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Laboratory sources of soft X-rays and extreme ultraviolet (EUV) based on laser plasmas produced with a gas puff target

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Electromagnetic radiation in the soft X-ray and extreme ultraviolet (EUV) wavelength ranges can be produced in a high-temperature plasma generated by interaction of high power laser pulses with matter [1-3]. It was demonstrated that laser plasma soft X-ray and EUV sources could be useful in various applications in physics, material science, biomedicine, and technology. However, conventional laser plasma sources based on a solid target have debris production problem. We have demonstrated that using a double-stream puff target, instead of a solid target, it is possible to develop highly efficient and debris-free laser plasma soft X-ray and EUV sources [4-5]. The target is formed by injection of high-*Z* gas (xenon, krypton, argon, etc.) into a hollow stream of low-*Z* gas (hydrogen and helium) using a double nozzle. The nozzle setup consists of a central nozzle in a form of a circular orifice, surrounded by an outer nozzle in the form of a ring. The nozzle is supplied with gases from two electromagnetic valves mounted in a common body. Strong soft X-ray and EUV emissions from the double-stream gas puff targets, exceeding the emissions from solid targets, have been demonstrated [6].

In the paper laser plasma sources of soft X-rays and EUV based on a gas puff target, developed for various applications, including metrology and microscopy, photo-etching and processing of materials, surface modification, radiography and tomography, radiobiology and material damage, photoionization of gases and cold plasma formation, are presented.

The gas puff target approach was used for developing a compact laser plasma EUV source for metrology applications [7]. The xenon target was irradiated with 4 ns/0.5 J pulses produced with repetition rate of 10 Hz from a commercial Nd:YAG laser. Conversion efficiency of the laser energy into the EUV energy at 13.5 nm wavelength of about 2 % was measured in 7 % wavelength band, corresponding to about 0.5 % in 2 % band [8]. The source has been used in the measurements of optical characteristics of Mo/Si multilayer mirrors [9].

High-brightness soft X-ray source based on the gas puff target driven with the PALS laser facility [10, 11] has been used for the first time for processing materials. Direct photo-etching of inorganic (silicon) and organic (polymers) materials with nanosecond pulses of soft X-ray and EUV radiation was demonstrated [12, 13].

Efficient processing of organic polymers (PMMA and PTFE) has been also demonstrated with the compact EUV source for metrology, operating at 10 Hz [14] and strong temperature effect on soft X-ray photo-etching of PTFE was shown [15]. The same source equipped with a multi-foil optic collector [16] has been used to study the EUV emission from solids irradiated with intense EUV pulses [17]. A new technique for detection of surface changes of materials, utilizing scattered or luminescent EUV radiation, was proposed [18].

The use of a grazing incidence axisymmetrical ellipsoidal mirror as a collector strongly increased the EUV fluence on irradiated samples up to 100 mJ/cm^2 [19]. This made possible to increase dramatically the EUV ablation rates and improve micromachining of polymers. Efficient processing of non-organic materials (SI, Ge, NaCl, and CaF_2) has been also demonstrated [20]. Modification of polymer surfaces by creation of characteristic micro- and nanostructures was observed in case of irradiation with EUV pulses at relatively low fluence (<10 mJ/cm^2) [15, 21-25]. It was found that such EUV patterning of surfaces can be useful for biocompatibility control of polymers [26]. These studies resulted in development of the source dedicated for EUV processing of materials [27].

Laser plasma EUV source for processing is composed of a vacuum chamber in a form of a vertical column mounted onto a cubical base, housing a compact commercial Nd:YAG laser system (EKSPLA) generating 4 ns laser pulses with energy up to 800 mJ and vacuum pumping system. The source chamber is composed of three sections. Each section is pumped separately by oil-free vacuum pumps (differential pumping). In the first upmost section of the chamber the electromagnetic valve to produce a gas puff target and the laser beam focusing system are placed. The valve is mounted using the x-y-z translation stages, allowing placing the gas puff target in the required position with accuracy of about 10 μm. The gas puff targets are formed by pulsed injection of working gas (krypton, xenon or krypton/xenon mixture) into a stream of helium, using an electromagnetic valve system with a double-nozzle setup. The repetition rate of the system is determined by the repetition rate of the laser (10 Hz). The source is equipped with a grazing incidence axisymmetrical ellipsoidal mirror (RITE), to focus the EUV radiation. The mirror is mounted in the second, central section of the vacuum chamber. It makes possible to focus the EUV radiation onto a polymer sample mounted in the third section of the chamber, evacuated to high-vacuum. The EUV radiation is focused to a spot of about 1 mm in diameter with fluence up to 100 mJ/cm^2 for the xenon gas puff target [28].

The source has been used for EUV micromachining of poly(vinylidene fluoride) (PVDF). PVDF is an important fluoropolymer because of its piezoelectric, pyroelectric and ferroelectric properties. It is also known to have an extremely high chemical stability and electrical resistivity. Micro- or even nanopatterning of PVDF is highly desirable for applications in multifunctional and integrated devices. Many works have been performed on surface processing of PVDF using ion beams, synchrotron X-ray and UV laser radiation, however, irradiation of PVDF with these sources resulted in strong modification of the molecular structure in a near-surface layer of the polymer. Using the laser plasma EUV source dedicated for processing polymers we have demonstrated for the first time efficient micromachining of PVDF without changing the chemical structure of the unprocessed material [29]. PVDF foils of 50 μm thickness (Goodfellow) were irradiated with the EUV radiation through a contact metal mask with square orifices 60×60 μm^2. Micro holes etched through the foils have been obtained as a result of 1 min irradiation at 10 Hz repetition rate.

Investigation of the ablation products with QMS demonstrated a good agreement between the stoichiometric composition of PVDF molecules in the ablated and bulk polymer. XPS spectra, acquired for the polymer after ablation, are almost identical to the spectrum of pristine PVDF, indicating preservation of the chemical structure of the remaining material. However, XPS measurements performed on the polymer irradiated with low fluence (<10 mJ/cm^2) indicated strong chemical modification in the near-surface layer. In this case defluorination and thus carbon enrichment in the surface material was revealed [29].

We have demonstrated in our previous works that EUV radiation can be used for surface modification of polymers for biocompatibility control [26, 30]. Surface modification of PTFE, PVDF and PC polymers for biocompatibility control has been studied using the laser-plasma EUV source. Modified surfaces were characterized by SEM and AFM. Up to several hundred nanometers high wall-type micro- and nanostructures were formed [31]. Simultaneous treatment of polymer surface by EUV radiation and ionized nitrogen injected in the interaction region has been studied [32]. Chemical analysis by XPS revealed decreased oxygen contents in PC samples and nitrogen enrichment in PTFE [31, 33]. Exclusion of oxygen from polar groups leads to a polymer with increased hydrophobicity that was confirmed by contact angle measurements. Biocompatibility tests of PTFE and PVF samples modified with the EUV photons and seeded with fibroblasts have shown strong cells adhesion to polymer surfaces [34].

The compact laser plasma EUV source, additionally equipped with an ellipsoidal mirror with the Mo/Si coating and operating with the argon gas puff target, allowed producing quasi-monochromatic EUV radiation at 13.8 nm wavelengths [35]. This source has been successfully used for EUV nanoimaging with the spatial resolution down to 50 nm, proving possibility to develop a compact, desk-top EUV imaging tool [36-38]. The tool was also used for EUV imaging of nanostructures [39], crystalline thin films and nanofibers [40].

The compact EUV source has been also used for pulsed radiography and tomography of the gas puff targets. The beam of EUV radiation for backlighting of a target under study was produced by spectral selection of emission from xenon plasma using a Mo/Si multilayer mirror. To eliminate the visible light from the plasma a 200 nm thick Zr filter deposited on a 200 nm thick Si_3N_4 membrane was used. In this way quasi-monochromatic radiation at 13.5 nm wavelength with the bandwidth of about 1nm was obtained. The EUV shadowgrams of the targets were registered with the use of the back-illuminated CCD camera (Reflex), equipped with a 512×512 pixels CCD chip.

The EUV radiography setup was used for characterization of the multi-jet gas puff targets with modulated gas density for high-order harmonic generation (HHG) experiments [41] produced using the nozzles in a form of linear array of 5, 7 or 9 orifices. 2-D gas density map for the target produced using the nozzle with 7 orifices at the backing pressure of 4 bar and corresponding gas density profiles for various distances are presented in [42]. The radiography setup with the laser plasma EUV source has been also used for characterization of the dual-gas multi-jet gas puff target [43], the pulsed gas cells [44] developed for HHG experiments and the elongated plasma channels [45].

The same setup was used for the EUV tomography of the multi-jet gas puff targets. In this case the gas puff valve was mounted on top of a rotation stage to ensure 2π rotation of the nozzles while acquiring projections. The set of 900 EUV projections was used for tomographic reconstruction. The 3-D image of the multi-jet gas puff target has been obtained [46, 47]. A new technique for 3-D tomographic reconstruction of low density objects with the use of a compact laser plasma EUV source was demonstrated.

Laser plasma soft X-ray sources operating in the ‘water window’ wavelength range between 2.3 nm and 4.4 nm are used for microscopy of live biological objects. It was shown that the laser plasma source with argon or nitrogen gas puff targets is an efficient source of radiation in this range [48]. This source has been used for development of a compact, desk-top soft X-ray microscope, based on a laser plasma source, operating in the ‘water window’ range. The microscope was equipped with an ellipsoidal grazing incidence mirror coated with nickel as a condenser to focus soft X-ray radiation from the plasma onto a sample [49]. A Wolter type grazing incidence hyperboloid/ellipsoid axisymmetrical mirror was used as an objective to form a magnified image onto a CCD camera. The first soft X-ray microscopy images in the ‘water window’ spectral range of biological samples (onion skin cells) with sub-micron spatial resolution have been obtained [50].

Much better resolution was obtained using a Fresnel zone plate as a microscope objective. The Fresnel zone plate was 250 μm in diameter, with the outer zone width of 30 nm and the focal length at 2.88 nm f = 2.6 mm. Soft X-ray microscopy images of the test object (the TEM mesh) could be registered in 10 s exposure time (100 pulses). The spatial rsolution assessed by the knife edge test was equal to 60 nm. The resolution for a single-shot exposure was about 240 nm [51]. The microscope has been used for imaging of biological samples. Soft X-ray images of thin layers of saccharose (160 nm) and plasmid DNA deposited on the Si_3N_4 membrane have been obtained in 20 s and 50 s exposition times, respectively [51].

The laser plasma soft X-ray source with an argon gas puff target irradiated with the 4 ns/0.8 J/10 Hz Nd:YAG laser was developed for application in radiobiology experiments [52]. The source delivers approximately 6.83×10^{13} photons/4π in the wavelength range of about 2.5-4 nm. The low penetration depth of this radiation in biological samples and pulsed character of the source lead to high local dose loads and dose rates, respectively. The design of the source allows samples to be irradiated both in vacuum and in He-environment. Doses in a single pulse of soft X-rays of about 300 Gy for irradiation in vacuum and about 20 Gy for the He-environment irradiation were measured. Initial irradiation experiments carried out with plasmid DNA demonstrated that the source can be used in systematic studies of soft X-ray radiation damage to biomacromolecular samples and other biological specimen [53]. The source has been also used for the soft X-ray contact microscopy experiments [54].

The laser plasma EUV source for processing of materials has been also used in the first experiments on EUV photoionization of atomic and molecular gases. Gases were injected into the focus of the EUV beam using an additional gas nozzle mounted in the third section of the source chamber. Formation of low-temperature photo-ionized neon plasmas induced by nanosecond EUV pulses from the laser plasma source and by femtosecond EUV pulses from the FLASH free electron laser was studied [55]. Luminescence of helium and neon gases induced by the EUV pulses was measured [56] and significant differences between absorption spectra of neutral helium and low temperature photoionized helium plasmas have been detected [57]. Spectral investigations in the EUV/VUV region of photoionized plasmas induced in atomic and molecular gases using nanosecond EUV pulses were performed [58].

EUV photoionization of gases and formation of low-temperature plasmas have been also studied using more energetic EUV pulses generated as a result of irradiation of the gas puff target with laser pulses with time duration from 1 ns to 10 ns and energies from up to 10 J at 10 Hz repetition rate produced from the Nd:YAG laser system (EKSPLA) [59]. These preliminary investigations have shown applicability of compact laser plasma EUV sources for research in a new field of EUV-induced plasmas.

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