Supporting Information: Robustness and Accuracy Improvement of Data Processing with 2D Neural Network for Transient Absorption Dynamics

Ruixuan Zhao[a, ‡], Daxin Wu[a, ‡], Jiao Wen[b], Qi Zhang[c], Guanglei Zhang[a], Jiebo Li[a*]

[a] *Institute of Medical Photonics, Beijing Advanced Innovation Center for Biomedical Engineering, School of Biological Science and Medical Engineering, Beihang University, Beijing 100191, P.R. China*

[b] *School of Materials Science and Engineering Beihang University Beijing 100191, China*

[c] *State Key Laboratory of Molecular Reaction Dynamics Dalian Institute of the Chemical Physics Chinese Academy of Sciences Dalian, Liaoning 116023, China*

* jiebo39@buaa.edu.cn;

‡ These authors contribute equally to this work.

Including Fig. S1-3.

S1 Training set generation for 1DCNN

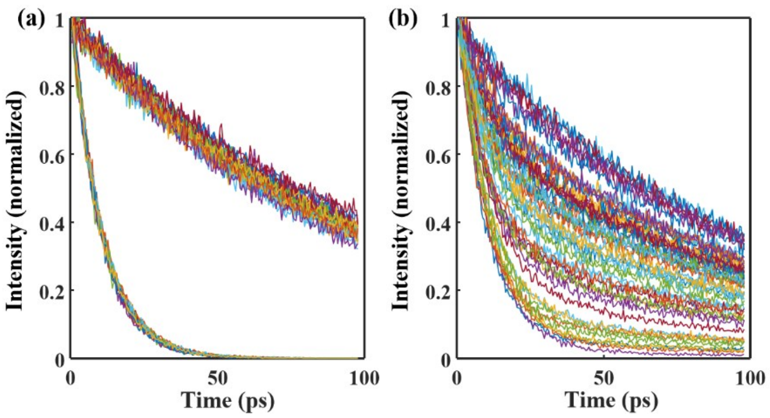
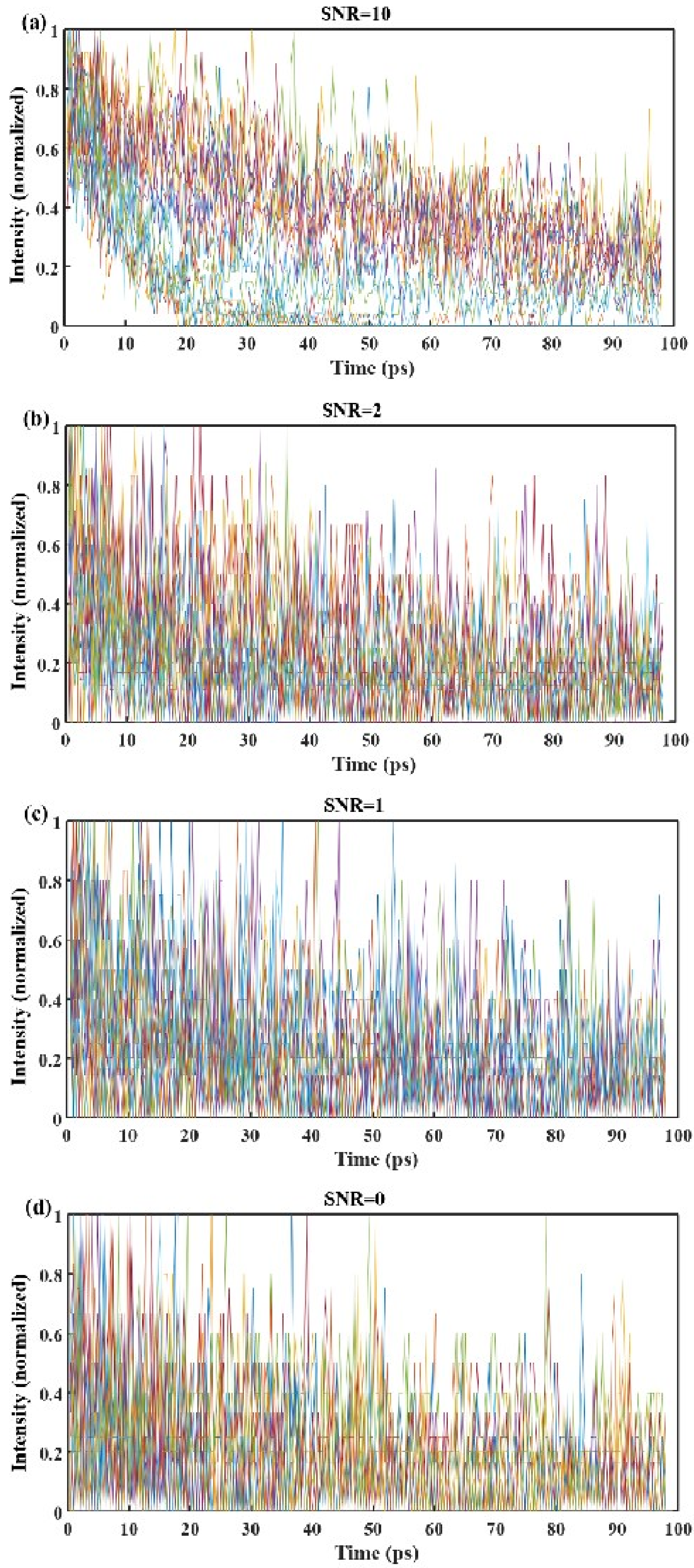


Fig. S1 (a)Generated single exponential training sample for 1DCNN. (b)Generated bi-exponential training sample for 1DCNN.

To obtain well trained model and achieve effective classification of exponential form for decay process, both single exponential decay and bi-exponential decay (Fig. S1(b)) were simulated in the form of Eq. 8 with addition of Poisson noise. Overall, 150 sets of spectrum were simulated in which each spectrum had 100 wavelength units. Furthermore, to promise the balance of different exponential forms in a training sample, different exponential decay was generated randomly (50% bi-exponential decay, 50% single-exponential decay) in a spectrum and the lifetime distribution was also determined randomly (μ = 10 ps or μ = 100 ps) in a single exponential decay (Fig. S1(a)). Specifically, in a bi-exponential decay, the amplitude fraction in Eq. 8 was set as a random number ranging from 0 to 1 so that effective sampling between two lifetime distribution could be achieved and the training effectiveness could be promised.

S2 Training set generation for highly noised data



(a)
SNR=10
Intensity (normalized)
Time (ps)
(b)
SNR=2
Intensity (normalized)
Time (ps)
(c)
SNR=1
Intensity (normalized)
Time (ps)
(d)
SNR=0
Intensity (normalized)
Time (ps)

Fig. S2 Generated highly noised data sample with (a) SNR = 10, (b) SNR = 2, (c) SNR = 1, (d) SNR = 0.

First, to characterize the noise background, signal to noise ratio (SNR) was defined as $10lg\frac{P_{signal}}{P_{noise}}$ (dB), and P_{signal} was calculated from $1/N\sum_{i=1}^{N}x[n]^2$, where x[n] stands for sampling points in exponential decay without noised background, so as the calculation process of $P_{noise.}$ As shown in Fig. S2, to test the robustness of our algorithm under highly noised data, 150 sets of spectrum were simulated respectively with different SNR (SNR = 0, SNR = 1, SNR = 2, SNR = 10). Furthermore, all the decay curves were simulated using Eq. 8 in biexponential form with life time in different distributions(t_1: μ = 10 ps σ = 0.5, t_2:μ = 100 ps, σ = 5, A_1: μ= 0.2, σ = 0.1), and the amplitude fraction A_1 was set as a random number ranging from 0 to 1 to promise impartial sampling.

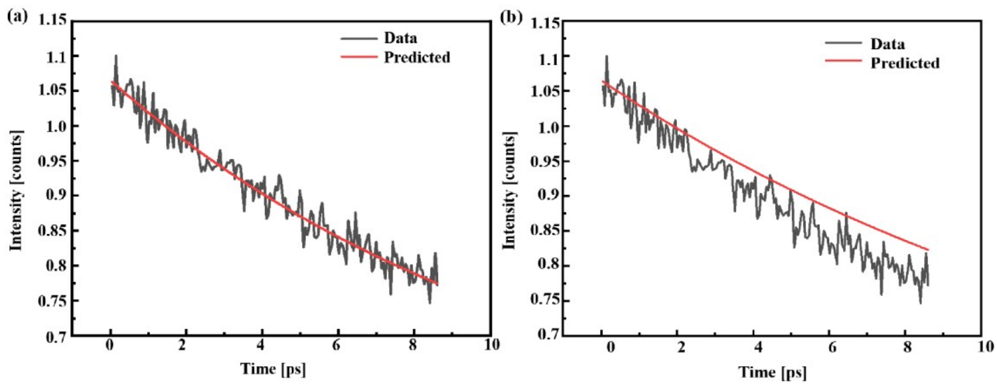


Fig. S3 Generated exponential decay and the prediction of lasso with different resolution ω (a) 3.5, (b) 3.6.

During exponential sampling process characterized by $\Gamma_k = \Gamma_0 \exp(k\pi/\omega)$, even Γ_0 could be fixed according to the limitation of detecting range of experimental instrument for specific task, the determination of resolution omega could still be an onerous task. As shown in Fig. S3(a), Γ_0 was set to 0.001 and omega was set to 3.5, as comparison, Γ_0, omega was set to 0.001 and 3.6 in Fig. S3(b). We could obviously differentiate that omega = 3.5 enables better fitting to the simulated decay curve. However, there only exists 1% discrepancy in ω1 and ω2, which indicates that omega possesses a relatively rugged solution space and it is arduous to search the reasonable resolution artificially.