

Supplementary Information for

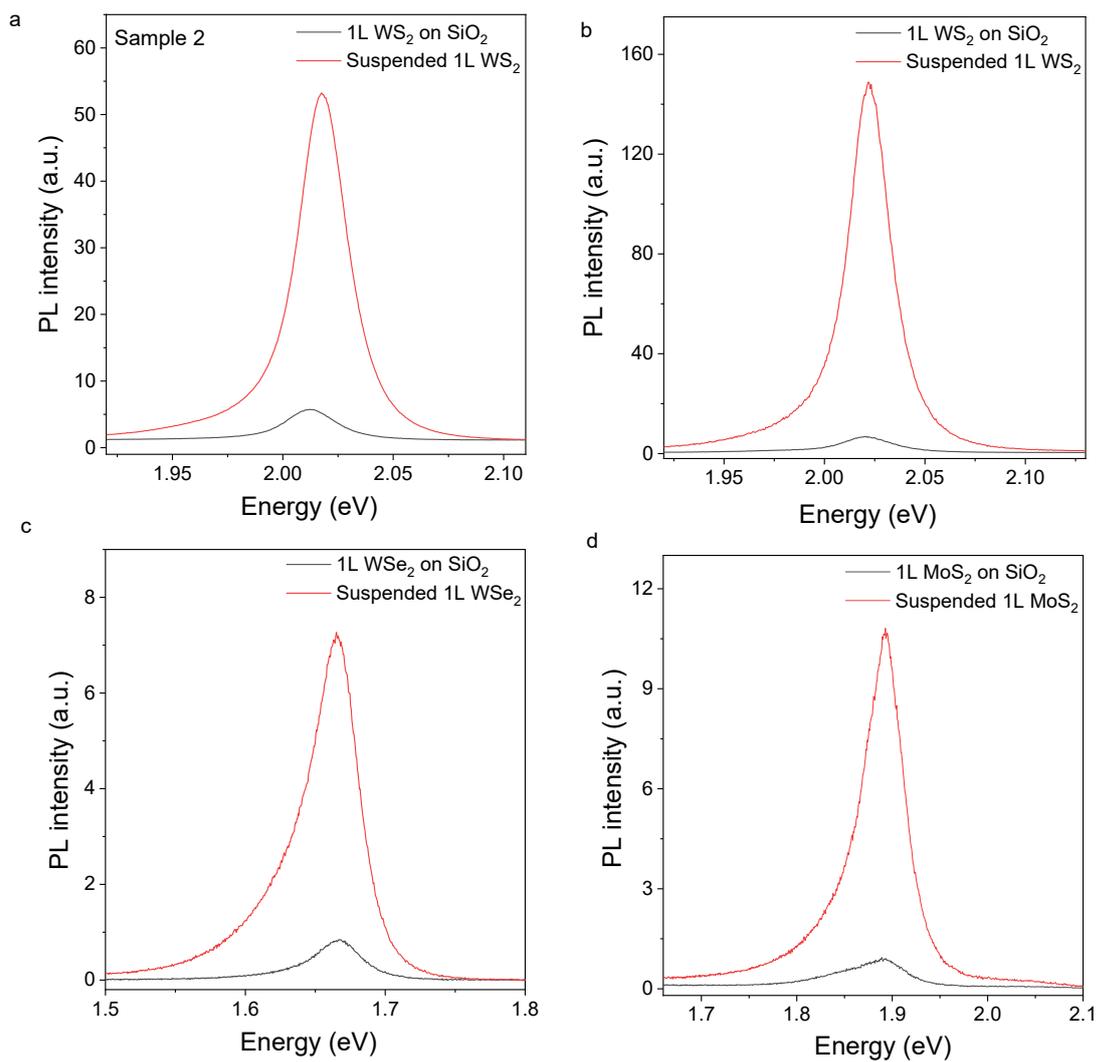
Enhanced Interactions of Excitonic Complexes in Free-standing WS₂

Xueqian Sun¹, Zhuoyuan Lu¹ and Yuerui Lu^{1,2*}

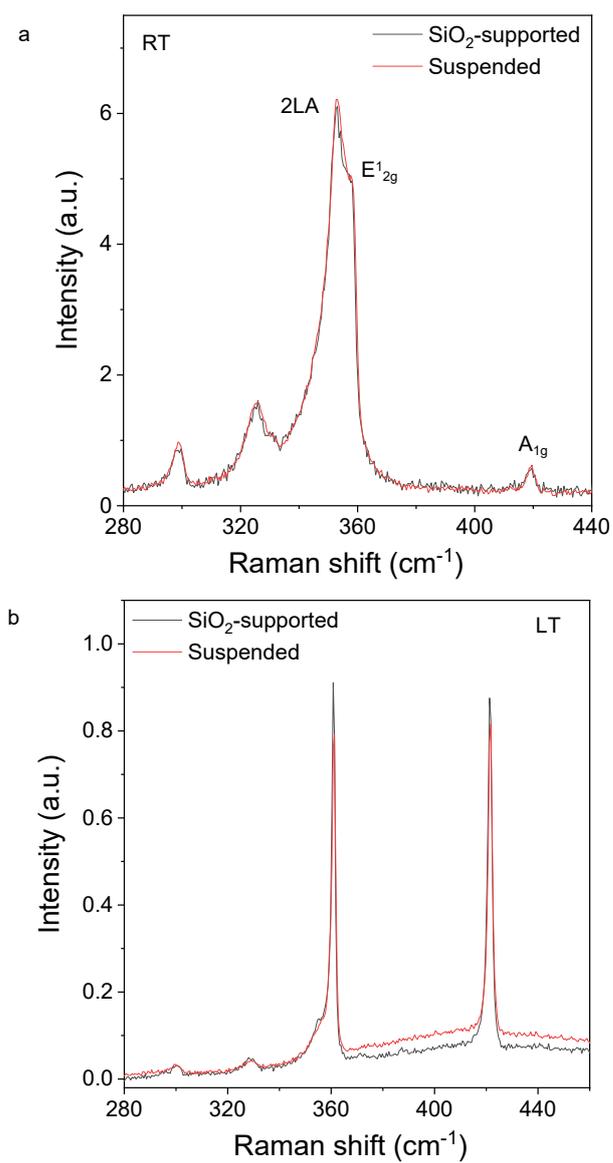
¹School of Engineering, College of Engineering and Computer Science, the Australian National University, Canberra, ACT, 2601, Australia

²Australian Research Council Centre of Excellence for Quantum Computation and Communication Technology, the Australian National University, Canberra, ACT, 2601 Australia

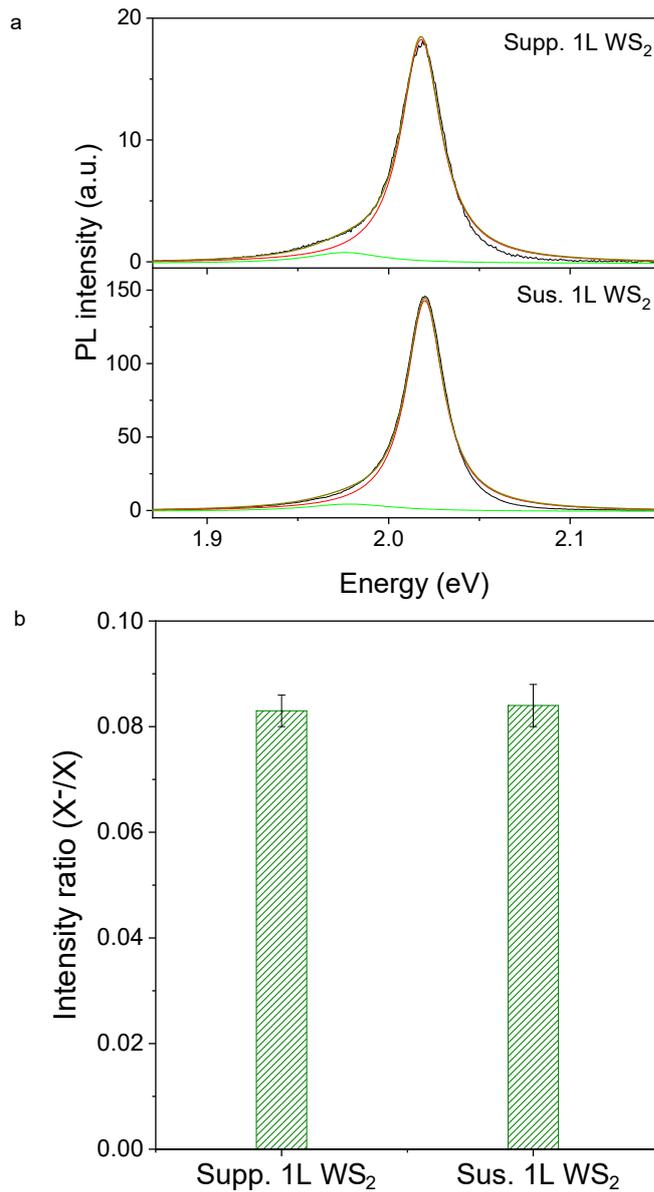
* To whom correspondence should be addressed: Yuerui Lu (yuerui.lu@anu.edu.au)



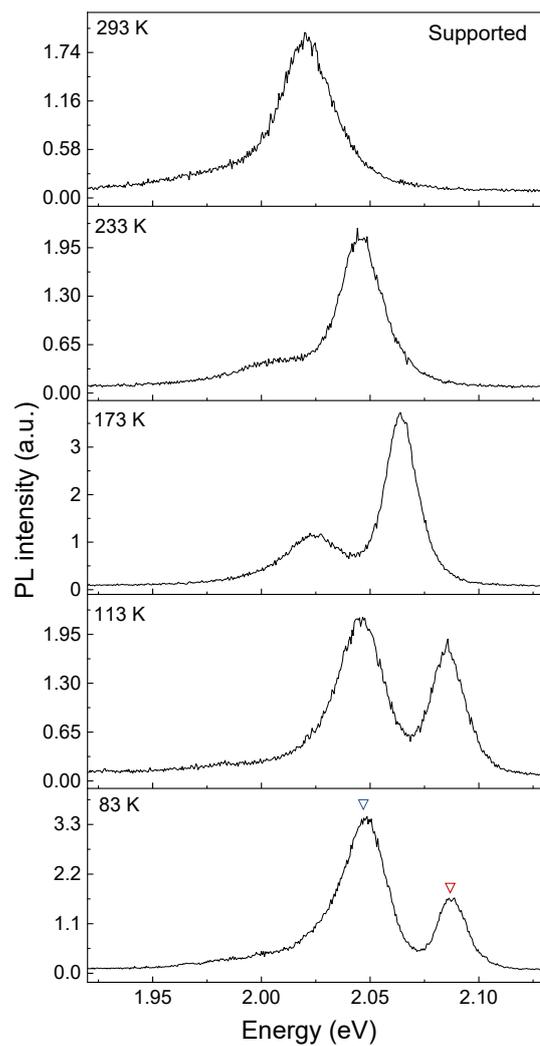
Supplementary Figure S1 | Measured PL spectra from 1L SiO₂-supported and suspended monolayer WS₂ sample 2 (a) and sample 3 (b), monolayer WSe₂ sample (c) and monolayer MoS₂ sample (d) at room temperature.



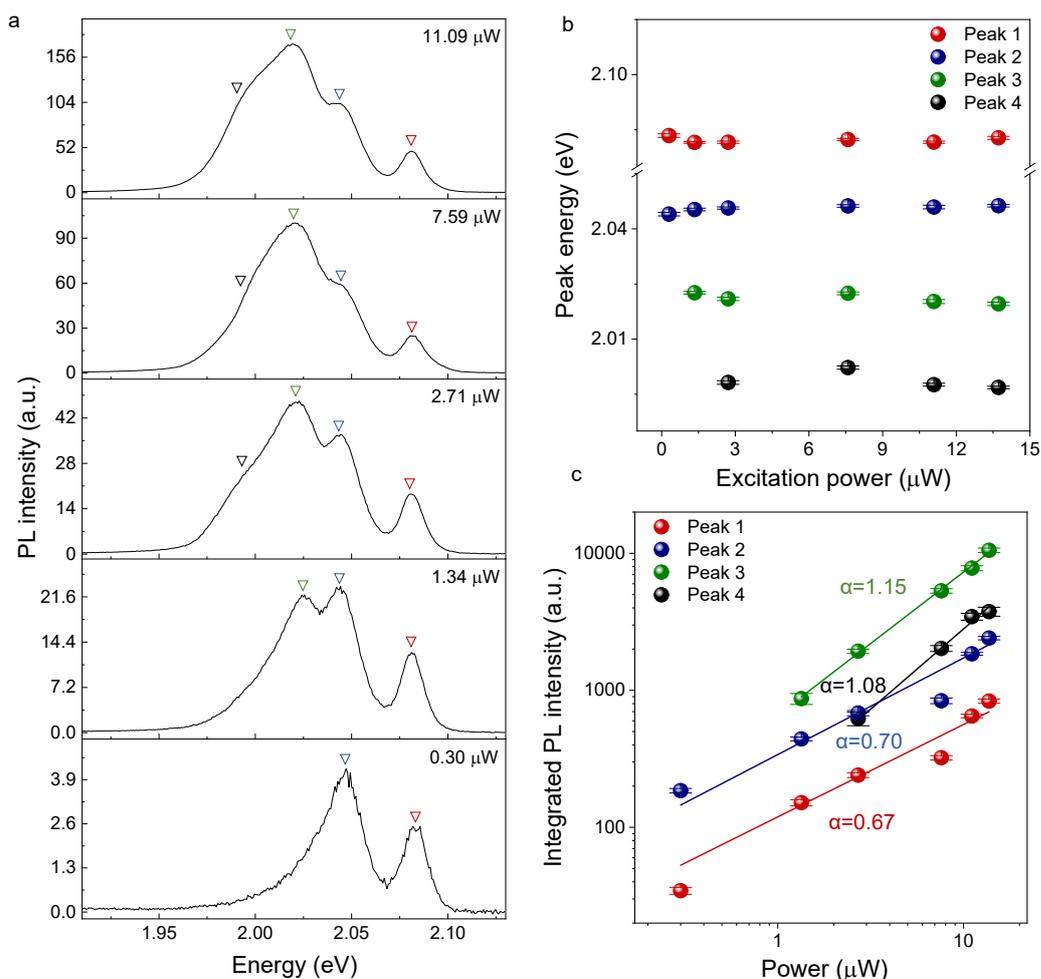
Supplementary Figure S2 | Measured Raman spectra from 1L SiO₂-supported and suspended monolayer WS₂ at room temperature (a) and 83K (b).



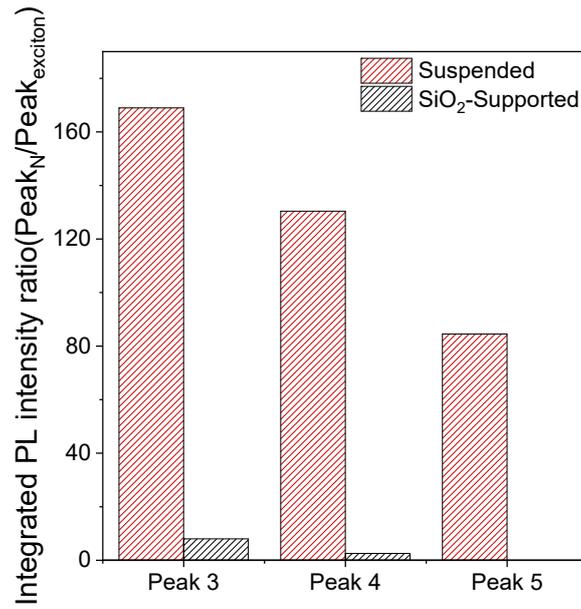
Supplementary Figure S3 | **a**, Measured PL spectra from the SiO₂-supported (top panel) and suspended (bottom panel) monolayer WS₂ at room temperature. The spectrum was fitted with trion (green curve) and exciton (red curve) peaks. Black and dark yellow curves are experimental data and accumulative fitting curve, respectively. **b**, The extracted intensity ratio of trion (X⁻) to exciton (X) for SiO₂-supported and suspended monolayer samples, indicating the value of around 0.083 ± 0.003 and 0.084 ± 0.004 , respectively.



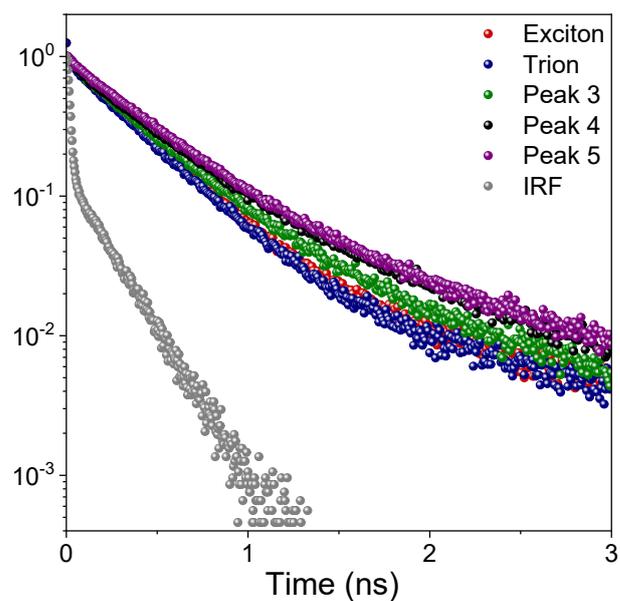
Supplementary Figure S4 | Measured temperature-dependent PL spectra from a SiO₂-supported WS₂ monolayer, under an excitation power of 0.3 μ W with a 532 nm CW laser.



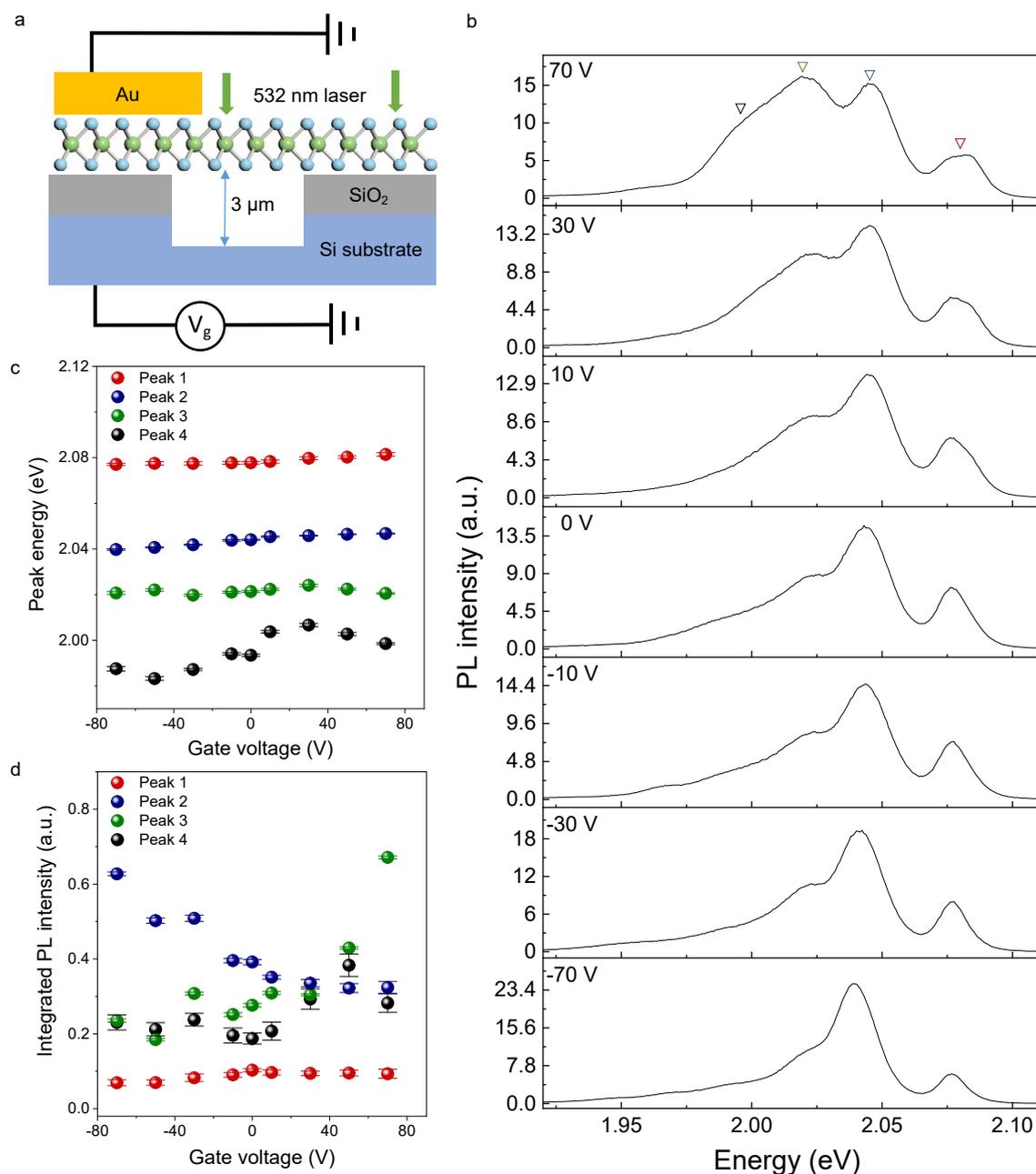
Supplementary Figure S5 | Excitation power-dependent PL spectra from the SiO₂-supported WS₂ monolayer at 83K **a**, Measured excitation power dependent PL spectra from the SiO₂-supported WS₂ monolayer with pumping power ranging from 0.3 μW to 11.09 μW . **b**, Extracted peak energy of exciton peaks as a function of excitation power. **c**, Extracted integrated PL intensity of the exciton peaks as a function of excitation power. The fitting curves give rise to a slope of 0.67, 0.70, 1.15 and 1.08 for Peak 1 to peak 4, respectively. The error bars in **(b)** and **(c)** represent the fitting uncertainty obtained from multiple fitting analysis (>3).



Supplementary Figure S6 | Extracted PL intensity ratio (η) of additional emission peaks from samples with different dielectric screening, suspended and SiO₂-supported structures, using the measured PL spectra displayed in Figure 3a and S5a at the same excitation power of 2.71 μ W.



Supplementary Figure S7 | Time-resolved PL emission (normalized) obtained from different emission peaks from freestanding WS₂ monolayer at 83K. The decay curve was fitted by deconvoluting the data from the instrument response function (IRF), yielding a lifetime of 377 ps, 368 ps, 476 ps, 524 ps and 552 ps, for exciton (red), trion (blue), biexciton quasiparticles (green, black and purple), respectively.



Supplementary Figure S8 | Electrical modulation of the exciton dynamics for SiO₂-supported WS₂ monolayer at 83K **a**, Schematic plot of a 1L WS₂ device structure, showing the sample used for modulating the doping in both freestanding and SiO₂-supported monolayer using back gate voltage. The gold electrode was grounded, covering both supported and freestanding samples, and a doped Si substrate was employed as the back gate to provide uniform electrostatic doping. **b**, Measured gate dependent PL spectra from a SiO₂-supported WS₂ monolayer at 83 K with gate tuning changing from from 70 to -70V. **c**, Extracted peak energy of exciton peaks as a function of back gate voltages. **d**, Extracted integrated PL intensity of the exciton peaks as a function of back gate voltages. The error bars in (c) and (d) represent the fitting uncertainty obtained from multiple fitting analysis (>3).