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Data-Driven Versus Physics-Informed Neural Networks for Nonadiabatic Semiclassical Mapping Dynamics

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Abstract: Semiclassical mapping dynamics offer a computationally tractable approach for simulating nonadiabatic processes in complex molecular systems. This work presents a comparative study of purely data-driven (DD) neural networks and physics-informed neural networks (PINNs) for learning the Markovian propagation of trajectories within the Meyer-Miller-Stock-Thoss (MMST) mapping Hamiltonian framework. Using the spin-boson model as a benchmark, we assess the performance of both approaches in reproducing the nonadiabatic dynamical details for classical mapping model (CMM) and symmetrical quasiclassical (SQC) dynamics. Our results demonstrate that PINNs, which explicitly incorporate the physical equations of motion, significantly outperform DD models, especially when trained with limited datasets. PINNs accurately capture the population dynamics and preserve the physical correlations in phase space, regardless of the underlying neural network architecture (fully connected network or gated recurrent unit) or the specific MMST Hamiltonian formulation. In contrast, DD models exhibit substantial inaccuracies and unphysical behaviors. This clearly shows the key benefit of embedding physical laws into machine learning frameworks to achieve data-efficient, accurate, and reliable simulations of nonadiabatic phenomena.

Key words: machine learning, nonadiabatic dynamics, physics-informed neural network, data-driven approach, mapping Hamiltonian.

1. Introduction

Nonadiabatic dynamics describe the molecular processes involving transitions between multiple electronic potential energy surfaces and are fundamental to a vast array of phenomena in chemistry, physics, and biology. These processes are characterized by the intrinsic coupling of nuclear and electronic motions, which is beyond the scope of the Born-Oppenheimer approximation. They govern the outcomes of photochemical reactions, facilitate charge and energy transfer events, and underpin critical biological functions such as photosynthesis [1–5]. Furthermore, nonadiabatic events are central

to solar energy conversion and the behavior of advanced materials. A deep understanding of these complex mechanisms is therefore indispensable for the rational design of novel molecules and functional materials.

However, despite their commonness, the simulation of nonadiabatic dynamics presents serious theoretical and computational challenges. The primary difficulty stems from the need to treat the coupled evolution of electronic and nuclear degrees of freedom (DOFs) quantum mechanically. It is a task that is computationally prohibitive for most systems, but some low-dimensional models. Consequently, various approximate methods have been developed, though these introduce their own complexities, including numerical stability, error accumulation, and questions about the validity of underlying approximations for certain processes.

In the quest for computationally tractable yet physically sound approaches, semiclassical methods have emerged as an attractive platform, aiming to balance the inclusion of essential quantum mechanical effects with the efficiencies of classical trajectory-based simulations. A cornerstone in this area is the family of semiclassical or quasiclassical methods based on the Meyer-Miller-Stock-Thoss (MMST) mapping Hamiltonian [6,7]. This mapping dynamics formalism provides a prescription to transform a discrete F-level electronic system into an equivalent mapping Hamiltonian. This Hamiltonian is expressed in terms of F continuous Cartesian phase-space variables for the electronic DOFs, and it allows the coupled dynamics of electronic and nuclear motion to be propagated via Hamilton's equations of motion. The MMST Hamiltonian serves as a foundation for diverse semiclassical methods like various linearized semiclassical dynamics (LSC) [8-12], symmetrical quasi-classical (SQC) windowing techniques [13-17], and classical mapping models (CMM) [18-22], to name a few. The specific semiclassical propagation scheme and observable formulation chosen can profoundly influence simulation accuracy, computational cost, and conservation properties, highlighting the complexity beyond the Hamiltonian [23].

Machine learning (ML), particularly techniques based on neural networks (NNs), has rapidly become a powerful tool in computational chemistry, applied to predict molecular properties, accelerate materials discovery, construct accurate potential energy surfaces, and simulate complex system dynamics [24-28]. The application of ML to nonadiabatic dynamics is a rapidly advancing frontier, offering strategies to develop PESs [29,30], directly propagate quantum wavepackets or reduced density matrices (RDMs) [31-33], and learn the evolution of semiclassical or mixed quantum-classical trajectories [32,34,35]. Purely data-driven (DD) NNs, such as convolutional neural networks (CNNs) [36], kernel ridge regression (KRR) [37,38], gated recurrent units (GRUs) [31,39], long short-term memory (LSTM) [33,40,41], and transformer [42] learn input-output relationships directly from large dynamical datasets. While powerful, these models typically require substantial training data and, without explicit encoding of physical laws, may not inherently respect fundamental principles like energy conservation, potentially leading to physically implausible predictions. In contrast, Physics-Informed Neural Networks (PINNs) [43-48] aim to bridge this gap by incorporating known physical laws, often expressed as differential equations or conservation principles, directly into the NN's loss function during training. This can improve data efficiency, enhance generalization, and produce more physically consistent predictions.

The temporal characteristics of dynamics, particularly the distinction between Markovian (future state depends only on the present) and non-Markovian (future depends on history) behavior, also significantly influence ML model design. The propagation of only the electronic RDM is typically non-Markovian if nuclear DOFs are unknown. This aspect was explored in our previous work [31], where we demonstrated from the Nakajima-Zwanzig generalized quantum master equation (GQME)[49,50] that the non-Markovian propagator for the RDM is fundamentally a linear map, provided that the historical information used for training is sufficiently long compared to the system's intrinsic memory time, for which we also proposed an estimator. Subsequent applications to various neural network architectures showed that when this condition of sufficient historical data was met, simpler linear-mapping NNs like linear fully connected networks (FCNs) could achieve high accuracy and even outperform more complex nonlinear architectures such as GRU or hybrid convolutional neural network-LSTM (CNN-LSTM) [32], which might otherwise be prone to overfitting. Instances where nonlinear NNs appeared superior typically corresponded to situations where the provided historical data length was shorter than the system's memory time, thus not conflicting with the underlying linearity of the true non-Markovian propagator. Such non-Markovian ML models, which use a segment of historical dynamics to predict a future time step, are referred to as short-for-long (SFL) type models.

In semiclassical mapping dynamics, however, the total knowledge of electronic mapping variables and nuclear phase space variables is available at each time slice. Training an ML model with this complete instantaneous state information should, in principle, allow for the description of Markovian dynamics, where the next state is predicted solely from the current state, thus simplifying the ML task. Previous work by Lan and coworkers utilized LSTM networks to train SQC mapping dynamics [35] (both electronic and nuclear variables) using short-time windows of trajectory data as input, an approach akin to training for non-Markovian dynamics; their ensemble averages of electronic populations showed good agreement with reference calculations.

In this study, we adopt a distinct Markovian approach to directly compare the effectiveness of purely data-driven NNs (such as FCNs and GRUs) against PINNs for predicting the propagation of nonadiabatic dynamics within the MMST mapping Hamiltonian framework. Importantly, we train our models with only one time step of complete state information (electronic mapping variables and nuclear phase space variables) as input to predict the state at the very next time step, genuinely treating the learning problem as Markovian. In our PINN implementation, which we use as a general term, the known form of the equations of motion (EOM) derived from the mapping Hamiltonian is incorporated into the learning process, while NNs may be employed to learn specific physical terms within these EOMs, such as potential energy surfaces, their gradients, or the zero-point-energy parameter. A primary goal of this study is to use a relatively small training dataset to showcase the potential benefits of incorporating physics-informed properties, i.e., the explicit form of the EOM governing the mapping dynamics, in accurately reproducing the step-to-step propagation. This paper will further detail the theoretical background of the methods, outline the computational approaches and model systems, present a critical discussion of the comparative results, and conclude with key findings and future outlooks.