Supporting Information

Observation of quantum-confined exciton states in monolayer WS_2 quantum dots by ultrafast spectroscopy

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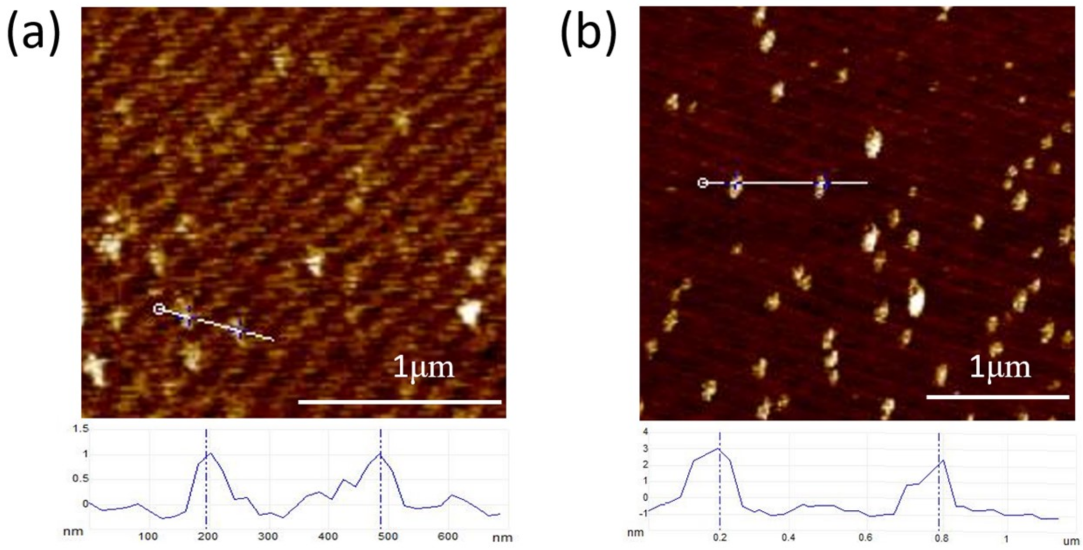

Figure S1. AFM images of (a) monolayer WS_2 QDs and (b) WS_2 nanosheets.

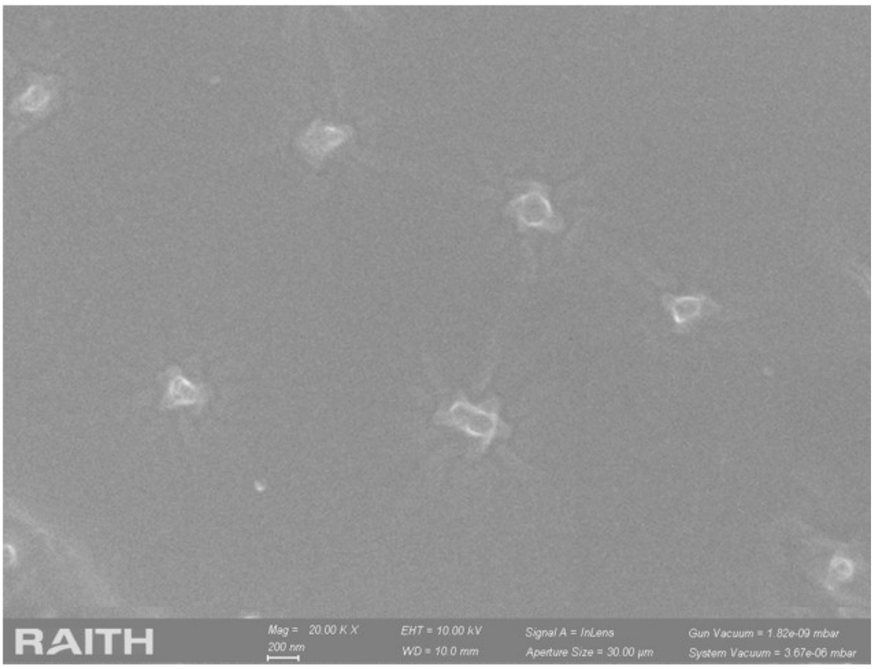

Figure S2. SEM image of WS_2 nanosheets.

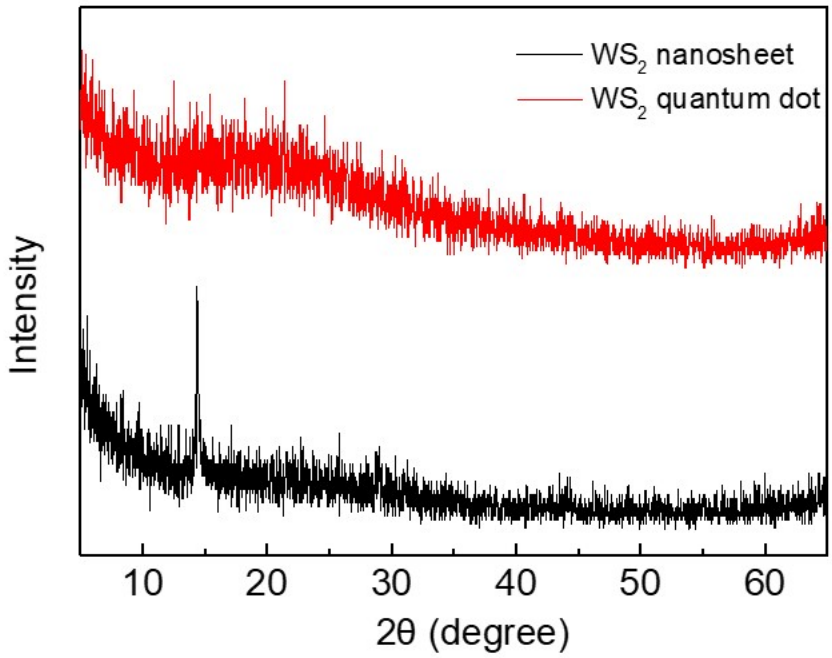

Figure S3. XRD patterns of monolayer WS_2 QDs and WS_2 nanosheets.

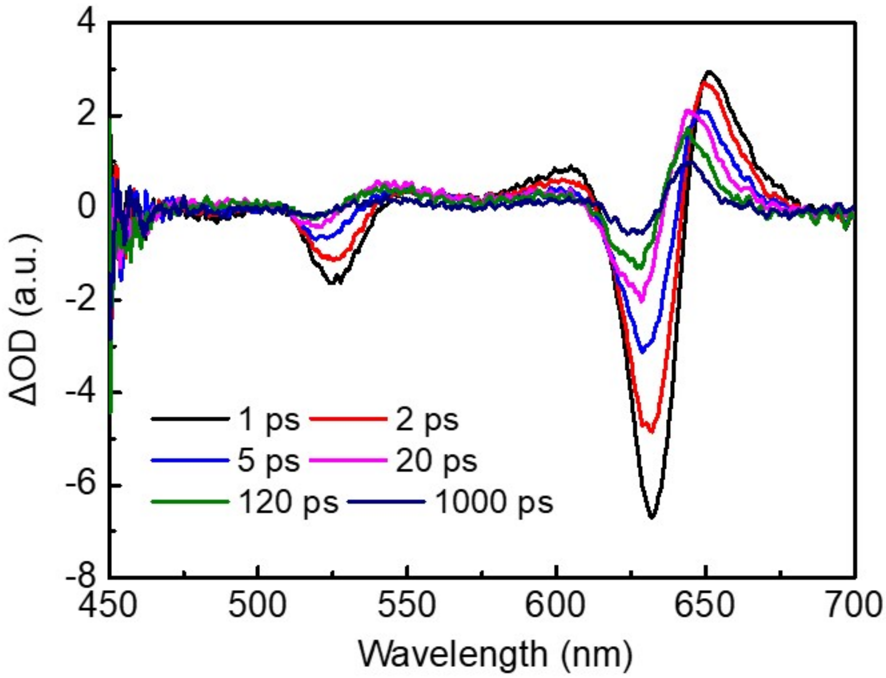

Figure S4. TA spectra of WS_2 nanosheets under 610 nm excitation at different delay times.

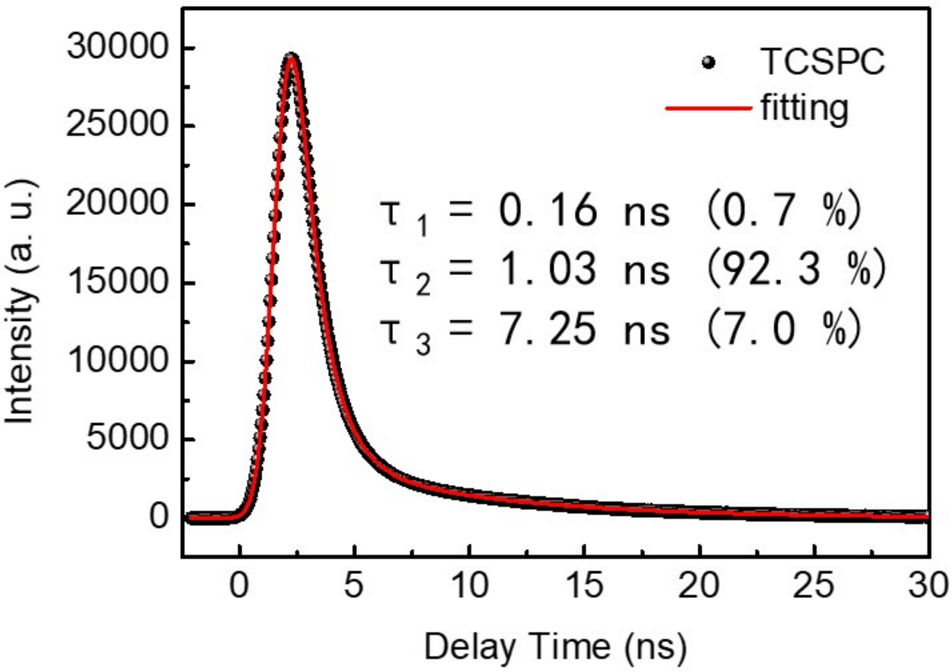

Figure S5 Time-resolved PL dynamics of monolayer WS_2 QDs measured by time-correlated single photon counting (TCSPC; the excitation wavelength is 430 nm, and the probe wavelength is 500 nm).

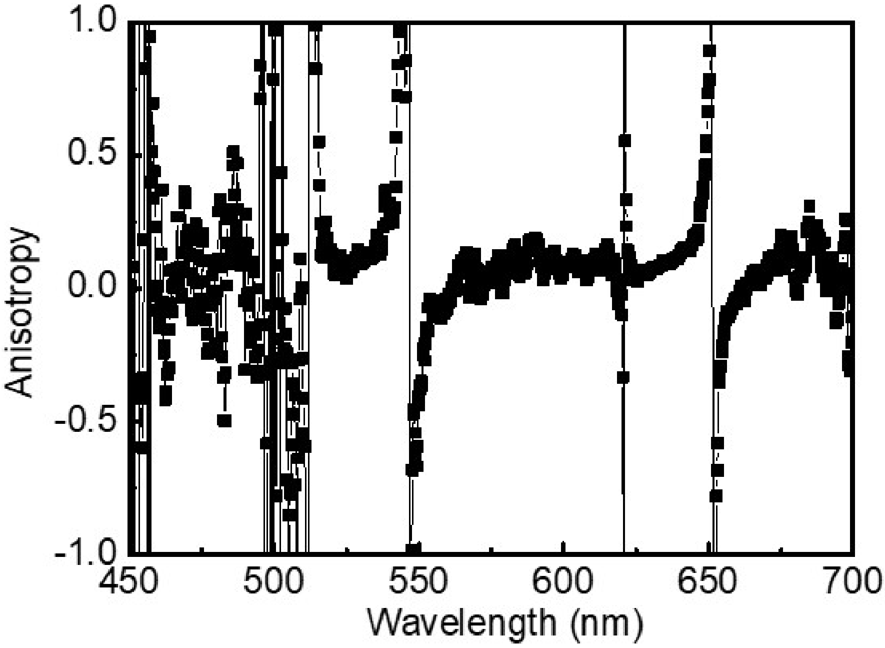

Figure S6. Anisotropic spectra of WS_2 nanosheets probed at 0.28 ps.

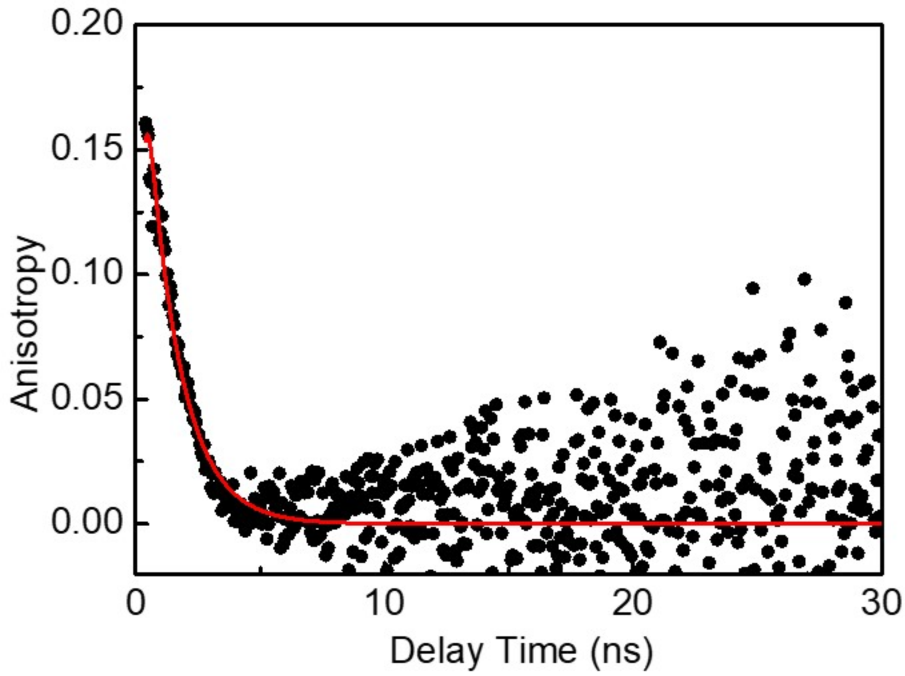

Figure S7 Time-resolved PL anisotropy dynamics of monolayer WS_2 QDs measured by TCSPC (the excitation wavelength is 430 nm, and the probe wavelength is 500 nm). It gives a rotational diffusion time (τ_{rot}) of 1.3 ns. According to the equation $\tau_{rot} = \eta V/k_B T$, where η is the viscosity, V is the volume of the rotating unit, k_B is the Boltzmann constant and T is temperature. At room temperature, η is 1.86 mPa·s for NMP,[1] $k_B T \sim 26$ meV. Assuming that rotational diameter is cube root of V, the resulted rotational diameter is ~1.4 nm for monolayer WS_2 QDs. It is consistent with the average height (~1 nm) of monolayer WS_2 QDs. This indicates the effective volume of the rotating unit is relative to the lateral thickness of monolayer WS_2 QDs, suggesting a pure rotation process in slip case. Noting that the calculated rotational diameter is slightly larger than the physical thickness of monolayer WS_2 QDs, it may reflect that the solvent molecules (NMP) could be involved with the rotational diffusion behavior of monolayer WS_2 QDs, implying the potential influence of solvent-QD interactions.

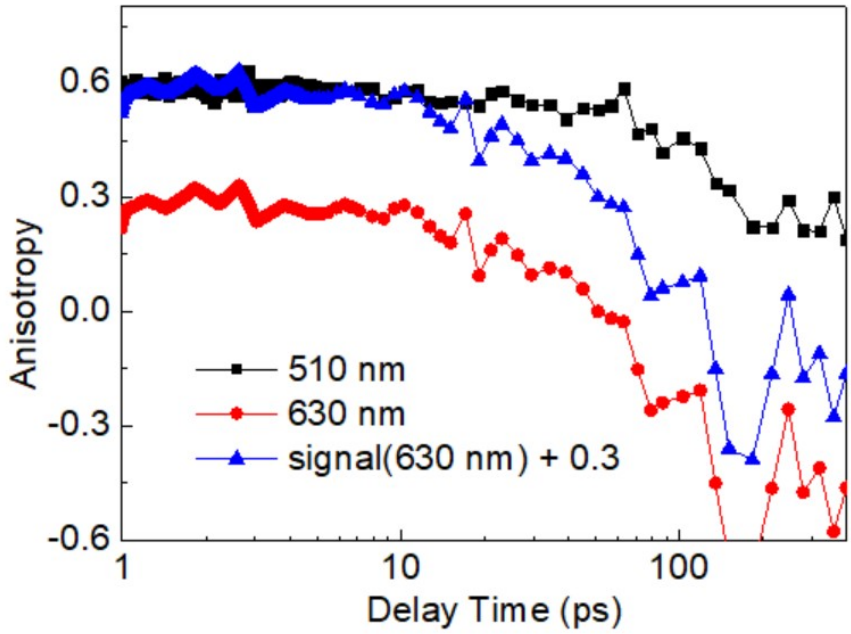

Figure S8. Anisotropic dynamics of monolayer WS_2 QDs under 520 nm excitation and probe at 510 nm (black line) and 630 nm (red line). The blue line is obtained from the method that lifting up the original anisotropic dynamics signals at 630 nm by a value of 0.3.

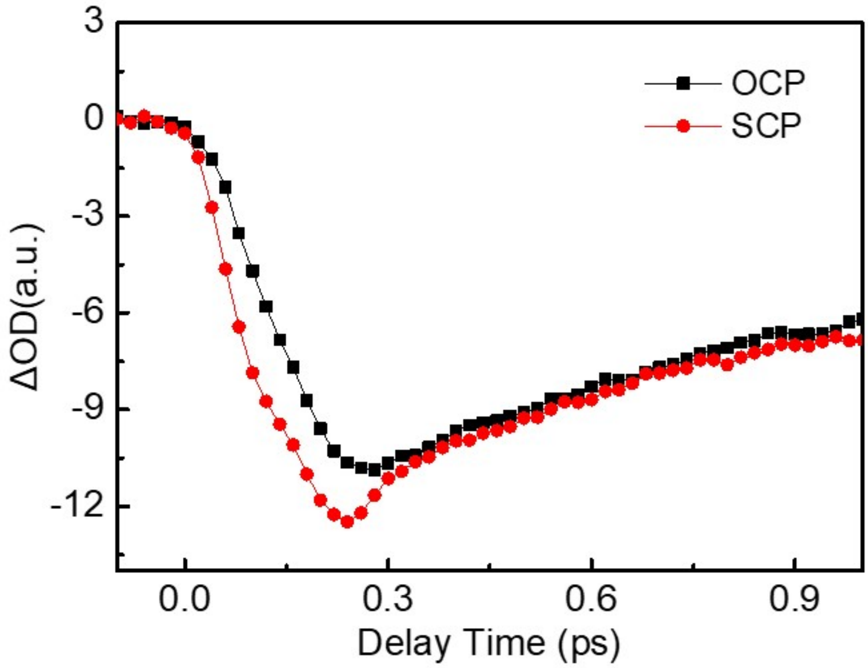

Figure S9. The SCP and OCP dynamics of A-exciton for WS_2 nanosheets probed at 634 nm.

Table S1. Best-fitted parameters for dynamics of exciton decay of monolayer WS_2 QDs and WS_2 nanosheets with a function of $I(t) \propto \Sigma_i A_i \exp(-t/\tau_i)$.

	τ_1 (ps)	τ_2 (ps)	τ_3 (ps)
WS_2 QDs (470ex)	1.4 (37%)	12 (50%)	184 (13%)
WS_2 QDs (570ex)	1.8 (39%)	13.8 (46%)	199 (15%)
WS_2 QDs (610ex)	0.95 (37%)	9.6 (50%)	247 (13%)
WS_2 nanosheets	0.57 (46%)	4.9 (41%)	703(13%)

Reference

1. A. García-Abuín, D. Gomez-Díaz, M. D. La Rubia, and J. M. Navaza. *J. Chem. Eng. Data.* 2011, **56**, 646–651.