Project title: Machine Learning for Molecular Quantum Dynamics

Supervisors: O. Kühn (German PI), M. Schuurman (Canadian PI)

Current state of the art

The simulation of molecular quantum dynamics of large molecular systems often rests on parametrized model Hamiltonians, describing coupled degrees of freedom in terms of low-order Taylor expansions [1]. For instance, the coupled intramolecular nonadiabatic electron-vibrational dynamics is usually described in terms of an expansion with respect to vibrational normal mode coordinates. For extended systems such as photosynthetic pigment-protein complexes or supramolecular architectures mimicking photosynthetic concepts, intramolecular electronic and vibrational degrees of freedom are additionally coupled to environmental modes such as phonons described by a spectral density. The complexity of such scenarios arises from the interplay between different time and length scales. As a consequence, regimes ranging from incoherent kinetics to coherent wave packet dynamics can be realized. In recent years there has been a controversial discussion about the so-called Quantum Biology, i.e. the functional relevance of quantum effects for biological systems [2]. Even in case that photosynthesis doesn't use quantum coherence for efficient light-harvesting, the discussion has brought quantum effects on the general agenda in Chemistry and Material Sciences [3, 4].

Theoretical prediction and characterization of quantum effects in complex molecular systems requires versatile methods for solving the dynamical equations. The standard for solving the high-dimensional Schrödinger equation is the Multiconfiguration Time-dependent Hartree method [5]. When it comes to condensed phase systems, a reduced density matrix description is more appropriate [1] although computationally very demanding if multiple degrees of freedom need to be included for non-Markovian dynamics [6]. In terms of numerical efficiency a stochastic unraveling of the non-Markovian dynamics leads to a scheme coined Hierarchy of Pure States (HOPS) that was shown to have good performance [7]. In HOPS the stochastic integro-differential equation is unraveled into a set of coupled differential equations for the physical and auxiliary state vectors, the latter capturing non-Markovian effects. The stochastic sampling and the propagation of the extended set of state vectors is still numerical demanding for larger system sizes. In order to achieve size-invariant scaling mesoHOPS has been developed, where dynamically relevant subsets of states are identified along with the solution of the dynamical equations [8]. An application to a photosynthetic antenna array was reported in Ref. [9]. Despite the fact that mesoHOPS has been demonstrated to have size invariant scaling, reaching the turn-over point in terms of system size can become computationally demanding such that systematic parameter scans or calculation of spectroscopic observables are challenging.

Employing the power of machine learning (ML) might become a game changer in quantum dynamics research. In Molecular Physics this has been widely recognized in the context of nonadiabatic trajectory simulations [10], where the goal is not the solution of the dynamical equations but to provide on-the-fly potential energy surfaces including nonadiabatic couplings (see also Ref. [11]). While there are approaches to solving the full dynamical equations for model systems using, e.g. Physics-Informed Neural Networks (PINN)[12] or time-dependent Neural Quantum States [13], there are only a few applications of ML techniques to the solution of the time-dependent Schrödinger or Liouville-von Neumann equation for molecular systems. For instance, Hammes-Schiffer et al. used propagator training to simulate proton transfer under the effect of a time-dependent force due to heavy atom motion [14], Zheng et al. developed a PINN to study a two-state plus two-coordinate model of photoisomerization of retinal in rhodopsin [15], and Bande et al used Fourier neural operators to study laser control of a one-dimensional model of a tunneling reaction [16]. In the context of this project most notable is the very recent work by Zhang et al. [17], who employed a NN to learn the stochastic propagator of the HOPS method (see below) and applied the method to spin-boson dynamics.

Research goals and working program

The overarching goal of this project is to develop and implement an ML-based method for quantum dynamics simulations of coupled degrees of freedom interacting with a heat bath. The methodology to be developed will be based on the non-Markovian stochastic Schrödinger equation in HOPS formulation. Reference is the recent work

on NN operator learning by Zhang et al. [17] who formulated the hierarchy of coupled equations for the physical and auxiliary state vector in terms of a time-evolution operator for stochastic quantum trajectories. Key to the NN design were deterministic Fourier layers to capture non-local correlations. Using this approach as a starting point our focus will be shifted towards larger systems in terms of size and dimensionality, implying a better description of real (supra-) molecular systems. Utilization of the mesoHOPS approach appears most promising in this respect. Identifying the NN architecture most compatible with the stochastic propagation of the physical and auxiliary states under the dynamic mesoHOPS truncation will be a key objective of this project. The developed method and numerical toolbox shall be applicable to describe effects of decoherence and energy dissipation for problems covering the range from decoherence of solution-phase photoinduced molecular dynamics through quantum control of molecular qubits to imaging real space mesoscopic transport in supramolecular architectures. The project will be connected to the theoretical efforts of the Ottawa group on developing ML approaches for parametrizing molecular excited-state potential energy surfaces under the influence of environmental fluctuation forces.

[1] V. May and O. Kühn, *Charge and Energy Transfer Dynamics in Molecular Systems*, Wiley-VCH, Weinheim, 2023.

[2] J. Cao et al., *Sci. Adv.* **6**, eaaz4888 (2020).

[3] J. D. Schultz et al., *Chemical Reviews* **124**, 11641 (2024).

[4] G. D. Scholes et al., *Phys. Chem. Lett.* **16**, 1376 (2025).

[5] H.-D. Meyer, *WIREs Comput. Mol. Sci.* **2**, 351 (2011).

[6] Y. Tanimura, *J. Chem. Phys.* **153**, 020901 (2020).

[7] D. Suess et al., A. Eisfeld, and W. T. Strunz, *Phys. Rev. Lett.* **113**, 150403 (2014).

[8] B. Citty et al, *J. Chem. Phys.* **160**, 144118 (2024).

[9] L. Varvelo et al., *J. Phys. Chem. Lett.* **14**, 3077 (2023).

[10] C. Müller et al., *Chem. Sci.* **16**, 17542 (2025).

[11] T. Y. Wang, S. P. Neville, and M. S. Schuurman, *J. Phys. Chem. Lett.* **14**, 7780 (2023).

[12] K. Shah et al., *arXiv*, 2210.12522 (2022).

[13] I. L. Gutiérrez and C. B. Mendl, *Quantum* **6**, 627 (2022).

[14] M. Secor, A. V. Soudackov, and S. Hammes-Schiffer, *J. Phys. Chem. Lett.* **12**, 10654 (2021).

[15] H. Zeng, Y. Kou, and X. Sun, *Molecular Physics*, e2501775 (2025).

[16] K. Singh et al., *J Comput Chem* **45**, 2360 (2024).

[17] J. Zhang, C. L. Benavides-Riveros, and L. Chen, *arXiv*, 2509.01049 (2025).