Generation and application of the soft X-ray laser beam based on capillary discharge

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Abstract. In this work we report on the generation and characterization of a focused soft X-ray laser beam with intensity and energy density that exceed the threshold for the ablation of PMMA. We demonstrate a feasibility of direct ablation of holes using a focused soft X-ray laser beam. Ablated craters in PMMA/gold-covered-PMMA samples were obtained by focusing the soft X-ray Ar⁺ laser pulses generated by a 46.9 nm tabletop capillary-discharge-pumped driver with a spherical Si/Sc multilayer mirror. It was found that the focused beam is capable by one shot to ablate PMMA, even if the focus is significantly influenced by astigmatism. Analysis of the laser beam footprints by atomic force microscope shows that ablated holes have periodic surface structure (similarly as Laser-Induced Periodic Surface Structure) with period ~2.8 μm and with peak-to-peak depth ~5-10 nm.

1. Introduction
Table top soft X-ray lasers based on capillary discharge produce hot and dense plasma which is a brilliant source of the EUV and soft X-ray radiation. For many years, researchers are looking for compact tabletop soft X-ray lasers that could be routinely used in application in numerous disciplines including

- physics of thin layers and physics of surfaces/interfaces,
- biological applications including imaging of cellular and sub-cellular structures, studies of membranes, radiation damage studies,
- materials science for real-time lattice dynamics studies,
- phase transition studies,
- element-state-selective radiation chemistry (so called molecular surgery),
- radiography, interferometry, shadowgraphy of dense plasmas with micrometer resolution,
- real-time studies of fast chemical reactions and liquid-phase chemistry,
- non-linear optics at ultrashort wavelengths,
- precise studies of atomic physics phenomena such as photoexcitation, photoionisation, inner-shell processes and threshold Raman processes,
development and testing of new kind x-ray detectors for various applications e.g. in biomedicine and astrophysics.

In future, when the wavelengths of soft X-ray radiation drop below 1 nm, the following applications will also be realized:

- a time integrated diffractometry of biological and inorganic materials,
- a medical diagnostic by coherent soft X-rays.

Moreover, certainly some of important applications still remain unforeseeable.

With demonstrated multi-hertz repetition rate operation and average powers of several mW and mJ-level pulse energy at 46.9 nm, capillary discharge-pumped lasers are the first compact lasers that reached this goal [1].

It is necessary to realize that besides laser-pumped and discharge-pumped soft X-ray lasers there are several alternative possibilities how to achieve coherent radiation in soft X-ray region: synchrotron sources [2]-[3], free electron lasers (FEL’s) [4]-[5] and harmonic up-conversion of high-power visible/IR lasers [6]-[7]. However, due to their size and cost - these sources are unsuitable for massive application.

In this work, we report on the ablation of PMMA/gold-covered-PMMA samples irradiated with an intense focused 46.9 nm laser beam. Firstly, the soft X-ray laser beam was focused on PMMA sample covered by a thin (transparent) layer of gold. In this case obtained footprints show that energy of the soft X-ray laser beam is sufficient for ablation of this layer (unfortunately, the focus is influenced by astigmatism). Secondly, PMMA sample was ablated by soft X-ray radiation emitted from the capillary. It was demonstrated that our Ar$^+$ laser with wavelength of $\lambda = 46.9$ nm (the energy of which has not yet been reliably measured, but is estimated to be tens or hundreds of $\mu$J) is capable to ablate PMMA/gold-covered-PMMA samples by a single shot.

2. Experimental apparatus

The high current flowing through the capillary results in pinching of the Ar plasma to form the hot thin plasma column along the axis. The experimental setup is shown schematically in Figure 1. The high current pulses used for excitation of the capillary plasma are produced by a Marx generator, coupling section and pulse forming line. This water filled fast capacitor is discharged through a self-breakdown spark gap pressurized with SF$_6$ gas to alumina capillary. The ~23 cm long capillary is directly joined to the spark gap (having one common electrode). The gas filling and pumping part is

![Figure 1 – Schematic presentation of experimental setup used to focus soft X-ray laser beam](image-url)
attached to the outer end of the capillary (through the orifice in the outer electrode). A pre-ionization current precedes the main discharge (typically it is initiated 10 μs prior to the spark gap breakdown and its current has usually amplitude ~10 A) and it is produced by an independent circuit. A detailed description of our capillary discharge driver CAPEX can be found in the recently published paper [8].

The laser beam is focused with a spherical (R=2100 mm) Si/Sc multilayer-coated mirror placed in a vacuum chamber. In our first experiments footprint of the soft X-ray laser beam was on-line detected by a “phosphor” screen (ZnS, density 8 mg/cm²)/photographic camera assembly. Nowadays, the soft X-ray laser beam is visualized by ablation of easily ablative material (PMMA/gold-covered-PMMA samples).

3. Experimental results
At first, the focused laser beam was registered in a visible region by the “phosphor” screen, which was covered by 0.4 μm aluminium filter to avoid the detection of visible plasma radiation [9]. It was found that the far field laser beam profile with a diameter ~5 mm has an annular shape (similarly as in [10]). In case of near field the output laser beam profile with a diameter ~ 0.6 mm has an intense single peak. Furthermore, the beam profile in the region around laser beam waist was influenced by astigmatism. In consequence of possible ambiguous interpretation of these results in visible region we are obliged to look for new diagnostic tools. During the later experiments PMMA/gold-covered-PMMA samples were selected as a target for focused soft X-ray laser beam.

First experiments with ablation of PMMA sample show that the focused beam of our laser is capable to ablate PMMA by one shot, even if the focus is significantly influenced by astigmatism (see Figure 2 – visible photo-pictures taken through optical microscope with geometrically reflected illumination) [11]. Analysis of these footprints by optical microscope between crossed polarisers shows that the sample has no residual stress caused by local heating. It seems that the ablation is not a result of thermal effects of radiation, but of quantum ones only. Unfortunately, following analysis of ablated PMMA samples by scanning electron microscope was not successful. Finally, the laser beam footprints were analysed by atomic force microscope (AFM). It was found that ablated part has on its

Figure 2 – PMMA ablated by one shot of Ar⁸⁺ laser (Left), PMMA ablated by five shots (Middle), PMMA ablated through Ni grid (step 100x100 μm, free windows 70x70 μm, traverses 30 μm) by five shots (Right); all these three pictures are in the same scale and with false colours.

Figure 3 – Analyzed parts (150x150 μm) of the spot shown in the middle of the Figure 2: a) 3D (Left) and 2D (Right) region in the right corner. b) 3D (Left) and 2D (Right) region in the middle-bottom part of the spot. Both parts contain the ablated and the not-ablated regions.
surface more or less contrast periodic structure with period ~2.8 μm and with peak-to-peak depth ~5-10 nm (see Figure 3). It offers to attribute these structures to LIPSS-II (“incoherent” Laser-Induced Periodic Surface Structure) [12], which depend mainly on fluency and interaction time of the laser beam, in other words on “melting effects” of the radiation. Contrary, LIPSS-I (“coherent”) depend on the wavelength, coherence, polarization and incidence angle of the laser beam [13].

The last experiment should have answer, how grid with smaller characteristic pattern influences LIPSS. Therefore, PMMA sample was ablated through Au grid (step 12.5x12.5 μm, windows 7.5x7.5 μm) (see Figure 4 a)). Analysis of these laser beam foot prints exposed through Au grid was realized by atomic force microscope. The period of the diffraction pattern (see Figure 4 b)) changes from ~800 nm (at the edge of window) down to ~125 nm (in the middle of the window). A clearly visible diffraction pattern (esp. at the edges) suggests that hardly any melting takes place.

As already mentioned, analysis of our first ablated PMMA samples by scanning electron microscope was not successful, but for this analysis the PMMA sample was covered by a thin (~40 nm) layer of gold. As it turned out, already the first test shots at this layer showed that the energy of our laser is sufficient for ablation of this layer. Gold-covered-PMMA samples were analyzed by AFM microscope. Typical ablated laser beam footprint (~100 μm) at focus (~1427 mm from the mirror) is shown in Figure 5 a) (Left). Laser beam footprint (~250 μm) taken far from the focus (~1450 mm from the mirror) has an annular shape similarly as in earlier experiments in visible region (see Figure 5 a) Right). Phase contrast on the right-top image in Figure 5 b) indicates that physical properties of underlying substrate are significantly different from those of the gold film. Analysis of the ablated spot in AFM working in extended TUNA mode (in which local conductance was
Figure 6 – Laser beam footprints on gold-covered-PMMA, the images were taken through optical microscope (grid 125x125 μm/div); false colours. (Middle and Bottom right images in Figure 5 b) show that the composite area, where gold is intermixed with PMMA, spreads several microns from the border of the laser spot. Any melting effects on the edge of ablated laser beam footprint are not observed. We scanned the laser beam footprint in Z-direction trying to find the best focus. It was found that astigmatism of our soft X-ray radiation is very strong, astigmatic difference being ~16 mm (see Figure 6). Later it turned out that the distortion of focal spot is caused by distorted mirror shape. Analysis of the last results with ablation of gold-covered-PMMA confirmed our assumption that in our case the quantum ablation plays a dominant role and the thermal effects are negligible.

4. Conclusion

Our laboratory is one of several successful laboratories in the world, where Ar$^{8+}$ ion laser is in routine operation and where discharge based soft X-ray lasers are further studied. It was demonstrated that focused beam of our soft X-ray laser ($\lambda = 46.9$ nm) is capable by one shot to ablate PMMA/gold-covered-PMMA samples, even if the focus is significantly influenced by astigmatism and mirror distortion. It was found that in case of large ablated pattern the PMMA sample has on its surface a periodic structure with period ~2.8 μm (like LIPSS-II structure). Contrary, at small ablated pattern a diffracted pattern (with periodicity down to ~125 nm) was observed. Analysis of PMMA/gold-covered-PMMA samples by atomic force microscope working in different modes shows that the ablation is not a result of thermal effects of radiation, but of quantum ones only.

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