## Measuring and controlling out-of-plane shape of free-standing 2D materials

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In this work, we show through transmission electron microscope (TEM) and atomistic simulations that the nonflatness of free-standing graphene, hBN, and MoS<sub>2</sub>, as well as their heterostructures varies depending on the material. Out of all studied materials, graphene is the least flat, followed by hBN and finally MoS<sub>2</sub>. For the heterostructures, the overall shape is determined to a large extent by the stiffer of the two materials.

In addition to measuring the out-of-plane shape, we can also control it *in situ* in one direction using a stretching holder. For these experiments, we glued the samples, transferred onto gold TEM grids with a perforated amorphous carbon film, onto the holder and applied mechanical strain with small incremental steps to avoid breaking the film during the experiment. Figure 1 shows the results of one such study. The deviation of circular symmetry of the diffraction pattern and the shape of the individual diffraction spots give us insight on the strain in the material and its out-of-plane shape, respectively.

Our results show that this simple method can be used to completely flatten the 2D materials in the direction of the applied force. At this point, the material exhibits an aligned set of one-dimensional corrugations. After the structure has been flattened, continuous mechanical deformation leads to a measurable strain in the structure.



Figure 1: Evolution of the diffraction pattern of graphene and individual diffraction spots during the experiment. (a-c) Diffraction pattern recorded at different stages of the experiment: (a) at the beginning, (b) towards the end and (c) at the end. All shown patterns were recorded at sample tilt  $a = 21^{\circ}$ . The dashed lines show the approximate tilt axis and the overlaid hexagons highlight the first set of diffraction peaks. The panels on the right show a zoom-in of the indicated diffraction spots in false color. (d) Orientation of the ellipse fitted to the diffraction patterns ( $\Theta$ ) and that of the diffraction spots ( $\theta$ ) during the experiment. (e) Ellipticity of the diffraction patterns (B/A, right y-axis) and spots (b/a, left y-axis). Standard errors from the fits to the measured values are contained within the markers. All diffraction pattern values are for a = 0° and spot values for a = 21°. In panels d and e, the values corresponding to the diffraction patterns shown in panels a-c are marked with corresponding labels. The x-axis values ( $\Delta$  $G/G_0$ ) indicate the relative change in the size of the gap in the sample carrier over which the sample was suspended.