Energy dispersive X-ray diffraction imaging

Jörn Donges[1, a], Andre Rothkirch[1,b], Thomas Wroblewski[1,c], Aniouar Bjeoumikhov[2,d],Oliver Scharf[3,e], Ivan Ordavo[4,5,f] and Sebastian Ihle[5,g]

[1]DESY-FS, Notkestr.85, 22607 Hamburg, Germany

[2]IfG, Rudower Chaussee 29/31, 12489 Berlin, Germany

[3]IAP, Rudower Chaussee 29/31, 12489 Berlin, Germany

[4]PNDetector GmbH, Emil-Nolde Str.10, 81735 München, Germany

[5]PNSensor GmbH, Römerstr. 28, 80803 München, Germany

[a]joern.donges@desy.de, [b]andre.rothkirch@desy.de, [c]thomas.wroblewski@desy.de, [d]bjeoumikhov@ifg-adlershof.de, [e]scharf@ifg-adlershof.de, [f]ivan.ordavo@pndetector.de, [g]sebastian.ihle@pnsensor.de

Keywords: Energy dispersive diffraction, X-ray imaging, X-ray camera, pnCCD

Abstract. Position resolved structural information from polycrystalline materials is usually obtained via micro beam techniques illuminating only a single spot of the specimen. Multiplexing in reciprocal space is achieved either by the use of an area detector or an energy dispersive device. Alternatively spatial information may be obtained simultaneously from a large part of the sample by using an array of parallel collimators between the sample and a position sensitive detector which suppresses crossfire of radiation scattered at different positions in the sample. With the introduction of an X-ray camera based on an energy resolving area detector (pnCCD) we could combine this with multiplexing in reciprocal space.

Introduction

X-ray diffraction imaging on polycrystalline materials [1] applies a collimator array in front of an area detector. These collimators suppress crossfire of radiation scattered from different locations of the specimen. A dedicated instrument using a micro channel plate in front of a CCD detector has been realized at HASYLAB beam line G3 [2] usually operating at energies up to about 11 keV. Further studies have been performed using long (300 mm) polycapillaries for X-ray bulk diffraction imaging [3] or arrays of boron silicate glass capillaries for neutron (bulk) diffraction imaging [4]. In these cases a flat beam has been applied to illuminate a slice through the specimen while with low energy X-rays mainly surface near regions have been probed with an extended beam.

X-ray diffraction imaging has been applied to investigate the spatial distribution not only of different phases (in this case also the fluorescence has been used [5]) but also of strain and (preferred) orientation. Furthermore the method is ideally suited for kinetic studies like recrystallization [2,6] (in such cases no prior knowledge of where an event occurs exist). In the framework of a study on the recrystallization of tungsten in the filament of an electric light bulb in cooperation with OSRAM [7] strong preferred orientation has been observed. To extend the measurement to manifold lattice planes while preserving the spatial information, it was favorable to maintain the geometry of the experiment (arrangement of incoming beam, sample and detector arrangement) which was achieved by changing just the wavelength. Exposures were taken at different wavelengths chosen according to the Bragg condition of the corresponding lattice planes. This can be regarded as first energy dispersive diffraction imaging.

Position resolved energy dispersive diffraction using a pencil beam (named energy dispersive X-ray diffraction tomography [8] was first performed on X-ray tubes with voxel sizes in the order of mm^3. With synchrotron radiation voxel dimensions in the order of some $(10\ \mu m)^3$ became accessible [9]. A drawback of this method was the time consuming scanning procedure leading to efforts to

combine collimator arrays with multi element energy sensitive detectors [10]. These efforts however suffered from rather low spatial resolution (300µm) and low transparency (~2%) of the collimator array.

Experiment

Capillary arrays have transparencies in the order of 75%. Such an array in combination with a pnCCD [11] energy resolving area detector, has already been used for full-field fluorescence imaging [12]. The detector used had an area of 12.7*12.7 mm² comprising 264*264 pixel of 48*48 µm² and could be read out with a frame rate of 400 Hz. Every pixel acts as individual energy dispersive point detector. However the charge cloud generated by a single event is smeared out over several pixels and the photon energy has to be determined from the sum of the charges collected in the connected pixels. The position of the event is given by the center of gravity of these pixels. For this reason the number of correctly recordable photons per readout is much less than the number of pixels. Further details of the detector including achievable count rate and the capillary optics are given in [13] and [12] respectively and will not be repeated here. The experiments were performed using the white beam at HASYLAB station F3 at the DORIS storage ring which is usually used for conventional energy dispersive diffraction [14]. Because of the horizontal polarization the capillary/detector combination was positioned at 90° in the vertical scattering plane.

The main motivation of this study was to obtain information from manifold lattice planes in a single shot. In the study of the filament mentioned in the introduction time consuming energy changes which then would have become unnecessary. Furthermore a simultaneous measurement would have enabled the continuous observation of dynamic processes like recrystallization. The complete information from all lattice planes would have given valuable insight into the dynamics of the process.

Fig. 1: Set-up for energy dispersive diffraction imaging on frost in transmission mode. The beam enters from the left through a slit system. The capillary optics is located in the cylinder above the specimen which is produced by exposing a two stage Peltier cooler to air.

However, the detector was not optimized for the intensities obtained with the full (6mm*6mm) unattenuated beam. The reason is that in contrast to the fluorescence experiment where the radiation from the specimen is isotropic the diffracted radiation from a crystallite is well collimated leading to a high local intensity on the detector. The energy resolution capability of the detector requires that the patterns generated by each single incoming photon in a frame must be separated. Therefore the incoming intensity had to be reduced. This was achieved by attenuators and/or by using a smaller beam.

Such small beams are needed in the transmission mode where a slice through the specimen is illuminated by a flat beam. Furthermore the sample itself functions as attenuator. Nevertheless the studies were restricted to materials composed of light elements because the detector is most efficient for energies below 25 keV.

Fig. 1 shows the setup for our studies on ice. For this purpose a Peltier cooler was installed below the beam. With this arrangement even dynamic studies like the resublimation of ice became possible. The growth of ice crystallites from the humidity of the air through the beam was monitored just by turning on the cooler. Unfortunately no continuous nucleation/growth curves could be observed, because Bragg reflections of crystallites disappeared again which is probably due to tilt and/or rotation of the crystallites during the growth of the frost layer. Crystallization studies from the liquid phase gave no usable nucleation/growth curves due to the high speed of crystallization and the low number of (coarse) grains fulfilling the Bragg conditions.

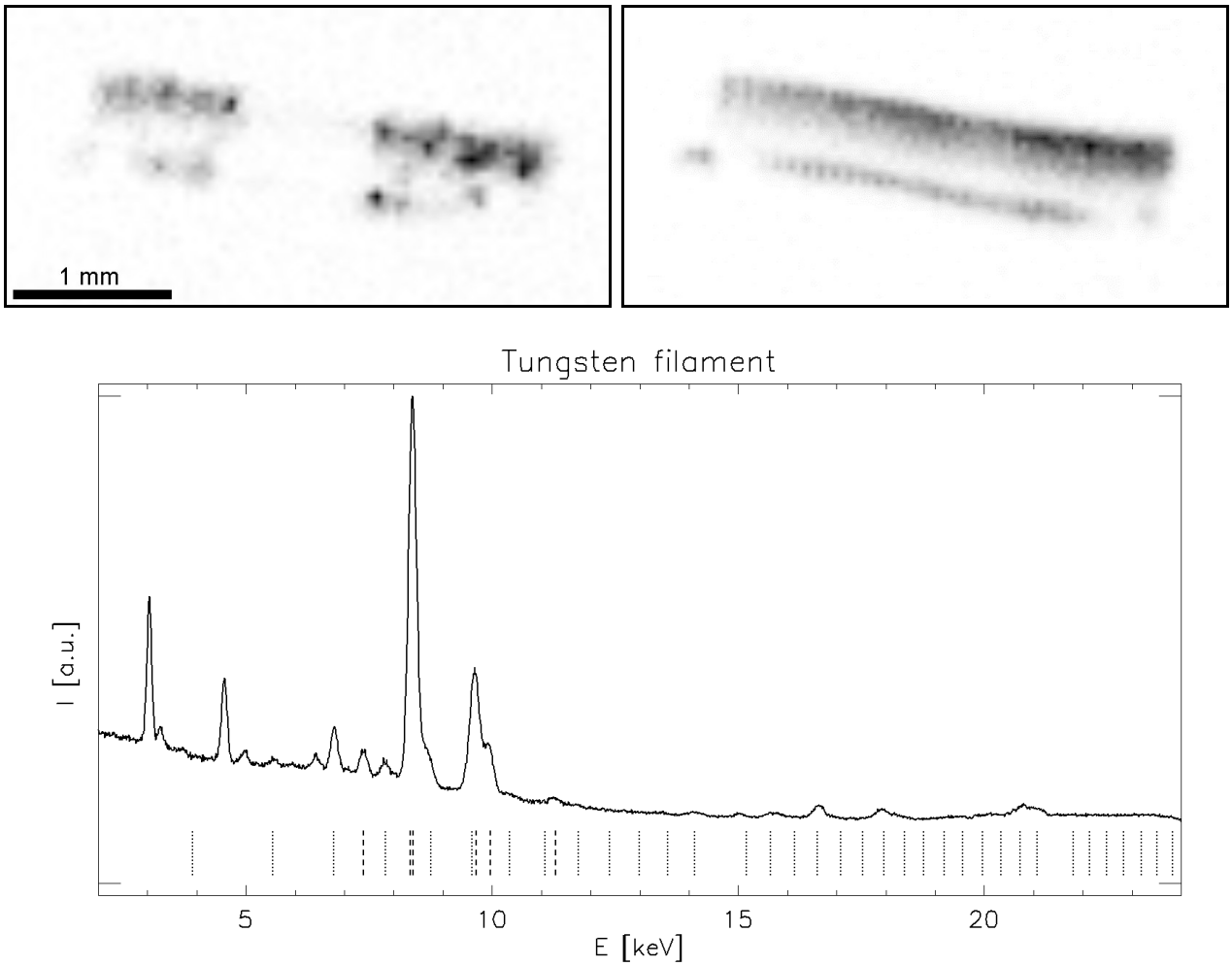

Fig. 2: Monochromatic images from a tungsten filament extracted from an X-ray colour data set. The left image at energies around 6.8 keV corresponds to the 211 reflection while the right one at energies around 9.7 keV shows mainly the Lß fluorescence. The energy spectrum averaged over the entire data set is shown below

Investigations in reflection mode were made on the tungsten filament of an electric light bulb (Kess halogen-lamp 10W). Fig. 2 shows fluorescence and Bragg reflections from the filament. While the W-fluorescence remains constant along the filament (the intensity gradient is due to the nonhomogeneous entrance slit not designed for such small sizes) the diffraction signal is higher at the ends of the filament compared to its central part which may be due to higher crystallinity.

Due to the limited count rate of the detector in single photon counting mode the picture could not be obtained in a single shot with a beam illuminating the entire specimen but a ~50μm high beam had to be used instead. The sample was then scanned vertically through the beam while the intensity in the detector was accumulated.

Summary

Energy dispersive X-ray imaging has been performed using a capillary array collimator in combination with an X-ray camera based on an energy- and position-sensitive pnCCD detector. Exposure times were in the order of minutes. To correctly reconstruct the energy of every incoming photon, the detector has to be operated in single photon counting mode. In this mode the probability of two or more photons to generate overlapping patterns must be kept small. For this reason the incoming beam has to be attenuated and only a fraction of the available intensity is used. At this reduced exposure level, times in the order of minutes were necessary to achieve reasonable statistics. Therefore the experiments presented here can be considered first proof of principle investigations of the capabilities offered by this kind of detector. However, it may turn out that the setup is well suited for measurements at conventional X-ray sources. Such studies are in preparation.

The final goal is however to use the entire intensity of the white spectrum of synchrotron radiation and the full size of the beam to image large areas of polycrystalline materials. In this case imaging of the tungsten filament shown in Fig. 2 could have been achieved in a single shot yielding simultaneous information about all crystallographic lattice planes. In the present situation pseudo-polycromatic imaging as described in [7] where several monochromatic images taken at different energies are combined is more efficient. This pseudo monochromatic approach is, however, not suited for the continuous observation of dynamic processes like (re)crystallization. Only in a limited number of cases the time scale of such processes can be modified by external parameters like temperature. It would, however, be of great value if the observations were not restricted to only one Bragg plane.

Of paramount importance is the increase in count rate but also improvements in spatial resolution and sensitivity at higher X-ray energies are desirable. . In this regard it must be noticed that preliminary studies at higher frame rates (up to 1000 Hz) than that used for the experiments (400 Hz) are ongoing and show encouraging results. Moreover the spatial resolution is not limited by the pixel size, but actually by the optics used. Spatial resolution below the pixel size can be achieved experimentally by means of more intelligent pattern reconstruction algorithms in combination with smaller polycapillary channels [15]. These improvements are still subject of investigations but they could be applied in the next run of experiments.

References

[1] T. Wroblewski, et al., X-Ray imaging of polycrystalline materials, Rev. Sci. Instrum. Vol. 66 (1995), 3560-2562

[2] T.Wroblewski , et al., A new diffractometer for materials science and imaging at HASYLAB beamline G3, Nucl. Instrum. Methods A 428 (1999) 570-582

[3] T.Wroblewski, A. Bjeoumikhov, X-ray diffraction imaging of bulk polycrystalline materials, Nucl. Instrum. Meth. A 538 (2005) 771-777

[4] T.Wroblewski, E. Jansen, W. Schäfer, R. Skowronek, Neutron imaging of bulk polycrystalline materials, Nucl. Instrum. Meth. A 423 (1999) 428-434

[5] T. Wroblewski, X-ray imaging of polycrystalline and amorphous materials, Advances in X-ray analysis 40, CD-ROM

[6] T.Wroblewski, Self-organized criticality – a model for recrystallization?, Zeitschrift für Metallkunde, 93 (2002) 1228-1232

[7] J. Almanstötter, P. Schade, D. Stein, T. Wroblewski, Diffraction imaging of recrystallization in tungsten wire coils for incandescent lamps, HASYLAB Annual Report 1999, 891-892

[8] G. Harding, M. Newton, J. Kosanetzky, Energy-dispersive X-ray diffraction tomography, Phys. Med. Biol. 35 (1990), 33-41

[9] C. Hall et al., Synchrotron energy-dispersive X-ray diffraction tomography, Nucl. Instrum. Meth. B 140 (1998) 253-257

[10] C.C.T. Hansson, K.H. Khor, R.J. Cernik, Coherent imaging using diffracted X-rays, Crystallography Reports, 55 (2010) 1162-1173

[11] L. Strüder, High-resolution imaging X-ray spectrometers, Nucl. Instrum. Meth. A454 (2000) 73-113

[12] O. Scharf, et al., Compact pnCCD-based X-ray camera with high spatial and energy resolution: A colour X-ray camera, Analytical Chemistry 83(7) (2011) 2532-2538

[13] W. Leitenberger, R. Hartmann, U. Pietsch, R. Andritschke, I. Starke, L. Strüder, Application of a pnCCD in X-ray diffraction: a three dimensional X-ray detector, J. Synchrotron Rad. 15 (2008) 449-457

[14] J. Staun Olsen, B. Buras, L. Gerward and S. Steenstrup, A spectrometer for X-ray energy dispersive diffraction using synchrotron radiation, J. Phys. E: Sci. Instrum. **14** (1981) 1154-1158

[15] I. Ordavo et al, A new pnCCD-based color X-ray camera for fast spatial and energy-resolved measurements, Nucl. Instrum. Meth. A 654(1) (2011) 250-257