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# Beamstop-based low-background ptychography to image weakly scattering objects

Juliane Reinhardt[a,*], Robert Hoppe[b], Georg Hofmann[c], Christian D. Damsgaard[d], Jens Patommel[b], Christoph Baumbach[b], Sina Baier[c], Amélie Rochet[c], Jan-Dierk Grunwaldt[c], Gerald Falkenberg[a], Christian G. Schroer[a,e]

[a] Deutsches Elektronen-Synchrotron DESY, D-22607 Hamburg, Germany
[b] Institute of Structural Physics, Technische Universität Dresden, D-01062 Dresden, Germany
[c] Institute for Chemical Technology and Polymer Chemistry, Karlsruhe Institute of Technology, D-76131 Karlsruhe, Germany
[d] Center for Electron Nanoscopy and Department of Physics, Technical University of Denmark, DK-2800 Lyngby, Denmark
[e] Department Physik, Universität Hamburg, Luruper Chaussee 149, D-22761 Hamburg, Germany

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ABSTRACT

In recent years, X-ray ptychography has been established as a valuable tool for high-resolution imaging. Nevertheless, the spatial resolution and sensitivity in coherent diffraction imaging are limited by the signal that is detected over noise and over background scattering. Especially, coherent imaging of weakly scattering specimens suffers from incoherent background that is generated by the interaction of the central beam with matter along its propagation path in particular close to and inside of the detector. Common countermeasures entail evacuated flight tubes or detector-side beamstops, which improve the experimental setup in terms of background reduction or better coverage of high dynamic range in the diffraction patterns. Here, we discuss an alternative approach: we combine two ptychographic scans with and without beamstop and reconstruct them simultaneously taking advantage of the complementary information contained in the two scans. We experimentally demonstrate the potential of this scheme for hard X-ray ptychography by imaging a weakly scattering object composed of catalytic nanoparticles and provide the analysis of the signal-to-background ratio in the diffraction patterns.

## 1. Introduction

X-ray imaging plays an important role in various fields of research such as materials science and chemistry. Detailed knowledge of the structure and morphology especially on the nanoscale is essential to understand and improve the functionality of, e.g., catalytic processes. Improving chemical reactions in terms of activity, selectivity, and lifetime has significant impact on the economy and the environment [1–3]. The function and efficiency of these processes strongly depend on the size, the shape, the oxidation state and the distribution of catalytic nanoparticles [4,5]. Since these nanoparticles typically range from one to several tens of nanometers in size [2], most high-resolution studies have conventionally been conducted using electron microscopy [6], including in situ studies [7–9].

Today, X-ray microscopy progresses towards similar spatial resolution and features valuable traits such as high penetration depth. This allows for the non-destructive investigation of bulk material and measurements in reactor cells [10] under realistic conditions in 3D. Notably, scanning coherent X-ray diffraction microscopy, also known as ptychography [11,12], has gained in importance. The technique is commonly used to image many kinds of specimens quantitatively with high spatial resolution and sensitivity well beyond the capabilities of conventional optics-based X-ray microscopes [11,13–15]. For ptychographic imaging, the specimen is scanned through the beam recording a far-field diffraction image of the scattered wave behind the specimen at each scan point. Due to an appropriate overlap between the illumination at adjacent scan points, a microscopic image in the form of the object's complex transmission function can be retrieved computationally from the ensemble of diffraction patterns using an iterative reconstruction process. Furthermore, by varying the X-ray energy around an absorption edge, ptychography allows for chemical imaging, determining the local chemical state of the given element [16–18]. Additional tomographic post-processing reveals the three-dimensional structure [19].

* Corresponding author.
*E-mail address:* juliane.reinhardt@desy.de (J. Reinhardt).

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In general, the spatial resolution and sensitivity of ptychography are limited by the coherent fluence on the sample [20], by the signal-to-background ratio in the acquired data, and the mechanical stability of the X-ray microscope [21]. With soft X-rays high spatial resolution on the nanometer scale has been demonstrated also with chemical contrast for low-Z elements in vacuum [22,23]. However, the application determines the photon energy. Accordingly, even higher penetration depths as well as the necessity to reach the absorption edges of other relevant catalyst materials such as Pd and Rh involve energies in the hard X-ray regime [24] where the photon–electron interaction is weaker. As a consequence, the diffracted signal may become difficult to detect over the background disturbances which are present in hard X-ray imaging setups that usually operate under ambient conditions. Whereas spatial resolution of less than 10 nm has been reported for strongly scattering samples even in the hard X-ray regime [14], imaging of weak objects is challenging [15]. For instance, catalytic nanoparticles in the range of 10 nm and smaller scatter three to five orders of magnitude less than the smallest features of a typical test pattern, e.g., the standard resolution chart manufactured by NTT-AT. In order to obtain higher signal from such weak objects, the fluence on the sample has to be increased by orders of magnitude for enhanced statistics and a clearer signal over noise. Usually, this is done by longer exposure or lowered requirements on coherence [25] that might lead to diminished reconstruction quality due to instabilities [26]. Moreover, background scattering from air molecules, other components such as windows or the detector material itself will increase concomitantly.

This work focuses on the improvement of the signal-to-background ratio in hard X-ray imaging by reducing the parasitic background signal. We present the approach of using an opaque beamstop that is placed far upstream of the detector in order to block the central beam as early as possible. In this way, the strongest source of parasitic scattering – the incoherent interaction of the intense central beam with matter – is eliminated. Consequently, the background is considerably reduced and the weak sample signal emerges clearly up to large angles in the diffraction patterns. Due to the absorption of the central beam, the low-frequency information in the diffraction patterns is lost and will be recovered by a second scan without beamstop.

In the next section, the experimental setup including the model sample and the data acquisition are briefly described. Following this, we show the result of the simultaneous reconstruction of two data sets – one with and one without beamstop – using the conventional ePIE algorithm with position correction. As supporting analysis, the signal-to-background ratio of the diffraction patterns is discussed afterwards. In conclusion, we demonstrate that a weakly scattering sample catalyst can be imaged with high resolution and a sensitivity increased by two orders of magnitude compared to previous examples [14,18] under ambient conditions using hard X-rays. The result can be directly applied to 3-dimensional data sets in prospective experiments.

## 2. Experiment

The experiment was carried out using the scanning microscope at the hard X-ray nanoprobe station at beamline P06 [27] of the synchrotron radiation source PETRA III at DESY in Hamburg, Germany. A scheme of the ptychographic setup with and without beamstop is shown in Fig. 1. At a photon energy of 11.919 keV, the sample was scanned through the coherent nanobeam produced by nano-focusing refractive X-ray lenses [28]. In the sample plane, the full-width-half-maximum lateral size of the amplitude distribution of the illuminating beam was about 140 nm with a flux of $4.4\times10^7$ Ph/s.

An ensemble of nanoparticles containing different catalyst materials on a 20 nm thick silicon nitride ($Si_3N_4$) membrane served as a well-defined but weakly scattering test object. To prepare this model sample, colloidal nanoparticles, i.e., platinum and palladium in acetone and gold nanoparticles in water, were mixed in a glass vial. The diameters of the particles range from less than 10 nm to about 100 nm. A single µL droplet of the mixture was deposited on a plasma-cleaned $Si_3N_4$ TEM chip (9 windows in total, eight windows of 100 µm × 100 µm and one of 100 µm × 350 µm in size). The plasma cleaning allowed for a fast spreading of the particles all over the TEM windows. The sample dried at room temperature under infrared light. Further details of the sample preparation are described by Hofmann [29]. Fig. 2a shows a scanning electron micrograph of the sample in backscattering contrast (SEM).

In order to acquire a quantitative high-resolution ptychogram of this sample, we recorded two ptychographic data sets, one conventional and one with a beamstop. For both data sets, the same sample area of 1 µm × 1 µm was scanned in 20 × 20 steps of 50 nm step size. At each position of the scan a far-field diffraction pattern was recorded with a *Pilatus 300k* pixel detector (DECTRIS, Baden, Switzerland) positioned 2080 mm behind the sample. This kind of single photon counting detector is notably suited due to the absence of readout noise and dark current. The ptychographic data set without beamstop (scan A) was acquired with a short exposure time of 0.3 s per scan point to collect the far-field diffraction pattern in the forward direction. The second ptychographic data set (scan B) was recorded with an exposure time of 3 s and using an opaque beamstop. This beamstop – made of steel, with a length of approximately 10 mm, and a diameter of 1.5 mm – was placed into the central beam about 500 mm upstream of the detector. In the last 500 mm of the air path near the detector, more than 90% of air scattering are generated. Therefore, with our approach, it is possible to suppress the major part of air scattering as well as background scattering by the detector material under feasible experimental conditions, i.e., a macroscopic beamstop which is almost scatterless and can be mounted with high stability. About $3\sigma$ of the approximately Gaussian shaped central beam were covered and the maximal count rate of the central beam was reduced by a factor of $10^5$. Accordingly, the beamstop can be considered as fully absorbing. By introducing the opaque beamstop into our setup, the relatively weak parts at larger scattering vectors $\vec{q}$ in the far-field diffraction patterns were recorded with low background noise.

## 3. Ptychographic reconstruction

The 441 diffraction patterns of the scan without beamstop (scan A) were reconstructed using an own implementation of the ePIE [12] algorithm. For the initial guess, a flat object function and a Gaussian shaped illumination were applied. The diffraction patterns were cropped to 256×256 px, resulting in a pixel size of 4.9 nm in the reconstructed images. The result for the phase reconstruction after 1000 iterations is shown in Fig. 3A. The main structure of the sample is reconstructed properly and single spherical particles down to a size of about 40 nm in diameter can be resolved individually.

For the ptychogram shown in Fig. 3A+B, scans A and B were jointly reconstructed, by using the conventional ePIE algorithm that was extended to correct for slight positioning inaccuracies and drifts between the two scans [30]. Identical to the reconstruction of the scan without beamstop, the diffraction patterns were cropped to 256×256 px and a flat object function and a Gaussian shaped illumination were applied as the initial guess. Both data sets were processed simultaneously, refining a shared object and a shared illumination function. While scan A (without beamstop) reliably provides the low-$q$ information in the diffraction patterns – albeit with a lower signal yield for higher angles – the diffraction patterns in scan B (with beamstop) provide the high-$q$ information, however, they lack the low-$q$ data behind the beamstop. Therefore, complementary masks were created: in the diffraction patterns of scan B, the pixels behind and in the immediate vicinity of the beamstop were masked to be freely adjusted by the algorithm. A complementary mask was applied to scan A. In this way, low-$q$ information comes from scan A – assuring the correct phase shift offset – while the scattering information of smallest structures at higher $q$ is provided by scan B. As a result, the complex transmission function of the object is still quantitatively recovered, but with