Supporting Information for

Direct growth of single- and few-layer MoS_2 on h-BN with preferred relative rotation angles

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Materials and Methods

CVD growth of single-layer and multi-layer MoS_2 on exfoliated h-BN

A two-zone furnace was used to grow single- and few-layer MoS_2 on exfoliated h-BN as shown in Figure 1a. H-BN flakes were exfoliated onto SiO_2/Si chips, which have a typical thermal oxide layer of 285 nm. Various thicknesses of h-BN flakes can be found on a chip after exfoliation. Then the chips with h-BN flakes were placed on top of the crucible with 200 mg powder-like MoO_3 nanoribbons. The MoO_3 crucible and another crucible with 2 g of S were then placed in a quartz tube. MoO_3 crucible was placed downstream in zone 2 of the CVD furnace, while S crucible was placed upstream in zone 1. At the beginning of the growth, zone 1 and 2 were both held at 105 °C for about 3 hours while flowing 200 sccm high-purity N_2 gas through the quartz tube. Then zone 2 was slowly increased to 500 °C and kept at 500 °C for 30 minutes to preheat MoO_3 precursor and the chips. Meanwhile zone 1 was kept at 105 °C. Then zone 2 was slowly increased to 800 °C and remained at 800 °C for 10 minutes while zone 1 was slowly increased to 400 °C and remained at 400 °C for 10 minutes at the same time. During this 10 minutes' reaction time, N_2 gas flow rate was changed to 10 sccm. After the reaction finished, the furnace cooled down slowly to room temperature.

TEM characterization and sample preparation of single-layer and multi-layer MoS_2 on exfoliated h-BN

TEM study on single-layer and multi-layer MoS_2 grown on exfoliated h-BN was performed on a FEI TitanX 60-300 microscope, which was operated under 60 keV to minimize the radiation damage to the samples. The Bruker windowless EDS detector installed on this TEM allows precise quantification of the ratio between different elements even for light elements such as B and N. TEM sample was prepared by "pick-up" method similar to reference 6 (in main text) but with polyethylene terephthalate (PET). Briefly, a small piece of PET was placed on the targeted flakes and then heated at 120 °C on a hotplate for 30 minutes. PET became soft from heating and then conformed onto the flakes on SiO_2/Si chips. After cooling down to room temperature, PET hardened again and can be removed by tweezers. PET with flakes was then placed on a quantifoil TEM grid and again heated up to 120°C on a hotplate for 30 minutes. Then PET/TEM grid was placed into dichloromethane and PET was dissolved leaving flakes on the TEM grid.

AFM characterization

AFM study was performed with an Innova ambient environment AFM under tapping mode, using a gold-coated Si tip.

Photoluminescence study

Photoluminescence was studied with a Renishaw inVia Raman microscope. The excitation laser has a wavelength of 488nm and a grating of 1800 1/mm is used. The photoluminescence spectra were taken with a range from 1.6 eV to 2.1 eV.

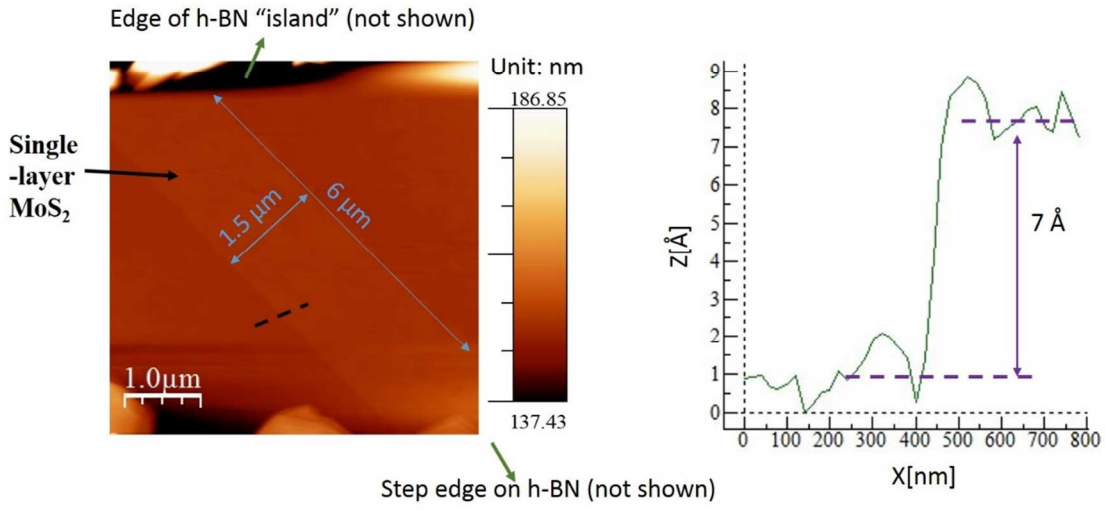

Fig. S1.

AFM image of a strip-like single-layer MoS_2 with a size 1.5 µm×6µm grown on h-BN. The line plot across the edge of flake shows that this is single layer.

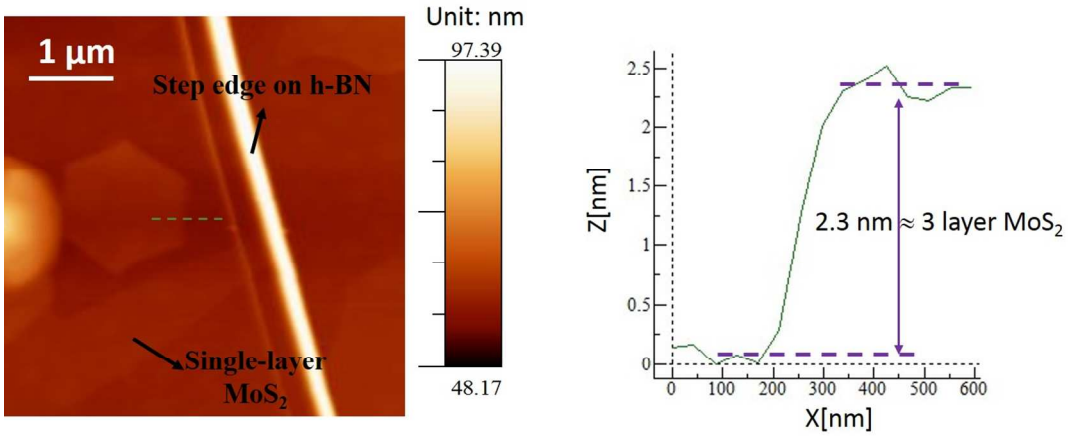

Fig. S2.

AFM image of smooth multi-layer MoS_2 with a size ~ 2 µm grown on hBN. The line plot across the edge of flake (dashed line) shows that this is 3 layer.

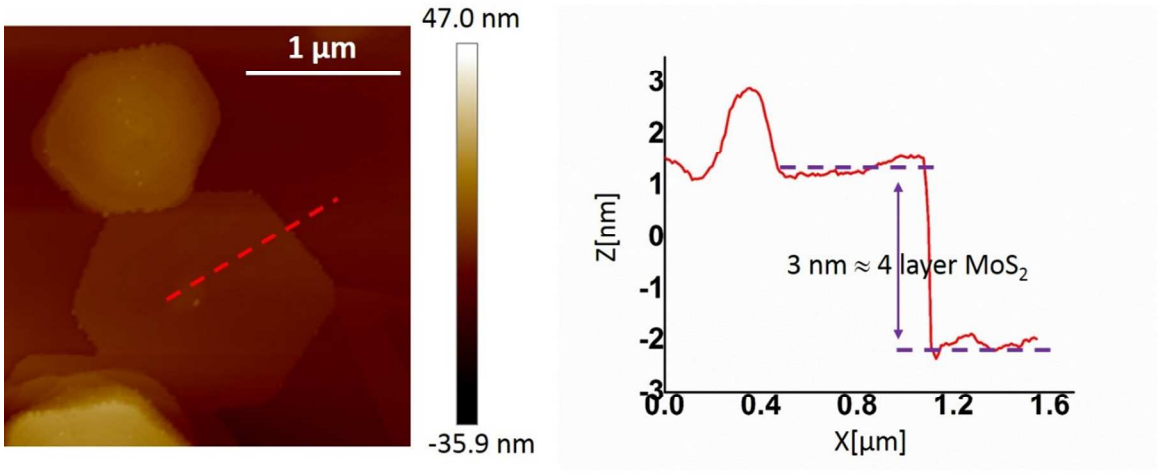

Fig. S3.

AFM image of a multi-layer MoS_2 with a protrusion in the center indicating the nucleation center. The line plot across the edge of flake (dashed line) shows that this is 4 layer and the nucleation center is 2 nm thicker than the flake.

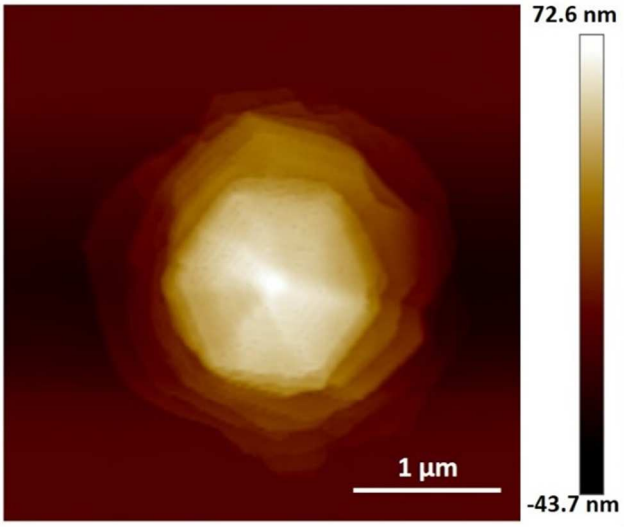

Fig. S4.
AFM image of one herringbone-structured MoS_2 island grown on h-BN.

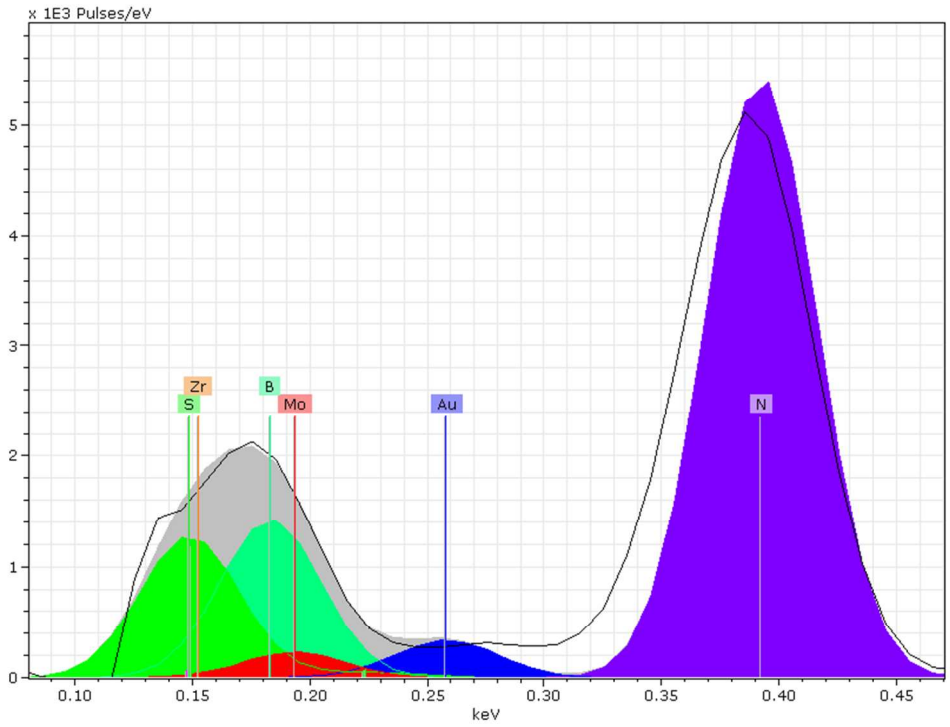

Fig. S5

Part of the EDS spectrum taken from MoS_2 grown on h-BN (in Figure 3), which includes B, N, Mo and S peaks located around 0.18 keV, 0.39keV, 0.19 keV and 0.15keV, respectively. As the concentration of B is calculated from this spectrum, some intensity from Mo peak can be included and this makes the elemental mapping of B takes the shape of Mo distribution as shown in Figure 3c and e.

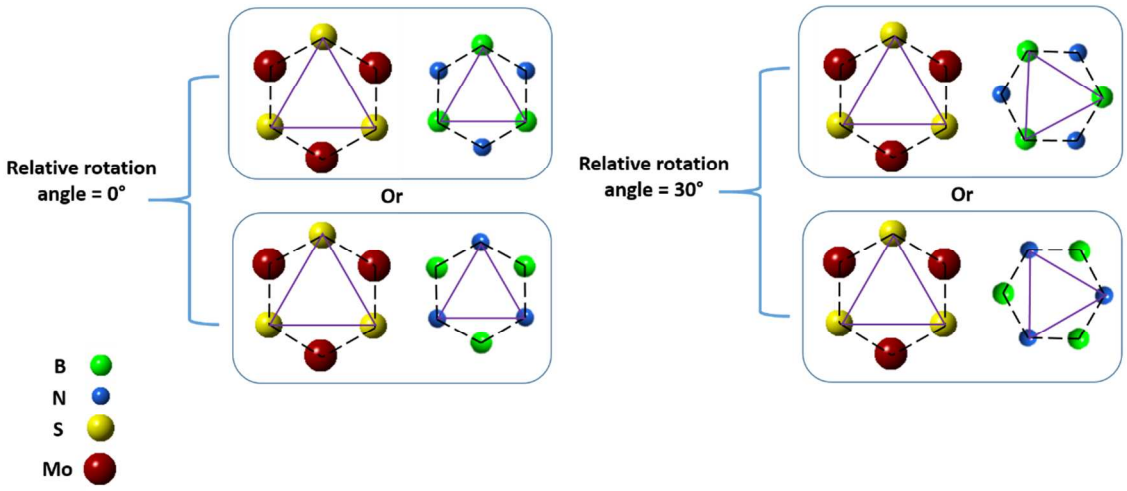

Fig. S6:

Schematics of sublattice orientations for MoS_2 and h-BN. The purple inner triangles connect similar atomic species and define a sublattice orientation. For the left schematics the sulfur directions are aligned with B (top) or N (bottom) directions. For the right schematics the sulfur directions are misaligned by 30^o from the B (top) or N (bottom) directions. If B and N become indistinguishable, then the sublattice misorientation angle θ is relevant only in the interval 0^o to 30^o, i.e. the reduced misorientation angle θ_{red} equals θ for $0^o<\theta<30^o$, and equals $60^o-\theta$ for $30^o<\theta<60^o$. Such a reduced representation is effectively used for Fig. 5. The symmetric full rotation angle histogram of Fig. S7 indicates there is no B or N distinguishability in the alignment, which supports such a reduced angle representation.

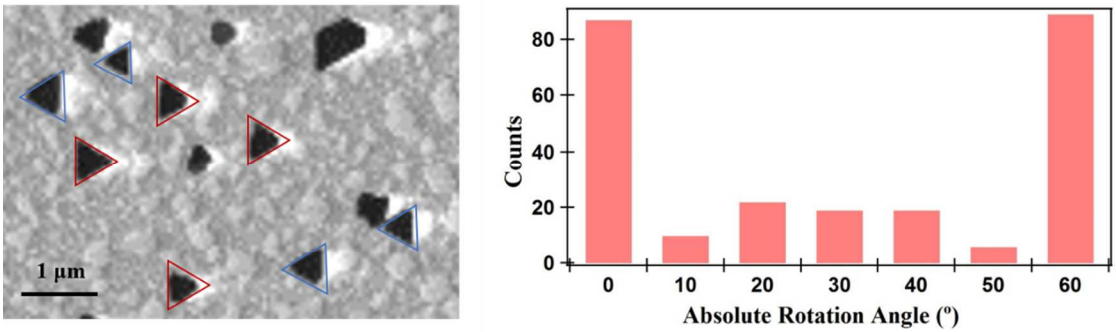

Fig. S7:

Left: SEM image showing small single-layer crystalline MoS_2 triangular islands grown on single-crystal h-BN. Blue and red triangles are guides to the eye for MoS_2 orientations; they are rotated 60° with respect to each other. These triangles identify the two most dominant orientations of MoS_2 islands. By complementary electron diffraction in TEM, we determine that the two dominant orientations are 0° and 60° absolute, relative to the h-BN substrate. We assign blue triangles 0° and red triangles 60°. The error for angle measurement here is within 4°. Right: Absolute rotation angle distribution based on SEM image with 264 MoS_2 near-identical islands grown on the same h-BN flake. The distribution is bimodal and symmetric, with dominant peaks centered at 0° and 60°.