Femtosecond-Laser-Driven Cluster-Based Debris-Free Soft X-ray Source for Nanostructure Imaging

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Intense soft X-ray emission was obtained from the plasma produced by the irradiation of the clusters (10% CO_2 + 90% He gas mixture) by femtosecond laser pulses. Soft X-ray flux of the strongest Oxygen spectral lines (~ 1.9 nm) reaches $2.8x10^{10}$ photons/(sr · pulse) (~ 3 μJ) and corresponds to the brightness $1.6x10^{23}$ ph/s/mm^2/mrad^2/0.1%BW. Modeling of the radiation spectrum shows that the total X-ray flux of this polychromatic source in the 1-30 nm spectral regions is 2 - 3 orders of magnitude higher than the flux of the single Oxygen spectral line. Absorption images of the samples with micro- and nanoscale features illuminated by the developed source were recorded by the LiF crystal soft X-ray detector. Radiography experiments show that this debris-free plasma source could be particularly useful for the imaging of the ultrathin (nanoscale) foils or biological structures. Even if the foil is essentially transparent for the soft X-ray radiation (like 100 nm thick Zr foil) image contrast could be significantly increased due to the influence of phase-contrast effect by placing the detector at the proper distance from the sample.

Keywords: fs laser interaction with clusters, laser plasma X-ray sources, phase-contrast soft X-ray imaging.

1. Introduction

Among different applications of the x-ray sources micro radiography is one of the most important and allows obtaining hidden information about internal sample structure. At the same time traditional conventional radiography based on the hard X-ray sources has some serious limitations due to the low absorption of such radiation in ultrathin samples or samples consisting of materials with low atomic numbers. To overcome this problem phase contrast techniques were developed [1]. Typically phase contrast method uses X-ray optics or free space propagation [2] for the recording phase changes on the propagated wave, which induced by the sample. However both such techniques require sources, which produce either spatial or temporal coherent radiation. Another possibility to increase imaging contrast is shifting of the energy spectrum of the probing radiation to the lower energies, which allows one to obtain good absorption images of the ultrathin structures due to the high interaction efficiency of the soft x-rays with a matter. Indeed a low energy radiation like soft X-ray or VUV is fully absorbed already by few micrometers of material which intensify investigation of such samples. Thus soft X-ray in the 1-30 nm spectral regions is the best candidate for the imaging of ultrathin sample.

There are few types of the laboratory scale sources which can produce radiation in this spectral region: X-ray lasers, high order laser harmonics and laser produced plasmas [3-5]. In spite the fact that all of these sources are suitable for the imaging applications only laser produced plasma (LPP) can be used for the imaging of the large (up to few cm^2) samples since it emits radiation in the whole spherical angle 4π. The LPP source size, radiation spectrum, X-ray flux, the brightness and the quantity of parasitic debris will depend on the used laser and on the target material. Plasma produced by a femtosecond laser radiation [6, 7] is a very attractive X-ray source because the size of such plasma usually is a very small one and lower quantities of debris are produced. Especially challenging to use as a target cluster media, which is debris free, can significantly increase the efficiency of fs laser pulses coupling into the target [8,9] and generate an ultrabright X-ray radiation.

In the current work we used as a target cluster media (mixture of 90% atomic He and 10% molecular CO_2 gases) irradiated by fs Ti:Sa laser pulses to create bright debris-free plasma X-ray source which emits mainly in the

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1-30 nm spectral region and allowed us to achieve a very high X-ray flux and the source brightness. Parameters of this source were measured using the x-ray Focusing Spectrometers with Spatial Resolution (FSSR). The laser plasma source was applied for the absorption radiography of the different samples with dimensions up to ten's mm^2. The LiF crystal detectors [10] were used for the registration of the 2D soft x-rays images. Obtained images had 700 nm spatial resolutions and allow resolving thickness variations on the 10 nm scale. We also observe contrast enchantment in the images of the transparent objects when we placed detector at some distance from the sample. Propagation based phase contrast was the mechanism of the contrast improvement in this case. It was possible due to the partial spatial coherency of the developed LPP source.

2. The characterization of the soft X-ray source

Different types of nanostructure targets have been proposed recently to increase the efficiency of fs laser pulses coupling into the target and consequently to enhance generation of X-ray emission. The simplest way to produce targets with nanoscale dimensions is to use the expansion of different gases in supersonic nozzles [11]. The mixture of He gas with some molecular gases is a very attractive for production of clusters target. Indeed, such gases combination will give the advantage in clusterization process due to the expansion of molecule gas together with He gas [12]. In addition, such mixture is optically thin for the useful SXR and VUV emission generated by cluster plasma.

In our experiments especially designed [13] supersonic gas jet nozzle with input and output diameters of 0.5 and 2.0 mm, respectively, and a length of 75 mm was used to produce clusters from pure CO_2, N_2O or mixture of 90% He and 10% CO_2 gases. Big clusters with diameter up to 1 μm have been produced by such nozzle. To heat the clusters the 36 fs Ti:Sa laser pulses with energy of 160 mJ were focused in the focal spot about 50 μm at the distance 1.5 mm below the nozzle outlet.

Intensities and spatial dimensions of the Oxygen Ly_α (653.7 eV) and He_β (665.7 eV) lines were measured by two FSSR Spectrometers. These spectrometers allowed measuring of the plasma dimension, which emits soft X-ray radiation in the direction of laser beam propagation as well as in the transversal direction. The measured number of photons produced by the mixture cluster target was 2-8 times higher compare with the pure CO_2 or N_2O cluster targets and reaches values of $2.7x10^{10}$ and $2.8x10^{10}$ ph/sr·pulse (about 3 μJ) for Ly_α and He_β, respectively. The brightness of both lines is also higher for the used gas mixture and has $1.6x10^{23}$ and $1.3x10^{23}$ ph/s/mm^2/$mrad^2$/0.1%BW.

Modeling of the obtained spectrum made it possible to measure electron temperature (~ 110 eV) and electron density (~ 10^{20} cm^{-3}). Using such plasma parameters modeling of soft X-ray emission in the spectral range (1-30 nm) was done using FLY code [14]. It was found that the 98% of the generated photons (including the photons from bremsstrahlung) are in the 1-5 nm spectral range and the total number of photons in this spectral region is at least 10^{12} ph/sr·pulse.

2. Absorption images

Soft X-ray images of the different samples were registered by means of LiF crystal detectors [10]. The 2-mm-thick LiF crystals have circular shape with a diameter of 20 mm. Investigated samples were placed in the same holder with the LiF detector in contact with the crystal surface or at some distance from it. Sample holder were mounted in the vacuum chamber at the different sides of LPP source at the distance 230 - 500 mm far from it. At such distances whole surface of the sample was uniformly illuminated. Thus we were able to obtain 2D SXR images of the samples with submicron spatial resolution over entire crystal surface. Due to the high X-ray flux, several minutes (about 1000 laser shots) were enough to produce high quality images.

To demonstrate that such SXCI system has high spatial resolution on the big field of view and high dynamic range test sample with 1500 lpi mesh and dry mosquito was made (fig. 1). Sample was placed in contact with LiF detector (diameter 20 mm).

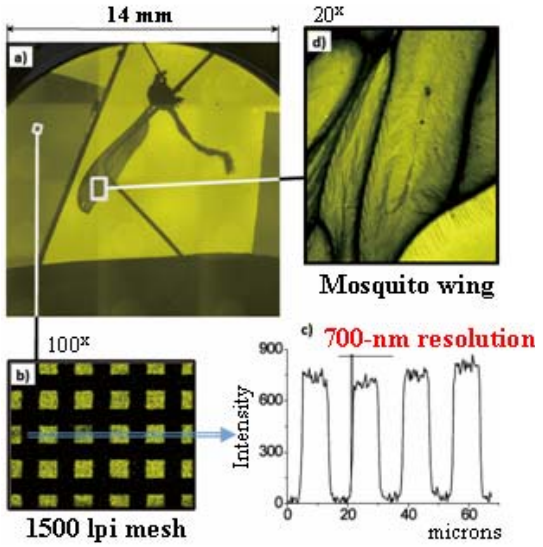
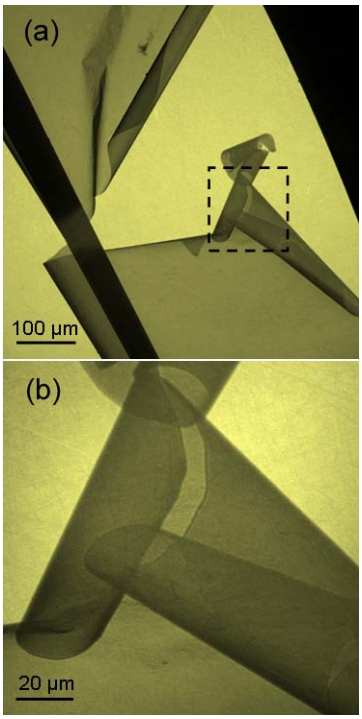


Fig.1 Images of the 1500 lpi mesh and mosquito: (a) On the large field of view; (b) Portion of image was read-out with the 100^x optical magnification; (c) intensity profile demonstrating 700 nm spatial resolution; (d) Mosquito wing fragment read-out with 20^x magnification. Hairs with dimensions 2-3 μm are clear seen.



Luminescent patterns registered at LiF crystal were readout using fluorescence microscope (FV300, OLYMPUS). To stimulate luminescence a 488 nm Ar laser was used to pump LiF crystal while image was readout in the spectral range > 510 nm. Despite the not perfect matching of pumping laser wavelengths with absorption band of LiF (see [6,10] for details) obtained luminescence was strong enough and images have good space resolution in the full area of the LiF detector (see part of the image in Fig.1). Spatial resolution about 700 nm was obtained for both open and covered by polypropylene 1500 dpi meshes (Fig.1b,c) located at different parts of LiF crystal. Image of the dry mosquito has also high contrast and spatial resolution. Tiny details of wing structure (micron sized hairs) are clearly resolved throughout the whole wing (fig. 1d). Then we applied our soft X-ray source for the imaging of the ultrathin foils. For example, in the fig. 2 there are absorption images of the 100 nm thick zirconium foil. The foil structure is perfectly visible in these images. One can easily find overlapping of a few foil layers and swirl peaces in the small-scale image (fig 2b).

3. Phase-contrast imaging

However, we found that in some cases even soft X-rays cannot make good absorption images of the ultrathin foils which consist of materials with low atomic number *Z*. For example, we obtain contact image of the 1000-nm thick polypropylene foil and it had a very poor contrast (fig 3a).

Then we placed detector 3 mm far from the sample. Recorded image (fig. 3b) had higher contrast. It means that due to the free propagation of the wave between sample and detector planes the image contrast has been enhanced. This is well known phenomenon, named propagation based phase contrast (PBPC). However, as it was shown previously for the hard X-ray sources, the spatial

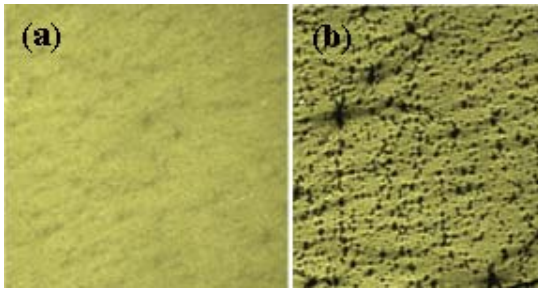


Fig.3 Image of the surface of the 1 μm thick polypropylene foil (0.7x0.7 mm fragment): (a) Contact image, zero distance between source and foil, (b) Phase-contrast image, detector placed 3 mm far from the source.

coherency of the X-ray radiation is necessary to create PBPC images [2]. As it is well known, generally laser produced plasma radiation is not temporally coherent. But in the case of registration radiation at big distances from the LPP source some spatial coherence appears. We found, that the soft X-ray radiation of our fs LPP source has also some spatial coherence at big distances from the target. For example, we observed diffraction fringes at the vacuum-foil interface (edge) in the images of the Zr foil when the distance between source and detector was 500 mm and between foil and detector - 3.8 mm (fig. 4). We modeled soft X-ray intensity near the foil edge by calculating Fresnel diffraction integral. Good agreement between calculated and measured intensity (dashed and solid lines in the fig. 4d, correspondingly) was achieved assuming 200 μm source diameter and uniform broad spectrum in the 1-20 nm spectral region. Our modeling shows that spatial coherency lengths reached ~ 2 μm in this particular case. Used for modeling values differ from the source size and spectrum which was found by the modeling of the Oxygen spectral lines profiles. Difference in the plasma source size explained by the limited spectral range of the spectrometers, practically source size is larger due to contribution of the less ionized ions. Also spectrum used for the fringe modeling appears to be more uniform in the wide spectral range then the spectrum predicted by the steady state kinetic code. This is probably due to low intensity radiation at the large wavelengths ($\lambda > 5$ nm) which has a much larger lifetime then the intense K-shell lines of the multicharged ions (1 - 5 nm) thus compensating difference in the intensity. So, modeling of the SXR images could provide one with additional information about plasma size and spectrum.

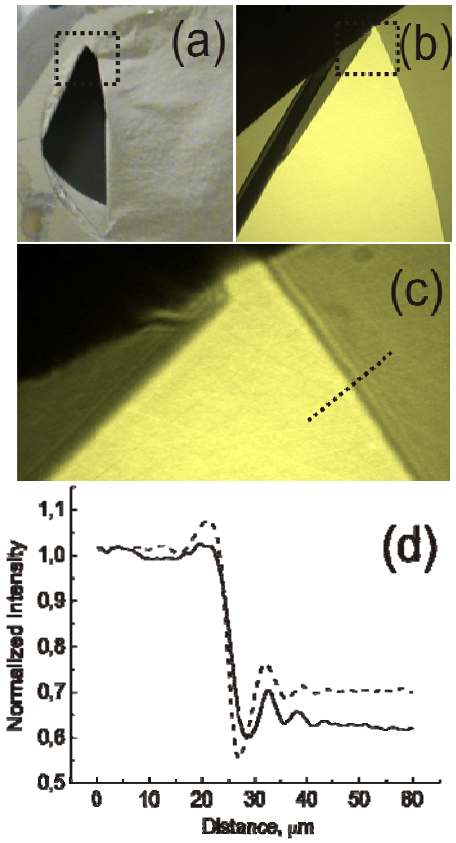


Fig.4 Image of the 100 nm thick Zr foil obtained on the LiF crystal placed 3.8 mm far from the foil: (a) Photo of the sample with the damaged part; (b), (c) Soft X-ray image fragments of the damaged part (read-out with 10^x and 40^x magnifications, respectively); (d) Solid curve – experimental intensity profile at the vacuum-foil interface showing oscillations, dashed curve – intensity calculated at the foil edge.

We observed other interference phenomenon at the images obtained with FLP source. For example, in the fig. 5a there is the image of the local inhomogeneity at the surface of the 500 nm thick Zr foil. Image was obtained at the LiF detector placed 3.8 mm far from the foil. One can notice the light spot in the center of the shadow and light stripe around this circle defect. We calculated image of the circle defect with 10 μm diameter and thickness 20 % larger than the thickness of the rest foil. Modeled (fig. 5b) and experimental image are in the good agreement.

4. Summary

Detailed study of the subpicosecond debris–free cluster plasma soft X-ray sources was performed. To maximize the x-ray flux radiation particular attention was devoted to the optimization of the nozzle and gas parameters. Investigation of the Oxygen spectra in the 1.84-1.92 nm showed that the x-ray flux in the strong Oxygen lines was about 2.8×10^{10} ph/sr·pulse while a peak

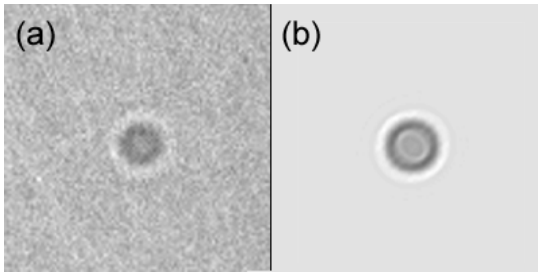


Fig.5 Images of the defect (thickness variation) in the 500 nm Zr foil. Diffraction maximums are clearly visible both in the (a) experimental and (b) calculated images.

brightness reached 1.3×10^{23} ph/s/mm^2/mrad^2/0.1%BW. Developed polychromatic source was applied for the micro radiography of the biological specimen and ultrathin foils. Soft X-ray images were registered by the LiF crystals, which allowed achieving submicrometer spatial resolution and high contrast even in the contact images. LiF crystal detectors placed at some distance from the investigated transparent samples detected enhancement of the images quality due to the propagation based phase contrast mechanism.

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