

# Supplementary Information for Analysis of interpretable data representations for 4D-STEM using unsupervised learning

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## 1 Supporting Tables and Figures

Table S1: Parameters and Runtimes for Unsupervised Learning Models

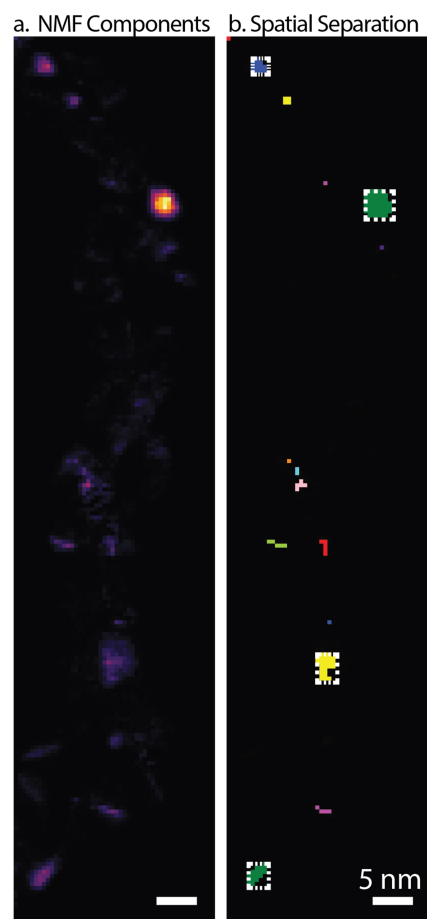
Dataset	Feature	Input Components	Correlation Threshold	Runtime [Minutes]*
Ag1	RV	30	0.30	26
Ag1	AA	50	0.40	21
Ag1	BD	60	0.20	95
Ag2	RV	25	0.45	21
Ag2	AA	50	0.40	65
Ag2	BD	75	0.25	181
Ag3	RV	25	0.45	19
Ag3	AA	50	0.40	65
Ag3	BD	60	0.25	88
Pd@AuAg NW	RV	12	0.40	5
Pd@AuAg NW	AA	50	0.48	54
Pd@AuAg NW	BD	100	0.18	270

\*Runtime was measured using a MacBook Pro equipped with a 2.3 GHz 8-Core Intel Core i9 processor

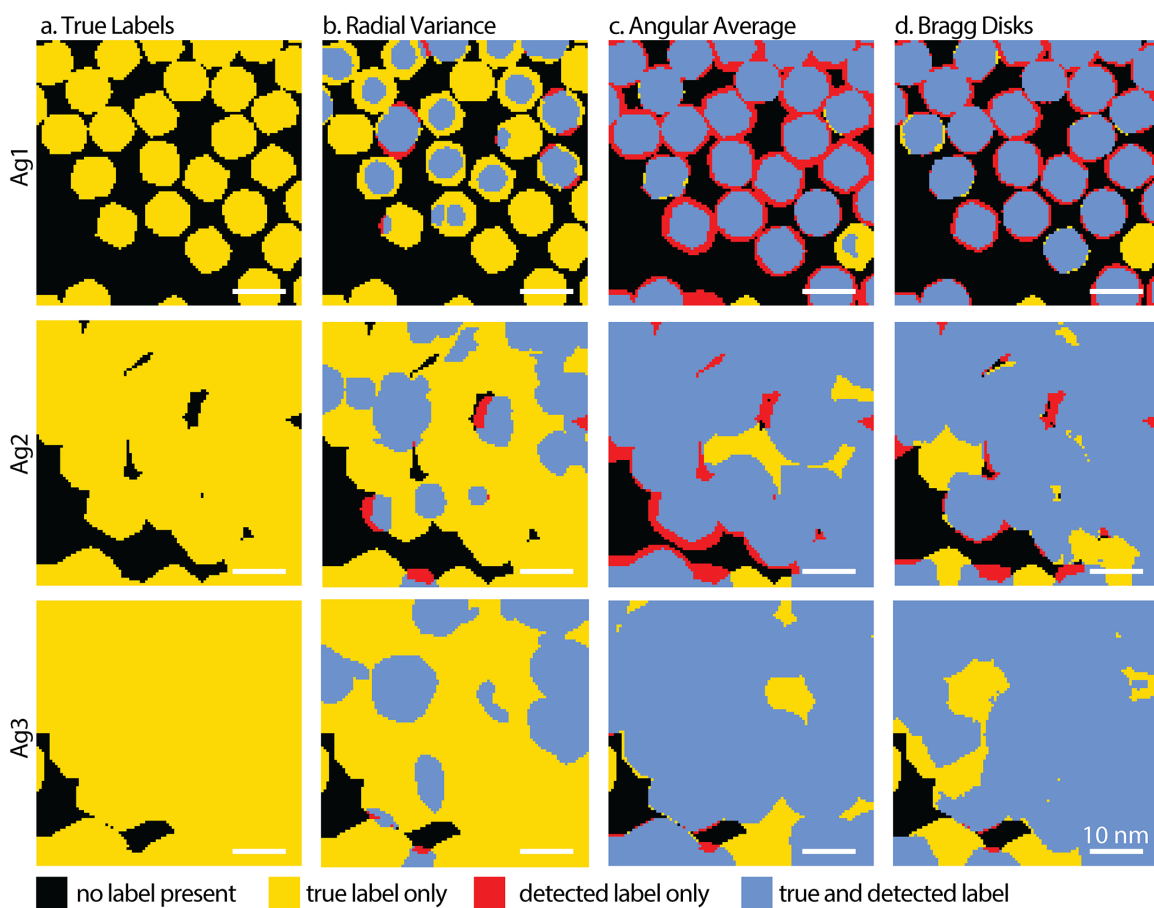
Table S2: Area-Based Metrics

Dataset	Feature	$TPR_A$	$FPR_A$	$FNR_A$	$TNR_A$	$Precision_A$
Ag1	Radial Variance	33.0	2.6	70.0	97.4	93.4
Ag1	Angular Average	95.8	35.4	4.2	64.6	75.3
Ag1	Bragg Disk	94.3	20.3	5.7	79.6	83.9
Ag2	Radial Variance	33.0	2.6	70.0	97.4	93.8
Ag2	Angular Average	92.9	44.6	7.1	55.4	92.6
Ag2	Bragg Disk	87.7	24.2	12.3	75.8	95.7
Ag3	Radial Variance	28.0	4.1	72.0	95.9	99.2
Ag3	Angular Average	91.0	8.6	9.0	91.4	99.5
Ag3	Bragg Disk	84.7	3.6	15.3	96.4	99.8

True positive rate ( $TPR_A$ ), true negative rate ( $TNR_A$ ), false positive rate ( $FPR_A$ ), false negative rate ( $FNR_A$ ), and precision ( $Precision_A$ ) of the three simulated Ag datasets using the area-based metrics described in the methods section of the main text. The area-based metrics score how well the features were able to predict the presence of any non-distinct crystalline region in the dataset.



**Figure S1:** Methodology of spatial and size refinement. a) Virtual image created from an individual radial variance feature component. b) Spatially separated grains from an individual input column, with those selected as independent clusters after size refinement in white dashed bounding boxes and those without bounding boxes excluded from analysis.



**Figure S2:** Number of grains per pixel for the Ag film simulation. a.) Binary area label, where the gold indicates where a grain is present and the black indicates a region with no grain. Maps showing where true labels, detected labels, and overlap are present in the b) Radial Variance, c) angular average d), and the Bragg disk features. The red labels count against the area-based precision metric as these indicate false positives, while the gold regions (false negatives) do not count against the area-based precision metric.

Table S3: Grain Match - Average Metrics

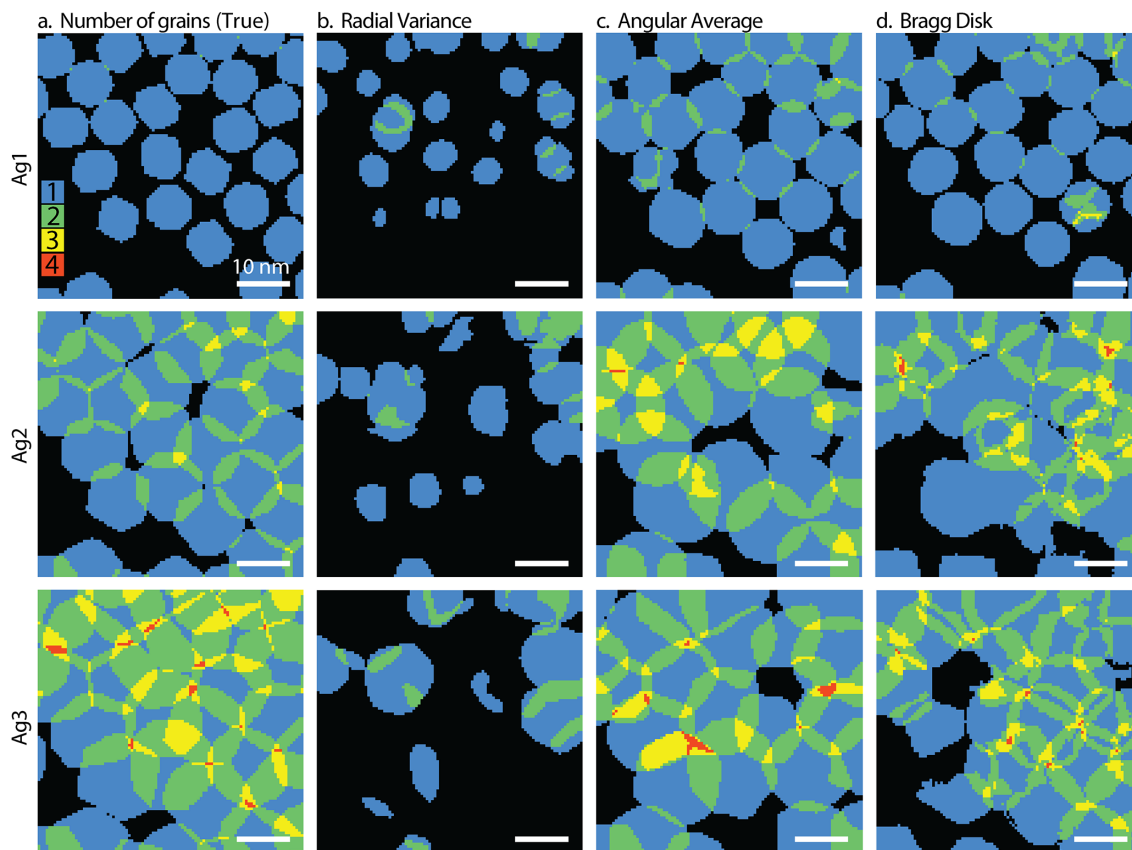
Dataset	Feature	$TPR_G$	$FPR_G$	$FNR_G$	$TNR_G$	Dissimilarity Score
Ag1	Radial Variance	38.6	0.1	61.4	99.9	17.0
Ag1	Angular Average	86.4	0.6	13.6	99.4	12.2
Ag1	Bragg Disk	89.5	0.3	10.5	99.7	12.6
Ag2	Radial Variance	36.9	0.3	63.1	99.7	30.6
Ag2	Angular Average	89.3	1.2	10.7	98.8	19.0
Ag2	Bragg Disk	70.2	0.9	29.8	99.1	21.7
Ag3	Radial Variance	33.5	0.3	66.5	99.7	58.1
Ag3	Angular Average	73.6	0.3	26.4	99.7	34.4
Ag3	Bragg Disk	48.8	0.1	51.2	99.9	32.0

The average values for the true positive rate ( $TPR_G$ ), true negative rate ( $TNR_G$ ), false positive rate ( $FPR_G$ ), and false negative rate ( $FNR_G$ ) of the three simulated Ag datasets using the grain-based metrics described in the methods section of the main text. The grain-based metrics score how well the features were able to predict the presence of a specific spatially distinct crystalline region in the dataset.

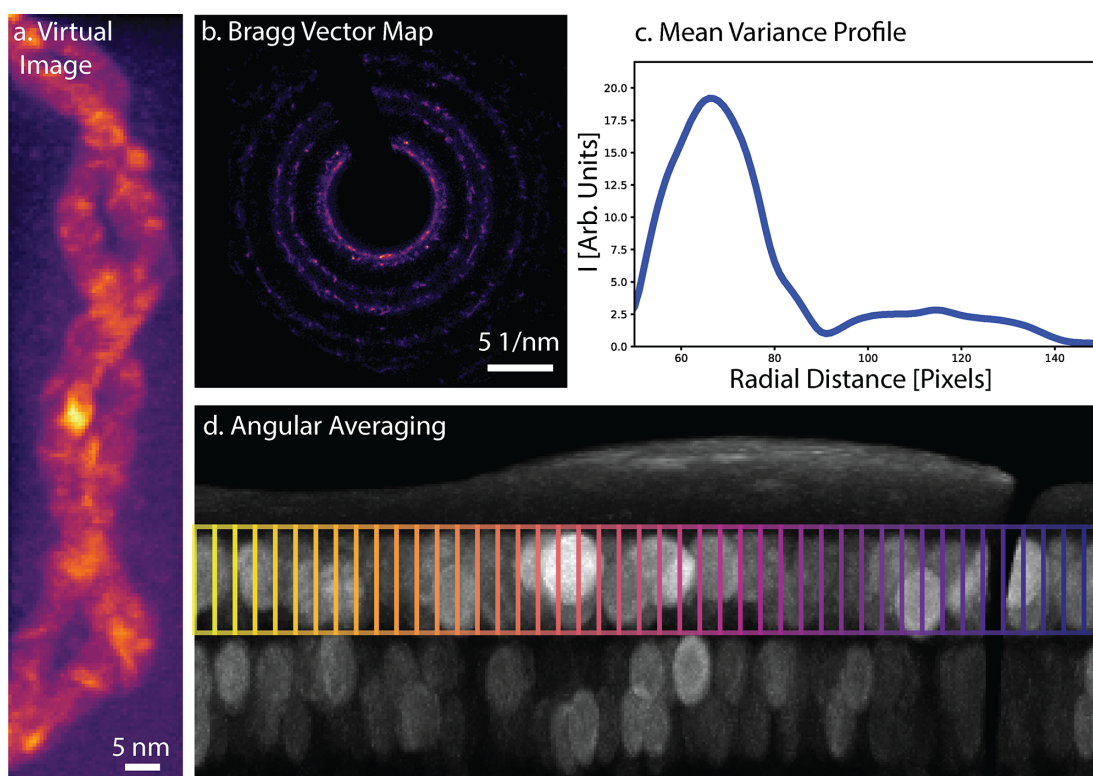
Table S4: Grain Match - Feature Metrics

Feature	$TPR_G$	$FPR_G$	$FNR_G$	$TNR_G$
Radial Variance	36.5	0.2	63.5	99.8
Angular Average	83.2	0.7	16.8	99.3
Bragg Disk	67.4	0.4	32.6	99.6

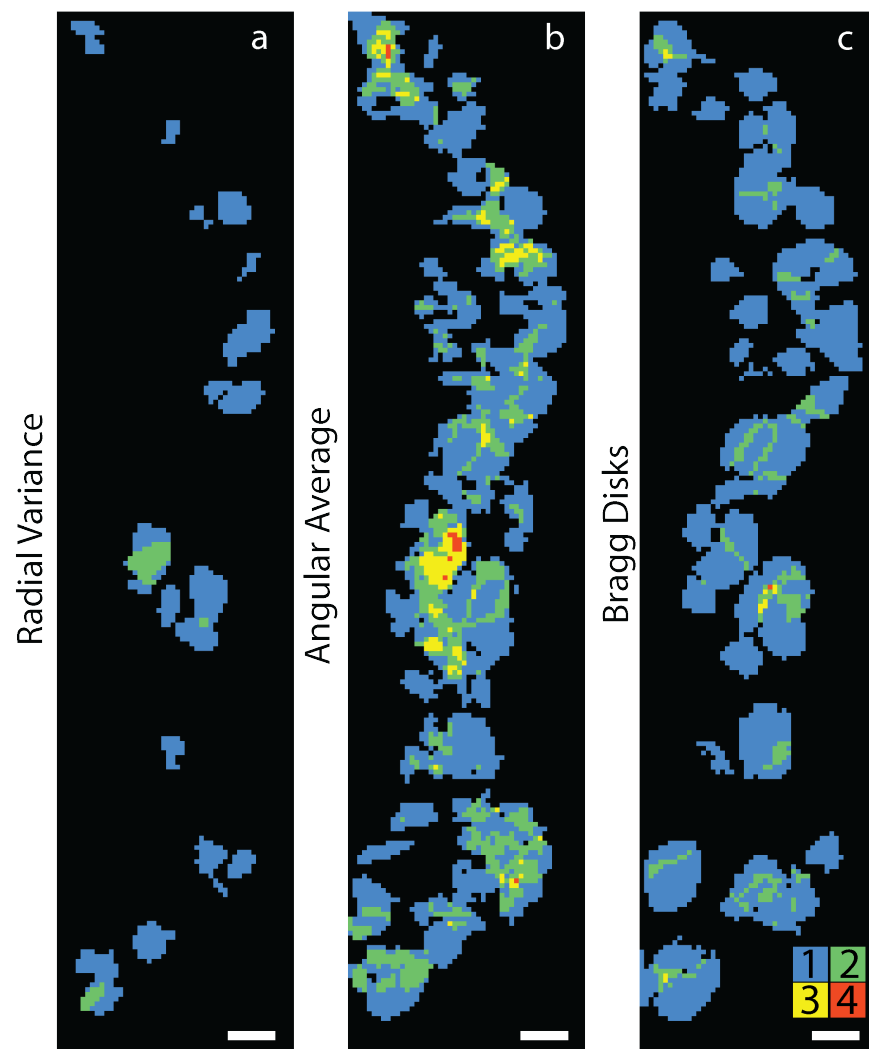
The average values for the true positive rate ( $TPR_G$ ), true negative rate ( $TNR_G$ ), false positive rate ( $FPR_G$ ), and false negative rate ( $FNR_G$ ) by averaging all scores calculated across the three Ag datasets by feature representation using the grain-based metrics described in the methods section of the main text. The grain-based metrics score how well the features were able to predict the presence of a specific spatially distinct crystalline region in the dataset.



**Figure S3:** Number of grains per pixel, simulated data. Number of a) known or detected grains per pixel in the 3 simulated datasets using RV b), AA c), and the BD d) features.



**Figure S4:** Feature Representations for Pd@AuAg Nanowire. a) Mean virtual image. b) Histogram of detected Bragg disks within the dataset. c) Average variance profile. d) Representation of the angular average feature set performed on the transformed max diffraction pattern.



**Figure S5:** Number of grains per pixel, Pd@AuAg NW. Number of clusters per pixel detected using the a) radial variance, b) angular average or c) Bragg disk feature. Scale bar 5 nm.