Non-adiabatic Semiclassical Simulations: Focusing on What Really Matters

Petr Slavíček*[1]

[1]*University of Chemistry and Technology, Prague, Technická 5, Prague 6, 16628*
e-mail: petr.slavicek@vscht.cz

Computational photodynamics in the visible and UV domain is gradually becoming a mature field, allowing for almost routine simulations of ultrafast photochemical processes taking place on tens to hundreds of femtoseconds. At the same time, the simulations are often disconnected from the need of computational practitioners and experimental chemists. In my presentation, I will concentrate on pragmatic approaches to photodynamics, allowing for stable simulations of larger systems and for longer timescales. In particular, I will discuss techniques of non-adiabatic dynamics [1] and the available toolbox of electronic structure theory. Attention will be paid to the question of initial condition sampling.[2,3] I will also briefly discuss photochemical simulations in the liquid phase. The computational techniques will be demonstrated on recent studies from our laboratory, e.g. photochemistry of bilirubin subunit taking place on femtosecond to picosecond time scale [4] and simulations of the birth of solvated electron upon radiolysis.[5]

[1] J. Suchan, J. Janoš, P. Slavíček, J. Chem. Theory Comput. 16, 9, 5809–5820.

[2] Š. Sršeň, J. Sita, P. Slavíček, V. Ladányi, D. Heger, J. Chem. Theory Comput. 16 (2020) 6428–6438

[3] J. Suchan, D. Hollas, B.E.F. Curchod, P. Slavíček, Faraday Discussions 212 (2018) 307.

[4] J. Janoš, D. Madea, S. Mahvidi, T. Mujawar, J. Švenda, J. Suchan, P. Slavíček, P. Klán, J. Phys. Chem. A 124 (2020) 10457-10471

[5] V. Svoboda, R. Michiels, A. C. LaForge, J. Med, F. Stienkemeier, P. Slavíček, H.J. Wörner, Science Advances 6 (2020) eaaz0385, doi: 10.1126/sciadv.aaz0385.