Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Wrinkle and Near Resonance Effect on the Vibrational and Electronic properties in Compressed Monolayer $MoSe_2$

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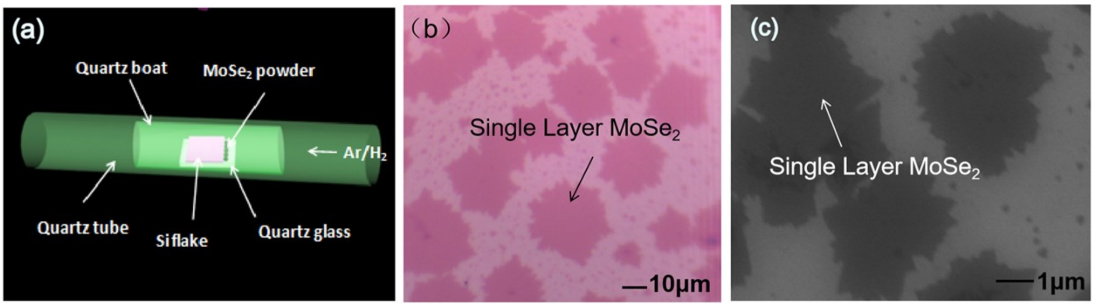

Figure S1. **(a)** Schematic illustration for the growth of monolayer $MoSe_2$ on Si/SiO_2 substrate. $MoSe_2$ powders were placed in a quartz tube as the precursor. Argon and H_2 were used as the carrier gas. **(b)** Optical image and **(c)** scanning electron microscopy (SEM) image of monolayer $MoSe_2$ deposited on Si/SiO_2 substrate.

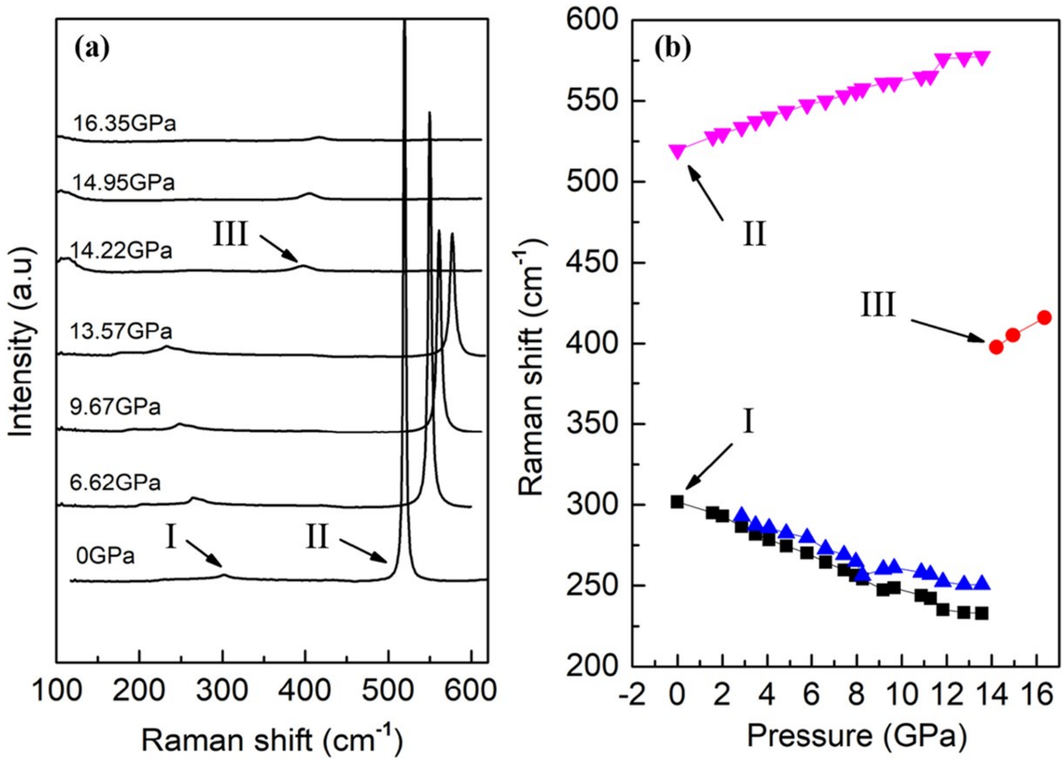

Figure S2. **(a)** The Raman spectra of Si/SiO_2 substrate at selected pressures. **(b)** Pressure dependent Raman frequencies of mode I, II, and III in Si/SiO_2 substrate. A novel mode marked as “mode III”emerges after 14.22 GPa.

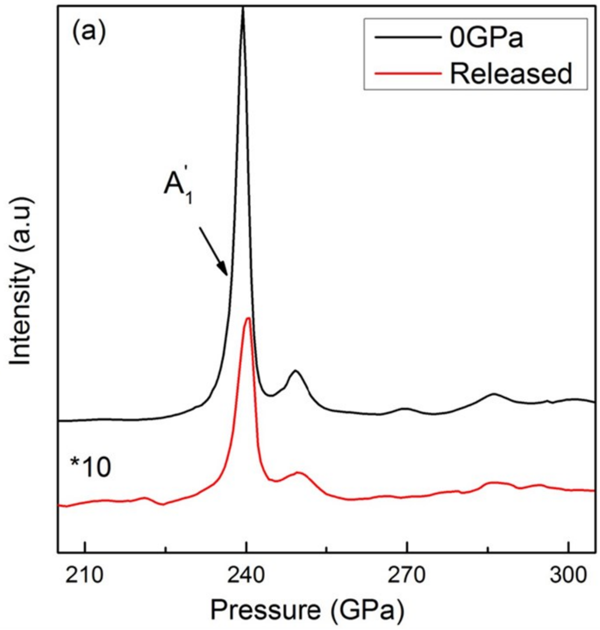

Figure S3. (a) The Raman spectra of monolayer $MoSe_2$ supported on Si/SiO_2 substrate at ambient condition and after removing pressure. When compared with the Raman spectra of monolayer $MoSe_2$ at ambient condition, the Raman frequencies after removing pressure exhibit a slightly blue shift, which should be related with the residual strain from the substrate deformation. The intensity of released Raman on Si/SiO_2 seriously decreases, which can be attributed to the structural distortions of monolayer $MoSe_2$ owing to the distortion of Si/SiO_2 substrate under high pressure.

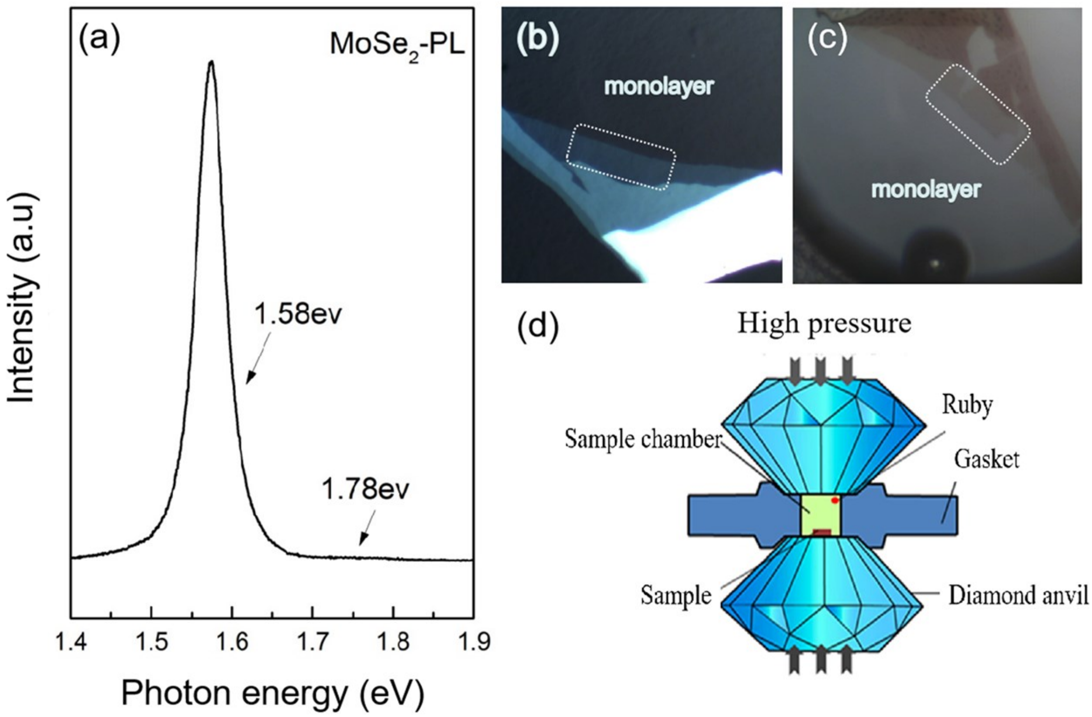

Figure S4. (a) The PL spectra of exfoliated monolayer $MoSe_2$ at ambient condition. A and B excitons were located at 1.58eV (785nm) and 1.78eV (697nm), respectively. (**b)** The image of monolayer $MoSe_2$ exfoliated onto the PDMS substrate. **(c)** The image of exfoliated monolayer $MoSe_2$ on diamond anvil. **(d)** Schematic diagram of the DAC for high-pressure PL, Raman and absorption experiments.

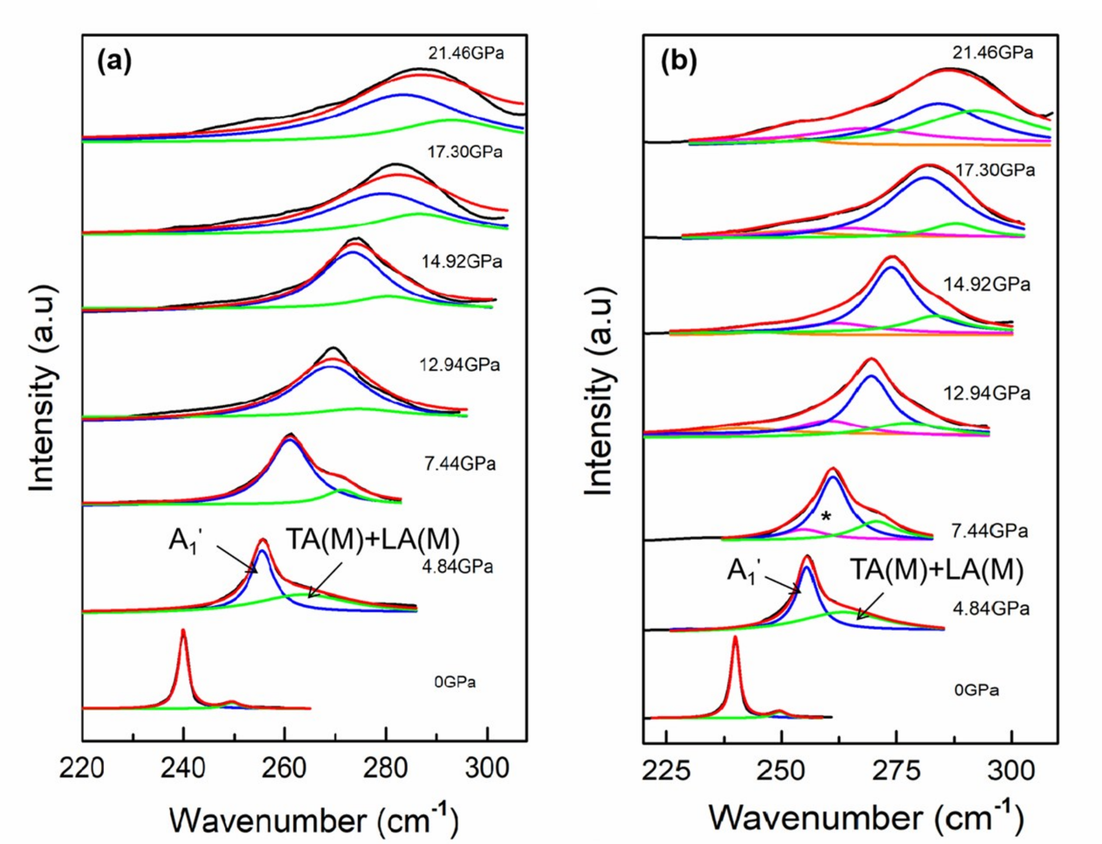

Figure S5. (a) A two-peak fit of the Raman spectra of compressed monolayer $MoSe_2$ at the range of 220-310 cm^{-1}. **(b)** A three-peak fit of the Raman spectra of compressed monolayer $MoSe_2$ at the range of 220-310 cm^{-1}.

The discussion about the adhered state in PVD-grown monolayer $MoSe_2$

According to the previous report (*Nature Materials*, **2011**, 10, 282-285.), the surface corrugations of graphene on SiO_2 are much larger as compared to that on hBN, (Figure 1) which can be attributed to the fact that, the graphene is well conformed to the substrate and the planar nature of hBN as compared with the amorphous SiO_2. That is, the 2D materials should be well conformed to the substrates. Furthermore, considering the fact that, our monolayer $MoSe_2$ is prepared by PVD method, the topographic image of monolayer $MoSe_2$ should be fully adhered to the substrate thanks to the deposition process.

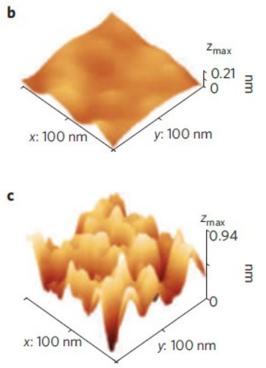

Figure 6 (b) STM topographic image of monolayer graphene on hBN showing the underlying surface corrugations. (c) STM topographic image of monolayer graphene on SiO_2 showing markedly increased corrugations. *Nature Materials*, **2011**, 10, 282-285.