Information efficient 4D-STEM for quantitative insight into structure-property relationships in large-scale $PtSe_2$ films

Hanako Okuno[1], **Samuel Dechamps[1]**, **Alexandre Camathy[1]**, **Djordje Dosenovic[1]**, **Eva Desgué[2]**, **Pierre Legagneux[2]**, **Nicola Vigano[1]**

Univ. Grenoble Alpes, CEA-Grenoble, IRIG-MEM, 38000 Grenoble

Thales R & T,

Hanako.okuno@cea.fr

Two-dimensional (2D) layered materials have attracted much attention for future (opto)electronics due to their extraordinary properties. However, their controlled synthesis remains a key challenge, as intrinsic atomic defects can be unavoidable sources of discrepancies between experimental measured properties and theoretically predicted ones. In recent years, atomistic studies based on high-resolution analytical techniques (e.g., STEM, STM), combined with DFT calculations, have greatly advanced the fundamental understanding of atomic defects in 2D materials and their influence on local electronic properties. However, such insights need to be complemented by quantitative analyses clarifying the distribution of defects for understanding their real properties at large scale.

In this work, we demonstrate a new characterization approach based on large scale 4D-STEM analysis, combined with atomic-resolution STEM and DFT calculations, correlating structural and electrical properties in MBE-grown 15ML semi-metallic $PtSe_2$ films [1]. Atomic resolution STEM analyses are performed to classify various growth-related defects, and which identified in-plane and out-of-plane domain junctions as "tuning-knob" for large scale sheet conductance. Usually, the in-plane domain junctions are considered the dominant defects, affecting sheet conductance by acting as tunnelling barriers for charge carriers. Additionally, our DFT calculations revealed that twisting interface between out-of-plane domains influence the local intrinsic electronic properties of multilayer $PtSe_2$ films, due to their strong interlayer coupling. 4D-STEM is performed to acquire datasets containing diffraction patterns (~200000 images) recorded at each probe position scanning over micron-scale areas. A robust numerical framework constructs a high throughput database associating each real-space position with accurately measured crystal orientations (θ) [2], as well as their intensity and multiplicity. These quantitative databases are subsequently used for visualization for selected structural parameters, providing detailed information on both in-plane and out-of-plane domain distributions, and thereby enabling the localization and quantification of domain junctions and interfaces. Finally, a clear correlation between the degree of misalignment between superimposed domains within the film and the sheet conductance is demonstrated through quantitative analyses.

References

[1] *E. Desgué et al., arxiv.org/abs/2503.20659 (2025)*

[2] D. Dosenovic et al., 2D Materials, 10 (2023) 045024.

Figures

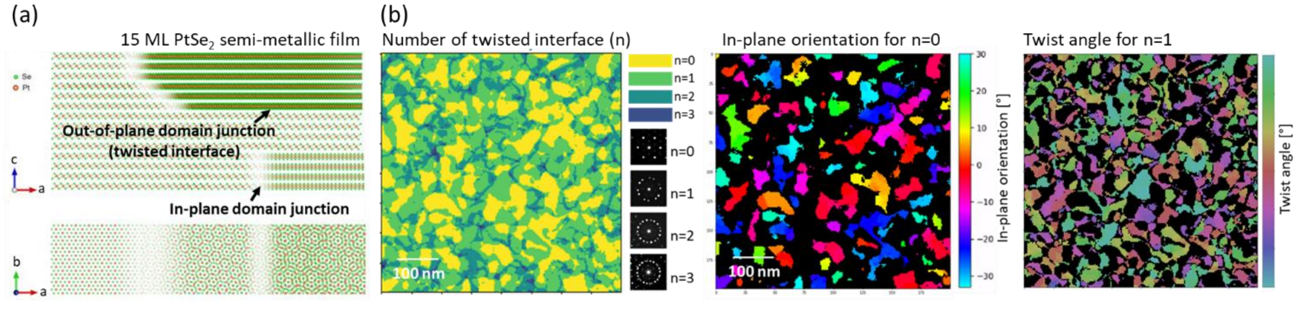

Figure 1: (a) Schematical domain configuration within a 15 ML semi-metallic $PtSe_2$ film and (b) examples of quantitative visualization of in-plane and out-of-plane information from 4D-datasets