

Anisotropic strain in epitaxial single-layer molybdenum disulfide on Ag(110).

Supplementary Information

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S1 Line shape of core levels

TABLE I. Line shape parameters for the fits of the Mo 3d and S 2p spectra acquired at 360 eV and 260 eV photon energy, respectively, for 0.75 ML of SL MoS₂ grown on Ag(110). L is the Lorentzian width, α is the asymmetry parameter and G is the Gaussian width.

Mo 3d	L (eV)	α	G (eV)	BE (eV)
3d _{5/2}	0.2	0.08	0.35	229.33
3d _{3/2}	0.38	0.08	0.35	232.48
S 2p	L (eV)	α	G (eV)	BE (eV)
2p _{3/2} top	0.17	0.06	0.21	162.27
2p _{3/2} bottom	0.17	0.06	0.21	162.51
2p _{1/2} top	0.18	0.06	0.21	163.47
2p _{1/2} bottom	0.18	0.06	0.21	163.71

S2 Direct MoS₂ growth on substrate at high temperature

An attempt to grow MoS₂ was done by following the pathway described in earlier reports[1, 2] for the growth of transition-metal dichalcogenides on Au(111). Mo was deposited in H₂S background pressure of

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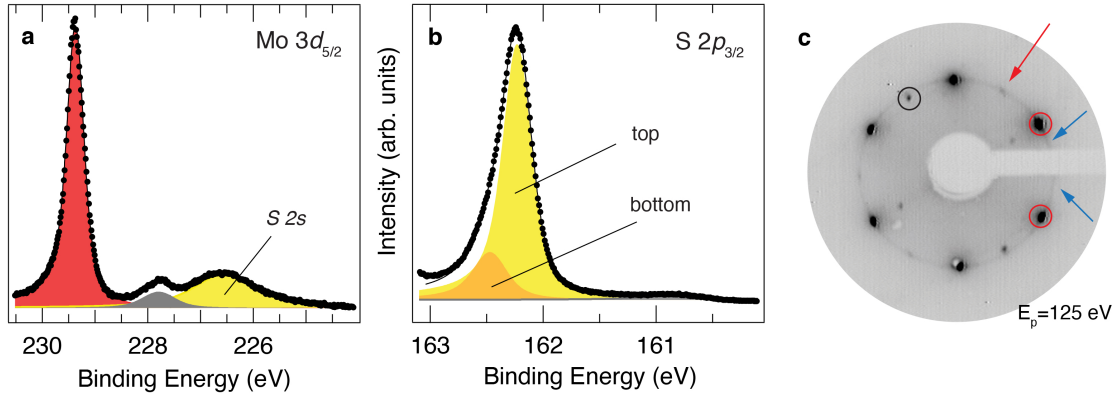


FIG. S1. High-resolution XPS spectra of the (a) Mo $3d_{5/2}$ and (b) S $2p_{3/2}$ core levels for direct growth of SL MoS₂ on Ag(110). (c) LEED pattern at $E_p = 125$ eV for a coverage of 0.7 ML of MoS₂. The principal spot corresponding to MoS₂ is indicated by the red circle, while the black circles mark the spots from Ag(110) surface. The red arrow marks the spot due to the MoS₂ domains rotated by 30° with respect to the main MoS₂ domain orientation, while the blue arrows indicate the streaks due to the randomly oriented domains.

5×10^{-7} mbar on the Ag(110) sample kept always at 800 K. The deposition was continued up to the final MoS₂ coverage of ca. 0.7 ML, with a monolayer (ML) representing one complete layer of MoS₂ on Ag(110), amounting to Mo surface density of 1.15×10^{15} atoms/cm² for the MoS₂ lattice parameter of 3.15 Å. The desired coverage of the MoS₂ single layer was obtained by evaporating controlled amounts of Mo. Assuming a layer-by-layer growth, an assumption corroborated by the absence of double layers detected in ARPES measurements, we measure the total intensity of the Ag $3d_{5/2}$ spectrum ($h\nu = 420$ eV that corresponds to a kinetic energy KE=52 eV,) before and after the growth of MoS₂. The intensity change is then compared with the intensity variation of the Au $4f_{7/2}$ spectrum measured at $h\nu=136$ eV (KE=52 eV) between the clean Au(111) and after the growth of SL MoS₂/Au(111). In the latter case, the MoS₂ coverage can be determined quite precisely from the decrement of the clean surface component of the Au $4f_{7/2}$ core level, as shown by Bana *et al.* [1]. In both cases, the kinetic energy of the measured photoelectrons is ca. 52 eV and the same experimental conditions are used. Therefore, it is reasonable to assume that the screening of the signal from the substrate due to the MoS₂ ad-layer should be of the same magnitude for the two substrates, owing to the unchanged inelastic mean free path of the photoelectrons having the same kinetic energy.

Figure S1 shows the high resolution core-level x-ray photoelectron spectra (XPS) of the Mo $3d_{5/2}$ and the S $2p_{3/2}$ core levels, measured after the growth. We observed the Mo $3d_{5/2}$ component at binding energy (BE)=229.33 eV (Figure S1a), corresponding to MoS₂. The grey component was associated to Mo atoms that did not react with S to form MoS₂ [3]. Moreover, we observed that S $2p_{3/2}$ has two components at

BE=162.27 eV and 162.51 eV) core levels (Figure S1(b)), assigned to S atoms in the top and bottom layer of SL MoS₂, respectively.

Unlike the data shown in the main text, the LEED pattern obtained with this growth protocol (Figure S1(c)) showed the presence of ring-like streaky features, marked with blue arrows, and of additional spots azimuthally rotated of ca. 30° with respect to the main MoS₂ spots (red circles). The ring-like structure indicates the formation of azimuthally randomly rotated domains of MoS₂, while the diffraction spots indicated by the red arrow correspond to MoS₂ domains rotated by 30° with respect to the main orientation of SL MoS₂, whose presence may be indicative of structural defects such as grain boundaries, which may eventually degrade the properties of SL MoS₂.

S3 Two-step MoS₂ epitaxial growth

As mentioned in the main text, the formation of an ordered MoS₂ single layer with just two mirror orientations was obtained with a two-step procedure: 1. We dosed a small amount of Mo (0.07 ML) in H₂S background pressure with the sample at RT, followed by an annealing up to 800 K in H₂S. 2. We directly dosed Mo on the substrate kept at 800 K, still in H₂S background pressure, up to the final coverage of about 0.75 ML.

In Figure S2 we report the evolution of the Mo 3d during the annealing in H₂S atmosphere that follows the initial deposition of Mo at room temperature of step (1). The left panel shows a 2D image reporting the time evolution of the Mo 3d spectra acquired along the annealing, by means of fast-XPS. The green continuous line on the right side shows the sample temperature, which was initially risen with a ramp of 1 K/s and then kept constant at 800 K. From this graph, we can immediately observe a distinct change in the binding energy of the Mo 3d peak when the temperature goes above 700 K. This modification is accounting for the conversion from partially sulfided species produced at RT to an ordered layer of MoS₂ [3].

In Figure S3 we show the evolution of the Mo 3d_{5/2} while dosing Mo in H₂S back-pressure directly after step #1 on the substrate kept at 820 K. We reached a final coverage of ca. 0.75 ML. We observed that only species with a BE compatible with single-layer MoS₂ were present on the surface at the end of the growth. We speculate that the step #1 of the growth process is needed to create MoS₂ islands that acts as seeds guiding the ordered growth of step #2.

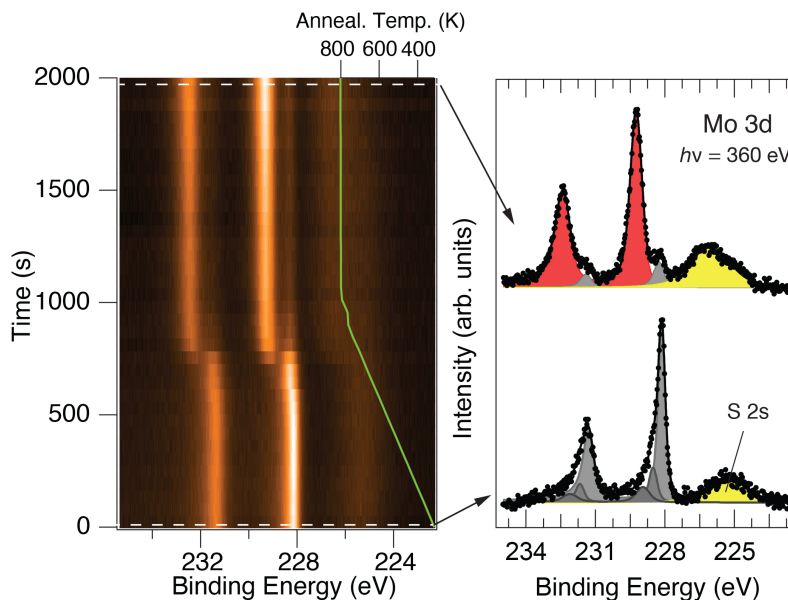


FIG. S2. Evolution of the Mo 3d core level during the annealing in H_2S atmosphere, measured by means of fast-XPS. Two Mo 3d spectra, and their deconvoluted components, selected at the beginning and at the end of the annealing ramp are shown on the right-end side of the figure. The photon energy employed was $h\nu = 360$ eV.

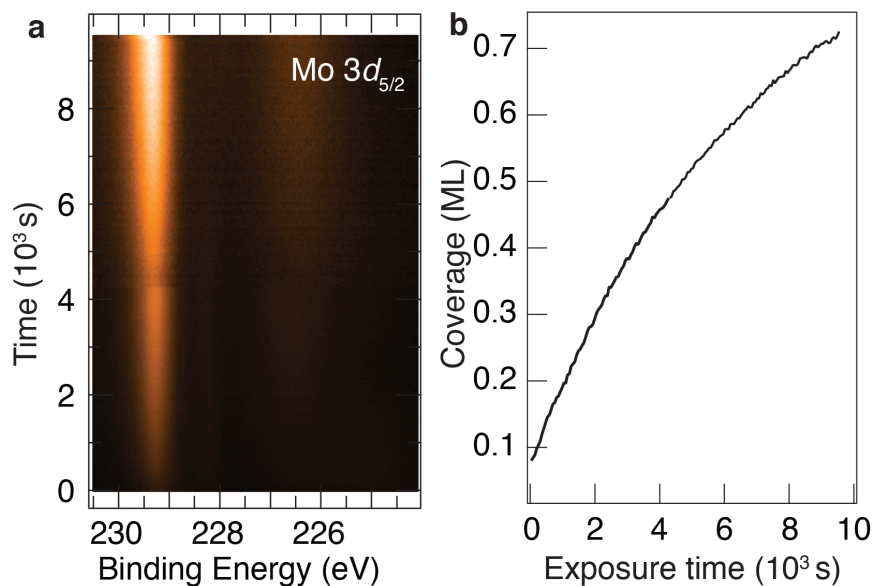


FIG. S3. (a) Evolution of the Mo $3d_{5/2}$ core level during step #2 of the MoS_2 growth. (b) Evolution of the MoS_2 coverage, proportional to the intensity of the Mo $3d_{5/2}$ peak (grey), as a function of deposition time.

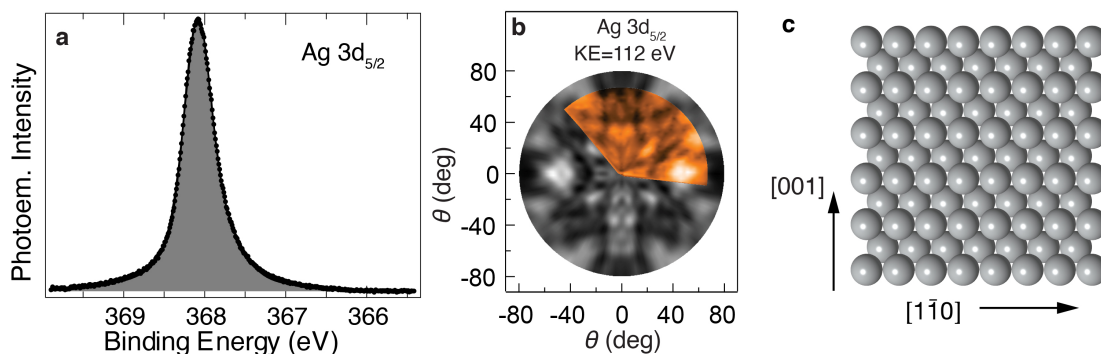


FIG. S4. (a) Ag $3d_{5/2}$ core level acquired at normal electron emission ($h\nu=480$ eV). (b) XPD pattern for Ag $3d_{5/2}$, measured with a photon energy of 480 eV (KE=112 eV). (c) Ball model for the Ag(110) clean surface associated with the pattern on the left.

S4 Orientation of the Ag substrate

An evidence supporting that the MoS₂ layer is aligned with one of the two crystallographic directions of the Ag(110) substrate, as already observed in LEED, can be obtained by performing XPD measurements to determine the orientation of the substrate, namely acquiring the Ag $3d_{5/2}$ core level.

Figure S4 shows (a) the high-resolution Ag $3d_{5/2}$ core level acquired at normal electron emission and (b) the associated XPD pattern (color) for the clean Ag(110) substrate before the growth of MoS₂. The pattern is two-fold symmetric, as expected from the rectangular lattice of the (110) face of a *fcc* crystal. The simulated XPD pattern (grey) was obtained via multiple-scattering calculations on a 3 layer slab of Ag, at KE = 112 eV, with the atoms in all three layers emitting simultaneously because no surface shifted core level component could be detected in the Ag $3d_{5/2}$ spectrum.

The comparison between the simulation and the experimental XPD pattern was carried out following an R-factor analysis [4] allowed the individuation of the orientation of the Ag(110) crystal, which is that one shown in Figure S4(c)

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