ESRF	**Experiment title: Structure and strain of a monolayer molybdenum disulfide, a two-dimensional crystalline membrane with out-of-plane degrees of freedom**	**Experiment number**: 32-03-735
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Monolayer MoS_2 flakes have been successfully grown on a Au(111) surface in the ultra-high vacuum environment of the INS2 instrument at the BM32-CRG/BM32beamline at ESRF by a combined use of molecular beam epitaxy (MBE) and chemical vapor deposition (CVD), while the process was monitored in real time by grazing incidence surface X-ray diffraction (GIXRD). Up to 0.6ML of MoS_2 have been grown. A long range order coincidence superstructure (moiré) concordantly oriented with the Au substrate has been quantitatively characterized via Surface X-Ray Diffraction (SXRD).

THE GROWTH

During the experiment we used a set of Au single crystal samples cut along the (111) face and showing the typical (22 × √3) herringbone reconstruction. The growths were performed according to the method already established in Aarhus laboratories [1], by cycling Mo deposition and annealing treatments in H_2S gas atmosphere. More specifically, Mo was deposited by an e-beam evaporator at the very slow calibrated rate of 0.02ML/min at room temperature and 5×10^{-6}mbar of H_2S pressure for 8min, in order to reach the nominal coverage of 0.2ML. The gas was injected in the growth chamber via the new gas line recently installed at BM32. After that, the sample was heated up to 650°C for 30min and then cooled down to room temperature without stopping the H_2S injection. This process was cycled until the limit target of 0.8ML was reached. According to [1], this partial coverage is considered as the step at which the by layer starts seeding.

RESULTS

Through GIXRD performed during each stage of the process, we observed that, at least at room temperature, the H_2S atmosphere where the sample is steadily maintained does not affect the pristine herringbone reconstruction of Au (Fig. 1). This is in contrast with other observations at room temperature reported in literature [2]. The reconstruction is lifted instead when Mo (or Mo-S) clusters crystallize on the surface: Fig. 1 shows in fact that the typical peaks for herringbone reconstruction surrounding the Au crystal truncation rod (CTR) degenerate as soon as Mo is deposited on top.

We devoted a first sample where 0.3ML of MoS_2 were deposited on top to the study of the MoS_2 behavior under annealing. Evolution of the (100) MoS_2 peak in function of the increasing temperature is visible in Fig. 2. As expected, the peak becomes sharper and higher in intensity as the temperature grows, which is a qualitative evidence of the domain size growth and the lattice constant refining. Above 650°C the spectrum does not improve significantly anymore, whereas the material starts degrading between 770 and 800°C. We used this temperature as reference to optimize the following cycles of growth.

Subsequently, we prepared two samples respectively of 0.6 and 0.8 ML of partial coverage, for ex-situ STM measurements (performed later by some of us at Inst. Néel in Grenoble) and quantitative diffraction *in-situ* characterization. Remarkably, the two combined approaches reveal the formation of single layer thick MoS_2 flakes characterized by a bulk-like lattice constant (3.15Å), and, besides that, a moiré superstructure that originates from the coincidence between MoS_2 and Au lattices. Reciprocal space maps together with radial scans along the two highest symmetry in-plane directions around the substrate Bragg peaks and CTRs clearly display the above mentioned results (Fig. 3). Satellite superstructure peaks emerging between MoS_2 and Au in the radial scan, as well as the remainders in the maps, form a hexagonal pattern perfectly aligned with both substrate and overlayer crystallographic directions and centered around Au reflections (this observation disproves the previous one reported in [3]). The periodicity of the peaks connected to the superstructure provides an estimation of the supercell size which has been calculated of 33.3 Å, in good agreement with [1]. The rocking scans collected for all the measurable MoS_2 and superstructure peaks reveal low mosaic spread for the MoS_2 grown islands (around 0.5°), which is a demonstration of the good epitaxial alignment with respect to the Au substrate. Lastly, quantitative out-of-plane rod and crystal truncation rod (including the specular (00ℓ) CTR) measurements have been performed during the allocated beamtime, whose deep quantitative analysis is still in progress: rods display quite strong oscillations (Fig. 4) from which it is possible to deduce precisely the atomic structure of the two-dimensional layer and its interface with the substrate.

REFERENCES

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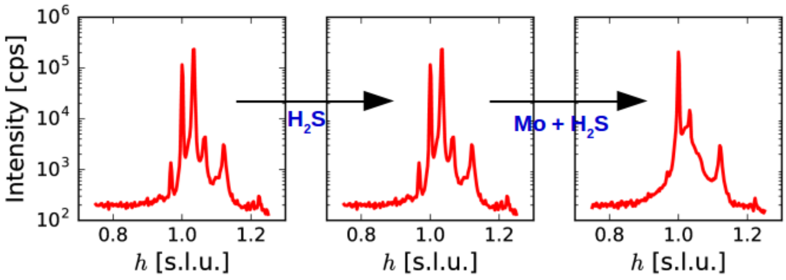

Fig1: effects of H_2S exposure and Mo deposition at room Temperaure on the Au(111) herringbone reconstructed surface. In thei mage only the spectral range between 0.7 and 1.3 Au surface lattice units is shown.

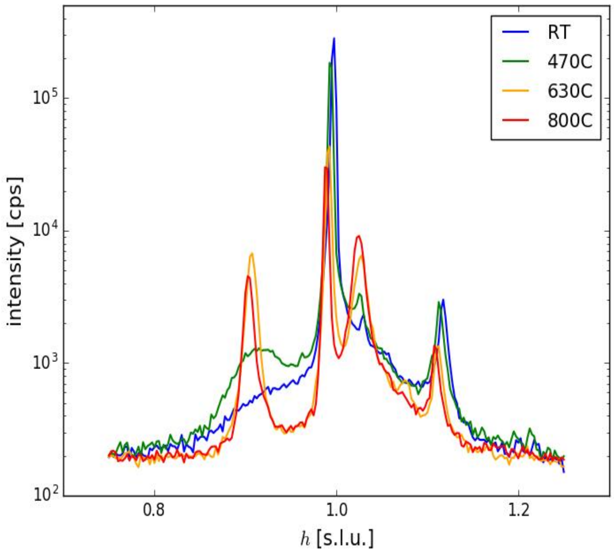

Fig2: Evolution in function of the annealing temperature (and in H_2S atmosphere) of the diffraction pattern. The MoS2 peak, marked in the figure by the blue arrow, becomes higher and sharper until 800°C, when degradation occurs.

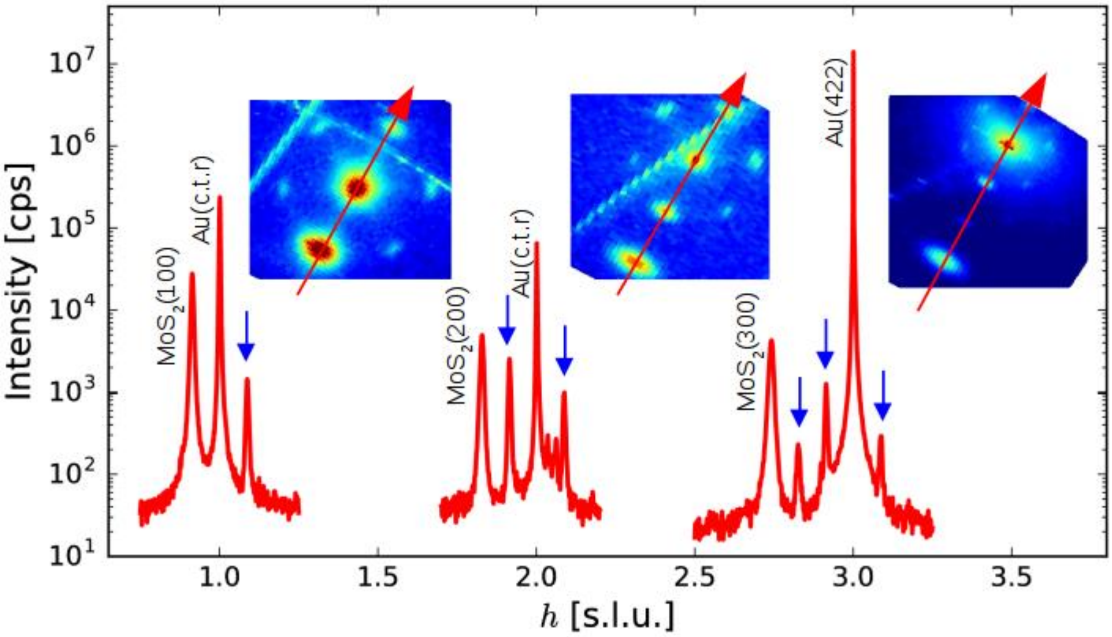

Fig3: Reciprocal space maps together with radial scans along the two highest symmetry in-plane directions around the substrate Bragg peaks and CTRs

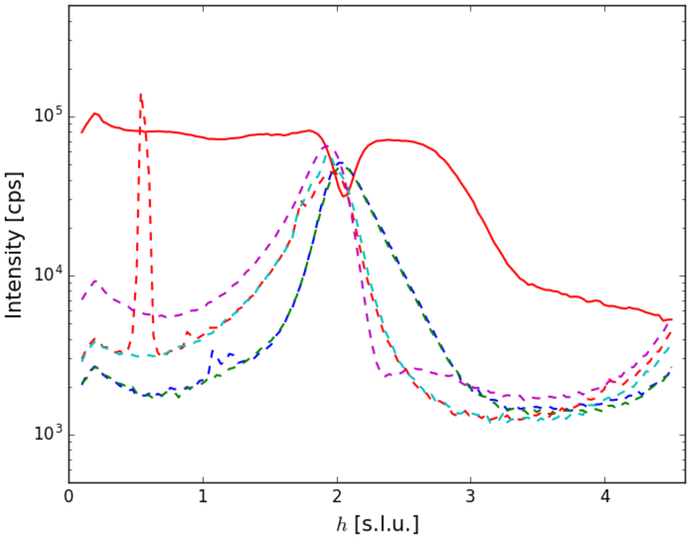

Fig4: MoS_2 (10*l*) rod (continuous line) and superstructure rods (dashed lines) measured around the first order CTR