Supplementary Information

Forecasting of In Situ Electron Energy Loss Spectroscopy

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1 Optimized Hyperparameters

Table 1: Optimized hyperparameters for the EELSTM model.

Hyperparameter	Search Space	Optimal Value
Learning Rate	$[\cdot 10^{-6}, \cdot 10^{-1}]$	$\cdot 10^{-4}$
LSTM Layers	[1,4]	3
Nodes in Layer	[64,512]	256
Dropout	[0.01,0.3]	0.2
Input Window	[3,15]	8
Batch Size	[20,100]	40

2 Autocorrelation Analysis

In some cases, the last input in a sequence is a better predictor of future sequences than an LSTM model. In light of this, autocorrelation calculations between the EELS spectra inform how many timesteps in advance must be predicted to ensure actionable predictions. Supplementary Figure 1 shows the result of the autocorrelation calculation from the dataset shown in the main text. The shaded area represents number of input window timesteps where the autocorrelation value falls into an acceptable range, whereas the blue dots represent the autocorrelation values for each input timestep. The smaller the autocorrelation value, the less likely that the model copies seasonal trends into its prediction and the more accurate the prediction will be. As shown in this figure, the minimum preferred number of input timestep is 6 for the EELSTM model, and the autocorrelation value converges to zero starting at 10 timesteps. This finding also verifies that our data are not subject to explicit autocorrelation.

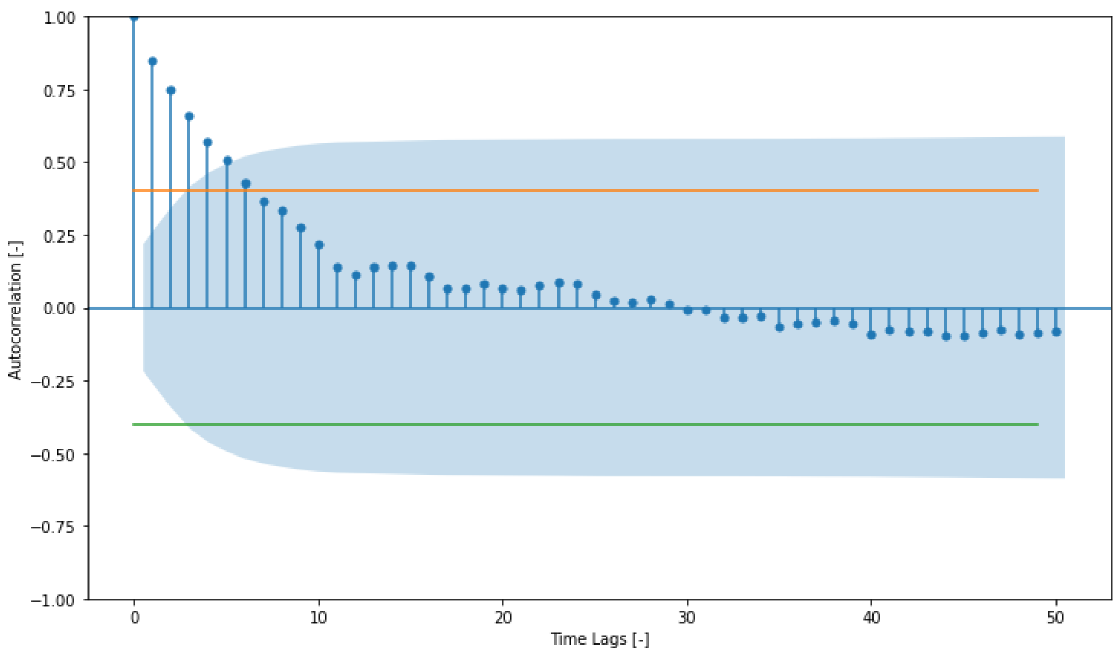

Figure 1: **Autocorrelation analysis of the dataset shown in the main text.** The shaded area represents the 95 % confidence interval. The horizontal lines depict an autocorrelation of ± 0.4, which is the preferred maximum range for our model. Both the autocorrelation and the time axes are unitless.

3 Graphical User Interface

The model described in the main text is linked to a graphical user interface (GUI) built using the Python Flask web framework, shown in Supplementary Figure 2. This interface provides real-time visualization of raw data and model forecasts. The GUI includes four steps: **Data Import**, **Spectrum Visualization and Model Selection**, **Forecast Display**, and **Forecast History**. First the user selects an appropriate EELS spectrum image (SI) data in the native Gatan *.dm4 format. This data should contain the EELS datacube in a time series. The backend of the GUI uses HyperSpy to parse the individual EELS spectra. After selecting the spectrum, the user is sent to a dialog where they can visualize the SI time series dynamically via a slider bar. They can also input the dwell time and model parameters. After submitting the desired parameters, the user is presented with a forecast, input windows, ground truth data, and MSE. Finally, the user may access a history of prior forecasts, allowing them to easily compare to previous spectra and results.

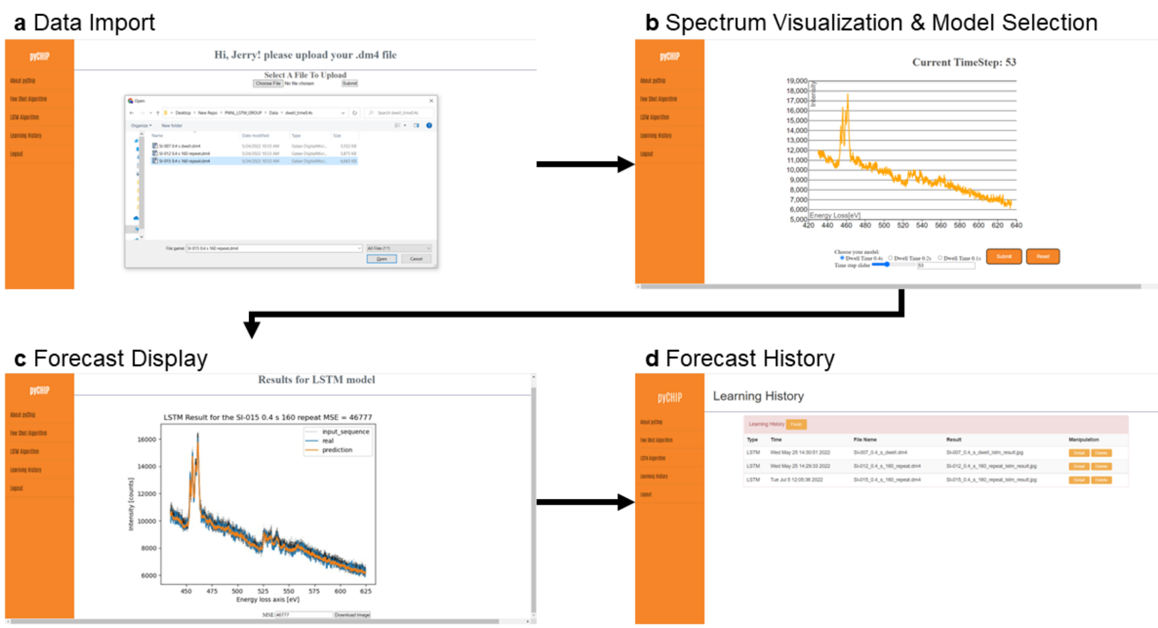

Figure 2: **Graphical user interface (GUI) for interacting with the EELSTM model.** (a–d) Usage steps include Data Import, Spectrum Visualization and Model Selection, Forecast Display, and Forecast History, respectively.