## **Supporting Information:**

Vertical Bridgman Growth and Optical Properties of CdSiP<sub>2</sub> Crystals

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- Fig.1S. Photograph of Polycrystalline CSP synthesized using the single-temperature zone furnace.
- Fig.2S. Powder X-ray diffraction spectra for the products reacted at different temperature.
- Fig.3S. High-resolution X-ray diffraction spectrum on the (004)-faced plate of CSP crystal.
- Fig.4S. Comparison of EDS components analysis between the inclusions and the CSP crystal.
- Fig.5S. Polarized transmission spectra of CSP for *o* and *e* light.
- Fig.6S.Comparison of refractive index between our fitted results and the results reported in Ref. 25
- Fig.7S.Comparison of phase matching curves between our results and the results reported in Ref. 25



Fig. 1S Polycrystalline CSP synthesized using the single-temperature zone furnace

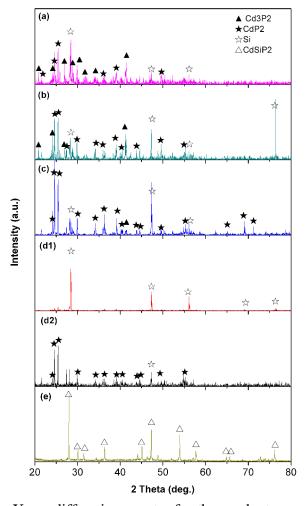


Fig. 2S Powder X-ray diffraction spectra for the products reacted at different temperature: (a) 500 °C, (b) 650 °C, (c) 700 °C, (d1) 780 °C (products at the top of the ample), (d2) 780 °C (products at the bottom of the ample) (e) 1050 °C.

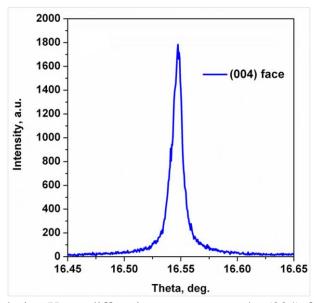
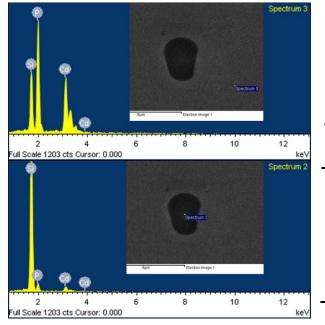


Fig.3S. High-resolution X-ray diffraction spectrum on the (004)-faced plate of CSP crystal.



Element	Weight%	Atomic%
Si	13.02	23.30
P	32.17	52.19
Cd	54.81	24.51
Totals	100.00	100

Element	Weight%	Atomic%
Si	84.48	91.61
P	5.78	5.77
Cd	9.66	2.62
Totals	100.00	100.00

Fig. 4S Comparison of EDS components analysis between the inclusions and the CSP crystal. The inset shows the micro-photograph of one inclusion and the position of the measurement.

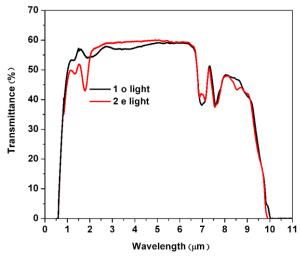


Fig. 5S Polarized transmission spectra for o- and e- light.

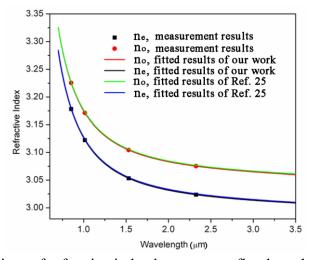


Fig. 6S Comparison of refractive index between our fitted results and the results reported in Ref. 25

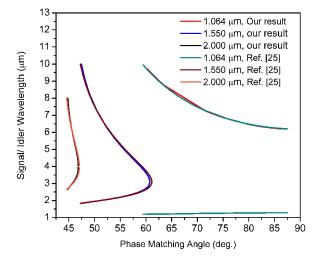


Fig. 7S Comparison of phase matching curves between our results and the results reported in Ref. 25