## Supplementary Information: Electrically switchable anti-ferroelectric bilayer In<sub>2</sub>Se<sub>3</sub> based opto-spintronics device

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**Figure S1** The spin-dependent photoresponse versus the photon energy  $E_{ph}$  of the circular polarized light. (a,c) The calculated spin dependent photoresponse for atomic structure  $\alpha_{out}$  when the photon polarization is fixed at  $\phi = 45^{\circ}$  and  $\phi = -45^{\circ}$ , respectively. (b,d) The calculated spin dependent photoresponse for atomic structure  $\alpha_{in}$  when the photon polarization is fixed at  $\phi = 45^{\circ}$  and  $\phi = -45^{\circ}$ , respectively. (e,f) The calculated spin dependent relative photoresponse change  $n_s$  versus the photon energy when the photon polarization is fixed at  $\phi = 45^{\circ}$  and  $\phi = -45^{\circ}$ , respectively. Here, the red and blue lines represent the spin up and spin down components. The polarization angle  $\phi$  is fixed  $\phi = 45^{\circ}$  in (a,c,e) and  $\phi = -45^{\circ}$  in (b,d,f). The triangle, asterisk and square points are obtained from DFT calculations and the solid lines are fitted curves based on the calculated points.



**Figure S2** (a,b) The spin dependent photoresponse versus the polarization angle  $\phi$  of CPGE for two In<sub>2</sub>Se<sub>3</sub> structures  $\alpha_{out}$  and  $\alpha_{in}$ , respectively. (c) The corresponding spin polarization of structures  $\alpha_{out}$  and  $\alpha_{in}$  versus the polarization angles  $\phi$ . (d) spin dependent relative photoresponse change versus the polarization angles  $\phi$ . The photon energy  $E_{ph}$  is equal to 0.8 eV. The triangle, asterisk and square points are obtained from DFT calculations and the solid lines are fitted curves based on the calculated points.