Supporting Information

Defect-engineered 2D Bi_2Se_3-based broadband optoelectronic synapses with ultralow energy consumption for neuromorphic computing

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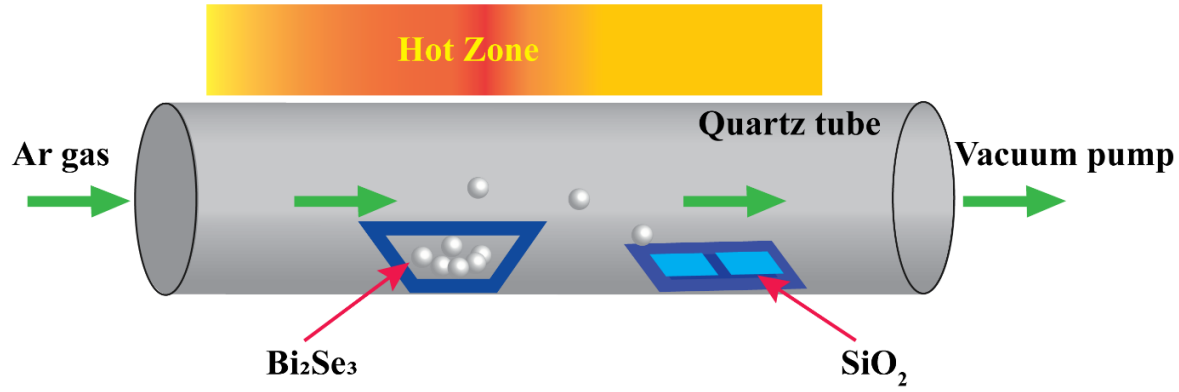

Fig. S1: Schematic diagram of the growth process of the ultrathin 2D Bi_2Se_3 film via CVD.

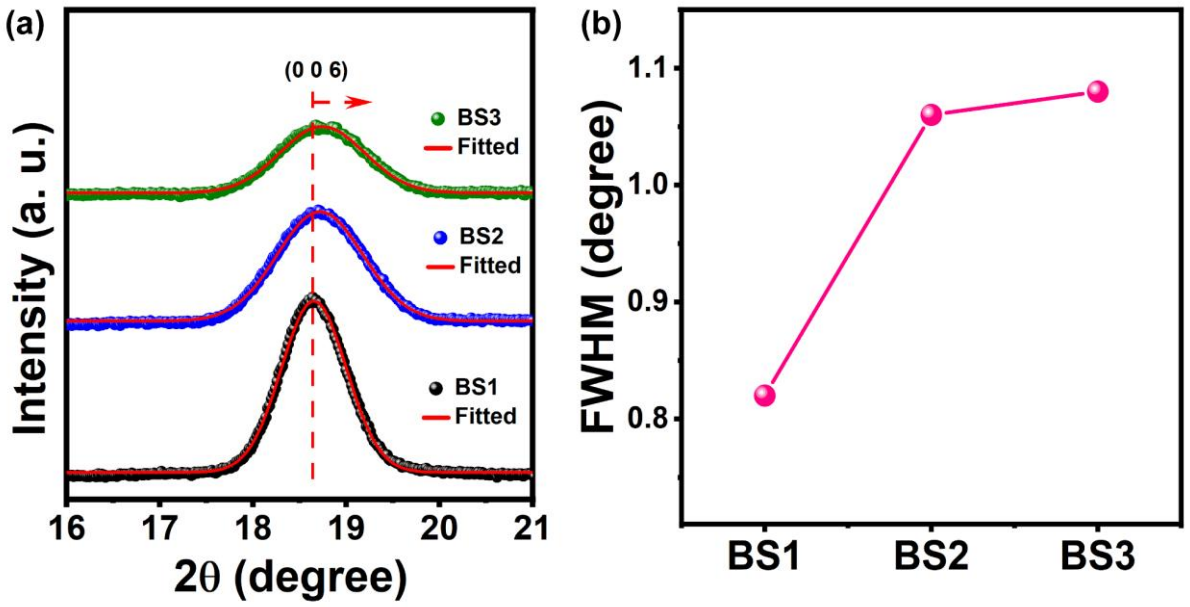

Fig. S2: (a) Fitted peak corresponding to (006) plane of BS1, BS2 and BS3. (b) Measured FWHM from the fitting of all the three samples.

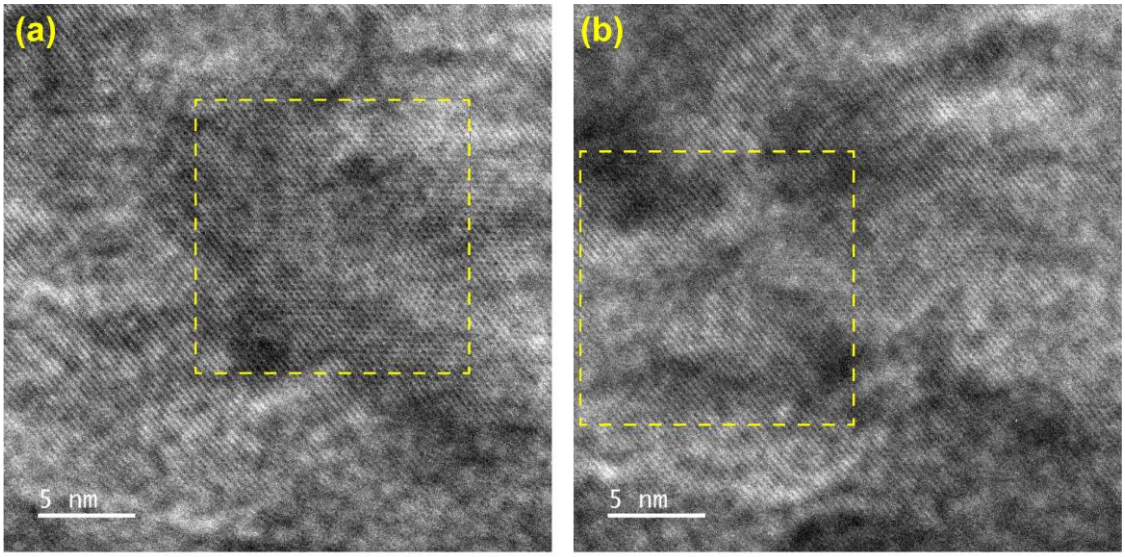

Fig. S3: (a) and (b) HRTEM images of the BS1 and BS3 samples. Square boxes show the position where IFFT was performed.

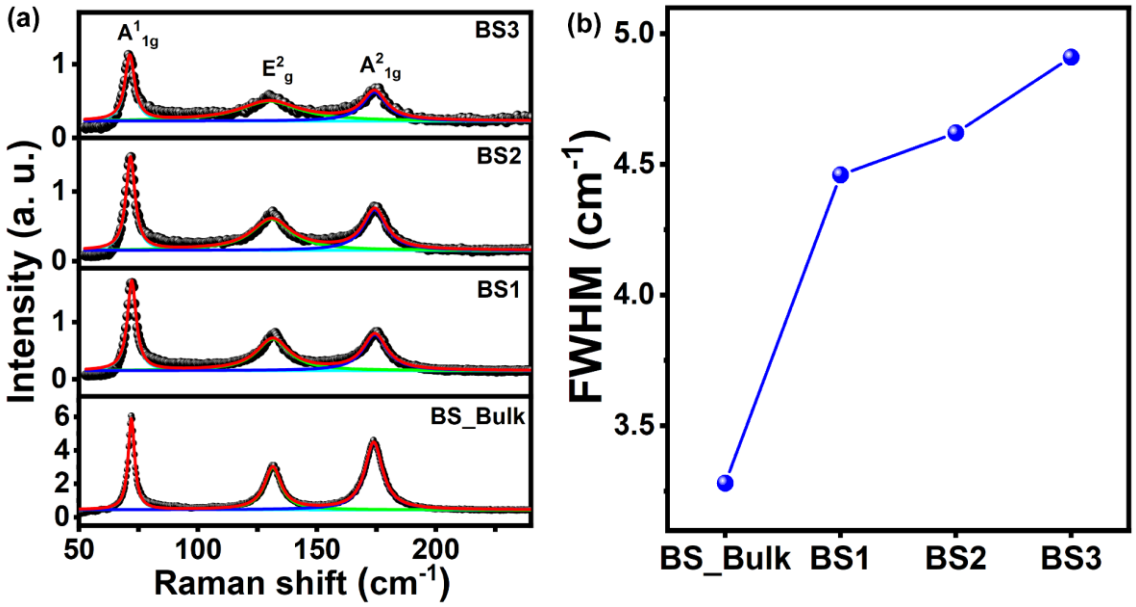

Fig. S4: (a) Fitted Raman spectra of bulk Bi_2Se_3, BS1, BS2, and BS3 and (b) Measured FWHM of the A^1_{1g} modes from the fitting of all three samples.

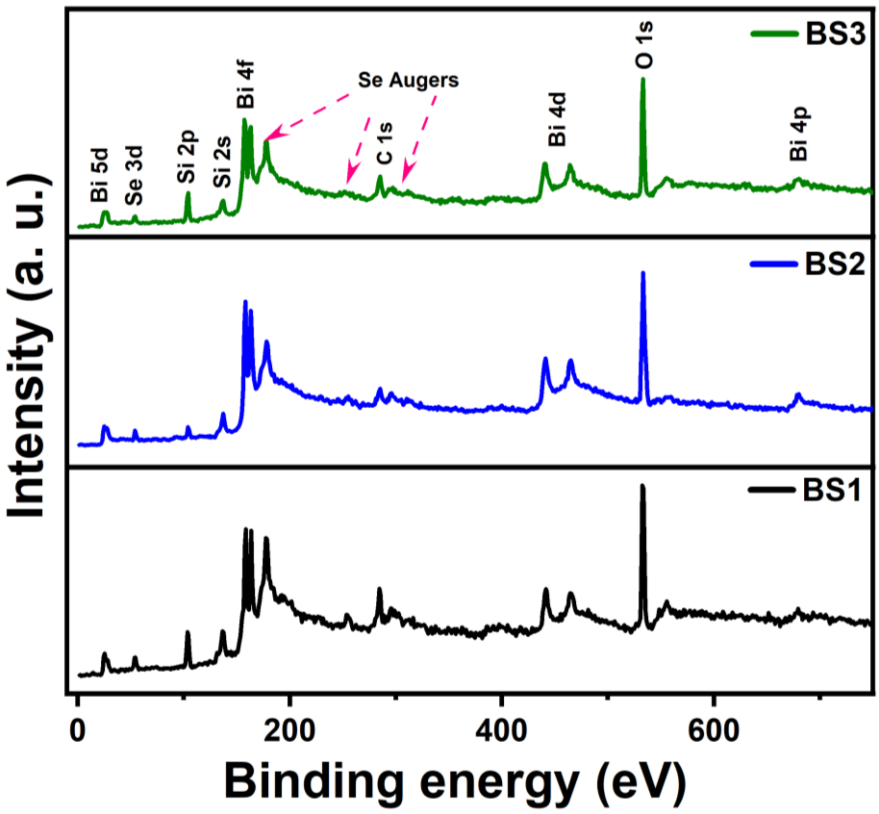

Fig. S5: XPS survey spectra of BS1, BS2, and BS3 showing all the elements present in the samples.

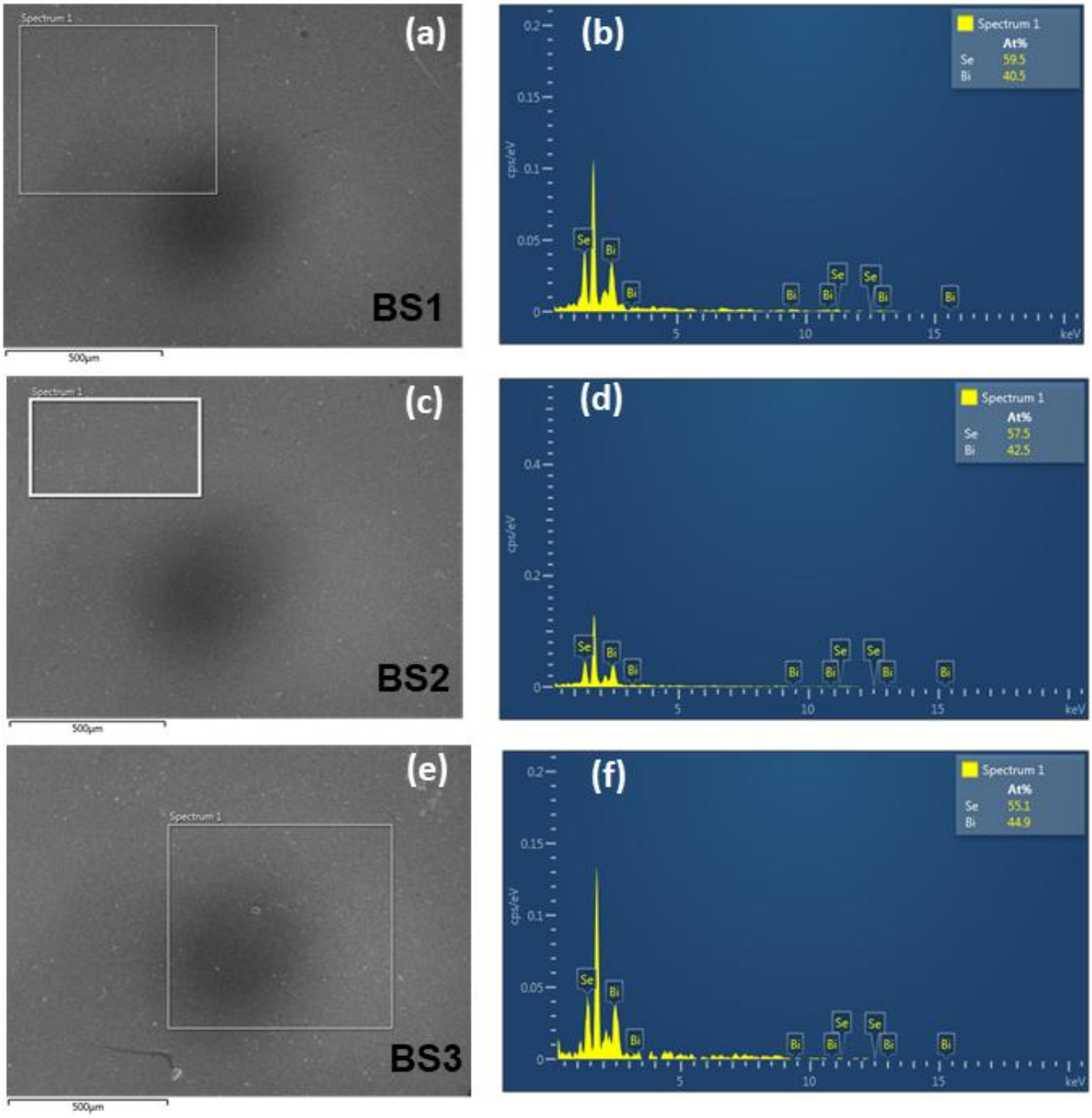

Fig. S6: EDS spectra of the as-grown Bi_2Se_3 thin films showing the atomic percentages of Bi and Se.

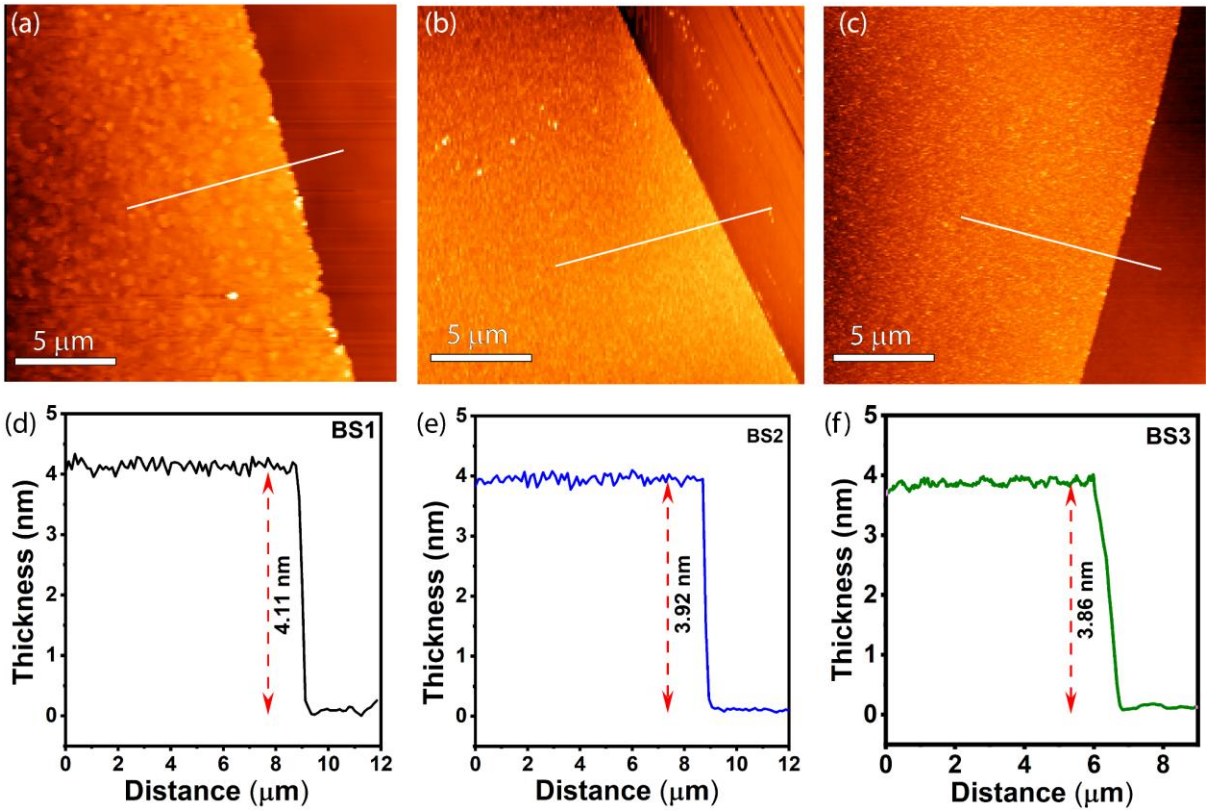

Fig. S7: (a), (b) and (c) show the AFM images taken at the film edges of BS1, BS2 and BS3. (d), (e) and (f) are the height profiles of the BS1, BS2 and BS3 films.

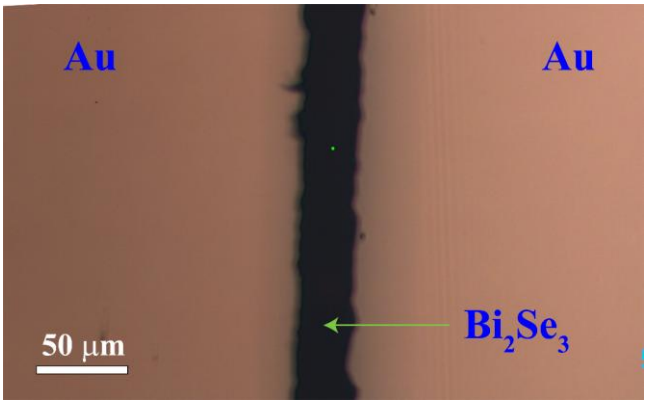

Fig. S8: Optical microscope image of the Bi_2Se_3-based OES device.

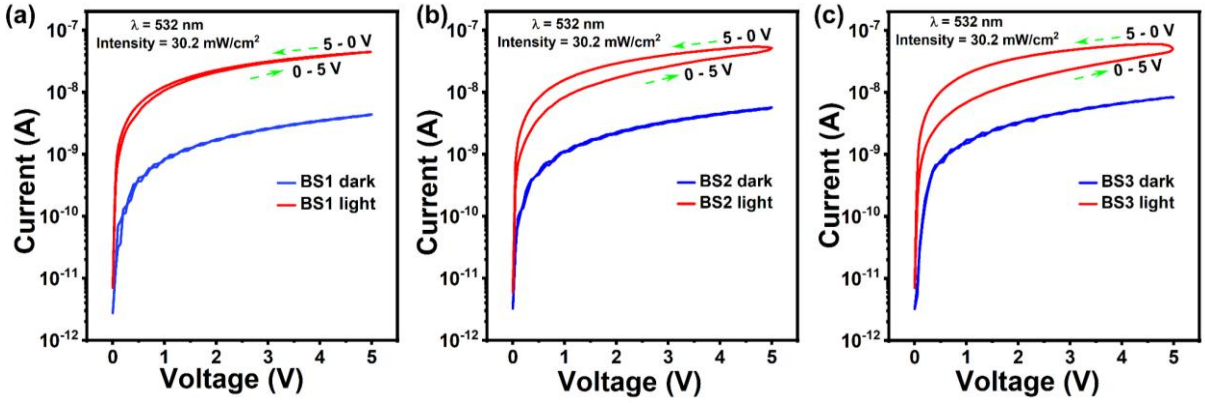

Fig. S9: (a), (b) and (c). Back-scanning I-V characteristics of the BS1, BS2 and BS3 based OES devices.

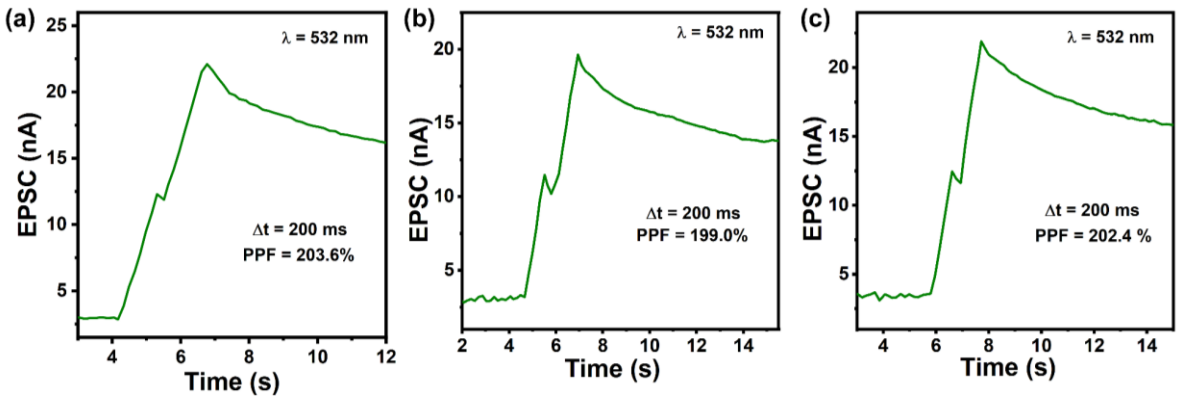

Fig. S10: (a), (b) and (c). The EPSC is triggered by a pair of optical pulses (532 nm, 30.2 mW/cm^2, 1 s) with an interval time of 200 ms for three repetitions with the time interval (Δt) between two consecutive pulses of 200 ms for maximum PPF index measurement.

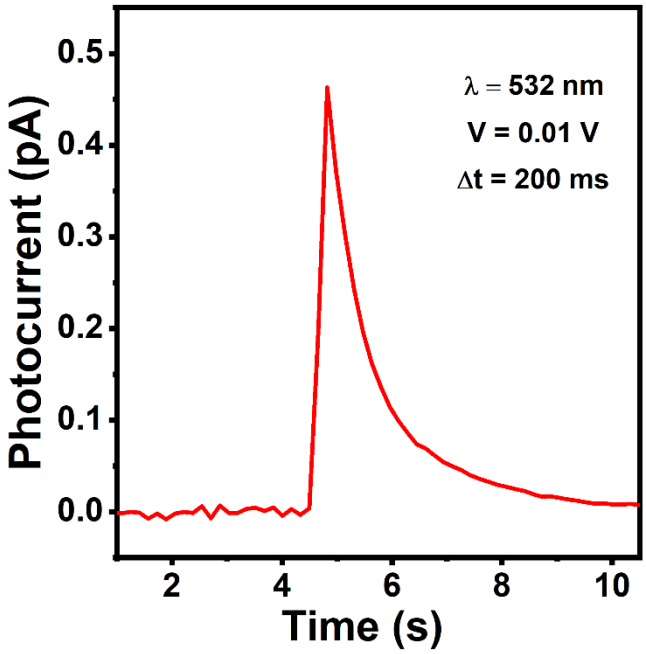

Fig. S11: Photocurrent response of the OES device after illumination of one single pulse to measure the lowest power consumption. V is the applied voltage.

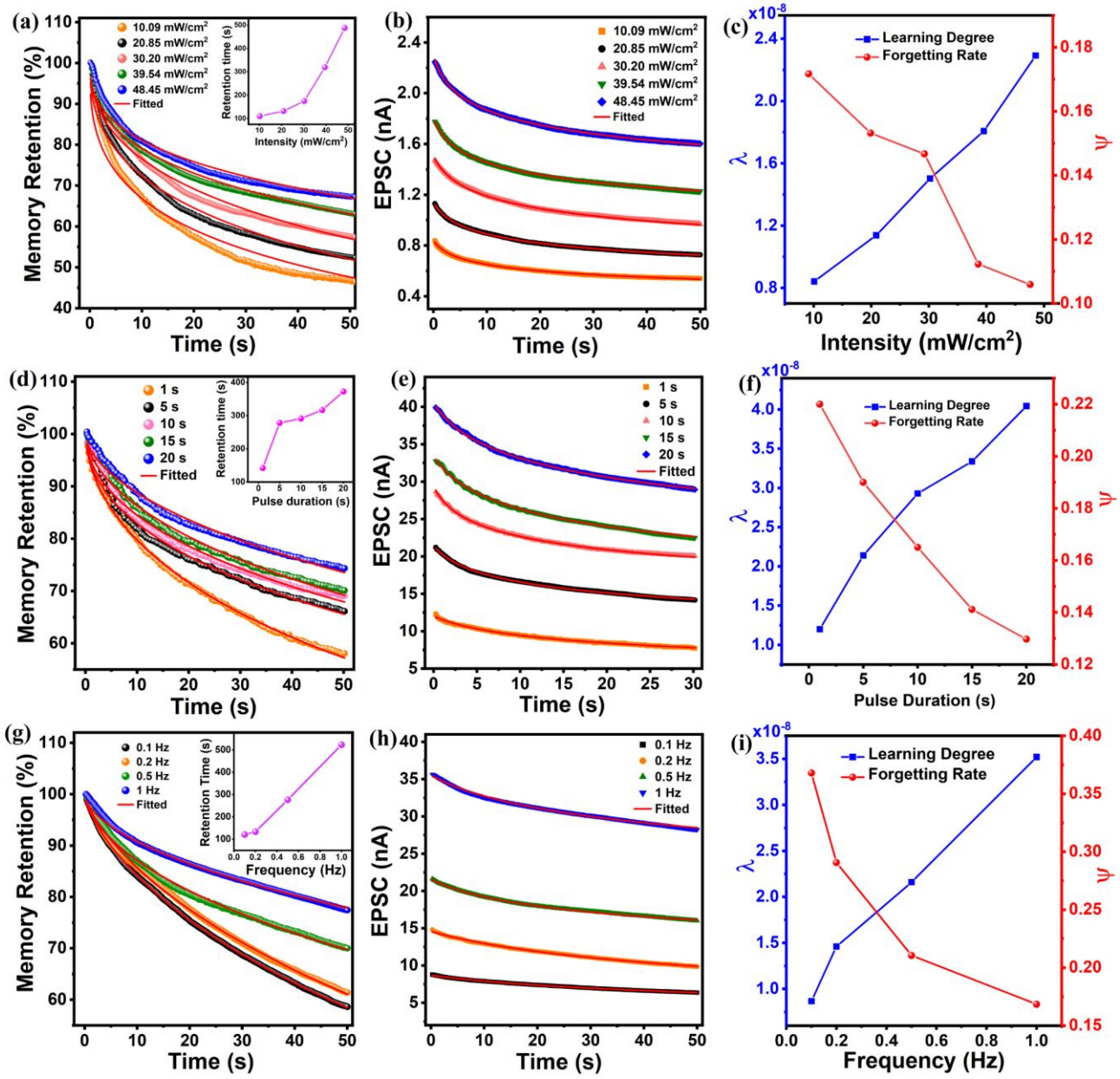

Fig. S12: (a), (d) and (g). Decay of the normalized memory retention change after stimulation by different light intensities, pulse durations and frequencies, respectively. The solid red line is the fitted curve by Kohlrausch function. The inset of each shows the change in the retention time with the corresponding variable. (b), (e) and (h). Decay of the EPSC after stimulation by different intensities, pulse durations and frequencies, respectively. The solid red line is the fitted curve by the Wickelgren power-law. (c), (f) and (i). The change of learning degree (λ) and forgetting parameter (ψ) with light intensity, pulse duration and frequency respectively.

Table S1. Summary of the values of parameters τ, λ and, ψ for all the cases.

Conditions		Retention time (τ), s	Learning degree (λ)	Forgetting rate (ψ)
Intensity (mW/cm^2)	10.09	109.8	8.41×10^{-9}	0.1717
	20.85	131.8	1.14×10^{-8}	0.1532
	30.20	174.7	1.50×10^{-8}	0.1467
	39.54	319.3	1.81×10^{-8}	0.1123
	48.45	487.9	2.29×10^{-8}	0.1059
Pulse duration (s)	1	142.3	1.20×10^{-8}	0.2216
	5	278.1	2.14×10^{-8}	0.1910
	10	291.3	2.93×10^{-8}	0.1653
	15	317.0	3.34×10^{-8}	0.1411
	20	372.5	4.04×10^{-8}	0.1297
Pulse number	5	112.04	1.54×10^{-8}	0.2850
	10	137.64	2.41×10^{-8}	0.2575
	20	214.93	3.09×10^{-8}	0.2262
	30	523.07	3.52×10^{-8}	0.1685
Pulse frequency (Hz)	0.1	120.71	8.67×10^{-9}	0.3679
	0.2	133.28	1.46×10^{-8}	0.2906
	0.5	276.85	2.16×10^{-8}	0.2105
	1	523.07	3.52×10^{-8}	0.1685

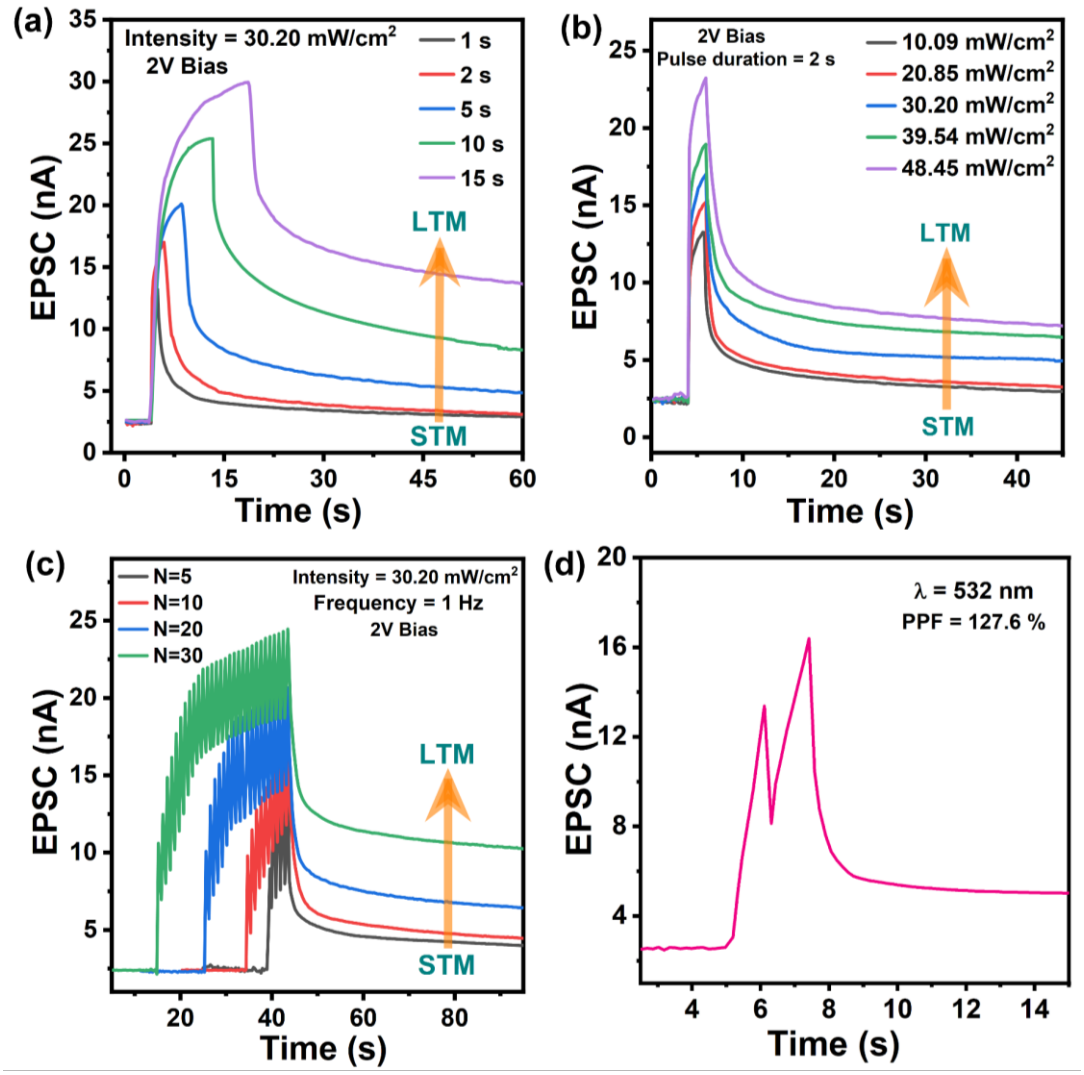

Fig. S13: (a) The transition of STM to LTM of the BS2 device at 2 V bias by increasing (a) pulse duration (532 nm, 30.2 mW/cm^2), (b) the light intensity (532 nm, 2 s), and (c) number of pulses (532 nm, 30.2 mW/cm^2, 1Hz). (d) the EPSC is triggered by a pair of optical pulses (532 nm, 30.2 mW/cm^2, 1 s) with an interval time of 200 ms for maximum PPF index measurement of the BS2 device.

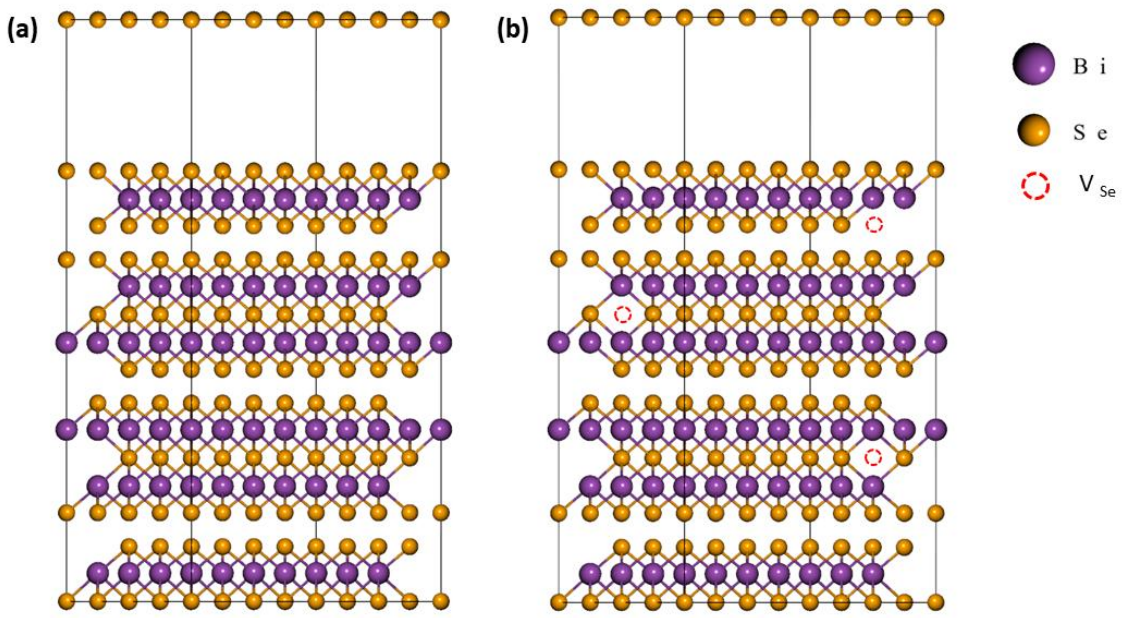

Fig. S14: Atomic structure of the 4-layer. (a) Bi_2Se_3 and (b) Bi_2Se_3 with Se vacancies.

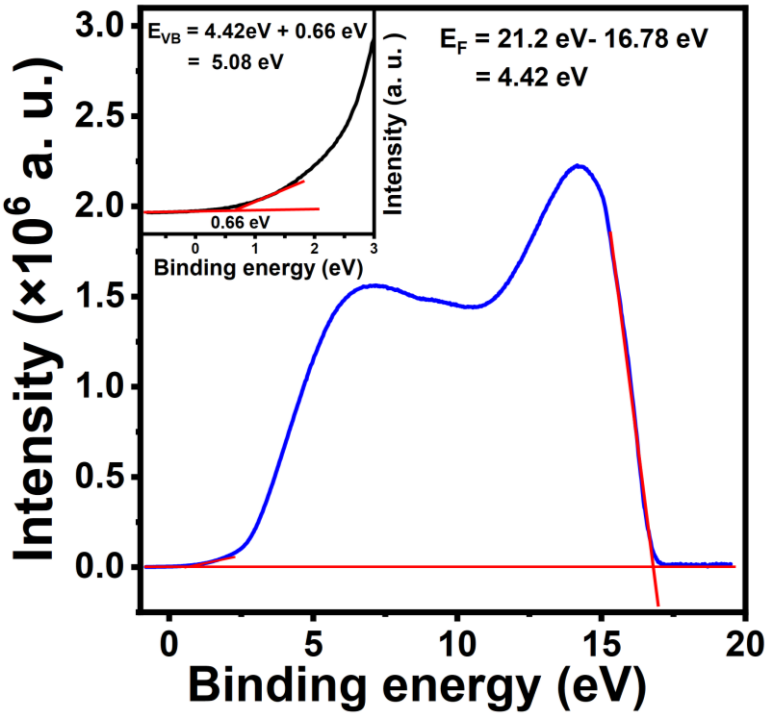

Fig. S15: UPS spectra and Fermi level analysis of intrinsic Bi_2Se_3 film; the inset shows the estimation of the maximum of the valence band.

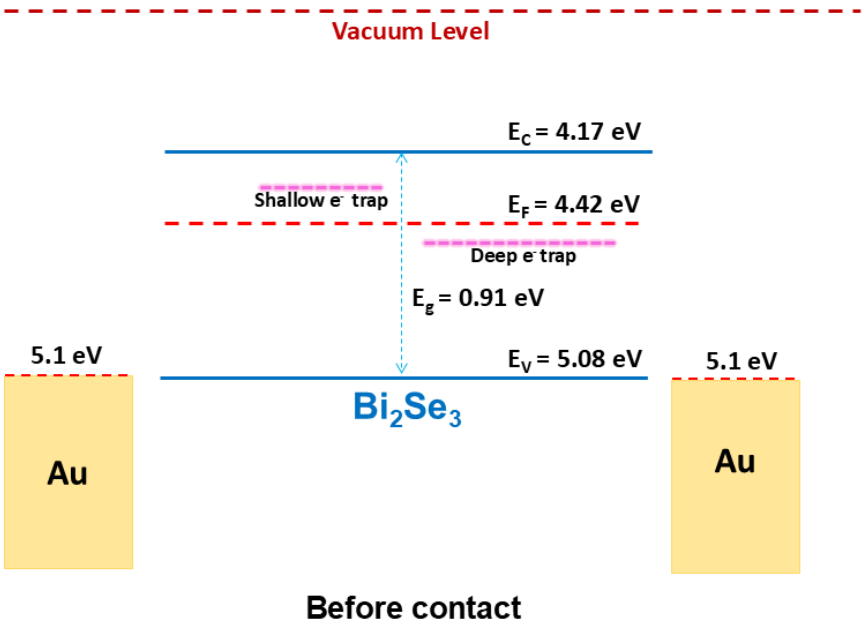

Fig. S16: Schematic of the band diagram of Bi_2Se_3 in conjunction with the Au electrodes before contact.

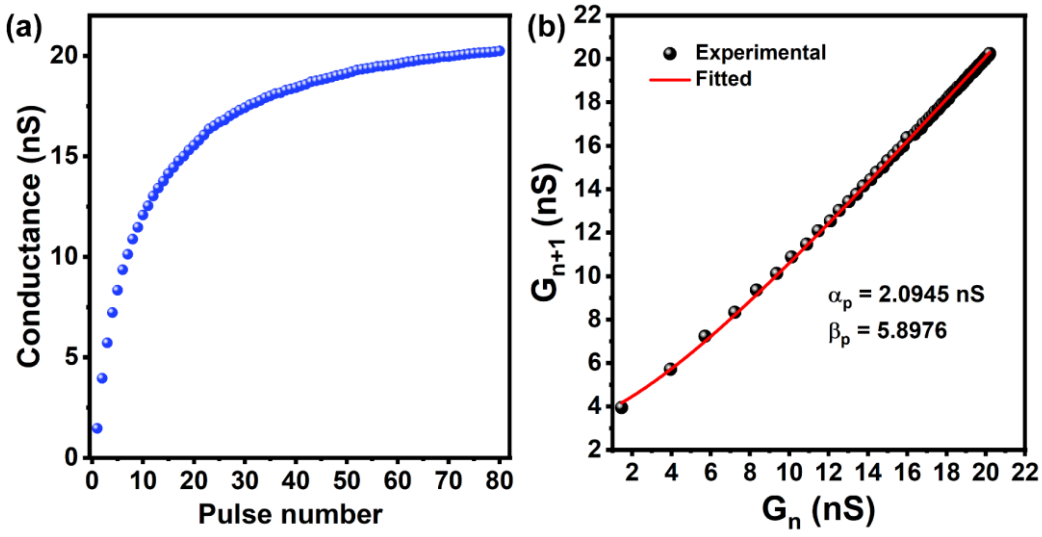

Fig. S17: (a) LTP curve shows the change of conductance with pulse number. (b) Fitted nonlinearity curve from the experimental data.