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Improving the spatial resolution of neutron imaging at Paul Scherrer Institut – The Neutron Microscope Project

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Abstract

Here we present results stemming from the first prototype of the Neutron Microscope instrument at Paul Scherrer Institut (PSI). The instrument is based on a very thin gadolinium oxysulfide ($Gd_2O_2S:Tb^+$) scintillator screen and a magnifying optics. The Neutron Microscope prototype has been tested at the ICON and the BOA beamlines at PSI and sub-10 μm features can be clearly resolved on a focussed ion beam (FIB) enhance test object – a gadolinium-based Siemens star. The spatial resolution of the images of the gadolinium-based Siemens star assessed by Fourier ring correlation was about 7.6 μm . The outlook for future improvement of the Neutron Microscope system is presented.

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1. Introduction

While X-ray imaging can be nowadays performed routinely with $1\ \mu\text{m}$ spatial resolution at many facilities worldwide, the spatial resolution of neutron imaging is currently limited to about 10 micrometres (Kaestner et al., 2011; Williams et al., 2012; Tremsin et al., 2012). The higher resolution neutron imaging has been flagged up as one of the key demands of neutron imaging user community for the future development in this field. There are ample domains that would profit from higher resolution neutron imaging, ranging from electrochemistry, materials for nuclear safety, soft matter, and soil physics as examples on the materials science side to imaging of various biological systems on the life science side.

As a result, the Neutron Microscope project has been initiated at Paul Scherrer Institut with the goal to develop a facility for high-resolution neutron imaging. The principal technical goal is to develop an instrument which would allow for a sub- $5\ \mu\text{m}$ image spatial resolution, while allowing the images to be taken in reasonable exposure times (i.e. sub-10 minutes for the single radiographies). The current capability of neutron imaging at PSI is limited to 27 mm field of view and $13.5\ \mu\text{m}$ pixel size (see Figure 1).

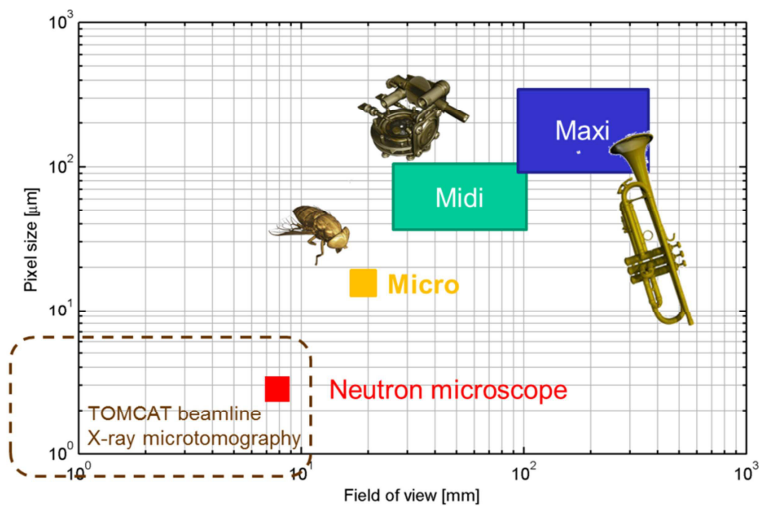


Fig. 1. Domains of neutron imaging facilities available at PSI. Note the overlap of the prospective Neutron Microscope with the domain of synchrotron X-ray microtomography beamline

1.1. The Neutron Microscope concept

The concept of our neutron microscope is, in principle, similar to the imaging concept (i.e. scintillator + camera) currently utilized for the neutron imaging at Paul Scherrer Institute (Lehmann et al., 1999) and also at the various synchrotron imaging beamlines (e.g. Stampanoni et al., 2007). In order to reach the project goals, the performance of all the individual components of the Neutron Microscope must be optimized. Within the initial stages of the Neutron Microscope project, the prototype No. 1.01 has been developed and tested at various beamlines at PSI.

The components optimized in the first phase of the project are

- Tailored magnifying optics of high numerical aperture
- High-resolution high-performance neutron-sensitive scintillator
- High-resolution, high-sensitivity camera
- Neutron-sensitive spatial-resolution test objects

In other words, the Neutron Microscope prototype 1.01 is a very high-resolution, high numerical aperture optical microscope with tailor-made high-resolution neutron-sensitive scintillator.

2. Experimental set-up

For the first prototype of the neutron microscope we decided to use objective with numerical aperture $NA=0.42$, exposure wavelength 436 nm, and the possible field of view of 15 x 15 mm. Originally, this objective served the purpose of being the reduction projection lens of a g-line waferstepper (Nikon NSR-1505G4C) lithography instrument (Mack, 2007). Even though the objective is not designed for the neutron imaging, it has provided sufficient intrinsic resolving power ($0.8 \mu\text{m}$) for our purpose. The photographs of the Neutron Microscope (as installed at BOA and ICON beamlines, respectively) are shown in Figure 2.

When coupled with a sCMOS camera (e.g. Andor Neo, ORCA Flash 4.0 Hamamatsu) with pixels size $6.5 \mu\text{m}$ the nominal pixel size of the images based on Neutron Microscope prototype is equal to $1.5 \mu\text{m}$.

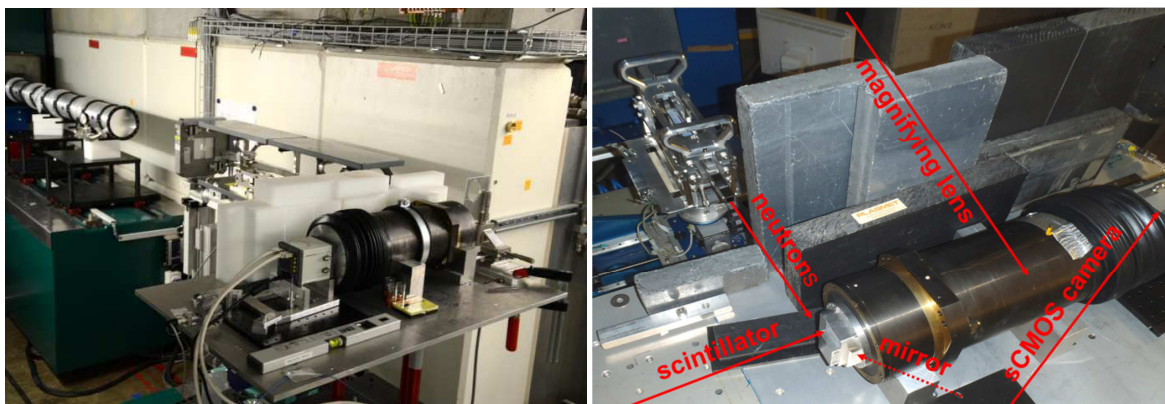


Fig. 2. Photograph of the experimental set-up of the first prototype of the Neutron Microscope installed at (left) BOA beamline and (right) ICON beamline.

The objective was equipped with an adapter that holds 5 x 5 mm scintillator screens and a small mirror that allows for the usage of the magnifying objective outside the neutron line of sight. The scintillator screen has been made of 0.535 mm thick Si-wafer on which a layer of $\text{Gd}_2\text{O}_2\text{S:Tb}$ was applied. The image of a FIB-milled recess revealing the scintillator structure and thickness (about $3.8 \mu\text{m}$) is shown in Figure 3. The magnifying lens can be connected with different types of detectors (CCD, sCMOS) that are to be placed on a linear positioning stage that is used for image focusing. In order to minimize the time needed for image focusing, the set-up is equipped with parabolic neutron guide (Rantsiou et al., 2014) that may provide about one order of magnitude higher neutron flux, albeit at the expense of the increase in the beam divergence. In order to utilize this higher flux for image focusing, $10 \mu\text{m}$ line patterns were milled into the structure of the scintillator at the corners of the expected field of view using FIB. Electron microscopy image of the line patterns is shown in Figure 3.

As the magnifying objective of the Neutron Microscope prototype 1.01 has been originally developed for monochromatic visible light, an optical filter was used in front of the CCD/sCMOS detector. The filter prevents the light of other peaks than the major peak of $\text{Gd}_2\text{O}_2\text{S:Tb}$ emission spectrum from reaching the detector.

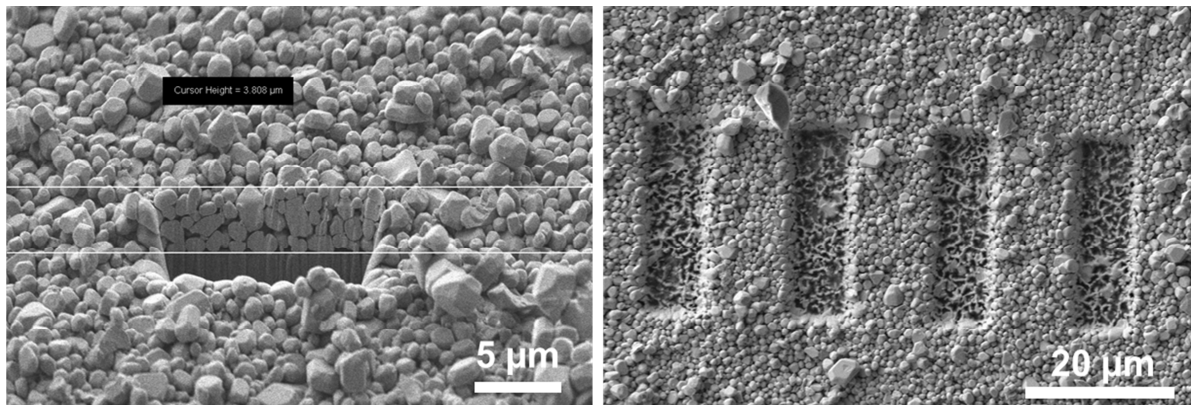


Fig. 3. (left) Electron microscopy image of the FIB-milled recess revealing the thickness (about 3.8 μm) and structure of $\text{Gd}_2\text{O}_2\text{S:Tb}$ powder scintillator utilized for the Neutron Microscope prototype 1.01, (right) 10 μm wide line patterns milled at the corners of field of view into the structure of scintillator for the purpose of image focusing.

An enhanced test object (Gd-Siemens star) was produced for the purpose of testing the resolving power of the new instrument. Five micrometres layer of Gadolinium was sputtered on the quartz glass substrate and micromachined first by ultra-short laser pulses and later by focused ion beam milling. While the micromachining with ultra-short laser pulses provided features (lines) down to 10 μm thickness (20 μm period), FIB milling was used for the enhancement of the test object to 6 μm line widths (12 μm period). As FIB milling on such large length-scales is relatively slow process, only one quarter segment of the Siemens star was machined using FIB.

The resolving power of the Neutron Microscope prototype 1.01 was tested at the BOA (Morgano et al, 2014) and the ICON beamlines. For the purpose of comparison, we present further the results from the ICON beamline and match them with the results based on the standard high-resolution setup (Kaestner et al., 2011) with 13.5 μm pixel size (MICRO set-up) that is used at ICON beamline for hitherto highest spatial resolution routinely available at PSI.

3. Results

Figure 4 provides the comparison of the neutron radiographs of the same object (Gd-Siemens star) using the standard Micro set up arrangement with the image taken using the Neutron Microscope prototype 1.01. The Neutron Microscope prototype image is based on twenty five single frames of the exposure time of 60 seconds, while the standard Micro set-up image based on five single frames of the exposure time 60 seconds only. In both cases was the Gd-Siemens star test object placed in the contact with the scintillator screen. Both set-ups were tested at the ICON beamline at the measuring position No.2.

Even though the pixel size of the images differs (13.5 μm and 1.5 μm , for standard Micro set-up and Neutron Microscope prototype, respectively), both images are cropped to show the same area of interest on the test object. The spatial resolution of the images can be suitably assessed by Fourier Ring Correlation (FRC) technique (van Heel and Schatz, 2005; Vila-Comamala et al., 2011). While the spatial resolution of the image based on the Neutron Microscope prototype is 7.6 μm , the resolution of the image based on the standard high-resolution setup is equal to 32.9 μm . This represents the improvement in the spatial resolution by about factor 4, which corresponds well with the magnification factor of the used optical objective.

Figure 5 shows the centre part of the neutron radiograph of the enhanced Gd-Siemens star that shown in Figure 4-right and is compared to the electron microscopy image of the same area. It is worth to note that even the slight changes in the thickness of the individual spokes that are revealed by electron microscopy of the Gadolinium Siemens star are exhibited as the subtle changes in contrast in the corresponding neutron radiography.

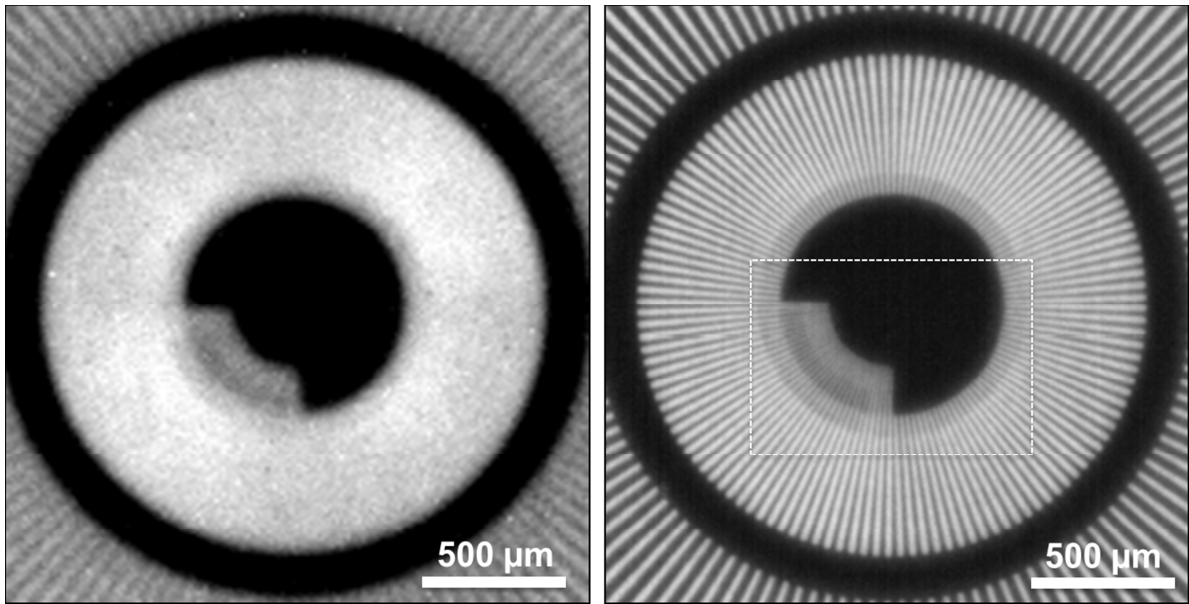


Fig. 4. Neutron radiograph of enhanced Gd-Siemens star (left) using the standard high-resolution setup (Kaestner et al., 2011) with $13.5\ \mu\text{m}$ pixel size and $20\ \mu\text{m}$ thick Gadox scintillator and (right) using the Neutron Microscope prototype 1.01 at ICON beamline with $1.5\ \mu\text{m}$ pixel size and the scintillator shown in Figure 3. The FSC-based spatial resolution of the images is equal to $32.9\ \mu\text{m}$ and $7.6\ \mu\text{m}$, for the standard high-resolution setup and for the Neutron Microscope prototype, respectively. The white dashed-line box shows the size of the zoom-in image in the Figure 5.

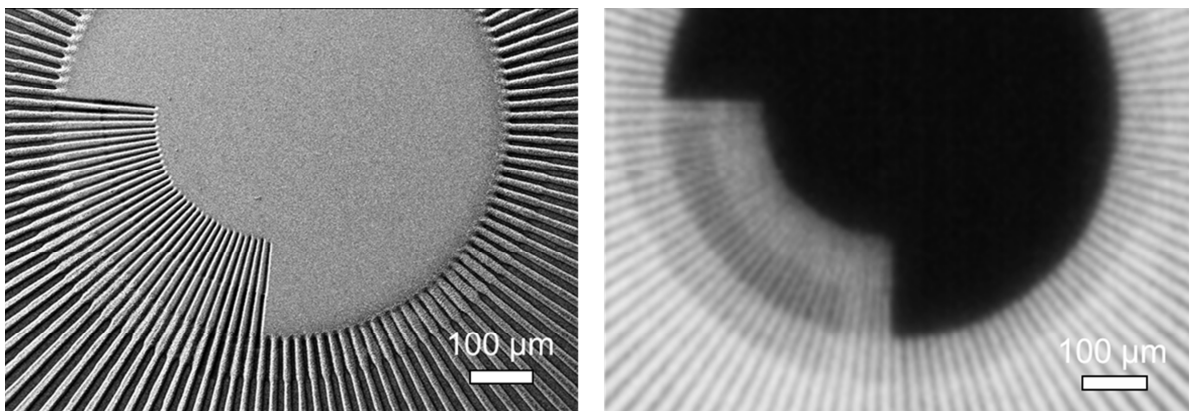


Fig. 5. (left) Electron microscopy image of the centre area of the enhanced Gd-Siemens star; the area indicated Figure 4-right. (right) The corresponding image based on the neutron radiograph

4. Discussion and outlook

The achieved four-fold improvement in spatial resolution will be further enhanced by the following developments of the higher versions of the Neutron Microscope facility.

4.1 Scintillator

The capture efficiency of the current scintillators is about 40%. In order to increase the capture efficiency of the high-resolution scintillators, the development of isotopically enriched materials (e.g. based either on ^{157}Gd , such as $^{157}\text{Gd}_2\text{O}_2\text{S:Tb}$, or on ^{10}B) will be developed. This will allow for the improvement in the temporal, and hence also spatial, resolution of the high-resolution imaging. The isotopically enriched materials are generally rather costly and therefore the challenge lies in the scintillator screen production using a rather limited amount of the input materials (both for the production of the isotopically enriched phosphor itself and the manufacturing of the scintillator screens using such isotopically enriched phosphors).

Another approach to enhance the spatial resolution is to attempt to decouple the spatial resolution of the scintillator from its thickness by means of its microstructuring. Such scheme is already successfully applied in the x-ray imaging community (Rutishauser et al., 2011). In this case, the neutron-sensitive phosphor is filled into the microchannels of the diameter smaller than the targeted resolution. In order to achieve a few micrometres resolution of the final neutron imaging system, the size of the structured microchannels should be about 1-2 μm . The first attempts for production of microstructured scintillators was performed recently (see Figure 6). Both above mentioned approaches (the microstructuring and the isotopical enrichment) can be also possibly combined together.

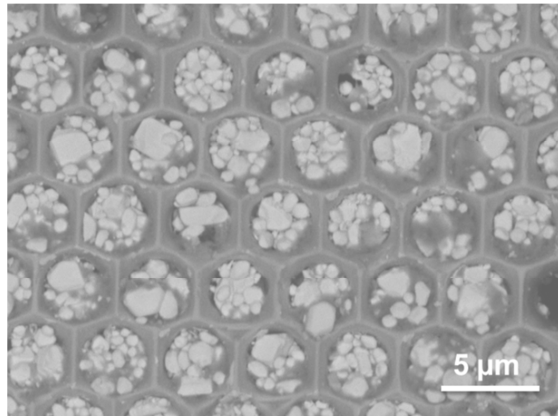


Fig. 6. First prototype of a neutron sensitive microstructured scintillator based on a porous Si-membrane and $\text{Gd}_2\text{O}_2\text{S:Tb}$ powder phosphor

4.2 Magnifying optics

The magnifying lens used in the case of the Neutron Microscope prototype 1.01 was not optimized for the purpose of neutron imaging. The design of the new lens system dedicated for neutron imaging is currently being pursued and will lead – due to optimized light transmission parameters – to the improvement in the temporal and hence of the spatial resolutions of the imaging system.

Also, the geometrical arrangement of the magnifying lens will be such that it will allow for observation of much larger samples (at least $10 \times 10 \text{ mm}^2$) without necessity to resort to the sample scanning and the subsequent image stitching. The field of view of Neutron Microscope prototype 1.01 is limited by the scintillator size to about $4 \times 4 \text{ mm}^2$ and by utilized cameras to about $3 \times 3 \text{ mm}^2$ only. In order to be able to observe the larger field of view, it is, therefore, planned to equip the Neutron Microscope instrument with a CCD/sCMOS camera of much large chip size (e.g. 4096×4096 pixels) .

4.3 Sample positioning and neutron beam collimator

In order to enable the inclusion of the Neutron Microscope instrument into the user-programme of Paul Scherrer Institut, a new sample positioning stage of the superior accuracy must be designed and implemented into the system. As it is planned that the Neutron Microscope instrument will be utilized at various beamlines at PSI (ICON, BOA, NEUTRA) and other external large-scale neutron facilities, the sample positioning stage will be designed as an integral part of a portable instrument.

It is foreseen that high-aspect ratio neutron beam collimators based on microchannel plates will be developed for the purpose of both the conditioning the divergence of the primary neutron beam and for the suppression of signal based on the scattered neutrons (Tremisn et al., 2011). As such collimator will lead to a decrease in the useful neutron beam intensity, the more efficient scintillators, tailored magnifying optics (as described in Chapters 4.1 and 4.2) will prove very important for the successful development of the Neutron Microscope facility.

5. Conclusions

The Neutron Microscope prototype 1.01 was developed within the first stage of Neutron Microscope project at PSI. The spatial resolution of the images of Gd-based Siemens star test object as assessed by Fourier ring correlation technique is equal to 7.6 μm . In this way, the spatial resolution of the neutron imaging facilities at was improved approximately by factor 4 (four).

The following further improvements are foreseen. The magnifying optics of superior light transmission parameter will be tailored for the purpose of the neutron imaging will lead consequently to the shorter image acquisition times and hence better spatiotemporal resolution. It is also foreseen that neutron-sensitive microstructured scintillators will be utilized to further improve the spatial resolution. Likewise, the scintillators based on isotopically enriched materials (e.g. $^{157}\text{Gd}_2\text{O}_2\text{S:Tb}$) will be developed and will enhance the scintillator absorption efficiency, hence the spatiotemporal resolution, significantly. With respect to the inclusion of such instrument into the standard user-programme of Paul Scherrer Institut, a sample positioning stage and a neutron beam collimator will have to be optimized and integrated into such high-resolution neutron imaging instrument.

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