Supporting Information

Robust Sample Preparation of Large-Area In- and Out-of-Plane Cross-Sections of Layered Materials with Ultramicrotomy

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Section I: Embedding, trimming and sectioning.

Embedding into epoxy. All the specimens discussed in this work were embedded in an Agar Low Viscosity resin, from Agar Scientific. The ratio between the two hardeners, VH1 and VH2, as well as the curing temperature was optimized in order to achieve the desired hardness of the epoxy blocks. The two other components of the recipe, namely the amount of low viscosity resin and of the accelerator, were kept fixed at their baseline values of 48 g and 2.5 g respectively. **Table S1** summarizes the different amounts of the components used for the premixed embedding epoxy mixture, from which their relative proportion can be determined.

The resulting epoxy mixture was further optimised by means of adjusting the curing temperature and the curing times. We found that a curing temperature between 60 ºC and 70 ºC together with around 16 and 9 hours of curing time represent reasonable parameters to ensure a suitable polymerization of the resin, since these choices result in epoxy blocks with consistent and reproducible properties.

Table S1. Components of the premixed embedding epoxy mixture. In each case we indicate its structure and the amount used for the optimal recipe used here.

Component	Structure	Weight (g)
Low Viscosity Resin	-	48
Low Viscosity Hardener (VH1)	(2-Nonen-1-yl) Succininc anyhrdide	12
Low Viscosity Hardener (VH2)	1,2,3,6-tetrahydromethyl-3,6-methanophthalicanhydride, MNA	40
Accelerator	Benzyldimethylamine, BDMA	2.5

The embedding procedure used to create the in-plane (for MoS_2) and out-of-plane (for WS_2) cross-sections differ in the relative orientation of the specimens within the epoxy block. As discussed in the main text (see **Figure 1**), this orientation is the main factor that needs to be considered before sectioning. In the case of in-plane cross-sections the embedding procedure is straightforward given that the whole process takes place at once. We used a cylindrical Teflon mold (**Figure S1a**) as a container for preparing the epoxy block. First, the wafer was placed below the cylindrical container, aligned with the hole. Afterwards, the hole was filled with the premixed epoxy. Then the wafer/epoxy/container system was placed inside a furnace where the epoxy was cured for 9 hours. **Figure S1b** shows the cured epoxy block containing the wafer once it has been removed from the cylindrical container. Such a curing time allowed detaching the wafer while maintaining the specimen inside the epoxy block (**Figure S1c**).

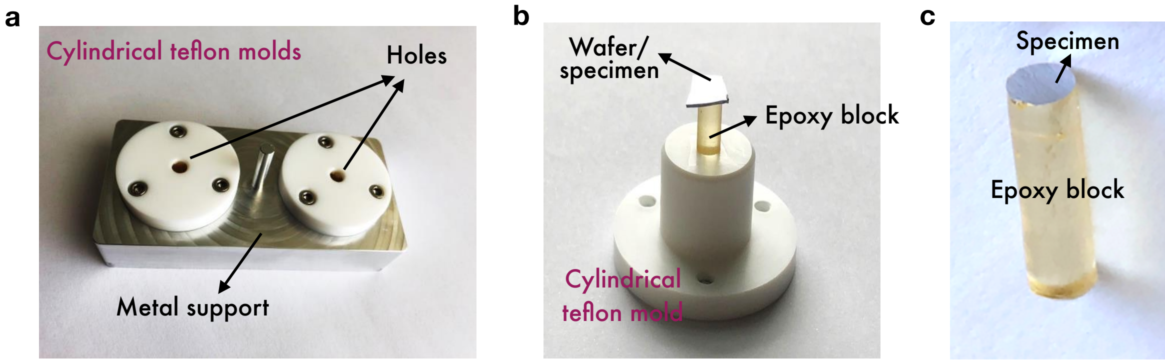

Figure S1. (a) The cylindrical Teflon mould used as container for the preparation of the epoxy blocks. **(b)** The cured epoxy block containing the wafer with the specimen and **(c)** the resulting epoxy block once we have detached the wafer containing the specimen.

Concerning the preparation of the epoxy blocks for the out-of-plane (WS_2) cross-sections, a two-step process was followed. Here a flat silicone rubber mold was used (**Figure S2a**). To begin with, half of the silicone mold was half-filled with the premixed epoxy. The wafer/specimen was then placed on top the epoxy, with the specimen side facing the epoxy (**Figure S2b**). The whole epoxy/wafer system was then placed inside a furnace and cured at a temperature of 70 ºC during 4 hours. We determined that this was the minimum amount of time required to ensure that the wafer/specimen could be detached from the half-epoxy block. This epoxy/specimen was again placed inside the silicone rubber mold and covered with epoxy and cured for a further 12 hours. **Figure S2c** displays the final sandwich-like assembly which we then use for the fabrication of out-of-plane cross-sections.

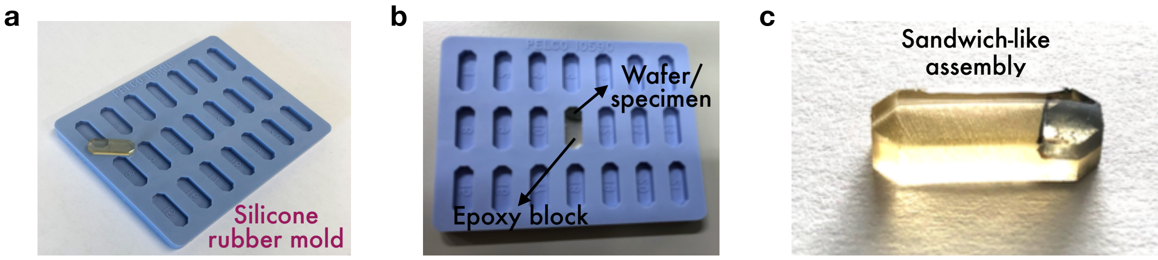

Figure S2. (a) The flat silicone rubber mould used to fabricate the out-of-plane WS_2 cross-sections. **(b)** The wafer/specimen on top of the half-filled epoxy in the silicone mold. **(c)** The final resulting sandwich-like assembly.

It is worth emphasizing that during the filling process it is crucial to reduce or ideally to avoid altogether the possible incorporation of air bubbles in the mixture. We found that by keeping the prepared epoxy mixture at a room temperature for between 10 and 15 minutes it was possible to reduce significantly the number of air bubbles formed during the preparation of the premixed epoxy.

Trimming and sectioning. The trimming and sectioning have been carried out using an RMC Ultramicrotome PowerTome PC from Boeckeler Instruments (**Figure S3a**). For the trimming of the cured epoxy block containing the specimen, a razor blade and a sculpt were used. Here it is very important to select the trimmed area accordingly to the orientation of the embedded specimen in the epoxy block. The obtained trapezoid-like shape face has a size of around 0.5 mm, with its area being 0.290 mm × 0.228 mm and 0.488 mm × 0.563 mm for the resulting out-of-plane and in-plane orientations respectively, see **Figures S3b** and **S3d**.

For the sectioning procedure, we employed a DiATOME diamond knife with a cutting angle of 35º and a clearance angle at 6º. The cutting speed and the sectioning thickness were generally set to 1 mm/s and a programmed thickness of 20 or 30 nm was adopted respectively. These ribbons of the slices were collected from the deionizer bath with a perfect loop and subsequently deposited onto a 300-mesh lacey carbon film copper TEM grid. These ribbons were then dried using a filter paper deposited onto the TEM grid (**Figures S3c** and **S3e**).

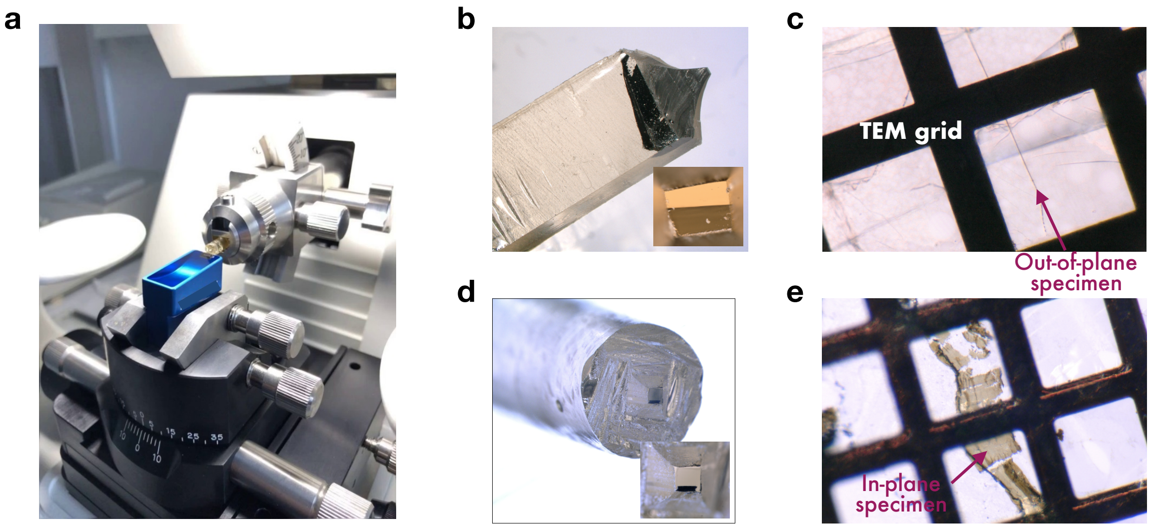

Figure S3. (a) The RMC Ultramicrotome used for the trimming and sectioning of our samples. **(b)** and **(d)** Resulting trapezoid-like shapes after the trimming of the cured epoxy block for the out-of-plane and in-plane specimens, respectively. **(c)** and **(e)** Optical microscope images of the TEM grids with the ribbons containing the specimens on top.

Section II: Structural characterization

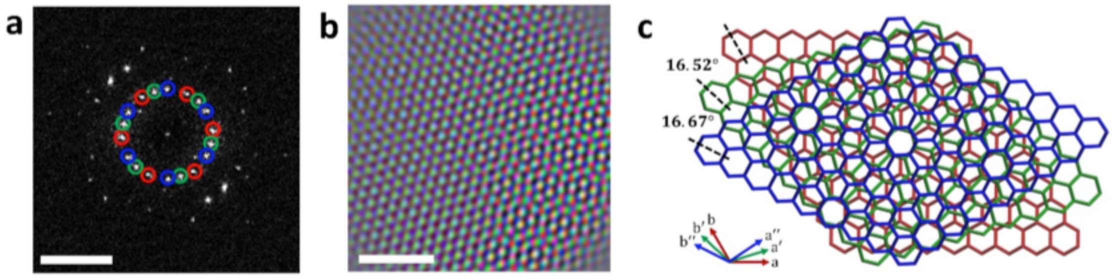

Figure S4. (a) A FFT taken from the high-resolution TEM image of a Moiré region in in-plane cross-section MoS_2, see **Figure 5c** from the main manuscript. One can observe three sets of Bragg reflections along the [0001] zone axis which correspond to three layers of MoS_2 with a relative rotation among them. **(b)** the corresponding inverse FFT. **(c)** A structural model representing these three MoS_2 layers rotated among them, and indicating the relative rotation angles. The scale bars in **(a)** and **(b)** are 5 nm^{-1} and 2 nm respectively.