Electronic Supplementary Information:

Straining Effects in MoS_2 Monolayer on Nanostructured Substrates: Temperature Dependent Photoluminescence and Exciton Dynamics

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Methods:

1. Transfer of CVD MoS_2 monolayer onto nanostructured surface

To transfer MoS_2 monolayer onto the nanostructured SiO_2/Si substrate, PMMA (methyl methacrylate) is spin coated on monolayer MoS_2 grown by chemical vapor deposition (CVD). The sample is baked at 150℃ for 2 minutes and soaked in a diluted potassium hydroxide (KOH) solution to separate MoS_2 from the substrate. The detached PMMA/MoS_2 stack is thoroughly cleaned by DI water and wet-transferred onto the nanostructured substrate. The transferred sample is air dried and PMMA is removed by sequentially soaking in acetone and isopropyl alcohol (IPA) for 30 minutes. The samples are then dried in nitrogen gas atmosphere.

2. Laser shock process

The whole laser shock system consists a transparent confinement, a laser source, an ultrathin metal foil and a layer of ablative coating. A pulsed Q-switch Nd-YAG laser (Continuum® Surelite III) was used as the laser source. The beam size is controlled with a focusing lens. The beam diameter was calibrated by a photosensitive paper (Kodak Linagraph, type: 1895) to be 3 mm. The confining media is clinical glass slide and the ablative coating (thickness in the range of 1-10 μm) is aerosol graphite painting sprayed on 4 μm thick aluminum foil (Lebow Company Inc, Bellevue, WA). Laser beam transmits through the confinement and is absorbed by the ablative coating, introducing vaporization effect and plasma expansion. The confining media delays and intensifies the plasma expansion. An X-Y stage is used to move the processing stage.

3. Characterizations

The prepared MoS_2 monolayer sample was characterized using scanning electron microscopy (SEM, Hitachi S-4800, acceleration voltage of 5 kV), Raman spectroscopy (Horiba, LabRAM HR-800, excitation laser wavelength of 532 nm) and atomic force microscopy (AFM, Veeco Dimension 3100).

4. PL measurement

The photoluminescence (PL) spectra were obtained on both the flat and nanostructured areas of the MoS_2 monolayer at different temperatures ranging from 10 to 300 K. During the measurement, the sample is excited by a continuous-wave 532 nm laser beam which is focused to a 1.4 μm spot on the sample by a long working distance objective lens (50×, 0.55 NA). The same objective lens collects the PL signal which is analyzed by a spectrometer and recorded by a TEC cooled CCD camera. The sample is placed in a microscopy liquid helium cryostat so that the sample temperature can be varied.

Figures:

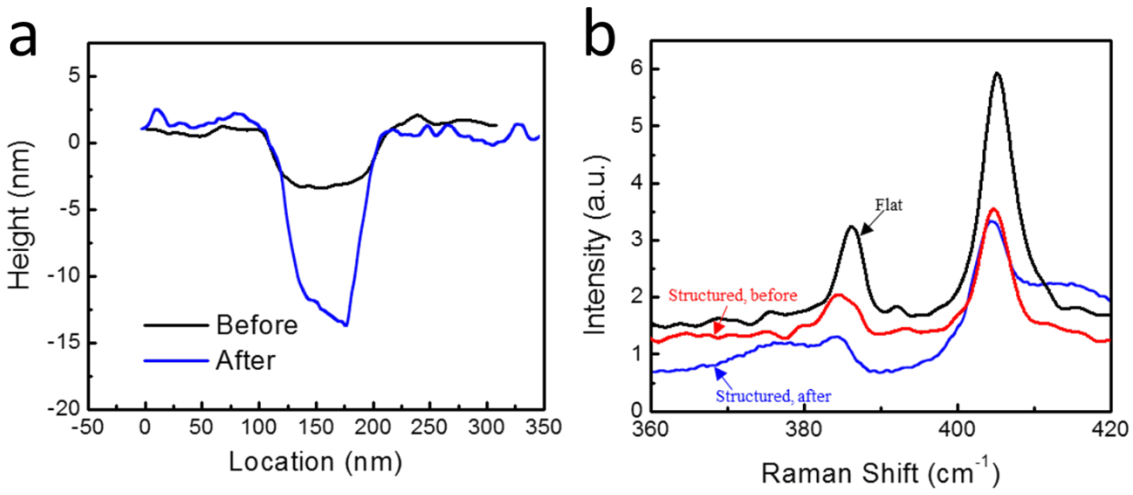

Figure S1: (a) AFM line profile of the surface morphology of nanodots array transferred with MoS_2 before and after laser shock treatment. (b) Raman spectra of MoS_2 at the flat and structured areas (Laser excitation wavelength: 532 nm) before and after laser shock treatment. An average tensile strain less than 1% is present on the structured area. And after laser shock treatment, the split mode with lower frequency (E_g^{-1}) is with a broad peak and is shifted to around 376 cm^{-1}, which is around 10 cm^{-1} shifts compared to the original peak (386 cm^{-1}) and 8.3 cm^{-1} shifts compared to the peak before laser shock (384.4 cm^{-1}). This indicates an additional strain of 2% is introduced based on the shift rates reported[1].

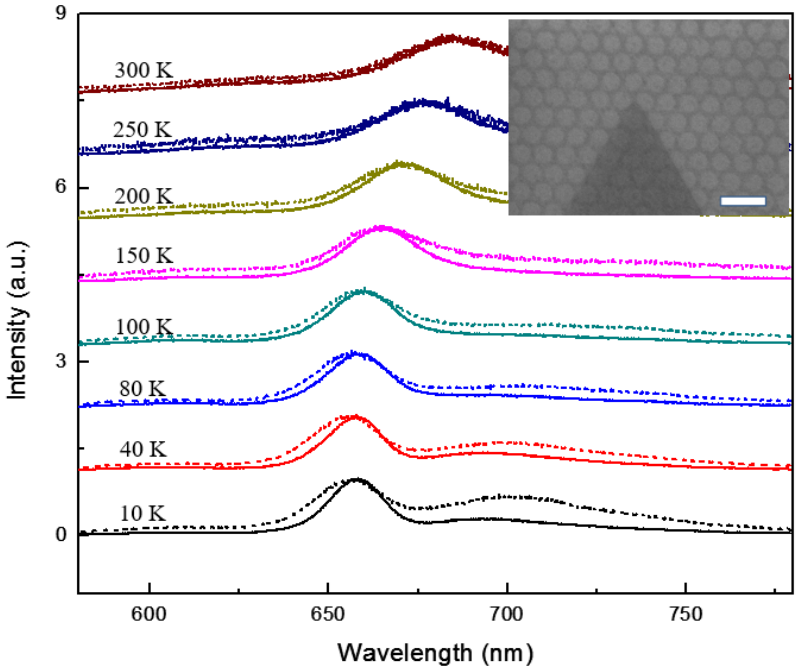

Figure S2: PL spectra of MoS_2 from the supported (solid lines) and suspended (dashed lines) areas at various temperatures. Inserted is the SEM image of the structure. Scale bar: 500 nm. The structural dimensions of the substrate nanofeatures strongly influence the strain levels and temperature dependent characteristics. For nanodots arrays, the introduced strains are biaxial and non-homogeneous.

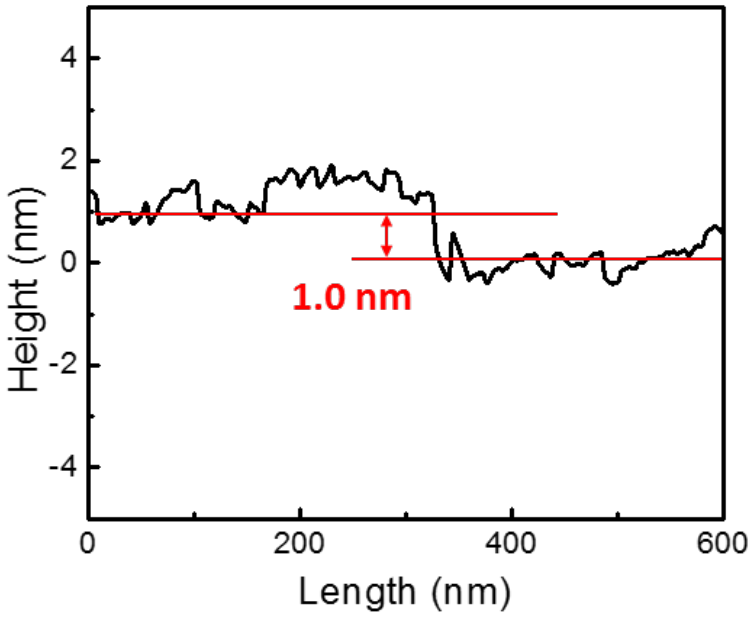

Figure S3: AFM line profile of the MoS_2 sample on a nanodot array showing the monolayer characteristic. The step height for a monolayer 2D MoS_2 could be larger than the actual thickness due to surface wrinkles of the material, residuals of polymer layers used during transfer process, and other molecules adsorbed on the surface.

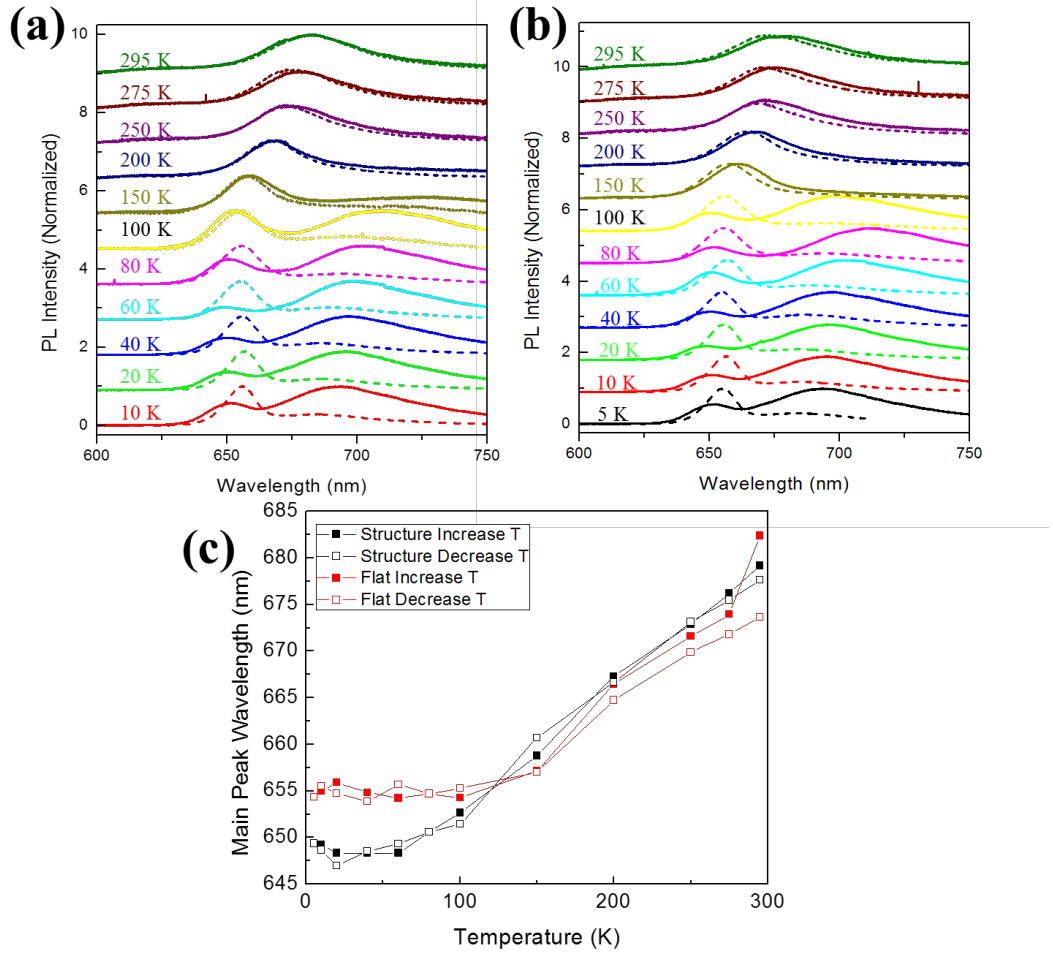

Figure S4: PL spectra of CVD MoS_2 on flat and structured areas with increasing (a) and decreasing (b) temperatures. (c) Peak positions of the PL spectra at different temperatures. The data shown here were taken on a different sample which was prepared using the same procedure. The peak positions of the monolayer CVD MoS_2 on both flat and structured areas are located at around 680 nm at room temperature, consistent with previous reports. The spectra obtained when decreasing temperatures from 300 K are identical to those obtained from increasing the temperature from 10 K, demonstrating a reversible substrate straining effect. The difference of the peak positions at flat and structured areas could be clearly visualized only at low temperature (below 100 K in the present case), agreeing with the findings discussed in the main text.

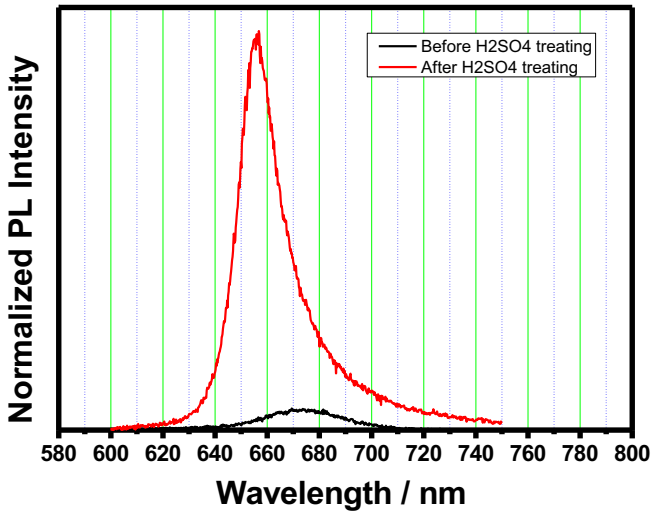

Figure S5: PL spectra of monolayer MoS_2 before and after concentrated hydrogen sulfuric acid. The monolayer MoS_2 was washed with concentrated (98%) hydrogen sulfuric acid for 10 s and dried with N_2 to blow the residue. A significant enhance photoluminescence was noticed on the flakes treated with concentrated hydrogen sulfuric acid. The peak position is blue-shifted by around 20 nm, due to the elimination of substrate-borne moisture.

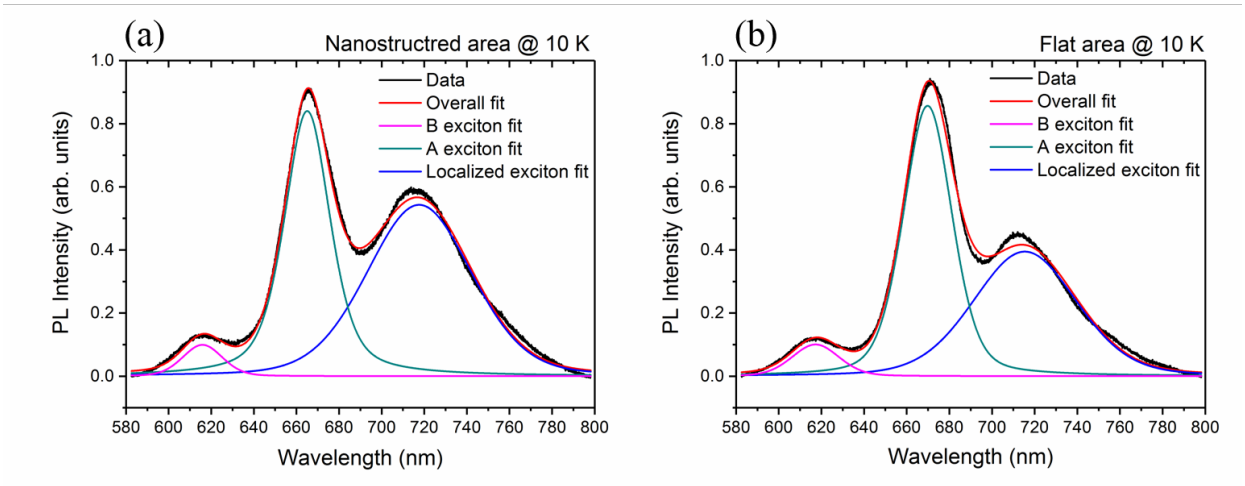

Figure S6: Multiple-peak fitting of the PL spectra from (a) nanostructured area and (b) flat area at a sample temperature of 10 K. The fit includes three peaks corresponding to B exciton, A exciton, and localized exciton resonances. The experimental spectrum, individual peak and overall fit are plotted in both cases.

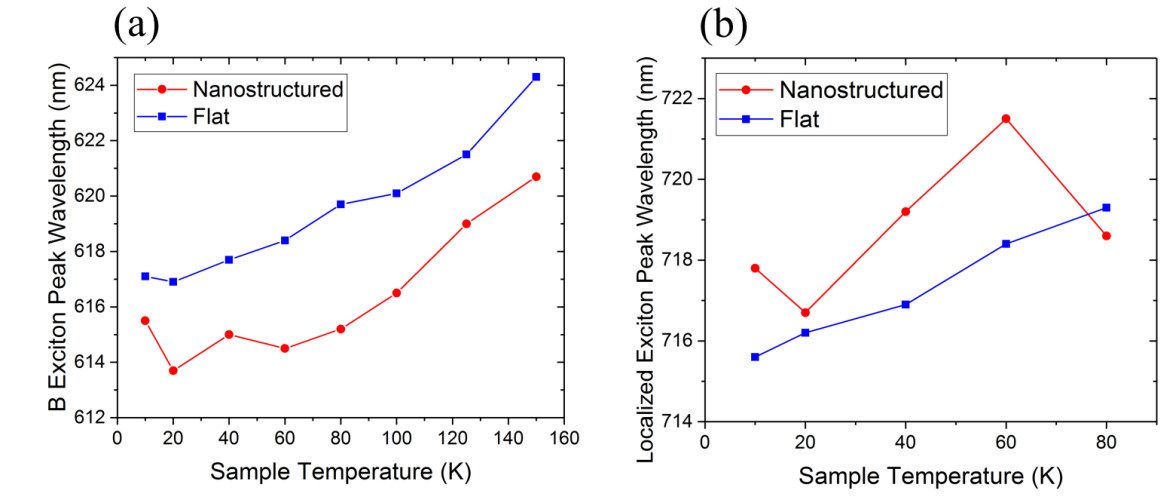

Figure S7: Peak wavelengths of (a) B exciton and (b) localized exciton in the flat (blue) and nanostructured (red) areas at various temperatures.

References:

1 J. U. Lee, S. Woo, J. Park, H. C. Park, Y. W. Son and H. Cheong, *Nature Communications*, 2017, **8**, 1370.