Defect-Type-Dependent Carrier Lifetimes in Monolayer WS_2 Films

*Yuanshuang Liu, Huan Liu, Jiangcai Wang and Dameng Liu**

State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China

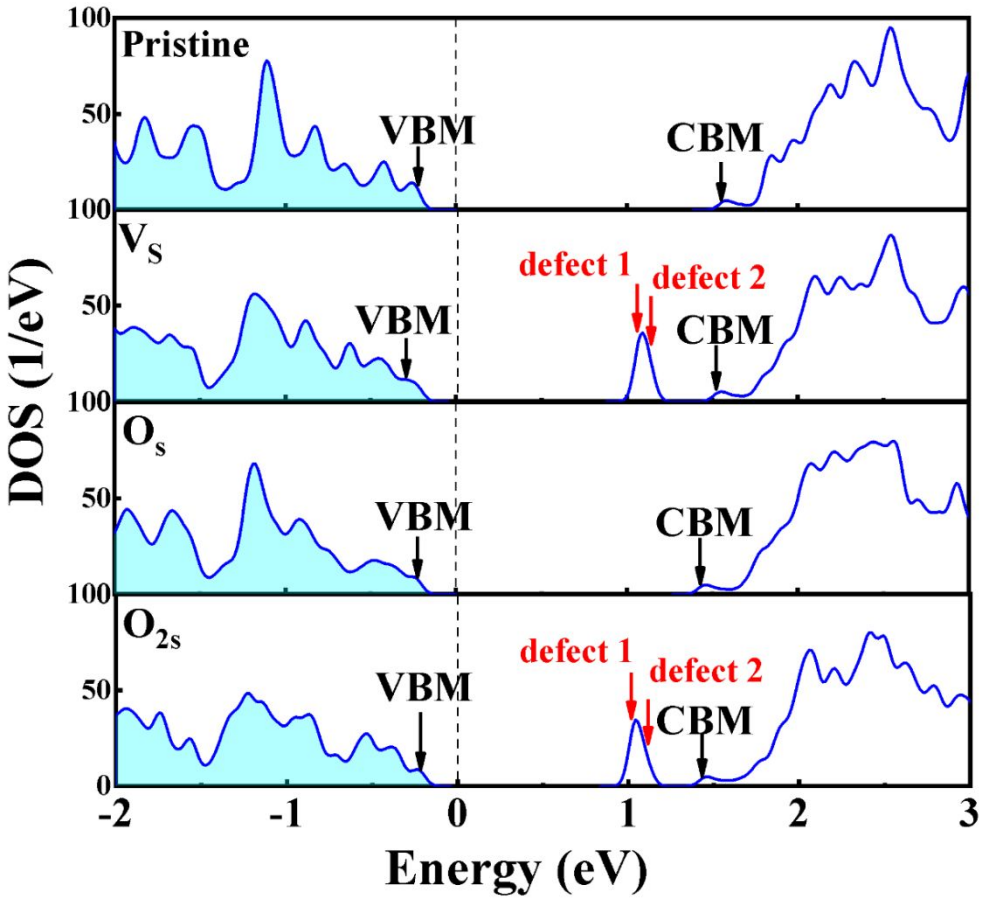

Figure S1. Density of states (DOS) of pristine and defective WS_2 monolayers. Both V_S and O_{2S} defects introduce two defect states in band gap, about 0.4 eV below the CBM.

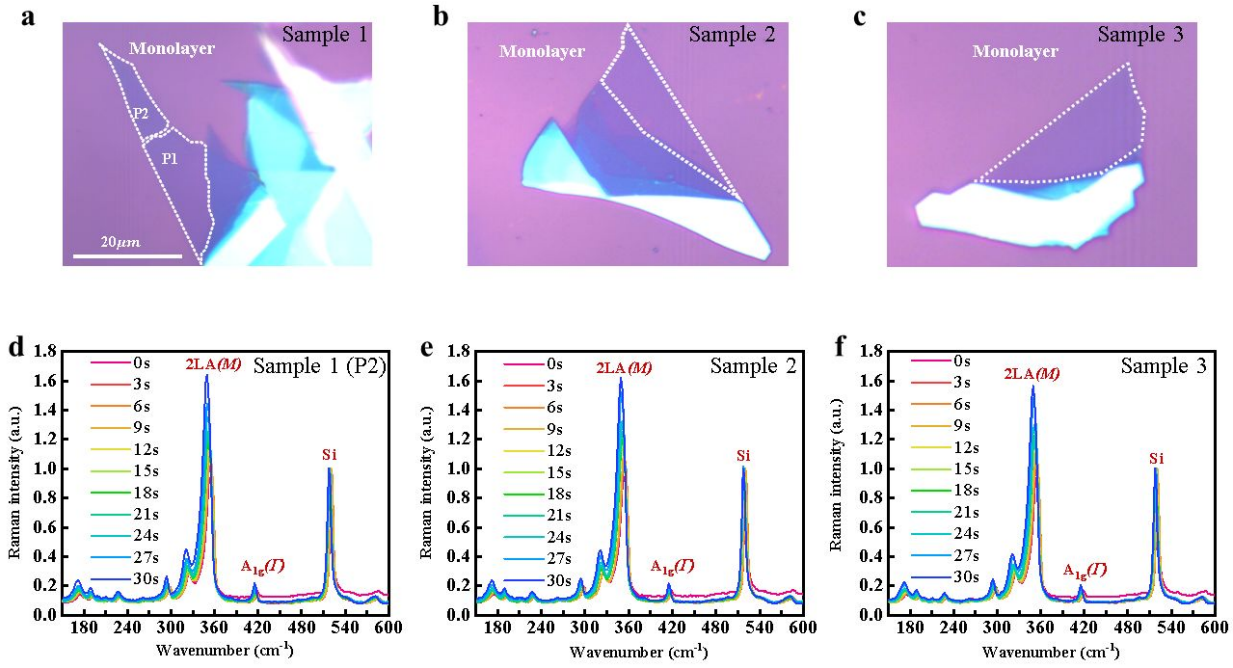

Figure S2. a)-c) Optical microscope image of different WS_2 samples on Si/SiO_2 substrate. d) Raman spectra at different location (P2) for WS_2 monolayer. e) and f) Raman spectra of sample 2 and sample 3 treated by O^{2+} plasma at different treatment times (calibrated by silicon Raman peak ~520cm^{-1}).

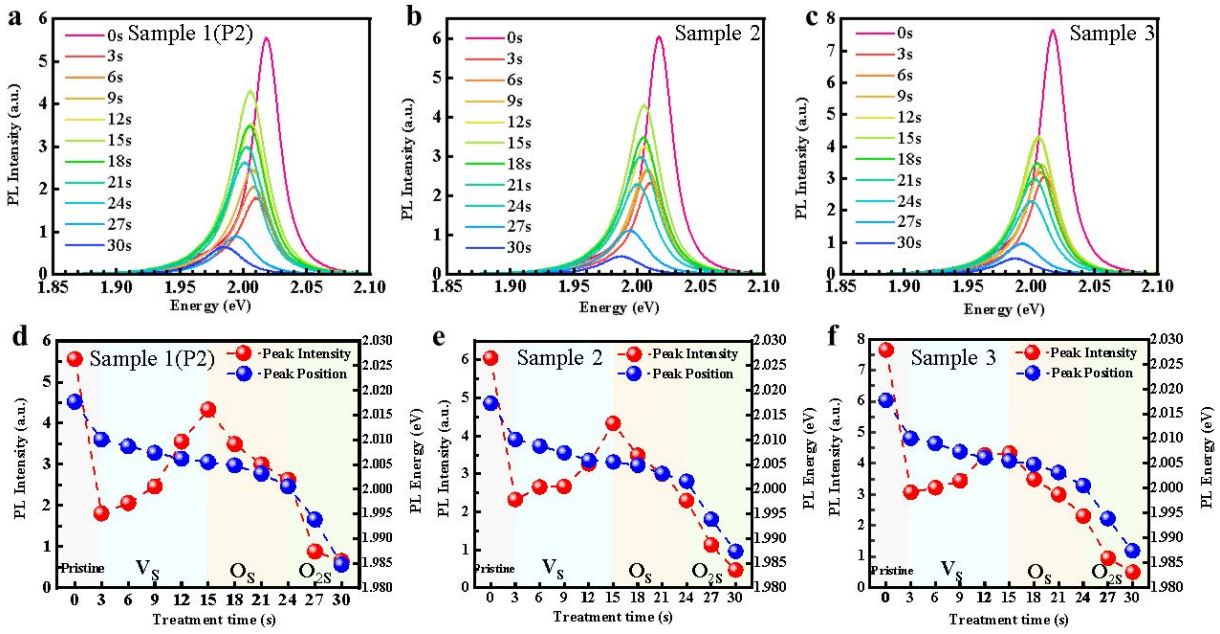

Figure S3. a) PL spectra at different location (P2) for WS_2 monolayer. b) and c) PL spectra of sample 2 and sample 3 treated by O^{2+} plasma at different treatment times (calibrated by silicon Raman peak ~520cm^{-1}). d)-f) PL intensity and energy as the function of the treatment time for different samples.

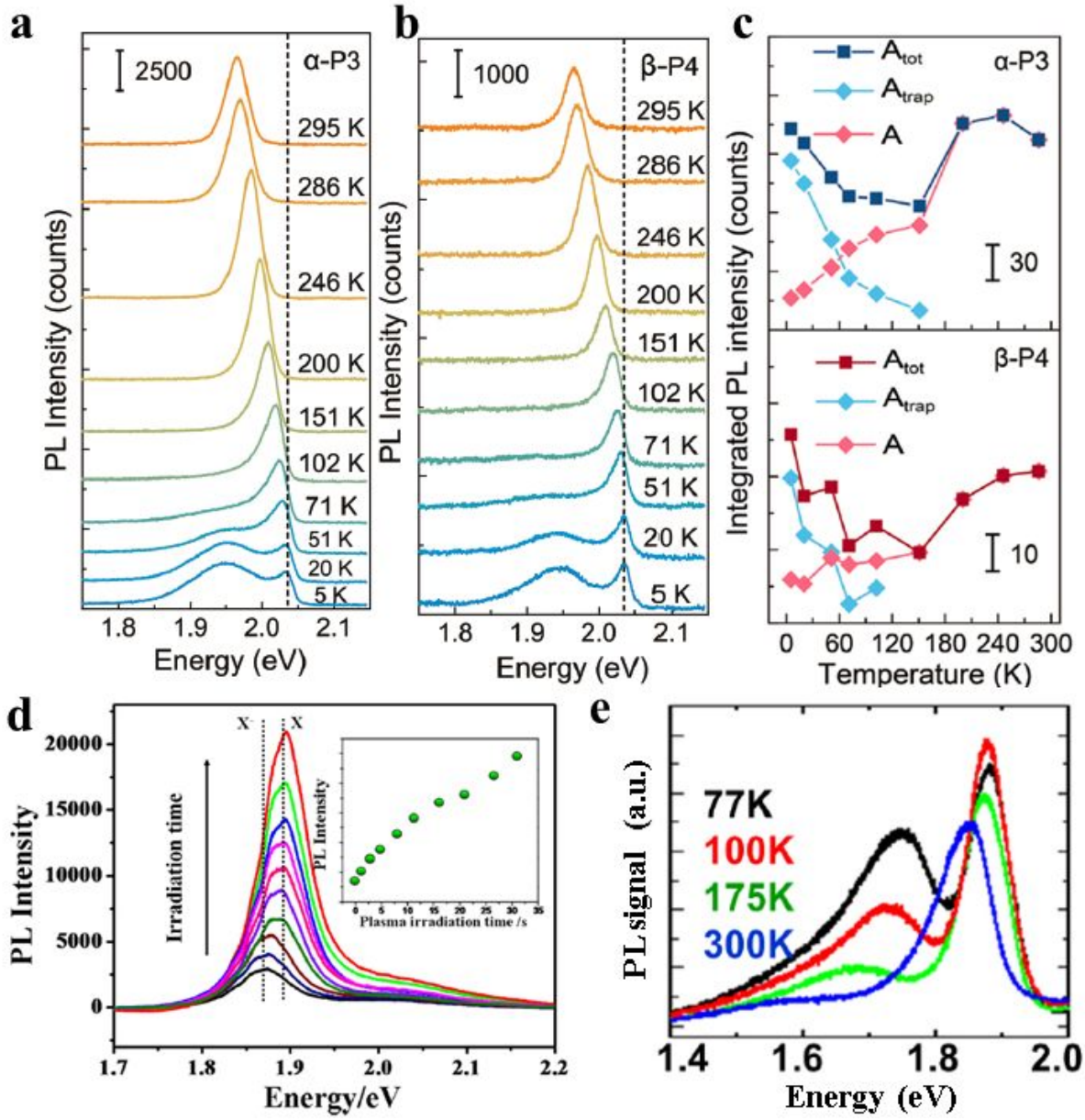

Figure S4. a)-b) The temperature-dependent PL spectrum of the two positions α ─ P3 and β ─ P4 in monolayer WS_2, respectively. c) The PL intensity of the A, A_{trap}, and A_{tot} at α-P3 (up) and β-P4 (down) as a function of temperature[1]. d) PL spectra of monolayer MoS_2 after oxygen plasma irradiation with different durations[2]. e) PL spectrum measured over the temperature range from 77 K to 300 K of a monolayer MoS_2, defect-induced PL peak (X_B) [3].

Table S1. Photoluminescence quantum yield (Φ) and radiative lifetime (τ_r) of WS_2 monolayer treated by O^{2+} plasma at different treatment times

Treatment Time (s)	Φ (%)	τ_r (ns)
0	0.0600	21.67
3	0.0187	26.69
6	0.0232	16.39
9	0.0261	13.41
12	0.0338	5.92
15	0.0604	5.30
18	0.0534	5.43
21	0.0488	5.94
24	0.0427	7.49
27	0.0132	18.24
30	0.0073	28.80

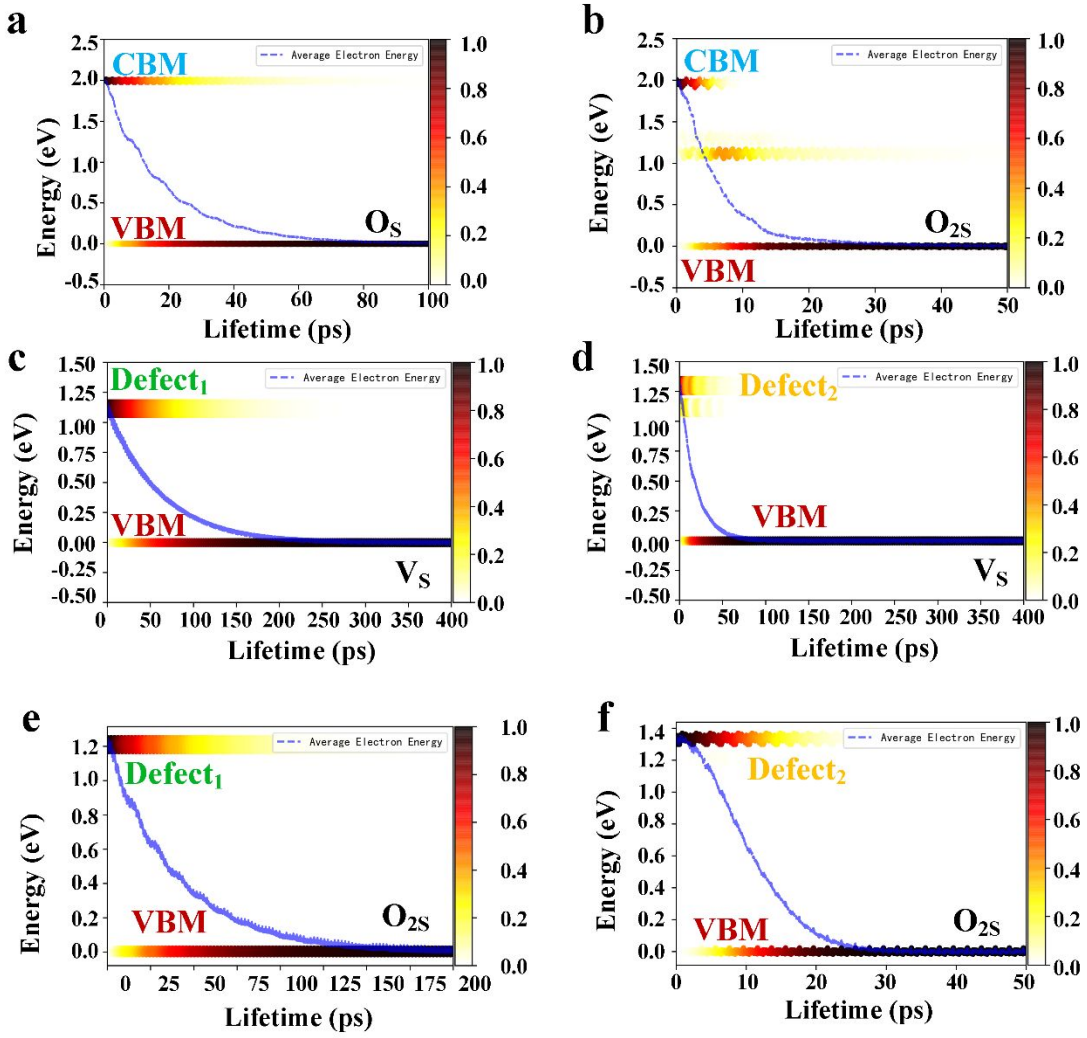

Figure S5. Nonadiabatic molecular dynamics results of defective WS_2 monolayer. a) and b) electron energy change from band edge in defective WS_2 with O_S defect and O_{2S}, respectively. c) and d) electron energy change from the first defect state (defect 1) and the second defect state (defect 2) in defective WS_2 with V_S, respectively. e) and f) electron energy change from the first defect state (defect 1) and the second defect state (defect 2) in defective WS_2 with O_{2S}, respectively.

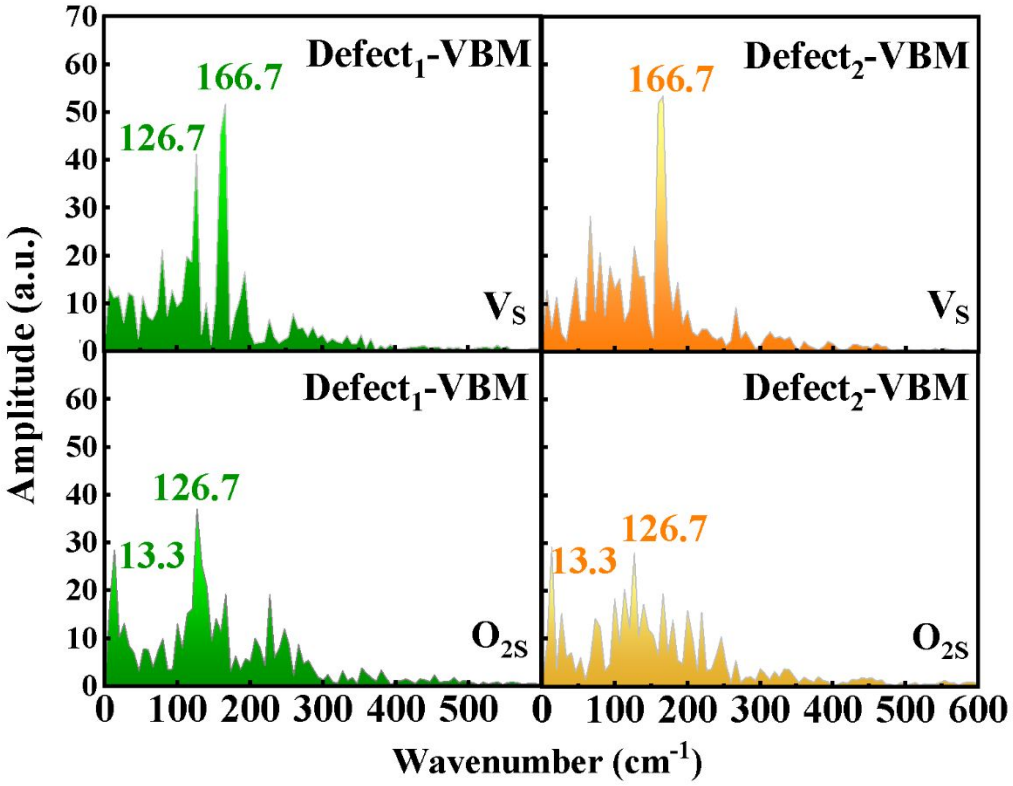

Figure S6. FT spectrum of between the first defect state (defect 1) as well as the second defect state (defect 2) and VBM in defective WS_2 monolayer with O_S defect and O_{2S}.

Reference

(1) Wu, K.; Zhong, H.; Guo, Q.; Tang, J.; Yang, Z.; Qian, L.; Yuan, S.; Zhang, S.; Xu, H., Revealing the Competition between Defect-Trapped Exciton and Band-Edge Exciton Photoluminescence in Monolayer Hexagonal WS_2. *Adv. Opt. Mater.* **2022**, 2101971.

(2) Nan, H.; Wang, Z.; Wang, W.; Liang, Z.; Lu, Y.; Chen, Q.; He, D.; Tan, P.; Miao, F.; Wang, X.; et al. Strong Photoluminescence Enhancement of MoS_2 through Defect Engineering and Oxygen Bonding. *ACS Nano* **2014**, *8*(6), 5738-5745.
(3) Tongay, S.; Suh, J.; Ataca, C.; Fan, W.; Luce, A.; Kang, J. S.; Liu, J.; Ko, C.; Raghunathanan, R.; Zhou, J.; et al.Defects Activated Photoluminescence in Two-Dimensional Semiconductors: Interplay Between Bound, Charged, and Free Excitons. *Rep* **2013**, *3*, 2657.