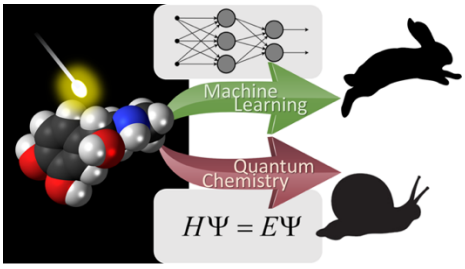
Machine learning for nonadiabatic and quantum dynamics

Pavlo O. Dral[1,2]

[1]*State Key Laboratory of Physical Chemistry of Solid Surfaces, College of Chemistry and Chemical Engineering, Fujian Provincial Key Laboratory of Theoretical and Computational Chemistry, Xiamen University, Xiamen, Fujian 361005, China*
[2]*Institute of Physics, Faculty of Physics, Astronomy, and Informatics, Nicolaus Copernicus University in Toruń, ul. Grudziądzka 5, 87-100 Toruń, Poland*

E-mail: dral@xmu.edu.cn



I will present our latest methods and tools for accelerating nonadiabatic dynamics with machine learning (ML).[1] They are the result of the eight-year effort[2-3] to design the fast and robust active learning protocol to build data-efficient machine learning models for trajectory surface hopping.[1] This protocol has the following key innovations and advantages:[1]

- depending on a system and reference electronic-structure method, the users can get the final results as fast as after a couple of days on a single GPU!
- multi-state learning model which has unrivaled accuracy for excited state properties (accuracy is often better than for models targeting only ground state!). Models can be used for TSH of multiple molecules (not just for a single molecule!)
- gapMD for efficient sampling of the vicinity of conical intersection
- samplings based on uncertainty in probabilities

This protocol and underlying methods are based on open-source MLatom (MLatom.com with many tutorials),[4] which was recently extended to support surface-hopping dynamics[5], also with the universal AIQM1[6] method yielding fast and reasonable photodynamics without the need for training. Deeper integration with Newton-X[7] to support more TSH algorithms is underway as well as integrating the new protocol for cost-efficient generation of the nonlinear time-resolved spectra from nonadiabatic dynamics trajectories[8]. An increasing number of such simulations can be performed online at XACScloud.com which I will use for demonstration of the MLatom capabilities.

Beyond on-the-fly surface hopping dynamics, we explored ML approaches for accelerating,[9-10] learning trajectories as a function of time,[11] and learning the entire trajectories[12] of the quantum dissipative dynamics. These are based on the MLQD program[13].

General discussion about the research in this field is given in our review[14] and book chapters[15-17].

[1] M. Martyka, L. Zhang, F. Ge, Y.-F. Hou, J. Jankowska, M. Barbatti, P. O. Dral. Charting electronic-state manifolds across molecules with multi-state learning and gap-driven dynamics via efficient and robust active

learning. *ChemRxiv, https://doi.org/10.26434/chemrxiv-2024-dtc1w* **2024**.

[2] P. O. Dral, M. Barbatti, W. Thiel. Nonadiabatic Excited-State Dynamics with Machine Learning. *J. Phys. Chem. Lett.* **2018,** *9*, 5660–5663.

[3] W.-K. Chen, X.-Y. Liu, W. Fang, P. O. Dral, G. Cui. Deep Learning for Nonadiabatic Excited-State Dynamics. *J. Phys. Chem. Lett.* **2018,** *9*, 6702–6708.

[4] P. O. Dral, F. Ge, Y.-F. Hou, P. Zheng, Y. Chen, M. Barbatti, O. Isayev, C. Wang, B.-X. Xue, M. Pinheiro Jr, Y. Su, Y. Dai, Y. Chen, S. Zhang, L. Zhang, A. Ullah, Q. Zhang, Y. Ou. MLatom 3: A Platform for Machine Learning-Enhanced Computational Chemistry Simulations and Workflows. *J. Chem. Theory Comput.* **2024,** *20*, 1193–1213.

[5] L. Zhang, S. V. Pios, M. Martyka, F. Ge, Y. F. Hou, Y. Chen, L. Chen, J. Jankowska, M. Barbatti, P. O. Dral. MLatom Software Ecosystem for Surface Hopping Dynamics in Python with Quantum Mechanical and Machine Learning Methods. *J. Chem. Theory Comput.* **2024,** *20*, 5043–5057.

[6] P. Zheng, R. Zubatyuk, W. Wu, O. Isayev, P. O. Dral. Artificial Intelligence-Enhanced Quantum Chemical Method with Broad Applicability. *Nat. Commun.* **2021,** *12*, 7022.

[7] M. Barbatti, M. Bondanza, R. Crespo-Otero, B. Demoulin, P. O. Dral, G. Granucci, F. Kossoski, H. Lischka, B. Mennucci, S. Mukherjee, M. Pederzoli, M. Persico, M. Pinheiro Jr, J. Pittner, F. Plasser, E. Sangiogo Gil, L. Stojanovic. Newton-X Platform: New Software Developments for Surface Hopping and Nuclear Ensembles. *J. Chem. Theory Comput.* **2022,** *18*, 6851–6865.

[8] S. V. Pios, M. F. Gelin, A. Ullah, P. O. Dral, L. Chen. Artificial-Intelligence-Enhanced On-the-Fly Simulation of Nonlinear Time-Resolved Spectra. *J. Phys. Chem. Lett.* **2024,** *15*, 2325–2331.

[9] A. Ullah, P. O. Dral. Speeding up quantum dissipative dynamics of open systems with kernel methods. *New J. Phys.* **2021,** *23*, 113019.

[10] L. E. Herrera Rodríguez, A. Ullah, K. J. Rueda Espinosa, P. O. Dral, A. A. Kananenka. A comparative study of different machine learning methods for dissipative quantum dynamics. *Mach. Learn. Sci. Technol.* **2022,** *3*, 045016.

[11] A. Ullah, P. O. Dral. Predicting the future of excitation energy transfer in light-harvesting complex with artificial intelligence-based quantum dynamics. *Nat. Commun.* **2022,** *13*, 1930.

[12] A. Ullah, P. O. Dral. One-Shot Trajectory Learning of Open Quantum Systems Dynamics. *J. Phys. Chem. Lett.* **2022,** *13*, 6037–6041.

[13] A. Ullah, P. O. Dral. MLQD: A package for machine learning-based quantum dissipative dynamics. *Comput. Phys. Commun.* **2024,** *294*, 108940.

[14] P. O. Dral, M. Barbatti. Molecular Excited States Through a Machine Learning Lens. *Nat. Rev. Chem.* **2021,** *5*, 388–405.

[15] L. Zhang, A. Ullah, M. Pinheiro Jr, M. Barbatti, P. O. Dral, Excited-state dynamics with machine learning. In *Quantum Chemistry in the Age of Machine Learning*, Dral, P. O., Ed. Elsevier: Amsterdam, Netherlands, **2023**; pp. 329–353.

[16] J. Westermayr, P. O. Dral, P. Marquetand, Learning excited-state properties. In *Quantum Chemistry in the Age of Machine Learning*, Dral, P. O., Ed. Elsevier: Amsterdam, Netherlands, **2023**; pp. 467–488.

[17] J. Li, M. Vacher, P. O. Dral, S. A. Lopez, Machine learning methods in photochemistry and photophysics. In *Theoretical and Computational Photochemistry: Fundamentals, Methods, Applications and Synergy with Experimentation*, García-Iriepa, C.; Marazzi, M., Eds. Elsevier: **2023**; pp. 163–189.