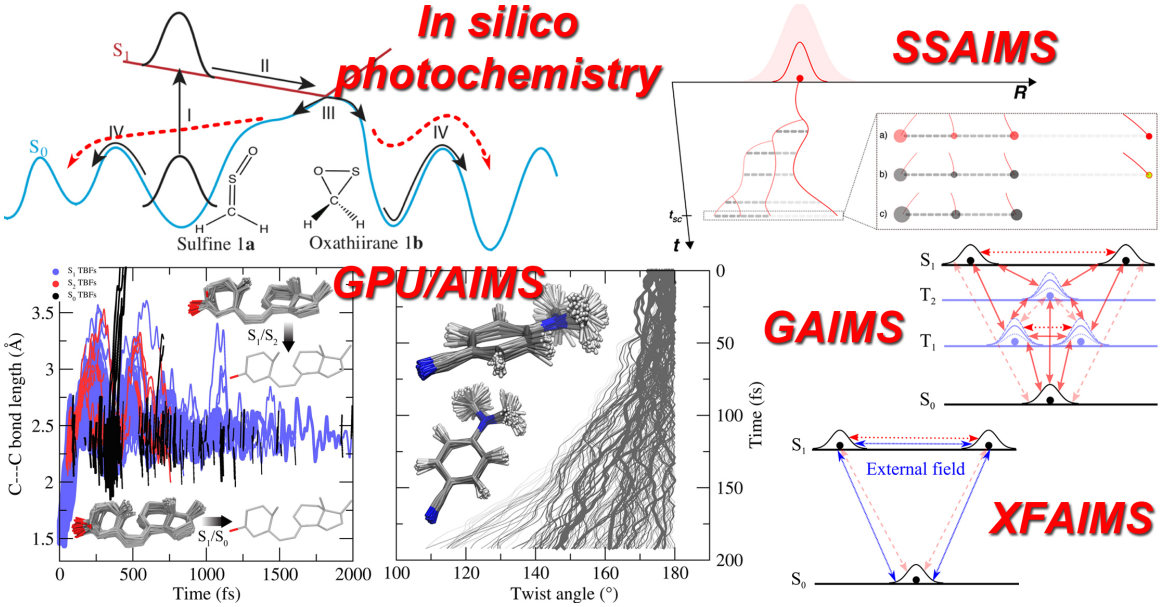
Ab initio multiple spawning (AIMS) is a theoretical tool that aims at an accurate yet efficient *in silico* description of photochemical and photophysical processes in molecules. AIMS describes the nonadiabatic dynamics of nuclear wavepackets by means of linear combinations of frozen Gaussians, whose number can be adapted when required. In this talk, I intend to present a survey of some recent developments and applications of the AIMS technique.

To study the excited-state dynamics of larger molecules, we recently interfaced AIMS with the GPU-based electronic structure code TeraChem. Combining the accuracy of AIMS with the efficiency of GPU-accelerated electronic structure calculations (LR-TDDFT or SA-CASSCF) allows indeed for a significant step forward in the simulation of nonadiabatic events, as it pushes the boundaries of the well-known compromise between efficiency and accuracy imposed by the computational cost of such dynamics. Thanks to this new interface, we could investigate the nonadiabatic dynamics of different medium-size organic molecules important in biological chemistry, organic electronics, and atmospheric chemistry.[1–3]

We also recently introduced two new theoretical developments: (*i*) a generalization of AIMS to the description of both internal conversion and intersystem crossing processes[4] and (*ii*) the inclusion of an external electromagnetic field to simulate photoexcitation processes.[5]



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