The Science and Technology of X-ray Lasers: A 2020 Update

D. Bleiner*[a,b],
[a]Empa Materials & Technology, Überlandstrasse 129 CH8600, Dübendorf, Switzerland;
[b]University of Zurich, Winterthurerstrasse 190 CH8057, Switzerland

The 17th International Conference on X-ray Lasers (ICXRL 2020) was a milestone in several ways. Firstly, was a milestone in this scientific community because for the first time the conference came to the participants' home, due to the outbreak of the pandemics, rather than the opposite. The organization of an online event was a challenge because was a brand new platform for both the organizers and the participants, and because it was put together in less than six months. The planned physical event in Rome, already tested with one year in advance, had to be withdrawn as it posed major risks to potential participants. ICXRL 2020 was milestone also because the increased proportion of papers on technological deployment of X-ray lasers. Of course, research on fundamentals and innovation still carried an important part of this thirty-four year event. Finally, ICXRL 2020 was a milestone also because there were participants from all five continents, no one excluded, and some were forced to incredible efforts to stay up and present their research. A great sign of inclusion. Last but not least, ICXRL 2020 saw four best presentation "Pierre Jaeglé" awards, equally split between female and male students: Adeline Kabacinski (LOA, France), Lydia Rush (CSU, USA, see ref. [33]), Felix Wiesner (FSU, BRD), Julius Reinhard (HIJ, BRD). Another great sign of inclusion.

In the present monograph, about 300 authors have gone even further and in the full SPIE spirit, made their research open access in written form. The exciting papers that are collected here, are a superb snapshot of where the community and the technology is in 2020. For practical purposes, the Editor has chosen to structure them in four sections. Each section is a major pillar in making a thriving science a robust technology to serve the wellbeing of society and the industry, without forgetting academia. Therefore, along with Empa Materials Science & Technology and SPIE, we are all indebted to the visionary companies that sponsored the event (see the front matter), which remained with the organizers even through the difficult times.

SESSION 1 -- X-RAY LASERS ON A TABLETOP: PAST & PRESENT

The utilization of X-ray lasers in the home laboratory is a forty year science, with future promises. Soon after the demonstration of optical lasers, the realization of coherent pulses "beyond ultraviolet" was on the table.

Mainly pushed by a few visionary theoreticians in the 1970s, and later on by late-night workers like Pierre Jaeglé [**1**], the field gained in momentum in the middle of the 1980s "Star Wars". The field actually took off from studies in X-ray absorption spectroscopy in a plasma, as "absorption anomalies".

The remarkable competence of the early workers in spectroscopy and quantum physics was matched by an incredible ability to produce and read the data. One initial mystery with the selenium X-ray laser was the absence of lasing on the 3p $^1S_0$ - 3s $^1P_1$ line at 18.3 nm that was

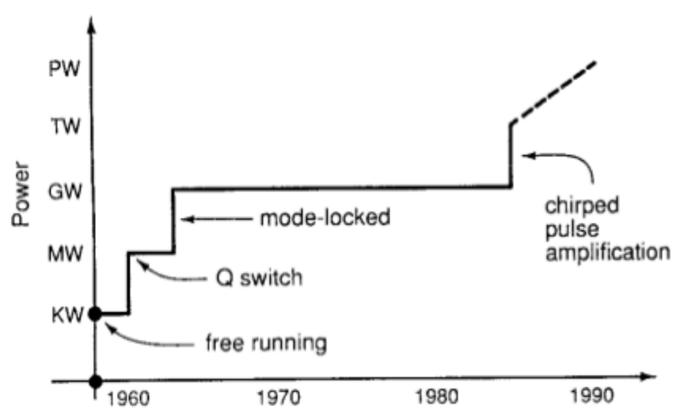

Figure 1: Development of peak power in laser technology.

International Conference on X-Ray Lasers 2020, edited by Davide Bleiner,
Proc. of SPIE Vol. 11886, 1188602 · © 2021 SPIE · CCC code:
0277-786X/21/$21 · doi: 10.1117/12.2604263

predicted to have the largest gain. At that time these scientists were only minimally supported by computers. Few large-scale laboratories in Italy, France, USA and the Soviet Union were receiving generous governmental funding. The competition between the United States and the Soviet Union was a significant driver in the development of the X-ray laser, and some details remain classified.

In the 1990s, X-ray lasing became a tabletop experiment. In 1992 the big breakthrough was the development of the pre-pulse technique. Different amplification principles were demonstrated as well as more efficient pumping schemes such as transient collisional excitation (TCE), grazing incidence pumping (GRIP) or travelling wave excitation (TWE). Nickel-like ions have the advantage of producing shorter wavelength lines than neon-like ones by a factor of two to three for similar excitation energies. The early nickel-like lasers were plagued with very low gain and never achieved saturated output. Yet they did reach wavelengths as short as 3.56 nm in nickel-like gold with huge pump energies.

Many advances took place in developing new materials [**2**]. Punctuated developments in laser amplification technology (**Fig. 1**) permitted to increase the peak power while shrinking down the setup footprint. Chirped pulse amplification was acknowledged internationally with the 2018 Nobel prize award. A milestone success for Mourou and Strickland, a milestone recognition for the entire high-power laser community.

Then in the last decade, workers started using them for applications. However much the capabilities of tabletop X-ray lasers have progressed, the tremendous impact of accelerator beamlines remains unmatched. It is clear that beamlines are limited in the access continuity and extend for a single user. Further, specific properties of the plasma-based X-ray lasers, such as the narrow linewidth, are unique [**3**]. Henceforth, there is a lot of expectation that in the future the open gap between tabletop setups and accelerator systems will be closed, such that a larger proportion of workers can be served.

This is promising a very powerful tool in the laboratory, with far-reaching impact on biology, chemistry and crystallography. One day it may even be used to resolve the structure of molecules with 24/7 throughput. Rocca has substantially contributed to this perspective, with the demonstration of high repetition rate X-ray lasers [**4**].

SESSION 2 -- ADVANCED X-RAY LASERS: FROM HIGH-END FACILITY TO TABLETOP

The realization of X-ray lasing has been accomplished by means of alternative mechanisms. None of these has yet established as the dominating one for technology transfer. Mainly this is due to open possibilities to combine the largest possible range of advantages and increase the technology readiness level. Proof-of-principle research is thus still very important as well as a combination of computational studies, experiments at compact systems and studies at large scale facilities. Further, the design and realization of experimental systems is an important part for the field's progress.

A few worldwide systems are based on a strictly tabletop architecture, as for instance presented in Hemani et al. **[5]** (Empulse in Switzerland). Nejdl et al. **[6]** (Extreme Light Infrastructure in Czech Republic) discusses a larger setup that still fit in a home laboratory. The definition of *tabletop setup* may thus be limited to any system with footprint smaller than 10 m^2, while one could define *laboratory setup* anything with footprint smaller than 100 m^2. Finally, anything larger than that should be called *high-end facility*.

In the efforts made to shrink down the footprint of coherent X-ray sources, some of the characteristics have to be compromised with respect to what found at a high-end facility. Hornberger et al. **[7]** discuss an innovative laboratory light source, which is a downsized synchrotron, thanks to the use of a laser-driven undulator. Alternatively, betatron sources have significantly contributed to the downscaling of accelerator beamlines for home lab operation. Betatron sources rely on the fs-laser drive of electrons in a gas jet. Chaulagain et al. **[8]** have imaged, with tomographic structure, the density distribution in such a gas medium.

Guilbaud et al. **[9]** have pushed the limit of state-of-the-art amplified spontaneous emission (ASE) plasma-driven lasers, showing the substantial improvement in pulse characteristics with seeding. The established use of harmonic generation pulses has been here extended by using vortex beams. Martinez Gil et al. **[10]** have presented the modelling framework to study plasma based seeded soft X-ray lasers. Kato et al. **[11]** review the mechanisms of ASE X-ray lasing comparing two alternative schemes, the recombination pumping and the collisional pumping.

Bencivenga et al. **[12]** presents non-linear techniques based on four-wave mixing able to probe ultrafast dynamics and quantum state correlations inaccessible by linear X-ray methods, tackling a major challenge for ultrafast X-ray science. While these techniques are largely carried out at free-electron lasers, Chau et al. **[13]** showed four-wave mixing accomplished on a tabletop setup. The field of non-linear spectroscopy in the home lab is substantially dominated by high-harmonic generation systems, which rely on non-linear photon conversion across a gas medium. Solid-state high harmonic generation is a relatively recent technique, as shown by Campi et al. **[14]**, that offers new insights into the dynamics of field light-matter interaction. For even more extreme sources, Jeong et al. **[15]** show that the relativistic-flying mirror is a promising candidate for generating attosecond intense electromagnetic pulses. Vagizov et al. **[16]** discuss a method for controlling the spectral and temporal characteristics of x-ray radiation produced by a radioactive or synchrotron Mössbauer source via its propagation through an optically thick sample of resonant nuclei with a modulated transition frequency.

SESSION 3 -- ADVANCES ON X-RAY LASER OPTICS & METHODS

One important use of coherent X-ray pulses is the enhanced resolution and the capability to nano-scale diagnostics. Several workers have combined computational and experimental studies to develop optical methods. The theoretical analysis finds a large challenge in the experimental demonstration, because minor deviation in the finishing quality and/or in the pulse wavefront may hamper the outcomes.

Begani-Provinciali et al. **[17]** present a new application of wavefront measurements, i.e. the tomographic reconstruction of a probed optically thin biosamples, by means of phase-contrasttomography (XPCT). Li et al. **[18]** studied the performance of Fresnel Zone Plates in medical imaging while controlling the number of zones or the illumination coherence. Pandey et al. **[19]** studied with wavefront diagnostics the effect of IR vortex beam on the X-ray generation, by means of high harmonic generation across a noble gas.

Besides, new concepts of optics fabrication and design are crucial to close the gap between theory and experiments. A Laue lens makes use of diffraction in the volume of a large number of crystal tiles mounted to concentrate incident radiation from a large collecting area in a small focal spot. Prasciolu et al. **[20]** report here on the use of multilayer Laue lenses to focus the intense X-ray Free Electron Laser (XFEL) beam at the European XFEL to a spot size of a few tens of nanometers. The presented computational results are subject of ongoing experimental study.

The coherence of the source, is obviously very important to the overall success of the optical method. Therefore, Dakroub et al. **[21]** have developed and implemented a diagnostic for the temporal characterization of seeded XUV laser pulses, based on laser-dressed photoionization in the sideband regime, using a self-made velocity map-imaging spectrometer as the central element. The technology readiness of research-level systems for high throughput imaging was reported by the CSU group. Wang et al. **[22]** demonstrate single-shot Fourier transform holography with a 7 μm diameter field of view and picosecond time resolution using a highly coherent ~5 ps pulse duration tabletop Ni-like molybdenum soft X-ray laser at 18.9 nm wavelength. This result is fantastic to show the capabilities of well-established ASE X-ray laser platforms.

Plasma X-ray laser was also used by Bleiner et al. **[23]** to image a Fresnel Zone Plate in a lensless configuration. Results show factor of 3 for the imaging resolution depending on the data processing method, and a major limitation given by the degree of the illumination homogeneity. Finally, van Tilborg et al. **[24]** investigated the use of laser-plasma betatron sources to carry out phase-contrast imaging at nano-scale.

SESSION 4 -- LABORATORY X-RAY LASERS: PRESENT & FUTURE APPLICATIONS

Coherent and incoherent X-ray sources are showing more and more their utility in a number of applications for diagnostics or processing materials. Some application are already well established at tabletop X-ray lasers. For some other ones, the standards are not yet permitting it, and either operation at high-end facilities or with incoherent X-ray sources is state-of-the-art. Nevertheless, the work of these authors shows a vision on how materials science or medicine may gain from the availability of laboratory X-ray sources.

The possibility to modify or structure materials with laser pulses with nano-scale accuracy is appreciated. Experimental result from Ishino et al. **[25]** for Si at λ=13.5 nm irradiation shows that there is a condition where the melting threshold becomes the damage threshold. Ablation experiments for Al and SiO_2 samples revealed that damage thresholds induced by the femtosecond and the picosecond soft x-ray lasers are equivalent, so that the picosecond laser ablation will be a good benchmark for the femtosecond laser ablation. The interactions of EUV light with matters along the ablation have potential advantage for the use in advanced materials processing. Tanaka et al. **[26]** have studied the charge states and their energy distributions in the EUV ablation plasma using an E×B mass-charge analyzer. The special interaction regime between extreme UV photons and matter poses challenges on the exact description under a robust physical model. Lolley et al. **[27]** have investigated the absorption coefficients for inverse bremsstrahlung and photo-ionization and the contributions of these processes to EUV absorption under different ionization models- In the case of both ionization models, inverse bremsstrahlung dominates the absorption process above temperatures of about 10 eV.

Artyukov et al. **[28]** have characterized the 3D imaging of porous materials with X-ray illumination, such to optimize the signal-to-noise ratio. In application of mirrors in high-end facilities, B_4C coating mirror deposited by reactive sputtering with nitrogen haven't taken into account. Based on the advantages of reactive sputtering technology, Wu et al. **[29]** investigated the B_4C coatings prepared by reactive sputtering with nitrogen. Billeter et al. **[30]** have investigated the role of surface-chemisorbed hydrogen in catalysts such as TiO_2 with respect to the electronic structure. Using resonant X-ray photons they were able to excite from core level to conduction bands and to defect states. The analysis showed that hydrogen promotes the occurrence of defect traps, while oxygen vacancies suppresses trap state emission. The modification of photocatalytically active materials such as TiO_2 by hydrogen is particularly interesting, because both the optical properties as well as chemical properties change.

Materials science largely relies on probing techniques. Laser-induced background spectroscopy (LIBS) is a well-established fast and semi-quantitative microtechnique. LIBS is limited by large fluctuations which limits its potential to map point-to-point variations in materials. Qu et al. **[31]** have therefore developed LIBS in the soft X-ray, in order to overcome issues with the continuum that covers the UV-visible, and also to collect the data in the pristine plasma.

A Further popular technique is Raman, a photon-hungry technique whose efficiency is predicted to boost at short wavelengths. In that case, ultranarrow lines are needed to access electronic transition shifts away from the fluorescence. While it is a long way to go to implement Raman in a lab setup in the X-ray, Sterzi et al. **[34]** have shown the potential of extreme UV Raman for water screening on the content of pharmaceuticals.

Rush et al. **[33]** have shown the possibility to carry out isotopic imaging with a X-ray laser coupled to a time-of-flight mass spectrometer, for the critical case of uranium isotope screening. Solis-Meza et al. **[16]** has investigated mass spectrometric signals on the same system to retrieve the laser-plasma temperature. Indeed, X-ray lasers have a tremendous potential for home lab spectroscopy, in bridging the gap with the bottlenecked access to beamlines. Jonas et al. **[35]** have shown the investigation of thin organic films offered by a laboratory X-ray absorption fine structure (XAFS) spectrometer for the soft X-ray range. The transmission spectrometer is based on a laser-produced plasma source in combination with a twin-arm reflection zone plate spectrometer. The merits of the spectrometer are demonstrated through the investigation of *poly[(9,9-dioctylfluorenyl-2,7-diyl)-alt-co-(1,4-benzo-{2,1',3}-thiadiazole)]* (F8BT), a poly-fluorene copolymer. Stiel et al. **[36]** have reviewed the topic with care for crucial details.

X-ray Lasers are showing a potential also for biomedical diagnostics. Müller et al. **[37]** have reviewed state-of-the-art X-ray imaging of human brain tissue, highlighting a perspective application for X-ray laser to fully access the molecular level. Osterwalder et al. **[38]** have also highlighted Hard X-ray micro computed tomography can be used for three-dimensional histological phenotyping of zebrafish embryos down to 1micrometer or below without the need for staining or physical slicing. Artyukov et al. **[39]** studied the deposition of sodium in the heart muscle, which has a remarkable impact on the function impairment. They used X-ray absorption and fluorescence microscopy as direct inspection techniques. Such direct measurements represent a milestone to prove the accumulation of sodium in the intercellular spaces.

REFERENCES

[1] Klisnick, A. First seeds of X-ray lasers: a tribute to Pierre Jaeglé, Proc. SPIE 11886, 1188603 (2021)
[2] Nilsen, J., X-ray lasers: The evolution from Star Wars to the table-top, Proc. SPIE 11886, 1188604 (2021)
[3] Tallents, G., A brief history of short wavelength coherent radiation sources, Proc. SPIE 11886, 1188605 (2021)
[4] Rocca, JJ. et al., Demonstration of Multi-Hz repetition rate x-ray lasers at shorter wavelengths, Proc. SPIE 11886, 1188606 (2021)
[5] Y. Hemani, M. Galimberti, D. Bleiner, Empulse: A Compact Terawatt Chirped Pulse Amplification Laser for Generating Coherent X-rays, Proc. SPIE 11886, 1188607 (2021)
[6] J. Nejdl et al. , Update on laser-driven X-ray sources at ELI Beamlines, Proc. SPIE 11886, 1188608 (2021)
[7] B. Hornberger, J. Kasahara, R. Ruth, R. Löwen, Inverse Compton Scattering X-ray Source for Research, Industry and Medical Applications, Proc. SPIE 11886, 1188609 (2021)
[8] U. Chaulagain et al., Tomographic characterization of gas jets for Laser-plasma acceleration with increased sensitivity, Proc. SPIE 11886, 118860A (2021)
[9] O. Guilbaud et al. , Solid-target seeded soft x-ray laser for short pulses and optical vortex amplification, Proc. SPIE 11886, 118860B (2021)
[10] P. Martinez-Gil, J.A. Moreno, J. Alvarez, A. Luque, F. Ribas, M. Ruiz, E. Oliva, 2D-3D multiscale modelling of inhomogeneous plasma amplifiers, Proc. SPIE 11886, 118860C (2021)
[11] Y. Kato, H. Daido, X-ray laser development at the Institute of Laser Engineering, Osaka University - with worldwide collaboration, Proc. SPIE 11886, 118860D (2021)
[12] F. Bencivenga et al., An approach for realizing four-wave-mixing experiments stimulated by two-color extreme ultraviolet pulses, Proc. SPIE 11886, 118860E (2021)
[13] Th.H. Chau, K.B. Dinh, A. Tran, P. Hannaford, L. Van Dao, Four-wave mixing with multi-color laser in extreme ultraviolet region, Proc. SPIE 11886, 118860F (2021)
[14] F. Campi et al., Efficient extreme-ultraviolet multi-band high-order wave mixing in silica Proc. SPIE 11886, 118860G (2021)
[15] T.M. Jeong et al., Ultra-strong attosecond laser focus produced by a relativistic-flying parabolic mirror, Proc. SPIE 11886, 118860H (2021)
[16] F. Vagizov et al., Temporal and Spectral Control of the X-ray Pulses in a Resonant Medium with a Modulated Transition Frequency, Proc. SPIE 11886, 118860I (2021)
[17] G. Begani-Provinciali et al., Phase-Contrast Tomography with X-ray Hartmann wavefront sensor, Proc. SPIE 11886, 118860J (2021)
[18] Y. Li O. De La Rochefoucauld, Ph. Zeitoun, Numerical Investigation of the Effect of Number of Zones On Fresnel Zone Plates Imaging Performance Under Different Coherence Conditions Proc. SPIE 11886, 118860K (2021)
[19] Pandey et al., Single-shot wavefront characterization of high topological charge extreme-ultraviolet vortex, Proc. SPIE 11886, 118860L (2021)
[20] M. Prasciolu et al. On the Use of Multilayer Laue Lenses with X-ray Free Electron Lasers, Proc. SPIE 11886, 118860M (2021)
[21] L. Dakroub et al. Temporal characterization of a plasma-based seeded XUV laser using a laser-dressed photoionization technique, Proc. SPIE 11886, 118860N (2021)
[22] S. Wang et al., Single-shot picosecond resolution Fourier transform holographic microscopy with large field of view using a compact soft X-ray laser, Proc. SPIE 11886, 118860O (2021)
[23] D. Bleiner, M. Ruiz-Lopez, Multiscale Lensless Imaging with a Plasma X-ray Laser, Proc. SPIE 11886, 118860P (2021)
[24] J. VanTilborg et al., Phase-contrast imaging with laser-plasma-accelerator betatron sources, Proc. SPIE 11886, 118860Q (2021)
[25] M. Ishino et al., Short pulse soft x-ray laser ablation research, Proc. SPIE 11886, 118860R (2021)
[26] N. Tanaka et al., Energy and charge distribution of Si ions in EUV ablation plasma, Proc. SPIE 11886, 118860S (2021)
[27] J. Lolley, G.J. Tallents, The role of photo-ionization in extreme ultraviolet ablation interactions, Proc. SPIE 11886, 118860T (2021)
[28] Artyukov, Coherent pore-scale imaging: phase contrast and ptychography, Proc. SPIE 11886, 118860U (2021)

[29] J. Wu et al., Composition analysis of boron carbide coatings prepared by reactive sputtering with nitrogen , Proc. SPIE 11886, 118860V (2021)
[30] E. Billeter et al., Hydrogen induced trap states in TiO2 probed by resonant X-ray photoemission, Proc. SPIE 11886, 118860W (2021)
[31] D. Qu et al., Dual Spectrometer for Simultaneous Visible and Extreme Ultraviolet LIBS, Proc. SPIE 11886, 118860X (2021)
[32] Sterzi et al. Short-wavelength Raman Spectroscopy for in-situ Water Analysis: A Feasibility study, Proc. SPIE 11886, 118860Y (2021)
[33] L. Rush et al., Imaging isotopic content at the nanoscale using extreme ultraviolet laser ablation and ionization mass spectrometry, Proc. SPIE 11886, 118860Z (2021)
[34] E. Solis-Meza et al., Time-of-flight mass spectrometry to determine plasma temperature from ablated target surfaces, Proc. SPIE 11886, 1188610 (2021)
[35] A. Jonas et al., Transient laboratory X-ray absorption fine structure spectroscopy on thin films demonstrated with F8BT, Proc. SPIE 11886, 1188611 (2021)
[36] H. Stiel et al., Ultrafast NEXAFS spectroscopy in the lab using laser-based sources and advanced X-ray optics, Proc. SPIE 11886, 1188612 (2021)
[37] B. Mueller et al, X-ray imaging of human brain tissue down to the molecule level, Proc. SPIE 11886, 1188613 (2021)
[38] M. Osterwalder et al., Three-dimensional Hard X-ray Microtomography of Zebra fish Larvae , Proc. SPIE 11886, 1188614 (2021)
[39] Artyukov et al., Study of sodium depositions in heart interstitium using methods of Xray absorption and fluorescent microscopy, Proc. SPIE 11886, 1188615 (2021)