Electronic Supplementary Information of "Strong Anisotropic Third Harmonic Generation in Layered Violet Phosphorus"

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The supporting file contains three sections to mainly illustrate the technical details of experimental measurements.

1. THG spectra of K9 glass substrate.

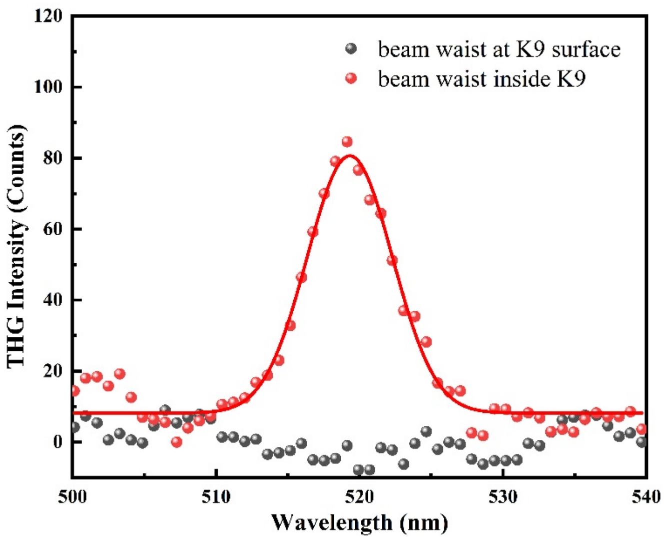

Figure S1. THG spectra, when the fundamental beam waist was focused on the surface (black dots) and inside (red dots) of the K9 glass substrate. The fundamental power was fixed 16 mW and the integration time of the spectrometer was set to 5 s. The solid red curve is a Gaussian fitting.

The THG of the K9 glass was observed (red dot) by increasing the fundamental power to 16 mW while slightly shifting the beam waist of the fundamental laser from the surface to the inside of the K9 glass substrate. To be specific, the THG spectrum when the fundamental beam waist was focused on the surface of the glass substrate (black dots) shows that the corresponding THG signal is too weak to be observed by our experimental setup. By focusing the fundamental beam waist into the glass substrate, a weak THG signal (red dots) can be observed.

2. AFM of layered VP samples.

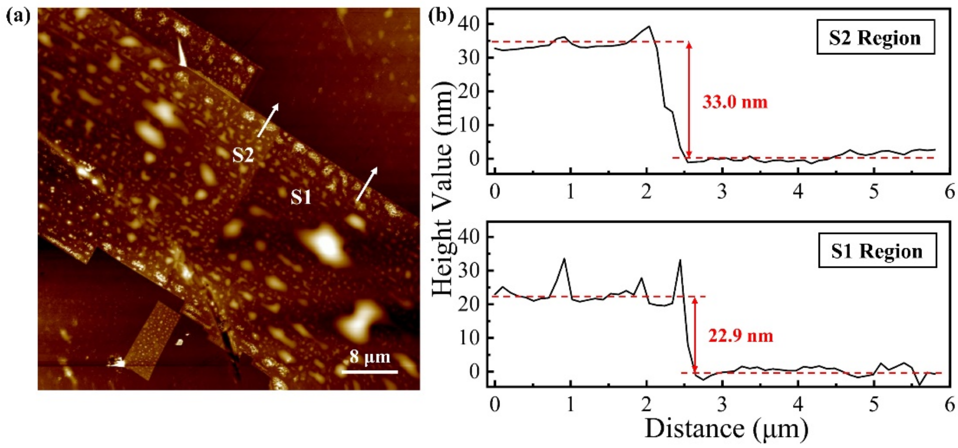

Figure S2. (a) Atomic force microscopy image of layered VP samples. (b) Thickness profiles of region S1 and S2, along the two white arrows in (a).

Figure S2(a) demonstrates the AFM image of layered VP samples investigated in the main text. Thickness profiles along the white arrows were extracted to obtain the thicknesses of region S1 and S2, as shown in Figure S2(b). The dashed red arrows represent the averaged thicknesses. In this way, the thicknesses of region S1 and S2 were about 22.9 nm and 33.0 nm, respectively.

3. Confocal detection for optical absorptance measurement.

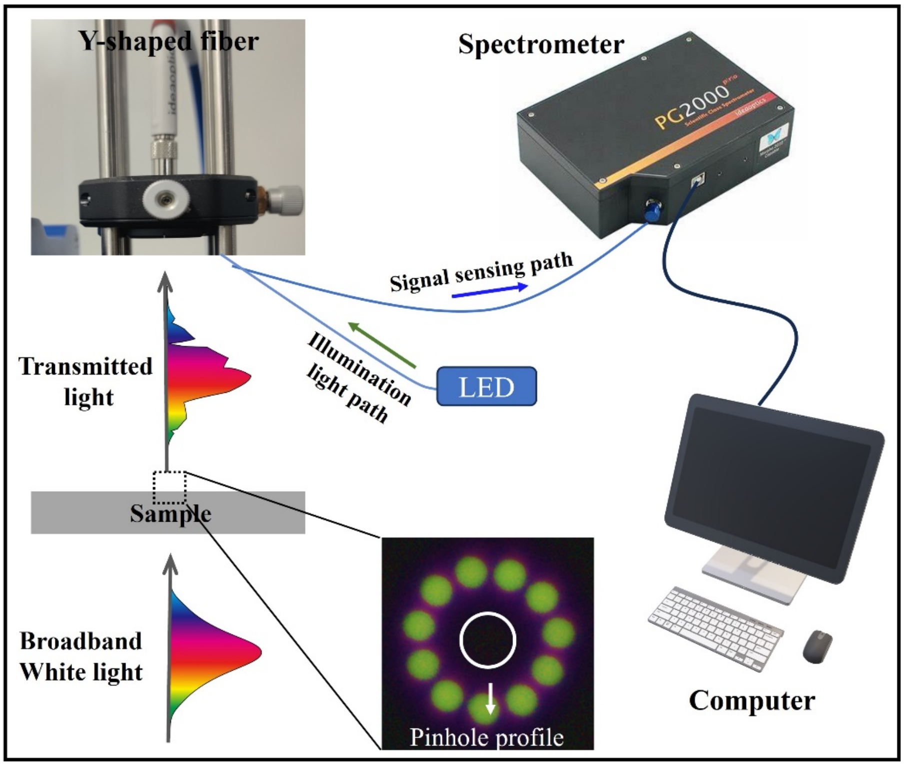

Figure S3. Schematic illustration of optical absorptance measurement.

Figure S3 illustrates the experimental setup of optical absorptance in the main text. A broadband white light illuminates the sample, and the transmitted light is collected by a Y-shaped fiber port and sent to a spectrometer, where the computer records and analyses the broadband white light spectrum and the transmitted light spectrum of the sample to obtain the optical absorptance. The Y-shaped fiber consists of a central core surrounded by 11 external cores. The central core of the Y-shaped fiber served as a pinhole of confocal detection. The central core is connected to a spectrometer for light spectrum sensing, and the outer cores are connected to a LED light source to finely localize the light sensing positions of the sample or the substrate.