Hard X-Ray Scanning Microscopy with Fluorescence and Diffraction Contrast

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Abstract. Based on nanofocusing parabolic refractive x-ray lenses we have developed and built a hard x-ray scanning microscope that was tested and put to use at beamline ID13 of the ESRF. It can provide a monochromatic hard x-ray nanobeam with lateral extension below 100 nm (down to 50 nm) and a flux up to $10^9$ ph/s in the energy range from 15 to 25 keV. The microscope exploits transmission, fluorescence, and diffraction contrast to obtain local elemental and nanostructural information from the sample. Tomographic scanning yields high resolution elemental maps from the inside of an object. Coherent x-ray diffraction imaging with nanofocused illumination yields images of objects with highest spatial resolution, e.g., 5 nm in a given example.

1. Introduction
One of the key strengths of hard x-ray microscopy is the large penetration depth in matter that allows one to investigate the inner structure of an object, also inside special sample environments. Scanning microscopy can exploit different x-ray analytical techniques as contrast mechanisms, such as diffraction, fluorescence, and absorption spectroscopy. In this way, one can obtain local structural, elemental, and chemical information from a specimen, respectively. This has applications in a variety of scientific fields, for instance in biomedicine, chemistry, materials, earth, and environmental science, and nanotechnology.

Based on refractive x-ray optics [1, 2], a hard x-ray scanning microscope was developed, tested, and put to use at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France.

2. Hard X-Ray Scanning Microscope
At the heart of a scanning x-ray microscope lies a high-resolution focusing optic that images the source onto the sample position in a strongly demagnifying geometry. The smaller the source and the more photons are emitted into the aperture of the optic, the more intensive is the microbeam. Therefore, highly brilliant sources are needed for this technique, such as undulator sources at third-generation storage rings.
In the hard x-ray range, refractive x-ray lenses are well suited to generate intensive microbeams [2]. Reducing the focal length of these optics is advantageous in two ways: the geometric image of the source decreases with decreasing focal length and so does the size of the diffraction limited Airy disc. The short focal length (10 to 30 mm in the hard x-ray range) is realized with so-called nanofocusing lenses (NFLs) [3]. With these optics, lateral beam sizes down to 50 nm have been reached [4]. Currently, they are made out of silicon by nanofabrication techniques, generating a line focus. In the future, their performance in terms of transmission and diffraction limit can be improved by using more transparent lens materials, such as boron or diamond. Theoretically, beam sizes down to below 20 nm are conceivable with these optics [3]. Further improvements of the focal size can be achieved with adiabatically focusing lenses [5].

![Figure 1](image1.png)

**Figure 1.** (a) Hard x-ray nanoprobe installed at beamline ID13 of the ESRF. (b) Detailed view of optics and sample.

![Figure 2](image2.png)

**Figure 2.** Calcium distribution on a virtual section through a leaf hair of *Arabidopsis thaliana*.

Fig. 1(a) shows a hard x-ray scanning microscope developed at Aachen University and TU Dresden in collaboration with beamline ID13 of the ESRF. In this microscope, two nanofocusing lenses are aligned behind each other in crossed geometry to generate a point focus at the sample position [Fig. 1(b)]. By appropriately prefocusing the beam using parabolic refractive x-ray lenses made of Be [2], efficient diffraction limited focusing is achieved. In the microscope the sample can be scanned along three translations and one rotation, allowing also for scanning tomography. Fluorescence, transmission, and diffraction contrast can be exploited.

3. Applications

The scanning microscope described in the previous section was tested and put to use at beamline ID13 of the ESRF. A variety of experiments were carried out, taking advantage of the different x-ray analytical contrasts.

Fig. 2 shows a tomographic reconstruction of the calcium distribution on a virtual section through a leaf hair (tichome) of *Arabidopsis thaliana*. To obtain this tomogram, the sample was scanned in translation (step size 100 nm) and rotation over 180 degrees in 60 steps, recording at each position in the scan a full x-ray fluorescence spectrum of the sample. After extraction of the calcium Kα line, the tomogram was reconstructed using a self-consistent tomographic reconstruction scheme that takes absorption of the incident and fluorescence radiation into account [6]. As the sample is very small, all corrections are small, too, and multiple scattering and secondary fluorescence effects are negligible.

Besides x-ray fluorescence, diffraction can be used as contrast, measuring, for example, the locally resolved diffraction pattern of individual SiGe Stranski-Krastanow islands [7].

In view of future applications of coherent x-ray diffraction imaging [9] at storage ring based or x-ray free-electron laser sources, we have investigated the coherence preservation in the
Figure 3. (a) Scanning electron micrograph of a cluster of gold nanoparticles on a silicon nitride membrane. (b) Coherent x-ray diffraction pattern recorded of the single gold particle pointed to by the arrow in (a). (c) Reconstruction of the gold particle from the diffraction pattern in (b), using phase retrieval techniques [8].

A nanofocused beam in a recent experiment [8]. A gold nanoparticle, pointed to by the arrow in Fig. 3(a) was illuminated by a nanobeam in the diffraction limited focus of the x-ray microscope at $E = 15.25$ keV. The lateral extension of the beam in terms of field amplitude was about $150 \times 150$ nm$^2$ (FWHM), fully illuminating the sample of about 90 nm diameter. The diffraction pattern shown in Fig. 3(b) was recorded in the forward direction in ten one minute exposures on a diffraction camera (FReLoN 4M, 50 µm pixel size, sample-detector distance 1.25 m). From this diffraction pattern, the gold particle in Fig. 3(c) was reconstructed with the unprecedented spatial resolution of 5 nm by phase retrieval techniques. This experiment shows that high resolution coherent diffraction imaging is possible with nanofocused illumination by refractive x-ray lenses. This is important for single particle experiments at future x-ray free-electron laser sources [10] and for high resolution scanning diffraction microscopy [11].

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