

BRNO UNIVERSITY OF TECHNOLOGY  
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**COMPACT, PLASMA-BASED  
SHORT-WAVELENGTH RADIATION SOURCES**

KOMPAKTNÍ PLASMOVÉ ZDROJE  
KRÁTKOVLNNÉHO ZÁŘENÍ

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## **KEY WORDS**

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## **KLÍČOVÁ SLOVA**

Plasma, zdroje krátkovlnného záření, rentgenové lasery, kapilární výboje, z-pinch

## **MÍSTO ULOŽENÍ HABILITAČNÍ PRÁCE**

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# CONTENTS

1 INTRODUCTION .....	5
2 PLASMA – THE FOURTH STATE OF MATTER .....	6
3 EXPERIMENTAL REALISATION OF THE SHORT WAVELENGTH RADIATION SOURCES .....	9
3.1 Conditions for realization of short-wavelength lasers.....	9
3.2 Lasing in the X-ray region.....	10
3.2.1 <i>Laser plasma based sources</i> .....	11
3.2.2 <i>Capillary-discharge based sources</i> .....	13
3.2.3 <i>Hybrid X-ray lasers</i> .....	16
3.3 Incoherent capillary-discharge based radiation sources.....	17
3.3.1 <i>Sources for EUV lithography</i> .....	17
3.3.2 <i>Plasma waveguides</i> .....	18
4 THEORETICAL INVESTIGATION OF THE CAPILLARY-DISCHARGE BASED RADIATION SOURCES .....	20
4.1 Description of the plasma dynamics in capillary z-pinch .....	20
4.2 Radiative and thermodynamics plasma properties.....	21
5 STUDY OF PLASMA – BASED SHORT WAVELENGTH RADIATION SOURCES .....	22
5.1 Investigation of ablative and non-ablative capillary-discharge based radiation sources.....	22
5.2 Construction of a capillary-discharge based laser.....	23
5.3 Application of short-wavelength radiation sources .....	25
SUMMARY .....	26
LITERATURE .....	28
SHRNUTÍ .....	34

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From 2001 until 2004 J. Kaiser was employed as a researcher at the Institute of Physical Engineering, Faculty of Mechanical Engineering, BUT, Czech Republic. From 2005 he is employed as a senior lecturer at the same institute.

His theoretical and experimental work is concentrated on the field of plasma physics. His main research interest is focused on the capillary discharges, plasma generation and study, soft X-ray lasers, theoretical models of discharges and laser induced breakdown spectroscopy-LIBS. He contributed to the foundation of a research group and specialized laboratory for investigation of LIBS technique and capillary-discharge based sources at the Institute of Physical Engineering, BUT. He established cooperation with several foreign research institutes and universities including the University of Pécs, Hungary; Univeristy of L’Aquila, Italy; GREMI Orleans, France; Synchrotron Elettra, Trieste, Italy; ENEA Rome, Italy. a Collaborative research work was conducted in these university laboratories and research institutes.

The research activities of J. Kaiser were supported with several international and national grants. Among those received were a post-doc grant “*Study of plasmas generated by capillary discharges*” presented by the Czech Science Foundation (GAČR 202/02/P113, 2002-2004) and he was involved in a solution of scientific problems addressed in the grant of “*Advanced functionally graded and nanostructured materials*” of the Ministry of Education, Youth and Sport of Czech Republic (CEZ: J22/98:262100002, 1999-2004). The results achieved were published in impacted international scientific journals (19 papers) and presented in the proceedings of 12 international conferences.

His teaching activities involves courses such as Introduction to Physical Engineering, Geometrical Optics, Labs for Physics I and II in Bachelor Study Programmes and courses Geometrical Optics and Engineering Optics in Master Study Programmes.

# 1 INTRODUCTION

Since the invention of the laser in 1960's, intensive research has been carried out in order to extend this device towards the shorter wavelengths, i.e. towards the X-ray region. Because scaling down the wavelength offers great opportunities for both science and technology, this area was extensively studied both experimentally and theoretically. In the habilitation work sources of short wavelength radiation that exploit high-temperature plasmas as the medium are discussed. This is the field, where up to date the most success has occurred.

The main emphasis is on compact, coherent and incoherent capillary-discharge based radiation sources. They are first introduced in a wider context, in comparison with other techniques and then discussed in details. The attached papers document the author's contribution to this field. The work is consisting of four main parts.

In the first part a historical outlook is presented. The term "plasma" is explained and the main directions of plasma research are mentioned. The conditions for employing plasmas for short wavelength radiation sources, together with the main fields of application of these sources are discussed. The conditions of the realization of X-ray lasers, and the different realization schemes are described.

The second part focuses on the experimental realization of the sources. The most important approaches from the past are presented together with the recent experimental results and possible future devices. This part includes also a chapter devoted to construction and diagnostics of the capillary-discharge based sources. The applications of the 46.9 nm capillary-discharge based soft X-ray laser together with the introduction to the research of incoherent capillary-discharge based radiation sources of EUV radiation are also discussed.

The third part is devoted to theoretical studies. Selected topics are presented, mainly regarding the plasma dynamics in pinching devices, together with the models of radiative and plasma properties. The physical processes in ablative capillary discharges are detailed.

The fourth part includes a summary of author's main results on this field. Together with a short description of the earlier outcomes connected to the investigation of ablative and non ablative  $\mu\text{s}$  discharges; the last chapter also contains a selection of eight commented publications [6], [17], [58], [92], [94], [104] – [106]. In these papers the more recent results are published.

## 2 PLASMA – THE FOURTH STATE OF MATTER

The word *plasma* (derived from Greek, meaning “moldable substance” or “jelly”) was introduced at the beginning of the 19th century by the Czech medical scientist Jan Evangelista Purkyně. Purkyně named *plasma* the transparent liquid, which remains when blood is cleared of its various corpuscles.

An American scientist, Irving Langmuir first used this term in 1927 to describe an ionized gas. The way electrified fluid carries electrons and ions reminded him of the way blood plasma carries red and white corpuscles [1]. Langmuir was investigating the physics and chemistry of tungsten-filament light bulbs in order to find a way to extend the lifespan of the filament. In the process he developed the theory of plasma sheaths, the boundary layers that form between ionized plasmas and solid surfaces. He also discovered that certain regions of a plasma discharge tube exhibit periodic variation of electric density, which we now term Langmuir waves. Langmuir’s research [2] forms the theoretical basis of most today’s plasma processing techniques for fabricating integrated circuits. After Langmuir plasma research spread in different directions, of which five are particularly significant.

*Study of the Earth ionosphere* - the earth ionosphere, a layer of partially ionized gases, was discovered by the development of radio broadcasting. This layer reflects radio waves and is responsible for the fact that radio signals can be received when the transmitter is over the horizon. By studying of the propagation of radio waves in ionosphere, the theory of electromagnetic wave propagation through non-uniform, magnetized plasma was developed. This theory was introduced at the beginning of the 20<sup>th</sup> century by scientists such as E.V. Appleton [3] and W.J.G. Beynon [4].

*Astrophysical studies* - astrophysics quickly realized that much of the matter in the universe consists of plasma, and that understanding the astrophysical processes requires deeper understanding of plasma physics. H. Alfvén [5] developed the theory of magnetohydrodynamics (MHD), in which plasma is treated as a conducting fluid. Several processes were described utilizing this theory; for example solar flares, the solar wind, and star formation. This theory was also adapted to describe the plasma dynamics in capillary-discharge based plasma generators [6].

*Study of the thermonuclear fusion* - the construction of the hydrogen bomb in 1952 caused a great interest in controlled thermonuclear fusion as a possible power source for the future. Independent research was carried out in several countries, mainly concerned to understanding how thermonuclear plasma can be trapped (in most cases by magnetic field) and investigating many plasma instabilities which may allow it to escape. However, in 1958 thermonuclear fusion research was declassified; after this 6-years period the theoretical plasma physics emerged as a mathematically rigorous discipline. Recently a fusion experiment managed to extract more fusion energy as was invested in the plasma, but we are still a long way from commercial use of such energy [7].

*Space plasma physics* - in 1958 J. A. Van Allen using data transmitted by U.S. Explorer satellite discovered the Van Allen radiation belts surrounding the Earth. This discovery started the systematic exploration of Earth’s magnetosphere via satellite [8] and opened up the field of space plasma physics. From fusion research space scientists use the theory of plasma trapping by magnetic fields, from ionosphere physics the theory of plasma waves, and astrophysics provided notions of magnetic processes for energy release and particle acceleration.

*Laser Plasma Physics* - the development of high-powered lasers during the 1960’s opened up the field of laser plasma physics. In this technique a high-powered laser beam focused to a solid target ablates the material and forms plasma at the boundary between the beam and the target. Laser plasma has fairly extreme properties (e.g. densities characteristic of solids). Main application of laser plasmas are expected in the field of inertial confinement fusion [9] and in the field of particle acceleration (to use the extremely strong electric fields generated when a high intensity laser pulse passes through plasma to accelerate particles) [10].

The demand to use high-energy photons (i.e. radiation from 0.5 to 50 nm) for scientific and industrial application (as biology, crystallography, micro-lithography, ballistic etc.) appeared a long time ago, already with the discovery of X-ray radiation in 1895 by W. C. Röntgen.

Laser plasmas were already employed as sources of radiation with extremely short wavelengths. In these applications the laser beam is usually focused at a solid, liquid or gaseous target [11], or to the plasma jet [12]. Recently it was shown [13] that plasma with parameters similar to various laser plasmas and suitable for specific application can be created also utilizing plasma generation methods that employ electric fields.

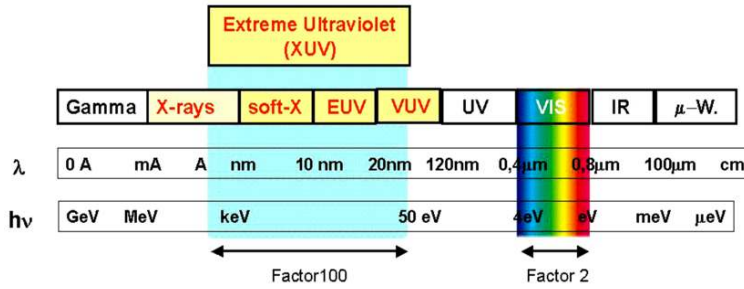


Figure 1. Spectral ranges of short-wavelength radiation sources together with the XUV range in which most of the application occurs [14].

The names and spectral limits of X-rays, soft X-rays and extreme ultraviolet radiation are not yet uniformly accepted. Figure 1 shows one of the nowadays used divisions of this spectral region, together with the XUV range, where most of the applications occur.

The term *laser* is an acronym for a radiation source based on *light amplification* by stimulated emission of radiation. Today, lasers deliver radiation over large portion of electromagnetic

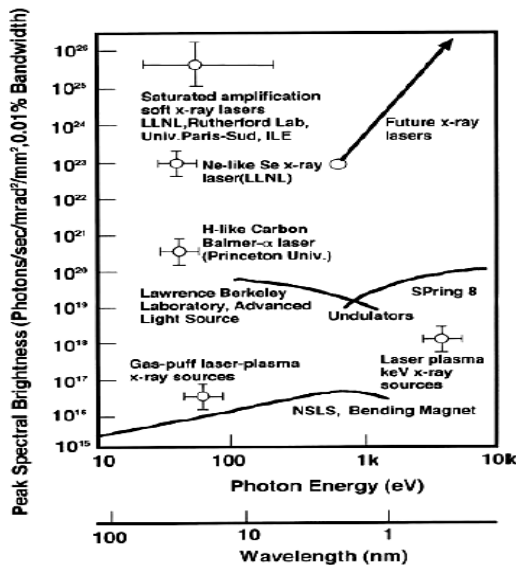


Figure 2. Peak brightness of the present X-ray sources as a function of photon energy in keV (X-ray wavelength in nm) [15].

spectrum, ranging from the far infrared to the X-ray region. X-ray lasers are lasers which deliver quasi-monochromatic, partially coherent photons in spectral range shorter than a few tens of nm [15]. Such kinds of sources have been recently developed worldwide. On Fig. 2) the peak brightness of present X-ray sources is summarized. As it was noted above, the different fields of applications mainly determine the interest of short wavelength radiation sources. There are several specific wavelength ranges of an extensive interest:

- Present-day manufacturing methods are very rough at the molecular level. For advanced future technologies, we will have to develop a new manufacturing technology which will let us build systems with elements that are molecular in both size and precision and are interconnected in complex patterns. Potential applications of the soft X-ray lasers include the technology capable

to fulfill these requirements (i.e. analysis and laser machining of materials on a very small scale) – nanotechnology. For example, computer chips are manufactured using projection lithography, in which each layer of the circuit is imaged onto the substrate material. Further generation of large-scale circuit (with resolution of 70-30 nm) can be produced by soft X-ray projection lithography, using laser plasma or capillary-discharge based source. Moreover, for wavelength-testing of the optics for lithography, it is necessary to have a coherent soft X-ray source. In the spectral range longer than the silicon *L*-edge (12.5 nm) the availability of good X-ray optics foresee the ideal wavelength of the soft X-ray testing laser.

- For biological application, mainly for imaging of biological samples the spectral region called water-window, placed between the carbon and oxygen *K*-edges (4.4 - 2.2 nm) is extremely interesting. Utilizing microscopes operating at X-ray wavelengths it is possible to observe living, *in vivo* biological objects, like cells, bacteria and viruses. A partially coherent illumination in these set-ups permits to improve spatial resolution by a factor of up to two [16]. Furthermore, a source of coherent (laser-like) water window radiation could be used to make a hologram of the sample to give three-dimensional images of living cells. Present soft X-ray microscopes usually use synchrotron radiation. Lasers emitting at this spectral range can represent more reasonable (cheaper and smaller size) alternatives of the radiation sources for soft X-ray microscopy.
- The spectral range shorter than 1 nm (often called X-ray or kilovolt X-ray) is important due the high photon energy. The photon energy is scaling with the wavelength  $\lambda$  as [16]:

$$h\nu(eV) = \frac{1239.842}{\lambda(nm)}, \quad (2.1)$$

where  $h$  is the Planck constant and  $\nu$  is the frequency. The X-rays beyond the photon energy of 3 keV are almost not absorbed by air of atmospheric pressure. They allow the development and application of other advanced techniques, such as non-destructive testing with phase-sensitive projection imaging [17].

- High-energy photons emitted by high-energy, ideally tunable soft X-ray lasers can also be used for the study of the high-lying energy levels of atoms and molecules. Often, these levels of molecules are dissociative and after their excitation the molecule breaks-up into smaller fragments. The study of such processes (called photochemistry) can result in finding applications in the field of excited-state chemistry.



### 3 EXPERIMENTAL REALISATION OF THE SHORT WAVELENGTH RADIATION SOURCES

#### 3.1 CONDITIONS FOR REALIZATION OF SHORT-WAVELENGTH LASERS

In principle, most lasers consist of three parts [18]: a pump source, an active medium and a resonator (see Fig. 3a). Almost any source of energy can, in some ways, be used to pump a laser.

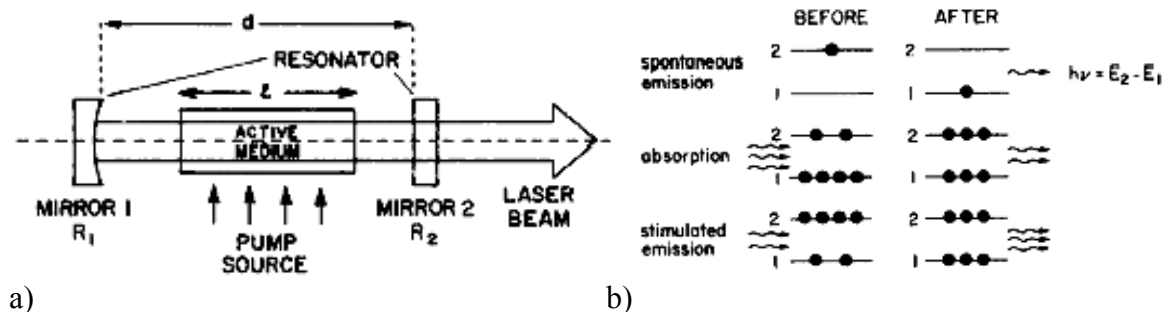


Figure 3. a) Typical parts of a laser. b) Fundamental processes for the interaction of radiation and matter. 1 and 2 can be any pair of levels in an atom, molecule or solid. For laser applications mostly level pairs with allowed radiative transitions between them are important [18].

For short-wavelength lasers gas discharges and plasma sources have a particular importance. The purpose of the pumping process is to establish population inversion in the active medium. Population inversion describes a condition (of the active medium) where the density of states at a higher energy is larger than the density of states at a lower energy. Interaction of light and matter is characterized by three scattering processes (see Fig. 3 b)

- Spontaneous emission – a higher excited state decays spontaneously to a state with lower energy and a photon is emitted.
- Absorption – a photon (light quantum) promotes the active medium from a state of lower energy to a higher energy state.
- Stimulated emission – photons stimulate an excited state to decay to a state of lower energy, emitting an additional photon.

In all these three processes energy and momentum are conserved. In consequence it means that in a stimulated-emission process the stimulated light is emitted in the same direction and with the same wavelength as the stimulated light.

The transition rate (the number of transition per volume and time) is proportional to the density of initial states for all three processes. In a medium with population inversion, more stimulated (and spontaneous) emission events will occur in any given time and interval. Consequently, more photons are generated per time than annihilated. The inverted active medium can amplify

- Spontaneously generated photons - in this case we speak about an *amplified spontaneous emission (ASE)* device, or
- Light inserted in the medium from the outside. This form is called an optical amplifier.

The easier way to increase the amplification of the laser is to use an artificially lengthened active medium – an optical resonator. In its simplest form it consist of two mirrors (from which one is semi-transparent) that enclose the active medium (see Fig. 3a). Spontaneously emitted photons in the direction of the resonator axis are amplified by the active medium and most of the light is reflected back into the active medium by the mirrors, where it is further amplified. When

the losses for the light per round trip are smaller than the gain per round trip, the light intensity inside the resonator will grow. It grows until the density of upper laser states that are produced per time by the pumping process approximately equals the density of photons generated by stimulated emission [18]. The laser output in this case has reached saturation.

Here we should note that saturation can be reached also in mirrorless ASE laser systems. In this case the spontaneous emission from a group of inverted atoms is linearly amplified by the same atoms, with an appreciable gain in at least one direction [19]. The output beam can be extremely bright, collimated and with a fair degree of spatial coherence. However, because the medium acts overall like a mirrorless laser with characteristics somewhere between a truly-coherent cavity-type oscillator and an incoherent thermal source, the output will be without any meaningful degree of temporal coherence [19].

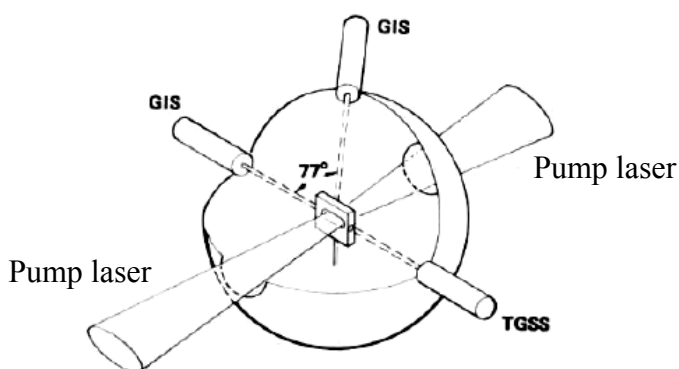
For short-wavelength laser operation, the suitable transition of the media capable of producing UV, VUV and X-ray photons have to be inverted by appropriate pumping mechanism. The requirements these media have to fulfill [18] are

- Transparency and sufficiently small absorption for the laser radiation,
- Capability to produce sufficiently high gain. This means in most cases an allowed optical transition and a relatively narrow line width yielding a high cross section for stimulated emission.
- The active medium should be invertible (at least in transient regime) - it means that it has to have a suitable energy level structure and favorable radiative and nonradiative transition rates.

### 3.2 LASING IN THE X-RAY REGION

The first clearly successful, but rather drastic X-ray lasing experiment was conducted in 1980. The X-ray beams created by underground nuclear explosion at the Nevada Test Site were used to pump the atoms in the system of metal rods. The X-ray laser pulse was emitted just before the whole experimental apparatus was evaporated by the explosion [20].

X-ray lasing in laboratory conditions was demonstrated in the mid eighties employing laser-produced plasmas as an active lasing medium at the LLNL (Lawrence Livermore National Laboratory) using an electron-collisional excitation scheme and at the Princeton University, where a recombination-pumping excitation scheme was used.



*Figure 4.* Experimental setup of the first laboratory soft X-ray lasing experiment. GIS – grazing incidence spectrometer TGSS – transmission grating spectrograph [21].

The first laboratory X-ray laser - Livermore's Novette, the precursor of the Nova laser, was starting to operate in 1984. The plasma was generated from solid targets and heated to the required temperature by the same high-power laser. Typically, the target was composed of a 75 nm layer of selenium vapor deposited on one side of a 150 nm thick formvar substrate [21]. A 532 nm laser light with a nominal pulse length of 450 illuminated it. The line-focused light of the pumping laser

created typical incident intensities of  $5 \times 10^{13} \text{ W cm}^{-2}$ . The targets were irradiated single-sided – in which geometry the target foil was hit by only one laser beam; or double-sided, when opposing laser beams irradiated a common area (Fig. 4). Using this configuration soft X-ray lasing was reached at 20.63 nm and 20.96 nm from the Ne-like selenium plasma.

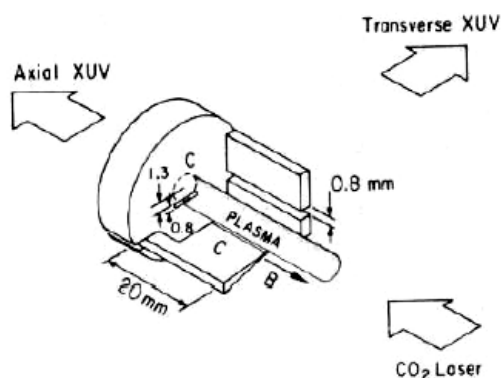


Figure 5. Schematic view of the carbon-disk target and laser illumination and observation geometries in the recombination-pumped soft x-ray laser. The carbon disk (C-carbon) was accomplished with a thin carbon blade (B) in order to create a more uniform plasma in the axial direction and to provide additional cooling by heat transport from the plasma to the blade [22]

In Princeton the laser plasma was created by an irradiation of a solid carbon target by a 75 ns (full width at half maximum – FWHM), 300 J, CO<sub>2</sub> laser (see Fig. 5). The enhancement of 95 of the hydrogen-like carbon 18.2 nm line, which corresponds to the gain-length product of  $\sim 6.5$  was obtained.

X-ray lasers were developed enormously since their first demonstration [23]. However, the shorter wavelength X-ray lasers in Ni-like ytterbium (5 nm), tantalum (4.5 nm), tungsten (4.3 nm) and gold (3.56 nm) have been obtained using the exploding foil techniques pumped by a single green laser pulse with several kJ energy at LLNL [24]; efforts were concentrated on increasing efficiency, extending lasing to shorter wavelength and to develop more compact - table-top sources, suitable for different applications.

Most of the nowadays X-ray lasers are collisionally pumped [15]. Recombination X-ray lasers [15] have been long viewed as an alternative for collisionally pumped systems. In spite the fact that they offered the potential of the higher conversion efficiency; to date it has been difficult to achieve large gain-length products.

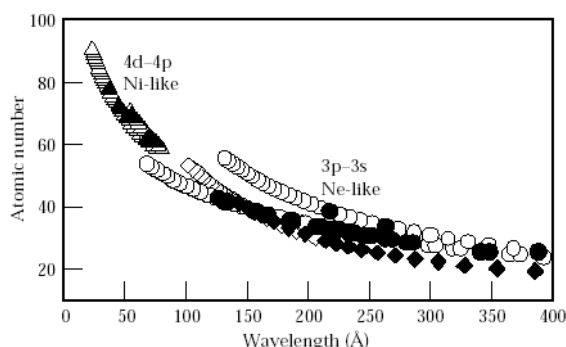


Figure 6. Collisionally pumped soft X-ray laser wavelengths as a function of atomic number. Open symbols represents calculated, solid symbols the measured wavelengths [25].

The dominating wavelengths of electron collisional excitation Ni-like and Ne-like soft X-ray lasers are shown in Fig. 6. The complete list (up to date 2002) of the experimentally demonstrated, electron collision excitation Ni-like and Ne-like soft X-ray lasers can be found in [15].

### 3.2.1 Laser plasma based sources

The successful demonstration of X-ray laser action by the large fusion systems initiated an intensive research in order to reduce the pumping energy requirements of these lasers. Until the

late 1980s the use of high-energy ns optical laser systems for pumping, considerably limited the practical applications.

After the demonstration of soft X-ray lasing at 19.5 nm and 28.5 nm in neon-like germanium ( $\text{Ge}^{22+}$ ) and copper ( $\text{Cu}^{19+}$ ) in 1987 by T.N. Lee *et al.* [26] using relatively low irradiance glass laser (600 J in 2 ns), the germanium laser coupled with solid slab technique (see Fig. 7) became popular worldwide.

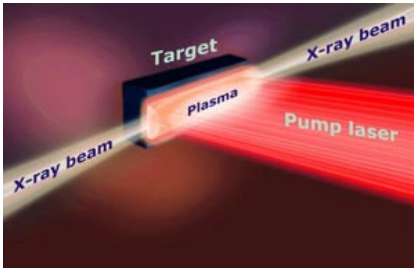


Figure 7. Sketch of a X-ray laser in a slab target geometry [27].

The plasma generated from the flat slab target has an electron density gradient that causes X-ray refractions. This behavior limits the achievable gain length and in most cases does not allow reaching real saturated amplification. To avoid this behavior, in 1986 J.G. Lunney proposed to use a curved slab target [28]. In the curved-slab target configuration (see Fig. 8) X-ray laser beam propagates along the gain region over a longer distance, when the target is bent at a curvature which is matched to the ray trajectory. Using this configuration an intensity enhancement by a

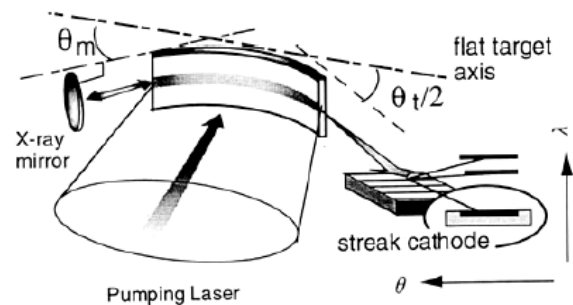


Figure 8. Schematic diagram of the curved-slab target.  $t$  and  $m$  represents the bend angel of the target and setting angle of the X-ray mirror, respectively [15].

factor 10 respect to the flat slab target was reported by R. Kodama *et al.* in 1994 [29]. Further improvements on this scheme were achieved by double pass amplification, using an X-ray mirror or polarizer.

The recent advances in laser-plasma based table-top soft X-ray lasers have been made possible mainly by the development of new, compact, high-power driving lasers. The ultra-short, excimer based systems [30], which have been available since the 1980s produce pulses of 0.3-0.5 ps

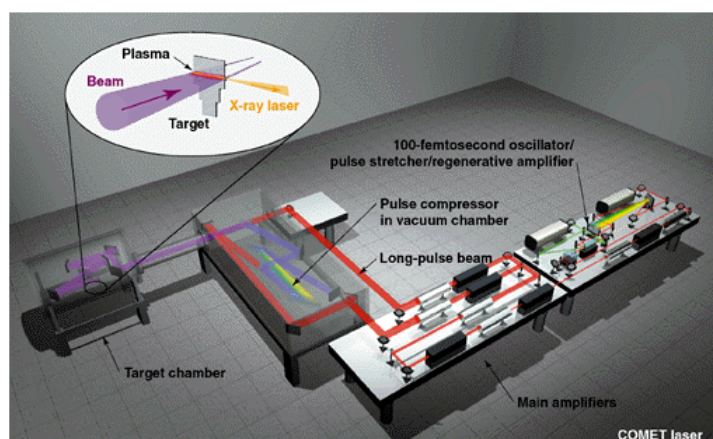


Figure 9. Experimental arrangement of the Livermore's COMET (compact multipulse terawatt) tabletop X-ray laser. The rendering shows the laser system and the target chamber [31].

duration. However, their output energy is limited by the low saturation fluency ( $\sim \text{mJ}/\text{cm}^2$ ). The majority of present compact laser systems use CPA (chirped-pulse amplified) solid-state amplifiers [32]. The CPA technique allows taking full advantage of the high-saturation fluences ( $\sim 1 \text{ J}/\text{cm}^2$ ), broad gain bandwidth and relatively long upper laser lifetime of solid state amplifier media such as Ti:sapphire, Nd-glass and Cr:LiSAF. In the CPA technique a short (typically fs) seed pulse is first stretched in duration, then amplified and finally recompressed to ideally its initial duration (see Fig. 9).

### 3.2.2 Capillary-discharge based sources

Since the first publication on wall stabilized electric arc by H. Maecker in 1956 [33], capillary discharges formerly called also as “creeping”, “guided”, “sliding”, “gliding” sparks [34] or “cascade arcs” have been intensively investigated. The cylindrical symmetry of these sources allowed expressing the energy balance equations in a comparatively simple form. This advantage has been used in the study of different physical mechanism involved in this type of plasma (see for example [35] and references therein).

The expected military application for electrothermal launchers (rail guns) [36] resulted in an extensive theoretical and experimental study of these discharges from the late 1960s.

Prior to their use in soft X-ray laser research, discharges in evacuated capillaries have been also applied in the field of spectroscopy, microscopy and lithography. The search for alternatives of laser drivers in X-ray laser schemes at the late 1980s initiated the development of soft X-ray lasers based on capillary discharges.

In 1987 F.C. Young *et al.* from Naval Research Laboratory, Washington, USA proposed to use sodium-bearing capillary-discharge plasma for X-ray laser experiments [37]. The NaF plasma was

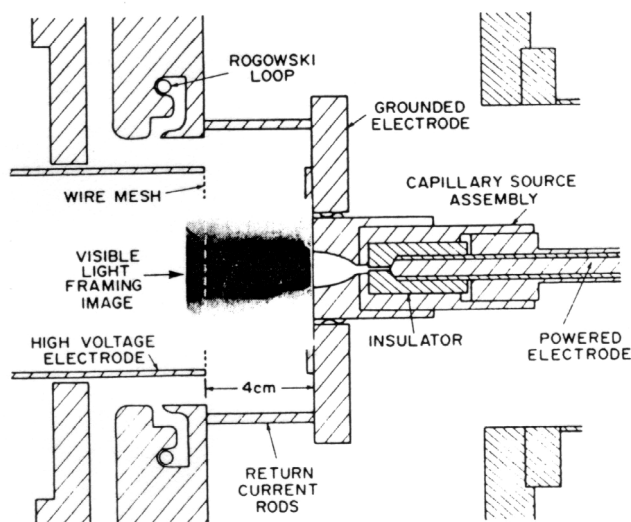


Figure 10. Schematic of the experimental setup for investigation of the Na/Ne X-ray laser [37].

created by a 60 kA, 3.4  $\mu\text{s}$  prepulse inside the capillary and the emerging plasma jet was subsequently excited with a high-current (1.2 MA) main pulse in order to produce an intense source of sodium *K*-shell X-rays (see Fig. 10). A peak power of 25 GW in a 20 ns pulse was measured for the He-like sodium ( $\text{Na}^{+9}$ )  $1s^2 - 1s2p^1P$  transition at 1.1 nm (He- $\alpha$  line). It was expected to use this line as a pump radiation for a sodium/neon line-coincidence XUV laser [38], [39]. Sodium and neon were selected because there is a natural line coincidence between the  $n=2$  to  $n=1$  transition in He-like sodium and the  $n=4$  to  $n=1$  transition in He-like Ne ( $\text{Ne}^{+8}$ ). The 1.1 nm radiation from He-like sodium can resonantly populate the  $n=4$  to  $n=1$  transition in He-like Ne with potential for lasing on the 4-3, 4-2, and 3-2 transitions at wavelengths of 23 nm, 5.8 nm, and

8.2 nm, respectively. However this scheme was intensively studied, no evidence on realization of utilizable Na/Ne X-ray laser was given.

The possibility to use capillary discharges directly for the excitation of recombination laser was suggested by J.J. Rocca *et al.* from Colorado State University, Fort Collins, Colorado, USA [40] in a paper published in 1988 and subsequently realized by a group of H.-J. Kunze from Ruhr University, Bochum, Germany in 1990 [41]. They considered a pulsed discharge, where the carbon plasma was produced from wall material and the recombination pumping was obtained by rapid conductive cooling by the capillary walls. In [41] an amplified spontaneous emission of the hydrogen-like carbon ( $C^{5+}$  - Balmer- $\alpha$  line) at 18.2 nm is described. The plasma was produced by ablating a wall of a small ( $< 1$  mm in diameter,  $< 50$  mm length) polyacetal -  $(CH_2O)_n$  capillary. The maximum gain observed was  $\sim 3.1$  cm $^{-1}$ .

In the beginning of 1990s the investigation on the possibility of using gas-filled capillaries started. In 1991 W. Hartmann *et al.* reported on a successful operation of an ultrafast, small-diameter, highly uniform  $z$ -pinch at currents of 35-50 kA, for a variety of gases and a large range of mass densities [42]. In the subsequent experiments in 1996 T. Wagner *et al.* showed a gain in a recombining  $z$ -pinch plasma at 52 and 49.8 nm [43]. The amplified XUV radiation originated from the  $4f-3d$  and the  $4d-3p$  transitions of Li-like oxygen ( $O^{5+}$ ). The plasmas were generated in a small diameter  $z$ -pinch discharge with currents of 40 kA.

Applying short-rise time (typically 10-40 ns) current pulses showed as key conditions for successful realization of collisional excitation pumping of gas filled (plastic) capillaries. The short current pulse heats the plasma fast and efficiently allowing the magnetic field to compress the

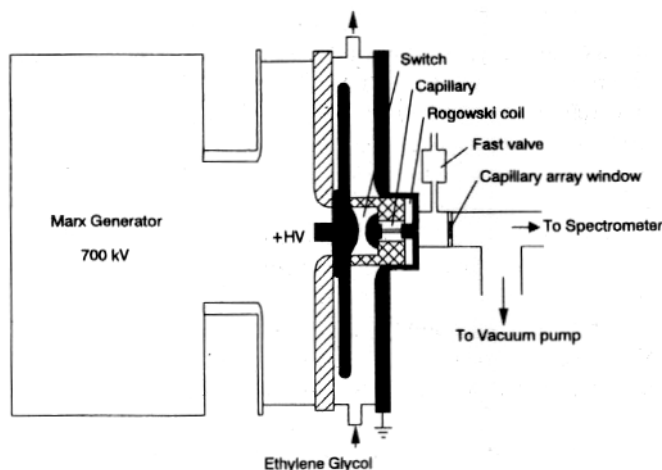


Figure 11. Schematic illustration of the fast capillary discharge setup [44].

plasma column, avoiding its cooling by interaction with the capillary wall. The collisional excitation capillary discharge soft X-ray laser was realized in the years 1992-1996 by J.J. Rocca *et al.* applying a compact laser driver. In this set-up the fast excitation current pulse is produced by discharging a water capacitor through a spark gap switch connected in series with the capillary load (see Fig. 11). In 1993 they reported on a strong amplification at  $J=0-1$ ,  $3p-3s$  transition (46.9 nm) on Ne-like Ar ( $Ar^{8+}$ ) [44]. The lasing was demonstrated in 1994 [45], and the saturation limit was reached in 1996 [46].

In the year 1996, amplified spontaneous emission at the  $J=2-1$ ,  $3p-3s$  transition (69.78 nm) of Ne-like Ar was reported by A. Hilderbrand *et al.* [47]. Gain coefficients up to  $0.25$  cm $^{-1}$  were reached, in the plasma column generated in up to 6 mm in diameter and up to 14 cm long capillary channels. The discharge was driven by a small capacitor bank having a capacitance of  $0.1$   $\mu$ F.

In 1997 lasing in Ne-like sulfur at 60.8 nm was demonstrated by the group of J.J. Rocca [48]. Ablating the wall of a 5 mm in diameter 2 cm long secondary capillary channel, with a slow current pulse delivering 200 J in about 50  $\mu$ s, produced the sulfur-vapor. The generated vapor was

injected into the main capillary channel through a hole in the ground electrode and was subsequently excited by a fast current pulse of 35-37 kA peak amplitude having a first half cycle duration of  $\approx 72$  ns [49].

The utilization of ceramic capillaries in 1998 was an important next step to develop a collisional excitation pumped high-average power tabletop soft-X ray lasers. The “ablation-free” sources have numerous advantages. To the contrary of the set-ups with ablative capillaries, they are instability-free, pollution-free, allow a high-reproducibility of the plasma emission and have a longer lifetime. By exciting a plasma column in  $\text{Al}_2\text{O}_3$  capillary, laser with average output power  $\approx 1$  mW ( $> 2 \times 10^{14}$  photons/s) and energy 135  $\mu\text{J}/\text{pulse}$  was reported by the group of J.J. Rocca [50]. The spatially coherent average power emitted at 46.9 nm (26.5 eV) by this compact, high-repetition rate (7 Hz) laser is comparable to that generated at this photon energy in a similar bandwidth ( $\Delta\lambda/\lambda=10^{-4}$ ) by a third generation synchrotron beam line.

Laser amplification at 52.9 nm in Ne-like Cl with a table-top capillary discharge source was reported in 2000 [51]. Laser output pulses with energies of as much as 10 m J have been obtained. The beam divergence was  $\approx 4$  mrad.

From 1999 the team of J.J. Rocca is building an apparatus (200 kA/10 ns) for amplification at shorter wavelengths using Ni-like ions. They are mainly interested in lasing line for Ni-like cadmium at 13.17 nm and Ni-like silver at 13.9 nm. The appropriate Ni-like spectra were characterized for cadmium [52] and silver [53] in 2003 and 2004, respectively. However, for example the 13.2 nm Ni-like cadmium-line ( $\text{Cd}^{20+}$ ) is clearly visible in the EUV spectrum; convincing evidence of lasing on these elements was not yet presented.

After the successful demonstration of soft X-ray laser action by J.J. Rocca, several groups worldwide reported on lasing obtained by similar type of sources.

In 2001, A. Ben-Kish *et al.* from Technion-Israel Institute of Technology, Haifa, Israel reported on a strong amplification of the  $3s-3p$  ( $J = 0-1$ ) transition in Ne-like Ar ions at 46.9 nm [54]. A gain coefficient of  $>0.75 \text{ cm}^{-1}$  and a beam divergence of  $<5$  mrad, measured along plasma columns of  $<150 \mu\text{m}$  diameter and up to 165 mm length was observed. Since this paper this group published no experimental work.

G. Niimi *et al.* from the group of E. Hotta (Tokyo Institute of Technology, Yokohama, Japan) reported on observation of a multi-pulse soft X-ray lasing in 2001 [55]. They also investigated in detail the role of predischARGE (preionization) current, in the argon capillary soft X-ray laser operation [56]. In 2003 they declared a lasing with an energy  $\sim 6 \mu\text{J}$ , gain of  $0.8 \text{ cm}^{-1}$  and  $Gl=12$  [57].

In 2002 a group, which the author of this habilitation work is a member of, succeeded in a construction of Rocca's type Ne-like soft X-ray laser (see Fig. 12) at the University of L'Aquila, L'Aquila, Italy [58]. The laser has been build in a frame of wide (Italian-Hungarian-Czech-Russian) international cooperation. The lasing was reached by a modification of a set-up originally proposed by S.V. Kukhlevsky *et al.* in 1997 for double-discharge scheme [59]. The first lasing action reported by this group at transition  $3p-3s$  ( $J = 0-1$ ) of Ne-like Ar was obtained by exciting the active medium with a current pulse of half-cycle duration 140 ns. The current was produced by discharging a 10 nF water dielectric capacitor, initially charged to voltages  $< 200$  kV by a six stage Marx generator, through 15 cm long 3 mm in diameter  $\text{Al}_2\text{O}_3$  capillary channels filled with Ar of 0.3 Torr pressure. The laser beam had 4 mrad divergence and time duration of 1.3 ns. Gain of  $0.6 \pm 0.1 \text{ cm}^{-1}$  was reached. Presently the laser operates in highly-saturated regime with output energy of  $\sim 300 \mu\text{J}$  in up to 55 cm long active medium. In this medium the laser intensity distribution reaches the divergence of 0.6 mrad, which approaches the limit of diffraction [60].

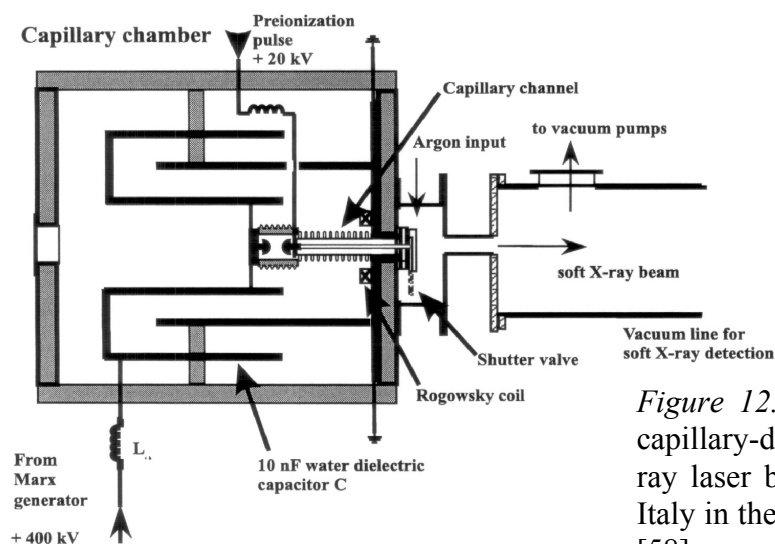


Figure 12. Experimental arrangement of the capillary-discharge based Ne-like Ar soft X-ray laser built at the University of L'Aquila, Italy in the frame of international co-operation [58].

The team of M. Vrbová from the Czech Technical University in Prague, Prague, Czech Republic is involved in investigation of physical processes in gas-filled [61], [62] and initially evacuated ablative capillaries [63]. They are investigating these sources in order to improve both capillary-discharge soft X-ray lasers and non-coherent EUV sources. For X-ray lasing at 13.4 nm, novel recombination schemes are considered using hydrogen-like nitrogen ions ( $N^{6+}$ ) [62]. In theoretical studies this team cooperates with a group of N.A. Bobrova from Institute of Theoretical and Experimental Physics, Moscow, Russia.

Promising results were reported in 2002 by the group of K. Kolacek from Institute of Plasma Physics, Academy of Sciences of the Czech Republic, Prague, Czech Republic, with theoretical support of the team of M. Vrbová. The laser line dominating in the soft X-ray spectrum has been observed [64]. The driver of the soft X-ray laser built by this group has similar parameters as the Rocca's first device [44].

The possibility to use ablative capillary discharges for production of coherent X-ray radiation was investigated also by the group of C. Fleurier at GREMI, Orleans, France. However they reported on observation of amplified spontaneous emission of  $H_{\alpha}$  and  $H_{\beta}$  lines of  $C^{5+}$  [65], presently the research is focused on non-coherent EUV sources [66].

Regarding the recent results in construction of soft X-ray lasers using evacuated ablative capillaries, it is noteworthy to mention a paper of S.S. Ellwi *et al.* [67] from a group of H.-J. Kunze. In the paper, published in 2001, the final evidence of lasing using charge exchange scheme is given. The charge exchange process occurs between ions produced in the neck of an induced MHD instability and low temperature ions present in the same plasma column. Using a straight capillary with a waved structure imprinted inside created the MHD instability. Time resolved diagnostics of plasmas produced by this source were done at GREMI Orleans and published in 2002 by S. Götze *et al.* [68].

### 3.2.3 Hybrid X-ray lasers

Further development of table-top soft X-ray lasers is expected by combining the two techniques, i.e. capillary discharge and laser-plasma produced sources. In this arrangement gas-filled or ablative capillaries are creating a medium, which can be pumped longitudinally by external laser pulse (see Fig. 13). The pump radiation can be guided inside the pre-formed plasma column and excite the active media with a length of a few tens of centimeters. Such a soft X-ray laser was demonstrated by K.A. Janulewicz and J.J. Rocca *et al.* in 2001 [69]. The capillary discharge was used to generate a sulfur plasma column with a large concentration of Ne-like ions and a radially concave electron density profile with minimum at the capillary axis. The subsequent



intense short laser pulse rapidly heated the electrons, producing amplification in the  $3p^1S_0-3s^1P_1$  transition of Ne-like sulfur at 60.8 nm. The gain-length product obtained exciting a 3 cm long capillary with a 0.46 J short laser pulse was  $\approx 7$ . Further work on this scheme is in progress [23].

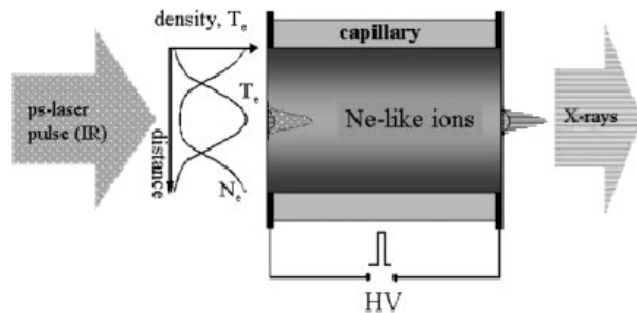


Figure 13. Experimental arrangement of the hybrid X-ray laser. The concave electron density distribution with minimum on the capillary axis, which is necessary for guiding of the pumping laser pulse, is shown on the left [23].

### 3.3 INCOHERENT CAPILLARY-DISCHARGE BASED RADIATION SOURCES

Among other, the incoherent capillary-discharge based radiation sources can be utilized as sources for EUV lithography and as plasma waveguides. These two particularly important applications will be detailed in the next two chapters.

#### 3.3.1 Sources for EUV lithography

In microlithography, the mercury-vapor discharge lamp (first at 463 nm  $-g$ -line and then at 356 nm  $-i$  line) has provided the illuminating flux for microlithography machines for over 20 years. The mercury  $i$ -line has been used to produce features below 0.35  $\mu\text{m}$ . Scaling down was possible by using shorter wavelengths, when the excimer lasers became available with sufficient intensity and flux in the 1980s. In particular the 248 nm KrF laser and the 193 nm ArF laser have been developed and used for imaging the mask patterns onto silicon wafers [72]. Further development of this technique, i.e. the possibility to create even finer structures requires moving towards shorter wavelengths. Moreover, sources for microlithography have to fulfill a number of system requirements, as high collectable in-band power, low debris production, high-repetition rate and pulse-to pulse repeatability [73]. Currently considered sources being able to become a EUV source for lithography are the xenon laser-produced plasmas for Mo:Be spectral window (that uses a cluster of xenon atoms generated by a supersonic gas nozzle), synchrotron/wiggler,  $z$ -pinch, capillary discharges and plasma focus. The detailed characterization of these sources goes beyond the range of this work. Comparison of EUV sources for lithography can be found in [72] and [73]. Detailed description on microlithography technique was published for example in [74].

In this work the discussion is limited to the capillary-discharge based EUV sources of some groups that most of them are involved also in X-ray laser research. Because The target of this chapter is to point out some directions of the research on this field instead of giving a detailed description of “state-of the art”.

The group of C. Fleurier has constructed high-repetition rate EUV sources [75]. The plasma column is created inside the gas-filled capillary channel. For achieving effective pumping, the inductance of the circuit was minimized by making a low-inductance connecting path (using metallic rings) between the knob-capacitors bank to the capillary (see Fig. 14). The electrical energy is stored in two capacitor banks connected in Blumlein configuration [66]. The capillary channel made up of  $\text{Al}_2\text{O}_3$  is positioned on the axis of annular capacitor bank. The two hollow electrodes made of tungsten allow probing of the radiation in both end positions. Flowing gas system is used with differential pumping across the hole in the ground electrode. The best result obtained on energy radiated in the band of  $(13.5 \pm 0.3)$  nm was 5 mJ/shot for 2J input energy [75].

Very good stability of the device was obtained by testing it in continuous mode at repetition rates of 100-150 Hz for time periods of several hours [75].

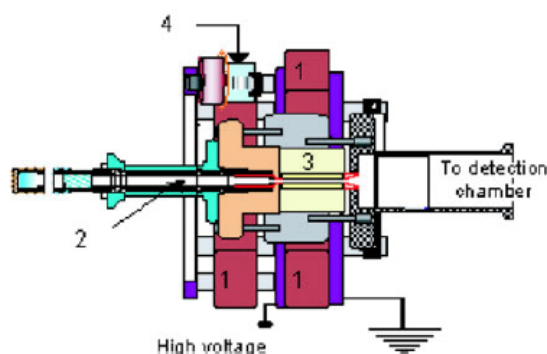


Figure 14. Schematic of the fast capillary discharge setup: 1-Knob capacitors, 2-Gas inlet, 3-Capillary, 4-Thyratron. [75].

Xe-filled fast capillary discharges are investigated also in the laboratory of M. Vrbová [61]. Her group is also involved in the study of the dynamics and emission characteristics of the source of C. Fleurier *et al.* described in the previous paragraph [76]. One-dimensional magneto-hydrodynamics (MHD) code was used to evaluate time and radial dependence of plasma properties. Plasma thermodynamics and radiative characteristics were evaluated by steady-state IONMIX code.

EUV source emitting at the wavelength range 11-13.5 nm, which utilizes xenon-filled capillary z-pinch plasma, is studied at the laboratories of E. Hotta. They reported recently on a source working with repetition rates 7-10 kHz. The device is equipped with a water-cooled ceramic capillary and electrodes and a solid state pulsed power generator. The high-repetition rate is achieved by using a stacked static induction thyristors as switching elements. A magnetic switch connected in series assists the semiconductor switch and provides a pre-ionization current [77].

At the University of L'Aquila (group of R. Reale and G. Tomassetti) a new EUV lithography source is built on the frame of Italian national FIRB (Prototype for electronic chip and opto-electronic device nanofabrication) project. The project is scheduled until 2006. The table-top (50 cm x 100 cm) EUV-L source is already realized and presently is tested.

### 3.3.2 Plasma waveguides

Ultra short laser pulses with high intensities ( $10^{18} - 10^{22}$  W/cm<sup>2</sup>) are used in a number of different applications which includes hybrid X-ray laser systems, electro-thermal launchers, laser-driven particle accelerators and nuclear fusion [78]. In general, the laser propagation distance is limited by diffraction and can be further limited by ionization-induced refraction. For this reason, optical guiding-transportation of the laser pulse over many Rayleigh lengths is needed. Appropriate conditions for the laser-pulse guiding can be reached also in plasma-filled channels. In this case, the preformed plasma column with a parabolic or step-like radial electron density profile and a refractive index peaked on axis is obtained by minimizing the local ambient electron density on the axis [79]. In addition, the laser-pulse guiding can be realized in a plasma waveguide formed in ablative capillaries near the axis [78] or in a non-ablative gas-filled capillary discharges [80].

Here two recent interesting results on capillary-discharge based plasma waveguides are introduced.

In 2002 A. Butler *et al.* reported on guiding of high-intensity laser pulses in non-pinchng fully ionized hydrogen plasma waveguide [81]. They succeed by a theoretical support of N.A. Bobrova and P. Satorov on guiding of a laser pulses of Ti:Al<sub>2</sub>O<sub>3</sub> laser with peak intensities  $> 10^{17}$  W cm<sup>-2</sup> in a 30 mm and 50 mm long, 0.4 mm internal diameter Al<sub>2</sub>O<sub>3</sub> capillaries (see Fig. 15). The storage capacitance used was 7.5 nF and it was charged to 17-30 kV. It is noteworthy that the (non-pinchng) capillary discharge was initiated with a very low amplitude (500 A) current pulse with a

half-period of  $\sim 380$  ns. The initial gas pressure varied between 82 and 248 Torr. The transmission reached for ideal conditions [81] was 90% and 80% for the 30 and 50 mm long capillaries, respectively.

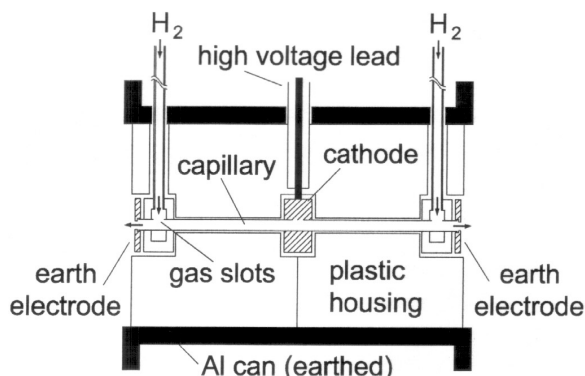


Figure 15. Schematic diagram of the capillary-discharge based plasma waveguide [81].

The group of J. J. Rocca reported in 2004, in a paper of B.M. Luther *et al.* [82] on utilization of a fast capillary discharge for guiding of intense laser beams. In opposite to the technique described above, in this case z-pinch discharge plasma-waveguide was created in  $\text{Al}_2\text{O}_3$  capillary of 3.2 nm in diameter, 5.5 to 11 cm long, filled with Ar at a pressure of 0.18 to 0.25 Torr. The discharge current pulses had a peak amplitude of 15 to 21 kA and a half period of  $\sim 120$  ns. Laser pulses with peak intensities up to  $2.2 \times 10^{17}$  W/cm<sup>2</sup> were guided. In a 5 cm long plasma column the transmission efficiency reached 75%. The maximum efficiency was observed just before the plasma reaches the conditions for lasing in Ne-like Ar. Figure 16 shows the parabolic electron density profiles suitable for guiding together with the near-field and far-field images of the exit beam shortly before the time of maximum compression.

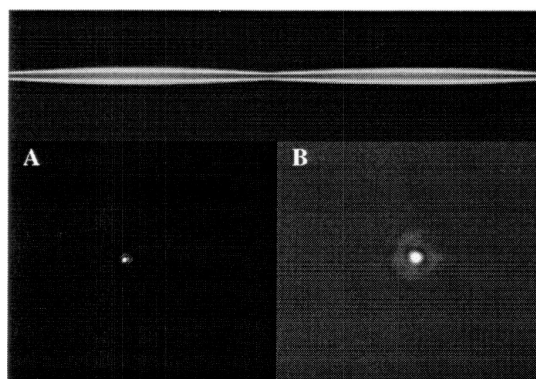
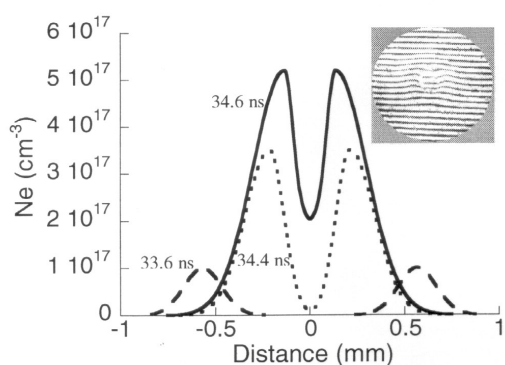


Figure 16. Electron density profiles obtained from interferograms corresponding to the early stage of guide formation in a 1.4 cm long capillary (left) together with near field (A) and far-field (B) images of the exit beam shortly before the time of maximum compression (right). On the top the corresponding beam propagation simulation is shown [82].

## 4 THEORETICAL INVESTIGATION OF THE CAPILLARY-DISCHARGE BASED RADIATION SOURCES

### 4.1 DESCRIPTION OF THE PLASMA DYNAMICS IN CAPILLARY Z-PINCH

In the  $z$ -pinch capillary discharges the plasma column inside the capillary is compressed by the magnetic field  $\vec{B}$  created by the current, which passes through the capillary channel. If the current density  $\vec{j}$  is sufficiently high, the magnetic field can drive the plasma sheet from the walls towards the axis (see Fig. 17). Plasma density and temperature are increased by the compression,

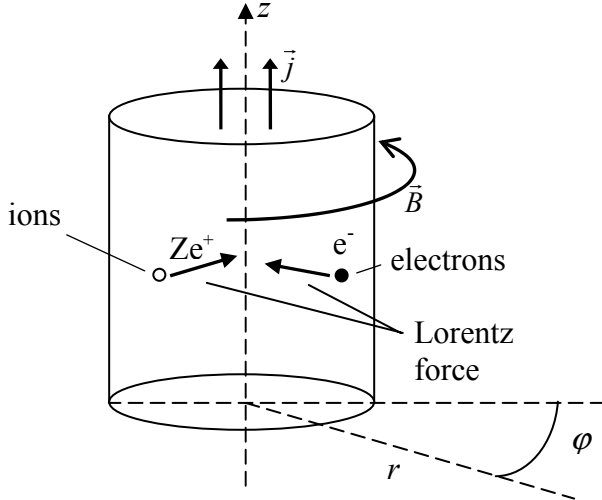


Figure 17. Schematic diagram of the plasma column inside the capillary channel compressed by Lorentz force [70].

which starts when the Lorentz force  $\vec{j} \times \vec{B}$  is higher than the kinetic pressure in the capillary. The pinch of the column formed this way continues until the kinetic pressure is sufficiently high to balance the magnetic field pressure [70].

The simplest way to describe the  $z$ -pinch formation is by means of the “snow-plow” model. The description of this model is presented for example in the book of A.V. Trivelpiece and N.A. Krall [83]. It is based on the presumption that initially a thin cylindrical layer, separated from the capillary wall is formed and all the electric current through the capillary passes through this sheet only. The radial movement of the created sheet is driven by the radial component of Lorentz force.

In the work of P. Vrba and M. Vrbová [84] the “snow-plow” model is used for the optimization of electrical pumping of capillary  $z$ -pinch X-ray lasers and to study the role of short-time current modulation caused by a multi-resonant electrical circuit on the  $z$ -pinch dynamics. Similar “simple” model [85] was utilized by our team for study of plasma evolution in argon-filled capillary  $z$ -pinch discharge. The results of this investigation were published in 2001 [86].

Another approach to the simulations of capillary discharge sources is using computer codes based on magneto-hydrodynamics (MHD) equations. This method was applied by H.-J. Kunze *et al.* in 1994 to analyze the possibility that charge exchange between plasma ions preceded by an  $m = 0$  instability is responsible for amplified spontaneous emission observed on the  $C^{5+}$  Balmer- $\alpha$  line emitted from a capillary discharge [87].

The capillary discharge MHD model is used also by the group of J.J. Rocca in order to model their source (see for example [88]).

MHD code was applied by the group of M. Vrbová to evaluate time and radial dependence of plasma properties for incoherent xenon capillary discharge source [76].

Extensive theoretical studies of capillary discharges using MHD simulations have been done by N.A. Bobrova *et al.* They considered capillary  $z$ -pinches in both, ablative and non-ablative capillaries. In 1998 they reported on results of their simulations [89] chosen to fit the published experimental conditions of Rocca’s soft X-ray laser [45]. The simulation showed a good

agreement with experimental results, for example the observed time of plasma compression has coincided with the simulated one. Moreover, in the work [90] published in 1996 they discussed the possibility of use of the capillary plasma to provide a guiding for ultrashort, high-intensity laser pulses. In 2001 they extended the model [91] for simulation of a non-pinching hydrogen-filled capillary discharge waveguide [81]. The MHD simulations were compared with the measurements of the electron density profile and good agreement was found. They showed that the mechanism of formation of the guiding electron density profile in this non-pinching device is very different from that which occurs in  $z$ -pinch capillary discharge waveguides.

The observations of active-plasma waveguiding of the laser built in L'Aquila, Italy [92] and the successful waveguiding experiment reported by B.M. Luther et al. [82] encouraged our group to extend the MHD model used to find appropriate conditions for soft X-ray laser and simulate the waveguiding conditions in argon filled capillary discharges [94]. The exact mechanism of the  $z$ -pinch compression of the plasma, which is considered in the simulation, was described in details by S.V. Kухlevsky in 2001 [93]. It was shown that the transverse electric field of the sliding surface discharge provides the instability-free compression and heating of the plasma.

## 4.2 RADIATIVE AND THERMODYNAMICS PLASMA PROPERTIES

Study of the radiative and thermodynamics properties in high-energy plasmas is important due several aspects. Investigating the spectral line balance one can obtain fundamental information on the plasma parameters, as ionization balance, rate processes, densities, temperatures, fluctuation levels etc.

The determination and modeling of thermodynamic and radiative properties of capillary-discharge plasmas is crucial for finding new lasing schemes and to optimize the already working sources. Several computer codes are available for describing the absorption, emission and transport of radiation for hot plasmas. Let us consider some programs used to describe plasma properties in capillary discharges.

The IONMIX code [95], developed by J.J. MacFarlane [96] can be applied for computing the equation of state and multigroup opacities for both LTE and non-LTE plasmas. The steady-state ionization and excitation populations are calculated using detailed balance arguments and rate coefficients based on the hydrogenic ion approximation. The code considers contributions from bound-bound, bound-free, free-free and electron scattering processes in evaluating extinction and emission coefficients at several hundred well-placed photon energies, which are then used to compute multi-group Planck and Rosseland mean opacities. More details on this code and the code can be found in [95].

The IONMIX code was applied by the group of M. Vrbová to evaluate the parameters for several types of capillary-discharge plasmas. As an example, the investigation of the thermodynamics and radiation properties of hot Xenon plasmas was presented in the work [61]. The code was also used for evaluation of ionization fractions and thermodynamic properties of a pinching discharge in nitrogen-filled capillary soft X-ray laser recombination pumping [62].

More recently, this group is accomplishing the simulations by applying the FLY code [97], for calculating the temporally resolved line X-ray spectra. The FLY code works as a post-processor of results obtained by MHD simulations [62].

Several computer codes are applied by the group of J.J. Rocca. For example in the work [88] the MHD code is coupled to the RADEX code [98] in order to find the gain coefficient and intensity for a new lasing scheme i.e. for  $\text{Ti}^{12+}$   $J = 0 - 1$  transition (at 32.6 nm) for different initial conditions. In the more recent work [97] the experimental Kr-like  $\text{Y}^{3+}$  intensity ratios are compared with collisional prediction of the HULLAC (Hebrew University - Lawrence Livermore Atomic Code) atomic physics package and the response of the system to a fast, transient excitation pulse is examined using the RADEX code.

## 5 STUDY OF PLASMA – BASED SHORT WAVELENGTH RADIATION SOURCES

### 5.1 INVESTIGATION OF ABLATIVE AND NON-ABLATIVE CAPILLARY-DISCHARGE BASED RADIATION SOURCES

The author of this work has been involved in the investigation of a short-wavelength radiation sources since the middle of 1990s. In 1998 he joined a research group of S.V. Kukhlevsky and A. Reale, which was working on a development of a capillary-discharge based soft X-ray laser. Originally the project proposed to use double-discharge (double-pulse) scheme [59] for both, enhance the gain of  $\text{Ar}^{+8}$  laser and to test the possibility to decrease the emission wavelength applying as an active media metal-vapor instead of argon [71].



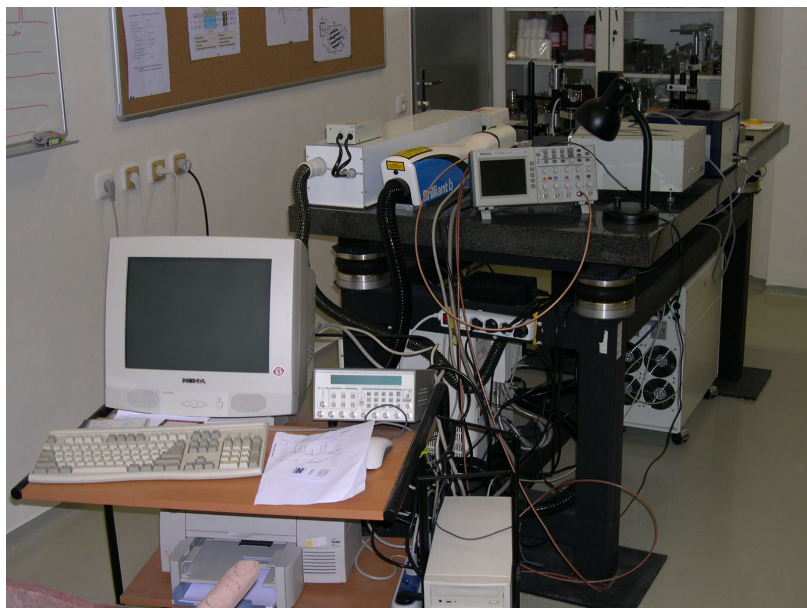
*Figure 18.* Overall view to the laboratory for investigation of capillary-discharge based radiation sources at the University of Pécs, Hungary.

In his earlier work the author was focusing mainly on a study of  $\mu\text{s}$ -scale capillary discharges. The experimental part of this research was conducted at the cooperating institute, University of Pécs, Hungary. With the contribution of the author an apparatus was built for investigation of both, ablative and non-ablative capillary-discharge based radiation sources [70], [101].

Discharges in ablative capillaries were investigated mainly in order to understand the physical processes involved in the discharge better. Among other, the role of the electrode ablation, which was not taken into consideration in the conventional models, was studied. The investigation of ablative capillaries was motivated also by the fact, that J.J. Rocca's first  $\text{Ar}^{+8}$  laser was realized utilizing discharges in ablative polyacetal -  $(\text{CH}_2\text{O})_n$  capillary [45]. Because at that time most of the groups working in this field used gas-filled or evacuated ablative capillaries, such discharges were intensively studied both experimentally and theoretically. The main results achieved in this field by contribution of the author were published in papers [99], [102] and in the Ph.D. thesis [70].

The investigation of discharges in non-ablative capillaries was conducted in order to create highly uniformed, high-density metal-vapor plasmas. The creation of the metal-vapor was based on the explosive ablation of electrode material in  $\mu\text{s}$ -discharge. Such kind of plasma can be used as an active medium in future capillary-discharge based lasers emitting at shorter wavelengths [52], [70]. Several diagnostic methods were used to investigate the created metal-vapor [103], from which the study of plasma homogeneity and purity [70] was partly accomplished at the LIBS laboratory of Brno University of Technology (see Fig. 19). It was shown in [103] that optimizing

the electrode configuration, capillary dimension and material, it is possible to create pure metal-vapor plasmas with peak densities above  $10^{19} \text{ cm}^{-3}$ . The plasma density can be controlled by varying the discharge parameters.



*Figure 19.* The setup for LIBS measurements at the Brno University of Technology, Czech Republic.

## 5.2 CONSTRUCTION OF A CAPILLARY DISCHARGE BASED LASER

From 2000 the main effort of the research team the author is member of was focused on a construction of Rocca's type  $\text{Ar}^{+8}$  laser. The source was built at the University of L'Aquila, Italy. Its present experimental arrangement is shown on Figure 12 (page 16).

The description of the earlier experimental setup, with a possibility to apply double-pulse excitation; together with the measured excitation current and X-ray intensity can be found in [104]. However, utilizing this setup the measured peak of the emission in the bandwidth of  $(47 \pm 2) \text{ nm}$  selected by a Sc/Si spherical converging multilayer mirror indicated an amplification, no laser action was observed.

In the paper [105] a detailed description of the excitation circuit and detection part is given. The measurements and characterization of XUV radiation emitted by the source was provided in three different spectral regions of interest. The temporal evolution of the radiation with energy of  $(50\text{--}72) \text{ eV}$  ( $17\text{--}25 \text{ nm}$ ), which is a spectral range demanded by a number of applications including XUV microlithography, and in the "water-window" ( $2.3\text{--}4.4 \text{ nm}$ ) region that is important for X-ray microscopy in biology, was measured utilizing calibrated PIN diodes and applying appropriate filters. The energy emitted per pulse in the full bandwidth ( $17\text{--}25 \text{ nm}$ ) and ( $2.3\text{--}4.4 \text{ nm}$ ) was estimated to be  $\approx 45 \text{ mJ/sr}$  and  $\approx 10 \text{ mJ/sr}$ , respectively. The  $(47 \pm 2) \text{ nm}$  region, which consists the desired laser line, was selected by Sc/Si multilayer mirror. We had no evidence of laser emission at such wavelength. On the other hand, a strong spontaneous emission has been observed not only around  $47 \text{ nm}$  but also at much higher photon energies, up to more than  $500 \text{ eV}$ . This result, combined with the pin-hole camera images demonstrated that an efficient  $z$ -pinch effect has been reached. The lack of lasing was attributed to the missing pre-ionization current.

Simultaneously with the experiments, the plasma evolution in the argon-filled capillary was studied also with theoretical models. Modeling activities took place mainly at the University of Brno, Czech Republic and University of Pécs, Hungary. The comparison of the experimental analysis of plasma evolution in the  $z$ -pinch with a theoretical model can be found in paper [6]. So-called 'simple model of  $z$ -pinch' was used to simulate the dynamics of the compression. Utilizing this model, the effects of the electrical circuit parameters and the initial discharge conditions on

the plasma evolution and the production of soft X-rays in the three different spectral regions discussed above were demonstrated. In the simulation a sinusoidal current shape was used. It was shown that the influence of electrical circuit is appropriately described by the charging voltage of the capacitor and the inductance that determines the current rise time and total energy put into the plasma. The dependence of different plasma parameters on the initial argon gas pressure in the capillary was studied both experimentally and theoretically. The experiments were conducted using calibrated PIN diodes covered by 0.8  $\mu\text{m}$  thick Aluminum or 0.5  $\mu\text{m}$  Vanadium filters in order to select radiation in the region 50-70 eV or 250-500 eV, respectively. The validity of a model was also verified by performing calculations of the experimental conditions reported in works describing amplification on 46.9 nm transition of Ne-like Ar in polyacetal and ceramic capillaries. Good agreement was found with results obtained in ceramic capillaries. It was shown that for polyacetal capillaries the mass ablation from the capillary wall should be taken into consideration.

In order to find appropriate plasma parameters for gain saturation in  $\text{Ar}^{+8}$  soft X-ray laser experiments, the gain in the  $3p^1S_0 - 3s^1P_1$  transition (46.9 nm) of  $\text{Ar}^{+8}$  was analyzed as a function of the electron temperature  $T_e$ , density  $N_e$  and plasma radius  $r_{pl}$  [106]. For the analysis a quasi-steady state approximation using a pure atomic kinetics code was applied. It was found that however the amplification exists in the large temperature/density domain ( $T_e = 60 - 150$  eV,  $N_e = 0.5 - 10 \times 10^{18}$   $\text{cm}^{-3}$ ), the limit  $G \sim 1.4$   $\text{cm}^{-1}$  required for the gain saturation in 15 cm long plasma column is reached in the extremely narrow density regions at high temperatures [106].



*Figure 20.* The capillary-discharge based soft X-ray laser at the University of L'Aquila, Italy [108].

The achievement of lasing by our group was reported in a paper [58]. The lasing was obtained utilizing set-up shown on page 16. From the construction point of view, as a key condition for stable operation of the source showed mainly

- the utilization of an independent circuit for pre-ionization and
- the modification/improvement of the water-insulated spark-gap.

The Ne-like Ar laser operating at 46.9 nm has been realized by exacting the active medium (the pre-ionized argon gas inside the capillary) with a current pulse having a peak value ranging from 26 to 40 kA and a relatively long half-cycle duration (140 ns). The current pulse was produced by discharging a 10 nF water dielectric capacitor through a low-inductance circuit consisting of the capillary tube and water spark-gap. The capacitor was initially charged up to 200 kV by a six-stage



Marx generator. The time evolution of the emission was monitored by a fast vacuum photodiode (XRD). The first spectroscopic identification of the radiation was realized utilizing a Sc/Si multilayer mirror by a normal incidence reflexivity of 35% at the wavelength 46.9 and with a bandwidth  $\pm 2$  nm centered in this wavelength. The time evolution of XRD signal obtained by applying the Sc/Si multilayer mirror clearly demonstrates that the intense peak of radiation at  $\sim 30$  ns after starting of the current pulse belongs to the  $3p-3s$  ( $J = 0-1$ ) transition of the Ne-like Ar at 46.9 nm. The intensity of the laser emission was investigated also as a function of the initial Ar pressure. The amplified spontaneous emission in a 3 mm in diameter, 151 mm long capillary channel and a peak discharge current of 32 kA was observed in a relatively small pressure interval, ranging from 0.2 to 0.4 Torr. The lasing line was identified also by further spectroscopic measurements. The typical example of the measured spectrum is shown on Fig. 21.

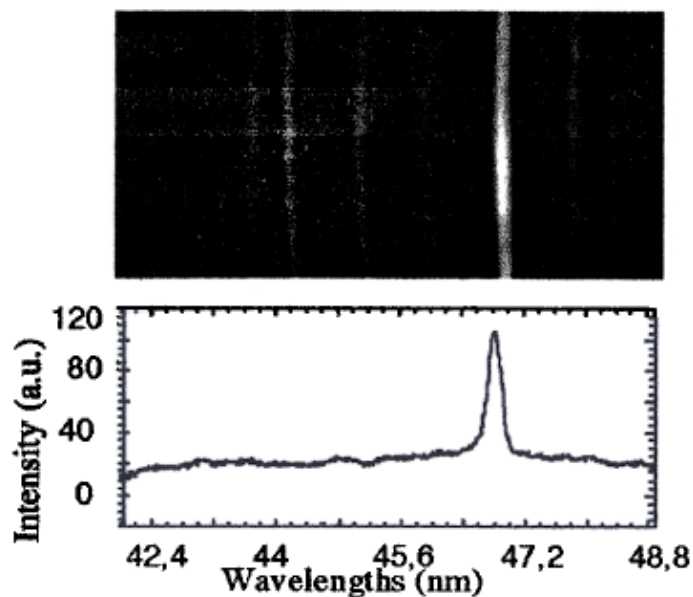


Figure 21. Measured spectra of the  $\text{Ar}^{+8}$  laser using 3 mm in diameter 20 cm long  $\text{Al}_2\text{O}_3$  capillary filled with 3.2 mTorr of Ar and pumped with a 30 kA current pulse [109].

The observation of the sub-milliradian divergence operation of the capillary-discharge soft X-ray laser is reported in the work [107]. In these experiments an active media was created inside the up to 30 cm long alumina ( $\text{Al}_2\text{O}_3$ ) capillary channel. The saturation behavior of the beam was investigated by measuring the laser pulse intensity (by XRD diode) as a function of the capillary length in the range of 8-30 cm at the optimized pressure 0.45 Torr. The saturation of the active medium was achieved at the channel length of  $\sim 16$  cm with the effective gain  $G_{\text{eff}} \sim 0.8 \text{ cm}^{-1}$ . The detection line for plasma imaging consisted of a MCP coupled to a phosphor screen and a CCD camera. In the far-field measurements the detection system was placed in a front of the laser beam at 140 cm distance from the capillary output. The near-field beam patterns were recorded by utilizing two Sc/Si multilayer mirrors in configuration that resulted on the detection plane a 1:1 image of the laser beam. During the investigation of the pressure-dependence of the measured far-field images three different profiles can be observed, from which one is particularly interesting. For a pressure interval 0.5 – 0.45 Torr the laser emission consists of two spatially separated structures, i.e. a very narrow axially centered spot and a thin ring. The most part of the laser pulse energy ( $\sim 80\%$ ) is contained in a central spot whose diameter at FWHM corresponds to a divergence of 0.7 mrad. This value, to our knowledge, represents the first observation of a sub-mrad divergence soft-X-ray laser obtained by a capillary discharge.

### 5.3 APPLICATION OF SHORT WAVELENGTH RADIATION SOURCES

The possibility to apply capillary discharges as plasma waveguides was discussed in Chapter 3.3.2. This application is investigated also by our group. Theoretical study of conditions for the

formations of plasma-based waveguide in non-ablative capillary  $z$ -pinch is presented in paper [94]. The waveguiding conditions were investigated for two different cases, i.e. in the  $z$ -pinch discharges pumped by high-amplitude fast current pulse and in the discharges with low-amplitude long current-pulse. Appropriate waveguiding conditions can be observed in both cases. On the base of these simulations, we showed that the ablation-free capillary  $z$ -pinch provides conditions suitable for guiding of high-intensity laser pulses. Experimental study of this phenomenon is under preparation on a modified setup at the University of L'Aquila.

The results described in the paper [17] correspond to the field of most recent interest of the author. The results described in this paper were obtained on the base of interdisciplinary research devoted to monitoring of accumulation of different metals in biological samples. In order to find appropriate plant species for detecting and natural removal of contaminants from polluted areas, the accumulation properties of various plants should be investigated. The mapping of possible biological structures, which can specifically accumulate metals within a given tissue can be realized by means of different techniques, such as X-ray microscopy in the water-window, X-ray fluorescence microprobes, fluorescence microtomography etc. For this purpose we are utilizing three different methods. The first two are based on X-ray microscopy and micro-radiography investigations, which make use both of soft X-rays generated either by plasma laser source ( $\sim 1$  keV) or synchrotron radiation at rather high photon energies (8.5-35 keV). The third one is the LIBS technique, which reveals as a powerful, low-cost and relatively simple method.

In [17] we report on some X-ray experimental techniques, i.e. on dual energy micro-radiography and tomography measurements, conducted at the Synchrotron Elettra, Trieste, Italy. The light source used (SYRMEP beamline) has appropriate parameters to conduct X-ray dual-energy micro-radiography (i.e. the analysis of the difference between the images obtained above and below the X-ray absorption edges) measurements for samples with selected metal contamination. The capability of these methods was demonstrated on mapping copper and lead in 2D and 3D samples. The samples we investigated were leaves or section of leaves (see Fig. 22), or

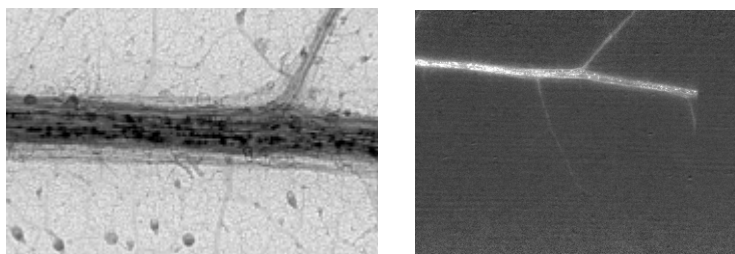


Figure 22. Results on the lead detection in 5days, 10mM PbAc treated *Helianthus annuus* sample. Detail of the treated sample (left) together with the metal distribution within the sample obtained by dual-energy analysis (right) [110].

section of roots. Each plant, from which the investigated samples were taken, was treated with different concentrations of contaminants (0 – 10 mM). It was also shown that with further improvement, relying mainly in the use of lower photon energies and carefully calibrated samples, accuracy appropriate even for analytical measurements can be reached.

## SUMMARY

In the habilitation work the results on the study, construction and application of plasma based short wavelength radiation sources are summarized. In spite to the fact that the main emphasis is on compact, capillary-discharge based sources, the other alternatives for generation are also introduced.

The experimental and theoretical approaches detailed in this work resulted in the early 1980s on the construction of X-ray lasers. However in the past 25 years these devices were improved considerable, we still do not have a laser emitting at wavelengths of several tens of nm commercially available. The main difficulties lie in the fundamental principles of interaction of radiation with matter; in creation of suitable active media and in controlling of its parameters. A

number of different schemes predict reasonably good conditions for achieving lasing at short wavelengths, but up to now only very few of them was realized experimentally. The recent advances in laser-plasma based (table-top) X-ray lasers have been made possible mainly by the development of new, compact, high-power driving lasers. The majority of present compact laser systems use CPA (chirped-pulse amplified) solid-state amplifiers. Typical wavelength range of these lasers is 10-20 nm and the output energy is in order of 10  $\mu$ J.

The search for alternatives of laser drivers in X-ray laser schemes at the late 1980s initiated the development of soft X-ray lasers based on capillary discharges. The present capillary-discharge based soft X-ray lasers emit radiation at 46.9 nm with pulse duration  $\sim$  2 ns and energy  $<$  1 mJ.

Further development of table-top soft X-ray lasers is expected by combining the two techniques, i.e. capillary discharge and laser-plasma produced sources. In this arrangement gas-filled or ablative capillaries are creating a medium, which can be pumped longitudinally by external laser pulse. The pump radiation can be guided inside the pre-formed plasma column and excite the active media with a length of a few tens of centimeters.

We should also note that due the possible application mainly for microlithography, the incoherent short-wavelength radiation sources are investigated extensively. Due their compactness and reliability capillary-discharge based sources seem to be good candidates for this application.

The research activities of the author are oriented to the investigation and application of short-wavelength radiation sources. The present research is based on his earlier works, mainly on:

- Construction and investigation of capillary-discharge based metal and dielectric vapor generator.
- Investigation of physical processes in ablative and non-ablative discharges.
- Study and development of capillary-discharge based soft X-ray laser.
- Application of short-wavelength radiation sources for investigation of biological samples.

During the studies the author of this work achieved the following main results:

- Including the role of electrode ablation into the models of  $\mu$ s (evacuated) capillary discharges. Presenting the “Surface – Explosion Theory” that shows the principal role of the explosive ablation of the electrode surface in the radial motion of mass and the generation of dielectric vapor plasmas for this type of discharges.
- Construction and study of the metal-vapor generator - creation of the pure, homogenous metal-vapor plasma column with peak density  $\sim 10^{19}$  cm $^{-3}$  by explosive ablation of electrode material (Fe) in  $\mu$ s-discharge utilizing a short ceramic (BeO) capillary. Such kind of plasma column can be used as an active medium in future capillary-discharge based lasers emitting at shorter wavelengths.
- Building a LIBS laboratory and investigating the metal-vapor purity by LIBS technique.
- Working out theoretical models for investigating the plasma dynamics and radiative properties in z-pinch discharges. It was found that however the amplification exists in the large temperature/density domain ( $T_e = 60 - 150$  eV,  $N_e = 0.5 - 10 \times 10^{18}$  cm $^{-3}$ ), the limit  $G \sim 1.4$  cm $^{-1}$  required for the gain saturation in 15 cm long plasma column is reached in the extremely narrow density regions at high temperatures.
- Participation on the construction of the capillary-discharge based Ar $^{+8}$  soft X-ray laser at the University of L’Aquila and achieving the lasing as the first laboratory in Europe.

The results obtained by the author contributed to the improvement of the capillary-discharge based short-wavelength radiation sources and made a solid base for the ongoing investigations in this field. The recent works on the study of the plasma-waveguiding and monitoring of the heavy-metal hyperaccumulation in biological samples foresee the areas of application of these sources.

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## SHRNUTÍ

Po dosažení laserové generace v roce 1960 byla intenzivně zkoumána možnost generování koherenčního záření na krátkých vlnových délkách - v oblasti rentgenového záření. Lasery vyzařující v této oblasti nabízejí řadu využití ve vědeckých i průmyslových aplikacích. Monografie je věnována zdrojům, které využívají ke generování krátkovlnného záření plasmu o vysoké teplotě, tj. oblasti kde dosud bylo dosaženo nejvýznamnějších výsledků. V práci je kladen důraz na popsání kompaktních, koherentních a nekoherentních zdrojů záření využívajících kapilárních výbojů. Nejprve jsou uvedeny v širším kontextu v porovnání s ostatními metodami a poté jsou diskutovány podrobně. Příložené publikace dokumentují přínos autora v oboru. Práce se skládá ze čtyř hlavních částí.

V první části je podán historický přehled. Je vysvětlen význam slova „plasma“ a jsou popsány nejvýznamnější směry výzkumu plasmu. Jsou diskutovány podmínky použití plasmu v zdrojích krátkých vlnových délek, spolu s hlavními aplikacemi těchto zdrojů. Navíc jsou zde uvedena schémata k realizaci rentgenových laserů. Druhá část se zabývá experimentální realizací těchto zdrojů. Nejdůležitější výsledky uplynulého období jsou popsány spolu se současnými postupy a do budoucna plánovanými uspořádáními. Samostatná kapitola je věnována konstrukci zdrojů a diagnostice plasmu generované pomocí kapilárních výbojů. Je diskutováno využití laseru  $\text{Ar}^{+8}$  založeného na kapilárním výboji, společně s úvodem do problematiky nekoherentních zdrojů EUV záření. Třetí kapitola je věnována teoretickým studiím. Jsou zde uvedeny modely pro popsání dynamiky plasmu v z-pinchi společně s modely pro výpočet parametrů plasmu a emitovaného záření. Jsou zde rovněž diskutovány fyzikální procesy odehrávající se během výboje v ablativních kapilárách. Čtvrtá část shrnuje přínos autora v oboru. Společně s krátkým výčtem výsledků z dřívějších prací, je zde přiloženo osm komentovaných publikací. V těchto člancích jsou popsány výsledky získané autorem z posledního období.