X-Ray Imaging of Polycrystalline and Amorphous Materials

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A novel method for obtaining position resolved information simultaneously from an entire plane of a polycrystalline or amorphous sample using the diffracted radiation or fluorescence has been developed. It makes use of a microchannelplate as a collimator in front of a position sensitive detector (CCD-camera). Experiments may be performed either in transmission or in reflection geometry. In the first case a ‘flat’ parallel beam illuminates a slice through the sample while in the second case the surface of the specimen is illuminated by a broad beam.

Typical fields of application are non-destructive investigations of the distribution of polycrystalline components in composites, reciprocal space mapping to determine the mutual influence of strain and orientation in adjacent grains in a polycrystalline material and analysis of the distribution of chemical elements or even valence states via their fluorescence.

Introduction

Fluorescence or radiation diffracted by a polycrystalline specimen are not or only partially collimated. This and the lack of optical components for hard X-rays have restricted position resolved experiments to time consuming scanning techniques. Recently a novel method using a microchannelplate as collimator in front of an area detector has been applied for the imaging with diffracted radiation from a polycrystalline sample\(^1\) and subsequently with fluorescence radiation\(^2\). This technique allows simultaneous imaging of areas in the order of few cm\(^2\) with resolutions in the order of several ten of microns. Depending on the sample and the properties to be investigated exposure times reach from some ten minutes (weak fluorescence) to less than a second (strong reflections). In the latter case the measuring time is dominated by the readout time of the detector and the time required to store the image. This allows the collection of a series of images each taken with different experimental parameters resulting in ‘spectra’ depending on one or more variables for each detector element. Characteristics of these spectra can be evaluated and presented in the form of ‘maps’.

Experiment

All measurement were made at the Kappa diffractometer at HASYLAB beamline F1 (bending magnet). The experimental set-up consisted essentially of a microchannelplate as collimator in front of a CCD camera as area detector. The microchannelplate used has a thickness of 4 mm and an active area of about 19 mm in diameter which is covered with channels of 10 micron diameter having a centre to centre distance of 12.5 micron. The angular acceptance is 2.5 mrad (0.14\(^\circ\)) resulting in a spatial resolution of 25 microns for a sample to detector distance of 10 mm. The CCD camera (HAMAMATSU C4880) has 1024\(^2\) pixels of 12 microns size resulting in a field of view of 12 mm times 12 mm. The readout of the camera could be triggered via the controls of the diffractometer allowing automated measurements.

The collimator-detector assembly may either be positioned at a 2-Theta angle corresponding to a Bragg-peak for diffraction experiments or off-peak for the measurement of fluorescence. Although this scheme is not dependent on a particular primary source, the use of synchrotron radiation is recommended both for diffraction because its high collimation well defines the angle of incidence and for fluorescence because of its tunability. This tunability not only allows the selective excitation of elements but also of different chemical states of one element (see fig.2) if a narrow band monochromator like perfect silicon crystals is used. Using a broad band monochromator like pyrolythic graphite\(^3\) or synthetic multilayers the incoming intensity may be increased by about two orders of magnitude at the expense of
energy resolution. This is still sufficient to discriminate between elements but not between their chemical states. Diffraction may also profit from broad band monochromators close to the minimum of the resolution function' and if the reflections are wide enough separated.

Exposures, images and Maps

Although even a single exposure can give valuable information about the distribution of an element (fluorescence) or the distribution and orientation of one compound in a polycrystalline sample (diffraction) one is often interested in the variation of these properties as function of experimental parameters. An example of such a scan is shown in figure 1.

Fig. 1:
3D reconstruction of the graphite distribution in a SiC-C composite. The volume shown is about 4*3*3 mm³. 34 exposures were taken in the xy-plane. The slice shows the intensity distribution in the yz-plane calculated from these exposures.

The sample was a carbon fibre reinforced SiC composite which was illuminated with a ‘flat’ beam (0.1 mm high) of 0.1 nm wavelength from a graphite double crystal monochromator. These parameters were chosen because even with a narrow band monochromator the graphite reflection were rather broad (about 2”) but the 002 reflection recorded in this experiment was still more than 5” separated from the next SiC peak. Furthermore high intensity was required because of absorption losses in this transmission geometry in which the beam illuminated a slice through the sample. Between two exposures the sample was moved 0.1 mm through the beam. Due to mechanical hindrance the minimal sample to detector distance was 50 mm limiting the spatial resolution.

Generally a series of exposures as function of external parameters delivers a spectrum for each pixel amounting to 1 million spectra in total. Automated data reduction is therefore of paramount importance. Usually one is not interested in the whole spectrum but only in few properties like the position and width of a peak. Figure 2 shows 8 exposures from an energy scan. The sample contained 3 different substances namely Fayalit (Fe₃SiO₄) and two glasses containing iron (CaFe₀.2₅Al₁₇.₅Si₆O₁₆ and KFeSi₃O₈). The whole data set consisted of 51 exposures taken around the Fe K-edge at energies from 7000 eV to 7050 eV in steps of 1 eV.
Fig. 2: Exposures from an energy scan at the Fe K-edge. The sample contains pieces of Fayalit (left), CaFe_{26}Al_{75}Si_{2}O_{8} (middle) and KFeSi_{10}O_{8} (right). The energies for the left series are 7000 eV, 7005 eV, 7010 eV, and 7015 eV and for the right series 7020 eV, 7025 eV, 7030 eV and 7035 eV. One can clearly distinguish between the Fe\(^{3+}\) of Fayalit and the Fe\(^{2+}\) of the right sample. The sample in the middle (nominal Fe\(^{2+}\)) shows an early onset of fluorescence and a small slope of the absorption edge. Additional fluorescence arises from the epoxy in which the samples were embedded. The area of each exposure is 12\(^{\times}\)12 mm\(^2\).

Fig. 3: Map of the positions of the absorption edges obtained from fluorescence images like in figure 2. The corresponding scale is shown below. It reaches from 7010 eV to 7030 eV (red)
Figure 3 shows a map of the sample in which the different colors indicate the energy of the absorption edge given by the maximum of the first derivative of the intensity of the fluorescence. Establishing this map revealed a problem associated with such measurements. Due to the limited angular acceptance of the microchannelplate only a small fraction of the (isotropic) fluorescence is detected leading to long exposure times and noisy data. This leads to difficulties in automated data reduction. Simple criteria like extrema of the data or their derivatives may yield wrong values even after smoothing or median filtering. More sophisticated routines like fitting with a function (gaussian) show numerical instabilities causing a breakdown of the program. To reduce these instabilities regions in which no change in the signal occurs are eliminated by thresholding (black regions in figure 3). But even then stability could only be obtained either by averaging over several pixels on the costs of spatial resolution or by using such a high threshold that great regions (especially in the central region of the sample where both the signal and its variation with energy are low) were excluded.

Recently a novel algorithm has been developed by Z. K. Silagadze. This ‘quantum mechanical’ algorithm suppresses the noisy features (readings) of the spectra and simultaneously amplifies the underlying characteristics (wave function). Using this algorithm before searching for the edge position the accuracy could tremendously be increased without degradation of the spatial resolution.

Fig. 4:
Exposures in the 200 reflection of β-brass for 3 different sample orientations (separated by 0.6°). For the entire data set 21 omega scans (step width 0.2°) were performed between which 2-Theta was changed in steps of 0.05° (41 times) to obtain reciprocal space maps. The area shown here is 0.96 × 0.96 mm².

Diffraction experiments normally do not suffer from low intensity because most of the intensity falling on a crystallite is reflected if it has the proper orientation. Figure 4 shows a part of three exposures taken at a sample of p-brass (200 reflection) at different sample orientations. Each exposure took less than a second so that a reciprocal space map with 21 (Omega) times 41(2-Theta) exposures could be collected in less than three hours. Readout and storage of the images dominated the collection time. By rearrangement of the data reciprocal spcemap for each data point could be obtained. (In this measurement superpixels composed out of 2×2 detector elements were read out. resulting in 5 ×5 ×12 data-sets.) As in the example above regions in which peak fitting should be performed were selected by thresholding. In this case no problems arose during fitting with a 2d-gaussian. The maps in figure 5 show the peak position in omega (lower) and 2-Theta (higher). They indicate that the grain consists out of several subgrains (which can already be seen from figure 4) and that there is a strain gradient.

All data processing was done with programs based on the IDL® programming language which already contained many routines for fitting and image processing. The possibility to associate
array structures with disk files was essential for the processing of these data sets, which amounted to 20 MB for the diffraction on the SiC-C composite, 100 MB for the fluorescence scan and 450 MB for the reciprocal space mapping.

Conclusion
Imaging using fluorescence or radiation diffracted by a polycrystalline sample yields within a short time an enormous amount of information about an inhomogeneous specimen. Methods to reduce this huge amount of data have been developed and yield maps of the relevant sample properties. It should be mentioned that all experiments have been performed using radiation from a bending magnet. Radiation from an insertion device (wiggler, undulator) will further improve the data quality especially for near edge fluorescence. Within the frame of this article only few selected examples could be presented which nevertheless show the potential of this novel method in many branches of material science.

Fig. 5:
Maps of the peak positions in omega (upper) and 2-Theta (lower figure) obtained from 21 (omega) times 41 (2-Theta) exposures like those shown in figure 4. It can clearly be seen that there is a continuous variation in the orientation of the lattice planes. From the lower map the strain distribution over the grain can be deduced.

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