

Predicting Open Quantum Dynamics with Data-Informed Quantum-Classical Dynamics

Pinchen Xie^{1,*}, Ke Wang², Anupam Mitra¹, Yuanran Zhu¹, Xiantao Li²

Wibe Albert de Jong¹ and Chao Yang^{1,†}

¹*Applied Mathematics and Computational Research Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

²*Department of Mathematics, Pennsylvania State University, University Park, Pennsylvania 16802, USA*



(Received 28 August 2025; revised 8 November 2025; accepted 19 December 2025; published 9 January 2026)

We introduce a data-informed quantum-classical dynamics (DIQCD) approach for predicting the evolution of an open quantum system. The equation of motion in DIQCD is a Lindblad equation with a flexible, time-dependent Hamiltonian that can be optimized to fit sparse and noisy data from local observations of an extensive open quantum system. We demonstrate the accuracy and efficiency of DIQCD for both experimental and simulated quantum devices. We show that DIQCD can predict entanglement dynamics of ultracold molecules (calcium fluoride) in optical tweezer arrays. DIQCD also successfully predicts carrier mobility in organic semiconductors (rubrene) with accuracy comparable to nearly exact numerical methods.

DOI: [10.1103/7lsx-ssjl](https://doi.org/10.1103/7lsx-ssjl)

Modeling a quantum device in a noisy environment can be simplified by focusing on a specific subsystem and treating the remaining degrees of freedom as an effective bath that interacts with the subsystem. There have been various methods that model equations of motion (EOM) of such an open quantum system with a trade-off between accuracy and efficiency [1–11]. On the high-accuracy end, pseudo-mode methods [5,10,12–14] expand the system’s Hilbert space with effective modes that encode the bath’s correlation function. On the high-efficiency end, the plain Lindblad equation sketches a dissipative environment without introducing auxiliary degrees of freedom. In between, mixed quantum-classical dynamics (MQCD) may strike a balance. With additional classical degrees of freedom, MQCD can describe coherent fluctuations that are otherwise reduced to mean-field dissipators in the plain Lindblad equation. However, phenomenological MQCD approaches such as Ehrenfest dynamics [15] and surface hopping [16] can suffer from unsystematic error control relative to exact quantum dynamics [17], hindering accurate predictions of material properties or device metrics. Furthermore, for systems where the environment does not have a particle-based semiclassical representation, phenomenological MQCD becomes irrelevant. For example, one needs to devise special-purpose MQCD models for cold-atom systems [18].

We introduce data-informed quantum-classical dynamics (DIQCD), a method that places MQCD in a general context by introducing variational capacity to the EOM. We developed a framework to optimize a general MQCD EOM, parameterized as a time-dependent Lindblad equation, using time-series data of quantum observable measurements. DIQCD is practical because it requires measurement data from the quantum subsystem rather than the environment. DIQCD is suited for sparse and noisy data due to its restricted variational flexibility, which prevents overfitting.

We demonstrate DIQCD’s applications with two distinct case studies. The first is on the dynamics of calcium-fluoride (CaF) molecular qubits [19]. We train DIQCD on sparse experimental data from a single CaF molecule and then predict two-qubit entanglement dynamics. The second is on quantum transport in rubrene crystal, where DIQCD, trained on data from simulating a single rubrene molecule, predicts carrier mobility with an accuracy similar to a nearly exact time-dependent density matrix renormalization group (TD-DMRG) simulation [20] but at a lower computational cost, bringing a new perspective to modeling bandlike transport.

Methods—In the following, we call the quantum subsystem of interest the “system” and the rest of the degrees of freedom the “environment.” The DIQCD approach is made up of a flexible EOM of the system, a loss function for optimizing the EOM, and the relevant training and simulation algorithms. The EOM is a Lindblad equation with a time-dependent Hamiltonian $\hat{H}_\epsilon(t)$:

$$\frac{d\hat{\rho}_\epsilon(t)}{dt} = -i[\hat{H}_\epsilon(t), \hat{\rho}_\epsilon(t)] + \sum_k \gamma_k \left(\hat{L}_k \hat{\rho}_\epsilon(t) \hat{L}_k^\dagger - \frac{1}{2} \{ \hat{L}_k \hat{L}_k^\dagger, \hat{\rho}_\epsilon(t) \} \right). \quad (1)$$

*Contact author: pinchenxie@lbl.gov

†Contact author: cyang@lbl.gov

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article’s title, journal citation, and DOI.

$\hat{\rho}_\epsilon(t)$ is the density matrix of the system. $\{\hat{L}_k\}$ is a finite set of static jump operators. $\hat{H}_\epsilon(t) = \hat{H}_0 + \hat{H}_c(t) + \sum_{j=1}^M f_j[\epsilon(t)]\hat{S}_j$ combines the static system Hamiltonian H_0 , the external control $\hat{H}_c(t)$, and perturbing Hermitian operators $\hat{S}_j$. $f_j[\epsilon(t)]$ is a scalar function of the multidimensional, classical dynamical processes $\epsilon(t)$ that encodes information about the environment. As a special case, Eq. (1) becomes Ehrenfest dynamics when $\epsilon(t)$ represents atomic nuclei evolving under the average potential energy surface $\text{Tr}[\hat{H}_\epsilon(t)\rho_\epsilon]$. In DIQCD, we allow $\epsilon(t)$ to be generic dynamical processes that can be integrated by an explicit Markovian integrator with or without auxiliary degrees of freedom $\xi(t)$. This means that $\epsilon(t + \delta t)$ and $\xi(t + \delta t)$ are determined by $\epsilon(t)$ and $\xi(t)$, together, in an integration scheme with δt as the time step. These requirements are imposed such that $\epsilon(t)$ can evolve concurrently with the structure-preserving integration [21] of the Lindblad equation. And eventually, the parameters controlling the EOM of $\epsilon(t)$ can be optimized together with other system parameters (such as γ_k) through chain rules, such that the behavior of $\hat{\rho}_\epsilon(t)$ can match time-series data of the system. A Langevin process $\epsilon_0(t)$ is an example of a flexible classical process. The evolution of $\epsilon_0(t)$ can be approximated by [22] $\epsilon_0(t + \delta t) = e^{-\delta t/\tau}\epsilon_0(t) + A\sqrt{1 - e^{-2\delta t/\tau}}\xi_0(t)$ for some parameters τ and A . $\xi_0(t)$ is Gaussian white noise. Simpler examples of flexible processes include periodic signals and Gaussian white noise. Complex examples include molecular dynamics on a parameterized potential energy surface.

In DIQCD, the expectation of any system observable $\hat{O}$ at time t is given as $O(t) = \langle O_\epsilon(t) \rangle_\epsilon = \langle \text{Tr}[\hat{O}\hat{\rho}_\epsilon(t)] \rangle_\epsilon$, where $\langle \cdot \rangle_\epsilon$ represents the average over realizations of $\epsilon(t)$. DIQCD is trained on time-series data at discrete time points $\{t_1, t_2, \dots\}$ with the mean-squared loss $\mathcal{L} = \sum_{ij} [O_i(t_j) - O_i^*(t_j)]^2$. The data $O_i^*(t)$ comes from measurements performed on either an experimental or a simulated system. For example, for an atom-based quantum computing platform, $\hat{O}_i$ can be a Pauli operator acting on the i th qubit, and $\epsilon(t)$ conceptually encapsulates dynamical information about optical traps, environmental radiation, etc. In DIQCD, one does not need to model $\epsilon(t)$ with precise knowledge of the environment. Various classical processes can be incorporated. A guiding principle for practical modeling is to employ a collection of processes characterized by flexible time scales [23], initialized to broadly encompass the range of time scales from δt up to the quantum coherence time and extending to infinity to represent shot-by-shot noise. Other than that, DIQCD does not specify the choice of $\{\hat{S}_j\}$ or the choice of system degrees of freedom. These considerations should be made with a phenomenological understanding of a quantum device.

The DIQCD model is trained using a forward-back-propagation loop, in which $\rho_\epsilon(t)$ and $\epsilon(t)$ are propagated from $t = 0$ to T via straightforward numerical integration

of Eq. (1) and the associated classical EOMs. We calculate the loss function $\mathcal{L}$, backpropagate the loss over the entire trajectory of $\rho_\epsilon(t)$ and $\epsilon(t)$, and update all flexible parameters with accumulated gradients. Details on the implementation of DIQCD, i.e., the Python package QEpsilon [24], are reported in End Matter.

CaF molecular qubits—Reconfigurable optical tweezer arrays of ultracold molecules have emerged as a promising platform for precision measurement and quantum simulation [19,25–27]. A precise *ab initio* model to capture multiscale quantum dynamics of this platform is not yet available. DIQCD can circumvent this difficulty and establish a reliable and scalable dynamical model based on data collected from a minimal unit of the platform: a single trapped molecule.

For example, we consider a recent realization of the platform [19] (see End Matter for details), where two CaF molecules are trapped in respective optical tweezers separated by a distance of d . The trapping potential is illustrated in Figs. 1(a) and 1(b). Let $\mathbf{R}(t) = (\mathbf{r}_1(t), \mathbf{r}_2(t))$ be the center-of-mass positions of the two molecules [28]. The quantum system here is relevant to two hyperfine states $|\uparrow\rangle$ and $|\downarrow\rangle$ of each molecule. Let $\hat{S}_i^\alpha = (\hbar/2)\hat{\sigma}_i^\alpha$ ($i = 1, 2$; $\alpha = x, y, z$) be spin-1/2 operator for molecule i . An effective Hamiltonian $H_\epsilon(t)$, used by DIQCD to describe the system, contains the one-body terms

$$H_\epsilon^{(1)}(t) = \sum_{j=1}^L \epsilon_{1,j}(t)\hat{S}_1^z + \sum_{j=1}^L \epsilon_{2,j}(t)\hat{S}_2^z \quad (2)$$

and the two-body dipole-dipole interaction

$$H_\epsilon^{(2)}(t) = J(\mathbf{r}_1, \mathbf{r}_2)(\hat{S}_1^x\hat{S}_2^x + \hat{S}_1^y\hat{S}_2^y). \quad (3)$$

Here, $J(\mathbf{r}_1, \mathbf{r}_2) = (J_0/r_{12}^3)(1 - 3\cos^2\theta')$ with J_0 being the coupling constant, r_{12} being $\|\mathbf{r}_1 - \mathbf{r}_2\|$, and θ' being the angle between $(\mathbf{r}_1 - \mathbf{r}_2)$ and the quantization axis (y axis).

From phenomenological understanding of the environments, we take an ansatz with $L = 6$: $\epsilon_{i,l}$ ($i = 1, 2$; $l = 1, 2, 3, 4$) are periodic processes with fixed frequency $\omega_l = l \times 60$ Hz and flexible [30] amplitudes. $\epsilon_{i,5}$ are flexible, overdamped Langevin processes. $\epsilon_{i,6}$ are time-independent, shot-by-shot noises with uniformly distributed initial values sampled from a flexible interval. Here, $\epsilon_{i,l}$ ($l = 1, 2, 3, 4$) represent line noises from power sources. $\epsilon_{i,6}$ represents noises with a time scale above ms. The Langevin process $\epsilon_{i,5}$ with a continuous spectrum approximates the mix of other sources of noise with time scales comparable to execution time. The choices of these processes are not unique, but should be organized in a way that adaptively covers a large range of time scales encompassing the time scale of system dynamics.

The DIQCD EOM is then given by Eq. (1) with one-body jump operators $\{\hat{S}_i^x, \hat{S}_i^z\}_{i=1,2}$ and two flexible

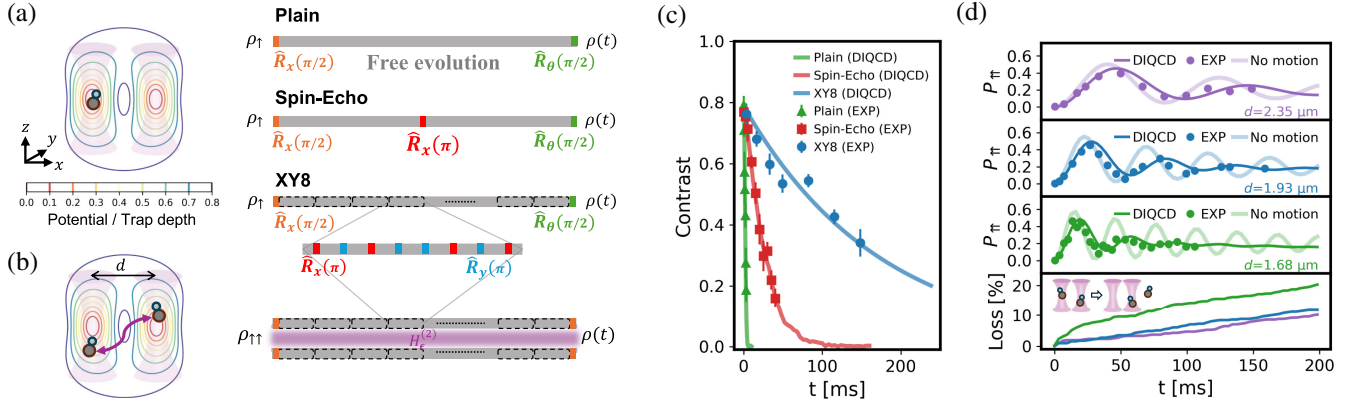


FIG. 1. (a) Optically trapped CaF molecule and three control schemes used in Ramsey experiments. Contour lines represent the equipotential surface of the trap, with energy levels indicated in the color map. (b) Pair of CaF molecules and the control scheme for Bell state generation. (c) Contrast $C(t)$ (scaled by ζ) as a function of circuit duration t . Circle, square, and triangle markers represent $C^{\text{EXP}}(t)$ data. (d) Top three panels show $P_{\uparrow\uparrow}$ (scaled by ζ^2) as a function of t . The bottom panel shows the rate of qubit loss as a function of t .

damping rates γ_x and γ_z , as a minimal parameterization of one-body dissipation. The EOM of $(\mathbf{r}_1, \mathbf{r}_2)$ is standard isothermal molecular dynamics (see End Matter) without nonadiabatic force because $\text{Tr}(\rho H_\epsilon^{(2)})$ is negligible for all values of d used in experiments.

We train the DIQCD model on data from a series of one-molecule Ramsey experiments [19], where the motion of the molecule's position is irrelevant. Experiments were performed with initial state $|\uparrow\rangle$ and three external control schemes as illustrated in Fig. 1(a): a plain scheme, a spin-echo scheme, and an XY8 scheme. These schemes probe the environment over different time scales [31], allowing DIQCD to distinguish noise components across these scales. Since the external controls consist of short pulses, we do not model them as a continuous control $\hat{H}_c(t)$, but as instantaneous one-body unitary rotations $\hat{U} = \hat{R}_{\hat{n}}(\cdot)$ along axis $\hat{n}$, followed by an instantaneous quantum error channel. Their combined effect is given as $\rho(t + \delta t) = \sum_{\alpha=1}^4 \hat{K}_\alpha \hat{U} \rho(t) \hat{U}^\dagger \hat{K}_\alpha^\dagger$, where the Kraus operators $\hat{K}_\alpha$ represent a depolarization error channel [32] controlled by a flexible scalar p . The collected data from the Ramsey experiments is the spin-up probability $P_\uparrow(t, \theta)$ as a function of circuit duration t and the angle θ associated with the last control pulse [see Fig. 1(a)]. Let $C(t) = |P_\uparrow(t, \pi) - P_\uparrow(t, 0)|$. Experiments yield 24 $C^{\text{EXP}}(t)$ data points shown in Fig. 1(c), with a state preparation fidelity $\zeta \approx 0.79$. The parameters embedded in $\{\epsilon_{1,j}\} \cup \{\epsilon_{2,j}\} \cup \{\gamma_x, \gamma_z, p\}$ are optimized by matching $C(t)$ predicted by a single DIQCD model, but with three control schemes, with all 24 $C^{\text{EXP}}(t)/\zeta$ data points. The solid lines in Fig. 1(c) are $\zeta C(t)$ from the optimized DIQCD, showing excellent agreement with experiments across multiple time scales. Such success would not be achieved by the plain Lindblad equation. All parameters in DIQCD converge to substantially nonzero values [33],

indicating a separation of noise over frequency regime. The optimized magnitude of the shot-by-shot noise ($\epsilon_{i,6}$) is comparable to other sources of decoherence, suggesting that low-frequency noises may be further disentangled if one can perform experiments probing a larger time scale.

Next, we model a two-molecule system by including the thermal dynamics of $\mathbf{R}(t)$ and the dipole-dipole interaction $H_\epsilon^{(2)}(t)$ in DIQCD. Radial and axial temperatures of the molecules are set to $T_r = 6 \mu\text{K}$ and $T_a = 18 \mu\text{K}$ (from Raman thermometry [29]) for isothermal simulation of $\mathbf{R}(t)$. The resultant DIQCD model is not trained on any two-molecule data. We validate its prediction against experimental results on Bell state creation. [See Fig. 1(b) and End Matter]. The ideal (without decoherence or molecular motion) probability of observing $|\uparrow\uparrow\rangle$ from the final measurement is $P_{\uparrow\uparrow} = \sin^2(J_0 t / 4\hbar d^3)$. The oscillation in $P_{\uparrow\uparrow}$ is damped in experimental measurements $P_{\uparrow\uparrow}^{\text{EXP}}$, plotted in Fig. 1(d) for different tweezer separations d . DIQCD can predict $\zeta^2 P_{\uparrow\uparrow}$ [solid lines in the top three panels of Fig. 1(d)] in excellent consistency with $P_{\uparrow\uparrow}^{\text{EXP}}$. Moreover, molecular dynamics predict the rate of qubit loss [34] [see Fig. 1(d)] due to molecules hopping from one optical trap to another. The importance of molecular dynamics is revealed when we simulate DIQCD with molecules fixed to tweezer centers—the resultant $\zeta^2 P_{\uparrow\uparrow}$ [semitransparent lines in Fig. 1(d)] shows underestimated damping and overestimated frequency of oscillation.

One can generalize the DIQCD model to a larger array of tweezers with pairwise dipolar interactions or extend the approach to quantum devices involving instantaneous feedback action [35]. Furthermore, DIQCD can model molecules cooled to almost the motional ground state by replacing classical molecular dynamics with ring-polymer path-integral molecular dynamics [36].

Rubrene crystal—The development of flexible electronics requires screening of high-mobility organic semiconductors [37]. Here we show the relevance of DIQCD with first principles-based prediction of the intrinsic carrier mobility (μ) of rubrene crystal, which has an excellent hole mobility [38,39]. The high mobility is due to the coherent quantum transport predominantly along the π -stacking direction. The effective model for transport in rubrene crystal can therefore be reduced to a 1D Holstein model with the Hamiltonian $\hat{H} = \hat{H}_e + \hat{H}_{ph} + \hat{H}_{ep}$. Let n be the label of a rubrene molecule on the 1D molecular lattice along the stacking direction. Let $\hbar = 1$. The single-carrier tight-binding Hamiltonian $\hat{H}_e = V \sum_n (c_{n+1}^\dagger c_n + c_n^\dagger c_{n+1})$ describes intermolecule carrier hopping. The harmonic Hamiltonian $\hat{H}_p = \sum_{n,m} \omega_m b_{n,m}^\dagger b_{n,m}$ is associated with the bosonic vibrational modes of each molecule. The interaction $\hat{H}_{ep} = \sum_{n,m} g_m \omega_m (b_{n,m}^\dagger + b_{n,m}) c_n^\dagger c_n$ is associated with electron-phonon coupling, of which the strength is characterized by the polaron binding energy $\lambda = \sum_m g_m^2 \omega_m$.

Modeling quantum transport is challenging when λ is of the same order of magnitude as V [40], a regime where Marcus theory is inaccurate and traditional MQCD approaches offer results with uncontrolled error. While TD-DMRG can simulate unitary dynamics driven by $\hat{H}$ with nearly full accuracy, its application is restricted to small length scales, which allows for the determination of μ with the Kubo formula, but not real-time, mesoscale transport [41]. By contrast, by treating the carrier as the system and bosonic modes as the environment, we demonstrate that DIQCD can predict μ with accuracy comparable to TD-DMRG, while offering superior numerical efficiency. DIQCD requires training data from only a single rubrene molecule since the main mechanism slowing down the coherent transport considered here is phase decoherence from local system-environment interaction.

We adopt the model of $\hat{H}$ with $V = 83$ meV, $\lambda = 73$ meV, and nine renormalized vibration modes for each molecule with $g_m \in [0, 1]$ and $\omega_m \in [84, 1594]$ cm $^{-1}$ (see End Matter for details). These parameters were obtained in Refs. [20,42] from a hybrid-level density functional theory [43–45]. Here, we are mainly interested in deriving μ near the room temperature. For $T = 200, 250, 300, 350$, and 400 K, we generate one-molecule data by simulating the unitary evolution driven by an effective spin-boson Hamiltonian,

$$\hat{H}^{\text{eff}} = \sum_{m=1}^9 \omega_m b_m^\dagger b_m + \sum_{m=1}^9 \frac{1}{2} g_m \omega_m (b_m^\dagger + b_m) (1 + \hat{\sigma}_z), \quad (4)$$

with the initial state $(\sqrt{\phi}|\uparrow\rangle + \sqrt{1-\phi}|\downarrow\rangle) \otimes |n_1, n_2, \dots, n_9\rangle$. The Pauli operators ($\hat{\sigma}_{\alpha=x,y,z}$) are associated with the two states $|\uparrow\rangle$ and $|\downarrow\rangle$, representing the presence and absence of a carrier, respectively. The bosonic

state $|n_1, n_2, \dots, n_9\rangle$ associated with nine vibrational modes is sampled from the Boltzmann distribution $\exp(-\beta \sum_{m=1}^9 \omega_m b_m^\dagger b_m)$. The mean and standard deviation (concerning the sampled trajectories) of $\langle \hat{\sigma}_x(t) \rangle$ and $\langle \hat{\sigma}_y(t) \rangle$ ($t \in [0, 100]$ fs) are obtained for each T from a sufficient number of quantum trajectories initiated with the same ϕ [46] but different pure states.

These data are used to optimize a one-site DIQCD model with $\hat{H}_e^{\text{eff}}(t) = \epsilon_0 \hat{\sigma}_z + \sum_{m=1}^9 \frac{1}{2} \epsilon_m(t) (1 + \hat{\sigma}_z)$ and the jumping operator $(1 + \hat{\sigma}_z)/2$ (γ is the damping rate), for each T separately. Here, ϵ_0 and γ are flexible, time-independent scalars. $\epsilon_m(t)$ is chosen to be a periodic sinusoidal signal with flexible amplitude, fixed frequency ω_m , and random initial phases. These choices are motivated by the fact that, at high temperatures, the quantum statistics of phonons converge to classical statistics of harmonic oscillators. Traditional MQCD approaches [47] adopt this rigid classical approximation also at low temperatures, leading to uncontrolled error from quantum kinetic energy. By contrast, DIQCD can incorporate excessive kinetic energy with a flexible classical environment [42]. Indeed, after training the parameters of $\{\epsilon_0, \gamma\} \cup \{\epsilon_m\}$ with data associated with the time interval $t \in [0, 70]$ fs, DIQCD models agree well with the data on $\langle \hat{\sigma}_x(t) \rangle$ and $\langle \hat{\sigma}_y(t) \rangle$ [see Fig. 2(b) for the case of $T = 300$ K as an example].

The one-site model is generalized for the lattice as

$$\hat{H}_e(t) = \hat{H}_e + \sum_n \sum_{m=1}^9 \epsilon_m^{(n)}(t) c_n^\dagger c_n \quad (5)$$

with the jump operator $\hat{L}_n = c_n^\dagger c_n$ (damping rate is γ) on any site n . We estimate the carrier mobility by simulating Eq. (1) with the initial condition that one carrier is localized on the middle site of a finite molecular lattice of size $L = 150$ and intermolecular distance $D = 7$ Å. The diffusion of the carrier at $T = 300$ K is illustrated by Figs. 2(d) and 2(e), showing a long-term behavior consistent with classical Fick's law. The mobility is calculated as $\mu(T) = (eD^2/2k_B T) \lim_{t \rightarrow \infty} (d/dt) \langle \text{Tr}[\rho_\epsilon(t) n^2] - \text{Tr}^2[\rho_\epsilon(t) n] \rangle_\epsilon$. The estimation obtained from DIQCD agrees closely with experimental estimation of intrinsic mobility [38], as well as the μ calculated from TD-DMRG [20] for a system with $L = 21$ and $D = 7$ Å. The comparisons are shown in Fig. 2(c), which also reports the μ that we calculated from the Ehrenfest approach [47] (with nonadiabatic force). As expected, the Ehrenfest approach deviates more significantly from TD-DMRG and DIQCD at lower temperatures. It is unclear whether the minor but persistent discrepancy between DIQCD and TD-DMRG is due to the finite-size error in TD-DMRG or the approximation made by DIQCD. However, more importantly, DIQCD and TD-DMRG yield almost the same temperature-induced variation in μ , indicating that DIQCD can mimic subtle variations in the quantum

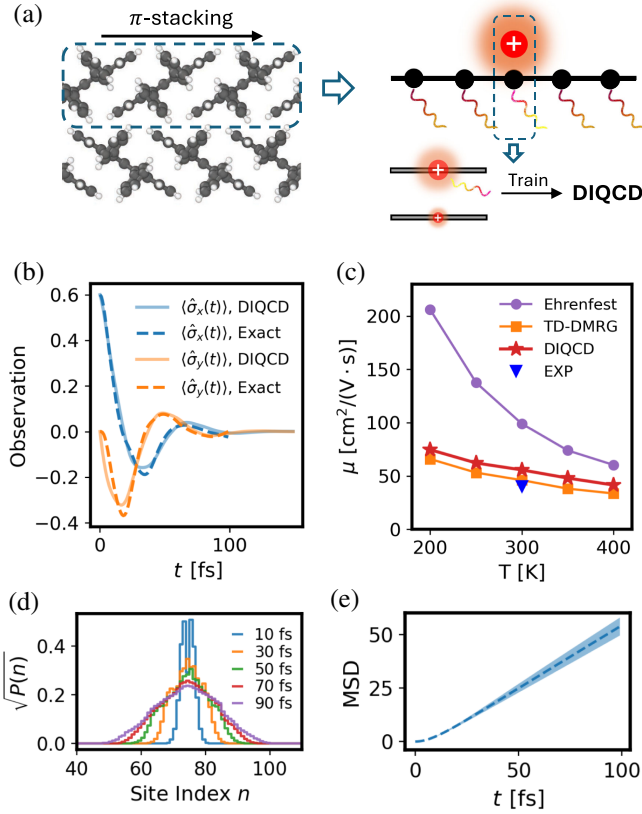


FIG. 2. (a) Rubrene crystal (left) and the effective model for carrier transport (right). (b) Comparison of data and DIQCD on average $\langle \hat{\sigma}_x(t) \rangle$ and $\langle \hat{\sigma}_y(t) \rangle$ for $T = 300$ K. (c) Carrier mobility as a function of temperature. The blue triangle is an experimental estimation of the intrinsic mobility [38]. (d) Square root of site occupation on the $L = 150$ molecular lattice. Snapshots are obtained from a DIQCD simulation lasting 100 fs for $T = 300$ K. (e) Mean squared displacement ($\text{MSD} = \langle \text{Tr}[\rho_\epsilon(t)n^2] - \text{Tr}^2[\rho_\epsilon(t)n] \rangle_\epsilon$) as a function of time for $T = 300$ K. The shades represent standard deviation.

kinetic energy that outweighs the classical fluctuation in shaping the transport.

Our approach can be readily applied to a two-dimensional lattice model. Point defects can also be introduced by training several one-site DIQCD models that combine into an inhomogeneous lattice model.

Discussion—DIQCD strikes a balance between accuracy and efficiency. In both case studies, DIQCD is trained on data from localized subsystems, reflecting typical real-world data collection. The resultant model is then generalized to the whole system by introducing known long-range interactions (dipolar interaction and intermolecular hopping). A key presumption here is that the noise in long-range interaction is either negligible or can be described with a known model, which holds for our two case studies. If this assumption does not hold, DIQCD would require training on data from a larger portion of the system to capture correlated noise. This may require

approaches like TD-DMRG or multiconfiguration time-dependent Hartree [48] if the data source is numerical simulations.

Although explicit semiclassical approximation are avoided, DIQCD requires a clear understanding of the quantum device and the observables that govern the dynamical process relevant to device metrics. This challenge is similar to selecting a “good” set of collective variables for molecular conformational dynamics. Although physical intuitions are available for the case studies presented here, for general open quantum systems, one needs a systematic method for defining a canonical transformation that reveals the most relevant low-dimensional observables for data collection.

The Python package QEpsilon is publicly available at Github [24]. A tutorial for using QEpsilon is provided in the online documentation [49]. All results reported in this Letter can be reproduced through annotated code examples (“QEpsilon/examples/CaF_qubits” and “QEpsilon/examples/Rubrene_Crystal”) within the package.

Acknowledgments—P. X. thanks Yukai Lu for his tremendous help with the specifications of the CaF qubit experiments and his contributions to preparing the manuscript. P. X. thanks Weitang Li for his help with the specifications of the Holstein model of rubrene. We thank Zhen Huang, Lin Lin, and Roberto Car for fruitful discussion. P. X. was supported by the Alvarez Fellowship of Lawrence Berkeley National Lab under Contract No. DE-AC02-05CH11231. K. W. and X. L. were supported by the NSF under Grants No. DMS-2111221 and No. CCF-2312456. C. Y. and Y. Z. were supported by the U.S. Department of Energy, Office of Science, Accelerated Research in Quantum Computing Centers, Quantum Utility through Advanced Computational Quantum Algorithms, under Grant No. DE-SC0025572. A. M. and W. A. d. J. were supported by the U.S. Department of Energy under Contract No. DE-AC02-05CH11231 Office of Science, Accelerated Research in Quantum Computing Centers, FAR-QC. Calculations were performed on the National Energy Research Scientific Computing Center (NERSC), a U.S. Department of Energy Office of Science User Facility operated under Contract No. DE-AC02-05CH11231.

Data availability—The data that support the findings of this article are openly available [24].

- [1] G. Lindblad, On the generators of quantum dynamical semigroups, *Commun. Math. Phys.* **48**, 119 (1976).
- [2] V. Gorini, A. Kossakowski, and E. C. G. Sudarshan, Completely positive dynamical semigroups of n -level systems, *J. Math. Phys. (N.Y.)* **17**, 821 (1976).
- [3] A. Garg, J. N. Onuchic, and V. Ambegaokar, Effect of friction on electron transfer in biomolecules, *J. Chem. Phys.* **83**, 4491 (1985).

- [4] Y. Tanimura and R. Kubo, Time evolution of a quantum system in contact with a nearly Gaussian-Markoffian noise bath, *J. Phys. Soc. Jpn.* **58**, 101 (1989).
- [5] B. M. Garraway, Decay of an atom coupled strongly to a reservoir, *Phys. Rev. A* **55**, 4636 (1997).
- [6] L. Diósi and W. T. Strunz, The non-Markovian stochastic Schrödinger equation for open systems, *Phys. Lett. A* **235**, 569 (1997).
- [7] Y. Tanimura, Stochastic Liouville, Langevin, Fokker-Planck, and master equation approaches to quantum dissipative systems, *J. Phys. Soc. Jpn.* **75**, 082001 (2006).
- [8] R.-X. Xu and Y. Yan, Dynamics of quantum dissipation systems interacting with bosonic canonical bath: Hierarchical equations of motion approach, *Phys. Rev. E* **75**, 031107 (2007).
- [9] D. Suess, A. Eisfeld, and W. Strunz, Hierarchy of stochastic pure states for open quantum system dynamics, *Phys. Rev. Lett.* **113**, 150403 (2014).
- [10] D. Tamascelli, A. Smirne, S. F. Huelga, and M. B. Plenio, Nonperturbative treatment of non-Markovian dynamics of open quantum systems, *Phys. Rev. Lett.* **120**, 030402 (2018).
- [11] K. Wang and X. Li, Simulation-assisted learning of open quantum systems, *Quantum* **8**, 1407 (2024).
- [12] X. Li, Markovian embedding procedures for non-Markovian stochastic Schrödinger equations, *Phys. Lett. A* **387**, 127036 (2021).
- [13] G. Park, Z. Huang, Y. Zhu, C. Yang, G. K.-L. Chan, and L. Lin, Quasi-Lindblad pseudomode theory for open quantum systems, *Phys. Rev. B* **110**, 195148 (2024).
- [14] Z. Huang, G. Park, G. K. Chan, and L. Lin, Coupled Lindblad pseudomode theory for simulating open quantum systems, *arXiv:2506.10308*.
- [15] P. Ehrenfest, Bemerkung über die angenäherte gültigkeit der klassischen mechanik innerhalb der quantenmechanik, *Z. Phys.* **45**, 455 (1927).
- [16] J. C. Tully, Molecular dynamics with electronic transitions, *J. Chem. Phys.* **93**, 1061 (1990).
- [17] J. E. Subotnik, A. Jain, B. Landry, A. Petit, W. Ouyang, and N. Bellonzi, Understanding the surface hopping view of electronic transitions and decoherence, *Annu. Rev. Phys. Chem.* **67**, 387 (2016).
- [18] A. L. Shaw, Learning, verifying, and erasing errors on a chaotic and highly entangled programmable quantum simulator, Ph.D. thesis, California Institute of Technology, 2024.
- [19] C. M. Holland, Y. Lu, and L. W. Cheuk, On-demand entanglement of molecules in a reconfigurable optical tweezer array, *Science* **382**, 1143 (2023).
- [20] W. Li, J. Ren, and Z. Shuai, Finite-temperature TD-DMRG for the carrier mobility of organic semiconductors, *J. Phys. Chem. Lett.* **11**, 4930 (2020).
- [21] Y. Cao and J. Lu, Structure-preserving numerical schemes for Lindblad equations, *J. Sci. Comput.* **102**, 27 (2025).
- [22] Z. Zhang, X. Liu, K. Yan, M. E. Tuckerman, and J. Liu, Unified efficient thermostat scheme for the canonical ensemble with holonomic or isokinetic constraints via molecular dynamics, *J. Phys. Chem. A* **123**, 6056 (2019).
- [23] Such as the damping time in a Langevin process and the angular frequency of an harmonic oscillator.
- [24] QEpsilon, <https://github.com/salinelake/QEpsilon> (accessed: 2025-08-26).
- [25] A. M. Kaufman and K.-K. Ni, Quantum science with optical tweezer arrays of ultracold atoms and molecules, *Nat. Phys.* **17**, 1324 (2021).
- [26] S. Burchesky, L. Anderegg, Y. Bao, S. S. Yu, E. Chae, W. Ketterle, K.-K. Ni, and J. M. Doyle, Rotational coherence times of polar molecules in optical tweezers, *Phys. Rev. Lett.* **127**, 123202 (2021).
- [27] Y. Bao, S. S. Yu, L. Anderegg, E. Chae, W. Ketterle, K.-K. Ni, and J. M. Doyle, Dipolar spin-exchange and entanglement between molecules in an optical tweezer array, *Science* **382**, 1138 (2023).
- [28] For the experimental molecular temperature [19] without further cooling [29], the quantum fluctuation in $\mathbf{r}_1$ and $\mathbf{r}_2$ is insignificant.
- [29] Y. Lu, S. J. Li, C. M. Holland, and L. W. Cheuk, Raman sideband cooling of molecules in an optical tweezer array, *Nat. Phys.* **20**, 389 (2024).
- [30] The word “flexible” indicates quantities that will be optimized with data.
- [31] L. Viola, E. Knill, and S. Lloyd, Dynamical decoupling of open quantum systems, *Phys. Rev. Lett.* **82**, 2417 (1999).
- [32] $\hat{K}_0 = \sqrt{1 - p}\hat{I}$, $\hat{K}_1 = \sqrt{(p/3)}\hat{\sigma}_x$, $\hat{K}_2 = \sqrt{(p/3)}\hat{\sigma}_y$, and $\hat{K}_3 = \sqrt{(p/3)}\hat{\sigma}_z$.
- [33] See Supplemental Code in [24] for the evolution of all parameters during the training.
- [34] The qubit loss is severe for $d < 1.68 \mu\text{m}$ due to parametric heating of molecules. For the same reason, the current DIQCD model with a fixed molecular temperature cannot be directly applied to $d < 1.68 \mu\text{m}$.
- [35] H. M. Wiseman and G. J. Milburn, Quantum theory of optical feedback via homodyne detection, *Phys. Rev. Lett.* **70**, 548 (1993).
- [36] D. Marx and M. Parrinello, *Ab initio* path integral molecular dynamics: Basic ideas, *J. Chem. Phys.* **104**, 4077 (1996).
- [37] J. Mei, Y. Diao, A. L. Appleton, L. Fang, and Z. Bao, Integrated materials design of organic semiconductors for field-effect transistors, *J. Am. Chem. Soc.* **135**, 6724 (2013).
- [38] J. Takeya, M. Yamagishi, Y. Tominari, R. Hirahara, Y. Nakazawa, T. Nishikawa, T. Kawase, T. Shimoda, and S. Ogawa, Very high-mobility organic single-crystal transistors with in-crystal conduction channels, *Appl. Phys. Lett.* **90**, 102120 (2007).
- [39] X. Ren, M. J. Bruzek, D. A. Hanifi, A. Schulzetenberg, Y. Wu, C.-H. Kim, Z. Zhang, J. E. Johns, A. Salleo, S. Fratini *et al.*, Negative isotope effect on field-effect hole transport in fully substituted ^{13}C -rubrene, *Adv. Electron. Mater.* **3**, 1700018 (2017).
- [40] This happens to be the case for most high-mobility organic semiconductors.
- [41] The success of TD-DMRG is also limited to 1D due to the 1D nature of the matrix product state and the numerical inefficiency of higher-dimensional tensor networks.
- [42] Y. Jiang, X. Zhong, W. Shi, Q. Peng, H. Geng, Y. Zhao, and Z. Shuai, Nuclear quantum tunnelling and carrier delocalization effects to bridge the gap between hopping and bandlike behaviors in organic semiconductors, *Nanoscale Horiz.* **1**, 53 (2016).

- [43] A. D. Becke, Density-functional thermochemistry. III. The role of exact exchange, *J. Chem. Phys.* **98**, 5648 (1993).
- [44] C. Lee, W. Yang, and R. G. Parr, Development of the Colle-Salvetti correlation-energy formula into a functional of the electron density, *Phys. Rev. B* **37**, 785 (1988).
- [45] M. J. Frisch *et al.*, *Gaussian 09 Revision D.01 ed.* (Gaussian Inc., Wallingford, CT, 2009).
- [46] Results are not sensitive to the choice of ϕ .
- [47] L. Wang, D. Beljonne, L. Chen, and Q. Shi, Mixed quantum-classical simulations of charge transport in organic

- materials: Numerical benchmark of the Su-Schrieffer-Heeger model, *J. Chem. Phys.* **134**, 244116 (2011).
- [48] H.-D. Meyer, U. Manthe, and L. S. Cederbaum, The multi-configurational time-dependent Hartree approach, *Chem. Phys. Lett.* **165**, 73 (1990).
- [49] Documentation of QEpsilon, <https://qepsilon.readthedocs.io/en/latest/> (accessed: 2025-08-26).
- [50] R. Kubo, The fluctuation-dissipation theorem, *Rep. Prog. Phys.* **29**, 255 (1966).

End Matter

Appendix A: QEpsilon—A schematic representation of the structure of QEpsilon is shown in Fig. 3. QEpsilon can be roughly divided into the following modules: (1) a library of second-quantized bosonic, spin, and tight-binding operators as the basis for composing a many-body Hamiltonian and the jump operators; (2) a library of typical classical-process integrators for parameterizing $\epsilon(t)$; (3) a user interface for composing a many-body mixed system with these libraries; (4) structure-preserving integrators for simulating unitary and Lindblad dynamics (we adopt the $M = 1$ integrator in Ref. [21] for Lindblad equation); and (5) a mechanism to train a DIQCD model on time-series data with backpropagation. These modularized components allow for training and large-scale simulation in a unified, GPU-enabled framework. More technical details of QEpsilon can be found in the online documentation of the open-source package [24].

Appendix B: CaF molecular qubits—Optical tweezer: A single optical tweezer has a Gaussian trapping potential $U_T(r, z) = -V(w_0/w(z))^2 e^{[-2r^2/w^2(z)]}$ with respect to radial distance r and height z from the center. $V_m = k_B \times 1.28$ mK is the maximal depth of the trap. $w_0 = 730$ nm is the waist of the beam while

$w(z) = w_0 \sqrt{1 + (z/z_R)^2}$ with the Rayleigh range $z_R = \pi w_0^2/\lambda$. $\lambda = 781$ nm is the wavelength. When performing a two-qubit gate, the central depth of the trap is limited to $V \approx k_B \times 0.13$ mK.

XY8 sequence: The size of each XY8 block (see Fig. 1) is 1.6 ms for one-qubit experiments and two-qubit experiments with $d < 1.68$ μm and 3.2 ms for two-qubit experiments with $d \geq 1.68$ μm .

CaF molecules: The coupling constant for dipole-dipole interaction is $J_0 = 1942.3 \hbar \times \text{Hz} \times \mu\text{m}^3$ [19]. When performing the two-qubit gate, Raman thermometry gives a rough estimation of the molecular temperature that amounts to $T_r = 6$ μK for radial motion (parallel to the x - y plane) and $T_a = 18$ μK for axial motion (z direction). Note that the molecular temperature changes with the depth of the optical trap. So the temperature here is lower than the temperature estimated at full depth [19].

Bell state creation: The scheme is illustrated by Fig. 1(b). The initial state is $|\uparrow\uparrow\rangle$. Two $\pi/2$ pulses along the x axis are applied at the start and end of the scheme. XY8 sequences are applied consecutively for error suppression.

Molecular dynamics of $R(t)$: A Langevin integrator [22] is used to simulate the molecular dynamics of $R(t)$ and its velocity $v(t)$, given as

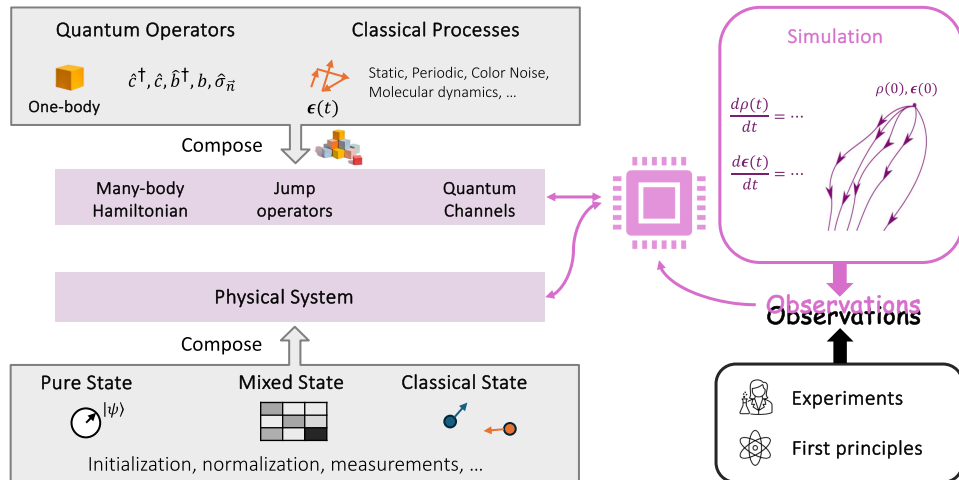


FIG. 3. Structure of QEpsilon.

TABLE I. Frequency of kept vibrational modes and the renormalized coupling constants.

m	1	2	3	4	5	6	7	8	9
ω_m/cm^{-1}	84	214	632	1002	1206	1351	1364	1535	1594
g_m	0.96	0.37	0.25	0.20	0.15	0.31	0.13	0.20	0.31

$$\begin{cases} v_i^{\text{mid}}(t) = v_i(t) + F_i(t)\delta t/m \\ R_i^{\text{mid}}(t) = R_i(t) + 0.5v_i^{\text{mid}}(t)\delta t \\ v_i(t + \delta t) = e^{-\delta t/\tau}v_i^{\text{mid}}(t) + A_i\sqrt{1 - e^{-2\delta t/\tau}}\xi_i(t) \\ R_i(t + \delta t) = R_i(t) + 0.5v_i(t + \delta t)dt. \end{cases} \quad (\text{B1})$$

$F_i(t)$ is the gradient force from the optical trap on the i th ($i \in [1, 6]$) component of R . The radial (x and y directions) and axial (z direction) motion are controlled [50] with the same damping time ($\tau = 100$ ms) but different amplitude (A) of white noise (ξ) to maintain their respective temperature $T_r = 6$ μK and $T_a = 18$ μK .

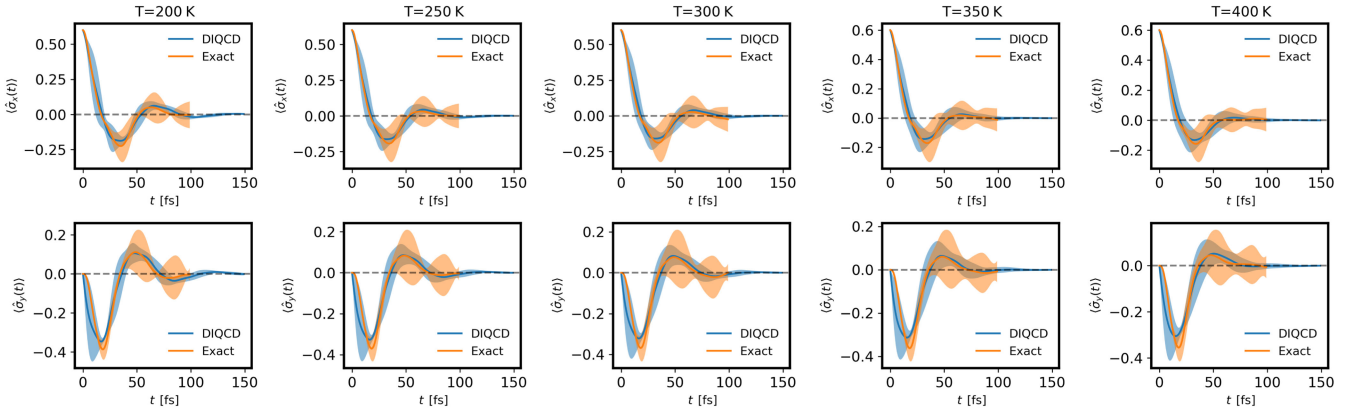
Training: We train the DIQCD model with all 24 one-qubit data (see Fig. 1) simultaneously. For each epoch, we simulate DIQCD three times, respectively, with plain, spin-echo, and XY8 control schemes. The batch size for each simulation is 512. The time step for integration is 0.01 ms for the plain scheme, 0.05 ms for the spin-echo scheme, and 0.1 ms for the XY8 scheme. The loss function is $L = \frac{1}{7} \sum_{i=1}^7 (C_i^{\text{M,Plain}} - \zeta^{-1} C_i^{\text{D,Plain}})^2 + (1/10) \sum_{i=1}^{10} (C_i^{\text{M,Echo}} - \zeta^{-1} C_i^{\text{D,Echo}})^2 + \frac{1}{7} \sum_{i=1}^7 (C_i^{\text{M,XY8}} - \zeta^{-1} C_i^{\text{D,XY8}})^2$. The superscripts “M” and “D” indicate the model and the data, respectively. $C_i^{\text{D,Plain}}$ is the i th ($i = 1, 2, \dots, 7$) data point associated with the plain scheme. $C_i^{\text{M,Plain}}$ is the corresponding DIQCD model prediction. The meanings of $C_i^{\text{M,Echo}}$, $C_i^{\text{D,Echo}}$, $C_i^{\text{M,XY8}}$, and $C_i^{\text{D,XY8}}$ are self-evident. The DIQCD model is trained for 200 epochs, with the standard ADAM optimizer in PyTorch with a fixed learning rate of 0.1 for all parameters but the damping rates γ_x and

γ_z . The learning rate for damping rates is set to 0.001. The loss saturates after roughly 50 epochs. The final productive DIQCD model is chosen to be the model trained after all epochs. The training is done with QEpsilon and a CPU node on Perlmutter. Training with a GPU is not necessary for a small quantum system.

Appendix C: Rubrene crystal—Electron-phonon coupling: Parameters for electron-phonon coupling in rubrene were derived in Refs. [20,42] from density functional theory calculations with B3LYP functional approximation [43], the 6-31G(d) basis set, and the Gaussian 09 software [45]. Each rubrene molecule has 210 vibrational modes, with tens of them contributing to electron-phonon coupling. To reduce the complexity of the effective Hamiltonian describing electron-phonon interaction, one can keep only a few modes with significant electron-phonon coupling and renormalize their coupling constants to conserve the total reorganization energy. The effective model adopted here keeps nine modes, with frequency (ω_m) and coupling constant (g_m) listed in Table I.

Data generation: We generate our data by simulating the plain unitary quantum dynamics with a time step of 0.01 fs. We use $\phi = 0.1$ throughout the work. The total sample time for each unitary simulation is 100 fs. For each temperature ($T = 200, 250, 300, 350$, and 400 K) separately, we generate 64 trajectories of unitary evolution. Then, we calculate the mean and standard deviation on observing $\hat{\sigma}_x(t)$ and $\hat{\sigma}_y(t)$ at $t = 0, 1, \dots, 99$ fs. The simulation is done with QEpsilon and one NVIDIA A100 GPU.

Training: For each temperature separately, we train a DIQCD model with a subset of one-molecule data associated with the sampling time interval $t \in [0, 70]$ fs. The loss function is $L = \sum_{i=1}^{70} \{ \text{Mean}^{\text{M}}[\hat{\sigma}_x(i \cdot \text{fs})] - \text{Mean}^{\text{D}}[\hat{\sigma}_x(i \cdot \text{fs})] \}^2 + \{ \text{Mean}^{\text{M}}[\hat{\sigma}_y(i \cdot \text{fs})] - \text{Mean}^{\text{D}}[\hat{\sigma}_y(i \cdot \text{fs})] \}^2 + 0.1 \{ \text{STD}^{\text{M}}[\hat{\sigma}_x(i \cdot \text{fs})] - \text{STD}^{\text{D}}[\hat{\sigma}_x(i \cdot \text{fs})] \}^2 + 0.1 \{ \text{STD}^{\text{M}}[\hat{\sigma}_y(i \cdot \text{fs})] - \text{STD}^{\text{D}}[\hat{\sigma}_y(i \cdot \text{fs})] \}^2$. The superscripts “M” and “D” indicate the model and the data, respectively. “MEAN”


 FIG. 4. Mean and standard deviation (concerning the ensemble of sampled trajectories) of $\langle \hat{\sigma}_x(t) \rangle$ and $\langle \hat{\sigma}_y(t) \rangle$ from the final DIQCD models and the data associated with exact unitary simulation.

and “STD” represent mean value and standard deviation concerning the 64 trajectories, respectively. Note that the loss concerning standard deviation weighs 0.1 due to its minor regularization role. The forward evolution of DIQCD uses a batch size of 512 and a time step of 0.05 fs. Each DIQCD model is trained for 200 epochs of forward evolution, with the standard ADAM optimizer in PyTorch with a fixed learning rate of 0.3. Model parameters converge after roughly 150 epochs. The final productive

DIQCD model is chosen to be the model trained after all epochs. The training is done with QEpsilon and one NVIDIA A100 GPU. A comparison between the final DIQCD models and the data is provided in Fig. 4.

Simulation: Simulation of the DIQCD models for a molecular lattice of size $L = 150$ is done with QEpsilon and one NVIDIA A100 GPU. The batch size for forward evolution is 512, and the time step is 0.01 fs. The total sample time is 100 fs for calculating the carrier mobility.