Supplementary Information

Epitaxial growth of single-crystal violet phosphorus flakes on silicon substrates

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Experimental Session

Scanning electron microscopy (SEM). The morphologies of the as-produced violet phosphorus were taken on a FEI Quanta 250F scanning electron microscope.

Powder X-ray diffraction (XRD). X-ray diffraction patterns were obtained from a Bruker D2 PHASER using Cu/Kαradiation (λ=1.5418 Å) at 40 kV and 30 mA.

Raman spectroscopy. Raman spectroscopy wastaken in a back-scattering geometry using a single monochromator with a microscope (Reinishaw inVia) equipped with CCD array detector (1024×256 pixels, cooled to -70ºC) and an edge filter. A piece of violet phosphorus crystal was irradiated with a 633nm laser.

TEM, HRTEM and SAED measurements. TEM, HRTEM images and SAED patterns were acquired using Talos F200X electron microscope with an acceleration voltage of 200 kV. The violet phosphorus sonicated in ethanol for 15 min was dropped onto copper grids coated with ultrathin amorphous carbon films and then dried under ambient conditions.

Lateral size measurements. The image J software was used for size measurements.

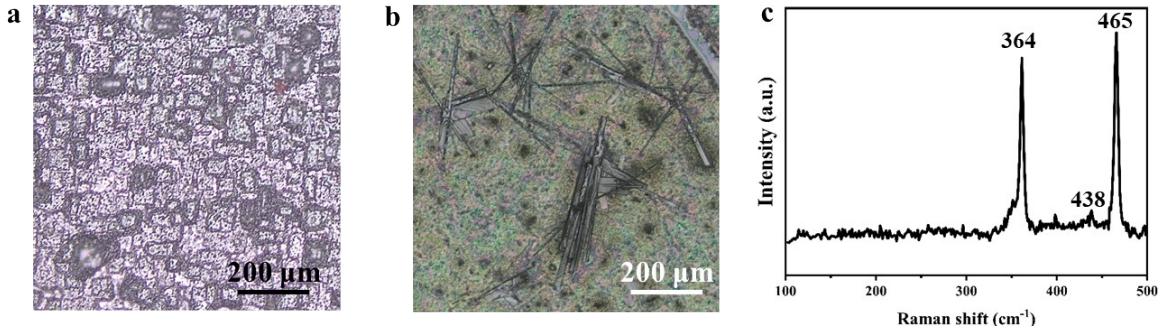

Fig S1. Optical images of the silicon substrate after 10 h reaction with a temperature gradient of (a) 0 ℃ and (b) 20 ℃. (c) Raman spectrum of samples grown on the silicon substrate after 10 h reaction with a temperature gradient of 20 ℃.

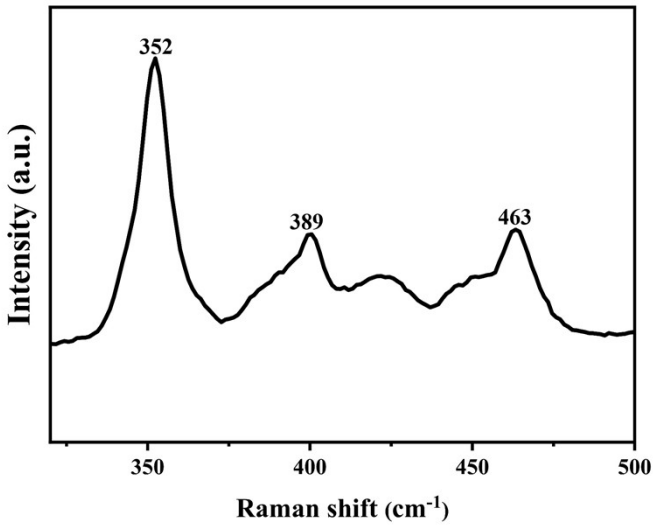

Fig S2. Raman spectrum of particles grown on bare silicon substrate.

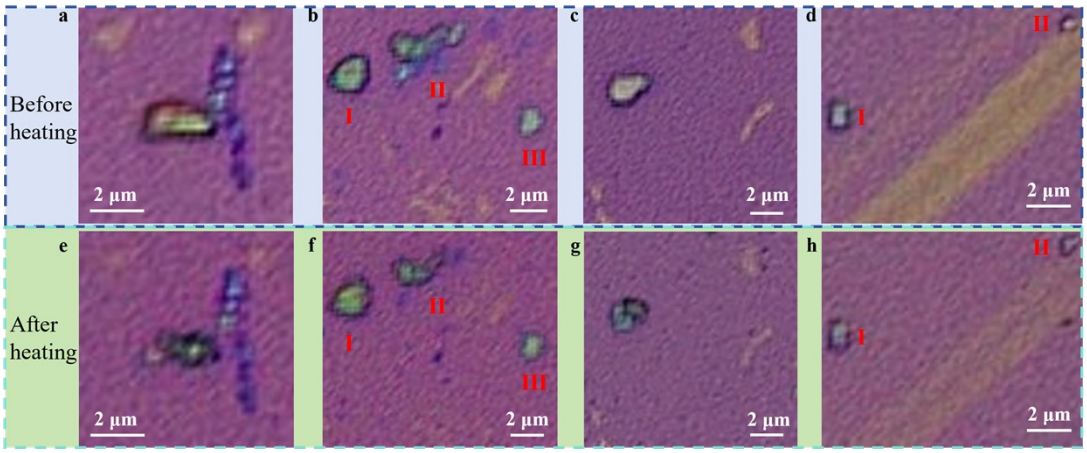

Fig S3. Optical images of VP flakes on silicon substrate (a-c) before and (e-f) after heating process according epitaxial growth experiment without any reactants for 1 h.

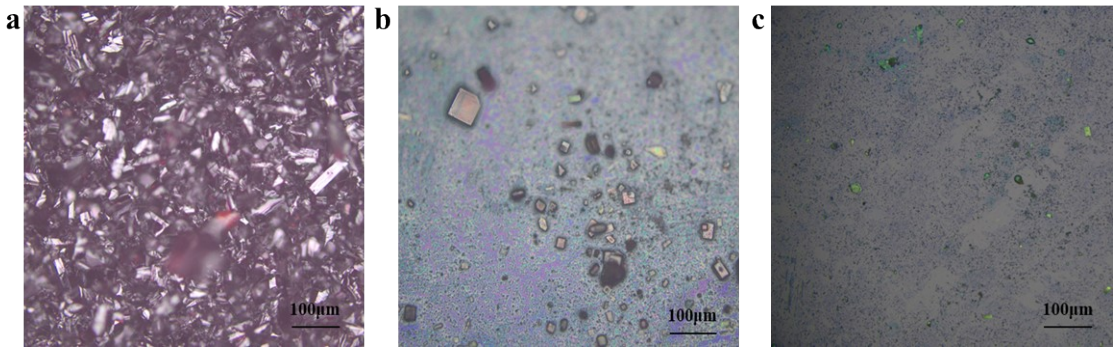

Fig S4. Optical images of the as-grown VP flakes on silicon substrates with different concentrated seeds after CVT growth for 10 h. (a)0.06 mg/mL (b) 0.03 mg/mL and (c) 0.015 mg/mL

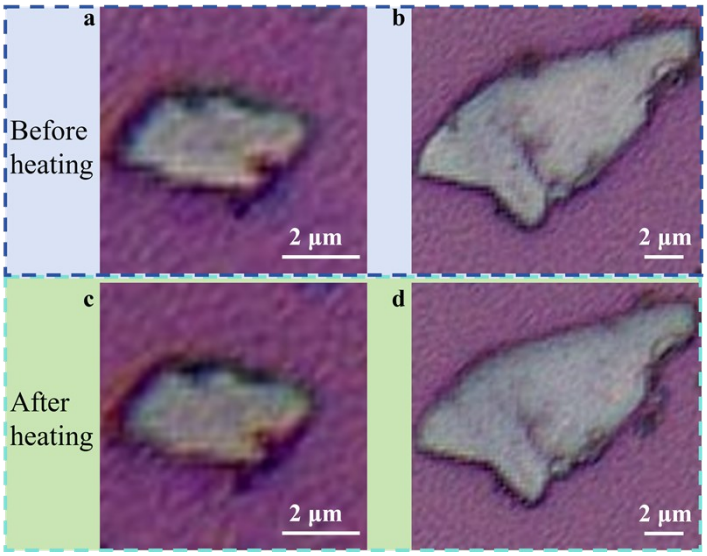

Fig S5. Optical images of thick VP flakes on silicon substrate (a-b) before and (c-d) after heating process for 1 h. (a, c) thickness of 1.9 μm, (b, d) thickness of 2.1 μm.

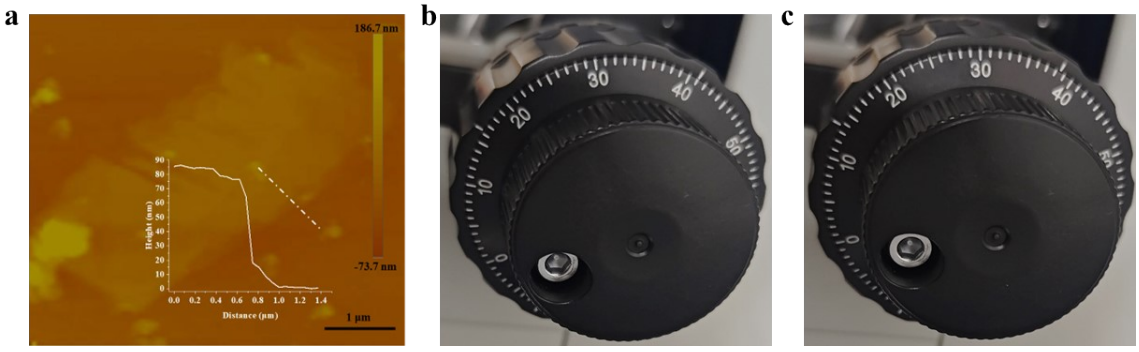

Fig S6. (a) AFM images and height profiles of the violet phosphorene nanosheet seeds on SiO_2/Si substrate. (b-c) The focus positions between substrate surface and flake surface of violet phosphorus flakes grown on the silicon substrate under optical microscope.

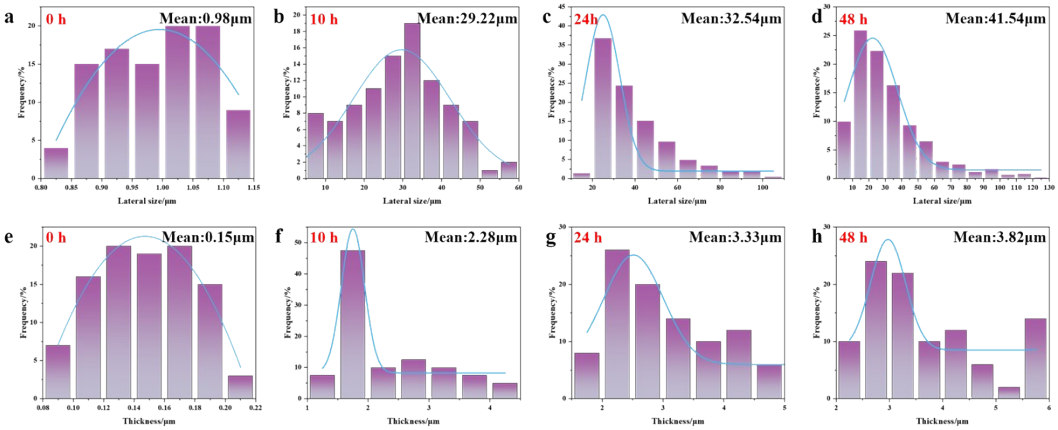

Fig S7. Lateral size distribution of the as-grown VP flakes on silicon substrates after reaction for (a) 0 h, (b) 10 h, (c) 24 h and (d) 48 h. Thickness distribution of the as-grown VP flakes on silicon substrates after reaction for (e) 0 h, (f) 10 h, (g) 24 h and (h) 48 h.

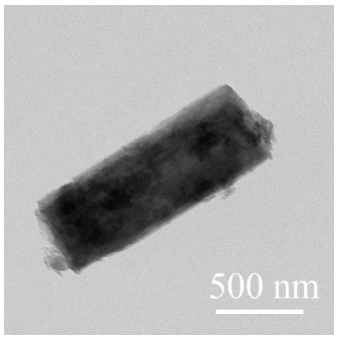

Fig S8. TEM image of a violet phosphorene nanosheet exfoliated from the as-grown violet phosphorus flakes.

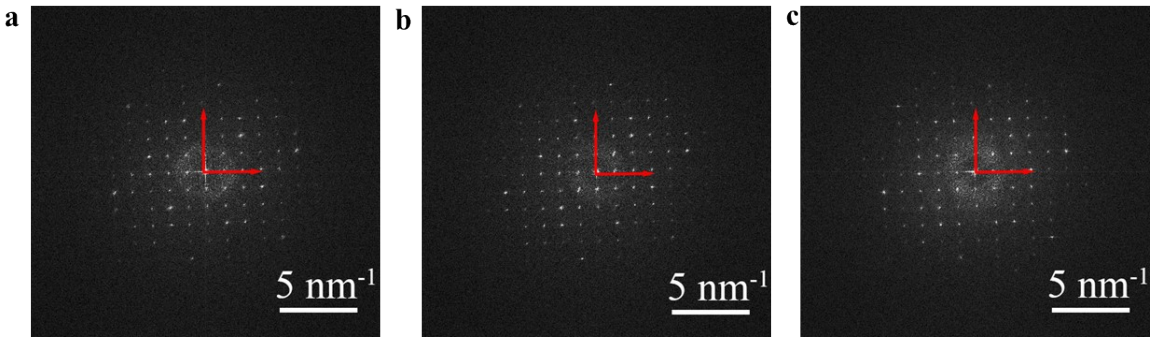

Fig S9. Selected-area electron diffraction patterns (SAED) patterns with the zone axis [001] of violet phosphorus with three different regions.

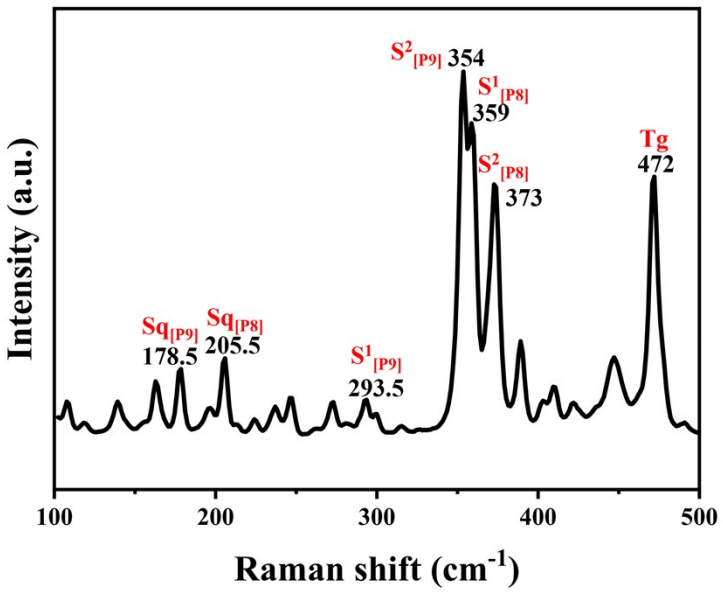

Fig S10. Raman spectrum of violet phosphorus crystals.