## Supplementary Information

## Ultra-high nonlinear absorption coefficients based on multiphoton-excited self-trapped excitons in perovskiteinspired copper halides

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Fig. S1 (a) XRD pattern of thermal-evaporated  $CsCu_2I_3$  thin film. (b) XRD pattern of thermal-evaporated  $Cs_3Cu_2I_5$  thin film.



Fig. S2 (a) PL and PLE spectra for thermal-evaporated  $CsCu_2I_3$  thin film. (b) PL and PLE spectra for thermal-evaporated  $Cs_3Cu_2I_5$  thin film.



Fig. S3 (a) TRPL decay spectra for thermal-evaporated  $CsCu_2I_3$  thin film. (b) TRPL decay spectra for thermal-evaporated  $Cs_3Cu_2I_5$  thin film.



Fig. S4 (a) Upconversion PL spectra excited by 840 nm laser for the thermal-evaporated  $CsCu_2I_3$  thin film. Inset is dependence of the upconversion PL intensity on the laser pulse energy for the  $CsCu_2I_3$  thin film. (b) upconversion PL spectra excited by 820 nm laser for the thermal-evaporated  $Cs_3Cu_2I_5$  thin film. Inset is dependence of the upconversion PL intensity on the laser pulse energy for the  $Cs_3Cu_2I_5$  thin film.