Supplementary Information

Trap-free exciton dynamics in monolayer WS_2 via oleic acid passivation

Dabin Lin,[a] Wenjun Ni,[b] Gagik G. Gurzadyan,[b] Fangteng Zhang,[a] Weiren Zhao,[a,c] Lin Ma*[a,c] and Zhaogang Nie*[a,c]

[a]School of Physics and Optoelectronic Engineering, Guangdong University of Technology, Guangzhou 510006, China.

[b]State Key Laboratory of Fine Chemicals, Institute of Artificial Photosynthesis, Dalian University of Technology, Dalian 116024, China.

[c]Guangdong Provincial Key Laboratory of Information Photonics Technology, Guangdong University of Technology, Guangzhou 510006, China.

E-mail: malin@gdut.edu.cn; zgnie@gdut.edu.cn

Exciton density calculation

We calculate the initial photogenerated exciton density N(0) from the incident fluence and the absorption efficiency, according to equation (S1):

$$N_0 = \varphi \frac{E\lambda}{hc}(1 - 10^{-A}) \qquad (S1)$$

where E is the fluence (J cm^{-2}) of excitation pulses, λ is excitation wavelength, A is the absorbance at excitation wavelength, φ is the ratio of exciton created per photon absorbed (assumed to be unity). The excitation intensities (fluences) applied in our study were in the linear-dependent range to avoid higher order nonlinear effect.

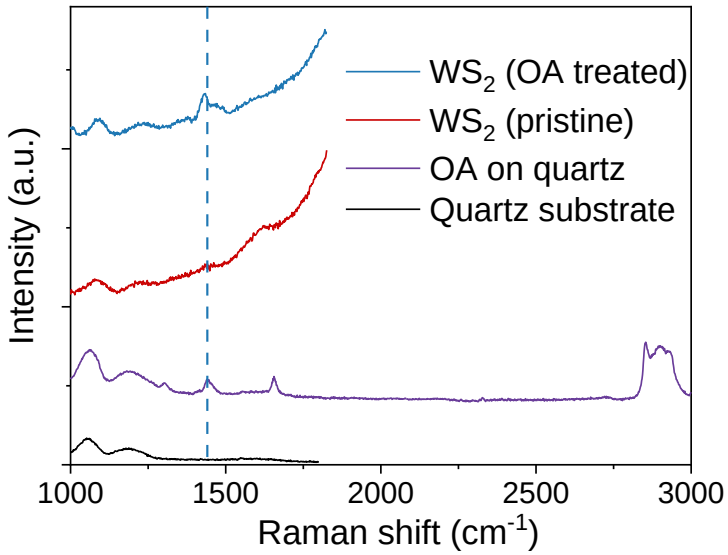

Figure S1. Raman spectra of OA treated WS_2 monolayer (blue), pristine WS_2 (red), oleic acid on quartz (purple) and quartz substrate (black) under excitation at 532 nm. The Raman signal at 1439 cm^{-1} which corresponds to the CH_2 scissoring mode of oleic acid is labelled by the dash line. The Raman signals above 1800 cm^{-1} for OA treated and pristine WS_2 were removed due to the influence from PL.

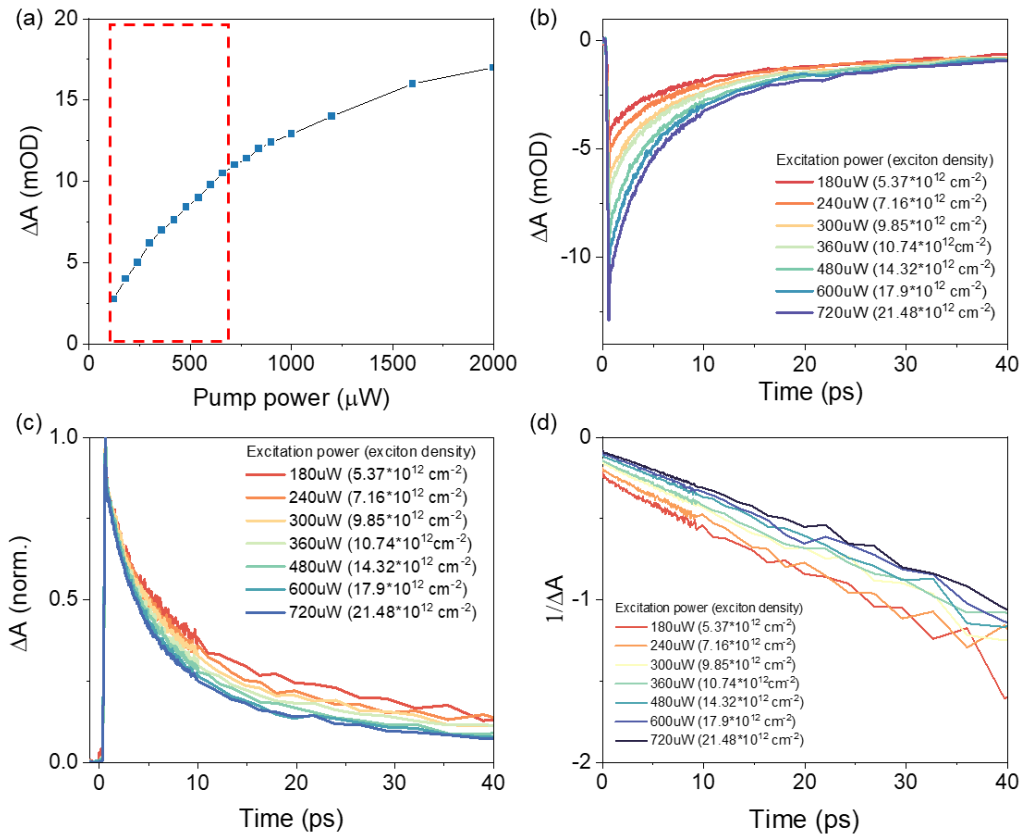

Figure S2. (a) Excitation power dependent TA signal at 1 ps in pristine monolayer WS_2. The dash rectangle indicates the power regime where the intensity dependent TA kinetics (b) were measured. (c) The normalized plot of (b) shows clear dependence of TA kinetics on the exciton density. (d) The relations between the reciprocal of ΔA (1/ΔA) versus time. The similar slopes are indicating similar EEA constant.

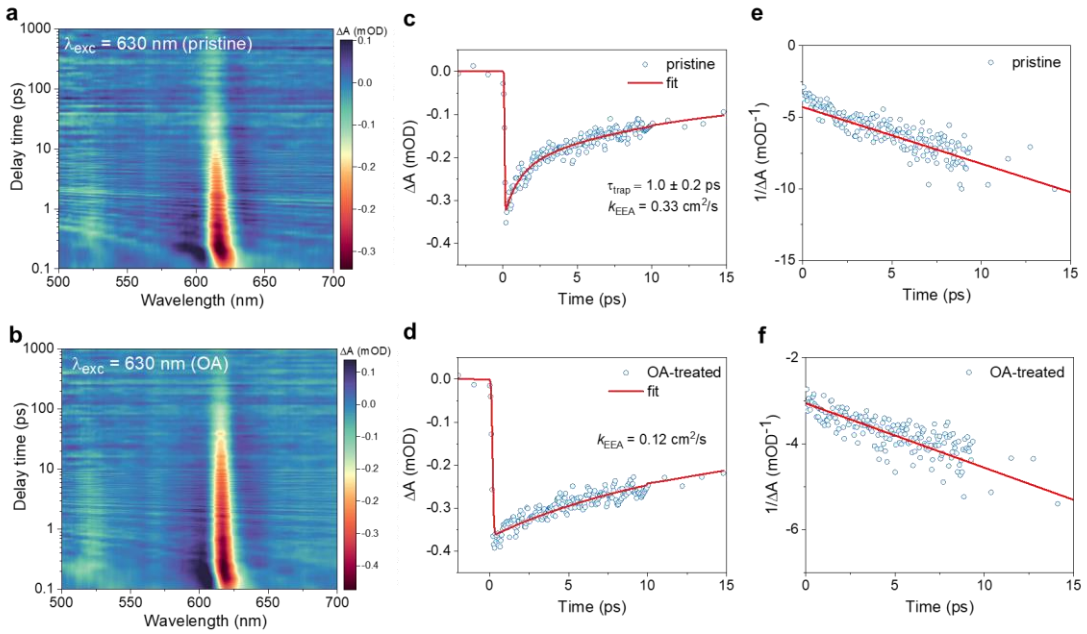

Figure S3. (a-b) 2D color contour plot of TA data; (c-d) TA kinetics; (e-f) The dependence of the reciprocal of ΔA (1/ΔA) versus time of pristine and OA treated monolayer WS_2 under excitation at 630 nm with low exciton density N(0) = $4.3*10^{11}$ cm^{-2}.

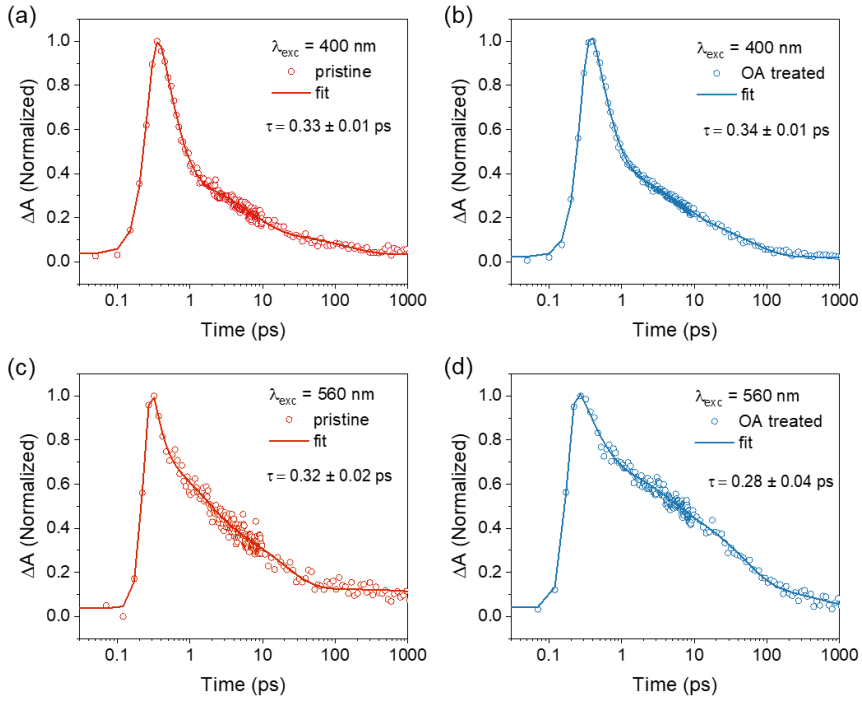

Figure S4. (a-b) Transient absorption kinetics of monolayer WS_2 before (a) and after OA treatment (b) under excitation at 400 nm. (c-d) Transient absorption kinetics of monolayer WS_2 before (c) and after OA treatment (d) under excitation at 560 nm. The solid lines are exponential fits to extract the ultrafast sub-ps time constant.

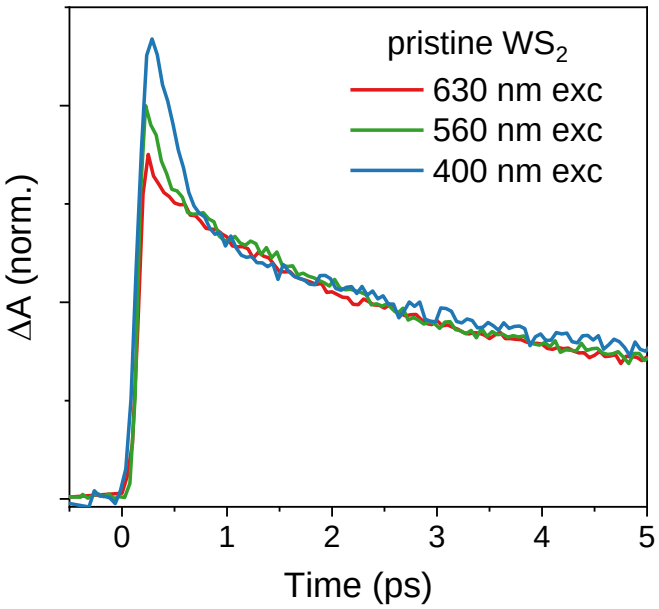

Figure S5. Normalized transient absorption kinetics at 614 nm of pristine WS_2 under excitation at 630, 560 and 400 nm.

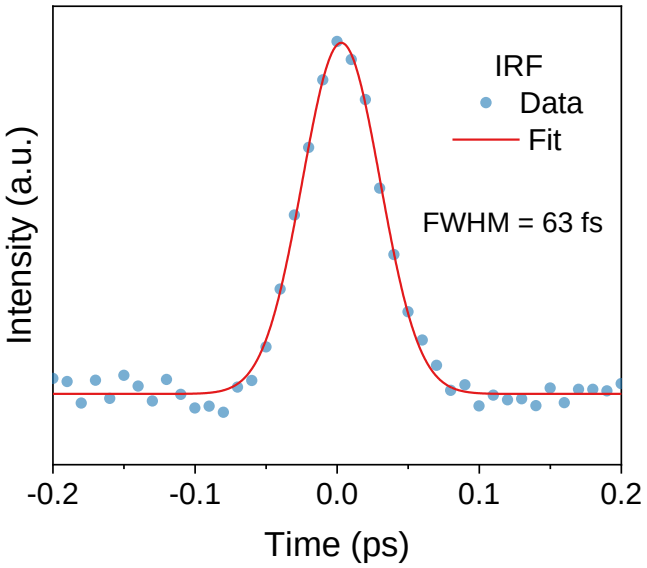

Figure S6. Instrument response function (IRF) of transient absorption setup determined from the Gaussian fit of Raman scattering signal in water.