Supporting information

Femtosecond optical switch using molecular two-photon absorption with multi-step charge dissociation

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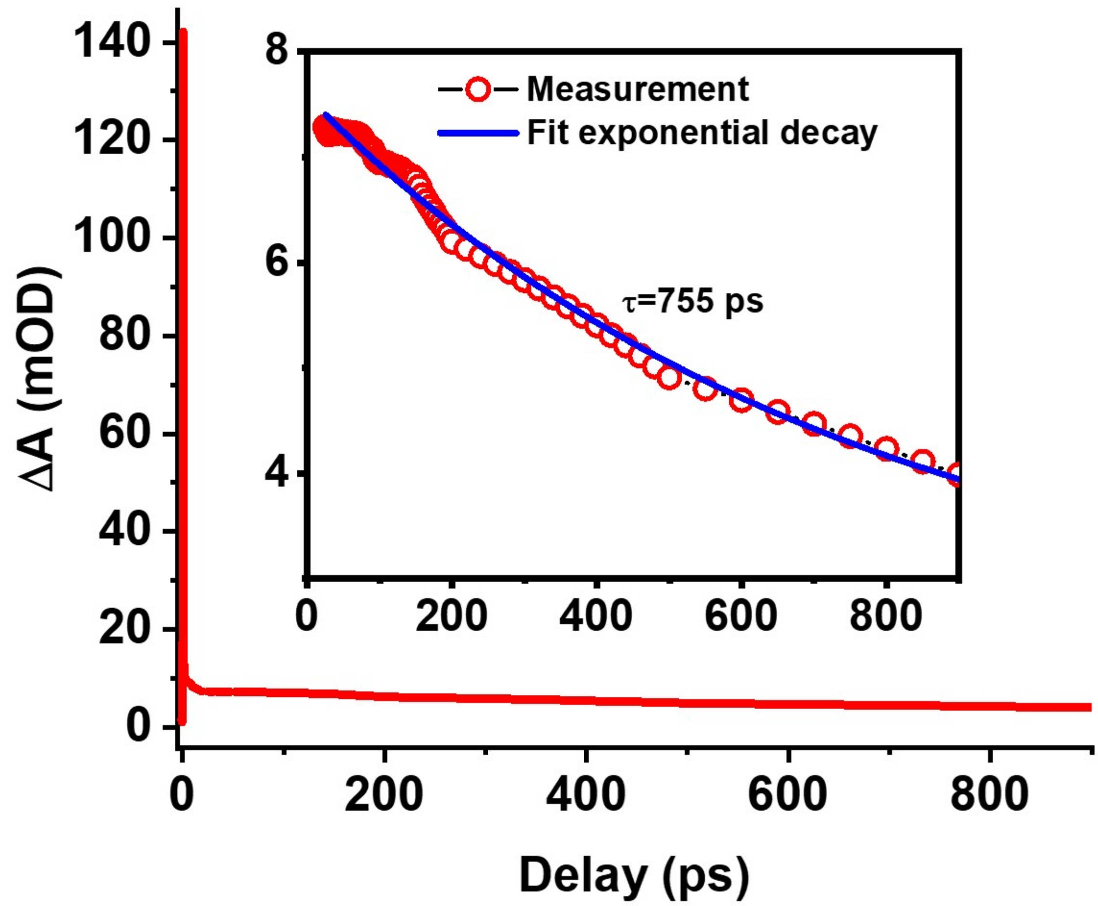

Fig S1. The TA dynamics measured at 510 nm using a pump at 800 nm and pump fluence of 1.43 mJ/cm^2.

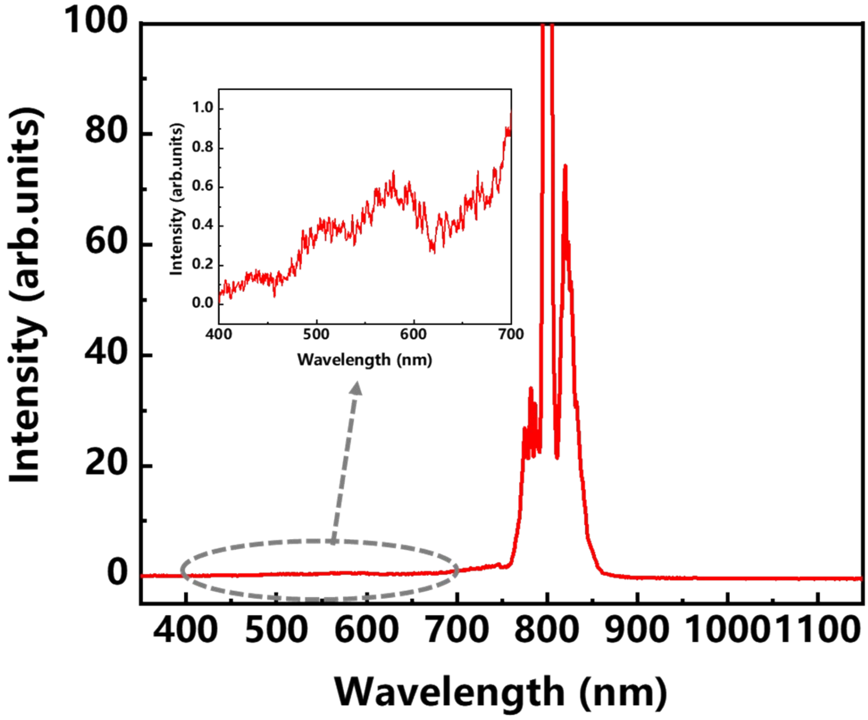

Fig S2. The supercontinuum spectrum that was used as the probe in the pump-probe measurements. Inset: The enlarged spectrum from 400 nm to 700 nm.

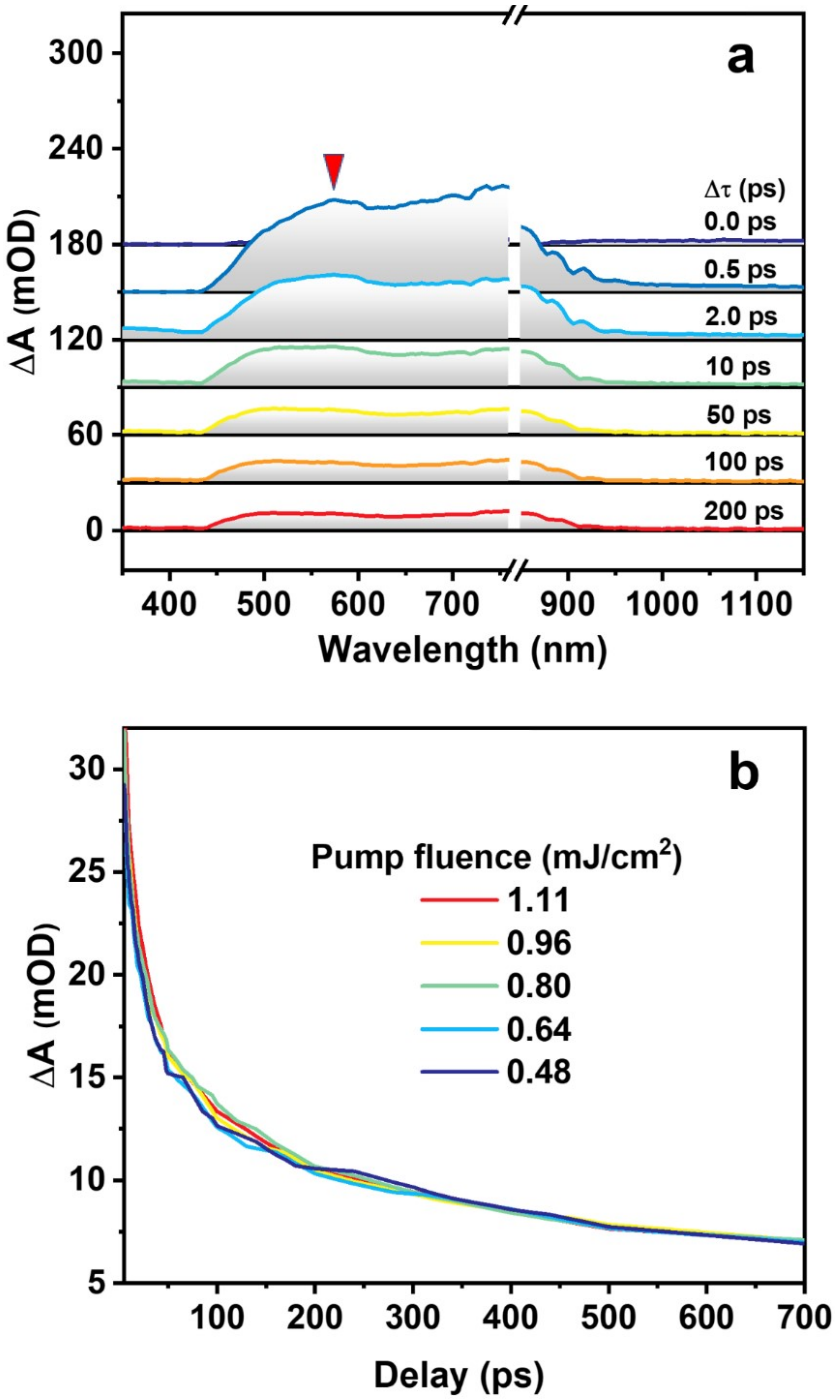

Fig. S3 (a) TA spectra measured at different time delays using pump pulses at 400 nm (3.1 eV). (b) Normalized TA dynamics at 580 nm at different pump fluence, corresponding to the peak wavelength highlighted by a triangle in (a).

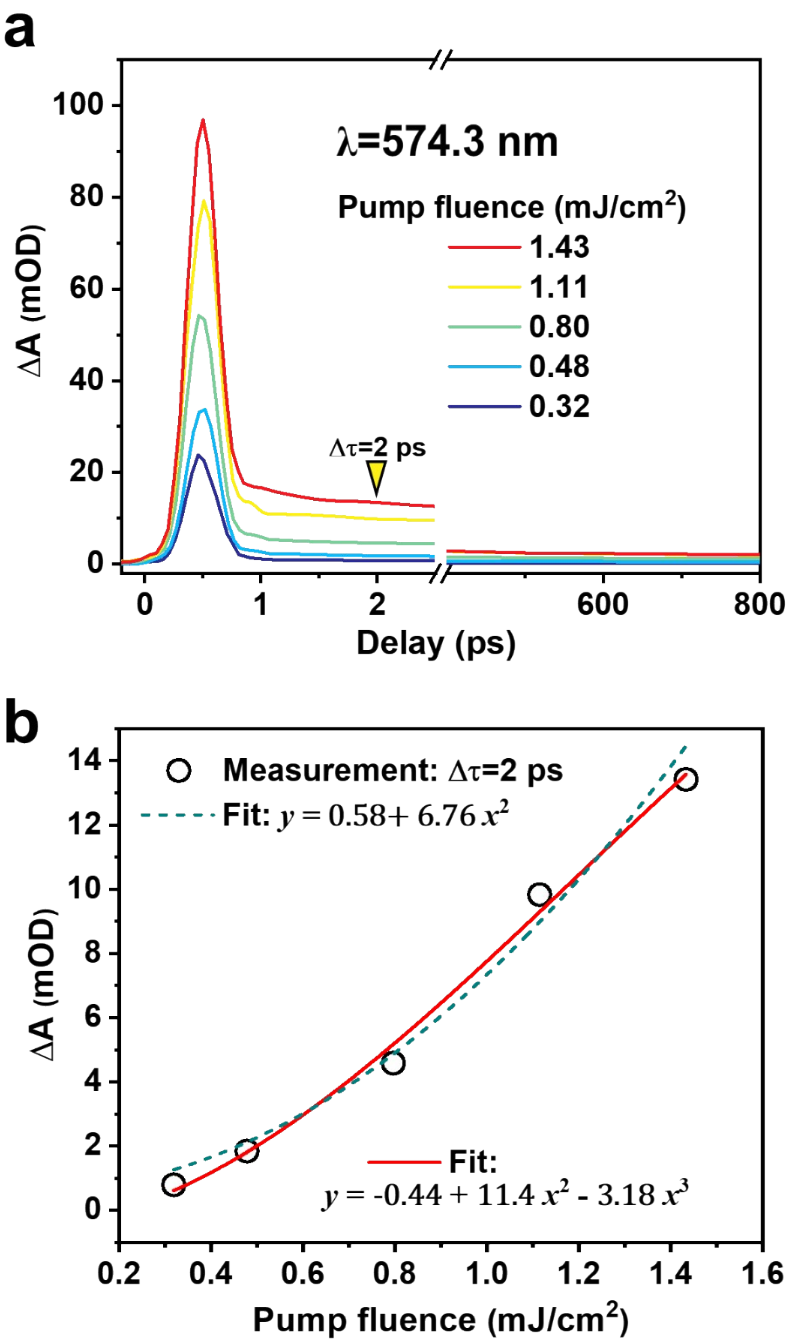

Fig. S4 (a) TA dynamics at 574.3 nm measured at different pump fluences using pump pulses at 800 nm (1.55 eV). (b) TA amplitude at Δτ=2 ps (yellow triangle in (a)) as a function of pump fluence. The TA signal caused by exciton

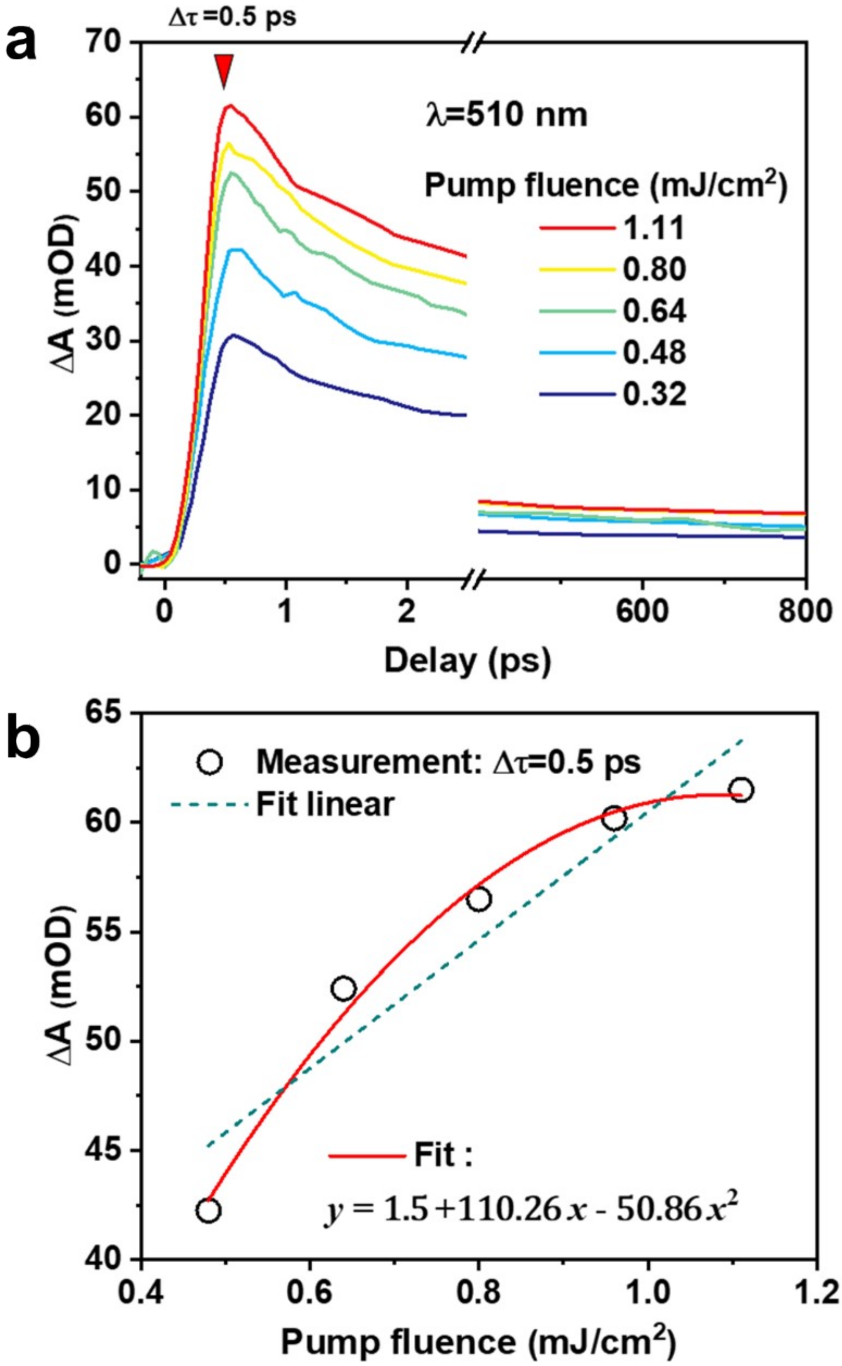

Fig. S5 (a) TA dynamics measured at 510 nm at different pump fluences using pump pulses at 400 nm (3.1 eV). (b) TA amplitude at a delay of □Δτ□=0.5 ps (red triangle in (a)) as a function of pump fluence. TA amplitude saturates with increasing pump fluence, which agrees with the reduction of electrons in the valence band due to the pumping and can be well fitted with a quadratic function.

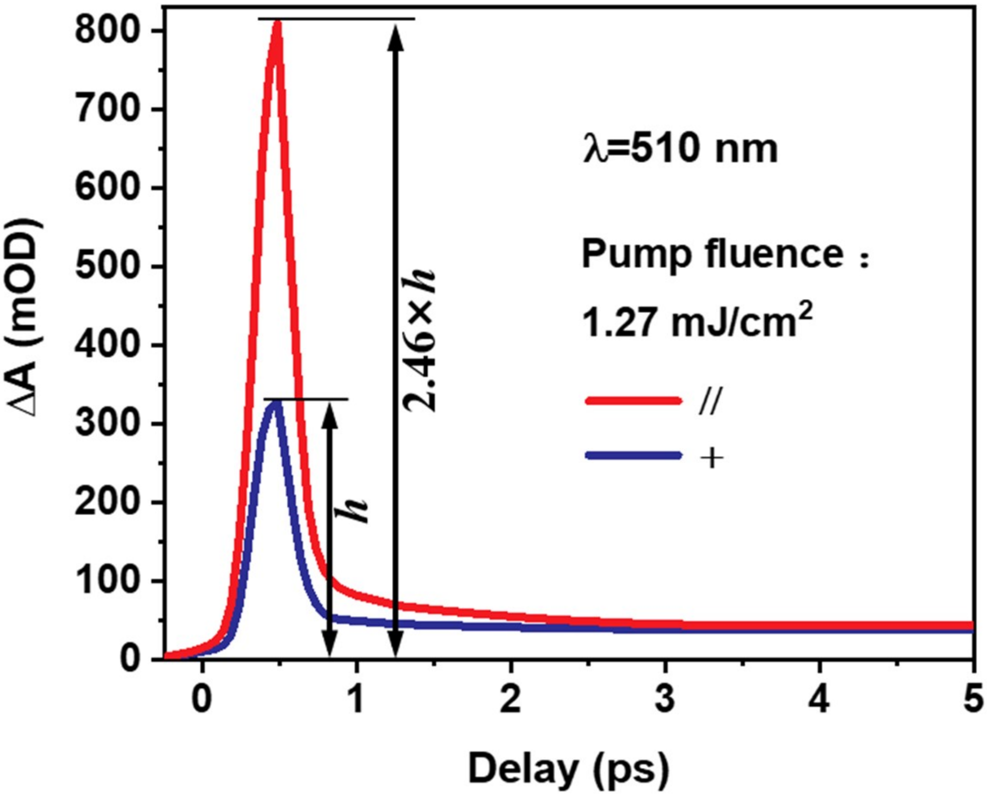

Fig. S6 TA dynamics at a probe wavelength of 510 nm measured with different configurations of polarization directions of the pump and probe pulses. The TA magnitude when the pump polarized is parallel to the probe (red curve) is 2.46 times large than when the pump polarized is perpendicular to the probe beam (dark purple curve).

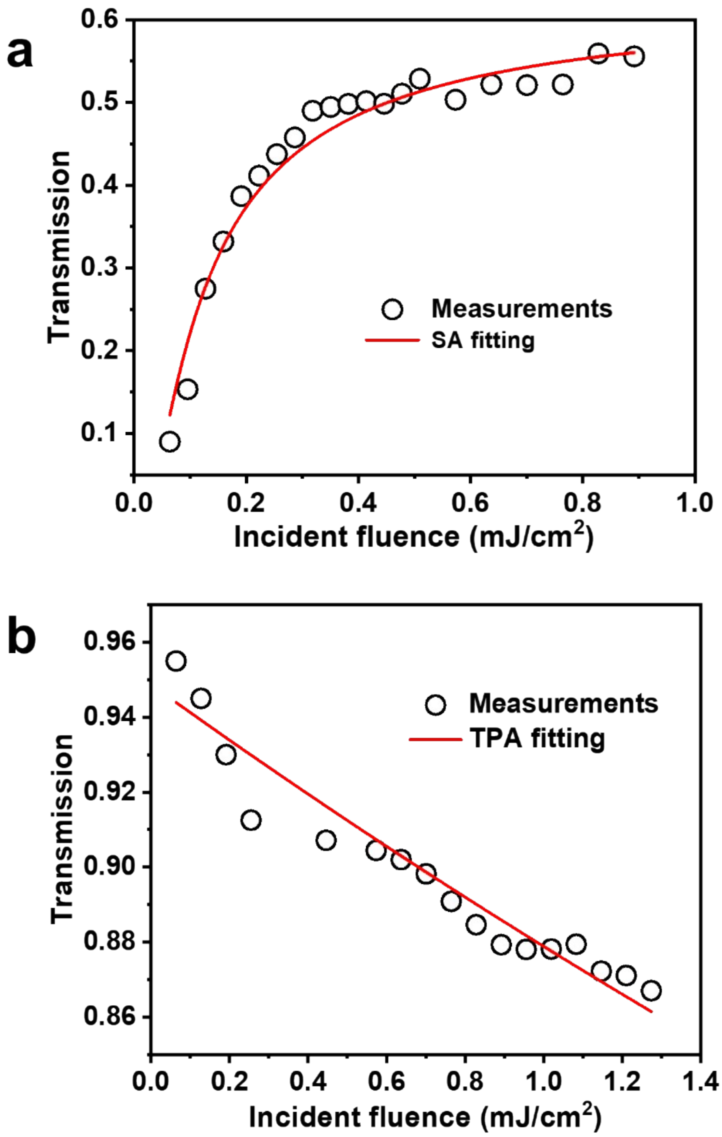

Fig. S7 (a) Transmission as a function of incident fluence under 400 nm laser excitation. The transmission increases with the incident pump fluence, showing a characteristic of saturable absorption (SA). (b) Transmission as a function of incident intensity for different thicknesses under 800 nm laser excitation. The transmission decreases with the incident pump fluence, showing a characteristic of two-photon absorption (TPA).