Blind Source Separation in SPED datasets: Machine learning assisted phase and orientation determination in multilayer oxide electronic thin film devices

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Scanning precession electron diffraction (SPED) datasets containing single diffraction patterns which include information from more than a single phase can be deconvolved by using machine learning algorithms. In this work we use blind source separation routines to extract components from a dataset of a planview prepared thin film stack. The morphology and type of grain boundaries – governed by local epitaxial conditions – in these devices have a strong influence on the macroscopic device behavior due to their inherent physical properties, i.e. localized high defect concentration [1, 2]. While 4D-STEM methodologies can resolve the relevant structural features [3], acquiring statistical information is a time-consuming process. Here, we present a procedure that allows for fast acquisition of spatially correlated, multi-phase information in a two (or more) layered thin film stack. By transmitting a (precessing) quasi-parallel STEM beam through the sample (see **Figure 1(a)**) a four-dimensional data set is generated. In our setup, an NBD probe (in 2D real space) is scanned over the sample and reciprocal space information is collected on a fast pixelated single electron detector (Merlin EM, Quantum Detectors). In the geometry described above a set of superimposed diffraction patterns of all sampled phases contained in the stack (see **Figure 1(b)**). In order to distinguish between phases, we applied machine learning (ML) based blind source separation (BSS) routines [4]. Using a list of components generated by unsupervised non-negative matrix factorization (NMF) and simplex volume maximization (SiVM), the raw data was filtered. Recent developments in ML mainly contributed to the advancements in deep learning, have brought large improvements to interdisciplinary fields, e.g. signal processing and microscopy [5]. Deep neural networks (DNN) therefore present a promising extension to the ML based BSS routines. Deconvolving multi-phase superimposed SPED datasets, as presented here, can offer a unique and statistical way of analyzing complex crystalline thin film stacks with a high spatial resolution.

[1] Petzold S, Zintler A, *et al.*, *Adv. Electron. Mater.* ***10*** (2019) [2]
Xue K-H, Blaise P, *et al.*, *Appl. Phys. Lett.* ***20*** (2013)
[3] Ophus C, *Microsc. Microanal.* ***3*** (2019)
[4] Duncan N. Johnstone, Phillip Crout, *et al.*,(2020)
[5] Borodinov N, Neumayer S, *et al.*, *Npj Comput. Mater.* ***1*** (2019)
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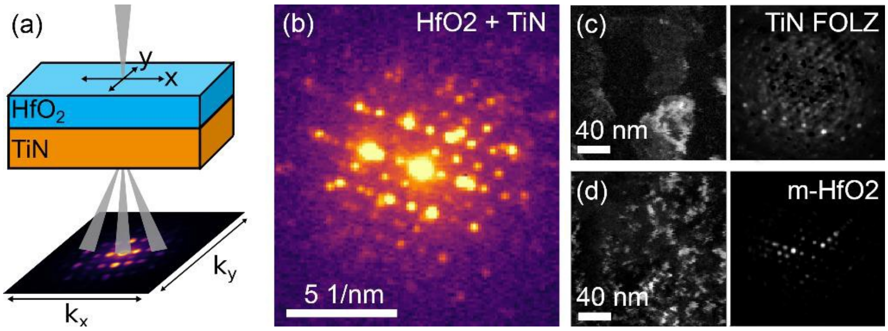

Figure 1: (a) Quasi-parallel 4D-STEM (SPED) and plan-view sample geometry, (b) single color-coded superimposed diffraction pattern of a HfO2 (blue) and a TiN grain (red), (c,d) Loadings and factors of two exemplary components of the SiVM result of the same area, revealing the different grain sizes observed in the layers.