Overcoming the field-of-view restrictions in soft x-ray holographic imaging

C Tieg\textsuperscript{1}, R Frömter\textsuperscript{2}, D Stickler\textsuperscript{2}, H Stillrich\textsuperscript{2}, C Menk\textsuperscript{2}, S Streit-Nierobisch\textsuperscript{3}, L-M Stadler\textsuperscript{3}, C Gutt\textsuperscript{3}, O Leupold\textsuperscript{3}, M Sprung\textsuperscript{3}, G Grüb\textsuperscript{3} and H P Oepen\textsuperscript{2}

\textsuperscript{1}European Synchrotron Radiation Facility (ESRF), B.P. 220, F-38043 Grenoble Cedex, France
\textsuperscript{2}Institut für Angewandte Physik, Universität Hamburg, Jungiusstrasse 11, D-20355 Hamburg, Germany
\textsuperscript{3}Deutsches Elektronen-Synchrotron (DESY), Notkestrasse 85, D-22607 Hamburg, Germany
E-mail: carsten.tieg@esrf.fr

Abstract. We present a new concept for imaging by soft x-ray holography. Microscopy-like imaging capabilities were achieved by the separation of mask and sample. The use of two independent silicon nitride membranes, one for the field-of-view-defining mask and the reference beam, and the other for the sample, allows to image different areas on the sample. The movement of the field-of-view across the sample is realized by a piezomotor-driven sample stage that permits relative and stable positioning with nm-precision. We demonstrate the capabilities of the x-ray holographic microscopy (XHM) technique by showing images with 60 nm spatial resolution of an artificially structured 100 nm thick gold film with a lateral size of 19 \times 4 \mu m^2.

1. Introduction
Very few imaging techniques feature at the same time element selectivity, nm resolution, magnetization sensitivity, flexible sample environment, and the capability to control external parameters like temperature and magnetic field. A recent photon-based imaging technique that combines these characteristics is soft x-ray holography. Since its first demonstration in 2004 \cite{1}, soft x-ray holography has proven to be a powerful tool for the study of magnetic systems, though the first images of non-magnetic specimen were already taken in the early 90s \cite{2}. The method’s inherent magnetization sensitivity, which is based on the x-ray magnetic circular dichroism effect at the $L_{2,3}$ absorption edges, was readily exploited for the study of layered systems with perpendicular magnetic anisotropies, like [Co/Pt] \cite{1, 3, 4, 5} and [Co/Pd] \cite{3, 4, 6, 7} multilayer systems. In addition, the potential for biological imaging was recently demonstrated by a study of diatoms and fibroblast cells \cite{8}. In the hard x-ray regime it was shown that holographic imaging can be combined with phase retrieval methods in order to achieve diffraction-limited spatial resolution \cite{9}. There are some advantages of soft x-ray holography in comparison to transmission soft x-ray microscopy, which offers similar key features and a spatial resolution of 12 nm \cite{10}. First, with soft x-ray holography one can achieve nm spatial resolution with \mu m stability of the setup when using a combined mask-sample structure. And second, there is more room available in the vicinity of the sample for its manipulation.

Synchrotrons are the common x-ray sources for this imaging technique. However, it was recently demonstrated that tabletop soft x-ray sources based on pulsed lasers can be used as
well [11]. Soft x-ray holography is a promising tool for the study of ultrafast magnetization processes using the temporal beam structure of the upcoming free-electron laser sources [12]. However, up to now a widespread use of this new technique has been hindered by certain limitations imposed on the field-of-view (FOV) and the sample.

In soft x-ray holography, samples are commonly deposited on a transparent silicon nitride membrane. On the side facing the beam the membrane accommodates an opaque metal film with apertures for the FOV-defining object hole and one or more holes to define the reference waves [13]. The back-side of the same membrane is used as a substrate for the sample. The object beam, which traverses the sample, interferes with the unperturbed reference beams and produces a holographic image on the sensor of a CCD camera. The intensity of the hologram carries the phase relation between object and reference beam. The real-space image of the non-periodic sample structure is reconstructed by a fast Fourier transform (FFT) of the hologram. This can be done in real-time during the experiment. Magnetic contrast is achieved by taking the difference between holograms recorded with opposite photon helicity at the $L_3$ absorption edge. Tuning the photon energy to the low-energy side of the resonance allows utilizing the phase contrast. This can be advantageous for radiation sensitive samples [6].

Though the ultimate spatial resolution of the reconstructed image is in principle limited by the wavelength, it is in practice the size of the reference hole that matters [1]. The best resolution reached up to now is $30 - 50$ nm for a wavelength of 1.59 nm [1]. Besides the technical difficulties to mill narrow reference holes with high aspect ratios into the opaque film, it is not advantageous in terms of the signal-to-noise ratio to aim for reference holes with sizes as small as 10 nm and below. To achieve a reasonable contrast in the reconstructed image, the intensity of the reference beam should be sufficiently high in comparison to the object beam. For a given resolution, the upper limit for the size of the FOV-defining object hole is therefore determined by the size of the reference hole. For a typical mask with an object and reference hole diameter of 2 $\mu$m and 0.1 $\mu$m, respectively, the intensity ratio of object to reference beam before traversing the sample is 400:1. This ratio is slightly lowered by the attenuation of the object beam intensity as a result of the absorption by the membrane and a thin metallic sample. For example, the x-ray transmission at the energy of the Co $L_3$ edge of a 200 nm thick membrane and a 10 nm thick Co sample is 0.75 and 0.6 [14], respectively. This gives an absorption-corrected intensity ratio of 180:1. A second reason for the size limitation of the FOV is the degree of coherence of the beam. To obtain an interference pattern, the transverse coherence length has to be larger than the distance between object and reference hole. Otherwise the contrast decreases in the reconstructed images. This effect has been recently demonstrated by comparing images of the same sample recorded with different transverse coherence lengths which were obtained by varying the wavelength [8]. A coherent beam is commonly prepared by the use of a pinhole. Placing a pinhole with a diameter of $d = 50 \mu$m for soft x-rays with a wavelength of $\lambda = 1.5$ nm at a distance of $z = 1$ m in front the sample yields a transverse coherence length of $\xi_t = \lambda z / 2 \pi d = 5 \mu$m only. Hence, images with a large FOV of about 50 $\mu$m and a spatial resolution better than 100 nm are therefore not feasible with this conventional mask-sample design. Finally, we also want to mention the limitation imposed by the CCD detector properties on spatial resolution and the maximal FOV that can be imaged. While the former requires a large chip size, the latter needs a small pixel size, which is technologically challenging to achieve with a single device.

In this work we show that the limitations on the FOV can be circumvented by separating the mask from the sample. This is realized by using two membranes. The separated mask allows to move the FOV together with the reference holes across the sample and thus to image different areas on the sample. Like in microscopy, a series of overlapping real-space images can be matched together to give a large-scale image of the sample. We demonstrate the potential of this new concept, for which we use the term x-ray holographic microscopy (XHM), by presenting images of an artificially structured Au film.
2. Experimental Setup

The key to overcome the restriction of a position-fixed and size-limited FOV in soft x-ray holography is to separate the imaging elements from the actual sample by using two silicon nitride membranes. The principal setup is illustrated in Fig. 1. We have used commercial 0.5 \( \times \) 0.5 mm\(^2\) Si\(_3\)N\(_4\) membranes with a thickness of 200 nm supported by a 5 \( \times \) 5 mm\(^2\) silicon frame. The upstream membrane acts as support for the opaque 800 nm thick Au film. We used focussed ion beam milling (FIB) to mill a 2 \( \mu \)m aperture for the object beam and three holes with diameters of 100, 320 and 350 nm for the reference beams into the Au film. The milling process of these high-aspect-ratio holes was performed from the metal side. This gives rise to a conical shape with the smaller hole diameter on the membrane side, as indicated in Fig. 1. The downstream membrane, which is mounted face-to-face to the mask-carrying membrane, supports the sample. It is fixed to a piezomotor-driven in-vacuum x/y-stage. The stage allows with nm precision and stability that both membranes can glide smoothly with respect to each other. In order to guide the reference beams through the sample membrane without being absorbed, we have milled a trench into it. Width and length of the trench were adjusted to accommodate the required lateral travel range of the sample.

![Figure 1](image-url)

Figure 1. Schematic side view of the imaging setup used for x-ray holographic microscopy (XHM). Mask and sample are supported by two separate Si\(_3\)N\(_4\) membranes. The mask support is fixed while the sample support can be moved in the plane perpendicular to the beam. By the relative movement of both membranes, the position of the field-of-view, which is defined by the object hole in the mask, can be changed. A trench in the sample membrane allows within a certain travel range an unperturbed passage of the reference beam through the sample.

For aligning mask and sample in the beam we have used a retractable photo diode. The measurements were performed at the soft x-ray beamline ID08 of the ESRF. A description of the setup can be found elsewhere [5]. The energy was tuned to 778 eV, corresponding to the Co L\(_3\) absorption edge. The intense direct beam is blocked by a 0.7 mm diameter beam stop situated in front of a 16 bit CCD camera with 1300 \( \times \) 1340 pixels and a pixel size of 20 \( \times \) 20 \( \mu \)m\(^2\). The camera was placed 36 cm behind the sample, allowing to record a maximum in-plane momentum transfer from the center (\( q = 0 \)) to the edge of the chip of \( q_{\text{max}} = \frac{4 \pi}{\lambda} \sin \theta_{\text{max}} \approx 0.14 \text{ nm}\(^{-1}\). Here, \( 2\theta_{\text{max}} \) denotes the maximal scattering angle. This gives a spatial resolution \( \Delta x = 2\pi/2q_{\text{max}} \) imposed by the measurement geometry in the reconstructed images of about 20 nm. This value is below the size of the reference holes that defines the actual resolution limit [1].

3. Results and discussion

For this proof-of-principle experiment we have used a sample based on a FIB-structured Au film. Mask and sample are shown in Fig. 2. These scanning electron microscope (SEM) images were taken after the holography measurements. The mask was imaged from the membrane side while the artificially structured sample is seen from the metal film side (cp. Fig. 1). The mask hosts the FOV-defining object hole with a diameter of 2 \( \mu \)m, and three reference holes, drilled at a
distance of 7.5 µm from the object hole center. They are arranged in a three-fold symmetry. The reference holes have a conical shape with the smaller diameter on the membrane side as a result of the milling process from the Au side. The upper and bottom left reference hole have a diameter of 320 nm and 350 nm, respectively. They allow high-contrast imaging while the bottom right one with a diameter of only 100 nm is optimized for high-resolution imaging.

![Figure 2. SEM image of mask (a) and sample (b) (same length scale). The 800 nm thick Au mask accommodates a 2 µm object hole, and a reference hole with 350 nm diameter at the bottom left, 320 nm at the top and 100 nm at the bottom right. Note that the membrane was completely removed in the object hole. The position of these four apertures on the FIB-structured sample is indicated in (b). A transparent trench (horizontal black bar at the bottom) assures for different imaging positions that at least one of the two lower reference beams can pass the 100 nm thick Au film of the sample without being obstructed. For XHM, a series of holograms at adjacent FOV positions is recorded, and the individually reconstructed images are merged together.](image)

For the sample preparation, we deposited a continuous 100 nm Au film on the back-side of the downstream membrane and milled the word Hamburg into the film down to the Si₃N₄. The entire FIB structure spans an area of 17 × 4 µm². In order to achieve a complete removal of the Au within the letters, the FIB milling process was extended until part of the membrane had been removed. This is indicated in the sample cross-section in Fig. 1. However, owing to an inhomogeneous milling yield, some Au remained on the membrane in the form of crystallites. These crystallites appear in Fig. 2 (b) in medium gray. In the bottom part of the sample we milled a trench through the Au film and the membrane. The complete removal of the membrane is obvious from the darker gray value as compared to the letters. The purpose of the trench’s discontinuity below the letter b is to maintain mechanical stability of the sample membrane and the Au film. The vertical distance of the trench from the Hamburg-letters and its size were adjusted to the design of the mask in order to assure that at least one of the two bottom reference holes always lied above the trench while scanning the sample. The circles on the sample in Fig. 2 (b) replicate the mask. In principle, recording a hologram at two adjacent positions along the vertical direction is sufficient to image the whole vertical extent of a letter. Note that the upper reference hole is blocked. At a photon energy of 778 eV, the 100 nm Au film and the 200 nm sample membrane attenuate the intensity of this reference beam to about 15%.

Fig. 3 shows as an example the hologram and its FFT image of the sample region at the lower right part of the letter H. The SEM image in the inset of (a) gives the position of the object hole in front of the sample. The hologram was recorded with a total exposure time of 20 s. The beam stop blocks the direct beam in the center. Three point-symmetric pairs of this sample region appear in the FFT reconstruction. The sub-image α and its complex conjugate α* have the
Figure 3. A hologram (a) and the real part of its Fourier transform (b). It was recorded while the bottom right part of the letter H was placed behind the object hole. The SEM image in (a) shows the position of the object hole on the sample. The FFT image contains the real-space image of the sample from all three reference holes, each producing a point-symmetric pair of the FOV. Sub-image $\gamma$ and its complex conjugate $\gamma^*$ correspond to the 100 nm reference hole. The lowest spatial resolution. They correspond to the 350 nm reference hole. Although blocked, the 320 nm reference hole gives rise to sub-image $\beta$, which has a weaker contrast as compared to $\alpha$. We have used sub-image $\gamma$ for reconstructing the entire sample. It corresponds to the 100 nm reference hole, and has a spatial resolution of 60 nm using the 20%–80% knife-edge definition.

As in optical microscopy, scanning the sample with respect to the optics allows to image samples that exceed the size of a single FOV. This capability is demonstrated in Fig. 4. The image, spanning the entire sample, is composed of 40 single FOVs. Contrast and spatial resolution are high enough to identify the Au crystallite on the membrane that result from an insufficient FIB milling process. For example, a Au crystallite is visible at the left and right end of the horizontal bar in the letter H. They have sizes of about 100 nm and 350 nm, respectively. It is important to stress again that it is not feasible to image such a large area with one exposure by conventional holography based on a position-fixed object hole of this size. In addition to the requirement of a much larger transverse coherence length, the relative intensity of the reference beam compared to the object beam would not be sufficient to obtain a reconstructable hologram if the size of the reference hole was not increased as well.

Figure 4. Image of a FIB-structured 100 nm thick Au film obtained from merging 40 single reconstructed holograms.
We have also used our x-ray holographic microscopy setup for magnetic domain imaging. We refer to our recent work where we demonstrate this capability by a study of the magnetic domain structure in a \([\text{Co}_{0.7\,\text{nm}}/\text{Pt}_{2.0\,\text{nm}}]_8\) multilayer with a laterally varying anisotropy as a result of the exchange coupling to an adjacent Fe wedge \([15]\).

4. Summary
We have demonstrated a concept for x-ray holographic microscopy (XHM). It is based on the use of two separate silicon nitride membranes, one to accommodate the mask and the other to support the sample. The FOV-defining object hole, which is fixed in size and position on the mask membrane, can be moved across the sample with nm precision and stability by means of an x/y–stage. This design allows to record holograms at different sample positions. Hence, large sample areas can be imaged by combining individually reconstructed holograms. In comparison to conventional holography based on combined mask-sample structures \([1]\), the preparation effort is much less. The technically demanding mask fabrication by FIB milling is now independent of the sample preparation. In addition, the mask can be used for several samples. Our XHM technique opens the door for the study of properties that vary along lateral length scales exceeding the size of a single FOV with a diameter of only \(1 − 3\,\mu\text{m}\) \([15]\). We believe that this technique will lead to a much broader use of soft x-ray holography due to its flexibility and microscopy-like capabilities.

Acknowledgments
The authors thank R. Barrett for engineering support, G. Retout, R. Homs-Regojo, and T. Trenit for technical assistance. The group of Hamburg University acknowledges financial support by the German research foundation DFG within SFB 668, and the Free and Hanseatic City of Hamburg in the context of the “Landesexzellenzinitiative”.

References
[1] Eisebitt S, Lüning J, Schlotter W F, Lörgen M, Hellwig O, Eberhardt W and Stöhr J 2004 Nature 432 885
[2] McNulty I, Kirz J, Jacobsen C, Anderson E H, Howells M R and Kern D P 1992 Science 256 1009
[3] Hellwig O, Eisebitt S, Eberhardt W, Schlotter W F, Lüning J and Stöhr J, 2006 J. Appl. Phys. 99 08H307
[4] Haet T, Günther C M, Pfau B, Schabes M E, Thiele J U, Rick R L, Fischer P, Eisebitt S and Hellwig O 2008 Phys. Rev. B 77 184421
[5] Streit-Nierobisch S \(\text{et al.}\) 2009 J. Appl. Phys. 106 083909
[6] Scherz A \(\text{et al.}\) 2007 Phys. Rev. B 76 214410
[7] Günther C M, Radu F, Menzel A, Eisebitt S, Schlotter W F, Rick R, Lüning J and Hellwig O 2008 Appl. Phys. Lett. 93 072505
[8] Guehrs E, Günther C M, Könnecke R, Pfau B and Eisebitt S 2009 Opt. Express 17 6710
[9] Stadler L-M, Gutt C, Autenrieth T, Leupold O, Rehbein S, Chushkin Y and Grübel G 2008 Phys. Rev. Lett. 100 245503
[10] Chao W, Kim J, Rekawa S, Fischer P and Anderson E H 2009 Opt. Express 17 17669
[11] Sandberg R L, Raymondson D A, La-o-vorakiat C, Paul A, Raines K S, Miao J, Murnane M M, Kapteyn H C and Schlotter W F 2009 Opt. Lett. 34 1618
[12] Dür H A \(\text{et al.}\) 2009 IEEE Transactions On Magnetics 45 15
[13] Schlotter W F \(\text{et al.}\) 2006 Appl. Phys. Lett. 89 163112
[14] Nakajima R, Stöhr J and Idzerda Y U 1999 Phys. Rev. B 59 6421
[15] Stickler D, Frömter R, Stillrich H, Menk C, Tieg C, Streit-Nierobisch S, Sprung M, Gutt C, Stadler L M, Leupold O, Grübel G and Oepen H P accepted by Appl. Phys. Lett.