## **Supporting Information**

Narrow-gap physical vapour deposition synthesis of ultrathin SnS<sub>1-x</sub>Se<sub>x</sub> (0≤x≤1)

## Two-dimensional Alloys with unique polarized Raman spectra and high

## (opto)electronic properties.

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Fig. S1 Optical images of the SnS<sub>1-x</sub>Se<sub>x</sub> alloyed nanosheets by different wafer methods: (a) one wafer method. (b) two wafers covered each other.



**Fig. S2** (a) Schematic diagram for the traditional growth of 2D SnS<sub>1-x</sub>Se<sub>x</sub> alloyed nanosheets. The samples were grown on the top of the SiO<sub>2</sub>/Si substrate. (b) Enlarged image of the black ellipse, showing the detailed orientation growth of the alloyed



Fig. S3 SEM-EDS of the SnS<sub>0.5</sub>Se<sub>0.5</sub> alloyed nanosheets

Element	Atomic (%)	Theoretical Atomic (%)
S	22.02	20
Se	20.15	20
S/Se ratio	1.09	1

Table S1. The calculation result extracted from the corresponding EDS spectrum in



Fig. S4 SEM-EDS of the  $SnS_{0.75}Se_{0.25}$  alloyed nanosheets

Table S2. The calculation result extracted from the corresponding EDS spectrum in

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Element	Atomic (%)	Theoretical Atomic (%)
S	32.79	30
Se	10.08	10
S/Se ratio	3.25	3

Figure S3.



Fig. S5 SEM-EDS of the  $SnS_{0.25}Se_{0.75}$  alloyed nanosheets

Table S3. The calculation result extracted from the corresponding EDS spectrum in

<b>8</b>					
Element	Atomic (%)	Theoretical Atomic (%)			
S	12.23	10			
Se	28.94	30			
S/Se ratio	0.42	0.33			

Figure	S4.
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Fig. S6 SEM images of the  $SnS_{1-x}Se_x$  alloyed nanosheets via two wafers method (a)





Fig. S7 (a) Transfer characteristic curve of the device under drain voltage of 1 V under dark condition. Inset: the optical image of the device, scale bar: 10  $\mu$ m. (b) Output characteristic curves of the device under different V<sub>g</sub> values (from 80 V to -80

V using step of 40 V).



Fig. S8 Macroscopic scheme of NGPVD method, showing the detailed orientation

growth of the alloyed sample.

Si sub	strate	Тор	SiO <sub>2</sub> laye	r
N <sub>2</sub> SnS/SnSe powder	Sublimation	Diffu Adsorption Decompositi	Assemblage Islon on	Aechanical pumping Sn • S • Se
		Bottom		

Fig. S9 Microcosmic scheme of NGPVD method, showing the detailed orientation



growth of the alloyed sample.

Fig. S10 Normalized Raman spectra of the  $SnS_{0.5}Se_{0.5}$  nanosheets with different



**Fig. S11** The microcosmic scheme of FETs devices based on  $SnS_{1-x}Se_x$  alloyed samples. (a) and (b) for ultrathin sample. (c) and (d) for thicker sample (larger than 25

nm).