## Quantifying multiple crystallite orientations and crystal heterogeneities in complex thin film materials

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## **Supporting information**

## **GIWAXS acquisition parameters**

Samples were all collected at Cornell High Energy Synchrotron Source, but under different conditions. GIWAXS data for  $(C_6H_5C_2H_4NH_3)_2SnI_4$  and  $CH_3NH_3PbI_3$  thin films were collected at the D1 station. The beam energy was 13.0 KeV, with a sample to detector distance (SDD) of 190 mm. The incident angle for this sample was 0.25°. GIWAXS data for the ZnO thin films were collected at G1 station. The beam energy was 10.0 KeV, with a SDD of 97.8 mm. The incident angle for this sample above the films critical angle and below that of the substrate 0.32°. GIWAXS data for PEDOT thin films were collected at D1 station. The beam energy was 10.6 KeV, with a SDD of 173 mm. The incident angle for this sample was 0.14°.

## Weighted Amplitude prerequisite

As stated in the paper, the weighted amplitude must be corrected if there are amplitude values at 0° or 90° to avoid any miscalculation of the MF value. This is because each amplitude is in essence a single point narrow crystallite being quantified. It is easier to see the issue with the edge amplitude when quantifying wider crystallite sizes. Figure S1 shows a constructed set of gaussian fits that correlate to the simulated intensity. As GIWAXS diffraction is symmetric across these planes, it is known that for each gaussian at a given  $\varphi$ , a matching gaussian occurs at the same distance away from 0°. This overlap must be accounted for when quantifying the weighted amplitude. For a value at 0, there are actually two equivalent gaussians that contribute to the total intensity. In the case of MF, each value is centered around a specific  $\phi$ , and represents a discrete width equally in both directions. If we consider the discrete width of each respective component, while not paying attention to the intensity of this location, one major observation is that the signal centered at 0° or 90° contributes equally within the 0° to 90° and -90° to 0° and 90° to 180° respectively. This can be seen in Figure S2, which visualizes discrete azimuthal linescans with discrete components 5° in width, and a 2.5° space in between each component. This visualization shows that the total signal collected at 0° and 90° only contribute half of their signal within the 0° to 90° range. This can also be seen in **Figure S3**, where only half of  $\phi_s = 0$  is contributing to the orientation within the range 0° to 90°. Thus signal that is observed at 0° and 90° in an  $I(\phi)$  vs  $\phi$ plot must be divided by two when quantifying the weighted amplitude.

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This was correction was identified after the values used in section 3.2 where quantifying orientation along various angles did not perfectly match. In this case all interior values yielded an MF of 0.97, while values centered at  $\varphi = 0^{\circ}$  and 90° were equal to 0.96. All the distributions should exhibit the same shape and the MF values should also equal each other. The issue stems from fact that signal oriented at 0° or 90° has signal that contributes 50% within the first quadrant and 50% in the second and fourth quadrant of the graph (**Figure S2**). Experimentally, the second and fourth quadrant are not studied therefore this data needs to be corrected. This issue is visualized in Figure S1 shows an example where data 0° and 90° are 50% inside the quadrant of interest, and 50% outside quadrant of interest. By correcting the weighted amplitude at 0° and 90°, the MF values in this example are now all equal, which matches what is experimentally expected.



**Figure S1.** An overall Gaussian signal comprised of multiple contributing Gaussian signals, two of which overlap at  $\varphi_s = 0^\circ$ , showing why the weighted amplitude at the spectral edges are actually composed of two equivalent signals.



**Figure S2.** An azimuthal linecut with discrete points of widths of 5° and a space between points of 2.5° used to show the fact that data points collected at 0° and 90° contribute only 50% of their signal to the ODF of the material. The other 50% of their signal contributes to the neighbouring quadrants.



**Figure S3.** Discrete values in a Gaussian distribution centered around  $\varphi_s = 0^\circ$  extend in both directions, showing why the weighted amplitude at 0 or 90 should only use half of the signal.