4D-STEM/PNBD: Fast and easy powder electron diffraction in SEM

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Background:
We have developed a new method, named 4D-STEM/PNBD (i.e., four-dimensional scanning transmission electron microscopy/powder nanobeam diffraction), which can convert a modern SEM microscope to a simple, fast, and user-friendly powder electron diffractometer [1, 2]. The only hardware requirement is that the SEM microscope must be equipped with a 2D-array detector of transmitted electrons (also referred to as 2D-array STEM detector or pixelated STEM). In 4D-STEM/PNBD, we reduce a huge and complex 4D-STEM-in-SEM dataset to a single 2D powder diffraction pattern (as shown in the attached figure). The final powder diffraction patterns are equivalent to those from TEM/SAED as documented in our previous studies [1-3]. They can be compared to theoretically calculated powder X-ray diffraction patterns (PXRD) in order to identify the investigated nanocrystals [2, 3]. This contribution deals with the recent improvements of our method, which should make it even more user-friendly and robust [3, 4].

Materials and methods:
We performed TEM/SAED (selected area electron diffraction) and 4D-STEM/PNBD measurements on three types of nanocrystalline samples on an electron-transparent carbon film. The samples differed by signal-to-noise ratio (SNR), where signal and noise are represented by the intensity of diffraction peaks and the amorphous background, respectively. The samples could be grouped as follows: (i) Au nanoislands with high SNR, (ii) GdF3 and TbF3 nanocrystals with good SNR, and (iii) magnetic iron oxide nanoclusters with or without amorphous silica shell with intermediate SNR. The 4D-STEM/PNBD calculations were performed with recent version of our open-source Python libraries STEMDIFF (conversion of 4D datasets to 2D powder diffraction patterns; https://pypi.org/project/stemdiff) and EDIFF (conversion of 2D-diffraction patterns to 1D radially averaged diffraction profiles and their comparison with theoretically calculated PXRD; https://pypi.org/project/ediff).

Results and discussion: Our initial studies [1-3] showed that the 4D-STEM/PNBD works very well for reasonably small, highly diffracting crystals

with low absorption. If the crystals are thicker and/or surrounded by amorphous matrix, the extraction of 2D powder diffractogram can become difficult or impossible. This results from the lower-energy electrons in SEM (E < 30 keV), which suffer from higher absorption and inelastic scattering than the higher-energy electrons in TEM (E > 100 keV). The better 4D-STEM-in-SEM datasets and, subsequently, the higher-quality 2D powder diffractograms can be obtained with better hardware (i.e. better SEM microscope and/or pixelated STEM detector), better experimental parameters (optimized scanning speed, dwell time etc.), and better software (i.e. data processing). For given hardware and optimized experimental conditions, the decisive factor is the data processing. Consequently, we made several improvements of our STEMDIFF software. At first, we introduced better, multi-criteria filtering of the raw 4D-STEM-in-SEM dataset. The better filtering allows us to select the highly-diffracting locations and ignore the rest, which contain high noise. Moreover, it enables us to get the better estimate of the point-spread-function (PSF) of the primary beam, which can improve the quality of the individual diffractograms by means of 2D-PSF deconvolution. At second, we increased the speed of the above-mentioned time-consuming 2D-PSF deconvolution step ca 5x by introducing multicore processing. Last but not the least, we improved the user interface and extended the original software package STEMDIFF (the conversion of 4D-datasets to 2D diffractograms) with a sister package EDIFF (the conversion of 2D diffractograms to 1D profiles and their comparison with theoretically calculated PXRD patterns). The recent versions of STEMDIFF and EDIFF employ the well-established Jupyter notebooks as interactive templates for complete data processing (4D → 2D → 1D) without any third-party software. The notebooks are written and documented in such a way that the data could be processed in step-by-step way by any SEM user without detailed knowledge of diffraction theory. The above-listed improvements enabled us to process not only the 4D-STEM-in-SEM datasets of strongly diffracting samples with high SNR (such as Au nanoislands), but also samples with intermediate SNR (such as iron oxide nanoclusters enveloped with amorphous silica). The samples with low and poor SNR remain as a challenge for our ongoing work, which comprises advanced 2D-PSF deconvolution methods and automated, machine learning-based noise reduction.

Conclusion:
The 4D-STEM/PNBD method brings a simple, fast, and easy-to-use electron diffraction technique to SEM users. The classical SEM microscopes offer imaging modes (such as SE or BSE) and spectroscopy modes (such as EDX). The modern SEM microscopes equipped with pixelated STEM detectors add also the third mode – the electron diffraction. The pixelated detectors can be installed in a common SEM port like any other detector. The 4D-STEM datasets are easy-to-collect, but difficult-to-process – at least for non-crystallographers. Our STEMDIFF package reduces a complex 4D-STEM

dataset to a simple 2D diffractogram. The sister EDIFF package enables a fast conversion of 2D diffractogram to radially averaged 1D diffraction profile and its comparison with theoretically calculated PXRD diffraction pattern. Both packages aim to be as user-friendly as possible, so that the new 4D-STEM/PNBD method could be employed by all users of SEM microscopes.

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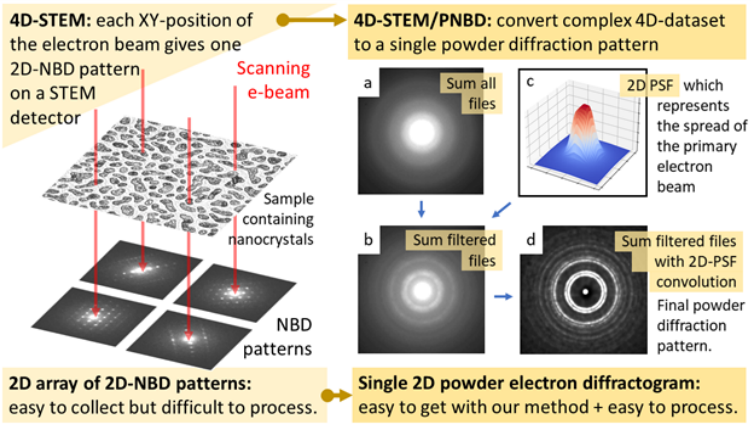
Graphic:



Keywords:

4D-STEM-in-SEM, powder electron diffraction, nanocrystals

Reference:

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