

## Supporting Information

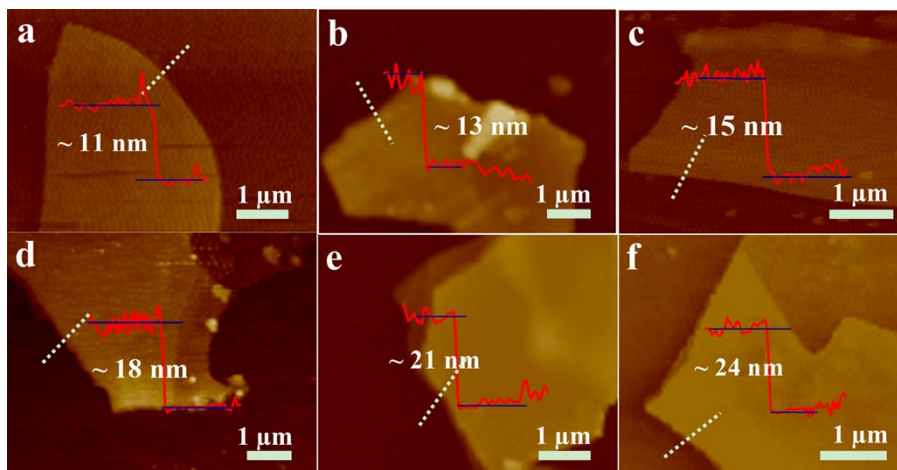
### **Modulation of opto-electronic properties of InSe thin layers via phase transformation**

Makkawi Osman,<sup>a</sup> Yanmin Huang,<sup>a</sup> Wei Feng,<sup>a</sup> Guangbo Liu,<sup>a</sup> Yunfeng Qiu<sup>a,\*</sup>

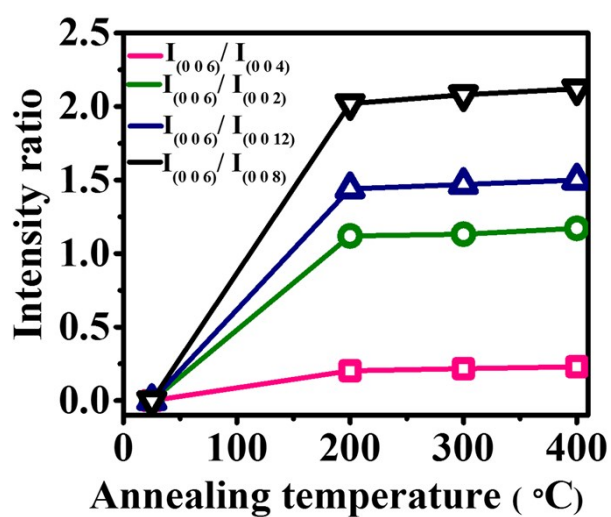
PingAn Hu<sup>a, b, \*</sup>

<sup>a</sup> *Key Lab of Microsystem and Microstructure of Ministry of Education, Harbin Institute of Technology, School of Materials Science and Engineering, No. 2 YiKuang Street, Harbin, 150080, China. E-mail: [hupa@hit.edu.cn](mailto:hupa@hit.edu.cn); [qiuyf@hit.edu.cn](mailto:qiuyf@hit.edu.cn)*

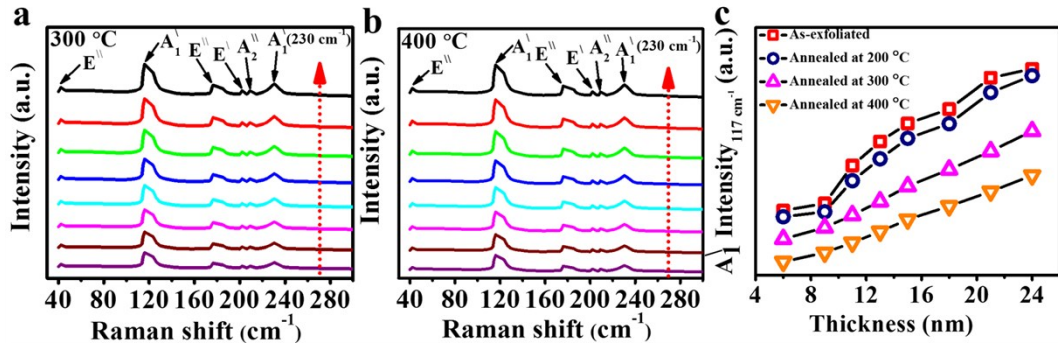
<sup>b</sup> *State Key Laboratory of Robotics and System (HIT), Harbin Institute of Technology, Harbin, Heilongjiang 150080, China.*



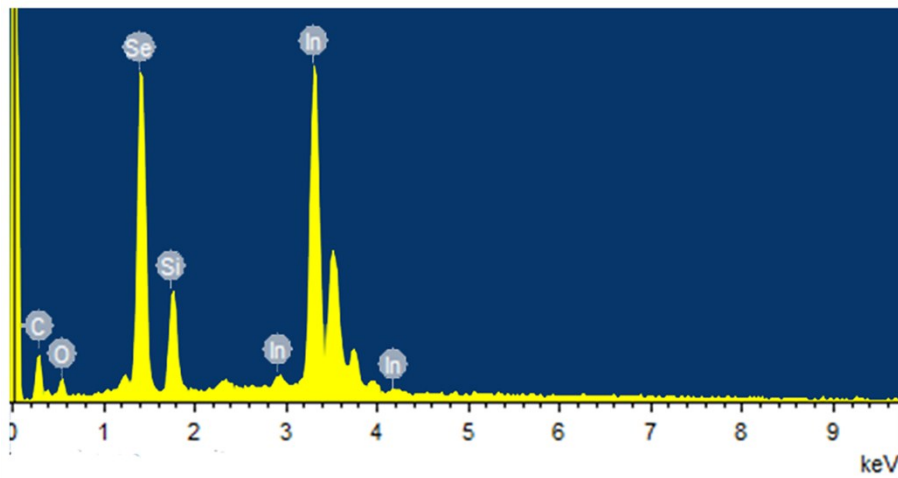
**Figure S1.** (a to f) AFM images of InSe nanosheets with different thickness of 11, 13, 15, 18, 21, and 24 nm. Insets are corresponding height profiles.



**Figure S2.** Intensity ratios of  $I_{(006)}$  of  $\gamma$ - $\text{In}_2\text{Se}_3$  to the other peaks of  $\beta$ -InSe.



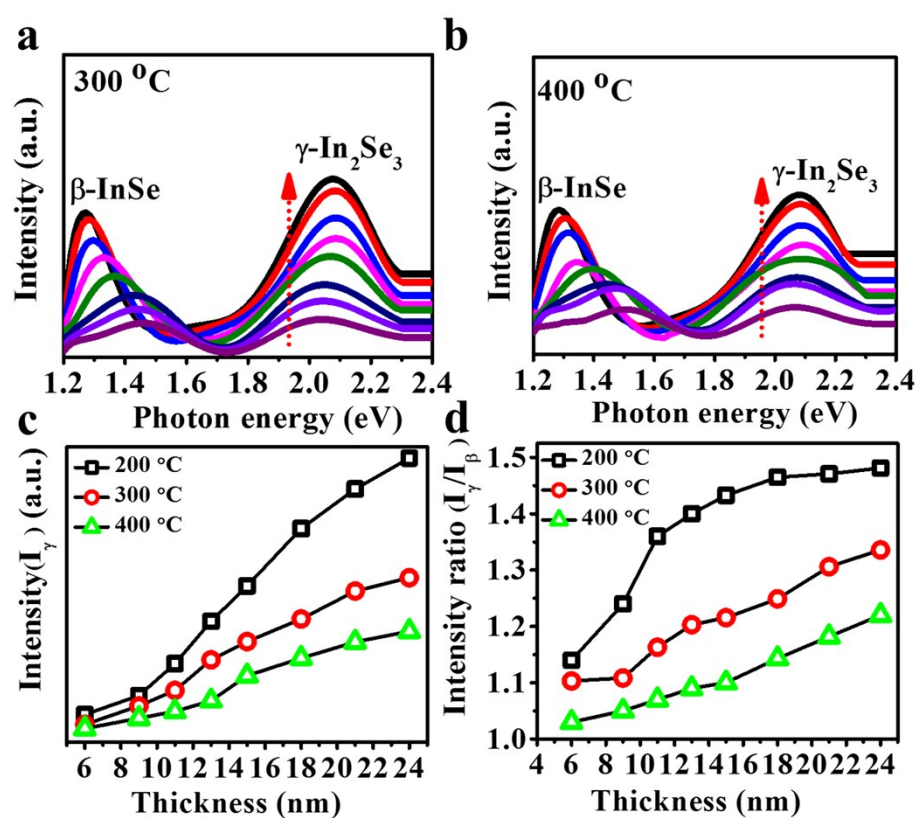
**Figure S3.** (a and b) Raman modes of annealed sheets at 300 °C and 400 °C, respectively. The thickness from bottom to top as indicated by dotted arrows in both a and b are about 6, 9, 11, 13, 15, 18, 21, and 24 nm, respectively. (c) Intensity of A<sub>1</sub><sup>'</sup> mode at 117 cm<sup>-1</sup> as a function of thickness in as-exfoliated sheet and annealed sheets at 200, 300, and 400 °C.



**Figure S4.** EDS spectrum of as-exfoliated InSe nanosheets.

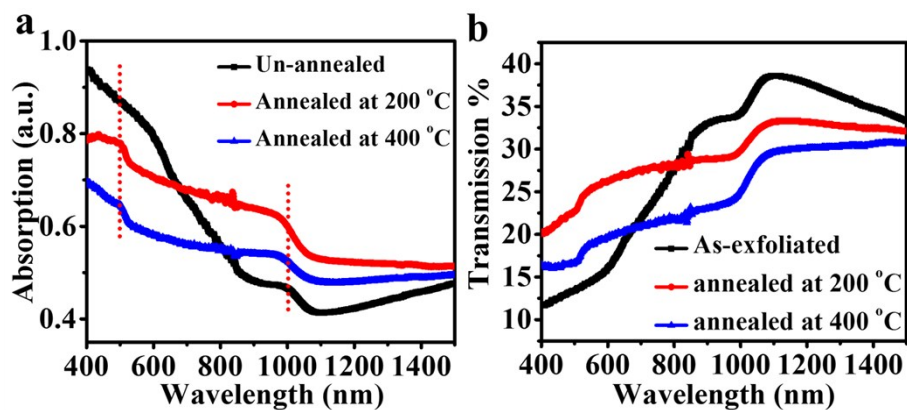
**Table S1.** Element composition and In/Se ratio of as-exfoliated and annealed sheets.

Samples	Atomic percentage In (%)	Atomic percentage Se (%)	Atomic ratio of In/Se
As-exfoliated	52.4	47.6	1.10
Annealed at 200 °C	47.2	52.8	0.87
Annealed at 300 °C	45.9	54.1	0.84
Annealed at 400 °C	45.3	54.7	0.83



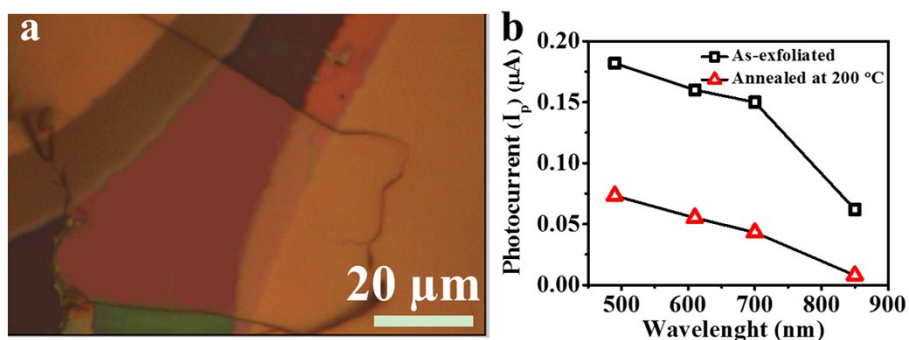
**Figure S5.** PL spectra of as-exfoliated InSe nanosheets and annealed sheets at 300 °C and 400 °C, respectively. The thickness from bottom to top as indicated by dotted arrows in both a and b are about 6, 9, 11, 13, 15, 18, 21, and 24 nm, respectively. (c)

PL peak intensity of  $\gamma$ -In<sub>2</sub>Se<sub>3</sub> as a function of thickness. (d) Intensity ratio of  $\frac{I_\gamma}{I_\beta}$  versus thickness at different annealing temperatures.



**Figure S6.** (a) Absorption and (b) transmission spectra of as-exfoliated InSe (black curve) and annealed samples at 200 (red curve) and 400 °C (blue curve), respectively.

InSe is exfoliated on the surface of quartz substrate.



**Figure S7.** (a) Optical image of InSe photodetector. (b) Photocurrents of as-exfoliated and annealed devices as a function of wavelength.

**Table S2.** Photodetector parameters of as-exfoliated and annealed sheet.

Photodetector	On/off ratio	Carrier mobility ( $\text{cm}^2 \text{v}^{-1} \text{s}^{-1}$ )	Photocurrent ( $\mu\text{A}$ )	Responsivity ( $\text{A/W}$ )	Detectivity (Jones)	Quantum efficiency (EQE) ( $\text{W/A}$ )
As-exfoliated InSe nanosheet	$5.51 \times 10^4$	10.32	0.182	$12.54 \times 10^4$	$1.72 \times 10^{15}$	$31.82 \times 10^4$
Annealed InSe nanosheet	$7.42 \times 10^3$	2.37	0.0732	$5.05 \times 10^4$	$1.30 \times 10^{15}$	$12.82 \times 10^4$