Fewest-Switch Surface Hopping with Long Short-Term Memory Networks

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Abstract

The mixed quantum-classical dynamical simulation is essential to study nonadiabatic phenomena in photophysics and photochemistry. In recent years, many machine learning models have been developed to predict potential energy surfaces in the ground and excited electronic states effectively, which accelerates the time evolution of the nuclear subsystem. Herein, we implement long short-term memory (LSTM) networks as a propagator to accelerate the time evolution of the electronic subsystem during the fewest-switch surface hopping (FSSH) simulations. A small number of reference trajectories are generated using the original FSSH method, and then the LSTM networks can be constructed, followed by careful examination of typical LSTM-FSSH trajectories that employ the same initial condition and random numbers as the corresponding reference. The constructed network is applied to FSSH to further produce a trajectory ensemble to reveal the mechanism of nonadiabatic processes. Taking Tully’s three models as test systems, the collective results can be reproduced qualitatively. This work demonstrates that LSTM is applicable to the most popular surface hopping simulations.
Dynamical simulations governed by the laws of quantum mechanism are indispensable for providing intuitive views of photophysical and photochemical processes.\textsuperscript{1–3} Although a fully quantum treatment on both nuclear and electronic motion should be employed in principle, such a simulation is too computationally expensive for medium-sized or larger polyatomic molecules. The requirement of prior knowledge about potential energy surface (PES) further hampers its application on realistic molecular systems with high dimensionality. As an alternative, the mixed quantum-classical molecular dynamics (MQC-MD) approach provides a powerful tool to study the mechanism of photochemical reactions, including the excited-state lifetimes and the quantum yields of photoproducts on different reaction channels. During MQC-MD simulations, the nuclei are represented as classical particles but incorporate quantum feedback from the electronic motion. Many theoretical models and algorithms for MQC-MD have been developed over the past decades.\textsuperscript{4–9} The most popular method is the fewest-switch surface hopping (FSSH)\textsuperscript{10,11}, in which the nuclei travel classically on a single PES in an adiabatic electronic state with a probability of switching to any other state. The hopping probability is extracted from the time evolution of electronic degrees of freedom. Despite its simplicity for implementation and great success in many applications\textsuperscript{12–14}, the computational cost on FSSH is still much more expensive than ground-state MD, especially with the growing demand of simulations on photodynamics in long timescale.\textsuperscript{15} At each time step, electronic structure calculations on ground and excited electronic states, including potential energies, gradients as well as nonadiabatic coupling vectors (NACVs) between different states, are usually required at a high level of theory. How to accelerate dynamical simulations remains an important topic if we are willing to extend FSSH to a broader prospect.

In recent years, we have witnessed the prosperity of machine learning (ML) assisted chemical researches.\textsuperscript{16–20} Several ML techniques such as kernel ridge regression and artificial neural network have been proved as promising tools to provide a relation between molecular structure and ground-state potential energy, so-called as ML-based PES or ML-based
force field. Because of its great potential to achieve high computational accuracy and high efficiency on MD simulations simultaneously, the extension to MQC-MD is attractive and seems to be straightforward. In principle, each of adiabatic PESs can be individually fitted as the same as the proposed ML-based force fields. However, the nonadiabatic coupling vectors between adiabatic states have become the main obstacle for a fully ML-based MQC-MD simulations. One solution is based on the Landau-Zener formalism such as Zhu-Nakamura dynamics, in which the probability of surface hopping can be obtained only from adiabatic energies and gradients, without the need to compute NACVs. It has been successfully applied to ML-based nonadiabatic molecular dynamics approaches recently, but the limitation of Landau-Zener formalism as well as its influence on final simulation results is not easy to realize or control. Another way is to improve ML models to fit the reference values of NACVs. It is worthy noting that machine learning on nonadiabatic coupling is much more difficult than its energy and gradient counterparts because NACV keeps almost negligible in most regions of PES but becomes very sensitive to small changes of nuclear positions in the vicinity of conical intersections. The subtle issue on phase correction makes it more complex. The construction on a high-quality ML database is also extremely time-consuming in practice on account of multi-configuration electronic structure calculations for accurate reference values.

In the above approaches, machine learning models are developed and applied to predict potential energies, gradients and nonadiabatic coupling vectors instead of electronic structure calculations, while the time evolution of nuclei and electrons is still governed by the Newton’s second law and the time-dependent Schrödinger equation, respectively. A time series of nuclear positions, velocities and elements of electronic density matrix is obtained and updated during simulations. It suggests employing machine learning to propagate nuclear and/or electronic subsystems based on historical knowledge extracted from the preceding trajectory data. On one hand, ML techniques such as recurrent neural network (RNN) and long short-term memory (LSTM) network have been introduced to classical MD
for realistic material and protein systems to solve Newton’s equations. On the other hand, several successful ML applications on quantum dynamics have been also proposed in recent years. For example, Lan and coworkers simulated electronic evolution of multi-configuration time-dependent hartree method with LSTM; Ullah and Dral studied quantum dissipative dynamics of spin-boson model and Fenna-Matthews-Olson complex using kernel ridge regression and convolutional neural network, respectively; Hammes-Schiffer and coworkers employed artificial neural networks to solve the time-dependent Schrödinger equation and propagate the wavepacket relevant to proton transfer systems. There are also reports of deep learning in prediction of Landau-Zener transitions as well as linearized semiclassical and symmetrical quasiclassical mapping dynamics. This inspired of us that machine learning can be implemented as propagator for electronic degrees of freedom in mixed quantum-classical molecular dynamics, possibly avoiding expensive computation or difficult prediction on NACVs.

In this Letter, we will show that LSTM networks can be incorporated into FSSH to evolve the electronic density matrix. Let us first give a brief review about LSTM. Neural network models represent arbitrary functions with highly interconnected nodes processing through one or several hidden layers. The input vector $x$ of the function is provided to the nodes in input layer, and the output vector $y$ is obtained from the nodes in output layer. For a conventional $L$-layer neural network, the connection between two adjacent layers can be expressed as

$$h^{[l]} = f \left( W^{[l]} h^{[l-1]} + B^{[l]} \right)$$

where $l = 1, 2, ..., L$. In the above equation, the $l$-th layer receives $h^{[l-1]}$ from the previous layer and sends $h^{[l]}$ to the next layer, $W^{[l]}$ and $B^{[l]}$ denote weight and bias parameters for the current layer, respectively, and $f$ is the activation function such as a sigmoid or hyperbolic...
Figure 1: Structures of LSTM unit (a) and multi-layer LSTM networks (b).
tangent function. Note that $h^{[0]} = x$ for input layer and $h^{[L]} = y$ for output layer. Recurrent neural network takes this form in general but accepts a sequence as input variables. A time series $x_1, x_2, ..., x_t$ is a typical input sequence, in which $x_{t+1}$ at the next time step can be predicted using neural network as

$$h_t = f_1 (W_{hx}x_t + W_{hh}h_{t-1} + B_h) \tag{2}$$

$$y_t = f_2 (W_{yh}h_t + B_y) \tag{3}$$

where $W_{hx}, W_{hh}$ and $W_{yh}$ are weight parameters, $B_h$ and $B_y$ are bias parameters, $f_1$ and $f_2$ can be selected as the same or different activation functions, $h_t$ is the hidden state vector that implicitly involves historical information of $x$, i.e., $x_1, x_2, ..., x_{t-1}$ through $h_{t-1}$ (see Eq 2), and $x_{t+1}$ is identical to or obtained from $y_t$. The hidden state and all input vectors at previous time steps are fully-connected in RNN and prone to an exploding or vanishing gradient. The former can be solved using gradient clipping, while the latter is always addressed using long short-term memory networks. In LSTM, $h_t$ is employed to represent the short-term state, and an additional vector $c_t$ is introduced to preserve the long-term state, which is so-called as memory. As shown in Figure 1(a), a gated cell is designed to decide what to store by a forget gate $f_t$, what to read by an input gate $i_t$, and what to write by an output gate $o_t$; that is

$$f_t = \sigma (W_{fx}x_t + W_{fh}h_{t-1} + B_f) \tag{4}$$

$$i_t = \sigma (W_{ix}x_t + W_{ih}h_{t-1} + B_i) \tag{5}$$

$$o_t = \sigma (W_{ox}x_t + W_{oh}h_{t-1} + B_o) \tag{6}$$

where $\sigma$ denotes the sigmoid function. The propagation of LSTM is given by
\[
\begin{align*}
\tilde{c}_t &= \tanh (W_{cx} x_t + W_{ch} h_{t-1} + B_c) \\
c_t &= f_t \circ c_{t-1} + i_t \circ \tilde{c}_t \\
h_t &= \tanh (c_t) \circ o_t
\end{align*}
\]

Here \( \circ \) denotes the elementwise product, \( y_t \) is obtained from \( h_t \) via a dense layer using Eq 3 with a hyperbolic tangent function, and \( x_{t+1} \) is identical to or obtained from \( y_t \). Eqs 4-9 can be briefly expressed as

\[
\{h_t, c_t\} = LSTM(x_t, h_{t-1}, c_{t-1}; W, B)
\]

In this work, a multi-layer LSTM is employed and shown in Figure 1(b); that is

\[
\{h_t^{[l]}, c_t^{[l]}\} = LSTM\left(h_{t-1}^{[l]}, h_t^{[l]}, c_{t-1}^{[l]}; W^{[l]}, B^{[l]}\right)
\]

where \( l = 1, 2, 3 \), \( h_t^{[0]} = x_t \), and \( h_t^{[3]} \) is applied to the dense layer to predict \( x_{t+1} \).

Here we implemented LSTM networks into the original version of FSSH. The extension to a variety of modified surface hopping algorithms is straightforward in principle. In the surface hopping scheme, the nuclear motion is determined by the gradients on a single adiabatic PES; that is

\[
M_a \ddot{R}_a = -\nabla_{R_a} E_k
\]

where \( R_a \) and \( M_a \) denote the position and mass of nucleus \( a \), respectively, and \( k \) denotes the adiabatic electronic state at the current time step with the relevant potential energy \( E_k \). The time evolution on electronic subsystem can be expressed as
Figure 2: Workflow for LSTM-FSSH: whole procedure with three steps (top), example on generation of sequences from a reference trajectory in step 1 (middle) and implementation on dynamical simulations in step 3 (bottom).
\[ \dot{\rho}_{jk} = -\frac{i}{\hbar} \rho_{jk} (E_j - E_k) - \sum_l \dot{\mathbf{R}} \cdot (d_{jl} \rho_{lk} - \rho_{jl} d_{lk}) \]  

(13)

where \(d_{jk}\) is the nonadiabatic coupling vector between state \(j\) and \(k\), and \(\rho\) denotes the electronic density matrix. According to the assumption of the fewest number of switches, the probability of nonadiabatic transitions from the current state \(k\) to state \(j\) in a time interval \(\Delta t\) is

\[ P_{k \rightarrow j} (t, t + \Delta t) = \max \left( 0, \frac{\int_{t}^{t+\Delta t} 2\text{Re} \left( \dot{\mathbf{R}} \cdot d_{kj} \rho_{jk} \right) d\tau}{\rho_{kk}(t)} \right) \]  

(14)

For a system only involving two electronic states \(j\) and \(k\), it can be simplified as

\[ P_{k \rightarrow j} (t, t + \Delta t) = \max \left( 0, \frac{\rho_{kk}(t) - \rho_{kk}(t + \Delta t)}{\rho_{kk}(t)} \right) \]  

(15)

In presence of multiple electronic states, Eq 15 is the summation over hopping probabilities from the current state to all other states and cannot be used directly. During FSSH simulations on molecular systems, however, the hopping is only allowed when the energy gap is lower than a certain threshold, e.g., 10 kcal/mol in practice. It means that in most regions of PES, the quantum subsystem can be reduced to the simple two-state case. It is no longer reliable in the vicinity of multistate intersections, in which some advanced nonadiabatic dynamical simulations beyond FSSH are usually required.

During conventional FSSH simulations, Eqs 12 and 13 are used to govern nuclear and electronic motion, respectively, and the hopping probability is calculated with Eq 14. In LSTM-FSSH, a LSTM network is built based on a small number of existing trajectories and works as a propagator of electronic degrees of freedom, which replaces Eq 13. In other words, \(\rho(t+\Delta t)\) is predicted by LSTM based on a time series in the preceding trajectory without solving Eq 13. The hopping probability is obtained directly using Eq 15 instead of Eq 14. The time evolution on classical subsystem can be also learned by LSTM or RNN.
as reported in recent works.\textsuperscript{38,39} On the other hand, several ML-based protocols have been performed excellently to predict accurate potential energies and gradients on the right-hand side of Eq 12, which may be a better choice for realistic molecular systems.\textsuperscript{52,53} Here the classical subsystem still evolves according to Eq 12 with analytical gradients of our test model systems. More benchmarks and further methodology developments on this issue are left for future work.

The workflow of LSTM-FSSH is summarized in Figure 2. We take a one-dimensional system with two electronic states as an instance and outline the procedure as follows:

Step 1. Database construction.

(1a) Conventional FSSH simulations with a time step $\Delta t$ are performed to generate a small number of reference trajectories. The nuclear position $x$ and velocity $v$, potential energies and gradients in two states as $E_0$, $E_1$, $G_0$, $G_1$, as well as two independent elements of electronic density matrix as $\rho_{00}$ and $\rho_{01}$, are recorded at each step.

(1b) A time series $x_1, x_2, \ldots, x_N$ with $N$ frames is extracted with a time step $\Delta t_s$ from one reference trajectory. Here $x$ consists of $x$, $v$, $E_0$, $E_1$, $G_0$, $G_1$, $\rho_{00}$, $\rho_{01}$ and/or arbitrary functions of the above nuclear degrees of freedom, and $\Delta t_s$ is equal to $\Delta t$ for simplicity.

(1c) The sequence obtained in (1b) is shifted forward by $M$ frames with the same time step to extract another time series until the end of this reference trajectory.

(1d) Repeat (1b) and (1c) on other reference trajectories to extract a large number of $N$-length sequences as the database for LSTM training.

Step 2. LSTM training.

(2a) Select 80\% of reference sequences randomly from the database to build the training set. The remaining sequences belong to the testing set.

(2b) Set initial values of hyperparameters of LSTM, such as layer sizes of network and batch sizes for minimization.

(2c) Perform the training of LSTM based on the training set. The mean squared error (MSE) of $\rho_{00}$ and $\rho_{01}$ is minimized using Adam optimizer, and the MSE for the testing set
is monitored to avoid overfitting.

(2d) Perform LSTM-FSSH with the same initial condition and random numbers for hopping of selected reference trajectories. If the generated trajectory agrees with the corresponding reference trajectory, the present LSTM model is acceptable. Otherwise, return to (2c) to rebuild LSTM using different hyperparameters and random numbers for training.

Step 3. FSSH simulation.

(3a) Initialize the nuclear position, velocity and electronic density matrix for one trajectory as the same as conventional FSSH simulation without machine learning.

(3b) Perform conventional FSSH simulation using Eqs 12, 13 and 14 for $N$ steps to produce an $N$-length sequence.

(3c) The generated sequence is provided into LSTM to predict electronic density matrix in the next step. Note that the dimensionalities of input and output features of LSTM can be different. Take our test case as an instance. The output layer usually has three nodes as $\rho_{00}$, $\text{Re}(\rho_{01})$ and $\text{Im}(\rho_{01})$, while the input layer includes all nuclear and electronic features.

(3d) The nuclear position and velocity are propagated to the next step using Eq 12, followed by the calculation on potential energies and gradients in all electronic states. The hopping probability between electronic states is obtained using Eq 15, and the nuclear velocity is adjusted if a switch occurs. This step is the same as conventional FSSH simulation without machine learning, except for the calculation on hopping probability.

(3e) Shift the $N$-length sequence forward by one step. Now the electronic and nuclear features updated in (3c) and (3d) is added into the sequence.

(3f) Return to (3c) to propagate the trajectory until the stopping criterion such as the maximum MD step is achieved.

(3g) Repeat (3a)-(3f) to produce a large number of trajectories. Study collective photodynamic behavior of the system based on trajectory ensemble.

It is worthy noting that step (2d) is the most essential during the whole procedure. Unlike the common case of ML training, step (2c) converges very quickly and seems to be insensitive
to hyperparameters in our test case. It indicates that the MSE for the testing set may be insufficient to reflect actual performance of LSTM on dynamical simulations. Actually, it is observed that a set of LSTM networks, all of which perform excellently in (2c), usually produce diverse FSSH trajectories with the same initial condition and random numbers. It may be due to the cumulative error of ML propagator after a long-time ML-driven dynamical simulation. The implementation in step (2d) is thus required. In this work, we compare the time evolution of electronic density matrix visually as a criterion of consistency between reference and LSTM-FSSH trajectories. The hyperparameters $M$ and $N$ in step 1 are also tuned, leading to a reconstructed database. In practice, the input features as well as the arbitrary functions mentioned in step (1b) may be even necessary to select again before returning to (2c). However, the additional computational cost is small because the reference FSSH trajectories have been produced in step (1a) and keep the same regardless of database reconstruction or input feature reselection.

Figure 3: Adiabatic PESs and nonadiabatic coupling vectors of the single-avoided crossing model (a), the dual-avoided crossing model (b), and the extended coupling Hamiltonian model (c). All quantities are in atomic units.

We employ Tully’s three models reported in the original FSSH paper\textsuperscript{10}: the single-avoided crossing model, the dual-avoided crossing model, and the extended coupling model as our test systems, which are supposed to cover most of the typical features in realistic molecular systems. The reference trajectories as well as the final collective results for comparison are obtained using the original FSSH method despite its well-known limitations such as...
the overcoherence problem.\textsuperscript{54–57} All parameters of Tully’s models are in atomic units. All trajectories start at \(x = -20.0\) on the lower surface with an initial momentum to the right and stop at \(x = \pm 25.0\). The time step \(\Delta t = 1.0\) for all simulations, and \(\Delta t_s\) for LSTM is equal to \(\Delta t\). The mass in Eq 12 is set as 2000 to mimic atomic nuclei. For each model and each method, 2000 trajectories are simulated with each initial momentum for collection. The training of all LSTM networks are implemented using Keras\textsuperscript{58} combined with Tensorflow\textsuperscript{59}. The Interface between FSSH propagator and LSTM networks are built using Keras2C\textsuperscript{60}.

The diabatic Hamiltonian of the single-avoided crossing model is given as

\[
V_{11}(x) = \begin{cases} 
A [1 - e^{-Bx}] & x > 0 \\
-A [1 - e^{Bx}] & x < 0 
\end{cases}
\]

\[
V_{22}(x) = -V_{11}(x)
\]

\[
V_{12}(x) = V_{21}(x) = Ce^{-Dx^2}
\]

where \(A = 0.01\), \(B = 1.6\), \(C = 0.005\), and \(D = 1.0\). The corresponding adiabatic PESs and NACVs are shown in Figure 3(a). We generate 20 reference trajectories without machine learning for each initial momentum. The sequences are extracted from all reference trajectories with \(M = 50\) and \(N = 100\), leading to 96235 sequences in total. The classical position \(x\), momentum \(p\), potential energies \(E_0\) and \(E_1\), as well as elements of electronic density matrix are employed as input features. The number of nodes for each LSTM layer and the dense layer are set as 32. The batch size for minimization is 64 with a learning rate as 0.001.

The excellent performance for the testing set in step (2c) is shown in Figure S1. As mentioned in step (2d), LSTM-FSSH simulations are implemented to produce three representative trajectories with different initial momenta. The time evolution of \(\rho_{00}\), \(\Re(\rho_{01})\) and \(\Im(\rho_{01})\) is compared to the corresponding reference trajectories with the same initial condition and random numbers for hopping (see Figure 4). At a low momentum of 5.0,
LSTM-FSSH keeps consistent with the reference for 12500 steps. The deviation takes place after the system departs from the coupling region, which will not affect collective results. At a medium momentum of 15.0 and a large momentum of 30.0, LSTM-FSSH agrees well with the reference except for a slightly fluctuation of $\rho_{00}$ after a sudden decay. The hopping event is observed only at a large momentum. The system switches to the upper surface at step 1882 and 1339 with and without LSTM, respectively. The difference probably originates from the cumulative error. Then we run 2000 trajectories with each initial momentum (60000 in total) using LSTM-FSSH. As shown in Figure 7, the collective result is almost the same as the original FSSH simulations. The resonance phenomenon at $7.0 < p < 9.0$ populated on R1 can be partially captured by machine learning with a smaller probability.

The diabatic Hamiltonian of the dual-avoided crossing model is

$$
V_{11}(x) = 0 \\
V_{22}(x) = -Ae^{-Bx^2} + E \\
V_{12}(x) = V_{21}(x) = Ce^{-Dx^2}
$$

(17)

where $A = 0.10$, $B = 0.28$, $C = 0.015$, $D = 0.06$, and $E = 0.05$. The adiabatic PESs and NACVs are shown in Figure 3(b). For each initial momentum, there are 50 reference trajectories generated without machine learning. The sequences are extracted with $M = 20$ and $N = 50$ to build the database with 355776 sequences in total. The input features are selected as the same as the first model. The number of nodes for each LSTM layer and the dense layer are set as 48. The batch size for minimization is 32 with a learning rate as 0.001.

First, it can be seen from Figure S1 that the performance for the testing set is remarkable. Second, three representative trajectories using LSTM-FSSH simulations are compared. As shown in Figure 5, the higher the initial momentum, the better the performance of machine learning. At a low momentum of $\ln(E_0) = -3.0$, LSTM-FSSH deviates away from the reference after step 3000. Both trajectories switch to the upper surface at step 2949, while
Figure 4: Electronic density matrix plotted as function of time for representative trajectories of single-avoided crossing model at a low (a), medium (b) and high (c) initial momentum. Different colors represent different simulation methods (black: original FSSH as reference; red: LSTM-FSSH).
an additional hopping to the lower surface occurs at step 4257 using LSTM. At a medium momentum of \( \ln(E_0) = -1.5 \), two trajectories share the same hopping event at step 1424. At a high momentum of \( \ln(E_0) = -0.5 \), the transition between two surfaces is never observed in the representative trajectories using LSTM, while the original FSSH simulation predicts an excited-state population lasting for a short time around step 800. Finally, 2000 LSTM-FSSH trajectories with each initial momentum (74000 in total) are implemented. The collective result shown in Figure 7 is consistent with the performance on representative trajectories. Our method gives a qualitatively correct result except an error within \(-3.0 < \ln(E_0) < -2.0\) where the increase of T2 population is missing. It is probably due to the relatively small number of T2 trajectories in our training set, but manually picking trajectories with a specified population is always unrealistic for polyatomic molecules.

The simulation results using another LSTM model with different hyperparameters are reported in Figures S1, S5 and S6. No problem can be observed from the MSE of the testing set in step (2c). However, the FSSH simulations with and without machine learning in step (2d) represent opposite tendencies of the time evolution of \( \rho_{00} \). At a low momentum, \( \rho_{00} \) keeps around 1.0 in the reference but decreases to 0.5 using LSTM after a long-time dynamics. At a medium momentum, \( \rho_{00} \) converges to 0.3 in the reference but returns to 0.8 using LSTM. It indicates large cumulative errors of this inappropriate LSTM model, leading to an incorrect collective result.

The diabatic Hamiltonian of the extended coupling model is

\[
\begin{align*}
V_{11}(x) &= A \\
V_{22}(x) &= -A \\
V_{12}(x) = V_{21}(x) &= \begin{cases} 
B \left[2 - e^{-Cx}\right] & x > 0 \\
Be^{Cx} & x < 0
\end{cases}
\end{align*}
\]

where \( A = 0.0006 \), \( B = 0.1 \), and \( C = 0.9 \). The adiabatic PESs and NACVs are shown.
Figure 5: Electronic density matrix plotted as function of time for representative trajectories of dual-avoided crossing model at a low (a), medium (b) and high (c) initial momentum. Different colors represent different simulation methods (black: original FSSH as reference; red: LSTM-FSSH).

\[ \ln(E_0) = -3.0 \]  
\[ \ln(E_0) = -1.5 \]  
\[ \ln(E_0) = -0.5 \]
in Figure 3(c). Note again that the original FSSH simulation results are employed as the reference of machine learning, and the decoherence correction, which is necessary for this model to reproduce its full-quantum dynamics behavior, is out of our scope in this work. We first implement FSSH simulations without machine learning to produce 50 reference trajectories at each initial momentum. However, all attempts on LSTM training with the above input features are unsuccessful in step (2d). As shown in Figure S7, the decrease of $\rho_{00}$ appears too early using LSTM; LSTM also fails to reproduce the strong oscillation of $\rho_{01}$, which is relevant to the coherence in the coupling region. Unsurprisingly, the collective result deviates away from the reference (see Figure S8).

The coupling between quantum and classical subsystems is the key factor of MQC-MD, which inspired of us that it may also play an important role on our problem. The refined selection on input features related to nuclear degrees of freedom is expected to improve the performance of LSTM on the prediction of electronic motion. We select the classical position $x$, momentum $p$, a function of nuclear degrees of freedom as $\frac{(G_k - G_j)\vert v \vert}{(E_k - E_j)^r}$ where $k$ denotes the current state and $j$ denotes another state, as well as $\rho_{00}$ are selected as input features. Totally 165693 sequences are extracted from all reference trajectories with $M = 50$ and $N = 100$. The number of nodes for each LSTM layer and the dense layer are set as 48. The batch size for minimization is 64 with a learning rate as 0.001. As shown in Figure S2, the time evolution of $\rho_{00}$ for three representative trajectories agree well with the reference. The collective results at low and high momenta match the original FSSH simulation, but the alternation of R1 and R2 populations at medium momenta disappears in LSTM-FSSH (see Figure S3). Here the off-diagonal elements of electronic density matrix are ignored. Although the hopping probability can be obtained in absence of $\rho_{01}$ using Eq 15, the phase of this term may strongly affect the population of reflection channels and should be included in machine learning models. However, the introduction of Re ($\rho_{01}$) and Im ($\rho_{01}$) to the current LSTM network fails in step (2d) after repeating attempts (see Figures S9 and S10).

We further construct multiple connected LSTM networks to enhance the accuracy of
Figure 6: Electronic density matrix plotted as function of time for representative trajectories of extended coupling model at a low (a), medium (b) and high (c) initial momentum. Different colors represent different simulation methods (black: original FSSH as reference; red: LSTM-FSSH). Switches between two connected LSTM networks are represented by vertical lines (orange: $1 \rightarrow 2$; green: $2 \rightarrow 3$; blue: $3 \rightarrow 4$).
LSTM-FSSH dynamical simulations. The first LSTM network is trained with 191903 sequences ($M = 25$ and $N = 50$) and a batch size of 32. When the trajectory starts at $x = -20.0$, it is employed until the system first reaches $x = -2.0$. The second LSTM is built using 35655 sequences ($M = 25$ and $N = 50$) and a batch size of 16. It follows the first network and continues to be employed until the system first reaches $x = 1.0$ or returns to $-2.0$. The third LSTM is constructed based on 175480 sequences ($M = 3$ and $N = 50$) and a batch size of 32. It is applied sequentially until the system first reaches $x = 4.0$ or returns to $-7.0$. The last LSTM is built with 471559 sequences ($M = 5$ and $N = 50$) and a batch size of 64. It is responsible when $x \geq 4.0$ or $\leq -7.0$. In addition, the velocity may reverse when $x \leq -2.0$ or $\geq 4.0$ at a very low or high momentum, respectively. In the former case, the first LSTM would switch to the third one immediately; in the latter case, the last LSTM would change to the third one when the system returns to $x = 4.0$. The third network would be kept until $x \leq -7.0$ in both cases. Examples are displayed in Figure S4. The classical position $x$, momentum $p$, potential energies $E_0$ and $E_1$, $\left(\frac{G_k-G_j}{(E_k-E_j)^2}\right)$, as well as $\rho_00$, Re($\rho_{01}$) and Im($\rho_{01}$) are selected as input features of all networks. The number of nodes for each layer is 72. The learning rate keeps as 0.001.

The comparison between three representative LSTM-FSSH trajectories and their corresponding references is shown in Figure 6. The multiple networks perform much better than our previous attempts using a single LSTM. The time evolution of $\rho_00$, Re($\rho_{01}$) and Im($\rho_{01}$) agrees with the reference except for a problem at a low momentum of 5.0, where $\rho_{00}$ descends too early and tends to oscillate using LSTM. Hopping events are observed only at a medium momentum of 15.0 with both methods, taking place at the same step as 1927. The collective result shown in Figure 7 is obtained from 2000 LSTM-FSSH trajectories with each initial momentum (60000 in total). LSTM-FSSH is qualitatively identical to the original FSSH simulations without machine learning. In addition, the MSEs for the testing sets with all LSTM models mentioned above are indistinguishable in Figure S1 despite different collective results, which highlights the importance of step (2d) on LSTM training.
Although the improvement of multiple LSTM networks is remarkable, a single LSTM is still recommended as the first choice on a new system. A close examination on representative trajectories indicates that the discrepancy between energy degeneration and strong coupling regions may lead to the difficulties on the extended coupling model. According to our experience, however, it is not frequent for realistic photochemical reactions.

Figure 7: Collective results of single-avoided crossing (a), dual-avoided crossing (b), and extended coupling models (c) obtained using original FSSH as reference (top) or LSTM-FSSH (bottom).

The application of this method on realistic molecular systems is attractive, but some nontrivial issues should be addressed in prior. First, although NACV is no longer necessary for two-state systems, the excited-state calculation on potential energies and gradients is still a bottleneck for medium-sized or larger molecules. Fortunately, ML-based force fields have been successfully applied to dynamical simulations on multiple PESs of several organic molecules in photochemistry.\textsuperscript{31,32,61–63} The combination of ML-based force fields and LSTM-FSSH is straightforward, in which the term on the right-hand side of Eq 12 would be obtained from ML prediction instead of electronic structure calculation. Second, the input
feature selection is more complicated for high-dimensional classical subsystem. Our preliminary results show that a large number of input features related to nuclear motion hampers the ability of LSTM to propagate electronic subsystem. The encoder network that generates a low-order latent space may be a promising choice to extract more concentrated input features. The use of encoder in LSTM-FSSH would be simpler than latent-space classical dynamics because the time evolution on nuclei is still implemented in the full-dimensional space. Third, a quantitative criterion in step (2d) is absent. Here we compare the representative LSTM-FSSH trajectories with the reference visually, which would be system-specific and laborious for realistic molecules. ML-based analysis of MQC-MD dynamics such as isometric feature mapping and multidimensional scaling provides a possible solution to this trouble. Finally, how to extend this method to study the dynamics around multistate intersections is unsolved in this work. We suggest making a pause for ML-driven simulation and switching to a more accurate MQC-MD approach when the system evolves in multistate intersection regions.

In summary, we have demonstrated how to construct long short-term memory networks as a propagator of electronic subsystem in fewest-switch surface hopping simulations. The single-avoided crossing, dual-avoided crossing and extended coupling models are employed as our test systems. Starting with a small number of full-length trajectories (20-50 for each initial momentum and each model) from the original FSSH simulations, LSTM networks can be built to reproduce the time evolution of electronic density matrix during a long-time FSSH dynamical simulation in presence of only 50-100 steps given as the initial input sequence. Then we simulate 2000 trajectories for each initial momentum and each model using LSTM-FSSH. The collective results is qualitatively consistent with the original FSSH as reference, even though the extended coupling model is much more difficult than other test cases and requires multiple connected LSTM networks. All results show that a lower initial momentum results in a longer-time trajectory with larger cumulative errors, which is an intrinsic drawback of LSTM propagators. Besides avoiding expensive computation on
nonadiabatic coupling vectors at least for two-state systems, the combination of LSTM and ML-based force field has a great potential to further accelerate surface hopping simulations on realistic molecular systems. We believe that LSTM is a powerful tool on mixed quantum-classical molecular dynamics to study photophysics and photochemistry more effectively.

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**Supplementary Information**

Electronic supplementary information (ESI) including performance of LSTM networks on testing sets, details on LSTM networks for extended coupling model and LSTM networks with bad performance is available.

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Supplementary Information for:
Fewest-Switch Surface Hopping with Long
Short-Term Memory Networks

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Performance of LSTM networks on testing sets

Figure S1: Comparison of LSTM predictions of electronic density matrix with reference for testing sets: single-avoided crossing model in the main text (a), dual-avoided crossing model in the main text (b), extended coupling model with multiple networks in the main text (c-f), dual-avoided crossing model with bad performance in Fig. S5 (g), extended coupling model with bad performance in Figs. S7 (h) and S9 (i), and extended coupling model with moderate performance in Fig. S2 (j). All values of $\rho_{00}$ are shifted by $-0.5$. 
Details on LSTM networks for extended coupling model

Figure S2: Electronic density matrix plotted as function of time for representative trajectories of extended coupling model at a low (a), medium (b) and high (c) initial momentum. LSTM network is constructed with input features as $x, p, |(G_k - G_j)|v|, (E_k - E_j)^2$ and $\rho_{00}$; all hyperparameters are listed in the main text. Different colors represent different simulation methods (black: original FSSH as reference; red: LSTM-FSSH).
Figure S3: Collective result of extended coupling model obtained using LSTM in Fig. S2 in comparison with reference.
Figure S4: Multiple connected LSTM networks (labeled as 1 in blue, 2 in green, 3 in orange, and 4 in red) are applied and switched during propagation on extended coupling model. Different initial momenta or random surface hopping events lead to different cases represented in (a-f).
LSTM networks with bad performance

Figure S5: Electronic density matrix plotted as function of time for representative trajectories of dual-avoided crossing model at a low (a), medium (b) and high (c) initial momentum. LSTM is built with the number of nodes for each layer as 32; other hyperparameters are the same as that used in the main text. Different colors represent different simulation methods (black: original FSSH as reference; red: LSTM-FSSH).
Figure S6: Collective result of dual-avoided crossing model obtained using LSTM in Fig. S5 in comparison with reference.
Figure S7: Electronic density matrix plotted as function of time for representative trajectories of extended coupling model at a low (a), medium (b) and high (c) initial momentum. LSTM is built with the same input features as the first and second test systems; 422341 sequences in database are extracted with $M = 20$ and $N = 50$; the number of nodes for each layer is 72 with a batch size of 64 and a learning rate of 0.001. Different colors represent different simulation methods (black: original FSSH as reference; red: LSTM-FSSH).
Figure S8: Collective result of extended coupling model obtained using LSTM in Fig. S7 in comparison with reference.
Figure S9: Electronic density matrix plotted as function of time for representative trajectories of extended coupling model at a low (a), medium (b) and high (c) initial momentum. LSTM is built with input features as \(x, p, \frac{(G_k - G_j)|v|}{(E_k - E_j)^2}, \rho_{00}, \text{Re}(\rho_{01})\) and \(\text{Im}(\rho_{01})\); 417096 sequences in database are extracted with \(M = 20\) and \(N = 50\); the number of nodes for each layer is 72 with a batch size of 32 and a learning rate of 0.001. Different colors represent different simulation methods (black: original FSSH as reference; red: LSTM-FSSH).
Figure S10: Collective result of extended coupling model obtained using LSTM in Fig. S9 in comparison with reference.