MI-506

Development of compound refractive lenses for hard x-ray full field microscopy, magnifying high resolution microtomography, and fluorescence microtomography

Final report

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1 Introduction

The aim of the project was to built and characterize parabolic refractive x-ray lenses made of different materials to improve their performance and to further develop experimental techniques based on them, such as full field imaging and microscopy, magnifying tomography, and fluorescence microtomography. Four experimental sessions were scheduled at ID22 during the term of the proposal. Table 1 show the details of the experimental sessions.

All major aims of the long term project were successfully addressed during

beamline	shifts	start date	Topic
ID22	15	27.06.2001	full field imaging and magnifying tomography
ID22	16	21.11.2001	first test of Be lens, demagnifying full field imaging, x-ray lithography tests.
ID22	15	01.05.2002	optical tests of Be lenses, full field imaging, microbeam production.
ID22	15	18.09.2002	optical tests of Si nanofocusing lenses, sub-micrometer fluorescence microtomography.

Table 1: Experimental sessions

the project. In addition, several new developments were made beyond the proposed goals.

During the proposal period we were able to advance the development of parabolic refractive lenses for different applications. One major breakthrough was the fabrication of beryllium lenses of high quality that made hard x-ray microcopy with about 100nm resolution possible [1, 2]. In addition, nanofocusing lenses were developed that can generate a microbeam in the range of hundred nanometers even at a short distance (i. e., 41m) from a (large) high-β source [3]. In addition, aluminium lenses were perfected and made available in large numbers such as to allow focusing of high energy x-rays [4] (up to 120keV). The proposed experimental methods were investigated during the experimental sessions. Magnifying tomography with Al lenses was demonstrated to yield 3D reconstructions with unprecedented resolution [in the hard x-ray range ($E = 25$keV)] of about 400nm [5]. By the second experimental session, the first Be lenses became available and were tested, while exploring the possibility of demagnifying hard x-ray lithography using refractive optics [6]. The third experimental session was dedicated to the characterization of Be lenses for both imaging and microbeam applications. The last experimental session was used to characterize the newly developed nanofocusing lenses (NFLs) [3] and to apply them to fluorescence microtomography. By combining Be lenses and NFLs, it was possible to acquire fluorescence tomograms with the unprecedented pixel size of 600nm. These results will be detailed in the following sections. Detailed reports on the experimental sessions are additionally available.

Six publications (two letters) have resulted from the experiments carried out during this project [6, 7, 5, 2, 3, 8]. [5] was selected to be published in [9], and [3] was editor's choice in *Science* **299** (5616), 1613 (2003). There are four publications in preparation that are a direct result of or based on results in this project [1, 10, 11, 12]. Two doctoral theses resulted from this work [13, 14] and three doctoral students will use parts of the results in their upcoming theses.

The lenses developed during this proposal have been made available to different beamlines of the ESRF, including ID11, ID13, ID15, ID18F, and ID22 and were used in many user experiments.

2 Development of lenses

In this section we describe the development of refractive lenses. Their application during the experimental sessions will be treated in the following sections.

Parabolic refractive x-ray lenses made of aluminium were available at the beginning of the project. They were improved further in view of their optical quality and used for the magnifying tomography and lithography experiments. In addition, they were used for high energy focusing at ID15, demonstrating their applicability at x-ray energies up to 120keV. They were applied for high energy focusing in user experiments [4]. By now, they can be made in large numbers and are made available to synchrotron radiation sources. The ESRF has acquired 1000 Al lenses and mountings since the beginning of the project for use at various beamlines. Al lenses are used in several user experiments at different ESRF beamlines.

Parabolic refractive x-ray lenses made of beryllium were developed during the project. They were used in three of the four experimental sessions. Their imaging properties were tested during the second and third experimental session. Microbeams generated by Be lenses were characterized during experimental sessions three and four. These lenses are by far more difficult to fabricate. Therefore, it is currently not possible to fabricate as many of them as those made of aluminium. However, a first set of six Be lenses was installed at ID13 for high resolution micro-SAXS experiments and a few user experiments were carried out with this setup. In addition, another 60 Be lenses will be delivered this year, 30 of which are planned to be used at the new protein crystallography beamline ID23.

Lenses made of Ni were successfully fabricated and tested for high energy applications. However, their efficiency is inferior to those made of aluminium up to 120 keV. In addition, they are more difficult to fabricate in large numbers. Therefore, we have reduced their production in favor of Al lenses.

Recently, we have developed parabolic refractive lenses with focal distances in the range of 10mm in the hard x-ray range (e. g., at $E = 25$keV). These nanofocusing lenses (NFLs) [3, 11] allow one to strongly demagnify an x-ray source onto the sample even at short distances (i. e., 41m) from a (large) high-β source (i. e., source size $900 \times 60\mu$m^2 at ID22) and thus generate also horizontally sub-micrometer foci in the first experiments hutch. In the fourth experimental session, we generated a microbeam with 380nm by 210nm lateral size at 41m from the undulator source at ID22. To reach these small foci, a demagnification of over 2000 is required that can not be reached by any other optic in the hard x-ray range. In addition, these lenses have potentially diffraction limits below 20nm and may be useful for microbeam experiments with highest spatial resolution. During a follow-up experiment (MI-649) we were able to generate a microbeam of $330 \times 110 nm^2$ in the first experiments hutch of ID22. The horizontal beam size was limited by the geometric demagnification of the large horizontal source. At low-β sections, the limitations on the horizontal beam size due to geometric optics are overcome and microbeams with a horizontal size below 100nm should be possible even at short distances < 50m from the source. We submitted a long term proposal for the development of a 100nm by 100nm microprobe for fluorescence microtomography together with the team of ID13. NFLs have first been made by e-beam lithography and subsequent reactive ion etching in silicon. Since the optical properties of NFLs made of boron, diamond and graphite are superior to those made of silicon, we currently develop the fabrication techniques for these materials. The first boron lens was successfully fabricated and tested during MI-649.

3 X-ray full field microscopy

To overcome the limitations of the spatial resolution in hard x-ray full field imaging, an object can be imaged with hard x-rays onto a high-resolution x-ray detector using a parabolic refractive lens. We demonstrated earlier, that x-ray microscopy with about 350nm resolution was possible using aluminium lenses [15].

During the first experimental session, we used optimized aluminium lenses for magnified imaging. It could be demonstrated, that imaging free of distortion can be achieved. Comparison with numerical models (of ideal lenses) showed good agreement even in details of contrast formation [16, 7].

In the third experimental session the imaging quality of Be lenses was characterized in detail. As the ESRF was unable to provide a high resolution camera of sufficient quality, all images had to be recorded on x-ray film. This significantly reduced the number of images taken (focusing sequences had to be evaluated after developing the film) and slowed down the process of evaluation, since the film needs to be digitized with high resolution and best possible dynamic range. Therefore, publication of some results has been delayed. However, we were able to demonstrate hard x-ray microscopy based on beryllium lenses with about 100nm resolution in a field of view of over 450μm (FWHM).[1] These results will be published soon [1].

Besides for magnifying imaging, refractive optics can be used to reduce the image of an x-ray mask onto an x-ray sensitive resist for lithography applications. This alleviates the requirements on the feature sizes in the x-ray mask to generate small lateral feature sizes in the resist. The main application of this technique would be in deep x-ray lithography, e. g., for LIGA. Together with S. Achenbach from the IMT in Karlsruhe, Germany, we assessed the deep x-ray lithography applications by reducing an x-ray mask

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[1] The gaussian aperture of the lenses leads to a gaussian intensity distribution in the field of view.

onto a resist covered substrate using aluminium refractive lenses during the second experimental session (for details, see report for that session).

For all full field imaging applications, the control of the lateral coherence of the illuminating radiation was crucial for high quality imaging. By using a diffuser (rotating slab of B_4C powder between two thin glass plates) the lateral coherence could be reduced. This reduces speckle in the images, increases the field of view and better matches the illumination to the aperture of the objective lens. For the lithography, a condenser lens made of Be (first Be lens) was used in addition.

4 Magnifying tomography

One of the key strength of hard x-rays is their large penetration depth in matter. This allows one to non-destructively image the interior of opaque objects. In combination with tomography, high resolution imaging and microscopy can be used to reconstruct the 3D inner structure of a sample.

During the first experimental session, we have combined magnifying imaging with tomography to increase the spatial resolution in 3D imaging over conventional projection imaging. During that session, we recorded a variety of magnified tomograms, in particular of a piece of an AMD K6 Microprocessor. Using this processor as a test sample, it was demonstrated, that using magnifying tomography the 3D structure of a sample can be reconstructed with sub-micrometer resolution (410nm) [5]. This result was selected for publication in the **Virtual Journal of Nanoscale Science & Technology** [9].

The resolution was mainly limited by the diffraction limit of the aluminium lenses. However, also the mechanics of the rotation stage were inadequate for this experiment. The rotation stage available has an eccentricity of about 1μm that results in reconstruction artifacts of 3μm in size. By numerically correcting for the eccentricity, the reconstruction to sub-micrometer resolution was possible.

5 Fluorescence tomography

Fluorescence microtomography requires a microbeam to scan the sample in translation and rotation, recording the fluorescence radiation at each point of the scan with an energy dispersive detector.

The microbeam generated by a Be lens was characterized in detail during the third experimental session. As the microbeam size is dominated by geometric optics for the given source size and distance from the source at ID22, the microbeam sizes obtained were similar to those previously obtained with Al lenses. However, microbeams could be generated at lower energies (down to 8keV) and with significantly higher flux ($\approx 10^{11}$ph/s) [2].

Using NFLs, microbeams significantly smaller than those achievable at ID22 by other optics were generated during the fourth experimental session [3]. The flux in this microbeam was slightly above 10^8ph/s.

In the fourth experimental session the aim was to acquire fluorescence microtomograms with highest spatial resolution. By using a Be lens to focus the x-rays into the aperture of two crossed NFLs, a 1.2 by 1.2μm beam with $4.6 \cdot 10^9$ph/s was generated at $E = 25$keV for the tomographic scan. Three fluorescence microtomograms of plant samples and one of a micrometeorite were recorded. Two of these tomograms were recorded with a step size of 600nm (highest resolution ever achieved in a fluorescence microtomogram). Self-consistent reconstruction [17] of the fluorescence data including absorption effects inside the sample was required, in particular for the strongly absorbing micrometeorite.

6 Future

In future experiments, we would like to use Be lenses to acquire magnified tomograms with 3D resolution in the range of 100nm. Besides high spatial resolution, Be lenses allow for a larger field of view and shorter exposure times. Within a project supported by the German BMBF (federal ministry of education and research) we have constructed a sample stage with low eccentricity (< 100nm) that could be used in conjunction with a Be lens for magnified tomography. A proposal for this experiment has been submitted for the next run.

Nanofocusing lenses have the potential of generating 100nm beams at short beamlines, allowing one to perform x-ray analytical techniques, such as diffraction, fluorescence analysis, absorption spectroscopy [18], or small angle scattering, with high spatial resolution. Together with the team from ID13 we have submitted a long term proposal for the development of these microbeam techniques, in particular for the development of fluorescence microtomography.

We continue to provide refractive optics to the synchrotron community.

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