

Supplemental material for

Ultrafast nonlinear optical absorption and carrier dynamics of CrPS<sub>4</sub> thin  
films

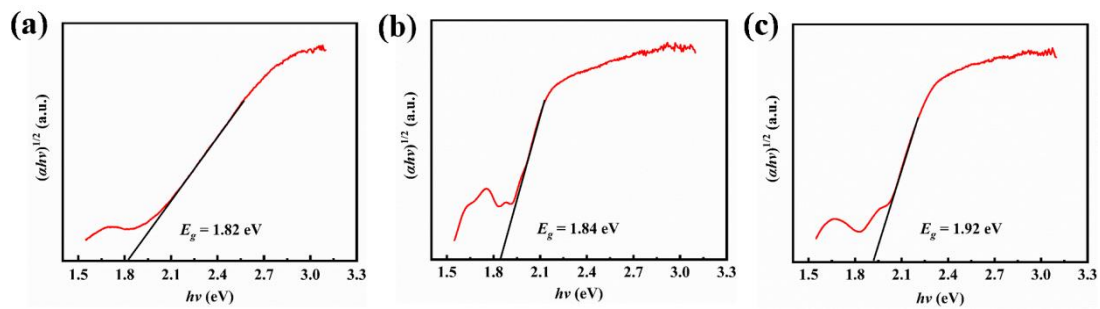
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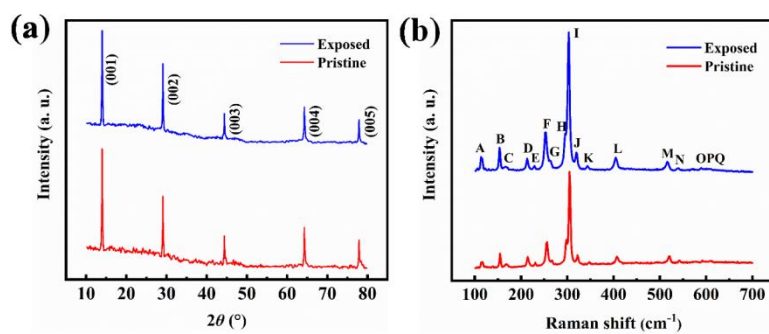
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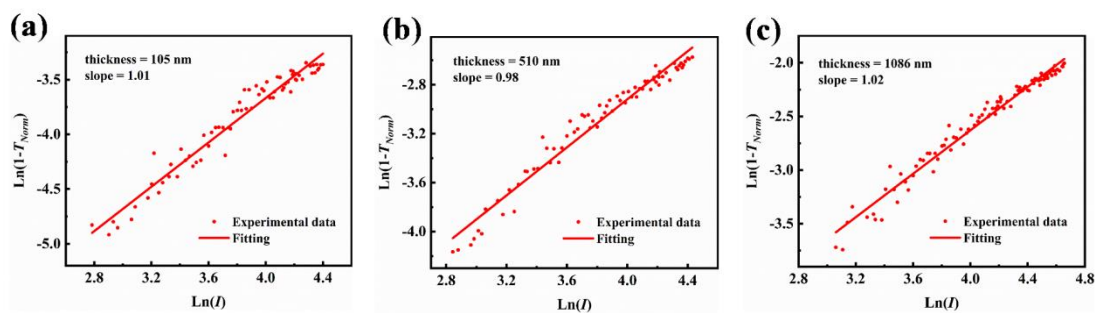
<sup>\*</sup>[sunlx@mail.sitp.ac.cn](mailto:sunlx@mail.sitp.ac.cn)



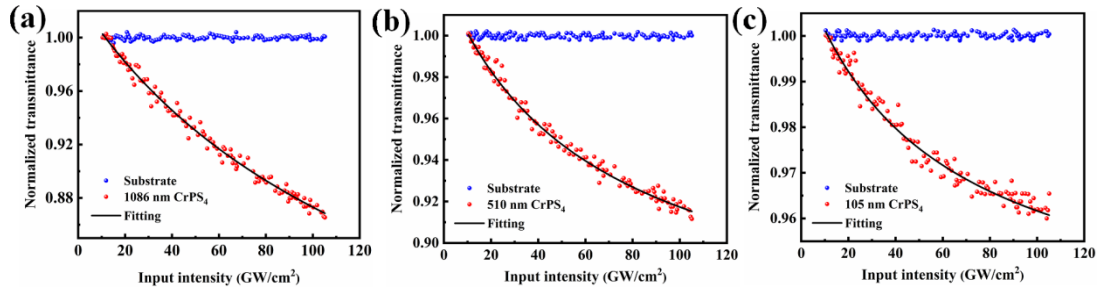
**Fig. S1.** The bandgap of CrPS<sub>4</sub> with different thicknesses. (a) 105nm, (b) 510 nm, and (c) 1086 nm.



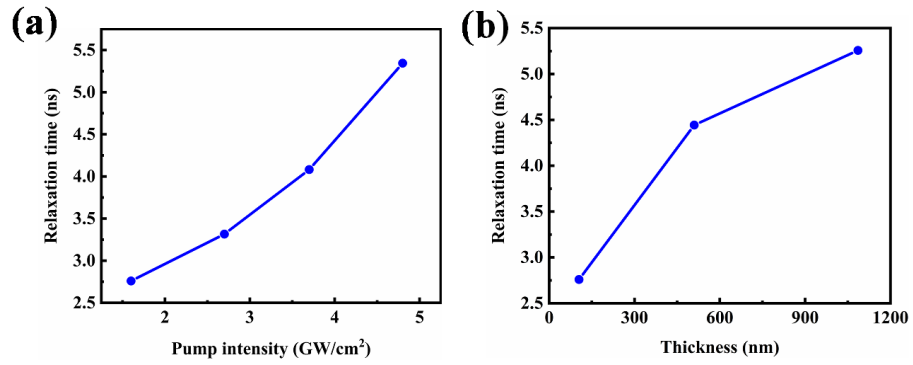
**FIG. S2.** The comparison of (a) XRD pattern and (b) Raman spectra of CrPS<sub>4</sub> before and after air exposure.



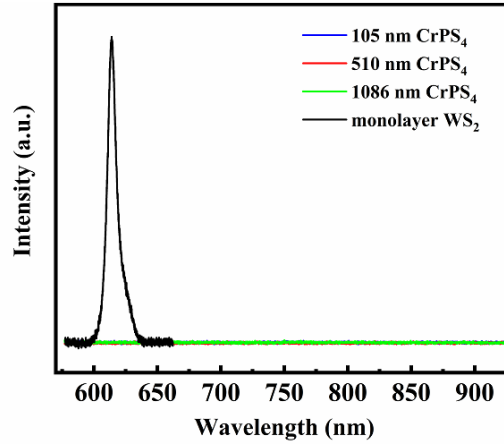
**Fig. S3.** Plots of  $\ln(1 - T_{Norm})$  versus  $\ln(I)$  of (a) 105 nm, (b) 510 nm, and (c) 1086 nm CrPS<sub>4</sub>.



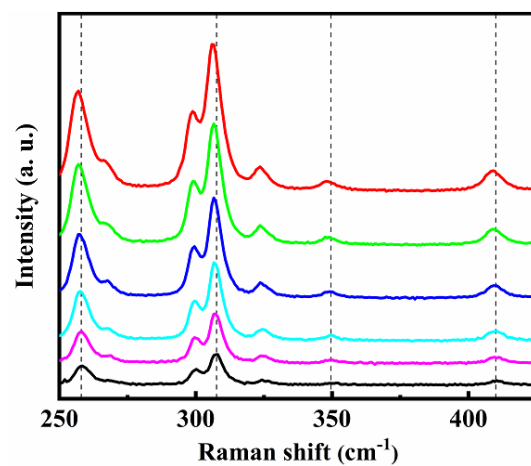
**FIG. S4.** Nonlinear absorption results of CrPS<sub>4</sub> and sapphire substrate.



**Fig. S5.** Relaxation time  $\tau_3$  with different (a) pump intensity and (b) thickness of CrPS<sub>4</sub>.



**Fig. S6.** Photoluminescence spectra of monolayer WS<sub>2</sub> and CrPS<sub>4</sub> at different thicknesses.



**Fig. S7.** Raman spectra from 250 to 425 cm<sup>-1</sup> excited at 532 nm under different powers corresponding to Fig. 5(b).