

Supplementary Materials

Near-Infrared Synaptic Responses of WSe₂ Artificial Synapse Based on Upconversion Luminescence from Lanthanide Doped Nanoparticles

Yaxian Lu, Chuanwen Chen, Qi Sun, Ni Zhang, Kun Lv, Zhiling Chen, Yuelan He, Haowen Tang and Ping Chen *

Center on Nano-Energy Research, Guangxi Key Laboratory for Relativistic Astrophysics,
School of Physical Science and Technology, Guangxi University, Nanning 530004, China

* Correspondence: chenping@gxu.edu.cn

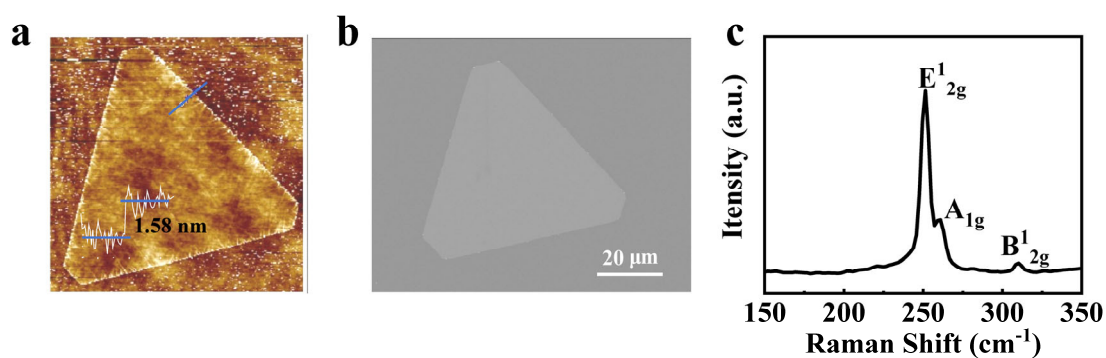


Figure S1. Characterization of 2D WSe₂ Materials. (a) AFM and (b) SEM images of 2D WSe₂. (c) Raman spectrum of WSe₂, excited by 532 nm.

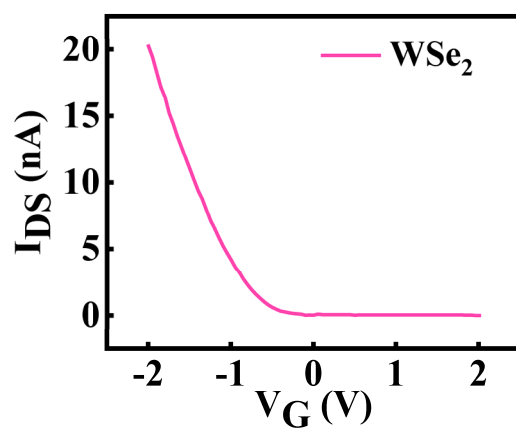


Figure S2. The transfer characteristic curve of WSe₂ with V_{DS} at 0.5 V.

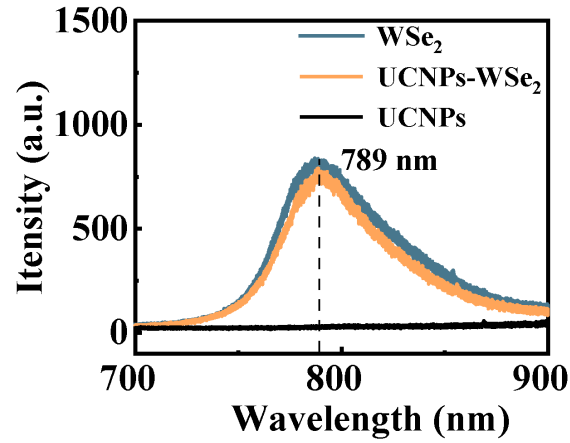


Figure S3. Characterisation of PL spectra. (a) WSe₂. (b) UCNPs-WSe₂. (c) UCNPs. Excited by 532 nm.

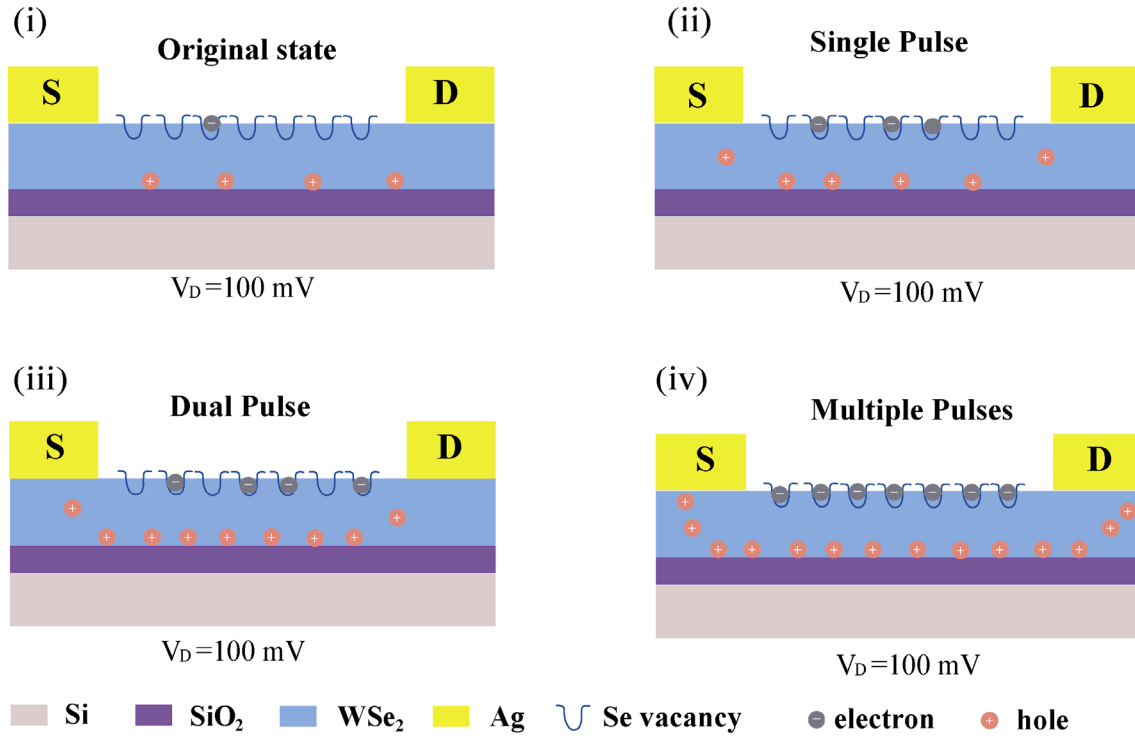


Figure S4. Working mechanism of UCNPs-WSe₂ device. Se vacancies capture electron situation under (a) initial state (b) single pulse (c) double pulse (d) multiple pulse irradiation.

Mechanism of optical energy transfer to the electrical synaptic plasticity

Original state: In the dark condition, the carrier concentration of WSe₂ is relatively low. When a bias voltage of 100 mV is applied, due to the limited number of mobile holes and electrons, only a weak dark current can be generated (Figure S4. (i)).

EPSC: When a 975 nm laser pulse is applied, Yb³⁺ absorbs effectively a 975 nm photon from ground state ²F_{7/2} to excited state ²F_{5/2}, then energy transfer to excited state ³H₅ of Tm³⁺, followed by relaxation to ³F₄ state by non-radiative relaxation. And ³F₄ state of Tm³⁺ will be pumped to ³F_{2,3} excited states after transferring

the second photon energy from Yb^{3+} . Thereby, $^1\text{G}_4$ and $^1\text{D}_2$ states will be generated after energy transfer the third and fourth photon from Yb^{3+} . And radiative transitions from $^1\text{G}_4$ and $^1\text{D}_2$ states give visible luminescence around 452 nm, 476 nm, 648 nm with $^1\text{D}_2 \rightarrow ^3\text{F}_4$, $^1\text{G}_4 \rightarrow ^3\text{H}_6$, $^1\text{G}_4 \rightarrow ^3\text{F}_4$ transitions, respectively[1]. WSe_2 reabsorbs these visible photons, leading to increase in the carrier concentrations within WSe_2 , thereby generating EPSC (Figure S4. (ii)) [2].

PPF: In the PPF process, the first 975 nm laser pulse triggers EPSC. Due to the electrons captured by Se vacancies not being fully released, the current fails to return to the initial level[3] (Figure S4 (iii)). When a second 975 nm laser pulse is applied, WSe_2 reabsorbs the visible light emitted by Tm^{3+} , promoting a further increase in carrier concentrations. This leads to a significant increase in the second pulse's photocurrent compared to the first. The process triggered by the successive action of two 975 nm laser pulses forms PPF.

LTP: On the basis of PPF, if 975 nm laser pulses are continuously applied, the carrier concentrations in WSe_2 will increase with increasing the number of pulses, which leads to a simultaneous elevation of the conductance. At this time, the increase in carrier concentrations results in a significant increase in the number of trapped electrons. After the excitation of the pulses, the current of the device recovers to the original state slowly because the trapped electrons are not fully released (Figure S4. (iv)). The sustained interplay of these processes ultimately gives rise to LTP.

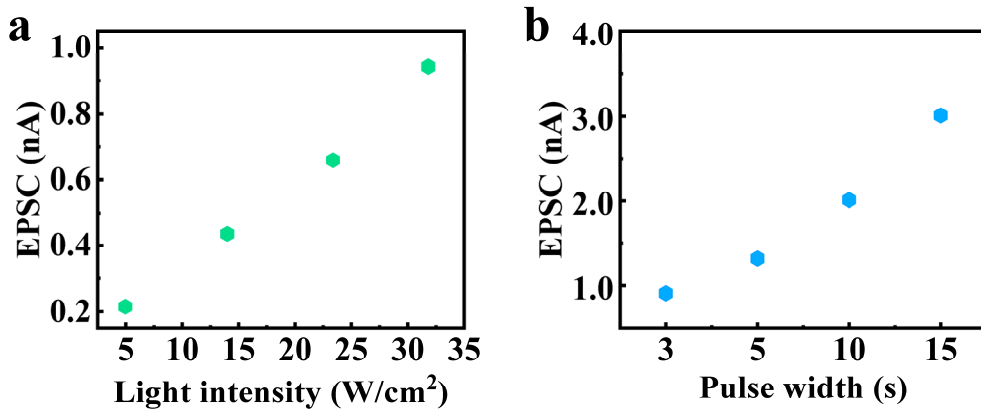


Figure S5. Extraction results of EPSC peaks of UCNPs- WSe_2 synaptic device under 975 nm excitation with (a) different power densities and (b) pulse widths.

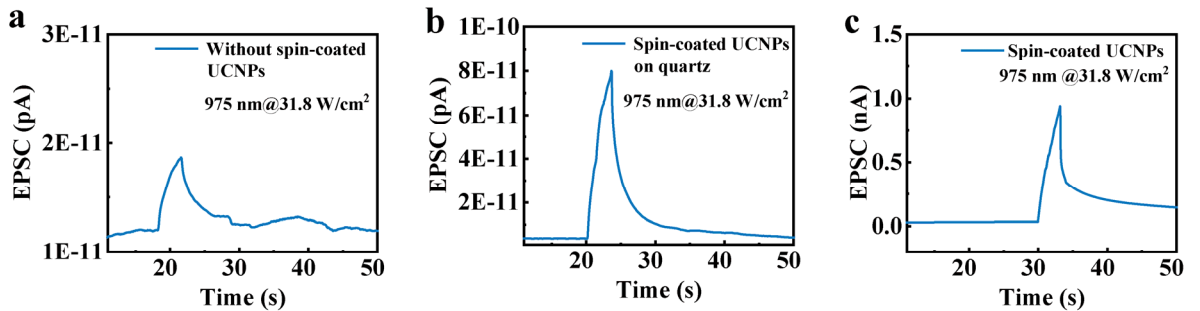


Figure S6. EPSC generated by WSe_2 synaptic device with (a) Without spin-coated UCNPs. (b) Spin-coated UCNPs on quartz. (c) Spin-coated UCNPs.

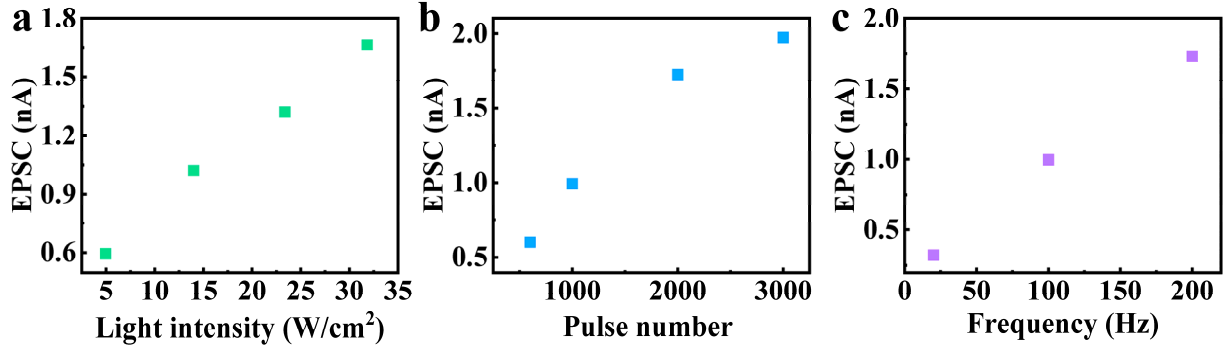


Figure S7. Extracted EPSC of UCNPs-WSe₂ synaptic device under 975 nm illumination with (a) different power densities, (b) pulse widths and (c) pulse frequencies.

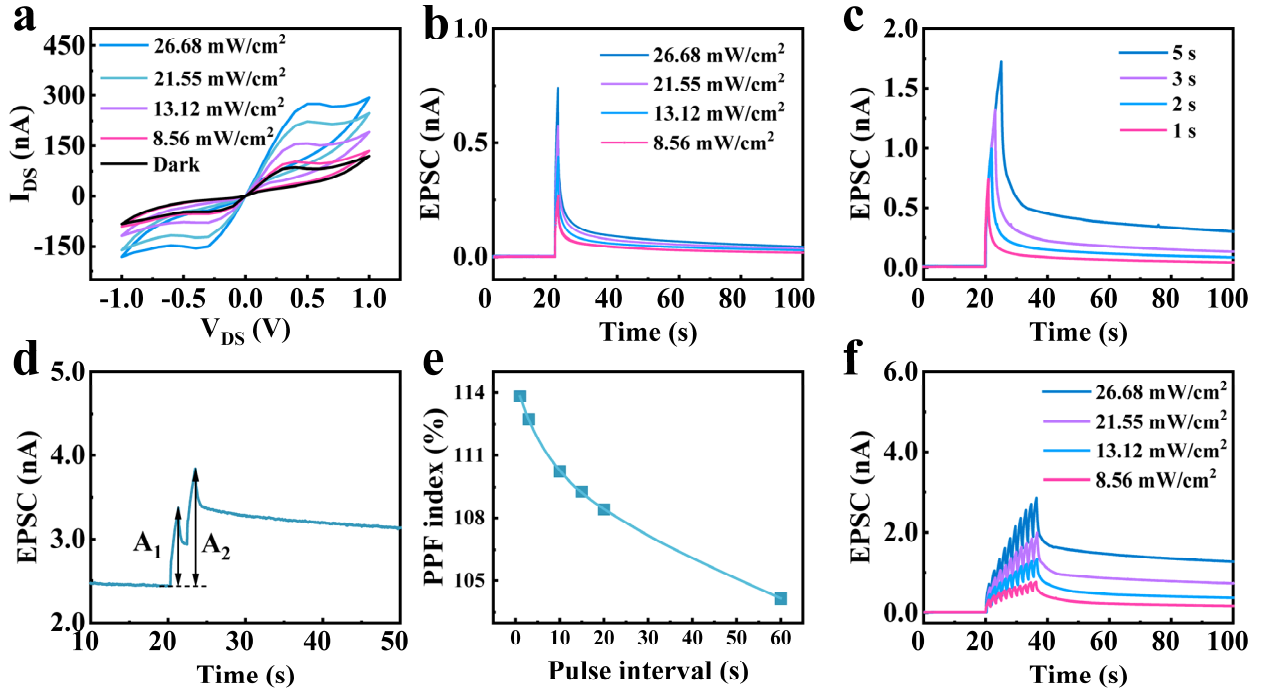


Figure S8. Synaptic response of UCNPs-WSe₂ synaptic device under 405 nm excitation. (a) I-V curves under darkness and 405 nm illumination with different power densities. (b) EPSC triggered by a 405 nm pulse with 1 s width at different powers. (c) EPSC triggered by 405 nm pulse of different pulse durations. (d) PPF caused by a pair of 405 nm light pulses with 1 s width and 0.5 s pulse intervals. (e) The dependence of PPF index on pulse intervals. (f) EPSC induced by 10 pulses at different power densities. All measurements are performed at 100 mV voltage.

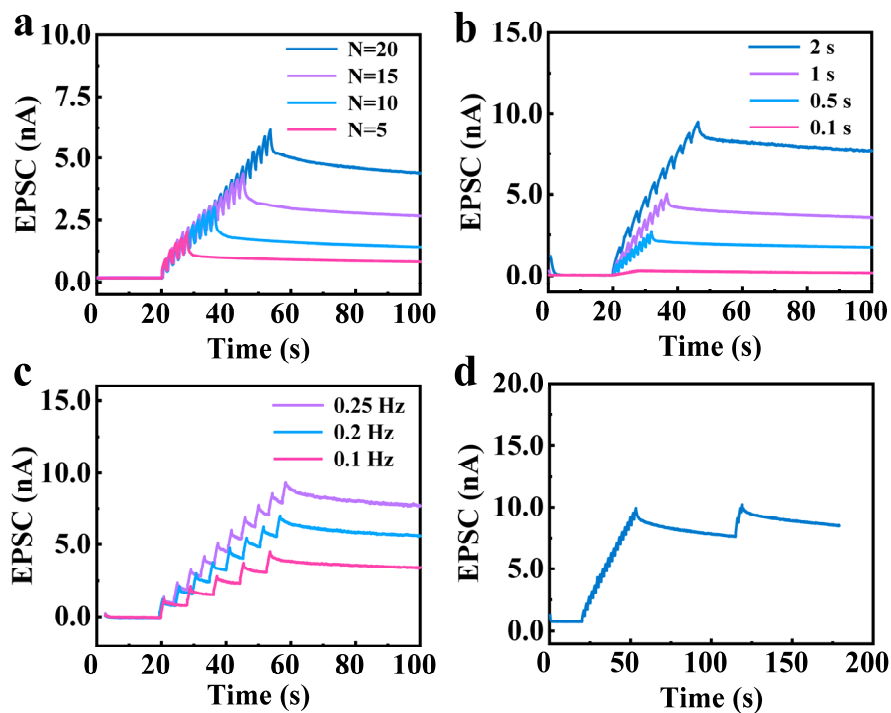


Figure S9. Synaptic response of UCNPs-WSe₂ synaptic device under 405 nm excitation. (a) EPSC triggered by 405 nm with various numbers of pulses. (b) EPSC triggered by different pulse widths with time intervals of 0.5 s. (c) EPSC with frequency from 0.1 Hz to 0.25 Hz. (d) The human body's learning-forgetting-relearning behavior under 405 nm pulses.

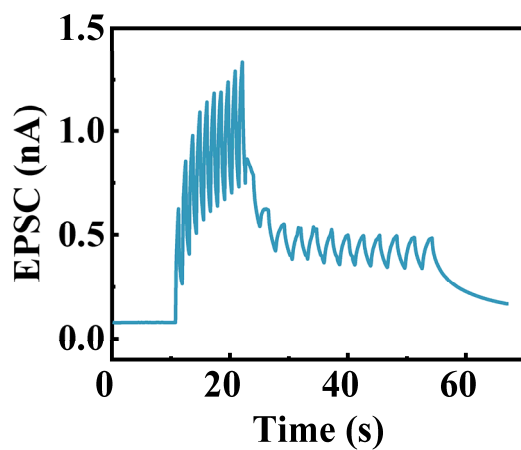


Figure S10. Two types of LTP inputs were modulated by 405 nm and 975 nm pulses under a reading voltage of 100 mV.

References

1. Wen, D. P.; Chen, P.; Liang, Y.; Mo, X. M.; Pan, C. F. Regulated Polarization Degree of Upconversion Luminescence and Multiple Anti-Counterfeit Applications. *Rare Met.* 2024, 43, 2172–2183, doi:10.1007/s12598-024-02675-z.
2. Gong, Y.; Xie, P.; Xing, X.; Lv, Z.; Xie, T.; Zhu, S.; Hsu, H.; Zhou, Y.; Han, S. Bioinspired Artificial Visual System Based on 2D WSe₂ Synapse Array. *Adv Funct Materials* 2023, 33, 2303539, doi:10.1002/adfm.202303539.
3. Guo, Z.; Liu, J.; Han, X.; Ma, F.; Rong, D.; Du, J.; Yang, Y.; Wang, T.; Li, G.; Huang, Y.; et al. High-Performance Artificial Synapse Based on CVD-Grown WSe₂ Flakes with Intrinsic Defects. *ACS Appl. Mater. Interfaces* 2023, 15, 19152–19162, doi:10.1021/acsami.3c00417.