Supporting Information for

Optical properties of thickness-controlled $PtSe_2$ studied via spectroscopic ellipsometry

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Details on Spectroscopic Ellipsometry measurement

The SE measurement was taken under room temperature at the incident angle of 65°. And the spot size of the ellipsometer is 25 μm×60 μm, while our samples are centimeter sized scale. During the measurement, we randomly selected different areas on each sample for measurement, and those corresponding Psi and Delta curves are almost completely consistent, which not only indicates the accuracy of the measurement, but also shows the pretty good uniformity of our samples. Fig. S1 shows the fitting result by Lorentz model. There is no difference between the measured experimental data (unfilled symbols) and calculated data (solid lines).

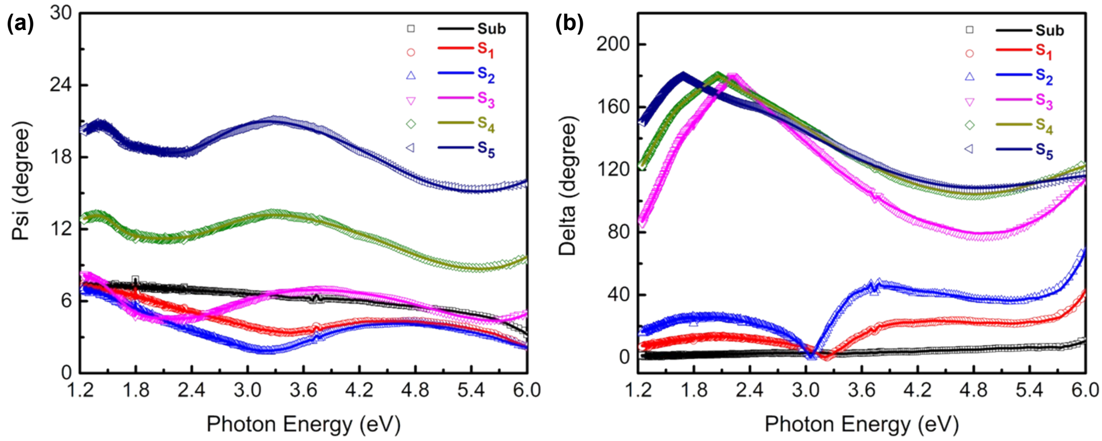

Fig. S1 Experimental and fitted SE data (a) Psi and (b) Delta of $PtSe_2$ films for various thickness at incident angle of 65°. The solid lines and unfilled symbols represent the fitted and the experimental data, respectively.

To check if there are any possible in-plane anisotropies, we rotated each sample's azimuthal orientation by every 15 degrees, from 0 to 75 degrees, to measure the ellipsometric data. The choice of rotation angle is based on the of tetragonal symmetry of sample structure. And as shown in Fig. S2, there is no obvious difference between each azimuthal orientation. Therefore, it is treated as in-plane isotropy in our data analysis.

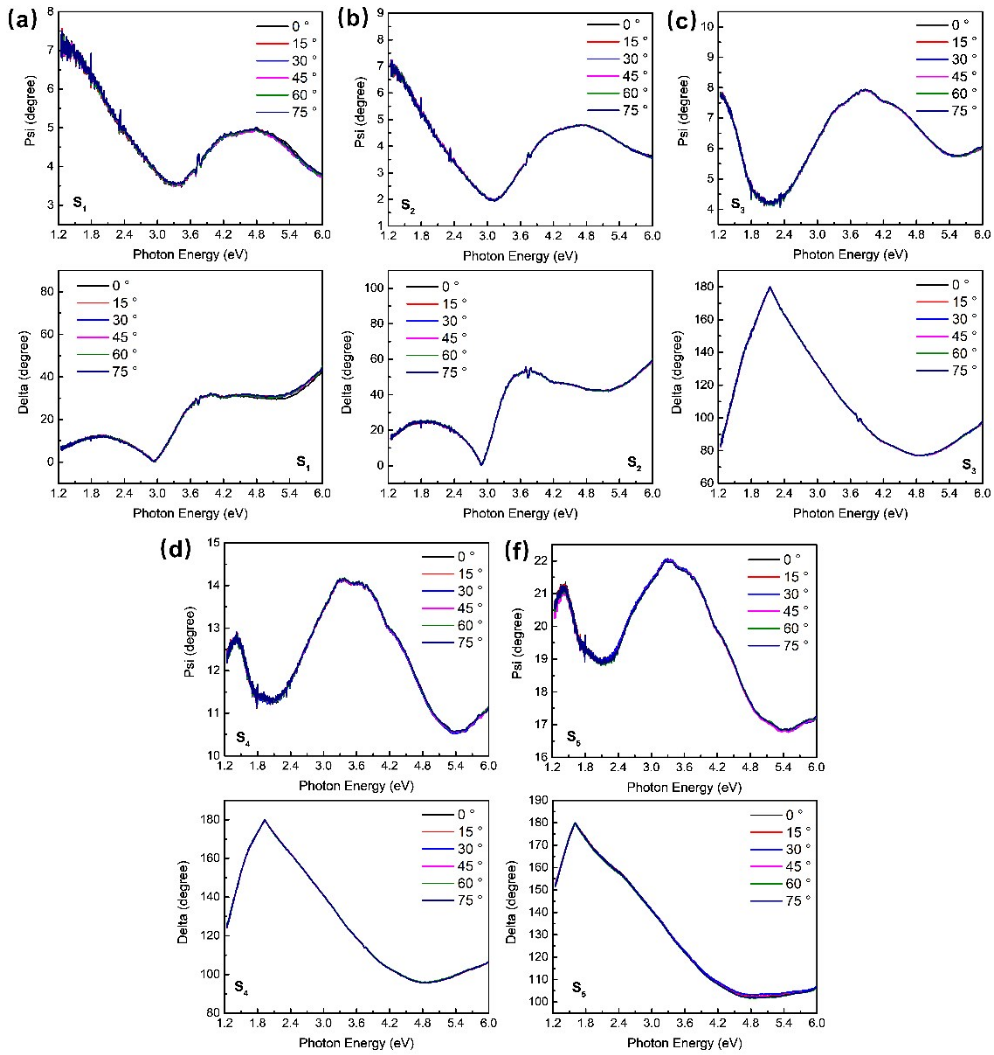

Fig. S2. Ellipsometric data of different azimuthal orientation. (a)-(f) is Psi and Delta data for Sample S_1 to S_5, respectively.

The AFM test

The AFM test was at air condition and working in the tapping mode. Fig.S3 shows the thickness obtained from AFM test which are consistent with the SE results. The sample S_1 is too thin to get a reliable thickness by AFM.

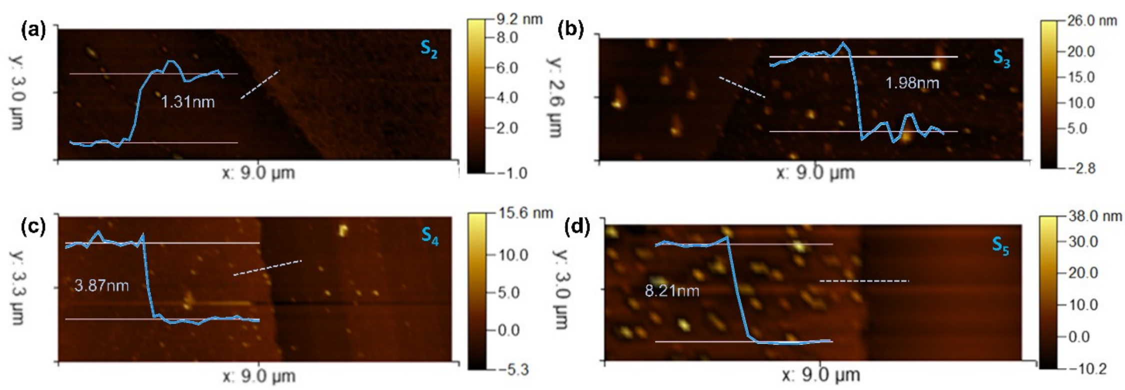

Fig. S3 Thickness measured by AFM. (a)-(d) are film thickness of sample S_2-S_5, respectively, measured along the marked blue dashed lines.

Absorption coefficient spectra and the Lorentz model parameters

The optical absorption coefficient spectra of five samples obtained via spectroscopic ellipsometry analysis is shown in figure S4.

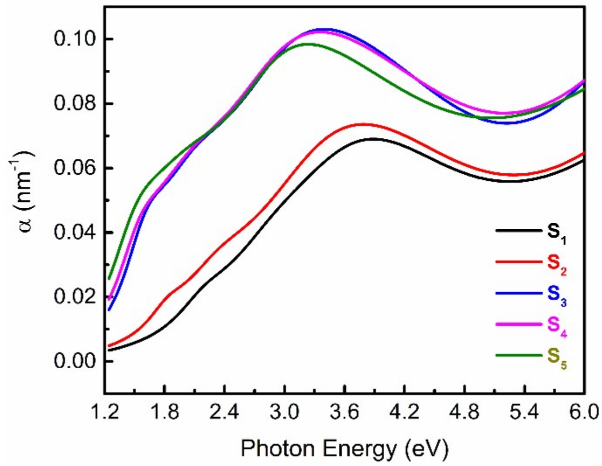

Fig. S4 Absorption coefficient spectrum of $PtSe_2$ thin films which also shows the thickness-dependence.

The absorption coefficient is calculated by:

$$\alpha = \frac{4\pi k}{\lambda} = \frac{4\pi}{\lambda}\left(\frac{-\varepsilon_1 + \left(\varepsilon_1^2 + \varepsilon_2^2\right)^{1/2}}{2}\right)^{1/2} \tag{1}$$

where k is the extinction coefficient, and λ is the wavelength, ε_1 and ε_2 are real and imaginary part of dielectric function, respectively. The dielectric function is extracted by the multi-oscillator Lorentz model:

$$\varepsilon = \varepsilon_1 + \mathrm{j}\varepsilon_2 = \varepsilon_\infty\left(1 + \sum_{i=1} \frac{A_i^2}{C_i^2 - E^2 - \mathrm{j}v_i E}\right) \tag{2}$$

where E is the photon energy, ε_∞ is the high-frequency dielectric constant, and A_i, C_i, and v_i are the amplitude, center energy and damping coefficient of each oscillator.

Table S1 lists the parameters extracted from Lorentz model. For the accuracy of the fitting, the same initial value was set for each fitting process, and the fitting was performed at least 10 times, thereby obtaining the error bars. Besides, a point-by-point method was also employed to check the results of Lorentz model. The point-by-point fit is a mathematical inversion method at each measured point of the SE data. In our fitting process, a comparison was made between the Lorentz model extracted dielectric function and mathematical inversion inverted results. We changed the number of oscillators and repeat the above analysis until the difference was not significant. And finally, a four oscillators Lorentz model was adopted.

Table S1 Lorentz model parameters

Sample		S_1	S_2	S_3	S_4	S_5
ε_∞	-	1.79±0.15	1.96±0.06	2.64±0.01	1.85±0.01	0.97±0.05
Oscillator 1	C_1/eV	2.18±0.11	1.81±0.01	1.58±0.02	1.52±0.01	1.47±0.01
	A_1/eV	1.46±0.43	1.09±0.07	2.43±0.04	2.80±0.04	4.55±0.26
	v_1/eV	0.70±0.17	0.49±0.03	0.56±0.01	0.58±0.04	0.67±0.03

Oscillator 2	C_2/eV	2.83±0.36	2.30±0.02	2.03±0.01	1.98±0.01	1.95±0.03
	A_2/eV	2.82±1.01	2.08±0.10	3.18±0.02	4.10±0.08	5.89±0.11
	v_2/eV	1.29±0.46	0.94±0.05	1.00±0.02	1.12±0.01	1.28±0.60
Oscillator 3	C_3/eV	3.56±0.13	3.32±0.01	2.92±0.01	2.88±0.01	2.84±0.02
	A_3/eV	4.87±1.08	5.59±0.11	5.46±0.01	6.22±0.03	6.89±0.19
	v_3/eV	1.89±0.12	2.02±0.03	1.82±0.02	1.77±0.01	1.49±0.04
Oscillator 4	C_4/eV	7.90±0.19	7.57±0.10	6.84±0.02	7.34±0.04	8.12±0.21
	A_4/eV	9.60±0.87	8.07±0.34	6.14±0.02	9.10±0.07	15.76±0.76
	v_4/eV	4.70±0.29	3.83±0.19	3.03±0.01	4.10±0.07	6.47±0.61