Visualizing and quantifying structural distortions of an unstable phonon-mode

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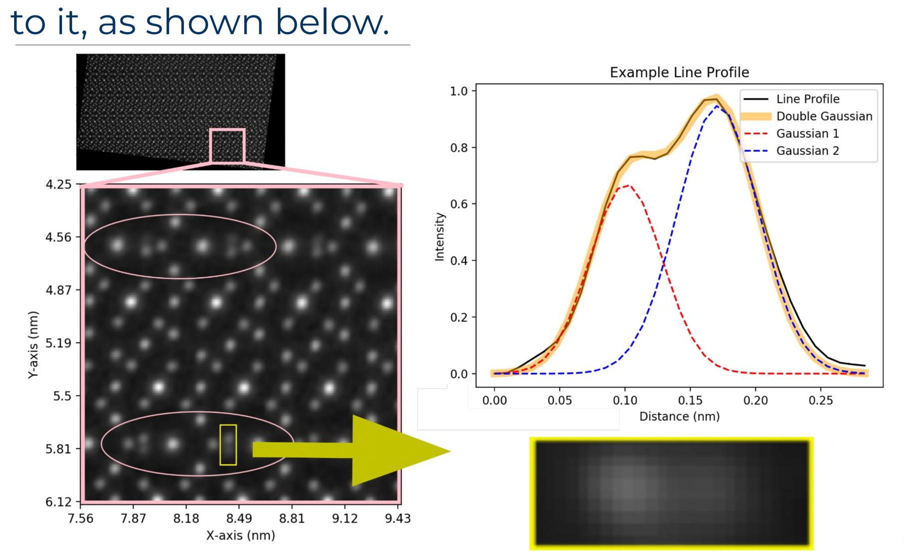
Abstract

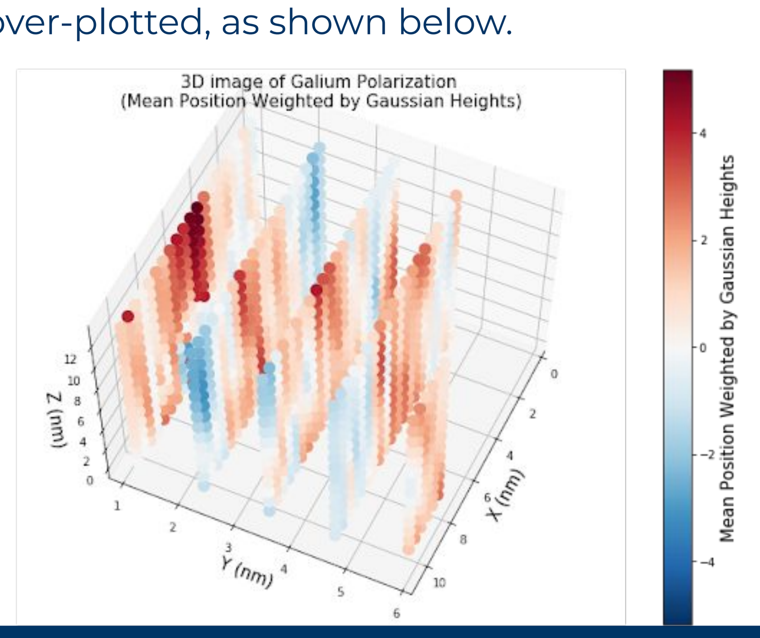
The crystal structure of strontium hexagallate hosts an unstable phonon mode that exhibits signs of spontaneous symmetry breaking, leading to the formation of nanometre-scale polar regions in the material. By analyzing the polarization of individual unit cells and its spatial variation from multislice-ptychography images, we quantify the size of such polar nano-regions. We used the Moran's I coefficient to quantify spatial correlations in the measured polarization, and observed a statistically significant correlation between the spatial position and polarization with a p-value ranging from 0.0020 - 0.018 for different sample regions.

Quantifying Polarization

- We use ptychographically reconstructed images of strontium hexagallate, creating a 3D rendering of the original sample by producing a series of 2D images or slices spaced at 1 nanometre intervals descending into the sample.
- We extract the line profile of the gallium atom and then fit double Gaussians to it, as shown below.

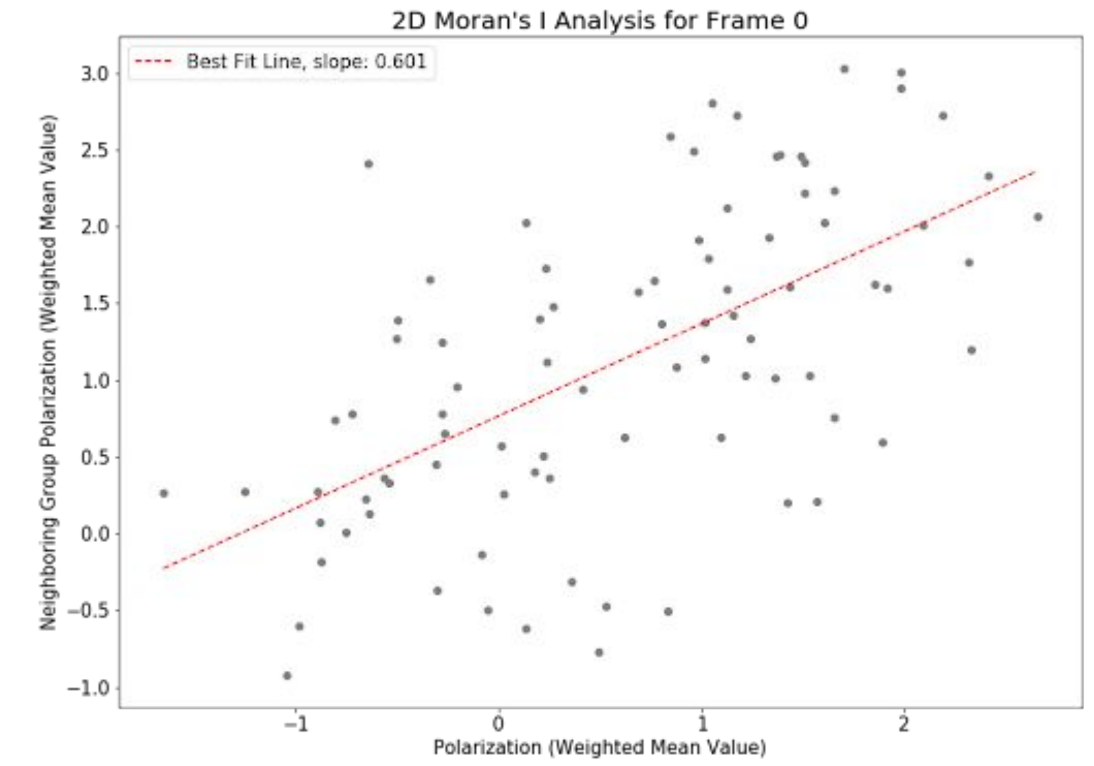


- We then use this fit to quantify the polarization by taking the averaging position of each Gaussian, weighted by the height of the Gaussian, producing a value called the mean weighted position.
- The mean weighted position is more positive for gallium atoms polarized up, more negative for gallium atoms polarized down, and zero for non-polarized gallium atoms.
- We then create a 3D visualization of the sample with the polarization of each unit cell over-plotted, as shown below.

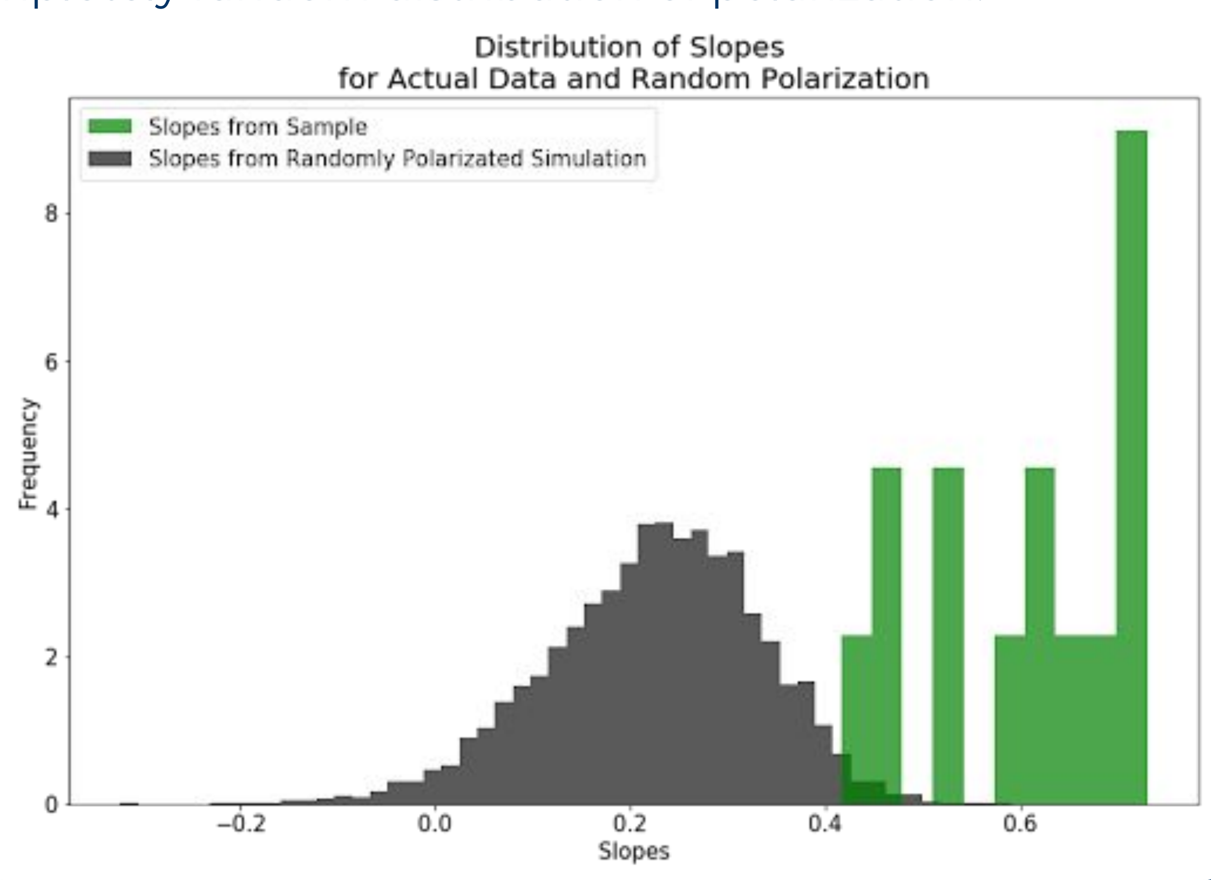


Establishing a Correlation between Position and Polarization

- To quantify whether there is a correlation between position and polarization, we utilize the Moran's I statistic.
- For each gallium atom, we take the mean polarization of every gallium atom within a radius of 2 nanometres. We then plot this value against the polarization of the original gallium atom. An example of this calculation performed on one ptychographic frame with a slope of 0.601 is shown in the figure below.



- In order to estimate the statistical significance of this slope, we calculate what the typical slope would be for our sample images if the polarizations are randomly distributed throughout the sample by randomly re-assigning each polarization value to a new gallium atom, then calculate the slope of that fictitious sample.
- This distribution of slope values (N=500) is shown in the figure below, where there is only a 1.8% overlap between the zero-correlation distribution of slopes and the actual observed distribution of slopes across all frames. This means that there is only a 1.8% chance of observing these slopes given a completely random distribution of polarization.



Conclusion

In conclusion, we notice a significant correlation between position and polarization, with the p-value ($p = (N_{OVERLAP}+1)/(N_{TOT}+1)$) ranging from 0.0020 - 0.018 across all ptychographic frames. Therefore, our samples of strontium hexagallate contain similarly polarized nano-regions. In future work, we seek to characterize the typical size of these polar nano-regions for the purpose of determining strontium hexagallate's utility in electronic memory storage.

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References

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