Ways of development of compact coherent femtosecond X-ray sources for applications in nano- and biophotonics

L. Mikheev¹,²

¹P.N. Lebedev Physical Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia
²National Research Nuclear University “MEPhI” (Moscow Engineering Physics Institute), Kashirskoe sh. 31, 115409 Moscow, Russia

E-mail: mikheev@sci.lebedev.ru

Abstract. Ways of the development of compact coherent sources of soft X-ray femtosecond pulses are discussed, which meet the requirements for the implementation of the “diffraction-before-destruction” approach in the lensless X-ray Coherent Diffractive Imaging (CDI) technique enabling quantitative 3D mapping of material structure with the nanoscale spatial resolution. An innovative hybrid (solid/gas) approach to produce ultra-intense femtosecond laser pulses in the visible is described in the context of its applications for laser driven high harmonic generation (HHG) and soft X-ray generation in laser plasmas due to recombination mechanism of excitation.

1. Introduction

Successful development of new technologies in nano- and bio-photonics strongly depends on the availability of a modern instrumentation for structural analysis. The most powerful and informative tool in the study of matter structure is the lensless X-ray Coherent Diffractive Imaging (CDI) technique enabling quantitative 3D mapping of materials with the nanoscale spatial resolution determined by the wavelength of a radiation source (e.g., [1]). In this technique, a sample is illuminated with a spatially coherent beam of light to obtain far-field diffraction patterns allowing the reconstruction of sample images with a computational phase retrieval algorithm. X-ray CDI has been successfully applied for nanoscale imaging with coherent soft x-ray radiation from synchrotrons, X-ray free-electron lasers (XFELs), laser driven high harmonic generation (HHG) and with a partially coherent compact gas–discharge EUV light source at 17.3 nm in both single and multiple shot modes [1-3].

It is well known that biological structures are highly sensitive to radiation damage. To solve this problem, a “diffraction-before-destruction” approach is used, which enables diffraction signals to be collected from a sample in a single femtosecond-duration pulse of high intensity before the onset of significant radiation damage. Among the above mentioned X-ray and EUV light sources, nowadays only XFELs, providing intense femtosecond pulses in diffraction-limited soft–X-ray beams, meet the requirements for implementation of the “diffraction-before-destruction” method in CDI.

However, despite the clear potential for x-ray microscopy, XFELs have limited availability because of their high cost and the enormous scale of linear electron accelerators of several kilometers in length. This limits the wide spread of ultrafast X-ray CDI technique and makes it relevant to the development
of alternative coherent X-ray sources for tabletop microscopy suitable for applications in academic and industrial laboratories.

One of the greatest challenges is the development of compact coherent X-ray sources radiating in the "water window" between the K absorption edges of carbon (4.37 nm) and oxygen (2.33 nm), where carbon is highly opaque, while water is largely transparent. This spectral range enables the observation of living biological structures in a natural aqueous environment with high contrast ratio and unprecedented spatial resolution.

2. High-order harmonic generation

Recently, intensive efforts have been made to develop such compact soft X-ray sources on the basis of laser driven HHG widely used to generate spatially coherent short wavelength radiation in gases and on the surface of solid targets (e.g., [4-6]). HHG can provide a single burst or a train of attosecond pulses which allow for applying X-ray CDI technique for ultrafast tabletop microscopy. HHG in gases is nowadays a routine operation. However, it has low up-conversion efficiency (typically less than 10^-5) and limited laser intensity scaling above 10^18 W/cm^2 because of phase mismatch due to plasma generation [4, 6]. These features impede HHG in gaseous media to be used for the development of highly intense X-ray sources which would be suitable for the "diffraction-before-destruction" CDI microscopy of biological structures.

Other concept relies on the generation of high-order harmonics by reflecting ultra-intense lasers from a solid surface, which has no limitation on driving laser intensities and promises much higher conversion efficiencies. According to the simplest oscillating plasma mirror model [7] the laser field induces a relativistic oscillation of the overcritical plasma surface, which results in Doppler frequency upshift of the laser light reflected by the dense plasma. This gives rise to the incident frequency harmonics emerged as an attosecond pulse train. HHG driven by a few-cycle laser field produces an isolated attosecond pulse [8], that is required to implement the "diffraction-before-destruction" method in X-ray CDI. Particle-in-cell (PIC) simulations [9] demonstrate a significant enhancement of conversion efficiency up to 20% with shortening driving laser pulse down to single-cycle pulse-width.

Up-conversion efficiency of HHG from solids displays strong driving laser frequency scaling [5, 10] so that shorter wavelength lasers are more efficient in the harmonic generation.

3. Soft X-ray lasers

Only collisional and recombination mechanisms of active media excitation in laser plasmas are of practical interest for the generation of coherent soft X-ray radiation. The first one was realized in laser plasmas with high electron temperature, providing a population inversion on transitions between excited states of ions, which typically lie in the range 10-50 nm. There is only one experimental result showing the X-ray emission at shorter wavelengths of 4.3 and 4.5 nm [11], close to the edge of the "water window". However, pulse energies reached in collisional soft X-ray lasers are too low for CDI applications.

The most promising way to reach desirable energy within the "water window" with soft X-ray lasers lies in the realization of recombination scheme of laser action on transitions to the ground state of recombining fully stripped ions, which are of great interest from the view point of scaling to shorter wavelengths. Optical-field-ionization (OFI) in a strong field of a linearly polarized laser beam is the most effective technique to produce fully stripped plasmas consisting of nuclei and cold electrons in a time much shorter than the time of their recombination. Low temperature of electrons enables their rapid recombination with nuclei resulting in the formation of the population inversion of low-lying levels of ions.

Residual energy of the electrons produced via OFI is proportional to I^2/\lambda, where I and \lambda are the intensity and wavelength of the pump beam, respectively. That is why the first gain observation in the recombination scheme was performed with the use of a KrF-laser [12] at the wavelength of 248 nm. In these experiments, quite a large gain of 20 cm\(^{-1}\) at the Lyman-alpha transition of the H-like LiIII ion (13.5 nm) was achieved. Subsequently, this result was confirmed in several studies performed at the
Berkeley National Laboratory [13] and Princeton University [14]. However, the gain saturation necessary for the efficient energy extraction from the gain medium was not demonstrated. This is due to the fact that the KrF-laser pulse-width lying in the subpicosecond time-domain is too long and leads to overheating electrons in the laser plasma. Pump pulse-widths shorter than 100 fs are unattainable for the KrF-lasers characterized by narrow gain bandwidth (~ 300 cm⁻¹).

Pulse-widths and intensities of the widespread Ti:Sapphire laser systems are much more suitable for the recombination scheme realization. However, the radiation of such systems lies in the near infrared (800 nm) and the electron residual energy turns out to be too high for the recombination scheme implementation. The most attractive for the excitation of lasing on the transitions to the ground state is the second harmonic of a Ti:Sapphire system that allows for increasing gain on these transitions by two orders of magnitude [15, 16]. In particular, the numerical simulation of the gain on the transition to the ground state of CVI H-like ions (3.4 nm falling within the “water window”) shows that gain as high as 180 cm⁻¹ can be attained at an intensity of (0.5-1) × 10¹⁹ W/cm² in a 20-50 fs pulse [16].

Achieving these parameters in a Ti:Sapphire laser system with nonlinear frequency doubling encounters serious technical difficulties in the manufacture of thin nonlinear crystals of large diameter, which have not yet been resolved. Development of hybrid (solid/gas) femtosecond systems in the visible can solve this problem and promises the output parameters, which meet the requirements of recombination X-ray lasers excitation.

### 4. Hybrid (solid/gas) femtosecond systems in the visible

An innovative hybrid (solid/gas) approach to produce ultra-intense femtosecond laser pulses in the visible was proposed at P.N. Lebedev Physical Institute of RAS (LPI, Moscow, Russia) [17] and thereafter it was developed in the course of experimental and theoretical studies carried out in cooperation with the Lasers Plasmas and Photonic Processes (LP3) Laboratory (Marseille, France) and the Institute of High-Current Electronics (IHCE, Tomsk, Russia) (see [18, 19] and references cited therein). This approach is based on original methods, developed at the LPI, of photochemical excitation of active gas media on broadband electron transitions in molecules (XeF(C-A), XeCl and KrF) and on amplification of the second harmonic of a Ti:sapphire femtosecond laser in these active gas media. As a result of these studies, two hybrid femtosecond systems in the visible (475 nm) have been built at the LPI and the IHCE: THL-30 with a design peak power of 5-10 TW and THL-100 designed for 50–100 TW peak power, respectively (figures 1,2). Both of them are based on application of the XeF(C-A) active medium in a boosting amplifier.

![Figure 1. THL-30: Photos of the Ti:Sapphire front-end (a) and XeF(C-A) amplifier (b).](image-url)
Figure 2. THL-100: Photos of the Ti:Sapphier front-end (a) and XeF(C-A) amplifier (b).

Pump technique is based on the conversion of e-beam energy to the VUV radiation of xenon at 172 nm used to excite the XeF(C-A) active medium. Principle of the converter operation is explained in figure 3 for the case of THL-30 system in which four 120 cm long × 15 cm wide radially converging e-beams are injected through 40 μm Ti foils into the converter filled with xenon at pressure of 3 bars. Xenon radiation excited by electrons comes through CaF$_2$ windows to a laser cell housed into the e-beam converter along its axis and filled with XeF$_2$/N$_2$ mixture at 0.25 bar. The XeF(C-A) active medium is excited due to photodissociation of the XeF$_2$ vapor by the 172 nm radiation. Principle of the THL-100 operation is practically the same besides using six radially converging e-beams in the converter and twice higher pump energy. More details on the subject can be found elsewhere [18, 19].

Figure 3. Cross sectional schematic diagram of the XeF(C-A) amplifier: (1) vacuum diode, (2) e-beam converter, (3) photolytic laser cell.

Laser XeF(C-A) transition is centered near 480 nm spectrally matching with the second harmonic of a Ti:Sapphire laser and has very broad bandwidth of $\Delta\lambda=70$ nm corresponding to the transform limited pulse duration of 5 fs. The wide emission continuum for the C–A transition is caused by strongly repulsive nature of the lower A state. The boosting XeF(C-A) amplifier is seeded by 50 fs pulses early down-chirped to 1-2 ps in a prism pair. Seed pulses of 0.1 TW peak power at 475 nm are produced with a solid-state Ti:Sapphire oscillator operating at 950 nm, regenerative and multipass amplifiers, and KDP frequency doubler spectrally matching the front end to the XeF(C–A) transition. Down-chirped pulses amplified in the boosting amplifier are recompressed in bulk fused silica compressor.

In pilot proof-of-principle experiments made in the THL-100 system, 14 TW peak power was obtained in a 50 fs pulse at the output energy of 0.7 J [19]. In the hybrid system THL-30 the output energy of 0.25 J was achieved, which allows a peak power of 5 TW to be obtained after temporal pulse compression to 50 fs [19].
5. Conclusions

Results obtained in the course of the novel hybrid concept studies demonstrate the high potential of the approach relying on the optically driven broadband active media in the visible. The most remarkable achievement of these studies is the record breaking peak power of 14 TW in the visible, which seems to be promising for the development of compact coherent X-ray sources based on the laser driven HHG from solids and recombination mechanism of X-ray generation in laser plasmas.

Acknowledgments

This work was supported by the Competitiveness Program of NRNU MEPhI.

References

[1] Chapman H and Nugent K 2012 Nature Photon. 4 833-9
[2] Seaberg M D et al. 2011 Opt. Express 19 22470–9
[3] Dstrcil M O, Ussmann J B, Udolf D R, Resenitz R B, Jianwei Miao, Brocklesby W S and Juschkin L 2015 Opt. Lett. 40 5574-7
[4] Eden J G 2004 Progr. Quantum Electron. 28 197-246
[5] Teubner U and Gibbon P 2009 Rev. Mod. Phys. 81 445-79
[6] Brabec T and Krausz F 2000 Rev. Mod. Phys. 72 545-91
[7] Lichters R, Meyer-ter-Vehn J and Pukhov A 1996 Phys. Plasmas 3 3425-37
[8] Heissler P 2012 Phys. Rev. Lett. 108, 235003
[9] Ma G, Dallari W, Borot A, Krausz F, Yu W, Tsakiris G D and Veisz L 2015 Phys. Plasmas 22, 033105
[10] Gibbon P 1996 Phys. Rev. Lett. 76 50-3
[11] MacGowan B J et al. 1990 Phys. Rev. Lett. 65 420-3
[12] Nagata Y 1993 Phys. Rev. Lett. 71 3774-7
[13] Donnelly T D et al. 1994 Proc. 4th Int. Conf. on X-Ray Lasers (Williamsburg, VA) (New York: AIP) p. 106
[14] Korobkin D V, Nam C H, Suckewer S and Goltsov A 1996 Phys. Rev. Lett. 77 5206-9
[15] Spence D J and Hooker S M 2005 Opt. Commun. 249 501-13
[16] Avitzour Y and Suckewer S 2007 J. Opt. Soc. Am. B, 24 819-28
[17] Mikheev L D 1992 Laser Part. Beams 10 473-8
[18] Mikheev L D, Tcheremiskine V I, Uteza O P and Sentis M L 2012 Progr. Quantum Electron. 36 98–142A reference
[19] Mikheev L D and Losev V F 2016 Multiterawatt Hybrid (Solid/Gas) Femtosecond Systems in the Visible. In “High Energy and Short Pulse Lasers”, ed. R Viskup (Croatia: InTech) chapter 6 pp 131-161