Anomalous enhancement of valley polarization in multilayer WS_2 at room temperature

Huimin Su, Chengrong Wei, Aiying Deng, Dongmei Deng, Chunlei Yang, Jun-Feng Dai

Supplementary Information

1. Second harmonic generation measurement

The angle-dependent intensity of parallel (black squares) and perpendicular (red circles) components of the second harmonic generation (SHG) signal are shown in Fig. S1b. The fitting of the experimental data to the expected quadratic dependence of $sin(3\theta)$ and $cos(3\theta)$ indicates the high crystal symmetry and orientation of the monolayer WS_2. The mapping of SHG intensity as fixed angle is shown in Fig. S1c. The SHG signal is uniform over the entire monolayer flake, which justify low grain boundaries on exfoliated WS_2 samples.

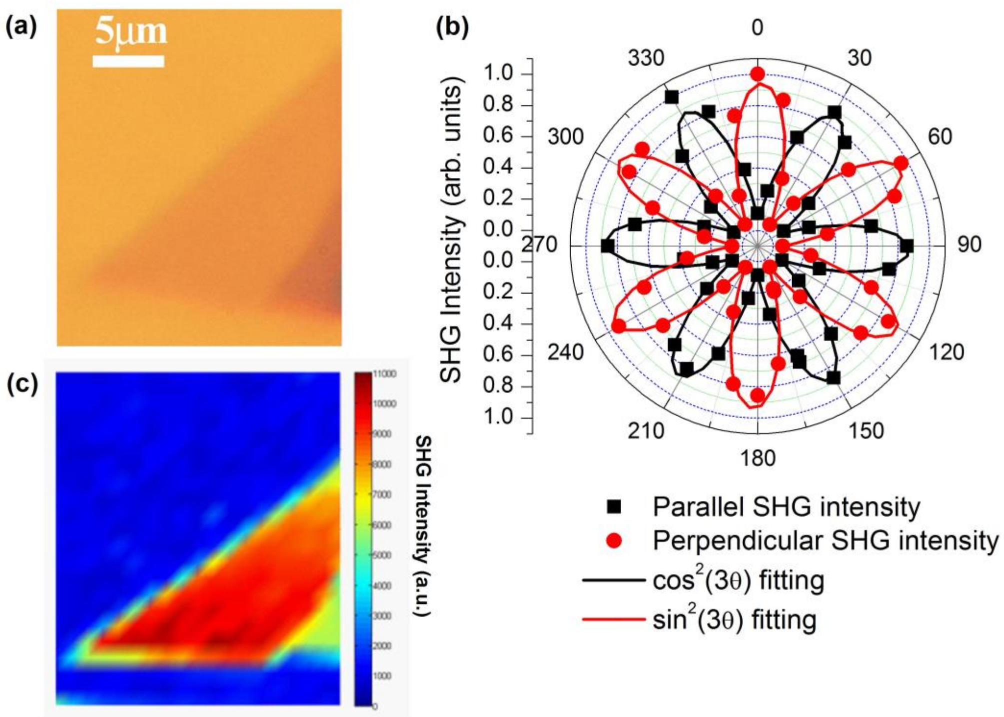

Figure S1: (a) Optical image of monolayer WS_2. (b) The intensity of parallel (blue squares) and perpendicular (black circles) components of second harmonic generation signal as a function of θ, the angle between the laboratory and the crystalline coordinates. The black (red) solid line indicates the expected $\sin^2 3\theta$ ($\cos^2 3\theta$) dependence. (c) The parallel component of SHG image of a monolayer WS_2 on 0^o angle.

2. Photoluminescence (PL) spectrum

The 2-dimensional optical image and mapping of PL of monolayer WS_2 are shown in Figure S2 (a) and (b), respectively, which supports high quality and uniform of monolayer WS_2 sample.

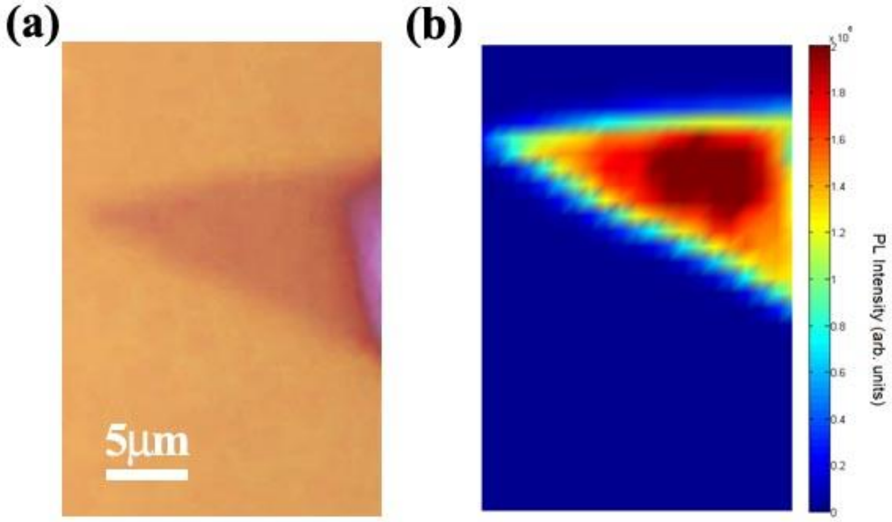

Figure S2: (a) optical image of monolayer WS_2 (b) the photoluminescence (PL) image of a monolayer WS_2.

3. Valley polarization measurement

The degree of circular polarization of photoluminescence is evaluated at PL peak from exciton emission with 10nm bandwidth by using the following equation:

$$P = \frac{I_{\sigma+} - I_{\sigma-}}{I_{\sigma+} + I_{\sigma-}}$$

where $I_{\sigma+}(I_{\sigma-})$ is the intensity of left-handed (right-handed) circularly polarized photoluminescence. The valley polarization as a function of emission energy is shown in Fig. S2 and Fig. S3.

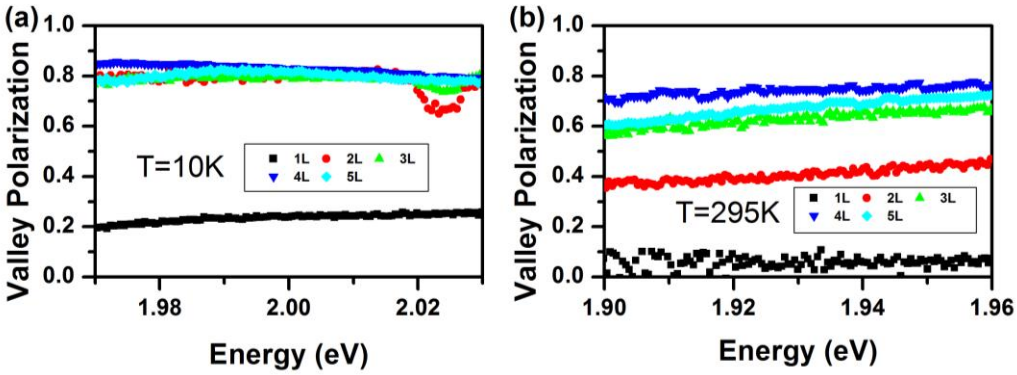

Figure S2: The valley polarization as a function of emission energy around PL peak for 1- to 5- layer WS_2. The single−frequency laser with an energy of 2.088 eV is used to excite monolayer and multilayer WS_2 at (a) 10 K and (b) 295 K.

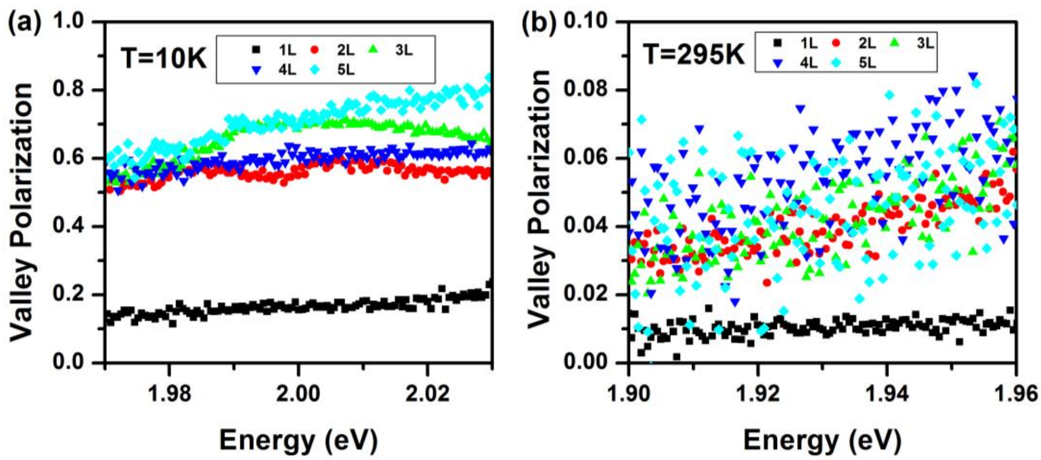

Figure S3: The valley polarization as a function of emission energy around PL peak for 1- to 5- layer WS_2. The single−frequency laser with an energy of 2.33 eV under the off-resonant condition, is used to excite monolayer and multilayer WS_2 at (a) 10 K and (b) 295 K.

4. Valley polarization in multilayer WS_2 samples at various temperature

The valley polarization in 2L, 3L 4L and 5L WS_2 samples as a function of temperature are shown in Fig. S4. The valley polarization in bilayer and trilayer WS_2 exhibits obvious temperature dependence above around 200K, while it is weakly temperature dependent in 4L and 5L samples. Above around 200K, the most apparent drop of valley polarization is observed on bilayer WS_2.

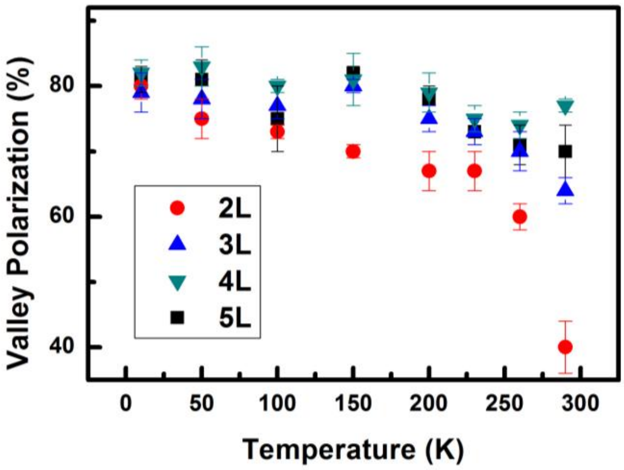

Figure S4: The valley polarization of multilayer WS_2 (including 2, 3, 4, 5 layers) as a function of temperature.