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The use of hot and dense microplasmas as gain media powered a renaissance of X-ray lasers on a tabletop. The research was boosted forty years ago in the Star Wars era for military applications. In the last decade, however the number of scientific and industrial applications has grown. Stimulated emission from a highly ionized shell is a transient process that is difficult to accomplish. Still both laser-produced plasma as well as discharge-produced plasmas have tacked the challenge. Switzerland is at the forefront in this unique field.

1. Introduction

The generation of laser light at wavelengths shorter than ultraviolet is a virtually impossible task. As dictated by the scaling of spontaneous versus stimulated emission at shorter wavelengths, the ratio between the Einstein coefficients A and B shows a power of 3 dependence with the wavelength. Since X-rays are 4 - 5 orders of magnitude shorter in wavelength than the UV-Vis, spontaneous emission dominates over stimulated emission by much more than 10^{12}-fold.

Indeed the "short wavelength range" comprises spectral different spectral domains with specific names and characteristics. Ranges such as extreme ultraviolet (EUV), soft X-rays (SXR), and hard X-rays (HXR) have been often confused. The physical boundary is defined using the refractive index jump between a strongly absorbing (e.g. EUV and SXR) to a notoriously penetrating (i.e. HXR) behavior while irradiating matter.

The accomplishment of "Light Amplification by Stimulated Emission of Radiation" (LASER) presumes a so-called population inversion. In a population inversion, there is a shift of the bound electron distribution toward upper energy levels, with transient vacancies at lower or ground levels. Such core excitation lifetime is all the shorter, the larger the energy gap. At energies of hundreds of eV's and more, as found in the short wavelength domain, the corresponding lifetime is too short for stimulated emission to be an active gain process.

A further challenge to make a X-ray laser happen, is the availability of a suitable gain medium. The need to extract high energies will inevitably cause the destruction of any gain material. Therefore, both liquids and solids are to be ruled out as X-ray gain media. Gas lasers are well-known even in the deep UV (exciplexes), but no ordinary substance would permit X-ray amplification. It needs an exotic one such as a plasma.

It must be clarified that so-called X-ray Free-Electron Lasers (XFEL's) are coherent short wavelength sources that do not work on the mechanism of stimulated emission on bound transitions. Such accelerators extract radiation from the oscillatory motion of charged particles, whereas the coherence can be increased by means of radiative feedback to the emitting particle bunch. Hence, not all coherent sources are, strictly speaking, "lasers" i.e. based on stimulated emission. Fig. 1 shows that all the available short-wavelength sources cover a wide range in brightness.

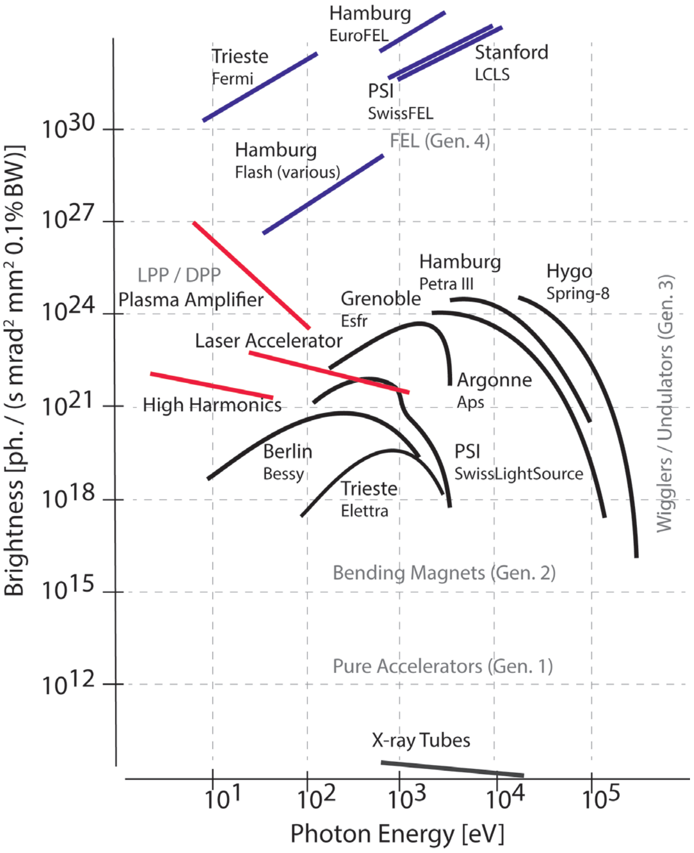

Fig. 1. Comparison of brightness for coherent short wavelength sources.

2. Basic Underlying Physics

2.1. Plasma Gain Medium

A viable option to stimulate the emission of EUV/SXR photons (20 - 500 eV) is using a gain medium with such large energy transitions, i.e. based on highly ionized atoms of a plasma [1]. The concurrence of such ions and a bath of free electrons, as it is in a plasma, is important. Highly ionized atoms of several charge units can be produced on a tabletop as a transient state in electrical discharges or alternatively by focusing a high peak power laser onto a solid target. Such expanding plasmas are very hot-and-dense at the birth moment while rarefy rapidly. The electron temperature and electron density characterize the medium (Fig. 2). For the sake of X-ray lasing across a plasma medium, the electron density is important to sustain an optimum number of excitation collisions between the free and the bound electrons [2]. This collisional excitation should bring the ion into a population inversion. If the collision rate is insufficient, the laser amplification is hindered. If the collision rate is too extensive, deexcitation or local thermalization also hinder

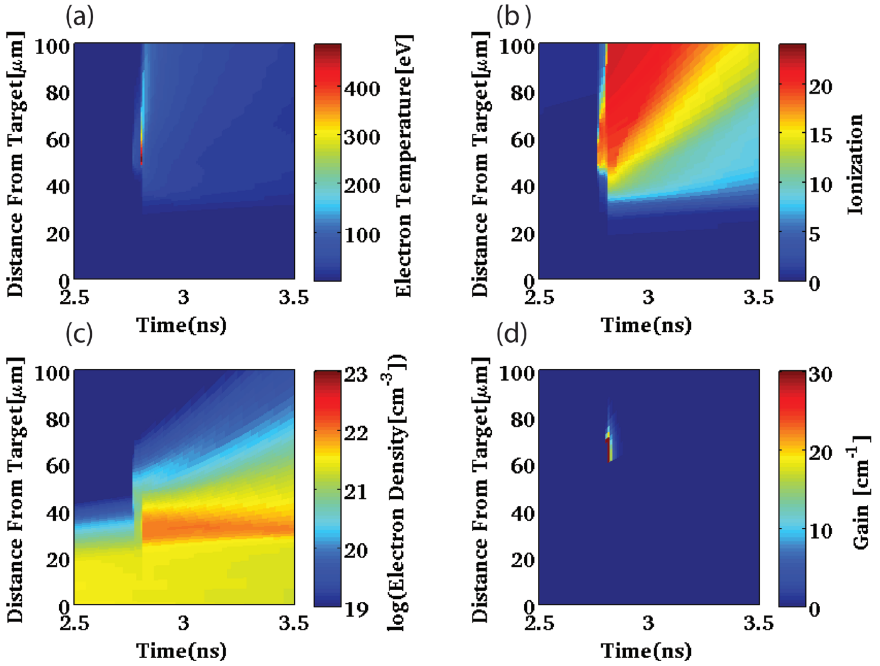

Fig. 2. Hydrodynamics as a function of time and expansion distance of a laser-produced plasma generated by irradiation of a Sn target with a pre-pulse (t = 0) and a main pulse (t = 2.7 ns). a) Electron temperature: one notes the short transient window for hot plasma lasing at the instant of main pulse delivery (see also d.); b) Ionization degree: at the hot condition a +22 ionization (Ni-like Sn) is accomplished, which due to plasma rarefaction does not recombine; c) Electron density; d) Calculated laser gain. From: Masoudnia & Bleiner [3].

population inversion. Obviously, the energy exchanged in the collision should be effective to promote the bound electron to the upper laser level, which demands the temperature of the plasma to be at a hot optimum. In previous publications, we have analyzed in detail the exact values of optimum conditions [2-5]. Such hot-and-dense plasma may support amplified spontaneous emission (ASE) across the plasma length. The requirements are strict and discussed in the next section.

2.2 Lasing by means of "Ionization Locking"

As the temperature does fluctuate across the plasma volume, also as a function of time, one needs to "lock" the related impact on the population inversion for a period long enough for the gain to establish. The gain relies on metastable levels in closed-shell ions working as upper laser level. Metastable levels are populated by means of collisional interaction (monopole excitation). These are between free electrons and bound ones in the ground level, where the selection rules do not allow a dipole transition. Henceforth, if the transition cannot be accessed radiatively (Fig. 3), the upper laser level state cannot relax spontaneously. The only ways to relax are either by stimulated emission, or by collisional de-excitation. The former contributes to gain, while the latter is realized at high electron densities.

As said, one needs to stabilize the ionization stage, otherwise the metastable state can be destroyed by means of ionization and/or recombination. This is done by means of ionization locking. Fig. 2.b shows the fractional abundance of ion stages for Sn calculated by means of coupling collisional and radiative rate equations (so-called non-local thermic equilibrium) for the hot and dense plasma core as well as for the corona. The plot shows that for closed shell configurations, i.e. +22, the specific ion stage is stable over a larger range than non-closed shell configurations.

For instance, a closed shell is the $^1S_0$ term of neon-like ions, i.e. with 10 residual electrons. Indeed, Ne-like argon (atomic number 18) giving soft X-ray lasing in a capillary Z-pinch discharge is eight-fold ionized Ar^{8+} (Ar-IX). Ne-like ions support lasing at modest energies of tens of eV that are possible on a tabletop. In order to extend the lasing to shorter wavelengths, higher atomic numbers are necessary. So far, Nickel-like ($^3F_4$), Zn-like ($^1S_0$) or Pd-like ($^1S_0$) ions have demonstrated lasing at energy as high as several hundreds of eV.

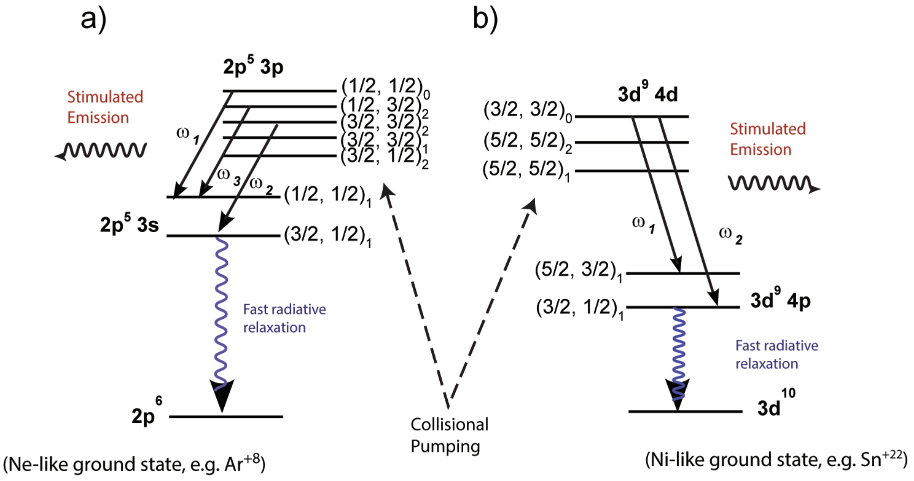

Fig. 3. Comparison of Grotrian schemes for the Ne-like and the Ni-like shells that are used to generate amplification across the 3p-3s and 4d-4p transition respectively. The upper laser level is metastable since it can be only populated by means of collisional excitation. The lower laser level is rapidly emptied thanks to dipole-allowed relaxation, which gives the upper-to-lower level population inversion.

An alternative approach to population inversion, instead of collisional excitation, relies on free electron recombination. While the collisional approach discussed above is easier to realize and scale, it is less Stokes efficient. The use of a hydrogen-like C^{5+} ion that produced lasing by recombination at $\lambda = 13.50$ nm (n = 4 to n = 2) and 18.22 nm (n = 3 to n = 2) has remained promising for long time to be scaled up. The underlying difficulty is that one needs a strong temperature contrast interface: a hot core where to breed high ionization radiators, directly in contact with a cold shell to induce massive recombination.

2.3 Advantage of the Plasma X-ray Laser

The accomplishment of amplified spontaneous emission (ASE) is based on fluorescence that stimulates emission in population-inverted ions. The sweep of this process across the plasma-column length gives an active gain. As the process occurs, the natural linewidth experiences gain narrowing. Henceforth, the plasma X-ray laser is extremely monochromatic, with linewidths much narrower than the natural width, in the range of $\Delta\lambda/\lambda < 0.001$ % or < 1 meV. This spectral purity is at the base of the enhancement of the peak brightness (Fig. 1), and permits high-resolution applications such as spectroscopy and/or lithography.

Furthermore, this is the only X-ray laser able to concomitantly emit two coherent lines at a significant energy span for interest in applications, as we demonstrated a few years ago [5]. While these lasers are extremely coherent in time, they are not so in the transverse direction. This means that the wavefront flatness is rather limited to approx 15 - 20 % of the pulse diameter, as it was shown before [6].

As the ASE is primed by fluorescence, the poor spatial coherence is not surprising. In order to improve the wavefront quality, and enhance collimation, a technique known as injection seeding has been used. This implies to prime the lasing using a good quality external pulse. One has to distinguish between self-seeding and external seeding. The former is accomplished with the concomitant (slightly delayed) generation of two plasma lasers, one injected into the other. In the case of external seeding, high-harmonic generation (HHG) is used which however makes the system grow in complexity.

3. Experimental Realization on a Tabletop Setup

The physics of laser action across a plasma is rich and dominated by unsteadiness and local transient processes. From an experimental standpoint, the overall process relies on specific critical steps to drive the amplification across a microplasma. To begin with, a pre-plasma has to be generated, a weakly ionized plasma that is suitable to prepare the main process of a hot-and-dense plasma gain-medium. A single-pass (no cavity possible) gain process can grow above a lasing threshold, if the plasma width-to-length is within an optimum aspect ratio. We derived the exact relation in a previous publication [7], which would explain why in some case the output is modest while in some other is at saturation. The drive-laser line-focusing (discussed below in sect. 3.1) or discharge confinement capillary geometry are thus crucial.

As the electron density is thicker in the core of the plasma and thinner at the margins, the opacity is also inhomogeneous. While the opacity of a medium determines its refractive index, one experiences larger refraction at the boundary of the plasma column. With that, the ASE tends to give a diverging X-ray laser pulse, as wide as 5 mrad at the output, with a donut profile. We have showed a solution to this issue, using a telecentric normal-incidence collector, fabricated with a multilayer coating [8]. Multilayers working as Bragg reflectors [9] permit to realize reflective optics for this wavelength range, to overcome the limitations of grazing incidence optics. State-of-the-art experimental concepts have been reviewed recently [10]. Here a short synopsis is given about plasma-based X-ray lasers. In general one distinguishes between two architectures (Tab. I): (i) optically-pumped, and (ii) electrically pumped.

Pumping	Optical	Electrical
Plasma Medium	Ne, Ni-like Ions	Ne-like Ions
Drive Power / Energy	0.2 - 20 TW / 0.3 - 30 J	1.5 GW / 2 J
Conversion Yield	$< 10^{-5}$	$\sim 10^{-5}$
X-ray Pulse Energy	~ 10 - 30 μJ	~ 20 - 50 μJ
Number of Photons	$\sim 10^{11}$	$\sim 10^{11}$
Wavelength	3.56 - 42.10 nm	46.87 nm
Linewidth	< 1 meV	< 1 meV
Pulse Duration	< 1.5 ps	~ 1 ns
Repetition Rate	< 10 Hz	1 - 100 Hz

Tab. I: Main characteristics of the alternative methods for plasma lasing.

3.1 Optically-Pumped X-ray Laser

One approach to generate hot-and-dense plasma gain-media is line-focusing a high peak power laser on a target [11]. Line focusing is important to produce high aspect ratio plasma columns by means of laser ablation [12]. Line focuses can be realized using tilted mirrors: the related astigmatism stretches the focal spot over a thin oval. Alternatively one could illuminate a spherical mirror parallel to the optical axis, such that the focal spot is affected by spherical aberration. Obviously, one could also obtain a line focus using a cylindrical lens: this practice is however disadvantageous because for high pulse intensities is hard staying below the damage threshold.

In fact, the drive-laser should have a pulse duration in the range of ps, leading to high peak powers. Shorter pulse duration of fs, although enhance the peak power, can deposit the energy in a time-scale too fast to exploit the full gain buildup time [4]. Obviously, long pulse durations of ns are too slow to pump the population inversion and ASE gain. The drive laser wavelength is typically in the infrared (IR), although visible pulses can penetrate deeper into the expanding opaque plasma.

A pre-pulse of 5 - 20 % the energy of the main pulse enhances the efficiency. The delivery of the main pulse with a few ns delay permits the pre-plasma to expand and relax the opacity and high-density gradients. The main pulse is thus advantageously coupled. Our group has also introduced a double pre-pulse technique to improve the control and alignment [6]. The pre-pulse was traditionally delivered from a separate laser than the main pulse: while the pre-pulse was a long pulse on-axis to the target, the main pulse was a ps-pulse at grazing incidence. The alignment of these two completely independent pulses, both in time and space, is crucial and critical. Jitter effects or minimal optics tilt would cause a detrimental mismatch of the two pulses on the target. Therefore, our group introduced the use of a dog-leg beam-splitter to generate a pre-pulse from the same laser used for the main pulse.

The pumping requirement (Fig. 4) to realize X-ray plasma lasing have substantially relaxed over the decades. If in the

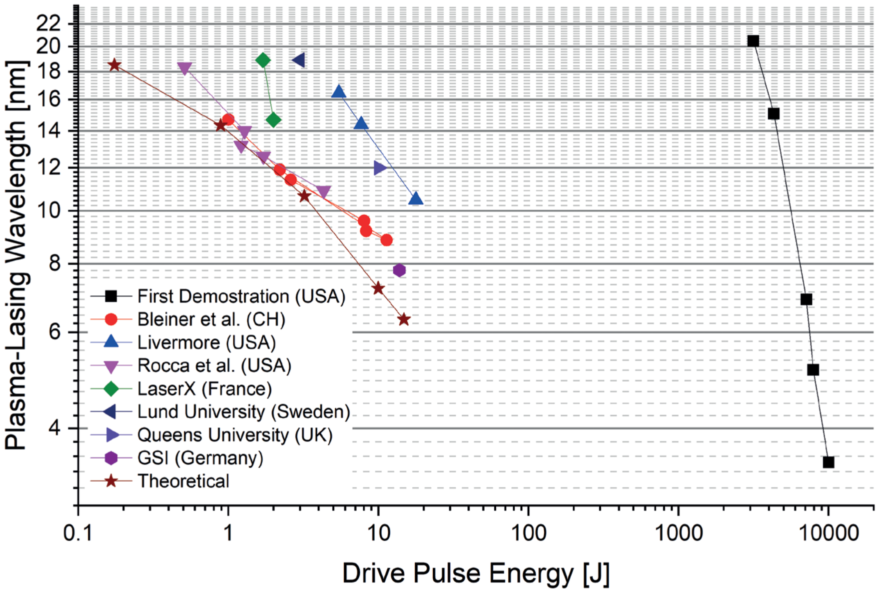

Fig. 4. Progress of the drive pulse requirement to accomplish tabletop X-ray lasers, from the first demonstration at the LLNL Laboratories forty years ago [13].

early times a multi-kJ laser was mandatory (which is an immense facility, with just a few pulses per day), nowadays a few hundreds of mJ can do the job.

3.2 Electrically-pumped X-ray Laser

While a kJ laser is a gigantic facility, a kV generator is a handy device. Henceforth, plasmas produced as electrical discharges have proven a suitable alternative to reduce the footprint and increase the average power (pulse repetition rate). The discharge in a capillary of approx. 20 cm produces the above-discussed plasma column. The pre-plasma is produced with a RF coil and a DC lead. The work gas is mainly Ar as other gases did not prove as effective. Ne-like Ar gives a strong lasing at 46.9 nm wavelength. The main pulse is delivered as a pulse-forming network drives a 40 kV / 40 kA signal, switched in 1 ns by a hydrogen thyratron.

The forward power transfer to the plasma column as load must be optimized by matching the impedance, using a so-called matching box.

4. Alternative Concepts for Short Wavelength Coherent Light

The focus of this update is tabletop X-ray lasers, based on plasma as a gain medium. There are however alternative concepts to realize tabletop high-photon-energy coherent light pulses. For completeness, we briefly mention them, pointing at the main differences with plasma-driven lasers. Coherent light is indeed a wider category than "lasers": the latter strictly implies energy extraction across a gain medium by stimulated emission.

A popular approach is that of utilizing parametric processes. In this case, there is no energy extraction from a medium. A non-linear medium is used for energy conversion involving a quadratic (or higher order) response of the polarization to a driving electric field. The non-linearity requires high amplitudes of the drive fields, i.e. as high as ~ GV/m (or > 0.1 TW/cm^2 in pulse peak intensity). The interaction happens instantaneously through a virtual state: energy and momentum are obviously conserved, making so-called phase-matching critical. High-Harmonic Generation (HHG) is the most popular of these parametric techniques. A high intensity pump beam across a non-linear medium, e.g. noble gas, will generate a comb of odd harmonics of the fundamental wavelength. The conversion yield drops after a cut-off harmonic order, such that at soft X-rays the photon budget is at best 10^7 photons. The details of HHG are beyond the scope of this article, see for instance [14-19].

A further approach to obtain coherent short wavelength light on a tabletop is that to develop scaled-down particle accelerators, in order to obtain synchrotron radiation in a similar fashion as it happens in the full-scale accelerators. We are involved in one such EU project within the Eupraxia consortium (http://www.eupraxia-project.eu/).

Let's consider the undulator equation which gives the wavelength of the n-th harmonic as a function of the period of the undulator poles (λ_u), the relativistic Lorentz factor γ and the deflection parameter K (function of the magnetic field) and the deflection angle θ:

$$\lambda_n \approx \frac{\lambda_u}{2\gamma^2}\left(1+\frac{K}{2}+\gamma^2\theta^2\right) \qquad (1)$$

One realizes that if λ_u is substantially scaled down, a comparable output wavelength λ_n is obtained at much lower γ. If a relativistic electron is driven by the oscillating field of a laser, instead of an accelerator of hudreds of meters, this condition is accomplished, such as in so-called betatron or inverse Compton setups. Similarly, the oscillatory drive field that can be realized on the crest of an array of nano-structures, to accomplish an accelerator on a chip. A few recommended papers go into the details of this interesting concept [10, 20-22].

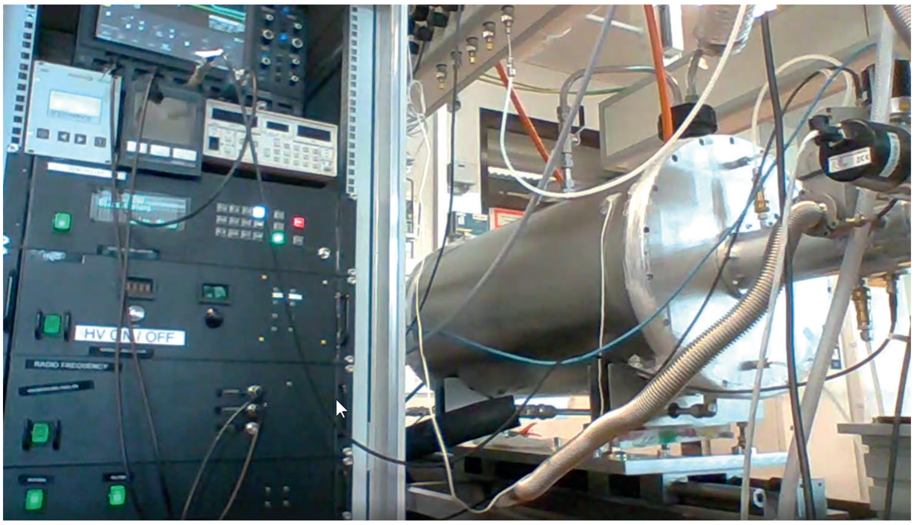

Fig. 5 The self-developed X-ray laser based on a electrical plasma discharge across a capillary.

5. Applications of tabletop X-ray lasers

There is a number of scientific and technical uses of EUV and/or SXR which become accessible on a tabletop [8]. The main strengths of such spectral range are the following three: (a) strong absorption, (b) nano-scale sensitivity, (c) efficient photolysis [23].

Defect inspection of nano-lithography masks is a technological application with huge economic impact [24]. The special physico-chemical processes that occur under the action of ionizing photons (radiolysis) are found also in nature in astrochemistry. Laboratory astrochemistry thus benefits for the possibility to produce and reproduce the quite exotic chemistry and radicals found in the extraterrestrial context. On the other hand , the wavelength range between the C_K edge (282 eV) and the O_K edge (533 eV) offers a strong contrast between organic and aqueous matter. This "water window" is thus attractive to perform in-vivo bio-microscopy.

The short wavelength favors the enhancement of the diffraction limit. As the latter scales as $\lambda/2$, a wavelength in the 10 - 100 nm means a substatial improvement of the spatial resolution. Nano-scale microscopy is thus possible without the use of electrons, but with similar advantages, and without sample preparation [25]. While illuminating the region of interest, chemical visualization in either pixel-scanning mode (mapping) or full-field snapshots (imaging) are possible [26, 27].

PD Dr. habil **Davide Bleiner** is the head of the Advanced Analytical Technologies at the Empa. He completed his PhD at the ETH Zurich in 2002 under the guidance of Detlef Günther on laser microanalysis. He has had postdoc positions at the Empa and then at the University of Antwerp. In 2007 he came back to ETH and worked as senior assistant in the Dept. of Mechanical Engineering, to develop a laser-produced plasma source for Extreme Ultraviolet Lithography. In 2011 he was awarded the SNF Professorship at the University of Bern in the group of Jürg Balmer and Thomas Feuerer. Finally, since 2014 he is at Empa. In 2019 he was listed in the top 100 analytical scientists by the magazine Analytical Scientist.

References

[1] D. Bleiner *et al.*, *Table-top x-ray lasers using a plasma gain-medium: limits and potentials*. Physica Scripta, 2014. **T162**.

[2] L. Masoudnia and D. Bleiner, *Optimum electron temperature and density for short-wavelength plasma-lasing from nickel-like ions*. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2014. **323**: p. 59-70.

[3] L. Masoudnia and D. Bleiner, *Hohlraum target for overcoming refractive losses in plasma x-ray lasers*. Laser Physics, 2013. **23**(5).

[4] L. Masoudnia and D. Bleiner, *Optimum Pump Pulse Duration for X-Ray Ar-Plasma Lasing*. Photonics, 2015. **2**(1): p. 164-183.

[5] L. Masoudnia, M. Ruiz-Lopez, and D. Bleiner, *Table-top two-color soft X-ray laser by means of Ni-like plasmas*. Physics of Plasmas, 2016. **23**(4).

[6] F. Staub *et al.*, *Soft-X-ray lasing in nickel-like barium at 9.2 nm using the grazing-incidence scheme*. Optics Communications, 2012. **285**(8): p. 2118-2121.

[7] D. Bleiner, *Saturation gain-length product during short-wavelength plasma lasing*. Applied Physics Letters, 2012. **101**(8).

[8] M. Ruiz-Lopez and D. Bleiner, *Implementing the plasma-lasing potential for tabletop nano-imaging*. Applied Physics B, 2013. **115**(3): p. 311-324.

[9] D. Bleiner *et al.*, *Actinic damage of Y/Mo multilayer optics in a table-top plasma-driven x-ray laser*. Appl Opt, 2014. **53**(22): p. 4894-902.

[10] D. Bleiner, *The science and technology of X-ray lasers: a 2020 update*. XVII International Conference on X-Ray Lasers. Vol. 11886. 2021: SPIE.

[11] D. Bleiner, J. E. Balmer, and F. Staub, *Line focusing for soft x-ray laser-plasma lasing*. Applied optics, 2011. **50**(36): p. 6689-6696.

[12] D. Bleiner *et al.*, *Laser-induced plasmas from the ablation of metallic targets: The problem of the onset temperature, and insights on the expansion dynamics*. Journal of Applied Physics, 2007. **101**(8).

[13] B. J. Macgowan *et al.*, *Demonstration of Soft-X-Ray Amplification in Nickel-Like Ions*. Physical Review Letters, 1987. **59**(19): p. 2157-2160.

[14] D. von der Linde *et al.*, *Generation of high-order harmonics from solid surfaces by intense femtosecond laser pulses*. Phys Rev A, 1995. **52**(1): p. R25-R27.

[15] S. Kim *et al.*, *High-harmonic generation by resonant plasmon field enhancement*. Nature, 2008. **453**(7196): p. 757-60.

[16] T. T. Luu *et al.*, *Extreme ultraviolet high-harmonic spectroscopy of solids*. Nature, 2015. **521**(7553): p. 498-502.

[17] K. Zhang *et al.*, *Shrinking the Synchrotron: Tabletop Extreme Ultraviolet Absorption of Transition-Metal Complexes*. J Phys Chem Lett, 2016. **7**(17): p. 3383-7.

[18] T. T. Luu *et al.*, *Extreme-ultraviolet high-harmonic generation in liquids*. Nat Commun, 2018. **9**(1): p. 3723.

[19] S. Roscam Abbing *et al.*, *Divergence Control of High-Harmonic Generation*. Physical Review Applied, 2020. **13**(5).

[20] E. Peralta *et al.*, *Demonstration of electron acceleration in a laser-driven dielectric microstructure*. Nature, 2013. **503**(7474): p. 91-94.

[21] N. V. Sapra *et al.*, *On-chip integrated laser-driven particle accelerator*. Science, 2020. **367**(6473): p. 79-83.

[22] D. Bleiner, *Tabletop Beams for Short Wavelength Spectrochemistry*. Spectrochimica Acta Part B: Atomic Spectroscopy, 2020.

[23] D. Bleiner, L. Juha, and D. Qu, *Soft X-ray laser ablation for nano-scale chemical mapping microanalysis*. Journal of Analytical Atomic Spectrometry, 2020. **35**(6): p. 1051-1070.

[24] J. E. Balmer, D. Bleiner, and F. Staub, *Extreme ultraviolet lasers: principles and potential for next-generation lithography*. Journal of Micro/Nanolithography, MEMS, and MOEMS, 2012. **11**(2).

[25] D. Bleiner *et al.*, *Evaluation of lab-scale EUV microscopy using a table-top laser source*. Optics Communications, 2011. **284**(19): p. 4577-4583.

[26] D. Bleiner *et al.*, *XUV laser mass spectrometry for nano-scale 3D elemental profiling of functional thin films*. Applied Physics A, 2020. **126**(3).

[27] R. Muller *et al.*, *Depth-Profiling Microanalysis of CoNCN Water-Oxidation Catalyst Using a lambda = 46.9 nm Plasma Laser for Nano-Ionization Mass Spectrometry*. Anal Chem, 2018. **90**(15): p. 9234-9240.