## Quantifying the local mechanical properties of twisted double bilayer graphene Electronic Supplementary Information

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## 1 Adhesion maps

The UFM response depends on the local mechanical properties of the material under the probe and the adhesion between the tip and the sample<sup>1–3</sup>. To rule out any adhesive contribution to our measured UFM response in TDBG, we performed adhesion maps with the PeakForce Quantitative Nanomechanics (QNM) method. Figure 1 shows the maps obtained. As the tip was not calibrated, the modulus and adhesion values obtained could not be quantified.

The maps were performed in the same twisted region as the UFM measurements shown in the main text. The moiré superlattice is not visible neither on the modulus nor on the adhesion maps. These results also highlight the higher sensitivity of the UFM technique for such samples as the domains walls are not visible with PeakForce QNM.



Figure 1 PeakForce Quantitative Nanomechanics (QNM) maps. a) Topography, b) Modulus, and c) Adhesion map. Scale bars: 400 nm.

## 2 Ultrasonic Force Microscopy calibration

Quantification of the UFM signal is not straightforward as explained elsewhere 1-3. However, for a given probe and assuming its apex remains unchanged during all experiments, it is possible to directly tune the UFM signal on materials with known mechanical properties.

This calibration method is particularly well suited for the experiments shown here, as we have several materials close to each other - namely silicon dioxide  $(SiO_2)$  and hexagonal boron nitride (hBN) - to fine-tune the UFM response.

Figure 2a shows a UFM image of hBN to  $SiO_2$  step. A line profile is also shown on Figure 2b.  $SiO_2$  has a modulus of 70 GPa<sup>4</sup> while hBN has an out-of-plane modulus of 38 GPa<sup>5</sup>. We should note that for 2D materials, as the out-of-plane Young's modulus is much smaller than the in-plane one, the mechanical response to a vertical indentation is dominated by the out-of-plane modulus. For example, hBN has an in-plane modulus of 811 GPa<sup>6</sup> while its out-of-plane modulus is 38 GPa<sup>5</sup>. The same happens for graphite, whose the out-of-plane modulus of few layer graphene is 39.5 GPa<sup>7</sup>.

The UFM method can probe quite deeply in the hBN/TDBG stack<sup>3</sup> and thus if the TDBG was placed on a material with a different Young's modulus, softer or harder, the quantification of the UFM signal would need to include the substrate contribution. As hBN's out-of-plane Young's modulus is very similar to that of few layer graphene, the measurements are directly sensitive to signal variation occurring in the TDBG layer.

Based on the measurement shown in Figure 2, we can calibrate a variation in the UFM signal with a variation in Young's modulus. In our case, 30% decrease in UFM signal corresponds to 45.7% decrease in Young's modulus. Using this calibration, we can then extract quantitative modulus values for the four layers graphene and then for the twisted area.



**Figure 2 Ultrasonic Force Microscopy calibration.** a) UFM map of the step between hexagonal boron nitride (hBN) and silicon dioxide (SiO<sub>2</sub>). b) Profile of the UFM signal along the blue linecut in (a). Scale bar:  $1 \ \mu m$ .

## Notes and references

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