Supporting information

Pressure-Induced K-Λ Crossing in monolayer WSe_2

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Sample fabrication and diamond anvil cell (DAC) device

The monolayer and bilayer WSe_2 samples were prepared by micromechanical exfoliation of a bulk WSe_2 crystal (provided by 2D Semiconductors, USA) on a thinned 270 nm SiO_2/Si substrate. Regions of mono- and bi-layer WSe_2 were identified by their optical contrast with an optical microscopy, seen in Figs.S1(a) and S1(b). The contrasts were confirmed using PL measurements. Figures S1(c) and S1 (d) show the schematic diagrams and photograph of the pressure chamber of DAC device. The center part of the DAC device is composed of a pair of diamond anvils with culet diameter of about 700 μm and a 500-μm-thick gasket (T301 stainless steel) as shown in Fig. S1(c) . The gasket is first incused, and then a hole is drilled in the center of the incused position. The pressure chamber of the DAC with a diameter of ~ 350 μm and a height of ~ 300 μm is made, as shown in Fig. S1(d). Liquid Argon is used as the pressure transmitting medium (PTM) in the DAC which shows a good hydrostatic pressure behavior at room temperature. Hydrostatic pressure generated inside the pressure chamber is monitored in situ using the ruby R1 fluorescence line shift. The pressure value can be calculated by the equation[1-3],

$$P = \frac{1094}{B} \times \left[\left(\frac{\lambda}{\lambda_0} \right)^B - 1 \right]$$

where P is the pressure in GPa, λ_0 and λ present the wavelength of ruby R1 line at zero pressure and the certain pressure P, respectively. B is a constant of 7.665 at hydrostatic and quasi-hydrostatic pressures. Figure S1(e) exhibits the shift of the ruby R1 line as increasing pressure from 0 to 5.43 GPa.

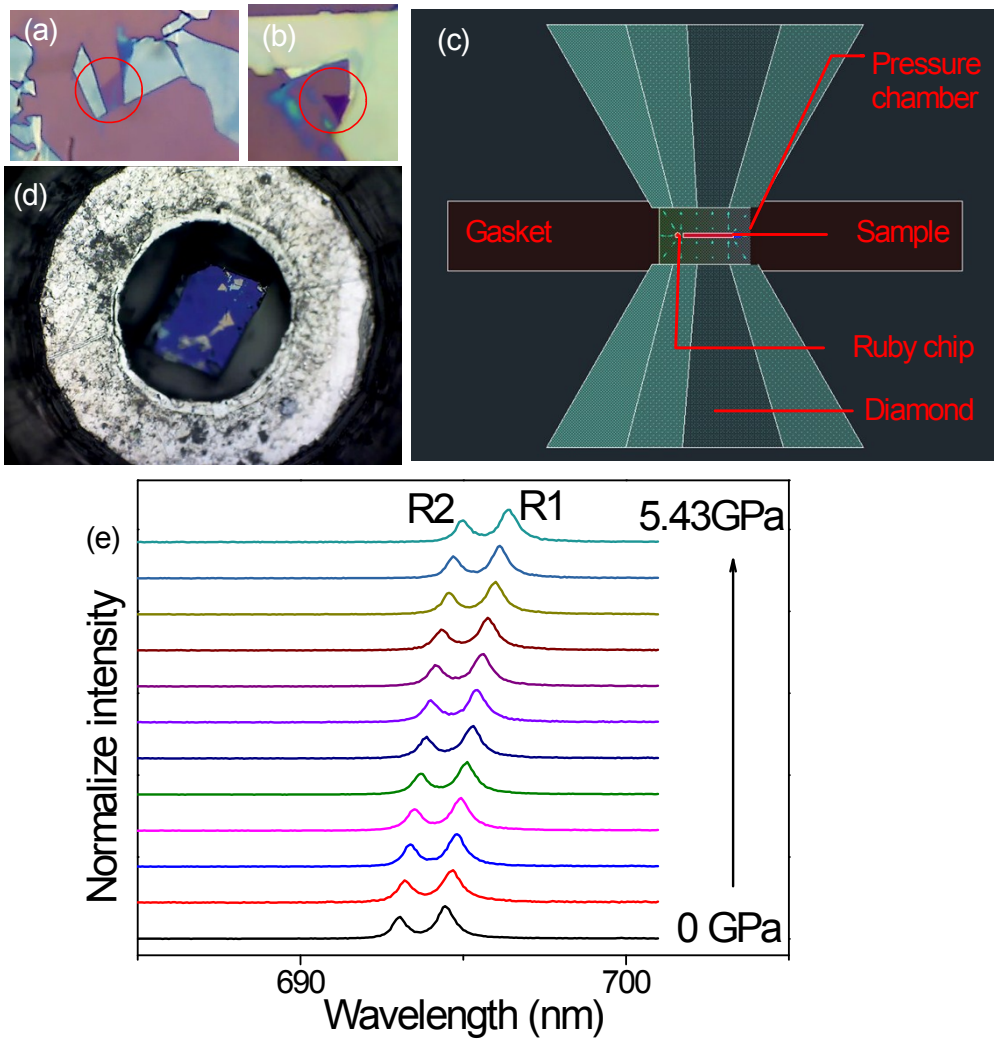

Figure S1. Optical microscopic image of the micromechanically exfoliated WSe_2 samples, both monolayer (a) and bilayer (b) are signed with red circles. (c) Schematic diagram of the DAC device, and (d) the photograph of the pressure chamber , together with the sample.. (e) PL spectra of the ruby as the pressure increasing from 0 to 5.43 GPa.

Micro-PL measurements under pressure

Micro-PL measurements were taken at room temperature using an optical confocal microscopy setup, as shown in Fig. S2. A continuous wave solid-state laser beam with a wavelength of 454 nm and an average power of approximately 0.1 mW was focused on the sample in the DAC device using an objective (NA=0.35). The emitted PL was collected by the same objective and analyzed using a 0.5 m monochromator equipped with a silicon charge-coupled device.

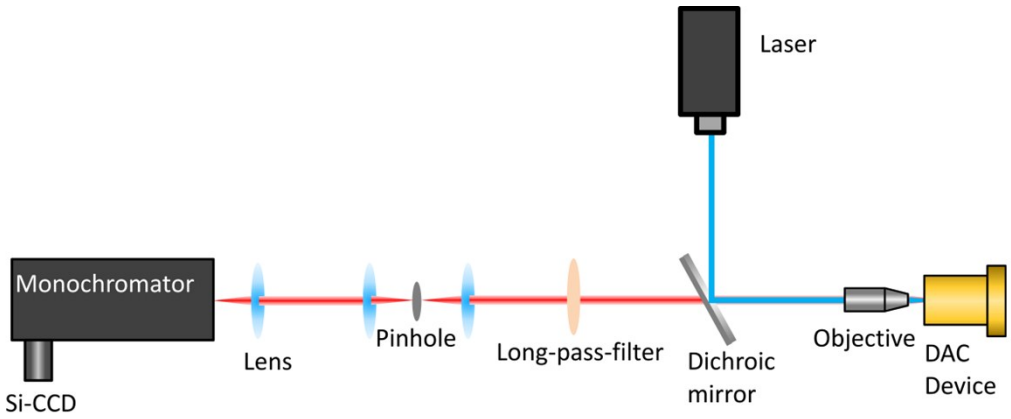

Figure S2. Schematic diagram of the experimental setup.

Pressure dependence PL spectra analysis

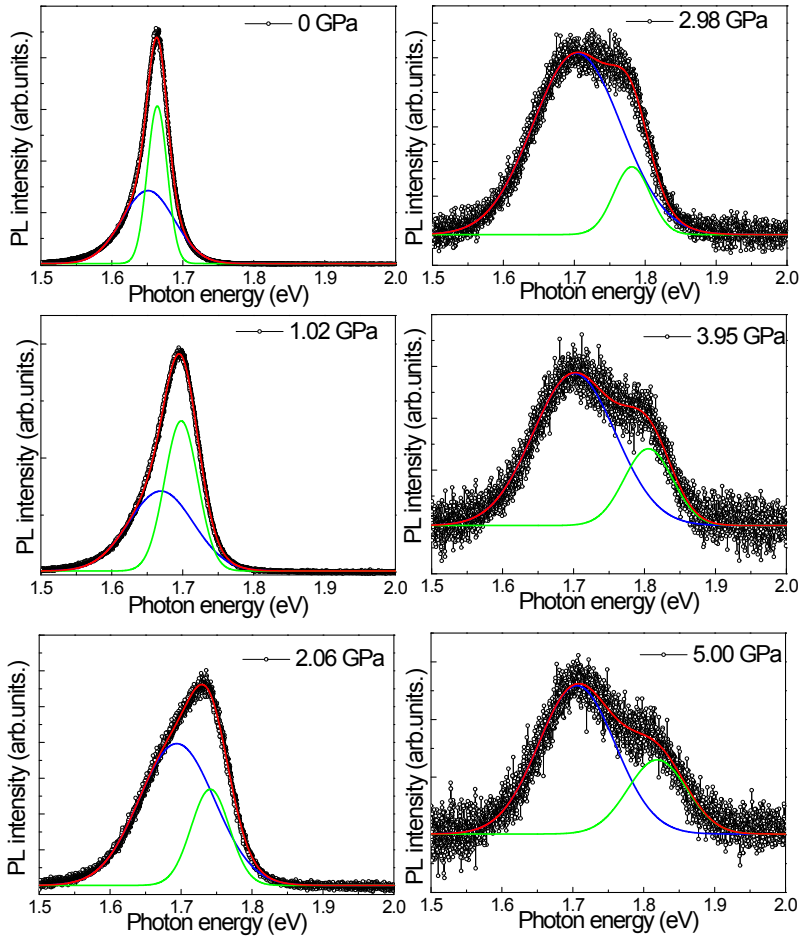

Figure S3. The PL spectrum corresponding to the exciton(peak X, green line) and negatively charged exciton(peak X^-, blue line) emissions under pressure of 0, 1.02, 2.06, 2.98, 3.95 and 5.00 GPa of monolayer WSe_2. Two spectrally-resolved peaks are

obtained by using Gaussian function fitting.

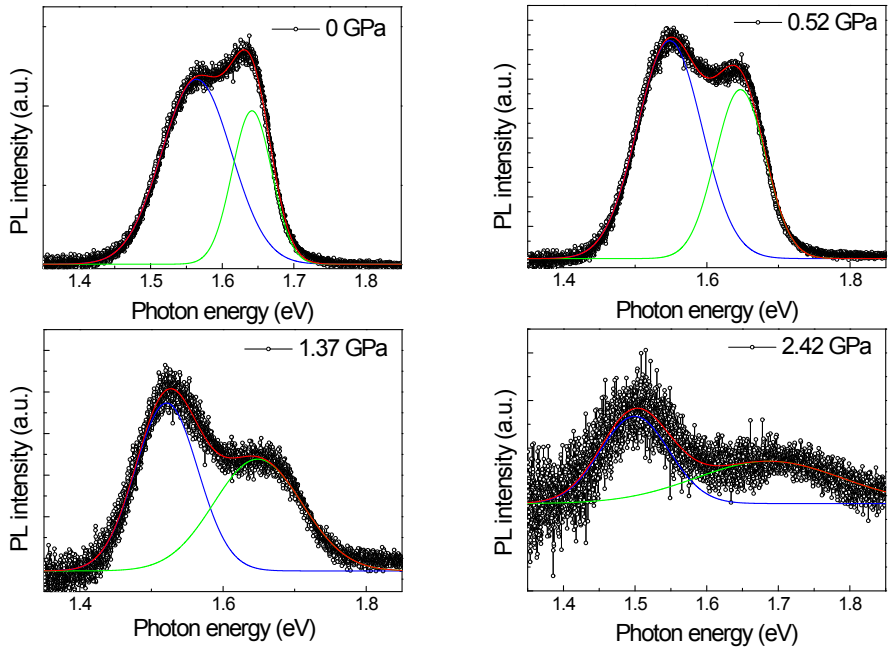

Figure S4. The PL spectrum corresponding to peaks X_K (green line) and $X_{人}$ (blue line) under pressure of 0, 0.52, 1.37 and 2.42 GPa of bilayer WSe_2. Two spectrally-resolved peaks are obtained by using Gaussian function fitting.

As can be seen in Figure S3, the PL spectrum under pressure of 0, 1.02, 2.06, 2.98, 3.95 and 5.00 GPa are displayed. The center photo energy of exciton and negatively charged exciton are positioned by two Gaussian function fit. As the evolution of both peaks shown in Fig. S3, peak X keeps blue shift as the pressure increased while peak X^- approximately shows not change at pressure 2.98 to 5.00 GPa. Moreover, the peak intensity as a function of pressure shown in Fig. 2(d) in main text can be observed visually In the process of fitting. Figure S4 displays the homogeneous information in bilayer.

REFERENCE:

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2. Piermarini, G. J.;Block, S.;Barnett, J. D. and Forman, R. A. Calibration of the pressure dependence of the R1 ruby fluorescence line to 195 kbar. *Journal of Applied Physics* **1975,** 46, 2774.
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