Revealing Defect-State Photoluminescence in Monolayer WS_2 by Cryogenic Laser Processing

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S1. Raman signatures of monolayer WS_2

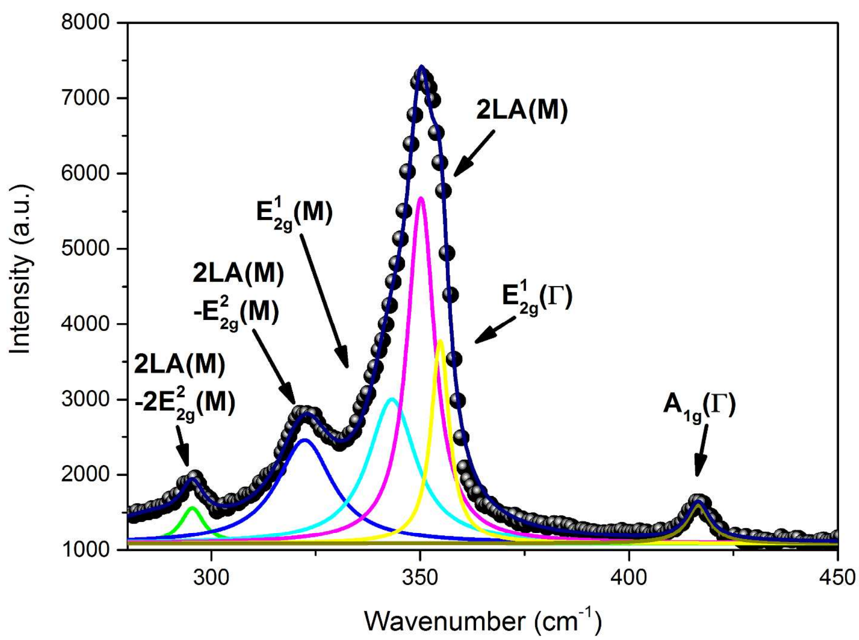

Figure S1. Raman signatures of monolayer WS_2.

S2. Another example of the emergence of peak U after laser processing in an

aged sample

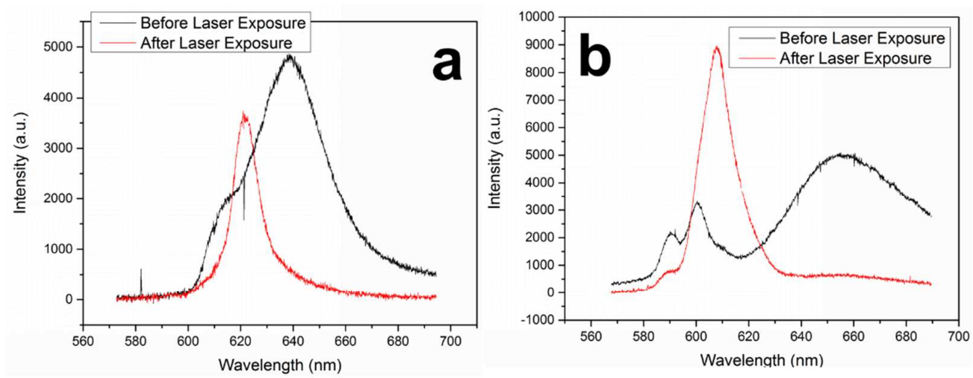

Figure S2. PL spectra showing another example of the emergence of peak U after laser processing from 8-month aged (a) non-transferred and (b) transferred WS_2.

S3. PL spectrum after laser processing at very low excitation power

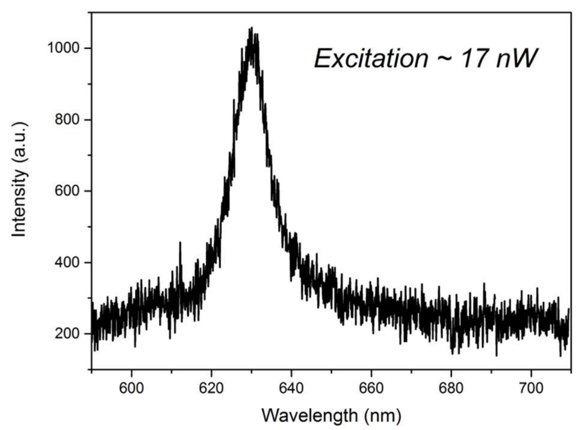

Figure S3. PL spectrum of laser treated aged sample under low excitation power of 17 nW.

S4. Testing Laser Induced Damage

Here we present Raman spectra and optical images before and after laser processing at power density of ~8 kW/cm^2 for 10 minutes on the same domain. There is no noticeable intensity or peak position change after laser processing. Optical images also shows that the laser treatment does not introduce any optical contrast variation.

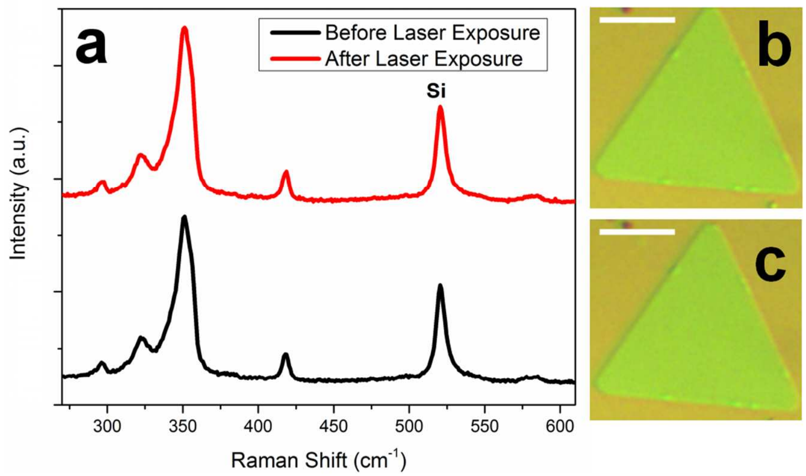

Figure S4. (a) Raman spectra of monolayer WS_2 before and after laser processing. Black curve: Before laser exposure. Red curve: after laser exposure. (b,c) Optical image of monolayer WS_2 (b) before and (c) after laser processing. Power density: ~8 kW/cm^2, time: 10 minutes. Scale bar: 15 μm.

S5. Power dependent PL color map

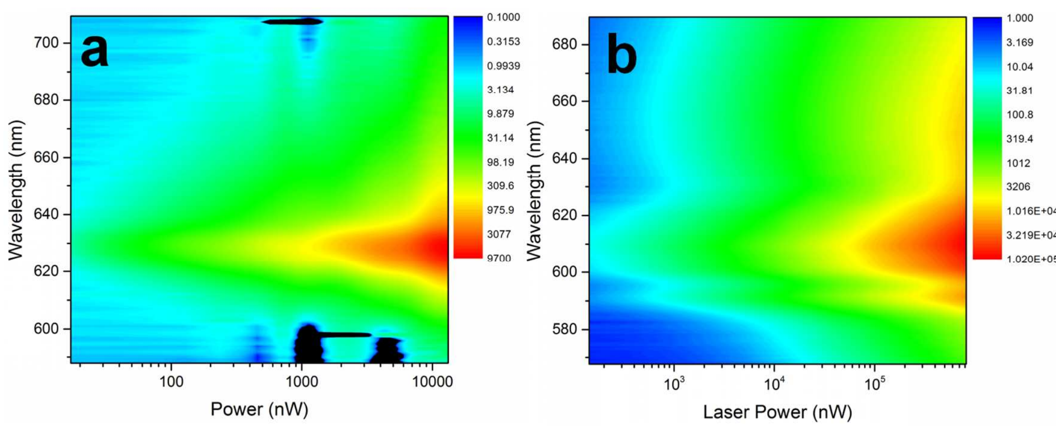

Figure S5. Power dependent PL map of (a) non-transferred and (b) after transfer after laser processing.

S6. Morphology characterization of WS_2 before and after heating treatment

Here we present the typical optical microscopy and SEM images of monolayer WS_2 with and without heating treatment, which clearly shows that there is no observable degradation even after 90 minutes annealing at 150 °C in air.

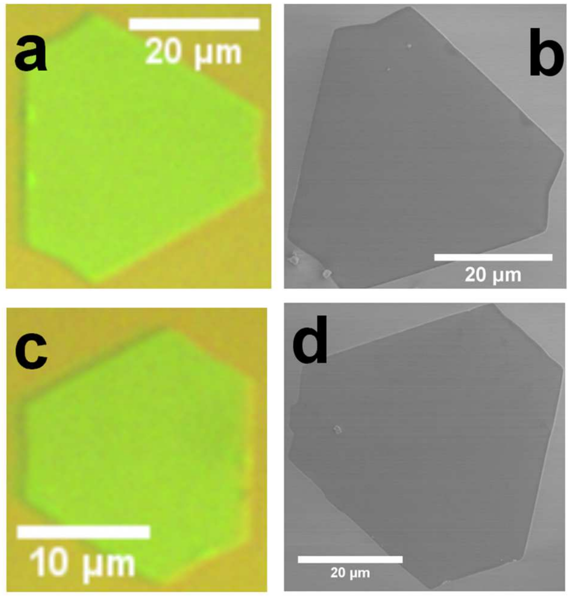

Figure S6. Optical images and SEM image (a,b) before and (c,d) after heating at 150 °C for 90 minutes. Scale bar: (a) 20 µm, (b) 20 µm, (c) 10 µm, (d) 20 µm .

S7. PL spectra of monolayer WS_2 as a function of ageing time in ambient condition

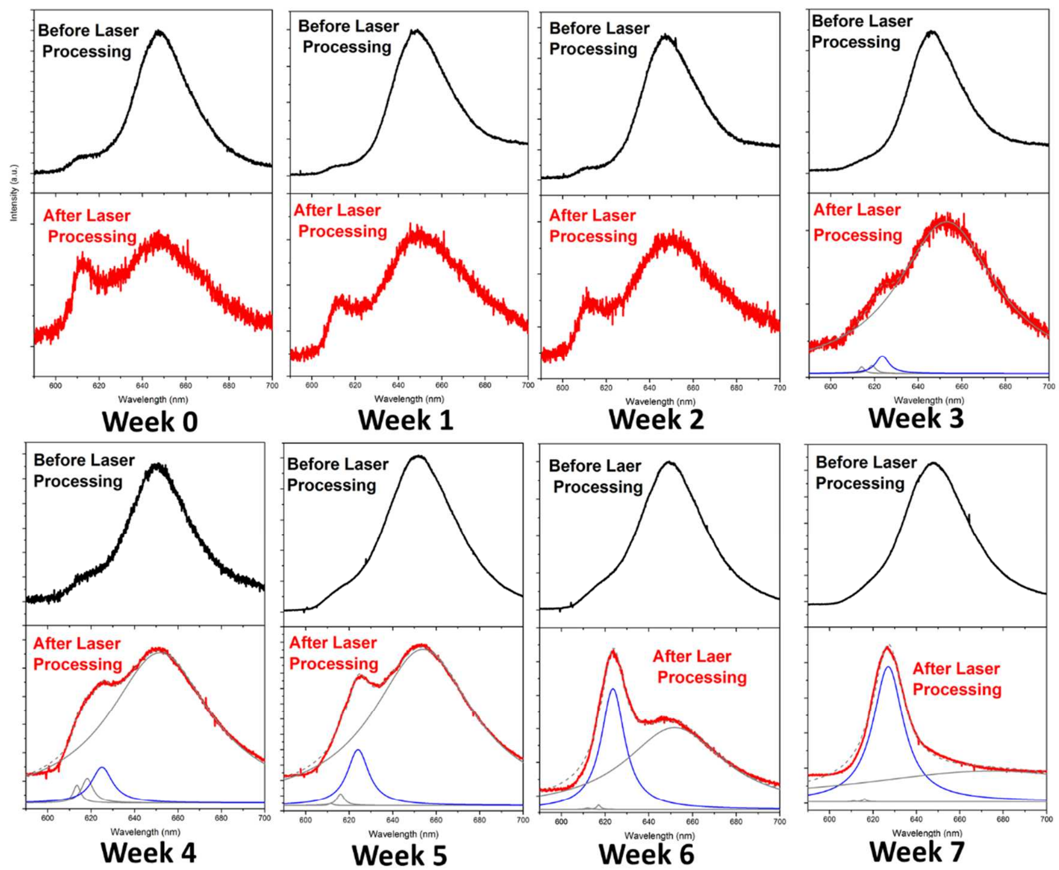

Figure S7. PL spectra before and after laser processing of monolayer WS_2 as a function of ageing time. From week 3 to week 7, Lorentzian peak fitting is performed to extract the contribution of peak U. Gray solid curves: contributions from exciton, trion and LS. Blue solid curves: peak U. Gray dashed curves: cumulative peaks.