Supporting Information

Synthesis of low-symmetry 2D $Ge_{(1-x)}Sn_xSe_2$ alloy flakes with anisotropic optical response and birefringence

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Supporting Figures and Tables

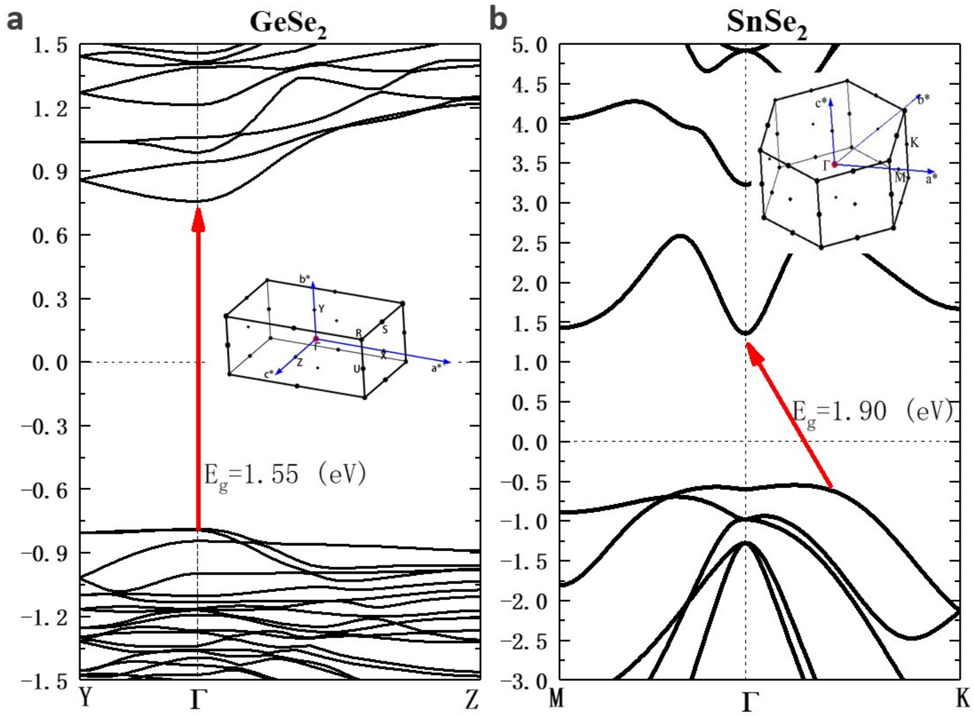

Fig. S1 Band structure and the first Brillouin zone of bulk $GeSe_2$ (a) and bulk $SnSe_2$ (b).

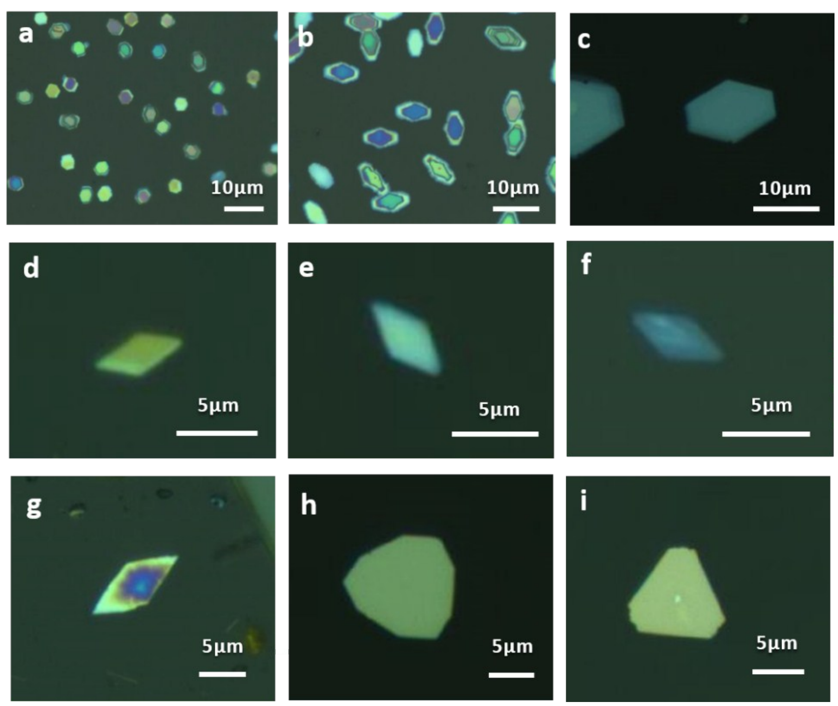

Fig. S2 (a-i) OM images of the $Ge_{(1-x)}\,Sn_xSe_2$ alloy flakes on the mica substrates under different growth conditions corresponding to Table S1.

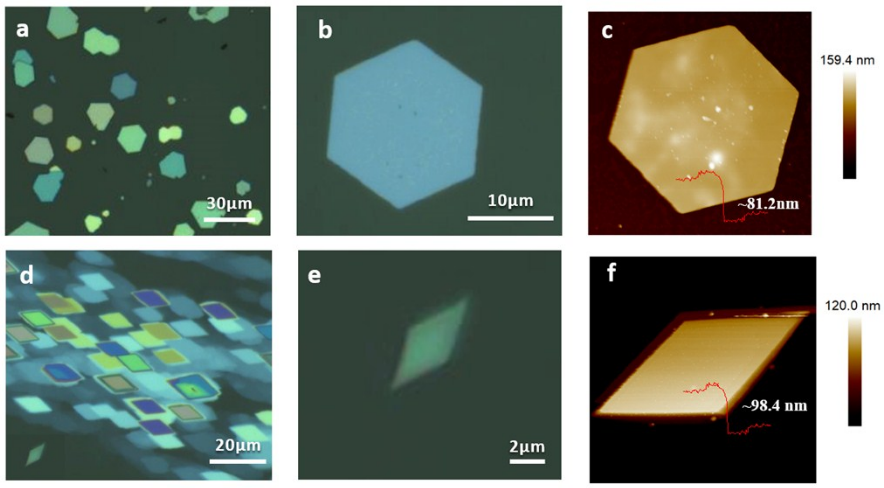

Fig. S3 (a-f) OM and AFM images of as-grown $SnSe_2$ (a-c) and $GeSe_2$ (d-f) flakes on the mica substrates.

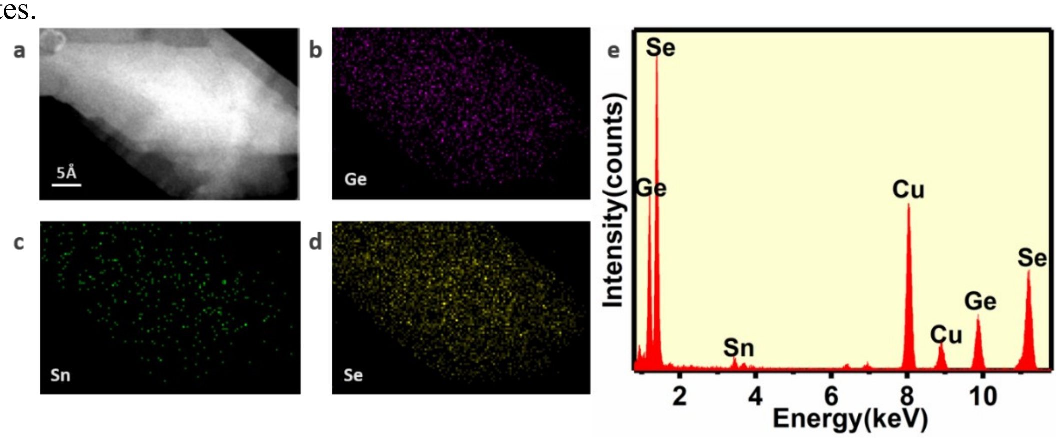

Fig. S4 EDS images of the $Ge_{0.86}Sn_{0.14}Se_2$ alloy flakes, which were synthesized at 500 ℃ for 10 min with the mixed gas (Ar/H_2) flow rate of 16 sccm, then cooling down to the room temperature naturally. (a) TEM image of a single as-grown alloy flake. (b-d) EDS mapping of Ge, Sn, and Se elements, respectively. (e) EDS spectrum of the as-grown flake.

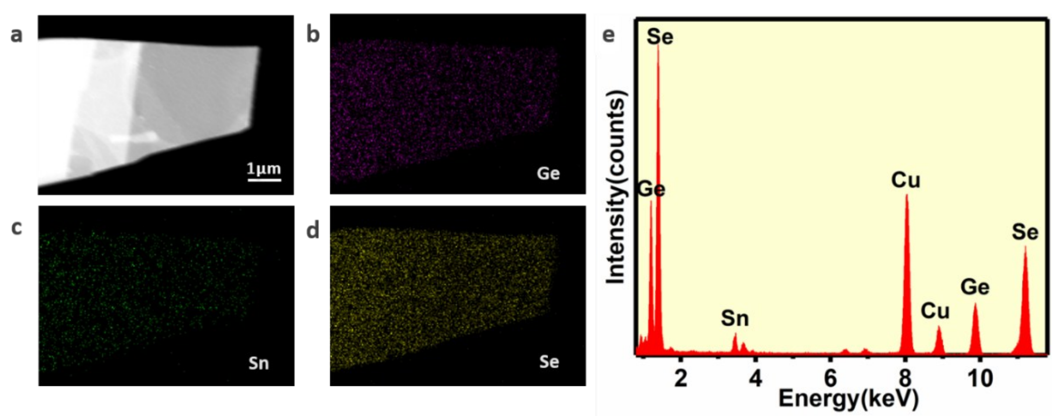

Fig. S5 EDS images of the $Ge_{0.74}Sn_{0.26}Se_2$ alloy flakes, which were synthesized at 550 ℃ for 10 min with the mixed gas (Ar/H_2) flow rate of 20 sccm, then cooling down to the room temperature naturally. (a) TEM image of a single as-grown alloy flake. (b-d) EDS mapping of Ge, Sn, and Se elements, respectively. (e) EDS spectrum of the as-grown flake.

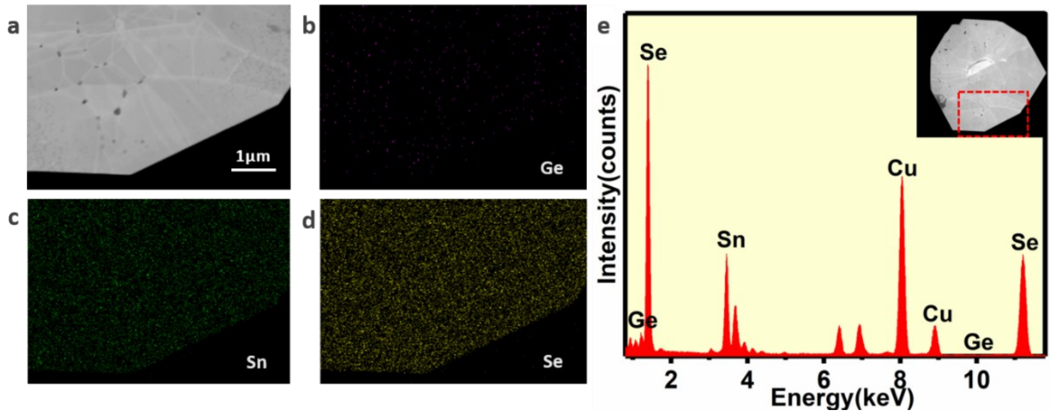

Fig. S6 EDS images of the $Ge_{0.03}Sn_{0.97}Se_2$ alloy flakes, which were synthesized at 550 ℃ for 10 min with the mixed gas (Ar/H_2) flow rate of 16 sccm, then cooling down to the room temperature naturally. (a) TEM image of a single as-grown alloy flake. (b-d) EDS mapping of Ge, Sn, and Se elements, respectively. (e) EDS spectrum of the as-grown flake.

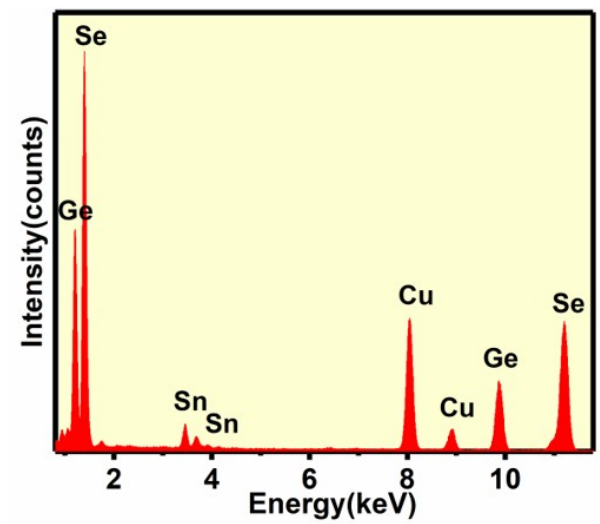

Fig. S7 EDS spectrum of the $Ge_{0.88}Sn_{0.12}Se_2$ flake.

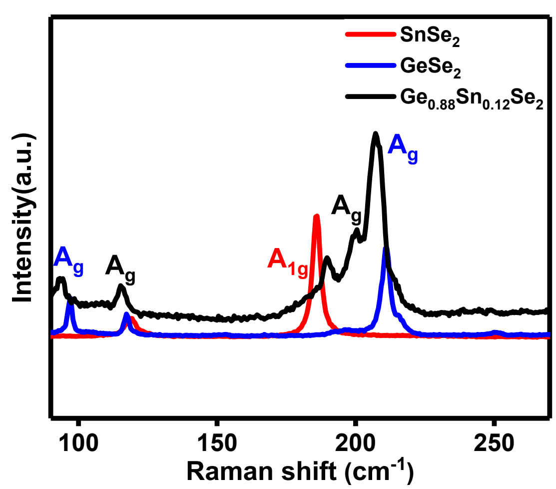

Fig. S8 Raman spectra of as-grown $SnSe_2$ (red line), $GeSe_2$ (blue line), and $Ge_{0.88}Sn_{0.12}Se_2$ (black line) flakes on the mica substrates.

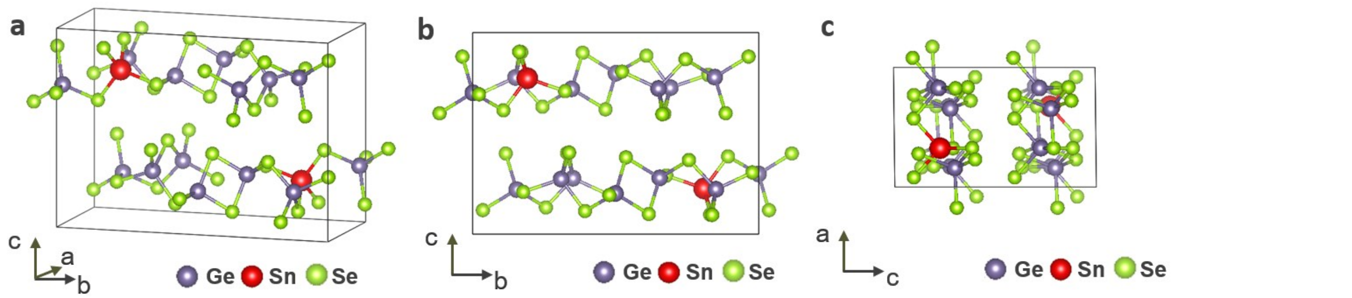

Fig. S9 (a-c) Schematic diagrams of the $Ge_{0.88}Sn_{0.12}Se_2$ crystal structure viewed from different directions.

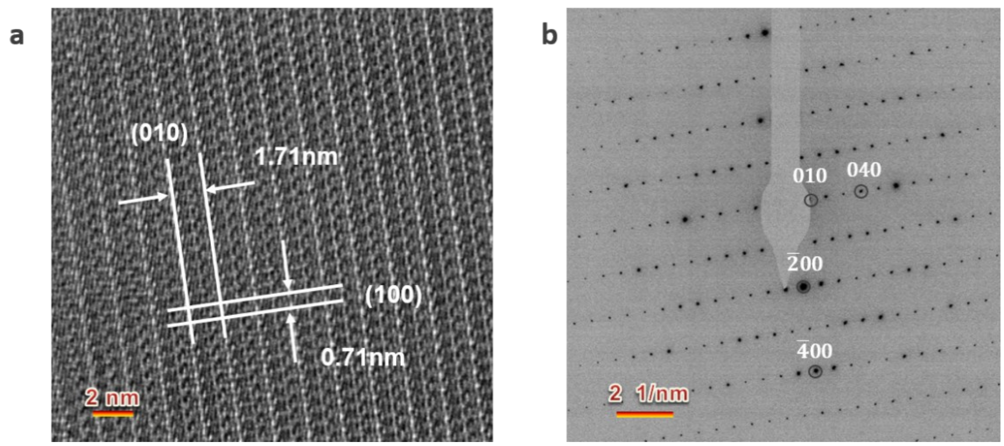

Fig. S10 (a, b) HRTEM (a) and SAED (b) images of the $Ge_{0.88}Sn_{0.12}Se_2$ flake.

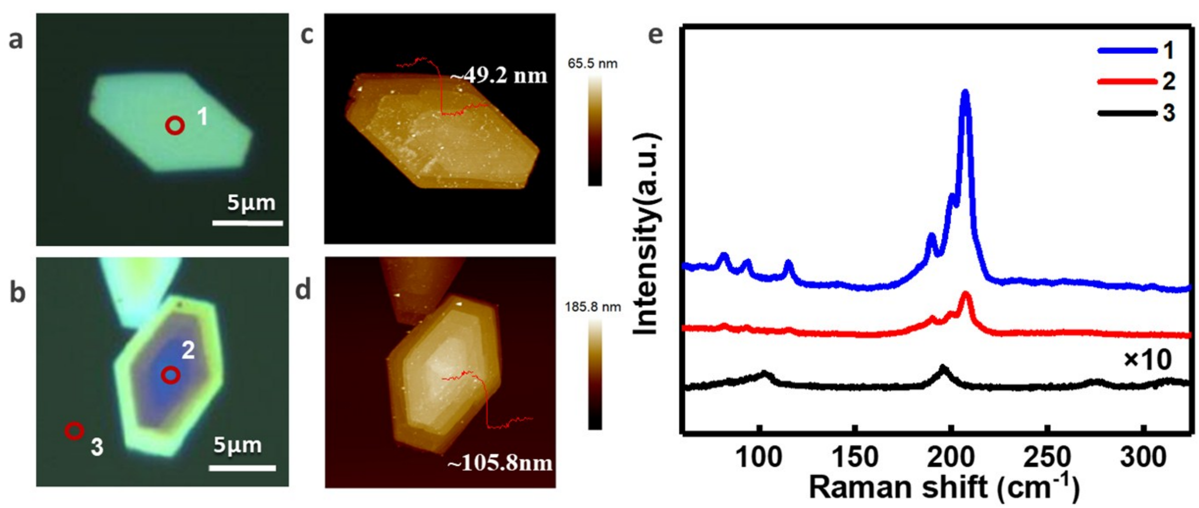

Fig. S11 (a-d) OM and AFM images of the 49.2 nm (a, c) and 105.8 nm (b, d) thick $Ge_{0.88}Sn_{0.12}Se_2$ flakes, respectively. (e) The corresponding Raman spectra of the $Ge_{0.88}Sn_{0.12}Se_2$ flakes with different thicknesses and the blank mica substrate. The spectrum of Area 3 was magnified ten times.

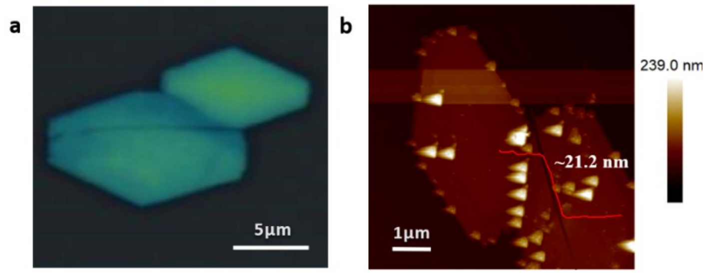

Fig. S12 (a) OM and (b) AFM images of a $Ge_{0.88}Sn_{0.12}Se_2$ flake used for the ADRDM characterization.

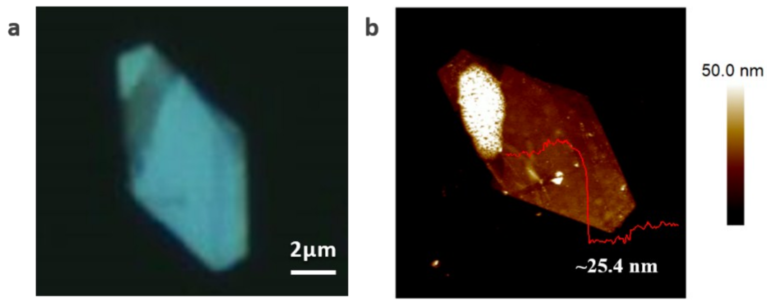

Fig. S13 (a) OM and (b) AFM images of a $Ge_{0.88}Sn_{0.12}Se_2$ flake on a quartz substrate.

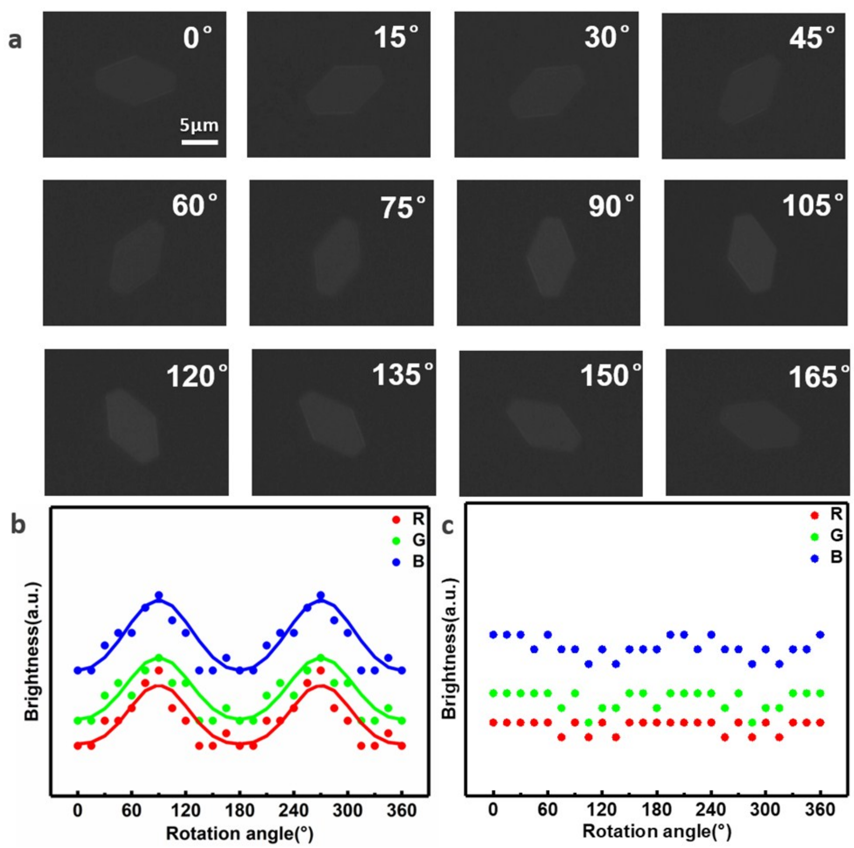

Fig. S14 (a) POM images of a $Ge_{0.88}Sn_{0.12}Se_2$ flake at different rotation angles (0 – 165°). (b, c) RGB optical brightness intensities of the reflected light from the $Ge_{0.88}Sn_{0.12}Se_2$ flake (b) and the blank 285 nm SiO_2/Si substrate (c), respectively.

Table S1. Different growth conditions of $Ge_{(1-x)}Sn_xSe_2$ alloy flakes.

No.	Holding temperature/℃	Holding time/min	Gas flow rate/sccm	Cooling method
1-a	515	7	16	rapid cooling
2-b	515	10	16	rapid cooling
3-c	515	10	16	natural cooling
4-d	500	30	12	natural cooling
5-e	500	30	16	natural cooling
6-f	500	30	20	natural cooling
7-g	500	10	16	natural cooling
8-h	530	10	16	natural cooling
9-i	550	10	16	natural cooling