	Experiment title: Formation and quantitative structure determination of $PtSe_2$ obtained by selenization of Pt(111)	**Experiment number**: 32-03-741
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$PtSe_2$ growth by selenization of Pt(111)

SL $PtSe_2$ was grown in UHV by a two-step process based on the direct selenization of a Pt(111) crystal surface. Firstly, several nm of amorphous selenium were deposited on top of Pt(111) until its reflection high-energy electron diffraction (RHEED) pattern disappears. Secondly, the sample was annealed from room temperature (RT) to 370°C. During the annealing, the platinum atoms bond to selenium at the interface forming a $PtSe_2$ monolayer, whereas the excess selenium desorbs from the surface.

A highly ordered $PtSe_2$/Pt(111) coincidence lattice

Figure 1a shows an 80° in-plane sector of the reciprocal space mapped with GIXRD. This map displays the successive Brillouin zones of Pt(111) (orange hexagons) having sharp Pt(111) peaks at their centres, with additional sharp peaks corresponding exactly to a (4×4) superstructure relative to the Pt(111) lattice. In Figure 1b, two high-resolution linear scans measure the scattered intensity along the radial direction (*h*00) (highlighted by the red frame in Figure 1a) before (blue) and after (orange) the selenization. In the latter case, the Pt peaks are pointed out by blue filled circles. $PtSe_2$ peaks, labelled with orange filled circles, are characterized by a periodic reciprocal lattice vector which is three quarters the Pt(111) surface one, consistent with previous reports. The observed peaks, marked by orange diamonds, are all perfectly aligned and equi-spaced. They are the signature of a surface hexagonal superlattice associated to the coincidence lattice between (3×3) $PtSe_2$ and (4×4) Pt(111) surface ones, with a lattice parameter of 11.1 Å. The reciprocal space is indexed using this large hexagonal surface superstructure unit cell, so that the Pt(111) and $PtSe_2$ Bragg peaks are at *h* and *k* surface reciprocal lattice units (s.r.l.u.) multiple of 4 and 3 respectively. The deduced $PtSe_2$ surface lattice constant, of (3.700±0.002) Å, is slightly smaller (0.7 %) than the relaxed bulk value of 3.724 Å. Remarkably, the superstructure peaks show strong intensities, similar or even exceeding those of the nearby $PtSe_2$ peaks (compare *e.g.* peaks at $h = 7$ with $h = 6$).

Periodic lattice distortions in SL $PtSe_2$ on Pt(111)

The compressive global strain and the almost perfect angular alignment are signatures of a strong interaction between $PtSe_2$ and Pt(111). Another signature are the strong intensities of the superstructure diffraction peaks, which point to sizeable lattice distortions, as well as the large intensity variations along the diffraction rods (Fig. 1c), which indicate that about 3 Pt(111) layers of the substrate undergo strong displacements due to this superstructure. We are presently finalizing the analysis of the in-plane projected structure. Details about the model can be found in a very soon to be submitted article (1). Fig. 1d shows the comparison between the experimental and theoretical in-plane diffraction diagram, which reveals a good agreement. The structural model comprises (3×3) unit cells of the SL $PtSe_2$ and (4×4) unit cells of the three topmost Pt(111) surface layers. Figure 1e shows the refined distorted atomic model (only 2 Pt planes shown). Within the coincidence supercells (red

frames), three main areas can be distinguished, referring to Pt atoms of the $PtSe_2$ layer sitting on the three-fold positions, on-top, fcc and hcp sites, labelled A, B and C, respectively. The other Pt atoms in the dichalcogenide layer (left side, in blue) shift along the supercell edge and mirror axes (continuous and dashed red lines) by displacements in the range of 0.40-0.55 Å towards the atoms A at the supercell vertices. This results in a contraction of the nearest-neighbour Pt-Pt distances around A. Similarly, some Se atoms, both in the top and bottom TMDC layers (left side, in green and yellow respectively) and Pt atoms of the first substrate layer (right side, in light grey) are displaced by similar extents, so that the atomic density is reduced (increased) around C (A). In the second Pt substrate layer, the displacement amplitudes are smaller, between 0.02 Å and 0.03 Å. Our in-plane model merely describes the atomic displacements occurring parallel to the surface plane. However, contraction and expansion of the distances between atoms parallel to the surface in a 2D material are likely to be accompanied by a wavelike corrugation. We expect here a similar corrugation within the supercell; the atoms around A (where projected lateral distances are smaller) being higher than those around B, themselves higher than around C, which is consistent with STM images reported in a previous work. In summary, the 1T structure of the TMDC material as well as that of the topmost substrate layers are perturbed by elastic deformations propagating at the interface. This does not correspond to a pure vdW epitaxy, where the 2D overlayer grows almost strain-free due to very weak interaction with its substrate. It rather implies a strong coupling between $PtSe_2$ and the Pt(111) surface.

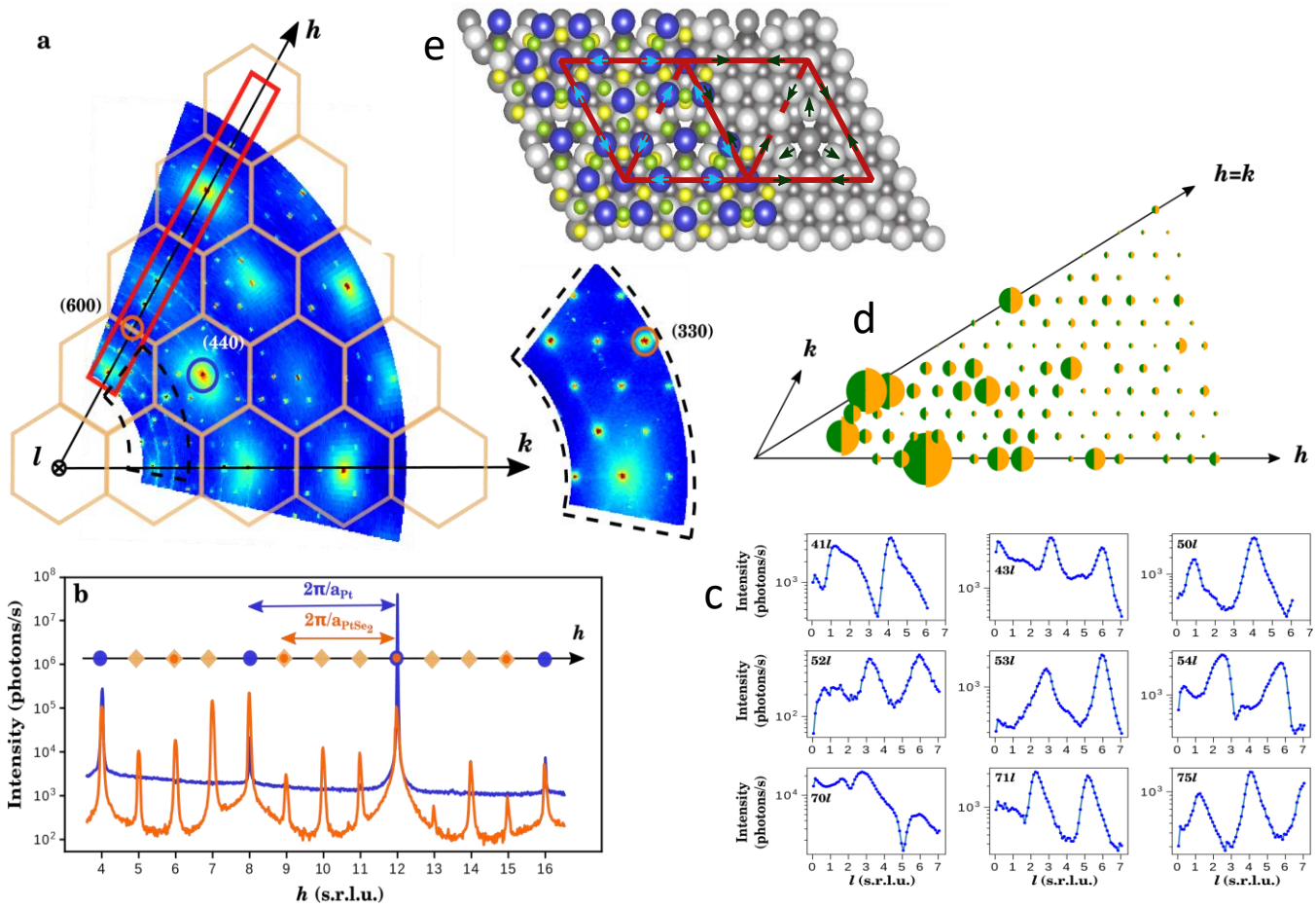

Figure 1: a) In-plane reciprocal space map measured by GIXRD on 1 ML $PtSe_2$ on Pt111. b) Radial scan along the h direction in superstructure reciprocal lattice units. c) Some rods (intensity as a function of perpendicular reduced coordinate ℓ in units of $2\pi/c$, c being the Pt(111) hexagonal unit cell lattice parameter of 6.8 Å, i.e. the distance between three Pt111 planes in the bulk). d) Comparison between experimental and theroretical superstructure in-plane diffraction intensities. e) Sketch of the projected structure with in-plane atomic motions.

References

(1) Sant et al., *Synthesis of epitaxial monolayer Janus SPtSe*, npj 2D Materials and Applications (2020) 4:41 ; https://doi.org/10.1038/s41699-020-00175-z and R. Sant, PhD-Thesis, Univ. Gre. Alpes, 2019.