Supplementary Materials

I. Experiment procedure

Our micro-ARPES measurements, performed at the Spectromicroscopy beamline at Elettra, followed the procedure described in detail in Ref. [14].

Fig. 1(a) in the manuscript shows the sample configuration in which the MoS_2 flakes are sitting on a Si substrate with cavities. The bright area of the PEEM image is from the cavity regions. The origin of the high emission signal is the geometric phase contrast described by Bauer *et al.* (see Ref. [18]). By tuning the voltage, constructive or destructive interference are developed due to the different path length of the electrons emitting from the terraces adjoining a step. Please find the details in Ref. [18].

II. Lattice relaxation

A. In-plane

To further address the intrinsic nature of the increase in the in-plane lattice constant of monolayer MoS_2**,** we show the comparison between bulk (yellow) and monolayer (pink) in Fig. S1 of supported monolayer MoS_2, which is suggestive of a similar increase as the suspended one.

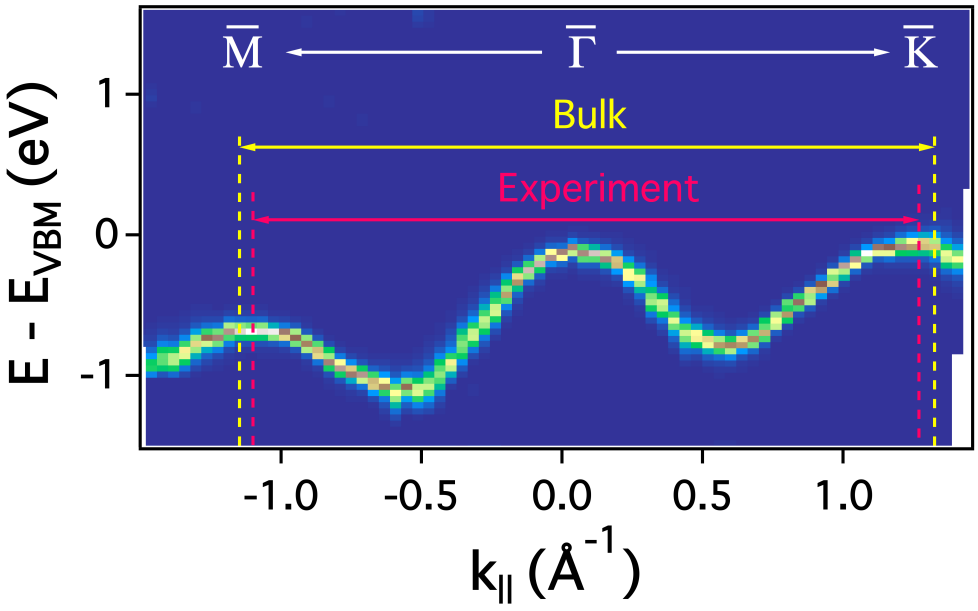

Fig. S1. 2D curvature plot of the uppermost valence band (UVB) of supported monolayer MoS_2 along high symmetric direction. Pink dashed lines mark the local maximum of the UVB extracted from ARPES measurement and the yellow dashed lines denote the positions of $\bar{M}$ and $\bar{K}$ using the lattice constant of bulk MoS_2.

B. Out-of-plane

Fig. S2 (a) shows our DFT calculated bands using the relaxed in-plane lattice constant (**a**=3.29Å) and the bulk interplane distance (**z**=1.586Å). Since the valence band at $\overline{K}$ is derived from in-plane orbitals including Mo $d_{x^2-y^2}$/ d_{xy} and S p_x/p_y (see Ref. [36]), the valence band maximum (VBM) is lowered in energy when using an expanded **a**. As a result, the VBM at $\overline{K}$ is lower than that at $\overline{\Gamma}$. This result indicates that we also need to expand the value of **z** in order to lower the VBM at $\overline{\Gamma}$, which is derived from the out-of-plane orbitals including Mo d_{z^2} and S p_z (See Ref. [36]). Fig. S2 (b) shows the DFT calculated bands using relaxed **a** and **z**, and we find that when increasing **z** by ~2%, the VBM at $\overline{K}$ and $\overline{\Gamma}$ are of the same height, and this calculated bands match the measured bands of suspended MoS_2 well.

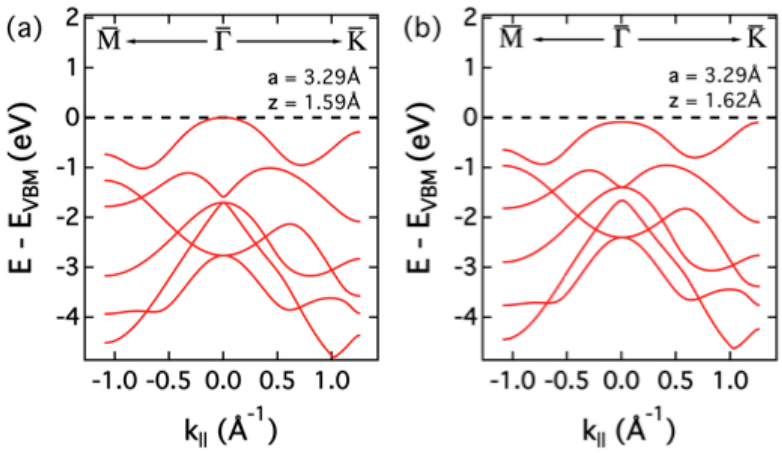

Fig. S2. (a) DFT calculated bands along high symmetry direction using relaxed in-plane lattice constant and bulk interplane distance. (b) DFT calculated bands along high symmetry direction using relaxed **a** and **z**.

We also compare our calculated bands using a relaxed **a** and **z** with those using the bulk lattice constants (full optimization gives a ~1% expansion of **a** and almost no change in **z**). As shown in Fig. S3 (a), using the VBM as a reference, all the bands using relaxed **a** and **z** (red solid curves) move upwards; as a result, the overall band width is smaller than that of the bands using bulk values (black dashed curves). Note that this upward shift is not a rigid offset. If we shrink the energy scale of the calculated bands using bulk lattice constants by ~11%, as shown in Fig. S3 (b), they match the bands using relaxed lattice constants very well. In other words, ~4% expansion of **a** and ~2% expansion of **z** result in ~11% shrinkage in energy scale for the calculated bands. Finally, to reiterate, our mention of the out of plane expansion simply notes that, to obtain a calculated band that is in reasonable agreement with measurement, a small change in the out of plane lattice constant is made. This is not a major finding of the paper, but will be of interest, we think, to the reader.

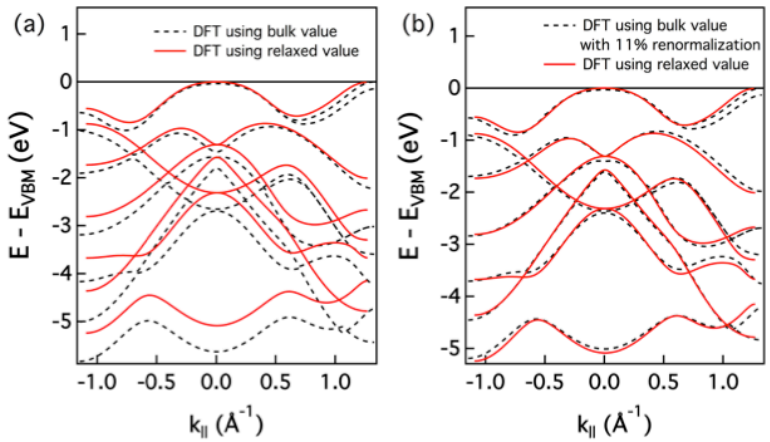

Fig. S3. DFT calculated bands of monolayer MoS_2 along high symmetry direction. Red solid curves are obtained by using the relaxed **a** and **z**. (a) The calculated bands using bulk lattice constant values (black dashed curves) are adapted from Ref. [25]. (b) We shrink the energy scale of adapted bands in (a) by 11%.