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Development and utilization of bright tabletop sources of coherent soft x-ray radiation

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Program Description

This project investigated aspects of the development and utilization of compact XUV sources based on fast capillary discharges and high order harmonic up conversion. These sources are very compact, yet can generate soft x-ray radiation with peak spectral brightness several orders of magnitude larger than a synchrotron beam lines. The work has included the characterization of some of the important parameters that enable the use of these sources in unique applications, such as the degree of spatial coherence and the wavefront characteristics that affect their focusing capabilities. In relation to source development, we have recently completed preliminary work towards exploring the generation of high harmonics in a *pre-ionized* medium created by a capillary discharge. Since ions are more difficult to ionize than neutral atoms, the use of pre-ionized nonlinear media may lead to the generation of coherent light at > 1 KeV photon energy. Recent application results include the first study of the damage threshold and damage mechanism of XUV mirrors exposed to intense focalized 46.9 nm laser radiation, and the study of the ablation of polymers with soft x-ray laser light.

Full characterization of the Electromagnetic Field Distribution in a Soft X-Ray Laser Beam

These compact table-top coherent soft x-ray sources open a wide field of applications in several disciplines using techniques such as interferometry, holography, and microscopy. However, despite its key role in applications, the wavefront has not been measured. With this objective a wavefront sensor for the soft X-ray wavelength range was developed and used to fully characterize the electromagnetic field distribution in a soft x-ray laser beam [1]. The sensor is based on the Shack-Hartmann concept and is composed of an array of 20×20 diffraction Fresnel lenses etched on a overcoated with a reflective multilayer (Bragg-Fresnel lenses). This design avoids optical aberrations and overcomes the limitations imposed by the strong absorption of all materials in this spectral range. Using this instrument the wavefront of a neonlike Ar soft x-ray laser ($\lambda=46.9$ nm) [2] was fully characterized in a single shot with an estimated resolution of $\lambda/100$. Figure 1a shows the experimental set up used in the measurements. The wavefront characteristics were observed to be dependent on the discharge pressure and on the capillary length, as a result of beam refraction variations in the capillary plasma. The results show a dramatic improvement of the soft X-ray laser beam wavefront with increased capillary plasma column length, leading to an improvement of the focal spot. Figure 1b illustrates the results obtained operating the discharge-pumped at a pressure of 420 mTorr, that is near the optimum for maximum amplification. The measurements show that for a 34 cm capillary about 70 percent

of the SXRL beam energy could be focused into an area about four times the size of the diffraction-limited spot, reaching intensities of $\sim 4 \times 10^{13} \text{ W/cm}^2$. The achievement of such high intensity in the soft X-ray wavelength range with a table-top device opens the possibility of new applications.

Fig.1: (a) Schematic representation of the experimental set used to measure the wavefront of a 46.9 nm capillary discharge soft x-ray laser showing the lens array and a measured image of focal spots. Measured wavefront shape (b), and corresponding intensity distribution (c) for an the laser beam produced operating the discharge at a an Argon pressure of 420 mTorr. The radius of curvature of the wavefront is 6.5 meters. The annular shape of the beam a is due to the refraction the laser beam in the plasma column. (Phys. Rev. Lett. 88, 183901 (2002))

Study of damage threshold and damage mechanism of XUV multilayer mirrors exposed to intense 46.9 nm laser pulses

High reflectivity XUV mirrors with high damage threshold mirrors are key elements for enabling numerous applications of the rapidly advancing high power coherent sources at these wavelengths, which include new table-top soft x-ray laser sources, high order harmonics, and free-electron lasers. This is particularly important as the peak power and fluence of XUV and soft x-ray sources have reached unprecedented values. For example the XUV radiation fluence at the exit of the plasma column in capillary discharge Ne-like Ar lasers operating at 46.9 nm can exceed 1 J/cm^2 [3]. Significant progress has been made in the developing of high reflectivity Sc/Si mirrors for the 35-50 nm range [4] with reflectance values as high as 43 percent in the vicinity of 47 nm [5]. However, the damage threshold of these mirrors when exposed to high peak powers of XUV light has not been studied. We have studied the optical damage mechanisms and damage threshold of Sc/Si EUV (35-50 nm range) mirrors exposed to high

power XUV laser radiation [6]. The study was conducted by focusing the output of a tabletop capillary-discharge Ne-like Ar laser emitting nanosecond duration pulses of ~ 0.13 mJ at a wavelength of 46.9 nm. The resulting damage of the coatings exposed to fluences ranging from 0.01 to 10 J/cm² was analyzed with optical microscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM), and with small-angle X-ray diffraction ($\lambda=0.154$ nm) techniques. Our results show similar values of damage threshold of ~ 0.1 J/cm² for Sc/Si multilayer coatings on Si and SiO₂ substrates, compared to 0.7 J/cm² found necessary to damage a bare Si substrate. The Sc/Si multilayers were deposited by dc-magnetron sputtering at 3 mTorr of Argon pressure on superpolished borosilicate glass. The multilayers on borosilicate glass consisted of 10 periods of Sc/Si layers, each with a thickness of ~ 26.7 nm, and a ratio of layer thickness $H(\text{Sc})/H(\text{Si}) \sim 0.7$. The multilayer coatings deposited on Si consisted of 33 periods of Sc/Si pairs with the same parameters as the borosilicate glass ones.

Figure 2 shows optical microscope images of damaged areas of coatings deposited on a Si wafer resulting from average XUV fluences of 0.15 J/cm² (a), 0.5 J/cm² (b) and 5 J/cm² (c). At 0.15 J/cm² At the lower fluences large areas with discoloration and undulations of the coating are observed. These areas are most likely produced by heat-triggered interdiffusion in the upper layers of the coatings. This surface modification, which already appears at fluences of ~ 0.08 J/cm², establishes the damage threshold for the Sc/Si multilayers defined in this work. In comparison, the onset of damage in bare Si substrates measured in this work appears at

Fig.2. SEM micrographs of the damaged areas of Sc/Si multilayer mirror coatings exposed to 46.9 nm laser beam fluences of 0.15 J/cm² (a), 0.5 J/cm² (b), and 5 J/cm² (c). *Optics Letters*, 26, 620, (2004)

significantly larger irradiation fluence of 0.7 J/cm². The areas with larger local fluences (Fig. 2b) are covered with cracks resulting from significant mechanical tensile stress generated by thermal expansion and the following cooling down process [8]. At even larger fluences of ~ 5 J/cm² the coating is fully evaporated from the center of the irradiated spot and the Si substrate is also damaged (Fig. 2c). Electron microanalysis data reveals that Sc is absent in the center part of the damaged region.

Small-angle X-ray diffraction analysis of samples irradiated with ≥ 0.1 J/cm² emission fluence shows a noticeable drop in the intensity of the diffraction peaks with respect to the unexposed areas. (Fig 3 a).

However, the peak's position remains approximately the same, indicating that the coating is only partially destroyed. This evidence suggests that while at these fluences the top layers of the coating are melted, the layers adjacent to the substrate remained unchanged. This interpretation of the X-ray diffraction data was confirmed by cross-section TEM imaging of the sample

exposed at 0.21 J/cm^2 . The TEM image of Fig.3 b shows that the top 700 nm of the coating are molten, while $\sim 180 \text{ nm}$ (7 periods) adjacent to the substrate are not destroyed. The molten layer constitutes an alloy of Sc_3Si_5 and crystal Si as determined from electron diffraction data. Analysis of the surviving multilayer coating beneath the molten layer indicates that changes in layer thickness have occurred within a distance of less than 2 periods from the molten region (Fig.3 c), thus the heat affected zone (HAZ) did not exceed $\sim 50 \text{ nm}$. Comparison of the layer structure in the HAZ with that of isothermally annealed samples indicates that the various stages of structural and phase transformations observed within a few periods of the coating under laser irradiation are the same as in samples annealed at different temperatures. The changes taking place in the Sc-containing layer nearest to the molten region (indicated by I in Fig. 3(c)) correspond to a stage of formation and crystallization of Sc_3Si_5 silicide that have been previously observed in isothermally annealed coatings at 430°C after 1 hour. In the next Sc-containing layer (indicated by II in Fig. 3(c)) only minor expansion of the ScSi silicide interface layers is observed, which is a result of solid state amorphization. Similar effects have been observed at annealing temperatures of less than 200°C .

Demonstration of an ultra-compact desk-top 46.9 nm discharge-pumped amplifier for high order harmonic amplification and other applications.

A new ultra-compact capillary discharge Ne-like Ar amplifier capable to amplify high order harmonic pulses was designed and constructed. It is designed to allow the free propagation of high order harmonic pulses along the axis for seeded operation. The amplifier provides a gain of approximately e^{10} at 46.875 nm using a 10 cm long plasma columns. An increase of the plasma column length to 20 cm produces saturation of the amplified spontaneous emission, generating an intense laser beam on its own. In this unseeded mode the laser produces pulses of up to 10 mJ of energy and $\sim 1.2 \text{ ns}$ duration. This soft x-ray laser amplifier has a dimension of about $40 \times 40 \times 15 \text{ cm}^3$ and can easily fit on top of a very small desk (Fig. 4a). It is to our knowledge the most compact source of coherent soft x-ray radiation presently available. Figure 4b shows the temporal evolution of the laser pulse measured with a vacuum photodiode. The laser beam was attenuated to avoid saturation of the photodiode. The intensity of the laser pulse is observed to

completely dominate the spectrally integrated spontaneous emission of hundreds of lines emitted by the hot dense plasma column. Fig 4c is a corresponding spectrum that illustrates the monochromaticity of the source. The observed linewidth is limited by the instrumentation, which can not resolved the estimated linewidth of $\Delta\lambda/\lambda \sim 5 \cdot 10^{-5}$. For all the tests reported herein the laser amplifier was operated at repetition rates of 0.5 to 1 Hz, but operation at higher repetition rates should be possible.

Femtosecond laser triggering of a sub-100 ps high voltage spark-gap

Applications experiments of this discharge-pumped amplifier, such as the amplification of high

Fig. 4. a) Photograph illustrating the size of saturated desk-top Ne-like Ar laser compared with a hand-held multimeter. b) Axial spectra of the capillary discharge plasma in which only the amplified laser line is observed.

order harmonic pulses requires a precise synchronization of events. To achieve necessary the low discharge jitter we have developed a high voltage spark-gap that is triggered by a femtosecond laser pulse of a few hundred microjoules of energy. Such trigger pulse can be extracted from the same Ti:sapphire laser amplifier that is used to create the high order harmonic seed pulses. We have developed a spark-gap switch with a shot-to-shot time delay statistical of $\sigma = 0.1\text{ns}$ for voltages of 10 KV [8]. At the high power levels required to pump the capillary discharge amplifier the jitter is higher, however sub-nanosecond jitter of the laser amplifier has been demonstrated under optimized conditions.

Generation of a discharge-ionized plasma channels for high harmonic generation from ions

We have conducted preliminary modeling and experimental work towards the generation of high harmonics in a *pre-ionized* medium created by a capillary discharge. Such pre-ionized nonlinear media may lead to the generation of coherent light at photon energies $> 1\text{ KeV}$. The use of ions as nonlinear media for x-ray generation was first proposed in 1973 [8] and HHG in

the strongly ionizing regime has been discussed in several works. However, only very recent work at Colorado has both definitively demonstrated that high harmonics can be observed from further ionization of an already fully-ionized gas [9] and has demonstrated that it is possible to apply techniques of quasi-phase matching to the process of HHG [10]. A periodically corrugated hollow fiber was used to avoid the destructive interference that would otherwise occur due to a phase-velocity mismatch between the driving laser and the short-wavelength light. Using this method, HHG was demonstrated in Ne at photon energies of ~ 300 eV, approaching the expected cutoff of 330 eV. This energy range is in the "water window," of interest for imaging of biological samples. However, in all this work, the ultrashort-pulse laser is doing all the "work" in ionizing the gas. Since the initial ionization generates only low-energy HHG photons, the energy expended in ionization is wasted, reducing the potential conversion efficiency. Using the phase-matched waveguide geometry, ionization-induced absorption is often a very significant limiting mechanism, absorbing as much as 50% of the incident light. Furthermore, ionization-induced defocusing is an even more significant limitation. Also, since the waveguide is a hollow glass tube, at some point the intensity of the driving laser becomes too high to sustain in the waveguide without damaging the walls, ultimately limiting the maximum intensity of the driver beam that can be practically used. The use of a pre-ionized waveguide has the potential to circumvent many of these limitations. High-harmonic generation in laser-generated plasma channels was proposed [5], but not studied experimentally. We plan to investigate the generation and quasi-phase-matched generation of high harmonics of femtosecond Ti:Sa laser pulses ($\lambda = 800$ nm) in a plasma waveguide created using an electrical capillary discharge plasma column with a tailored degree of ionization. In this scheme, an electrical discharge will create a totally ionized medium, where the degree of ionization is selected by choosing the discharge current. A pre-ionized plasma channel, through which the high-harmonic driving laser will propagate. This provides access to significantly larger I_p values than those of neutral atoms, contributing to the generation of shorter wavelengths. The driver laser intensity can be significantly increased before tunnel ionization again becomes an important loss. This will result in an increase in the cutoff energy. Driver laser energy loss due to optically-induced ionization is dramatically reduced. This can result in an increase in the efficiency of HHG. Finally, the radial distribution of the electron density profile can be concave, generating an index guide. This allows for a reduced laser intensity at the walls of the capillary, making it possible to guide pulses at the higher intensities required for shorter wavelength HHG, without damage to the walls.

With this objective we have started the development and characterization of a discharge-ionized gas-filled capillary channel in which both the degree of ionization and the electron density profile can be tailored for propagation of an intense ultrashort laser pulse. The amplitude and shape of the discharge current pulse are selected to produce the desired degree of ionization. Also, the discharge is designed to create an electron density profile with minimum electron density on axis and maximum density near the capillary walls, that will constitute an index waveguide for the light. Model simulations and preliminary experiments show that in small capillaries of 150-250 μm diameter filled with gases at pressures of the order of 5-10 Torr, a modest short current pulse with an amplitude of 10-200 A is sufficient to create a plasma column with mean degree of ionization Z ranging from 1 to 4.

Hydrodynamic simulations

Using a two-temperature one-dimensional Lagrangian code hydrodynamic/atomic code developed at CSU we have predicted the characteristics of the capillary plasmas and the discharge parameters that are required to optimize the degree of ionization and the guiding of intense light for high-harmonic generation. The atomic physics for the ionized gas of interest is

self-consistently simulated by computing the population rate equations using a collisional-radiative model that uses a cell approach to model the radiation transport. We have simulated the plasma columns that can be created in small-diameter argon-filled capillaries. Similar results could be obtained for neon and other noble gases of interest of HHG by properly adjusting the discharge parameters to compensate for the differences in ionization energy and transport coefficients. The results presented below correspond to a 200 μm diameter capillary channel filled with 10 Torr of Ar. These conditions are similar to what has been used in past experiments demonstrating HHG from Argon ions [10]. The gas is assumed to be excited by a 100 A current pulse of 500 ns FWHM duration. The horizontal axis represents the micro- capillary axis and the top of the graph represents the capillary wall.

In this discharge, the ohmic dissipation produced by the current pulse heats the free electrons and creates an azimuthal magnetic field. If the self-generated magnetic field is large enough for the magnetic pressure to overcome the kinetic pressure of the electrons and ions, the plasma column compresses. However, in the relatively low current discharge of interest here, the dynamics of the plasma are dominated by ohmic heating and heat conduction losses.

Shortly after the initiation of the current pulse the plasma is observed to expand as a result of the of the larger temperature increase in the axial region of the plasma zones due to ohmic heating. The outer zones remain colder due to heat conduction to the capillary walls. This generates a radial electron temperature profile with maximum on axis and minimum at the capillary wall, as illustrated in Fig. 5a. The electron temperature on axis is computed to approach 7 eV, while it remains several eV cooler in the periphery. With this temperature profile the plasma pressure of the inner zones exceeds the sum of the pressure of the outer most zones plus the magnetic pressure, resulting in the expansion of the plasma seen in Fig. 5b. Rapid initial radial oscillations in the plasma density are damped by the viscosity of the plasma. After about 300 ns the plasma reaches steady state, and the electron density distribution remain relatively constant until the end of the current pulse, when the plasma cools and recombines.

Figure 6 shows the radial lineout of the electron density 500 ns after the initiation of the current pulse when all plasma parameters have reached steady-state. This concave electron density profile with minimum electrons density on axis gives origin to an index of refraction distribution with maximum on axis, which guides the laser light. Fig. 7 shows the computed radial variation of the mean degree of ionization of the plasma for a 200 A discharge. The mean ion charge, Z , is predicted to increase with time to reach Ar IV ($Z=3$) at the axis about 300 ns after the beginning of the current pulse. Ar IV has an ionization potential of 59.8 eV. Moreover,

Figure 6. Radial lineout of the electron density profile at 500 ns after the initiation of the current pulse. The concave nature of the profile constitutes an index waveguide that can guide the laser light with decreased interaction with the walls.

Figure 7. Computed variation of the mean degree of ionization as a function of time. The capillary diameter is 200 μm , and the gas is Ar at an initial pressure of 10 Torr. The shape of the current pulse is that shown in Fig. 1 and the peak amplitude 200A. At 500 ns after the beginning of the current pulse the dominant ions species are Ar III and Ar IV.

the degree of ionization of the plasma can be tailored by changing the amplitude of the current pulse.

Preliminary results

While the discharge excitation parameters required are quite modest the generation of plasmas in such narrow capillaries with relatively fast current pulses can impose some challenges. We conducted a tests of plasma generation by pulse-discharge excitation into a 5 cm long capillary channel 200 micrometers in diameter, filled with Ar pressures between 5 and 10 Torr. Plasmas were excited by current pulses of about 500 ns duration and amplitudes ranging from 10 A to 100 A. The light emitted by the discharge over the visible and ultraviolet range was analyzed with a 0.6 meter spectrograph to characterize the degree of ionization. Figure 8 shows the relative intensity of lines from Ar I, Ar II and Ar III spectra as a function of capillary discharge current for discharge currents of 50, 70 and 100 A. The data shows it is possible to select the degree of ionization by controlling the discharge current.

Fig.8. Temporal evolution of the intensity of Ar I, Ar II and Ar III lines in a 200 μ m diameter capillary discharge plasma for three different discharge currents. The data shows the degree of ionization can be selected by adjusting the discharge current.

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