Supporting Information

Tunable Non-volatile Memory Based on 2D InSe/h-BN/GaSe

heterostructure towards potential multifunctionality

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The supporting information includes Figures S1-S9 and Table S1.

S1: The fabrication process schematics of the InSe/*h*-BN/GaSe FG-FET device



Figure S1. Schematic fabrication process of the 2D InSe/*h***-BN/GaSe heterostructure based floating-gate memory.** (a) preparation of GaSe nanosheets on the SiO₂/Si substrate. (b-i) construction of InSe/*h*-BN/GaSe heterostructure, including exfoliation, transfer and stacking process of *h*-BN and InSe. (j) Electron beam evaporation was used to define Cr/Au electrodes on the obtained heterostructures. S2: AFM images of the *h*-BN/GaSe, InSe/*h*-BN/GaSe heterostructures



Figure S2. Surface morphology of *h*-BN/GaSe and InSe/*h*-BN/GaSe heterostructures. (a) The topography of the *h*-BN/GaSe heterostructure in the stacking process, with surface roughness R_q about 0.12 nm after annealing. (b) The topography of the InSe/*h*-BN/GaSe heterostructure in the stacking process, with surface roughness R_q about 0.15 nm after annealing.



S3: OP and AFM images of the InSe, h-BN, GaSe

Figure S3. Morphology and thickness characterizations of the InSe, *h*-BN, GaSe nanosheets. (a-c) Optical images of InSe, *h*-BN, GaSe on Si/SiO₂ substrate. The typical size of each flake is around tens of micrometers. (d-f) AFM images of the InSe, *h*-BN, GaSe area within the corresponding circle in (a-c). Thickness line profiles of the InSe, *h*-BN and GaSe flakes measured at positions marked in red lines in (d-f), respectively. The results show that the thicknesses of InSe, *h*-BN and GaSe are about 13.48 nm, 6.74 nm and 11.43 nm, respectively.

S4: Optical microscope images of the devices with different *h*-BN thickness and corresponding AFM characterizations



Figure S4. Thickness characterizations of different *h*-BN related devices. (a-c) False-colour optical images of the InSe/*h*-BN/GaSe heterostructure Device 1-3 respectively. (d) AFM characterization of *h*-BN in the Device 1 in Figure (a). The thickness of the *h*-BN flake measured at the position marked in red lines, showing that the thickness of *h*-BN is about 29 nm. (e) AFM characterization of *h*-BN in the Device 2 in figure (b). The thickness of the *h*-BN flake measured at the position marked in red line, showing that the position marked in red line, showing that the thickness of *h*-BN is about 23 nm. (f) AFM characterization of *h*-BN in the Device 3 in figure (c). The thickness of the *h*-BN flake measured at the position marked in red line, showing that the thickness of *h*-BN is about 23 nm. (f) AFM characterization of *h*-BN in the Device 3 in figure (c). The thickness of the *h*-BN flake measured at the position marked in red line, showing that the thickness of *h*-BN is about 21 nm.

S5: More details about the Device 2



Figure S5. Memory characteristics of InSe/*h*-BN/GaSe FG-FET Device 2.

S6: More details about the Control Device 3



Figure S6. Transfer curves of InSe related devices under Control Device 3 configuration. (a) False-colour optical image of the Control Device 3. (b) Output characteristics (drain-source current I_{ds} vs. drain-source voltage V_{ds}) of the floating gate transistor in the ON state, for control gate bias V_{cg} from -5 V to 5V. The curves show almost linear at small V_{ds} , indicating good ohmic contacts.



S7: Photoluminescence (PL) characterizations of few layers InSe, GaSe

Figure S7. PL spectra of the InSe (a) and GaSe (b) flakes comparing with **bulk.** Corresponding band gap values are use in Figure 4.

S8: Extinction Ratio of the Device 4



Figure S8. More details in erasing non-volatile of InSe/*h*-BN/GaSe FG-FET Device 4. Temporal evolution of drain-source currents (I_{ds}) in the erase (ON) and program (OFF) states. The curves are acquired independently for the erase ($10^{-7}-10^{-6}$ A) and program ($10^{-11}-10^{-10}$ A) current states and plotted on a common time scale. The drain-source bias voltage is 5 V, with the $V_{cg} = \pm 60$ V and the duration of the control-gate voltage (V_{cg}) pulse is 2 s. The resulted extinction ratio is close to 10^3 .



S9: Simulation of synaptic features in the Device 3

Figure S9. **Simulation of synaptic features based on the floating gate FET Device 3.** (**a**) The generation of excitatory postsynaptic current (EPSC) when a pulse voltage of -15 V is applied to the device. (**b**) The generation of EPSC when negative pulse voltages of different magnitude are applied to the device. (**c**) Inhibitory postsynaptic current (IPSC) generation when a pulse voltage of + 5 V is applied to the device. (**d**) The generation of IPSC when forward pulse voltages of different amplitudes are applied to the device.

Configurations	<i>t_{InSe}</i> (nm)	<i>t_{h-BN}</i> (nm)	t _{GaSe} (nm)	Floating gate
Device 1	11	29	13	floating
Device 2	14	23	15	floating
Device 3	14	11	12	floating
Device 4	13	7	11	floating
Control Device 1	13	7	11	grounded
Control Device 2	15	8	_	-
Control Device 3	10	_	13	floating

Table S1. Details of Devices 1-4 and Control Devices 1-3.

Table S2. Overall performance of Devices 4.

Configurations	ON/OFF	$\Delta V(V)$	$V_{ds}(V)$	$V_{cg}(V)$
	10 ⁴	72	5	± 60
	10 ³	75	-5	± 60
Device 4	10 ³	58	-5	± 50
	10 ³	50	-1	± 60
	10 ³	50	1	± 60