Extreme ultraviolet and soft X-ray microscopy using a compact gas puff target laser-plasma sources

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In this chapter we present our recent experiments related to EUV and SXR microscopy using a compact, laser-plasma based EUV and SXR sources employing a double stream gas puff target. We present a tabletop, EUV transmission microscope at 13.8 nm wavelength, with a spatial (half-pitch) resolution of 50 nm. Those results might be useful in the future for the realization of a compact high-resolution tabletop imaging systems for actinic defect characterization. We also present applications of a desk-top microscopy using a laser-plasma EUV source based on a gas puff target for studies of morphology of thin silicon membranes coated with NaCl crystals and ZnO nanofibers. Short wavelength sources emitting in the “water window” spectral range between \( \lambda = 2.3 \) and \( 4.4 \) nm are often used for imaging of biological samples. Herein, we also present the results on developing a compact, desktop size, laboratory microscopy setup, based on SXR gas puff target source, which emits incoherent radiation in the “water-window”. Details about the source and the microscope as well as imaging results for test and biological objects will be presented and discussed.

Keywords: diffractive lenses; Fresnel zone plates; EUV/SXR imaging; EUV/SXR microscopy; X-rays; soft x-rays (SXR); extreme ultraviolet (EUV); “water-window”; Wolter optics; gas puff target; laser-produced plasma

1. Introduction

Future developments in nanoscience demand tools capable of capturing images with a nanometer spatial resolution. Various imaging methods and techniques are currently under active pursuit worldwide, one of them being an extreme ultraviolet (EUV) and soft X-ray (SXR) microscopy, based on Fresnel zone plates [1], where a direct path to improve the spatial resolution is to use a short wavelength radiation.

The resolution of the optical system is described by the Rayleigh criterion. This criterion states that the image of two point sources of monochromatic radiation with equal intensities in a noise free background will produce two Airy intensity patterns in the image plane. If the points are separated enough, then the image will be formed by two distinct Airy patterns, one for each source. The point sources are said in this case to be resolved. If the sources are closer, then their Airy patterns will start to overlap. If the two sources are mutually incoherent, the superposition of the two Airy patterns is an intensity addition. By bringing the two sources closer together the intensity between them increases and it makes more difficult to distinguish the individual source images. According to the Rayleigh criterion, the separation between the sources where two point sources are said to be just resolved corresponds to the superposition of the first null of the Airy intensity pattern from the first source with the maximum of the Airy intensity pattern from the second source. This distance defines the resolution of the optical system under incoherent illumination in the straightforward way and is equal to \( \Delta = k \lambda / N A \), where \( \lambda \) is the illumination wavelength and \( NA \) is a numerical aperture of the system. This equation assumes incoherent illumination for \( k = 0.61 \). According to Heck et al. [2] the value of \( k \) may change from ~0.34 up to 1 depending on the degree of coherence of the illumination, the illumination spectrum, and the type of the resolution test.

Above equation shows that one way to improve the spatial resolution of the imaging system is by reducing the illumination wavelength. This was the motivation to employ short-wavelength laser-plasma EUV/SXR source for illumination. Its wavelength is ~100-400x shorter than the visible light and allows to significantly improve the imaging resolution. This is also the reason why using SXR radiation the best spatial resolution obtained till now in Fresnel zone plate microscopes is 12nm using synchrotron radiation [3]. The work done so far in EUV and SXR imaging is very extensive and it is not possible even referencing it in this small chapter, thus we only present a few examples for further research. Imaging experiments were carried, using coherent and incoherent EUV/SXR sources. A 700nm half-pitch resolution images with the use of EUV recombination laser at \( \lambda = 18.2 \) nm has been reported in the early imaging work [4]. 75nm resolution was reported employing SXR laser at \( \lambda = 4.48 \) nm pumped by fusion-class NOVA laser limiting image acquisition by laser repetition rate to several shots per day [5]. Recently, different approaches to sub-micrometer resolution imaging have emerged due to the development of smaller-scale short wavelength sources such as high-order harmonics [6], SXR lasers [7] and incoherent laser-plasma based sources [8]. Compact EUV and SXR sources give the opportunity to perform the experiments without the necessity to employ large “photon facilities” with limited user access. Thus these sources have a huge impact on the speed of nanotechnology development since the experiments previously restricted only to large facilities, now can be performed in the laboratory environment.

The importance of the development and the commercial availability of the EUV/SXR sources, in particular the lack of commercial tools for actinic inspection, for the semiconductor industry, was already noted by Intel [9]. The usability of the existing EUV/SXR sources was proved by many experiments involving compact table-top EUV/SXR sources such as capillary discharge lasers for the Interferometric Lithography IL and wavelength resolution holoography [10,11], high-
harmonic generation sources [12] lens-less imaging [13], optically pumped EUV lasers in the EUV microscopy with the use of zone-plates [14] and many more. Using radiation from a capillary discharge laser, \( \lambda = 46.9 \) nm wavelength, EUV images were obtained with a spatial resolution of 120-150nm [15]. A 13.2nm wavelength radiation from Ni-like Cd EUV laser allowed for a 55nm in reflection mode [16] and sub-38nm resolution nano-imaging in transmission mode [14], using pulses of 11.8ps for pumping. Using a capillary discharge laser EUV microscopy in transmission mode was demonstrated recently with a single EUV laser pulse, leading to a 54 nm half-pitch spatial resolution and temporal resolution of ~1ns, as reported in [17] and was evaluated in more details using a correlation algorithm, reported in [18]. A quasi-monochromatic emission from incoherent SXR source based on liquid nitrogen, at \( \lambda = 2.88 \) nm, in the “water window” range, allowed to demonstrate SXR microscopy with sub-50nm spatial resolution (~17\( \lambda \)) [19]. Using xenon based gas discharge EUV source, Schwarzschild objective and Fresnel zone-plate optic for second magnification step, EUV imaging was demonstrated reaching the spatial resolution of ~100nm [20].

SXR sources emitting in the “water window” region between 2.3 and 4.4nm wavelengths [21] are also very important for imaging of live biological samples. High contrast in this spectral range is obtained due to a difference in absorption of different constituents of biological specimen. While water, present in a sample, has relatively small absorption coefficient in this spectral range, carbon, due to much higher absorption, gives very good contrast in the image. Thus this spectral range is perfectly suitable for imaging of biological specimen. SXR microscopy has been successfully employed mainly in transmission mode, either using diffractive optics, such as zone-plates [22], raster scanning the sample by focused SXR beam [23] or as a contact microscopy [24]. However in most of these important achievements synchrotron sources were mainly used for sample illumination, due to their obvious advantages. The main motivation for development of compact, table-top SXR sources is to open the possibility to perform experiments, without necessity to employ large “photon factories” such as synchrotrons or free electron lasers. These sources are the state-of-the-art devices with tunable, wide range output wavelengths, large photon flux and brightness, however, desktop sources, although exhibiting often worse parameters are still an important alternative to perform some experiments in a university laboratory for instance. While synchrotrons are always attractive SXR sources for leading-edge experiments, the growing importance of compact EUV and SXR sources can already be seen.

2. EUV imaging based on a laser-plasma gas puff target source

2.1 Quasi-monochromatic EUV source at \( \lambda = 13.8 \) nm and EUV microscopy setup

One of the compact short wavelength sources, capable of being a source for high resolution microscopy is a laser-plasma source based on a gas puff target (LP-GPT source). The EUV source, used in the experiment, has been developed for EUV metrology applications in the frame of MEDEA+ project [25]. For imaging employing Zone Plate (ZP) objectives a monochromaticity of the EUV radiation, produced using the LP-GPT source, is a key parameter. For this purpose a laser-plasma argon gas puff target based EUV source was optimized for an efficient EUV photon production. In our experiments, reported in [26], this source was characterized and optimized for quasi-monochromatic emission and is based on a double nozzle gas puff target that was later employed in applications requiring narrow-bandwidth EUV radiation, such as EUV microscopy with ZP objectives [27,28].

The EUV microscope, presented in this paragraph was described in more details in [27]. The scheme and experimental arrangement are shown in Fig. 1a) and b), respectively.

![Fig. 1 Scheme a) and experimental arrangement b) of the EUV microscope (not to scale) using a laser-plasma EUV source based on gas puff target, image taken from [28].](image)

To study the bandwidth influence on spatial resolution of the EUV microscope Ar and Xe plasmas were produced using pumping laser pulses from Nd:YAG laser (Eksma), with pulse duration of 4ns and energy 0.74J. The plasmas radiate in a very broad range of wavelengths, dominantly in the EUV range (5-50nm) and by using additional spectral filtering it is possible to shape the spectral emission of the source. The source can operate up to 10Hz repetition rate. A pressure of 2x10^-3 bar was constantly maintained in the chamber during the source operation. The experimental setup is
extremely compact. The microscope is located inside the vacuum chamber, 24cm in diameter and 35cm in length and the entire system fits on top of a single 2x0.6m² optical table.

The laser plasma source was optimized for efficient EUV radiation generation from Ar [25] and Xe [29] plasma. Argon plasma emits the radiation in 13-14nm band mostly in two strongest lines in at 13.793nm and at 13.844nm wavelength. Number of photons measured in 13-14nm band was equal to (8.8±0.5)·10¹⁰ in a single EUV pulse. The measured photon flux corresponds to 1.29µJ/pulse. Additionally the source size was estimated. Using a pinhole camera it was found that the source was an ellipse-shaped with the major and minor axes of equal to 1090µm and 390µm. For the Xe plasma, due to much more efficient EUV photon production and the risk of ablating the zone plate objective, we did not optimize the source particularly for Xe and still obtained the photon flux approximately 5 times larger than in case of Ar plasma, with similar plasma size. Xe spectrum is much more complex, quasi-continuous, filling in the entire reflection band of the condenser $\lambda=13-14$nm, with transitions identified in [30,31]. For both plasmas, to eliminate longer wavelengths ($\lambda>18$nm) a 100nm thick, 10 mm diameter, free-standing Zr filter (Lebow) was used. The Zr filter was positioned ~4-5mm upstream the object.

EUV radiation from plasmas was collected, focused and spectrally filtered by an ellipsoidal, off-axis, 80mm in diameter mirror with Mo/Si multilayers. The mirror was corrected for the spherical aberrations. The multilayers were optimized for 13.5+/-0.5nm (FWHM) wavelength range and incidence angle of 45 degrees. The theoretical reflectivity of the mirror at 13.5nm wavelength is 37.7% for an unpolarized light from the laser-plasma EUV source. The mirror was designed to image the plasma with a unity lateral magnification, having both the object and image distances equal to 254mm. It was developed in co-operation with Reflex s.r.o., (mirror substrate) and Fraunhofer Institut für Angewandte Optik und Feinmechanik (coating).

![Fig. 2 SEM images of two objects used during studies of object thickness influence on the EUV microscope spatial resolution: a) copper mesh, 4µm thick and perforated carbon foil, coated additionally by a thin layer of gold, total thickness of 70nm b). Image from [28].](image)

Second interesting experiment was related to study of the object thickness influence on spatial resolution. For that two different objects, with different thicknesses in relation to depth of focus (DOF) of the microscope, were used. First one is a copper G2000HS Fine Square Mesh (SPI Supplies) with 12.5µm period, 5µm width bar and 4µm thick (~11x DOF). The second one is a Quantifoil holey 300M carbon foil supported on a steel mesh (SPI supplies), 10nm thick according to manufacturer’s specifications, having 1.5µm diameter holes spaced on a square grid with 2.5µm period. To improve the optical contrast carbon foil was additionally coated with ~60nm thick layer of gold, having a transmission of 4.3% at 13.84nm wavelength, thus in total having thickness of ~70nm (0.2x DOF). Typical SEM images of the mesh and the perforated foil are shown in Fig. 2a) and b), respectively.

The objects, placed 254mm from the mirror, are imaged using a Fresnel ZP objective onto EUV sensitive CCD camera iKon-M (Andor) with 1024x1024 pixels and 13x13µm² pixel size. The ZP was fabricated by Zone Plates Ltd., using electron beam lithography in 220nm thick PMMA (polymethyl methacrylate) layer spin coated on top of a 50nm thick silicon nitride Si3N4 membrane. ZP diameter is equal to D=200µm, number of zones NZP=1000 and an outer zone width $\Delta r=50$nm. At $\lambda=13.84$nm from Ar plasma the focal length of the ZP was f=722.5µm and the numerical aperture was equal to $\text{NA}=0.138$. For both types of spectra and both objects the magnifications, ranging from 520-840x, were used, adjusted by changing the camera-ZP distance and refocusing. Geometrical numerical apertures of the collecting ellipsoidal mirror in horizontal and vertical directions are equal to $\text{NA}_{\text{H}}=0.11$ and $\text{NA}_{\text{V}}=0.15$, respectively, and are similar to the numerical aperture of the ZP, thus providing incoherent illumination [22]. The ZP was mounted on three axis translation stage driven by vacuum compatible step motor actuators (Standa). A piezoelectric, 25µm travel, single axis flexure stage, model NF15A (Thorlabs), was used to adjust precisely the object-ZP distance at a rate of 3V/µm with theoretical resolution reaching 10nm. To provide a conical illumination of the object and avoid stray light through the ZP a circular beam block, 12mm in diameter, was placed ~15cm from the ZP. In case of “binary” transmission mesh object and quasi-monochromatic radiation from Ar plasma to obtain a single EUV image 50 EUV pulses were required, at 2Hz repetition rate, while in case of quasi-continuum radiation spectrum from Xe, due to the increased flux, the required exposure dropped to 10 EUV pulses at 2Hz repetition rate. The source can operate at up to 10Hz repetition rate; however, the pressure buildup in the microscope chamber might cause re-absorption of EUV photons in neutral gas. The CCD camera was cooled down to -20 °C to decrease intrinsic noise during the image acquisition.

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2.2 Experimental results

Four sets of measurements were performed for two types of objects (mesh and foil) and for two plasmas (Ar and Xe). For each case the microscope alignment was optimized to provide a uniform illumination in the entire FOV, also, during the image acquisition, the object-ZP distance was changed using the piezoelectric stage by \( \Delta z \sim 330 \text{nm} \), corresponding to 1V/step, to obtain the sharpest possible EUV image, over the \( z \)-range of \(-20\mu\text{m}\) from the ZP focal point. The \( \Delta z \) was chosen to be smaller than DOF. From the entire set of images, for each object/bandwidth combination, the “sharpest” EUV image was chosen for subsequent resolution measurements. Resolution of the microscope was assessed by a well-established knife edge (KE) test. For incoherent illumination the 10-90% intensity transition across a sharp edge corresponds to a well-known Rayleigh resolution and to twice the value of half-pitch grating resolution of the optical system [1].

Typical EUV images of the mesh object under illumination by Ar (a,c) and Xe (b,d) plasma radiation are depicted in Fig. 3. Small rectangular boxed regions, where subsequent KE resolution measurements were carried out, are magnified to show an obvious resolution decrease by edge blurring for the case of Xe illumination, shown in Fig. 3 (c,d). Similarly, for the perforated foil object, Fig. 4 depicts the EUV images obtained for different illumination bandwidths. An additional object blurring can be observed due to the 10x wider illumination bandwidth from Xe plasma. The best KE half-pitch resolution was measured to be 51.0+/-10.6nm based on 6 independent measurements for thin, perforated foil object and quasi-monochromatic illumination from Ar plasma. Although, less prominent, the influence of the object thickness on the spatial resolution can also be found from the measurements, because a slightly worse resolution of 72.7+/-5.0nm was obtained for thicker mesh object. This corresponds to previously assessed spatial resolution of the microscope equal to 69.4+/-4.0nm, reported in [26]. A real resolution change was noted, however, for broad band, quasi-continuous illumination from Xe plasma. For “thin” object the measured spatial half-pitch resolution was 139.9+/-15.3nm and for the “thick” one 151.8+/-11.8nm.

![Fig. 3](image-url) EUV images of Cu mesh object with Ar a,c) and Xe b,d) plasma illumination. c,d) are the magnified subsections of larger EUV images indicated by boxed regions showing the edge in detail [28].

![Fig. 4](image-url) EUV images of perforated carbon/Au foil object with Ar a,c) and Xe b,d) plasma illumination. c,d) are the magnified subsections of the EUV images in boxed regions showing a single hole in detail [28].

2.3 Discussion of the results

The theoretical half-pitch resolution of the microscope can be expressed by \( r_{KE} = k\lambda / (2N_{zp}) = k\Delta r = 0.5\Delta \), where \( k \) is illumination dependent and resolution test specific constant [2], \( N_{zp} \) is the numerical aperture of the objective ZP, for incoherent illumination \( (k=0.61) \) this resolution is equal to 30.5nm, so it is better than the measured half-pitch resolution of 51nm in the best case. This is due to the fact, that the zone plate is a highly dispersive diffractive element. The theoretical resolution can only be achieved for monochromatic radiation, when the monochromaticity criterion described by equation \( \Delta \lambda / \lambda > N_{zp} \) will be fulfilled, namely the IRB will be larger than the number of ZP zones. In our case, even for the quasi-monochromatic radiation from Ar plasma, this criterion is not satisfied, thus the relatively small IRB of our EUV source will introduce some achromatic blurring to the image. Secondly the resolution loss is related to the object thickness. If the object thickness \( t \) does not fulfill the relation \( t << DOF \), its thickness will influence the resolution of the EUV images as well, thus perforated foil object, having thickness much smaller than the ZP depth of focus, allows to avoid any spatial resolution decrease due to the object thickness and to study only the dependence of the illumination bandwidth on resolution.
2.4 Single shot operation

The possibility of single-shot operation and temporal resolution equal to the duration of the EUV pulse (in this case ~3ns) was also tested using Cu mesh object a sequence of images was obtained with decreasing number of EUV pulses used for acquisition, as shown in Fig. 5. Starting from 50 EUV pulses down to 1 the signal to noise ratio S/N decreases as well, however, the main features of the mesh at single EUV pulse are present. The image is very noisy and grainy, indicating the necessity to increase the number of photons illuminating the CCD detector. The S/N can be improved by integrating the signal over small areas in the CCD, binning the image 4x4 pixels for example, thus improving S/N considerably, as can be seen in the rightmost image in the sequence in Fig. 5. This procedure, however, can be done for the cost of worse resolution, since the equivalent pixel size after binning becomes larger.

Fig. 5  EUV images of Cu mesh with decreasing number of EUV pulses used for image acquisition. Last image in the sequence was obtained after internal integration of the signal from 16 CCD pixels (binning 4x4 pixels) to improve S/N ratio.

2.5 Applications of EUV microscope for material science

2.5.1 Imaging of thin silicon membranes coated with NaCl crystals

Samples on thin silicon membranes were prepared and imaged in the EUV spectral range [32]. In the slots, made in silicon substrate 3x3mm² in size, a 15nm thick silicon membranes were made by deposition and etching. On top of these membranes a thin layer of a NaCl from saline solution was applied. Recrystallization of NaCl leads to the formation of micro-and nano cracks. The cracks in the salt layer and membrane itself are difficult to visualize using an optical microscope, as can be seen in Fig. 6a); we see only the large cracks of the order of 2-10 microns in size, while nanocracks are not visible. SEM images, Fig. 6b), allows to obtain images of the sample with very good spatial resolution and quality, but cannot directly assess the depth of the cracks in the layer of salt. EUV microscope image, however, cannot see the surface of the layer of salt, but the areas with thickness less than ~100nm are clearly visible (bright areas in Fig. 6c)). These areas are visible due to the optical contrast in the EUV spectral range; only the thin areas of the sample transmit EUV photons, which are then used to form a magnified image of the sample.

Characteristic crystals that formed on the surface of the membrane are too thick for direct observation, because the absorption length in the sample is of the order of several hundred nanometers. The recrystallization process results in the formation of crystallites, however, the introduction of stress on the surface of a thin membrane causes formation of local cracks. These cracks are very well visible in the extreme ultraviolet, which gives additional information on the same sample as compared with the images from the optical microscope and SEM.

Fig. 6  The images obtained using an optical microscope of thin silicon membrane coated with a layer of salt, using a microscope objective with 100x magnification, NA~1 a), a boxed area, marked by dashed line was then visualized using the experimental EUV microscope and SEM microscope. An image from the electron microscope SEM, image size ~54x54μm² - b), EUV microscope image made up of 16 partially overlapping subimages in 4x4 arrangement - c). Image taken from [32].

Additional information that we can obtain using EUV microscope is shown in Fig. 7, where we compare small fragments of the sample: SEM images (Fig. 7a) and Fig. 7c)) and images from the EUV microscope (Fig. 7b), Fig. 7d)) representing different areas of the sample. Some cracks in a sample, the size of about 100nm (4 pixels in the image), are deep all the way to the silicon membrane and are visible in both the SEM and EUV images (marked with white arrows) - as shown in Fig. 7a) and b), and some cracks (marked dotted arrow) are visible in the SEM image – Fig. 7c), but are not present in the EUV image – d). For this reason, some features, or structures in the sample look almost identical in the SEM images, but exhibit completely different characteristics, which can be seen in the EUV images. Fig. 7e) shows
the EUV intensity profile of an exemplary crack in the image, with a width of about 4 pixels ~100nm. More details were published in [33].

![Image](image1.png)

**Fig. 7** Comparison of SEM images of small fragments (a, c) and EUV microscope images (b, d) representing different areas of the sample. Some cracks in the sample, the size of about 100nm e), are all the way to the silicon membrane and are visible in both the SEM and EUV images (marked with white arrows) - a, b) and some cracks (marked by dashed arrow) are visible in the SEM image c), but are not present in the EUV image d). White marker has a length of 2μm. Intensity profile of some crack in the EUV image that has a width of about 4 pixels ~100nm e). Image taken from [32].

### 2.5.2 Imaging of ZnO nanofibers

In the frame of mutual cooperation, a nanofiber sample was also prepared. The fibers, 70-800nm in diameter, were made from zinc oxide (ZnO) in Environmental X-ray and Electron Microscopy Research Laboratory, in Biological Physics Group, Institute of Physics, Polish Academy of Sciences, in Professor Danek Elbaum’s group. ZnO nanofibers are produced by electrospinning method in a strong electric field, forming a long, thin cylindrical structure.

![Image](image2.png)

**Fig. 8** Comparison of images of ZnO nanofibers, image size ~20x20μm², obtained using different imaging methods. An optical microscope image a) using a 100x objective, NA ~1, the EUV microscope image - b) for 100 EUV pulses exposure, magnification 690x, NA ~0.138, c) – the SEM image acquired at acceleration voltage of 30kV, ~5000x magnification (sample-screen), from [32].

Fig. 8 shows, for comparison, images of nanofibers, placed on top of a gold mesh, obtained using different imaging methods. An optical microscope image a) using a 100x objective, NA~1, the EUV microscope image - b) for 100 EUV pulses exposure, magnification 690x (object-CCD camera), NA ~0.138, c) - the SEM image acquired at acceleration voltage of 30kV, ~5000x magnification (sample-screen). We note that the spatial resolution of EUV microscope image is much better than an optical microscope. The smallest structures, visible in EUV image are approximately 100nm. In addition, EUV microscopy uses EUV photons as carriers of information, requires no special preparation of the sample prior to imaging, as in the case of SEM. The results of this experiment are presented in [34].

### 3. “Water-window” imaging based on a laser-plasma gas-puff target source

Particularly suitable range of wavelengths for biological imaging is so called the “water-window” spectral range. X-ray sources, emitting in the “water-window” region between 2.3 and 4.4 nm wavelength [21], are thus important for biological applications. High contrast in this spectral range is obtained due to a difference in absorption of different constituents of biological specimen. While water, present in the sample, has relatively small absorption coefficient in this spectral range, carbon, due to much higher absorption, gives very good contrast in the image. Thus this spectral range is perfectly suitable for imaging of biological specimen.

A quasi-monochromatic emission from an incoherent SXR source, at much shorter wavelength, λ = 2.88 nm, in the “water-window” spectral range, based on liquid nitrogen, allowed to demonstrate SXR microscopy with a sub-50 nm spatial resolution [35]. Ethanol droplet based SXR source at λ = 3.37 nm, combined with a zone plate objective allowed to capture images with spatial resolution ~50 nm [36]. Liquid nitrogen based SXR source at λ = 2.48 nm was used to demonstrate recently a compact full-field soft X-ray transmission microscopy with sub-60 nm resolution, operating at 100 Hz repetition rate, with exposure times of less than 5 min using both dry and wet samples [37]. Tomographic high resolution imaging was also performed in the “water-window” spectral range with a diatom, acquiring a tilt series of 53...
images covering 180° with half-period spatial resolution of the tomogram approaching 140 nm [38]. Using a Schwarzschild reflective objective, with 32x magnification and NA = 0.2 images of test objects were acquired with a half-pitch spatial resolution better than 0.5 µm [39]. Those examples are only a small fraction of existing work, but due to the obvious reasons we cannot present it all herein.

3.1 “Water window microscopy system based on argon gas-puff target plasma source

3.1.1 Argon plasma based SXR source and microscope experimental setup

The “water-window”, argon-plasma based microscope was equipped with an ellipsoidal SXR condenser coated with nickel, to focus SXR radiation onto an object. A Wolter type-I reflective objective was used to form a magnified image onto an SXR-sensitive CCD (charge coupled device) camera in transmission mode. The use of the gas puff target eliminates debris production problem associated with solid targets. Radiation from the “water-window” spectral range was selected by a titanium filter. Test objects – two distinct patterns of copper meshes were imaged with a half-pitch spatial resolution approaching 1 m in a very compact set up and with short exposures. The experimental setup of the SXR microscopy system is shown in Fig. 9 and was described in more details in [40].

Existing gas puff target EUV source was modified for efficient emission of SXR radiation, including the “water-window” spectral region, reported in [41]. This source has an advantage over other compact sources that it is a debris-free source and has a possibility to change the working gases, thus allowing changing both the peak emission wavelength and the inverse relative bandwidth of the emission.

Fig. 9 Scheme of the system a), photograph with indicated components b). Image taken from [40].

For the SXR microscope Ar plasma was produced by focusing of the pumping laser pulses, from Nd:YAG laser (Eksma), with duration of 4 ns and energy of 0.74 J by an f = 25 mm focal length lens onto a gas puff target. The target is formed by two circularly symmetric nozzles. The inner nozzle, 0.4 mm in diameter, injects a small amount of working gas (argon) into the vacuum. The outer nozzle, ring-shaped 0.7-1.5 mm in diameter, injects a small Z-number gas, in our case helium, to narrow down the flow of the working gas, reducing its density gradient along the normal to the nozzle axis. The nozzle axis was positioned almost concentrically with the laser focal point, displaced 0.1 mm in the direction opposite to the condenser optic, to reduce the absorption of SXR radiation in a neutral gas from the target. The distance from the focal point to the nozzle plane was 1.5 mm to avoid nozzle damage by plasma formation. The plasma radiates in a very broad range of wavelengths, including SXR region and by using additional spectral filtering it is possible to tailor the spectral emission of the source. The source can operate up to 10 Hz repetition rate. A pressure of 1.5 x 10⁻⁴ mbar was constantly maintained in the microscope chamber during the source operation. The experimental setup is extremely compact. The microscope is located inside a vacuum chamber, 60 cm in diameter and 35 cm in height and the entire system fits on top of a single 1.8 x 1.2 m² optical table.

Radiation from the plasma was collected and focused by an ellipsoidal, axi-symmetrical nickel coated condenser mirror, developed by Rigaku, Inc. The condenser is a broad-band optic, capable of efficiently reflecting radiation from the EUV range down to SXR region. The distance between foci of the condenser was equal to 270 mm. The distance from the plasma, positioned in the first focal plane to an entrance plane of the condenser was 140 mm, while the distance from an exit plane to the second focal plane – 60 mm. To spectrally narrow the emission from argon plasma a 200 nm thick, 10 mm in diameter, free-standing titanium filter (Lebow) was used, positioned 21 mm downstream the condenser. Spectrally filtered radiation illuminates the sample, positioned 60 mm downstream the condenser, in its second focal point. Then the sample is imaged onto a SXR sensitive back-illuminated, 1024 x 1024 pixels, 13 x 13 µm² pixel size, CCD camera (i-Kon, DO-934N model, from Andor) by a Wolter type-I reflective objective [42]. The objective is composed of two axially-symmetric ellipsoidal and hyperboloidal nickel coated mirrors. The object plane of the objective, which coincides with the sample plane, is located ~150 mm from its entrance aperture, 15 mm in diameter with 14 mm in diameter central beam stop. The image plane is ~2190 mm from the objective’s exit aperture, 17 mm in
diameter with 15.5 mm diameter central beam stop. The magnification of the objective is thus equal to 14.6x and the image pixel size is 890 x 890 nm². Exit numerical aperture of the condenser is almost twice the objective entrance NA, providing incoherent illumination [2]. The condenser, sample stage and objective were mounted on three axis translation stages driven by vacuum compatible step motor actuators (Standa). To obtain a single image in the “water-window” spectral range 50-100 SXR pulses were necessary, at 10 Hz repetition rate. During image acquisition the CCD camera was cooled down to -10 °C to decrease the intrinsic, thermal noise of the detector.

Measurements of the source photon flux were performed using commercial AXUV100 silicon p-n junction photodiode, from International Radiation Detectors, Inc., and corrected for transmission of the Ti filter. Subsequent measurements yielded the number of photons in the focal plane of the condenser, equal to 3.6 x 10^10 photons/pulse, which corresponds to 2.3 µJ/pulse in band. Ar emission spectrum was measured in the wavelength range from 2-16 nm. It contains number of transition from 2.7-4.4 nm wavelength identified according to the data reported in [43], right in the range of “water-window”, usually defined as the wavelength range from 2.3-4.4 nm. A spatial distribution of the Ar plasma in the “water-window” spectral range was obtained using a pinhole camera. The measurements were performed with Ti filter, to assess the plasma size particularly in the “water-window” range. Laser drilled, 32 µm in diameter pinhole was positioned 291 mm from the plasma and 280 mm from the CCD camera (X-vision M25, Reflex s.r.o., Czech Republic), which was equipped with 512 x 512 pixels CCD chip, 0.5 x 0.5 in² in size. This results in a lateral magnification of 0.96x. For the measurements 300 SXR pulses were required. The argon plasma FWHM size was measured to be 240 x 130 µm².

3.1.2 Experimental results

As test objects, two transmission electron microscope (TEM) grids (meshes) (Tesla, Czech Republic) of different geometries were used. Typical scanning electron microscope (SEM) images of the objects are shown in Fig. 10.

![SEM images of the objects: square mesh a) and rectangular mesh b), from [40].](image)

A square-shaped mesh, shown in Fig. 10a), has a period of (123.8±0.5) µm and a bar with width (42.9±0.7) µm, while a rectangular mesh, shown in Fig. 10b), has a period of (83.2±1.4) µm and a slit width of (37.1±0.8) µm. The errors are associated with the accuracy of grating fabrication. The thickness of both grids was ≈13 µm. Dashed boxes indicate the regions of the sample imaged with the “water-window” microscope. To obtain the sharpest possible image in the “water-window” spectral range, series of images was recorded at various sample-objective distances, in the range of ~ +/−2 mm from the focal point of the objective. From the entire set of images, the “sharpest” SXR image was chosen for subsequent resolution measurements. Resolution of the microscope was also assessed by KE test.

![SXR images of the objects a), b), depicted in Fig. 10, obtained in the “water-window” spectral region. Dotted line indicates region where a lineout was made to assess spatial resolution based on the KE test. KE resolution test result c) showing Rayleigh resolution equal to 2.5 pixels = 2.2 µm or half-pitch spatial resolution equal to 1.1 µm.](image)

Typical images of the meshes, obtained under argon plasma illumination, filtered by 200 nm thick Ti filter are shown in Fig. 11a), for square mesh and b) for a rectangular mesh, respectively. Black dotted line, depicted in Fig. 11a), indicates the region, where subsequent KE resolution measurements were carried out. A typical KE lineout is depicted in Fig. 11c), where 10-90 % intensity transition in the normalized lineout through the sharp edge in the image is equal to
3.1.3 Imaging simplest biological material

Additionally, using the experimental SXR microscope the first images of biological structures (onion cells) were obtained. Fig. 12 shows the results of imaging using the optical microscope - image on the left, and in the "water window" spectral range using Ar plasma based SXR microscope - the images on the right. The spatial resolution should be improved in the future, however, undoubtedly the advantage of this system is the possibility of imaging in the "water window" relatively thick samples, in which case a thickness of approximately 40 microns, which is impossible when using EUV radiation range.

![Image of onion cells obtained using an optical microscope (picture left) and SXR microscope (images on the right)](image)

3.2 “Water window microscopy system based on nitrogen gas-puff target plasma source

In this section the laser-plasma SXR source, based on nitrogen gas-puff target, emitting quasi-monochromatic, incoherent radiation, in the WW spectral range and Wolter type-I objective was used to image various objects in the „water-window” spectral range. Wavelengths, near K-α oxygen edge are preferable due to largest absorption length of water, ~10 μm, which allows to image thicker samples. Additionally, monochromatic emission makes easier for any data processing, since only one wavelength needs to be considered. Herein we present a system, which captures magnified images of the objects with 850 nm half-pitch spatial resolution, for sample thicknesses of ~40 μm and exposure time as low a few seconds. Current setup allows for imaging in „water-window” spectral range using quasi-monochromatic radiation at \( \lambda = 2.88 \) nm, in a very compact table-top scheme, which might be of particular interest to various fields of science and technology.

3.2.1 Nitrogen plasma based SXR source and experimental microscopy setup

The experimental setup of the nitrogen-based SXR microscope system, described in details in [44] is very similar to one in paragraph 0., with the change of working gas from Ar to nitrogen. Measurements of nitrogen-based source photon flux in the focal point of the condenser were performed using commercial AXUV100 silicon p-n junction photodiode (International Radiation Detectors, Inc.), with Ti filter in place. Measured number of photons was equal to \((3.86 \pm 0.23) \times 10^9/pulse\) in the transmission band of the filter \((\lambda = 2.8 - 6 \text{ nm})\), which corresponds to energy of \(256 \pm 15 \text{ nJ/pulse}\) in band (20 independent measurements). The nitrogen plasma FWHM size was measured to be \(0.31 \times 0.47 \text{ mm}^2\) using a pinhole camera. Nitrogen plasma emission spectrum was measured in the wavelength range from \(\lambda = 1.5 - 6 \text{ nm}\). Measured spectrum is quasi-monochromatic, consisting of He-like nitrogen line at \(\lambda = 28.787 \text{ Å}\) and H-like line at \(\lambda = 24.779 \text{ Å}\), according to data reported in [42].

3.2.2 Experimental results and imaging of biological specimen

As test object, a square-shaped transmission electron microscope (TEM) mesh (Tesla, Czech Republic) was used. Typical scanning electron microscope (SEM) image of the object is shown in Fig. 13a), similar to Fig. 10a). The errors are associated with accuracy of grating fabrication. The thickness of the mesh is ~13 μm. Dashed box in Fig. 13a) indicate a region of the sample imaged with the “water-window” microscope. In order to refocus, a set of images was recorded at various sample-objective distances, ~ +/-2 mm from the focal point of the objective. From the series, the “sharpest” SXR image was chosen for subsequent resolution measurements using KE test. Typical SXR image of the mesh, binned 2x2 for better S/N ratio, is shown in Fig. 13b).
White dotted line indicates the region, where subsequent KE resolution tests were carried out. A typical KE lineout is depicted in Fig. 13c), where 10-90 % intensity transition in the normalized lineout is equal to ~1.7 μm (Rayleigh resolution). The value of half-pitch spatial resolution was assessed statistically, based on 10 independent measurements, and is equal to 0.85 +/- 0.12 μm. Some examples of “water-window” images of biological objects were obtained as well. SXR image of an onion skin cells are depicted in Fig. 13d). It depicts a few interconnected cells with dark cell walls and white cell interior. The advantage of “water-window” optical contrast near the oxygen-edge is obvious, since the sample thickness in this case is ~40 μm, which makes it completely opaque in the EUV spectral range for example.

3.3 Discussion of the results

The discrepancy between theoretical resolution of 17.1 nm (at λ = 2.88 nm – peak emission from nitrogen plasma), assuming incoherent illumination, and obtained mean value of 850 nm can be explained by several factors. Most important is that the Wolter objective is not a full aperture optic, but has a central beam block, forming a thin, annular shaped entrance aperture. This yields a point spread function (PSF) with slightly narrower central lobe, but much more pronounced secondary lobes, yielding half-pitch resolution equal to 304 nm, which is ~18x the diffraction limit. Second factor is the influence of modulation transfer function (MTF) of the CCD camera. Convolution of the two factors yields half-pitch resolution ~480 nm. Additional discrepancy is probably due to surface curvature errors, resulting in various aberrations of Wolter objective, optical quality of its mirrors, and finally surface roughness of the two-mirror optical system, being of highest importance [45].

4. Conclusions

In this chapter laser-plasma gas puff target sources were presented, suitable for EUV and SXR microscopy. Microscopy with Fresnel zone plates, requires a monochromatic radiation, which good approximation might be a quasi-monochromatic emission from Ar plasma EUV source at 13.84 nm wavelength. This source was actually used in the microscopy experiments yielding finally ~50 nm spatial resolution [27]. Applications of the EUV microscope in material science were also presented. Practical, compact and high resolution EUV full-field microscope that might be developed in the future might allow the study of ultrafast processes with nanometer-scale resolution and have a chance to become an enabling tool for a wide range of nanoscience and nanotechnology applications.

Ar-based and nitrogen-based SXR sources were successfully employed for SXR microscopes with reflective, dispersion-free Wolter-type I objective. The systems allow capturing magnified images of the objects, with magnification of ~15x, half-pitch spatial resolution down to 0.8μm and short exposure time. We also presented preliminary results of imaging biological material. Additionally, the lack of condenser and objective dispersion allows in-situ to change spectral emission from the source (by changing the working gas or the filters) and to perform imaging in the EUV range, λ = 17-70 nm using Al filter for example. This opens new possibilities to obtain different spectral information about the objects.
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References


