

• Supplementary File •

Photo-synapses based on single-crystalline VO₂ films for in-sensor information processing

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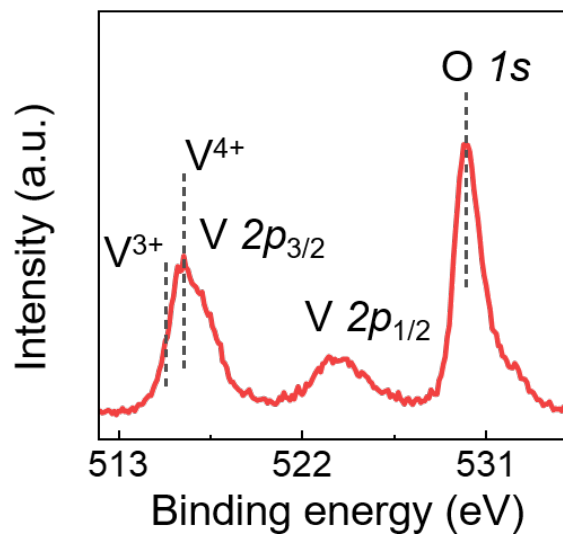


Figure S1 XPS spectra of VO₂ film.

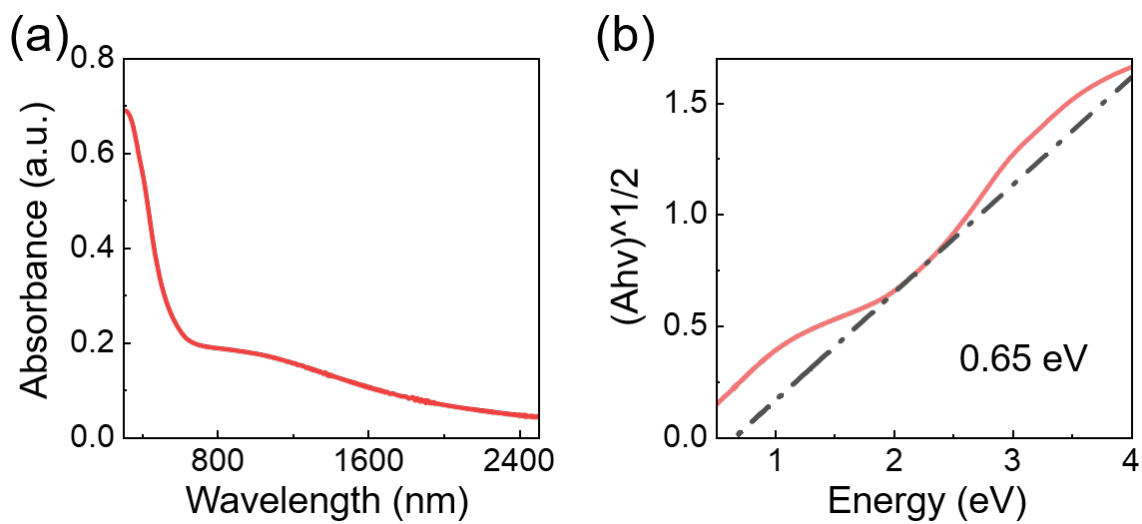


Figure S2 (a) UV-Vis absorption spectroscopy and (b) the corresponding Tauc plots of VO₂.

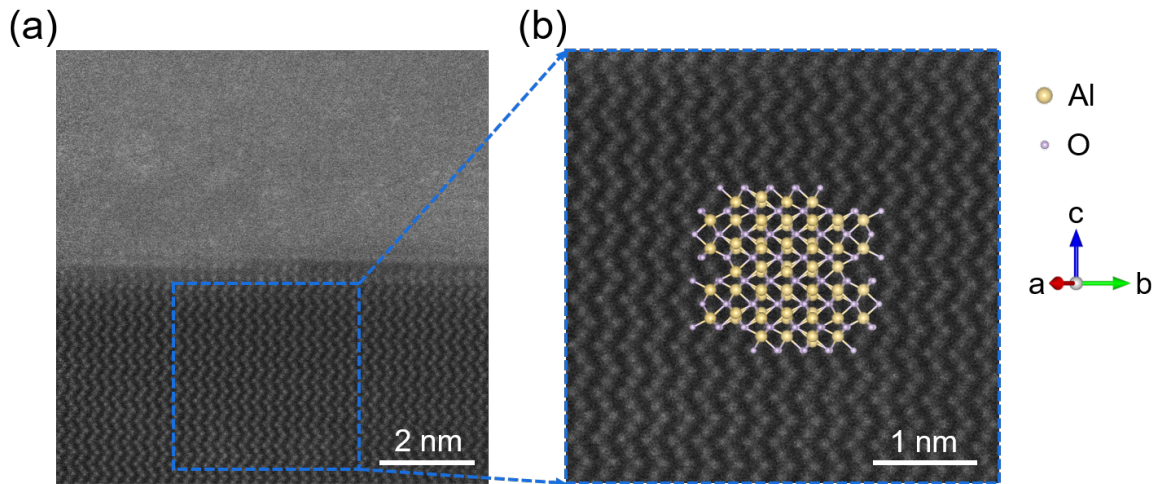


Figure S3 (a) Cross-sectional HAADF-STEM images in the interface region of VO₂/sapphire heteroepitaxy. (b) Crystal structure of sapphire.

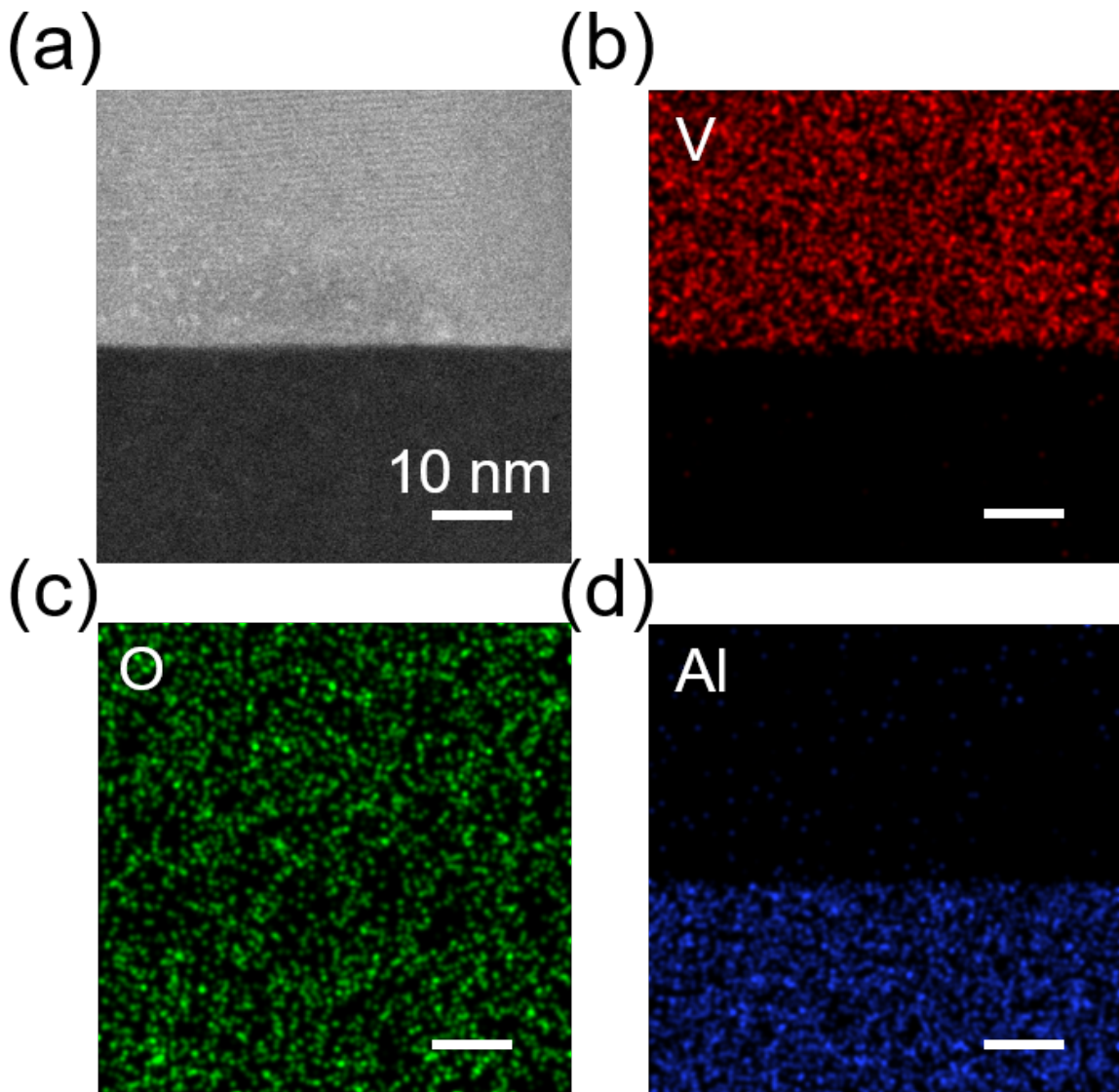


Figure S4 Cross-sectional HAADF-STEM image (a) and EDS mapping (b-d) at the interface of VO₂/sapphire heteroepitaxy.

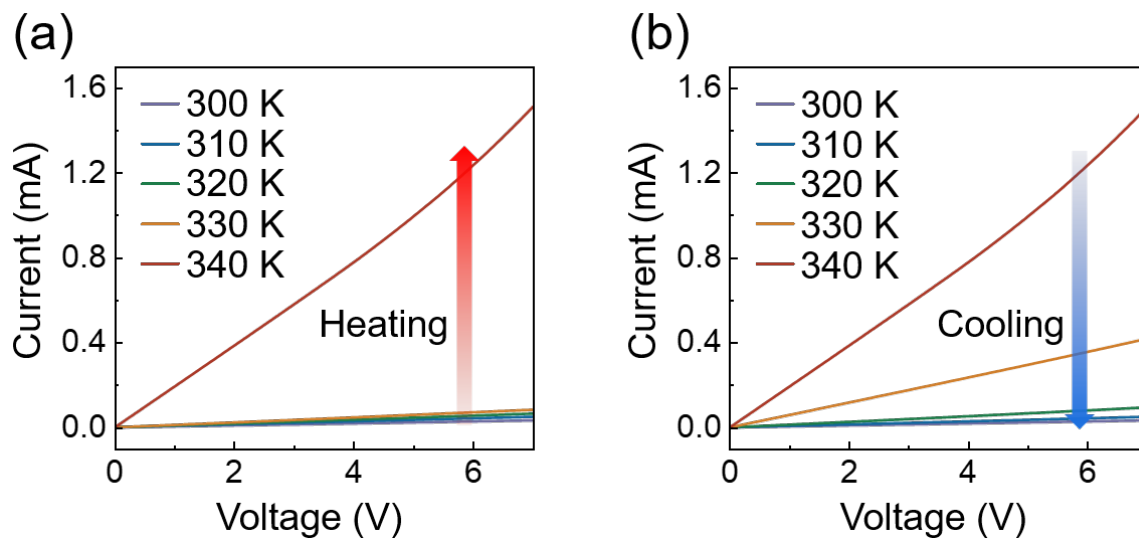


Figure S5 *I-V* curves of two-terminal VO₂ device at different temperatures during heating (a) and cooling (b) cycles, respectively.

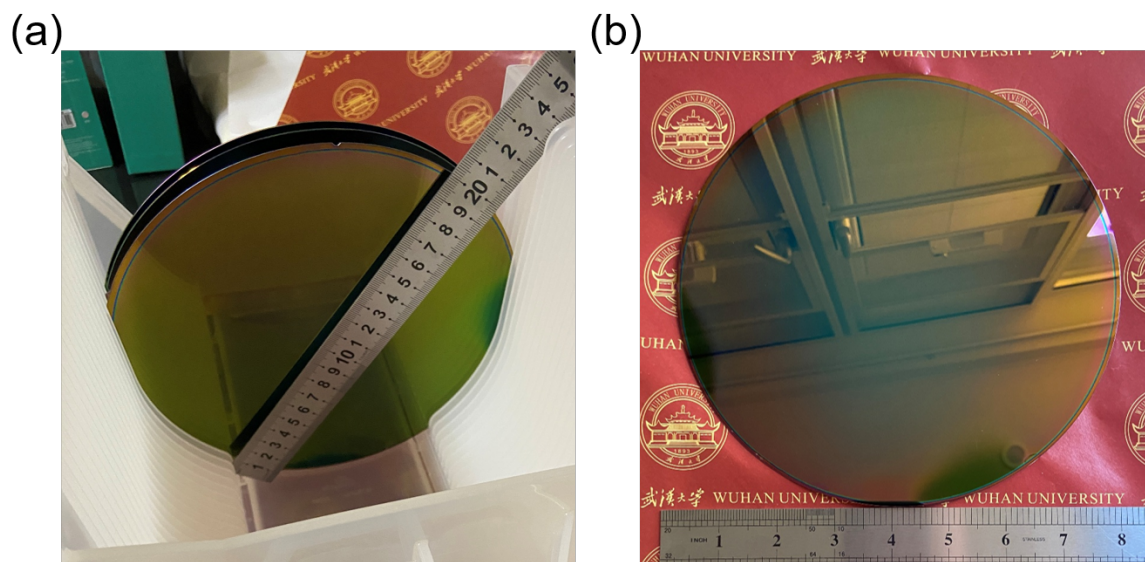


Figure S6 Photograph of 8-inch amorphous VO₂ film.

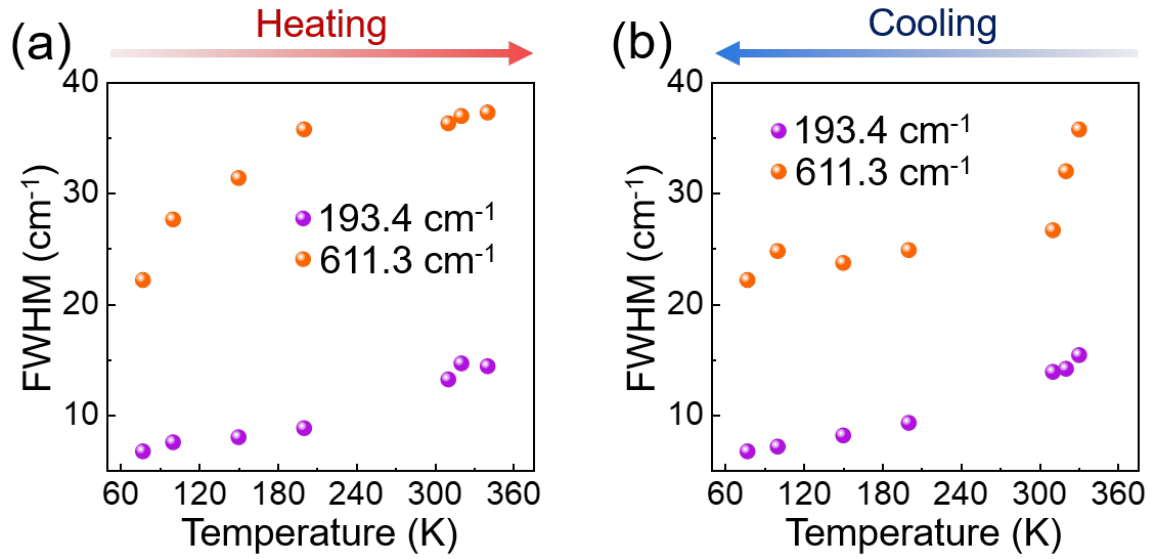


Figure S7 The variation in FWHM of the VO_2 film at 193.4 and 611.3 cm^{-1} as a function of temperature during heating (a) and cooling (b) cycles, respectively.

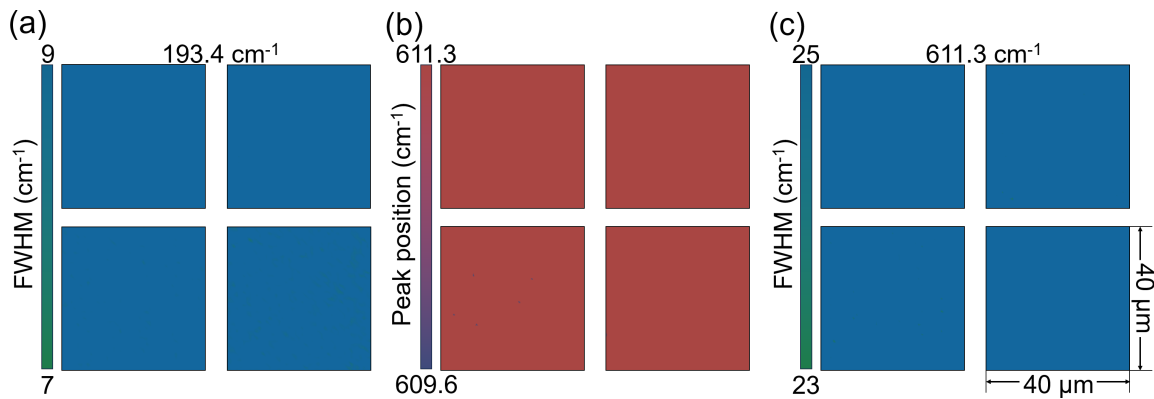


Figure S8 Raman mapping images of the FWHM (a) at 193.4 cm^{-1} , the peak position (b) and FWHM (c) at 611.3 cm^{-1} across 4 distinct film regions.

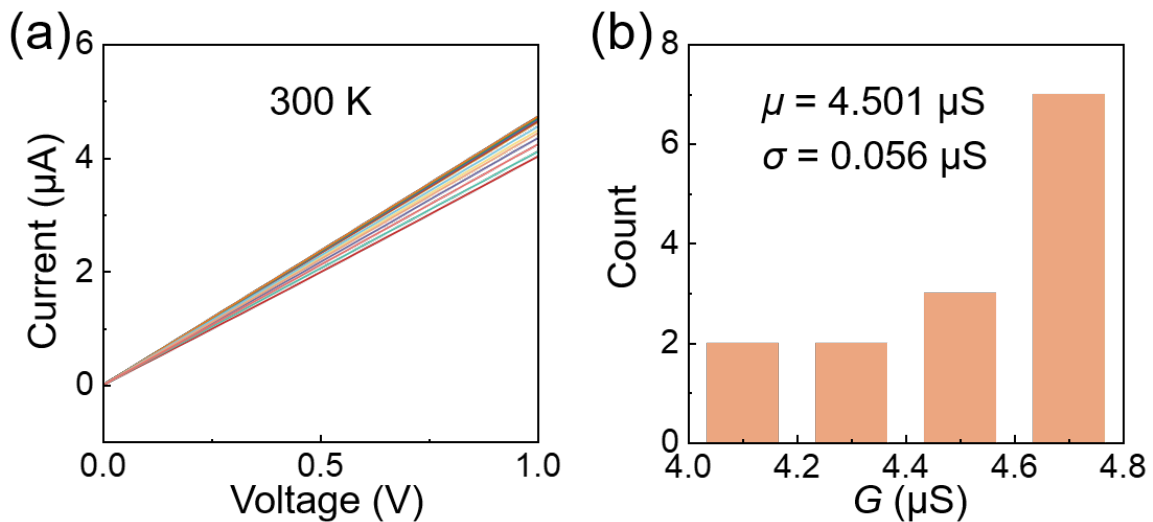


Figure S9 (a) Current-voltage curves of 15 VO_2 devices. (b) The conductance statistics were obtained from 15 devices. The mean and variance values were determined to be 4.501 and $0.056 \mu\text{S}$, respectively.

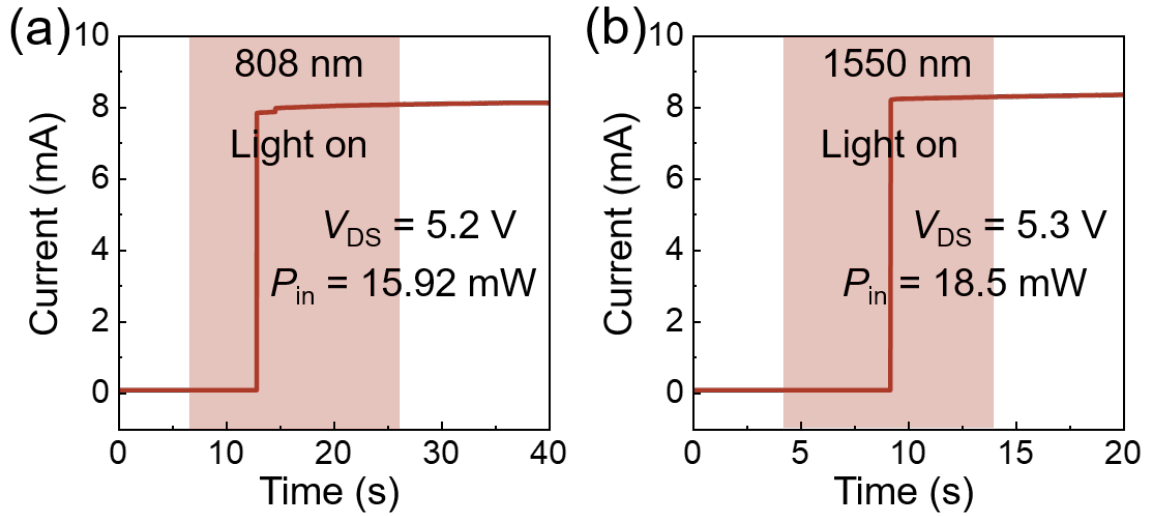


Figure S10 Current-time characteristic of the VO₂ device at wavelengths of (a) 808 and (b) 1550 nm, respectively. Bias voltage of (a) and (b) are 5.2 and 5.3 V, respectively.

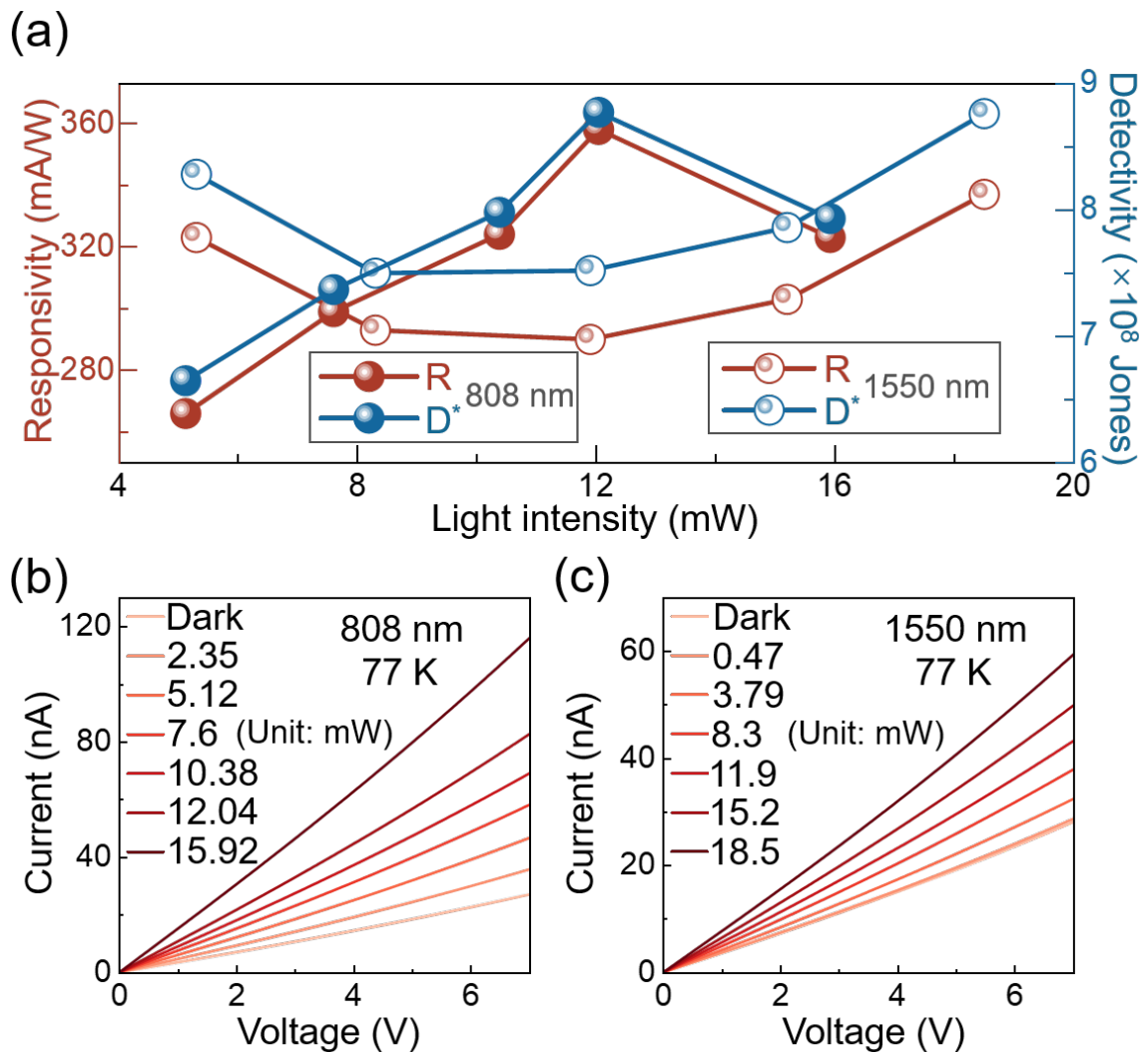


Figure S11 (a) Responsivity and specific detectivity of VO₂ device at 808 nm and 1550 nm. b, c) I - V curves of VO₂ device under 808 nm (b) and 1550 nm (c) light illumination with different incident light intensity at 80 K.

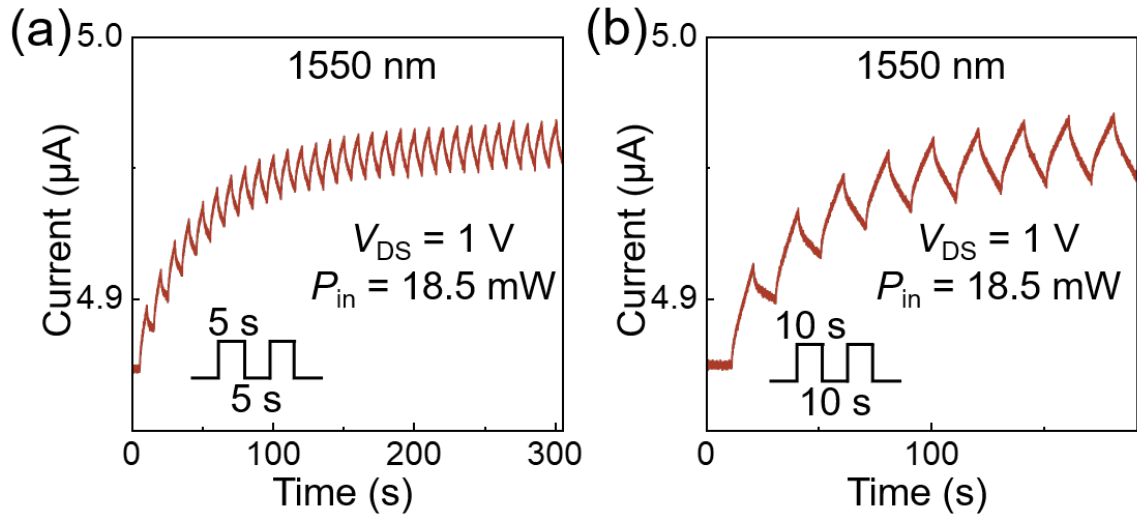


Figure S12 (a) EPSC triggered by a train of 1550 nm optical pulses with period of 10 (a) and 20 s (b), respectively.

Table S1 Actual photocurrent values and corresponding normalized current values for VO₂ photo-synapse device among different optical pulse sequences.

State	ΔI (nA)	Normalized (a.u.)
0000	0	0
1000	36	0.204
0100	40.9	0.232
0010	45.8	0.26
0001	56.2	0.319
1100	75.6	0.429
1010	80.2	0.455
0110	86	0.488
1001	90.8	0.515
0101	99.1	0.562
0011	106.4	0.603
1110	124.7	0.707
1101	128.3	0.727
1011	135.4	0.768
0111	140.1	0.794
1111	176.4	1