

**Electronic Supplementary Information for**

**Distinct Ultrafast Carrier Dynamics of  $\alpha$ -In<sub>2</sub>Se<sub>3</sub> and  $\beta$ -In<sub>2</sub>Se<sub>3</sub>:  
Contributions from Band Filling and Band Gap Renormalization**

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We used a home-built micro-absorption spectroscope to measure the optical absorption of the In<sub>2</sub>Se<sub>3</sub> crystal. According to Tauc et al.,<sup>1</sup> the absorption coefficient that near the

absorption edge of a direct-gap semiconductor is  $\alpha = A \frac{(\hbar\omega - E_g^{opt})^{1/2}}{\hbar\omega}$ , where  $\alpha$  is the absorption coefficient of In<sub>2</sub>Se<sub>3</sub> that can be calculated from its optical transmittance ( $T$ )

and its thickness ( $d$ ) as  $\alpha(\lambda) = -\frac{\log_{10}(T(\lambda))}{d}$ ,  $A$  is a material-dependent constant and  $E_g^{opt}$  is the optical energy gap of the material. The thickness of the In<sub>2</sub>Se<sub>3</sub> crystal used in our experiment was 42 nm. Therefore, the wavelength-dependent optical transmission for  $\alpha$ -In<sub>2</sub>Se<sub>3</sub> was shown in Fig. S1(a). The corresponding Tauc plots of optical absorption of  $\alpha$ -In<sub>2</sub>Se<sub>3</sub> was shown in Fig. S1(b). We found that the direct optical bandgap of  $\alpha$ -In<sub>2</sub>Se<sub>3</sub> is 1.43 eV, which was consistent with previous.<sup>2-3</sup>

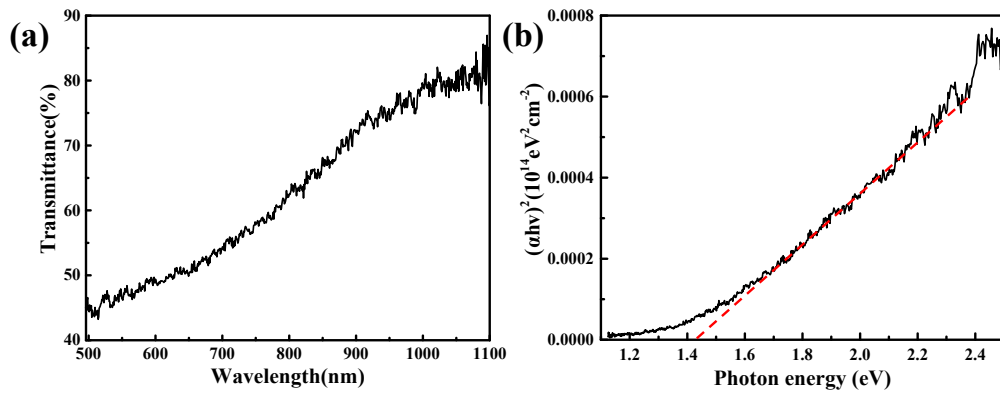


Fig. S1 (a) Wavelength-dependent optical transmission for  $\alpha$ - $\text{In}_2\text{Se}_3$ . (b) Tauc plots of optical absorption in  $\alpha$ - $\text{In}_2\text{Se}_3$ , the red dotted lines show the Tauc extrapolation of the absorption edge.

After annealing the  $\text{In}_2\text{Se}_3$  flake shown in Fig. 1(a) at 200 °C for 2 hours in argon atmosphere, the  $\alpha$ - $\text{In}_2\text{Se}_3$  was supposed to be converted to  $\beta$  phase.<sup>4</sup> Then we measured the optical absorption of  $\beta$ - $\text{In}_2\text{Se}_3$ . The wavelength-dependent optical transmission for  $\beta$ - $\text{In}_2\text{Se}_3$  was shown in Fig. S2(a). The corresponding Tauc plots of optical absorption of  $\beta$ - $\text{In}_2\text{Se}_3$  was shown in Fig. S2(b). We found that the direct optical bandgap of  $\beta$ - $\text{In}_2\text{Se}_3$  is 1.55 eV, which was consistent with previous.<sup>2-3</sup>

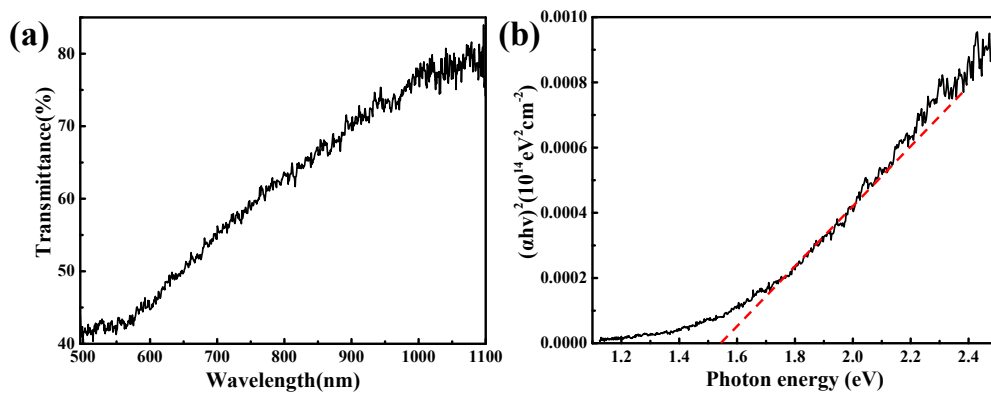


Fig. S2 (a) Wavelength-dependent optical transmission for  $\beta$ - $\text{In}_2\text{Se}_3$ . (b) Tauc plots of optical absorption in  $\beta$ - $\text{In}_2\text{Se}_3$ , the red dotted lines show the Tauc extrapolation of the absorption edge.

## References

- 1 J. Tauc, R. Grigorovici and A. Vancu, *Phys. Status Solidi B*, 1966, **15**, 627-637.
- 2 C. Julien, M. Eddrief, K. Kambas and M. Balkanski, *Thin Solid Films*, 1986, **137**, 27-37.
- 3 C. H. Ho, C. H. Lin, Y. P. Wang, Y. C. Chen, S. H. Chen and Y. S. Huang, *ACS Appl. Mater. Inter.*, 2013, **5**, 2269-2277.
- 4 X. Tao and Y. Gu, *Nano Lett.*, 2013, **13**, 3501-3505.