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Imaging modalities at the Swedish Materials Science beamline at PETRA III

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Abstract. High-energy synchrotron radiation has been demonstrated to be a powerful tool for materials characterization. The development of novel methodologies is still ongoing, driven by major technological advances regarding the available source brilliance and efficient large area detectors. The Swedish Materials Science beamline at PETRA III is dedicated to materials characterization by high-energy X-rays and scheduled to enter into user operation starting August 2019. The beamline has been designed in particular for the combination of two complementary techniques: wide and small angle scattering and imaging. The beamline design is presented briefly and the different techniques are reviewed with regard to the contrast mechanisms and the ability to obtain spatially resolved information.

1. Introduction

High-energy synchrotron radiation fills the important spatial and temporal resolution gap between electron microscopy and neutron diffraction, enabling the in situ investigation of bulk materials during e.g. thermo-mechanical processing. By combining different contrast mechanisms (e.g. attenuation, small and wide angle scattering) it has been demonstrated that important microstructure attributes such as crystallographic phase identification, orientation distributions, stress/strain state, size distributions and dislocation densities can be obtained. Initially, only averages over the illuminated sample volume were evaluated, but with the improving ability to provide intense micro-focused beams, various imaging methodologies have been developed exploiting tomographic reconstruction principles and/or scanning narrow illumination volumes across the sample. In some cases, area detectors can be employed for the simultaneous acquisition of scattering from many volume elements and therefore dramatically accelerate three-dimensional mapping as compared to point-by-point scanning. However, the applicability of specific methodologies is dependent on suitable microstructures and sample geometries.

PETRA III is currently the most brilliant high-energy X-ray source worldwide and the Swedish Materials Science beamline (SMS) has been designed in particular to perform multi-modal experiments providing spatial resolution. A number of techniques have been selected for implementation and in the following the status, underlying principles, potentials, and limitations are described briefly.

2. Beamline description

The SMS beamline comprises of a side branch (P21.1) dedicated for the study of diffuse scattering from single crystals, amorphous and nanocrystalline materials and a longer inline branch (P21.2) devoted to the combination of wide (WAXS) and small-angle X-ray scattering (SAXS) and imaging techniques with...
both broad and narrow bandwidth beams. In the following only techniques relevant to the characterization of polycrystalline bulk materials at P21.2 will be discussed.

P21.2 is served by an in-vacuum undulator, whose odd harmonics have a sharp central cone and are ideal for focusing and for small beam applications, while the donut shape of the slightly de-tuned even harmonics serve as a suitable source for large-beam experiments such as imaging.

A broad band double Laue monochromator, 100 meters downstream of the source, provides monochromatic ($\Delta E/E \approx 10^{-3}$) radiation ensuring high photon flux throughout the whole 40–150 keV energy range. Optionally, after collimation, a high resolution four bounce channel-cut monochromator can be used for experiments demanding very narrow $\Delta E/E \approx 10^{-4}$ energy distribution. The monochromatic beam may then be focused from several different locations with 1D or 2D compound refractive lenses (CRL) depending on the application. The largest unfocused beam size is approximately $2 \times 6 \text{ mm} \ (v \times h, \text{ FWHM})$ and the smallest achieved focus size (by CRLs) is about $1 \times 5 \mu\text{m}$ at the sample position.

3. Grain-resolved diffraction

Grain-resolved diffraction refers to situations where a sufficiently small number of grains (or subgrains) are illuminated by the incident beam, such that their diffraction spots do not extensively overlap on the detector [1]. The probability of spot overlap increases roughly with the square of the orientation spread (mosaicity) within the grains. Under favorable conditions, up to about one thousand grains can be illuminated simultaneously. Series of diffraction images are acquired while the sample is being rotated and the recorded reflections are assigned to individual grains by suitable software.

Three different methodologies can be delineated depending on the sample-to-detector distance (cf. figure 1a). If the sample-to-detector distance is comparable to the sample dimensions (‘near-field’), then the location of diffraction peaks on the detector is mostly dependent on the position of the diffracting units within the sample (at the cost of reduced strain sensitivity). By forward- or back-projection of the detected intensity distributions, crystallographic orientation maps of the illuminated sample volume can be evaluated (comparable to electron back-scatter diffraction surface orientation maps) [2]. The spatial resolution is limited by the detector resolution and typically about $2 \mu\text{m}$ can be achieved. The same detector design can be used for tomography, but thin scintillators are required resulting in poor detection efficiency and longer exposure times (see also section 4).

In the ‘far-field’ regime the sample-to-detector distance (typically about 1 m) is large compared to the sample diameter but small enough to still capture several complete diffraction rings on a large area detector. In general, no space constraints are posed for sample environments. Averaged quantities such as the position, size, crystallographic orientation, and the complete elastic strain tensor can be extracted. A test experiment$^1$ was performed on a well-annealed NiBi sample of about 1.1 mm diameter with approximately 40 $\mu\text{m}$ average grain size [3]. The full reconstructed volume with the 953 indexed nickel grains is shown in figure 1b. Using the full available beam intensity, a rotation speed of 25 $^\circ$/s would have been required to prevent detector saturation. Since the grain indexing is not yet limited by counting statistics even faster rotation should be feasible such that, in favorable conditions, one indexing scan could be accomplished per second. While the grain specific data enable the investigation of grain-to-grain interactions, it has also been demonstrated that by local averaging over an appropriate number of grains three-dimensional strain/stress mappings can be obtained [4]. If applicable, the grain-resolved technique is much faster than 3D-scanning of a local gauge volume as produced by a conical slit cell (cf. section 5.2).

The far-field detector setting can also be combined with a focused incident beam [5]. Spot overlap is minimized and intra-granular orientation variations can be resolved. However, a large number of images must be recorded (cf. section 5).

$^1$ The experimental details were as follows: beam energy: 59.87 keV, sample to detector distance: 984 mm, angular step size: 0.1$^\circ$, exposure time: 0.2 s, beam attenuation factor: 54, detector: Varex XRD 4343CT, pixel size 150 $\mu\text{m}$, number of pixels 2880$\times$2880
By increasing the sample-to-detector distance even further (typically about 4 m, ‘very-far-field’) the reciprocal space resolution can be increased such that the intrinsic peak profiles from metallic materials with low dislocation density can be characterized (at the cost of covering only small sections of a Debye–Scherrer ring) [6]. It is noted that the possibility of beam focusing with a very large focal length of about 40 m maintains the high reciprocal space resolution. The energy bandwidth can be cut down accordingly by the channel cut monochromator (cf. section 2).

4. Synchrotron micro-tomography

In synchrotron micro-tomography (SRµCT) a 2D projection image of the sample is acquired at each angular (ω) rotation-step, usually over a range of 180 degrees. The acquired projection images constitute the integral of the linear X-ray attenuation coefficient (µ) along the path of the X-rays. By utilizing either analytical or regularized iterative reconstruction methods, a 3D map of µ can be reconstructed for the entire illuminated volume. Transmitted X-rays undergo both absorption and phase shift, therefore the reconstructed µ is a combination of absorption, phase shift and their mixture. At small sample-to-detector distances—typically in the order of centimeters—the reconstructed µ primarily consists of the absorption part, which is optimal for strongly absorbing samples (high-Z elements). At high energies, however, µ becomes small for most elements, therefore the absorption contrast gets weaker. By increasing the sample-to-detector distance, the phase contrast contribution becomes more pronounced, which is advantageous for the investigation of poorly absorbing or phase samples and interfaces. On the other hand, an untreated phase contribution smears out the resolution at sharp interfaces. By using phase retrieval algorithms (PhR) the absorption and phase contributions may be separated. One such algorithm is Paganin PhR [7], which suppresses the absorption contribution resulting in predominant phase contrast.

For materials science related research, the combination of SRµCT with other X-ray techniques e.g. WAXS, SAXS or grain-resolved diffraction is particularly promising for revealing crystallographic information about a sample. However, this versatility comes at a cost as the currently available diffractometer at P21.2 is primarily optimized for interface diffraction and has a slight run-out error upon rotation.

The run-out error was characterized\(^2\) with a precisely manufactured steel ball immersed in glue in a Kapton tube. The projected images of the steel ball were fitted and the center positions were then used to perform post-alignment of the individual images, thus producing optimal slice reconstructions. Since the run-out error proved to be reproducible, a correction in the TomoPy [8] reconstruction workflow has been

\(^2\) The scanning parameters were the following; energy: 46 keV, sample-to-detector distance: 300 mm, exposure time: 0.4 s, sample rotation: 180°, no. projections: 1800, effective pixel size: 0.72 μm.
implemented and can be applied to correct the diffractometer imperfections for other measurements.

In SRµCT a common choice of detector is a scintillator combined with a 45° mirror, an objective and a CCD or CMOS camera. This setup has the advantage that the scintillator is more or less independent from the other parts of the detector, therefore can be changed upon demand. This is important, as the particular type and thickness of the scintillator determines the resolution and the required measurement time. The thicker the scintillator, the better the detection efficiency, but on the other hand the larger the smearing effect in the scintillator material. The small source size of PETRA III and the large distance from the source at P21.2 results in a particularly small virtual source size, therefore within reasonable sample-to-detector distances the resolution is determined solely by the detector—scintillator and camera.\(^3\)

The effect of the sample-to-detector distance (determining the degree of phase contrast) and the comparison of the analytical GridRec [9] reconstruction with and without PhR is shown in figure 2. The GridRec reconstruction works well for the strongly absorbing steel ball (figure 2a), but due to the lack of absorption contrast it performs poorly for the glue (figure 2b). Phase effects—even reconstructed without PhR—reveal tiny details (inclusions) in the steel ball (figure 2c), but only slightly increase the reconstruction quality for the glue (figure 2d). Using PhR exposes details in the glue (figure 2f), but at the same time hides small details in the steel ball (compare figure 2c and figure 2e).

5. Pencil-beam techniques

SRµCT (section 4) provides information on the 3D structure of multiphase materials, but lacks crystallographic information. Grain-resolved diffraction (section 3) requires a limited number of illuminated grains (to avoid spot overlap on the detector), which is often exceeded for fine grained microstructures. In these cases, pencil-beam techniques (when the beam size is comparable to the characteristic length in the sample, e.g. grain size\(^4\)) provide a unique tool to obtain local crystallographic information.

This technique employs scanning of a micrometer-sized beam across the sample cross-section with a diffraction or scattering pattern recorded at each scan step. This results in local information with a spatial resolution of the step and beam size perpendicular to the beam direction. Thus, structural gradients can be investigated by scanning along a line (1D profile) or across an area (2D maps) as long as the material is homogeneous in at least one direction, which is then oriented parallel to the incident beam. Good examples for this kind of profiling are materials after any kind of surface treatment (mechanical, chemical or irradiation), resulting in a gradient structure from the surface towards the bulk of the sample.

\(^3\) With a 300\(\mu\)m horizontal source size and 150 m source-to-sample distance, the detector may be placed as far as 500 mm from the sample before a virtual 1 \(\mu\)m resolution is worsened by source size smearing.

\(^4\) For WAXS CT the beam size should be significantly larger than the grain size such that the reconstructed voxels still contain a statistically relevant number of grains.
5.1. Pencil-beam tomography

Many materials are heterogeneous in all three spatial directions and profiling results in a diffraction signal, averaged along the incident beam direction. In this case, the combination of pencil beam mapping at various $\omega$ angles (cf. figure 1a) and tomographic reconstruction provides diffraction information in all three (spatial) dimensions. This method was proposed already in the 1980s [10], but reasonable measurement times and spatial resolution have only been achieved using the brilliant beams of third generation synchrotrons and focusing optics.

Compared to absorption tomography, a diffraction / scattering computed tomography dataset does not consist of a stack of 2D projections recorded at different rotation angles, but rather of a stack of grids of 2D diffraction / scattering patterns. Thus, large number of patterns has to be recorded and evaluated requiring high photon flux and automated data processing to obtain reasonable measurement times and analysis efforts. Furthermore, additional steps are required in the reconstruction process compared to absorption tomography. In most studies, each 2D pattern is integrated over the azimuthal angle to obtain 1D profiles (intensity as a function of the scattering vector / scattering angle), which is, however, only possible for rotationally symmetric diffraction / scattering patterns [11]. From these profiles, the intensity for each scattering vector bin is reconstructed using well-known tomographic reconstruction techniques (cf. section 4). Compared to absorption tomography, the computing time is multiplied by the number of scattering vector bins for reconstruction of the complete diffraction / scattering patterns. For this reason, diffraction studies often focus on the reconstruction of integrated intensities of a few peaks, indicating for example the spatial distribution of certain phases [12, 13].

Pencil-beam tomography can be used both with WAXS and with SAXS. While the actual measurements (except the larger sample-to-detector distance for SAXS) and the reconstruction are very similar, structural information from different length scales is obtained. While the WAXS signal yields information on the atomic level such as crystallographic phases, lattice parameters and crystallographic defects (dislocations, stacking faults), SAXS probes particle sizes, shapes and orientations in the nano- to mesoscopic range (up to about 1 µm). By placing multiple detectors at different distances to the sample, simultaneous SAXS/WAXS measurements provide a characterization technique covering a broad length scale [14].

Diffraction computed tomography has been used to characterize a large number of different complex materials. For example, Rietveld analysis of reconstructed 1D diffraction patterns has been used to determine quantitative phase compositions [15], atomic bond lengths [16] or average crystallite sizes [17] for each voxel. This shows that many evaluation techniques, that are known from conventional diffraction analysis, can be utilized in combination with pencil-beam XRD-CT scans to obtain a comprehensive understanding of micro- and particularly nanostructures in 3D. Modern instruments provide full XRD-CT scans in significantly less than one hour per slice, enabling in-situ experiments [15, 17]. The beam size in such experiments is always a compromise between high spatial resolution (small beam size), manageable measurement time and sufficient number of crystallites in diffraction conditions (large beam), since smooth Debye–Scherrer rings are required for many analysis techniques (except single grain techniques, cf. section 3). Texture distorts the quantitative analysis of reconstructed diffraction patterns, but usually still permits the determination of qualitative phase distributions [18]. Spotty Debye–Scherrer rings due to a limited number of diffracting crystallites result in artifacts in the reconstructed patterns, since individual grains coming to diffraction condition have significant impact on the integrated pattern. Thus, the beam size should be large enough to interact with a sufficient number of crystallites. Distinct strong spots from few large grains or a second coarse-grained phase can be removed via filtering to avoid reconstruction artefacts [18].

P21.2 is well suited for pencil-beam tomography, since the typical beam size of a few microns to a few tens of microns can be easily achieved and adjusted by corresponding positioning of the CRLs. The interface diffractometer provides all degrees of freedom required for such measurements.
5.2. Post-specimen apertures
Whereas XRD-CT is a powerful tool as long as azimuthal integration of the 2D diffraction patterns is possible, it becomes limited as soon as gradients along the azimuth are present. The most prominent examples are texture and residual stresses. Thus, direct restriction of the gauge volume via post-specimen apertures is often more effective compared to tomographic techniques for such measurements, in particular if only 1D or 2D slices are of interest.

The most widely used type of post-specimen apertures are conical slit cells. They consist of a strong absorber plate with conical openings in the size of a few tens of microns [19]. Only diffracted beams from a certain position in the sample along the beam can pass the conical slit, whereas those from other positions are absorbed by the plate. Thus, the gauge volume is limited in beam direction. The length of the gauge volume depends on the diffraction angle and usually counts several hundred microns [20, 21]. If needed the gauge length can be reduced via sample translation and reconstruction techniques and is then only limited by the width of the post-aperture slit [22]. Conical slits have been used to investigate 3D spatial distributions of residual stresses and textures in various materials [19–21].

One disadvantage of conical slits is that a conical slit is limited to the investigation of a pre-defined crystallographic symmetry group due to the fixed distance of its openings. Alternative aperture designs, such as spiderweb [23] slits have been proposed, but are not yet regularly applied.

Measurements with post-specimen apertures have not been conducted at P21.2 yet. However, the beamline is generally well suited for such experiments, which will be tested in the near future.

6. Conclusion and outlook
It has been discussed that P21.2 is capable of using several multi-dimensional imaging techniques. The combination of these techniques enables the investigation of structural characteristics from the Angstrom regime up to the millimeter scale, with spatial resolution down to the micrometer range. Future developments include the installation of a second diffractometer, optimized for rapid and precise rotation. With the advancement of the beamline control system and the incorporation of new fast detectors a considerable increase of the measurement speed is expected.

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