Supplementary Information for

Three-dimensional Spirals of Atomic Layered MoS_2

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S1: CVD Synthesis of MoS_2 spiral.

We grew MoS_2 spirals by CVD using MoO_3 and S as precursors. The substrate was loaded into the 1-inch CVD furnace and placed face-down above one crucible containing 5.0~7.5 mg of MoO_3 (≥99.5%, sigma-Aldrich) with another crucible containing 140 mg of S (≥99.5%, sigma-Aldrich). In order to increase the possibility to get spirals, S was loaded in the high temperature region close to MoO_3. In contrast, for monolayer growth, the substrate was loaded above one crucible containing 20 mg of MoO_3 together with another crucible containing 6.5~7.0 mg of S and the distance between S and MoO_3 is about 12 cm. MoS_2 spirals could be directly grown on diverse substrates (Fig. S1) and the optimized parameters have been summarized in Table 1.

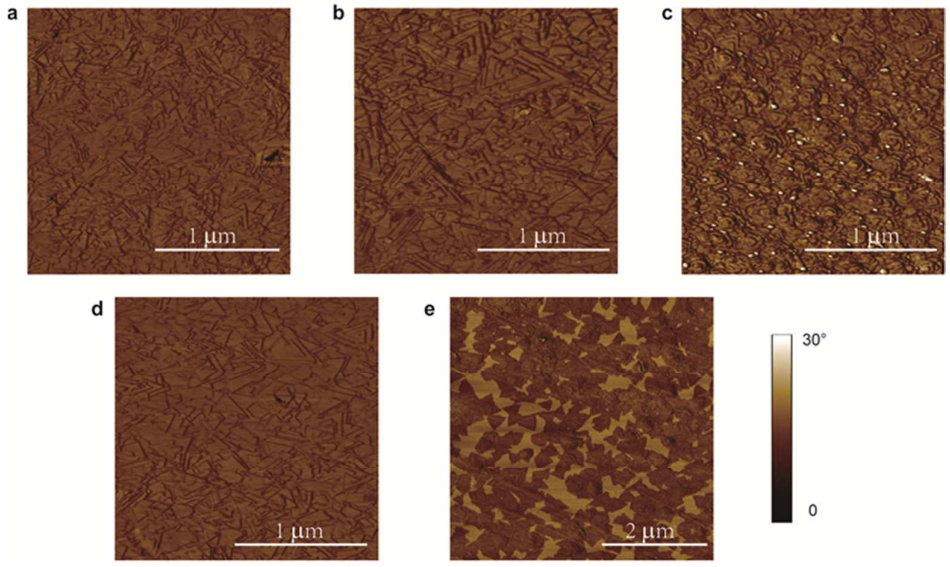

Fig. S1 a-e, CVD synthesis of MoS_2 spiral on varied substrates (a) Si, (b) mica, (c) SiO_2, (d) TiO_2 and (e) fused silica.

Table S1 Detail synthesis parameters for MoS_2 spiral

Substrate	Source temperature (℃)	Mass ratio between MoO_3 and S	Carrier gas flow rate (sccm)	Growth temperature (℃)	Growth time (min)	Pressure (Torr)	Domain size
Si	MoO_3: 650-700 S: 600-650	MoO_3/S 5.0~7.5 mg/140 mg	ultrahigh-purity N_2 10-15 sccm	700	5-20	ambient pressure	300-350 nm
SiNWs							
TiO_2							
SiO_2							
Fused silica							
Mica	MoO_3: 600-650 S: 550-600			650			
Fused silica	MoO_3: 650-700 S: 450-500	MoO_3/S 20 mg/6.5-7.0 mg		700			> 10 μm

S2: Structural characterization of spirals using XRD.

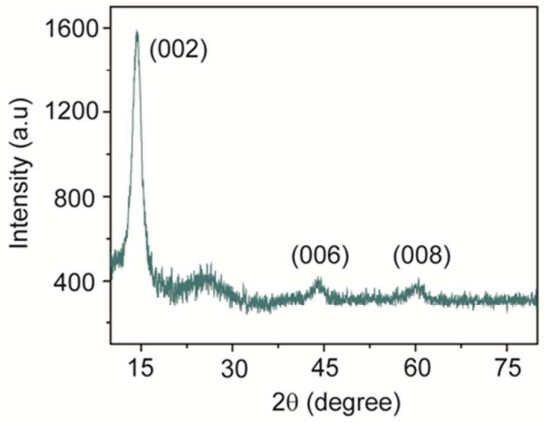

Fig. S2 XRD patterns of MoS_2 spirals. A pronounced diffraction peak locates at 14.4°, which corresponds to (002) crystal planes in Z direction. The wide peak signal between 20° and 30° originates from the fused silica substrate.

S3: Morphology characterization of one large MoS_2 spiral by AFM.

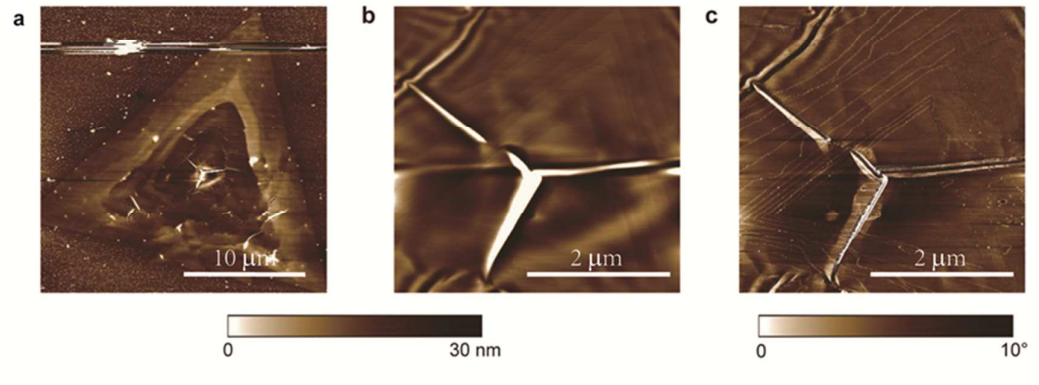

Fig. S3 a-c, Low- (a, height signal) and high- (b, height signal; c, phase signal) magnification AFM images of one large MoS_2 spiral with edge length ~20 μm. The wrinkles formed in the center were attributed to the topological deformation resulted in the process of temperature decreasing.

S4: Composition study of spirals by XPS.

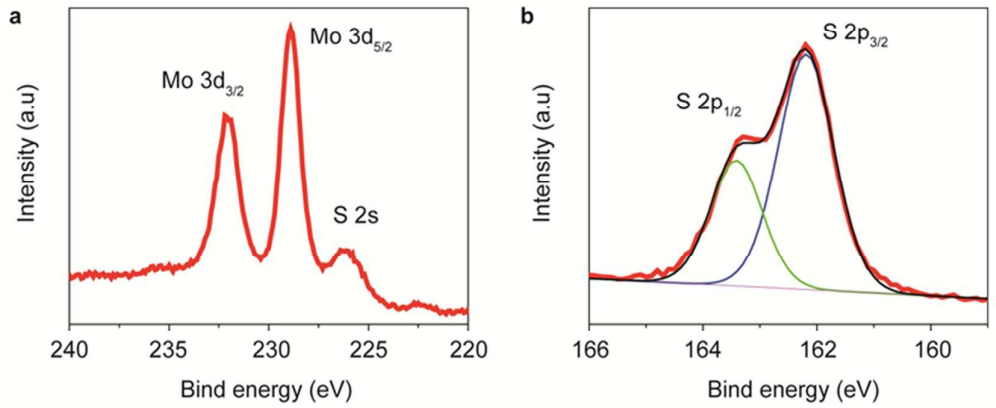

Fig. S4 a-b, High-resolution XPS spectra of Mo 3d, S 2s (a) and S 2p (b) peaks.

S5: Stability of different configurations in AA-stacked bilayer MoS_2.

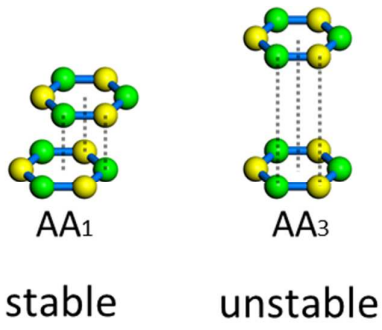

Fig. S5 Configuration of highly symmetric structures of AA stacked bilayer MoS_2. Local-density approximation (LDA) calculation shows that AA_1 is energetically stable, while AA_3 is unstable.

S6: LDA Band structure of AA-stacked MoS_2.

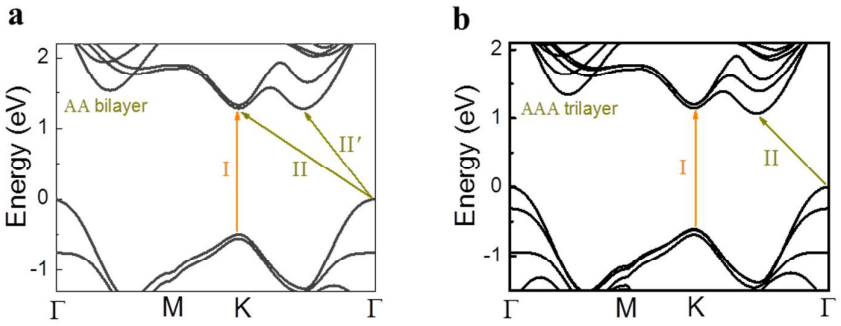

Fig. S6 a-b, LDA band structure of AA-stacked bilayer (a) and trilayer (b) MoS_2. The transition I corresponds to the main PL peak around 1.8 eV in the PL spectra, while transition II (or II') corresponds to the indirect photoemission. A supercell arrangement was used in the calculation, with the vacuum thickness set at 10Å to avoid interactions between the MoS_2 bilayer/trilayer and its periodic images. We employ norm-conserving pseudo-potentials with a plane-wave energy cutoff of 140 Ry. The structures were fully relaxed until the force on each atom is smaller than 0.01 eV/Å. Spin-orbit coupling was not included in our calculations.