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Spatial Order of Latent Variables to Characterize Multi-range Symmetry Lowering Distortions in a $Pd_3Bi_2Se_2$ Superconductor

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The spatial order of symmetry lowering variations from a high-symmetry host structure is often a critical target of (S)TEM characterization that can span multiple length scales. Even for a single crystal it can include the obvious identification of the host crystal structure, but also charge ordering, dopant/vacancy ordering, ferroic texture, charge density waves, and features of the TEM sample geometry such as plasmon oscillations with thickness. For multi-phase systems the moiré patterns resulting from projection of overlapping structures is increasingly a by-design functional property [1] and not just an imaging artifact. Moreover, this latter property is typically ignored in its most ubiquitous occurrence as reaction and reconstruction layers of nominally single-phase 3D materials at the entrance and exit surfaces. In this work we employ encoder networks for dimensionality reduction to isolate distortions from the highest symmetry primitive cell (the image-wide average) to independently evaluate their spatial order. We explore a case study of a superconducting ternary chalcogenide $Pd_3Bi_2Se_2$ thin film which is observed to exhibit such long-range spatially order distortions.

Dimensionality reduction tools are powerful for clustering and isolating independent features as orthogonal variables. In this application they disentangle low signal / subtle distortions from dominant features and, where multiple are present, from one another. Historically this is done agnostic to spatial relationships, e.g. PCA, NMF, k-means clustering, etc. but it can be incorporated in the encoding with convolution layers, or the latent variable loading maps can be spatially interrogated. A $Pd_3Bi_2Se_2$ thin film grown by Molecular Beam Epitaxy on $SrTiO_3$ [2] is observed to exhibit a significant long-range ordering of the nominally single-phase material. An atomic resolution HAADF image is shown in Fig 1a, with superlattice-type peaks visible in the FFT (Fig 1b-d). A primitive highest projected symmetry "unit cell" is defined as shown in Fig 1e, segmented, and spatially addressed as a matrix of "unit-cell" space. Although a minor spatial abstraction, it readily integrates with convolutional masks in the encoder layer to which a-priori restrictions of range and symmetry can be imposed. The latent component of the long-range distortions and their spatial periods are shown in Fig 2b for select latent variables from PCA. Three distinct periodicities are apparent, a short range in-plane unit cell doubling in #10 and #11, and longer range periods along ~[5,-2] (#7) and ~[2,-3] (#10 & #11) unit cells. For all, the signal variation is dominated by symmetry breaking on the Bi sites, possibly due to vacancy ordering as the material is known to be Bi deficient [2-3].

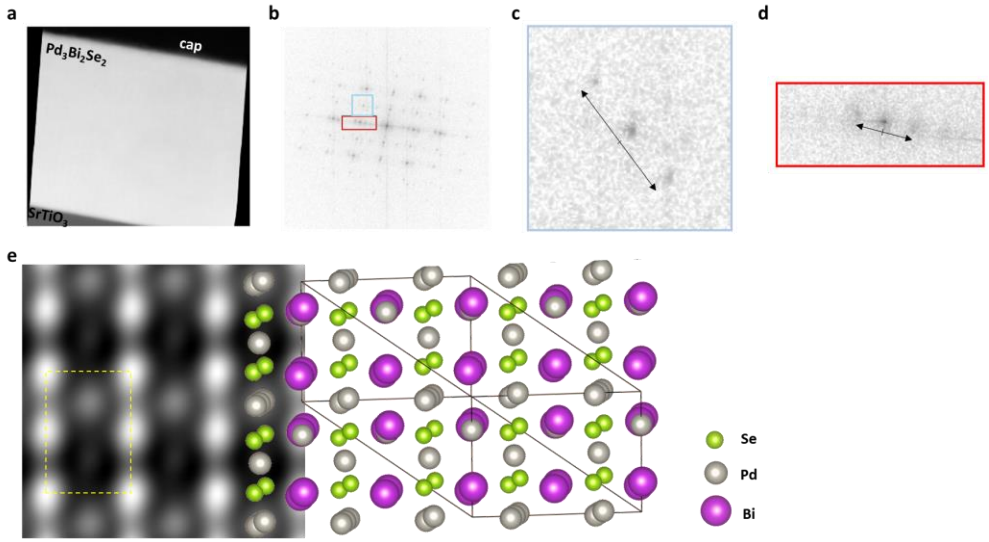

Figure 1. Superlattice-type ordering in $Pd_3Bi_2Se_2$ HAADF images. (a) Atomic resolution HAADF image. (b) Corresponding FFT. (c-d) Magnified views of the highlighted regions in the FFT showing superlattice-type satellite peaks along two different vectors. (e) Magnified HAADF of averaged structure, primitive "unit-cell" highlighted as dashed yellow line.

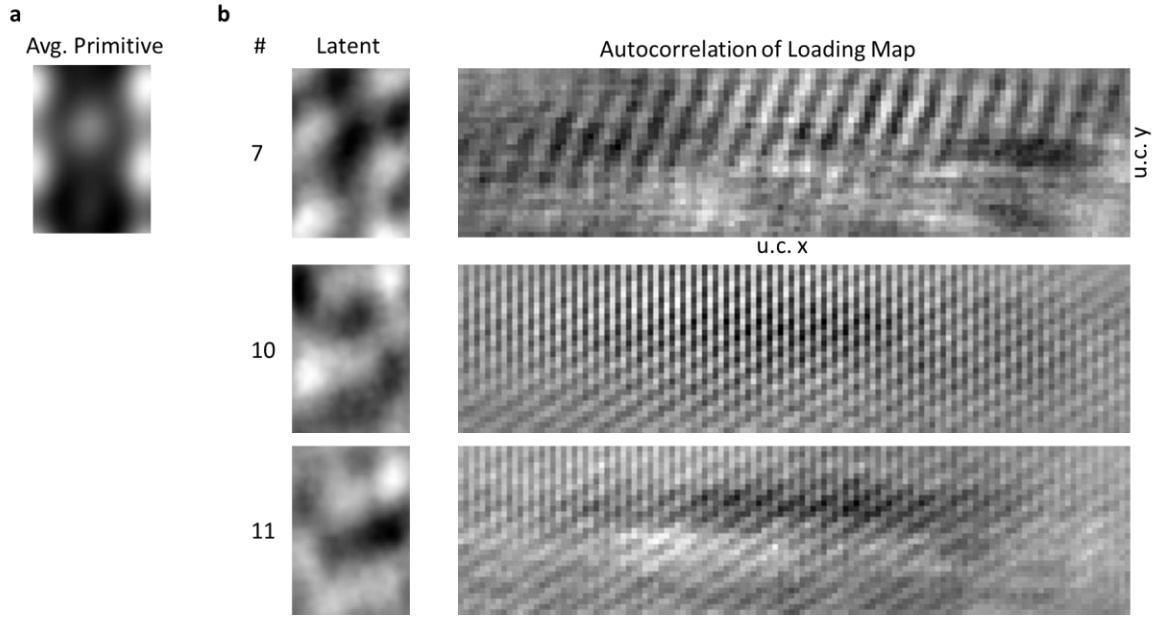

Figure 2. Spatial interrogation of PCA components. (a) Mean primitive "unit cell". (b) Select latent variables and their spatial distribution from autocorrelation of the loading maps in primitive unit-cell space.

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