On application of Deep Learning to simplified quantum-classical dynamics in electronically excited states

Evgeny Posenitskiy[1*], Fernand Spiegelman[2], Didier Lemoine[1]

[1]*Laboratoire Collisions Agrégats et Réactivité (LCAR), UT3-Paul Sabatier and CNRS UMR5589, 118 Route de Narbonne, Toulouse, France*

[2]*Laboratoire de Chimie et Physique Quantiques (LCPQ), UT3-Paul Sabatier and CNRS UMR5626, 118 Route de Narbonne, Toulouse, France*

*posenitskiy@irsamc.ups-tlse.fr

ABSTRACT

Deep Learning is applied to simulate non-adiabatic molecular dynamics of phenanthrene based on Time-Dependent Density Functional based Tight Binding (TD-DFTB) [1] approach for excited states combined with mixed quantum-classical propagation. Starting from the Tully's fewest-switches trajectory surface hopping [2] algorithm coupled to TD-DFTB, we examine the simplified treatment based on the Landau-Zener approximation and Deep Learning potentials for excited states. We first assess the accuracy of the TD-DFTB approach based on comparison with experimental and higher-level theoretical data. Using the recently developed SchNet [3,4] architecture for Deep Learning applications, we train several models and evaluate their performance. The main focus is given to the analysis of the electronic population of low-lying excited states computed with the aforementioned methods. We determine the relaxation timescales and compare them with experimental data. Our results show that the simplified approaches considered in this study do not yield accurate description of the electronic relaxation in phenanthrene as compared with the reference data. On the other hand, the SchNet performance allows high-throughput analysis at a negligible cost.

References

[1] S. Hammes-Schiffer and J. C. Tully; Proton transfer in solution: Molecular dynamics with quantum transitions. *J. Chem. Phys.* **1994**, 101, 4657-4667.

[2] T. A. Niehaus, S. Suhai *et al.;* Tight-binding approach to time-dependent density-functional response theory. *Phys. Rev. B* **2001**, 63, 085108.

[3] K. T. Schütt, P.-J. Kindermans *et al.*; SchNet - a deep learning architecture for molecules and materials. *J. Chem. Phys.* **2018**, 148, 24, 241722.

[4] K. T. Schütt, P. Kessel *et al.*; SchNetPack: A Deep Learning Toolbox For Atomistic Systems. *J. Chem. Theory Comput.* **2019**, 15, 1, 448–455.