Paper

New pico- and femtosecond laser based sources: from the Infra-Red to the XUV

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Abstract — In this paper, we review a number of recent developments concerning laser or laser-based sources generating pico- or subpicosecond pulses of light at wavelengths now ranging from the mid Infra-Red (50 µm) to the XUV (11.9 nm). Those include the new generation of „Chirp Pulse Amplification" laser systems, Free Electron Lasers operating in the IR or visible range, and a number of laser driven sources covering the XUV range, generally based on the high intensity irradiation of solid or gaseous targets. The main perspectives, applications and essential issues concerning such sources are discussed.

Keywords* — *lasers, short pulses, ultrafast phenomena.

Short pulse laser systems: the CPA/KLM revolutions

There has been a constant decrease of the available pulse durations ever since the laser existed. Picosecond lasers have been used for more than twenty years now, and subpicosecond lasers (very few of them really deserving the current „femtosecond" laser appellation) for more than ten. The essential moves of these past years have been the huge increase of the energy available per pulse, and a no less considerable spreading of the wavelength range over which short light pulses are available, which now spans from the mid Infra Red (IR: about 50 µm with the best IR Free Electron Lasers) to the XUV where subpicosecond pulses are available down to wavelengths in the 50 nm range, and picosecond X-laser pulses down to 11.9 nm.

One major breakthrough occurred in the late 80's with the invention of the „Chirped Pulse Amplification" (CPA) concept. Indeed the task of extracting the maximal possible energy from an amplifier system requires to work as close as possible to the „saturation fluence" which varies considerably with the type of medium used. As shown in Fig. 1, it is of the order of mJ/cm^2 for dyes, tens of mJ/cm^2 for excimers but can reach beyond the J/cm^2 for solid amplifying materials. An essential problem is then that one has also to stay below the „breakdown threshold" for the material used. This precludes the possibility of working at the saturation fluence in solid-state amplifiers in the pico- or subpicosecond regime. However, the saturation fluence can be reached if one works with pulse durations close to one nanosecond, because of the strong pulse duration dependence of the breakdown fluence. Since one is interested in amplifying short laser pulse, those are always associated with a significant bandwidth (the Fourier transform limitation imposes that $\Delta\omega\tau \geq k$, where $\Delta\omega$ is the bandwidth, τ the pulse duration, k being a parameter whose value depends on the exact pulse shape but is always of the order of 0.5). Any dispersive element, in which the long wavelengths travel generally faster than the short ones, will therefore produce a lengthening of the broadband pulse and result in a so-called „chirped pulse" in which the frequency will vary from „red" to „blue" along the pulse. This dispersive element can be any amount of dispersive material, or more elaborated optical systems (where dispersion is generally provided by sets of prisms or gratings). In the first case, the dispersion is always positive, whereas in the second, depending on the configuration, the dispersion can be either positive of negative, suggesting the idea that a short pulse can be expanded in time and then recompressed : this is the basic idea in CPA [1]. A short pulse (with pico- or subpicosecond duration) is produced in an oscillator, stretched in time up to durations in the nanosecond range, amplified (in conditions where the saturation fluence is reached below the damage threshold) and finally recompressed almost down to its original duration.

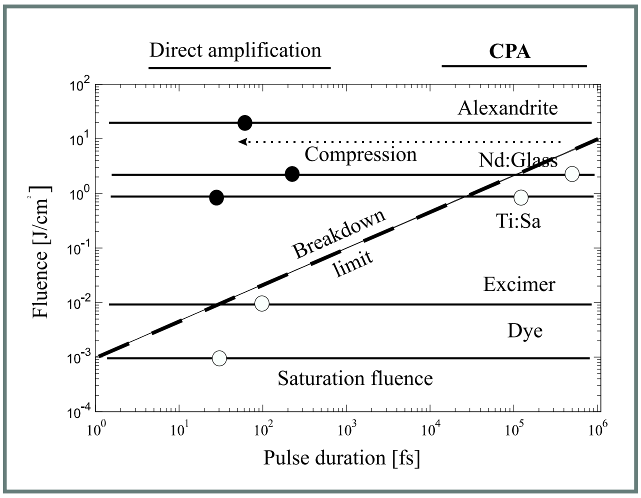

Fig. 1. Saturation fluence and breakdown limit for different materials (*by courtesy of F. Salin*)
An efficient amplification in solid state systems requires to work with pulses of nanosecond duration.

CPA was first applied to Nd:Glass systems and already allowed in the late 80's to reach peak powers in the terawatt range with table-top systems operating at 1064 nm with pulse durations close to one picosecond, opening a wide new field of applications (such peak powers had been so far reserved for giant multibeam laser systems). However the amplifying bandwidth of Nd:Glass was not yet sufficient to obtain „femtosecond" pulses, but using a combination of different glasses in the amplifying system, a number of large size lasers succeeded to produce pulses of duration down to 600 fs, with energies per pulse of the order of tens of joules, and the giant lasers now under construction (NIF in LLL–USA or LMJ in CESTA, Bordeaux, Fr) will most likely be equipped with a „PetaWatt" line allowing to reach laser intensities in excess of 10^{20} W/cm^2. Ultrashort pulses were still the speciality of dye and, to some extent, excimers systems until another revolution completely changed the picture, due to the emergence of a new class of oscillators based on the so called „Kerr Lens Mode-locking". Indeed, Titanium doped sapphire was expected, due to its large fluorescence bandwidth, to be an excellent candidate for the realization of solid-state femtosecond lasers. Moreover, in the process of designing a passively mode-locked oscillator based on this amplifying medium, the researchers discovered a quite unexpected possibility to obtain mode locking without inserting any Q-switching element in the cavity (hence the original appellation of this system: „magic" mode locking). Later on, this was found to be due to nonlinear effects occurring in the amplifying medium [2], hence the final appellation of „Kerr Lens Mode Locking", whose principle is depicted in Fig. 2. The cavity design includes two folding mirrors (symbolized by lenses for clarity on the figure), which focus both the pump and laser beam in the amplifying crystal. In the mode locked regime, the intensity is such in the crystal that nonlinear contributions to the refractive index (Kerr effect) turn the amplifying crystal (a plane parallel slab) into a lens, thus modifying the cavity stability, which is restored by a minor correction of the lenses position. The cavity can so be adjusted so that the best stability is obtained for the mode locked operation, conferring excellent stability to this type of lasers.

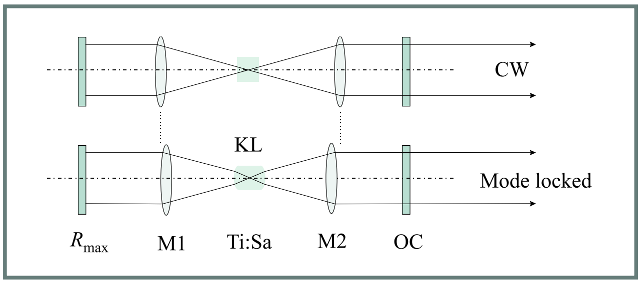

Fig. 2. Principle of the Kerr Lens Mode Locking
The cavity design has been simplified to the elements necessary for the understanding of the mode-locking principle. In the real set-up, lenses M1 and M2 are replaced by spherical mirrors. R_{max}: 100% reflecting mirror, OC : output coupler (3 to 10% transmission), KL : „Kerr Lens", in the mode locked regime only.

The real cavity design of course includes an internal negative dispersion line to compensate for Group Velocity Dispersion, mainly in the amplifying crystal. With such a set-up, pulse durations of the order of 30 fs are routinely achieved with both commercial and home-built systems, with energies per pulse of the order of a few nanojoules, repetition rates in the 80 MHz range, and excellent beam qualities. Because of this, intensities up to 10^{10} W/cm^2 at 800 nm can be obtained at a lens focus, and GW/cm^2 at 400 nm (after frequency doubling). This is more than needed for a number of nonlinear optics applications.

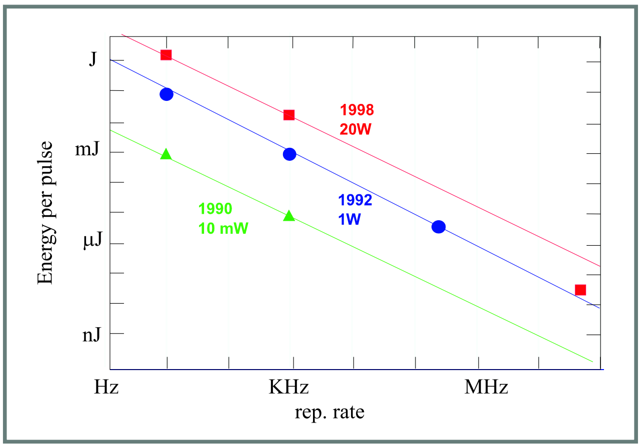

Fig. 3. State of the art concerning amplified Ti:Sa laser systems (*by courtesy of F. Salin*)

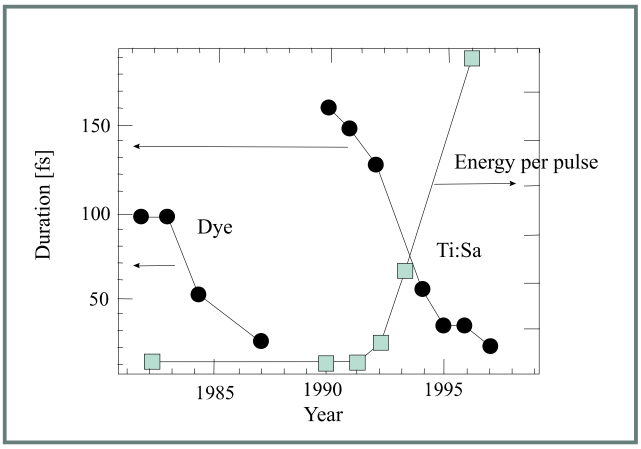

Fig. 4. Passed evolution on pulse duration (circles) and pulse energies (squares) for subpicosecond lasers

Amplification of such pulses using the CPA method in regenerative amplifiers yield energies per pulse of the order of a few millijoules, and can be obtained at repetition rates now reaching the kHz. Beam characteristics are usually quite good (nearly gaussian beams), allowing a number of very high intensity experiments (up to 10^{16} W/cm^2) with quite reasonable scale equipment. The use of extra multipass amplifiers can boost the energy per pulse to 100 mJ, and the peak intensity in the TW range, at repetition rates of about 10 Hz, a considerable progress compared to the Nd:Glass based CPA systems. The recent progresses in this respect are represented in Fig. 3 and 4. It is clear that one now disposes of laboratory scale systems allowing to investigate up to very high intensity effects at repeti-

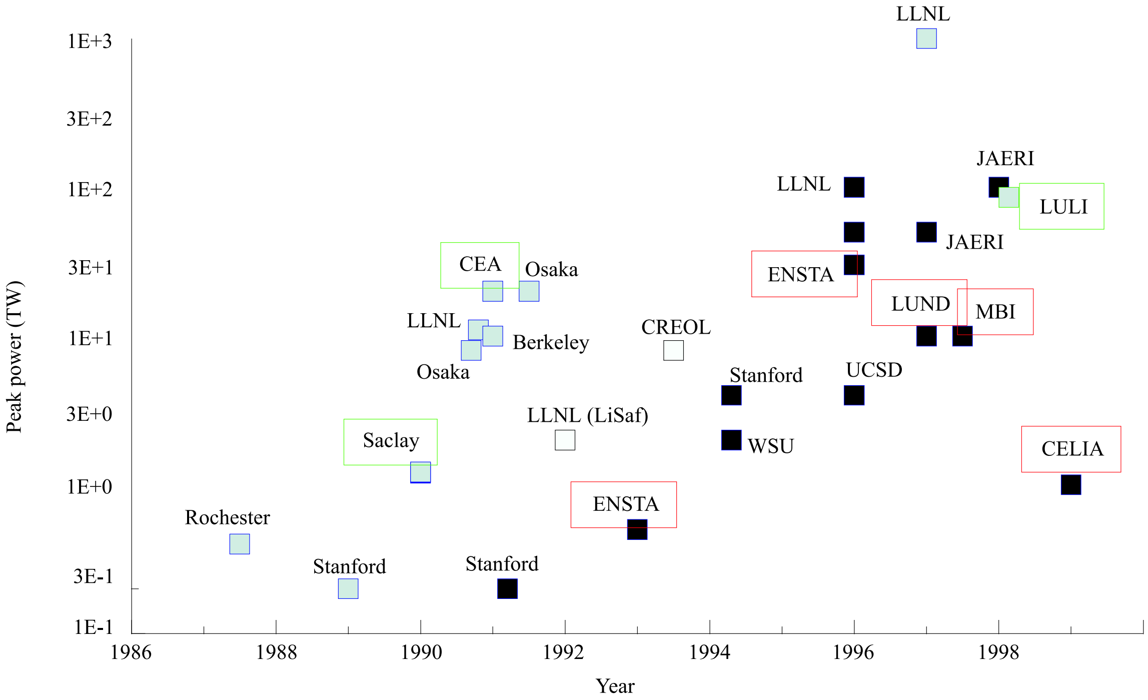

Fig. 5. Past evolution of the peak power of high intensity CPA based laser systems (*by courtesy of C. Leblanc and C. Barty*) In grey: Nd:Glass, Empty: LiSaf, Black: Ti:Sa; European based systems are circled.

tion rates which are orders of magnitudes above those of Nd:Glass based systems (whose rep rate would probably express best in milli or microhertz!). On the other side, Fig. 4 clearly shows the huge impact of both breakthroughs detailed above.

Figure 5 shows a world wide view of the evolution and of the present status in terms of intense lasers.

Note that some world records of a different type are not mentioned in this figure. In particular, concerning the pulse duration, a Ti:Sa cavity design employing „chirped mirrors" (multilayered mirrors reflecting in which different wavelengths have different penetration depths) produced pulses with a 5 fs duration [3], thus setting for such lasers a record that was only reached but very sophisticated dye based systems. Worth mentioning is also the operation of Ti:Sa subpicosecond oscillators using for energy extraction a cavity dumper. This reduces the repetition rate to the 100 kHz - 1 MHz range, but increases the pulse energy by the same amount compared to the classical design (up to µJ per pulse). The stability of such systems is however reduced and they are more complicated to operate.

Tunable short pulse optical sources

Both dye lasers and Ti:Sa, who lead the race towards shorter laser pulses are the prototypes of tunable laser sources because, as explained above, a broad emission band is necessary in both cases. However, the actual tunability of the operational short pulse lasers is quite reduced. In the case of Ti:Sa, it is indeed possible to slightly vary the centre wavelength of the laser (e.g. between 770 and 830 nm), but to the expense of a loss of performances. Concerning the dye systems, short pulse generation required (in the CPM design) the use of a couple of complementary dyes, the operation is essentially restricted to 620 nm. Essentially three solutions were found to achieve a broadband tunability: White Light Continuum (WLC) amplification, frequency mixing and, more recently, Optical Parametric Oscillators/Amplifiers (OPO/OPA) WLC amplification is based on the observation that focusing even a very small amount (microjoules) of energy into e.g. water (but any transparent material works to some extent) allows to generate, through a number of nonlinear optical mechanisms, a more or less broad continuum of white light centred around the pump wavelength, whose pulse duration equals that of the pump pulse. A straightforward solution is then to select a band in this WLC and to amplify it in a set of dye amplifiers. This solution has been used for several years to generate subpicosecond pulses almost throughout the whole visible, with pulse energies in the 100 µJ range in the yellow-red part of the spectrum (pumping with Nd:YAG second harmonic) and 10 µJ in the blue part (pumping with Nd:YAG third harmonic). However, in the latter case, the photostability of the dyes used (contrary to that of those used in the red) is not good, which is at the source of many complications. Likewise, on a general basis, use of dye amplification is at the source of many inconveniences (such as the use of poisonous solvents) so that the search for „all solid state" solutions has been active and recently successful.

If one disposes of both a tunable source in the yellow-red part of the visible spectrum, and of a number of fixed-wavelength sources (Ti:Sa and its different harmonics), it is possible to use sum-frequency generation to obtain tunable short pulse optical pulses at shorter wavelengths. Indeed, WLC amplification, sum frequency generation and harmonic generation combined allow to cover practically all the visible and a significant part of the UV (down to approx. 220 nm), with energies per pulse ranging from a few 100 μJ in the red to about 1 μJ in the UV. However, due to the limitations of the available NL crystals in particular, some holes remain in the tunability curve which can be filled using Optical Parametric Generation. Optical Parametric Generation (OPG) is a process which can be viewed as symmetric of sum frequency generation. In the latter case, the energy of two different photons is added in a nonlinear crystal to generate a photon with the sum energy. In OPG, it is on the contrary the energy of the incident photon (pump beam) which is split in two smaller energy photons. The obtained energies (usually referred to as „signal" and „idler", in a quite arbitrary fashion) are imposed by the necessity of conserving in this operation both the total energy and momentum, and are located on either sides of the „degeneracy wavelength" which is equal to twice the pump wavelength. The phase matching condition generally depends on the optical characteristics of the nonlinear crystal and of the geometry of the optical configuration, and tuning is simply obtained by rotation of the nonlinear crystal.

The signal created in the OPG can be further amplified in an Optical Parametric Amplifier based on the same principle. A filtered WLC can also be used as a seed pulse for amplification in OPA, as represented in Fig. 6, which shows the principle of a subpicosecond OPA system at 1.3 μm. Note that the amplifying stages have to be separated. The reason for this is that since the group velocity of the pump, signal and idler waves are different in the amplifying crystal, they slip off-phase as the thickness increases, an it is then necessary to use a new delay setting before starting the amplification again in a new crystal [4].

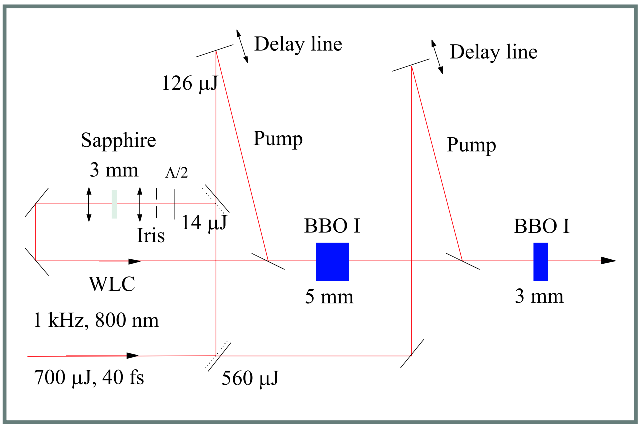

Fig. 6. Layout of a subpicosecond OPA system (*by courtesy of C. Leblanc*)
A white light continuum is generated in a Sapphire plate, and then amplified in a pair of BBO I type OPAs. The characteristics of this system are a pulse duration of 20 fs, a pulse energy of 50 μJ, at a wavelength of 1.3 μm.

A number of OPG/OPA pumped either by Nd:YAG lasers (tens of picosecond of pulse duration) or lately by Ti:Sa lasers, and their harmonics, are presently commercially available, covering the whole visible spectrum on one hand, but also a significant part of the Infra-Red. In the visible, they are clearly competitive compared to dye amplified WLC in the blue part of the spectrum where they are generally preferred, which is not quite the case in the red part of the spectrum, where dyes work best. It seems however that solutions employing different frequency mixing, whose tunable part is provided by a dye system in the red offer better efficiency and stability, but they are not commercially available and require the presence of a team with some expertise in nonlinear optics.

One of the main interest of the OPO is that they have a capability of extending the wavelength range in which short pulses are available to the mid IR (up to 7.5 μm at present, up to 12 μm probably possible), a very interesting region because it is that of vibrational spectroscopy. Up to a recent past this range was covered only by Free Electron Lasers (FEL), a quite specific type of source that we now discuss.

Free Electron Lasers

The principle of the Free Electron Laser (FEL) consists in using as the gain mechanism, instead of the stimulated emission, the synchrotron radiation emitted by free electrons bunches inside a specific alternating magnetic configuration known, in the Synchrotron Radiation language as an „undulator". The permanent magnetic field is sinusoidal in space, and the transverse acceleration of the electrons allows them to radiate along their propagation axis a synchrotron radiation at the „resonance" wavelength λ:

$$\lambda = \frac{\lambda_0}{2\gamma}\left(1+\frac{K^2}{2}\right) \quad \text{with} \quad \mathrm{K} = 0.94\,\lambda_0 \mathrm{B}_0(\mathrm{T})\,, \qquad (1)$$

where γ is the electron (or positron) normalized energy, λ_0 the magnetic field spatial period [cm] and B_0 its magnitude. It is clear from Eq. 1 that this source can be tuned, at a given beam energy by adjusting the magnetic field, and that a large tunability is in principle available also through the choice of the beam energy. The complex description of the field interaction with the particles is outside the scope of this paper (and the author's possibilities as well!), but it is enough to remember that FEL operate just as a standard laser whose gain medium is the relativistic electron beam. The laser gain increases when (i) the interaction length increases, (ii) the beam intensity increases, (iii) the beam energy decreases. Another peculiarity of FEL is that they produce a significant amount of harmonics of the laser.

The need for a relativistic particle beam implies that such lasers are built around particle accelerators, either linear, or as an insertion on a section of a Synchrotron Radiation machine. In all cases they are operated as a Large Scale Facility. However, their interest is such that there are a significant number of machines in operation, most of them as user facilities : about 10 machines in the USA, 5 in Japan and 7 in Europe are either in operation or construction.

For beam energies in the tens of MeV range, laser operation is achieved in the Infra-Red [5]. We have taken as an example the „CLIO" laser facility in Orsay. The laser is built around a „low" energy linear accelerator. It produces pulses with a specific time structure : a series of 1 to 10 µs macropulses with a repetition rate between 1 to 50 Hz, each of such macropulses consisting in a series of a few hundreds of micropulses, whose duration is pico and even subpicosecond (down to 300 fs have been achieved) depending on the cavity settings and of the position in the macropulse (shorter pulses are observed in the build-up of the macropulses). Fig. 7 illustrate the tunability of this laser for different beam energies. It shows that a very large wavelength range (more than one decade) can be obtained with this laser source. Peak powers at the maximum of the tuning curves reaches the 100 MW range, and about 5 MW are still obtained at the extreme wavelengths around 50 µm. On the same plot (in the lower left corner) is included the tuning curve of an IR-OPO pumped by a Nd:YAG laser. It illustrates the point mentioned above concerning the IR capabilities of OPO's. Though not as intense as the IR-FEL, they can still provide intensities in the 10 MW range, and they are by far not so complicated to operate as the LEL, and constitute a very serious alternative for the near IR, and are probably promised to a bright future.

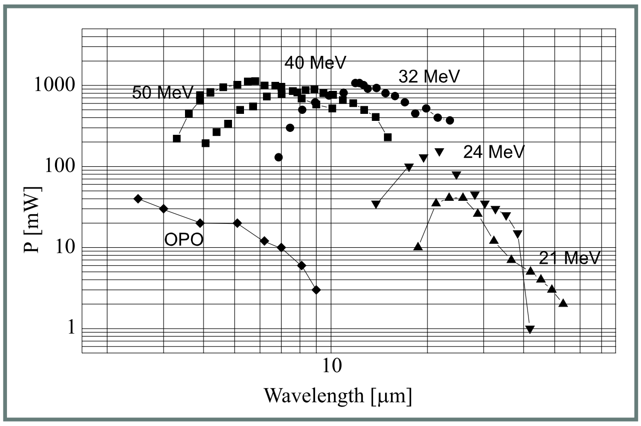

Fig. 7. Tuning curve of the IR-FEL „CLIO" (Orsay, France), as a function of the beam energy (*by courtesy of J. M. Ortega*)
In the lower left corner, the tuning curve of a Nd:YAG pumped OPO is presented for comparison.

Another exciting feature of FELs is their ability to operate simultaneously on two different wavelengths, as demonstrated in Fig. 8 [6]. This operated mode is obtained using two undulators with different spacing. The only principle restriction is that the two wavelengths must be obtainable with the same beam energy.

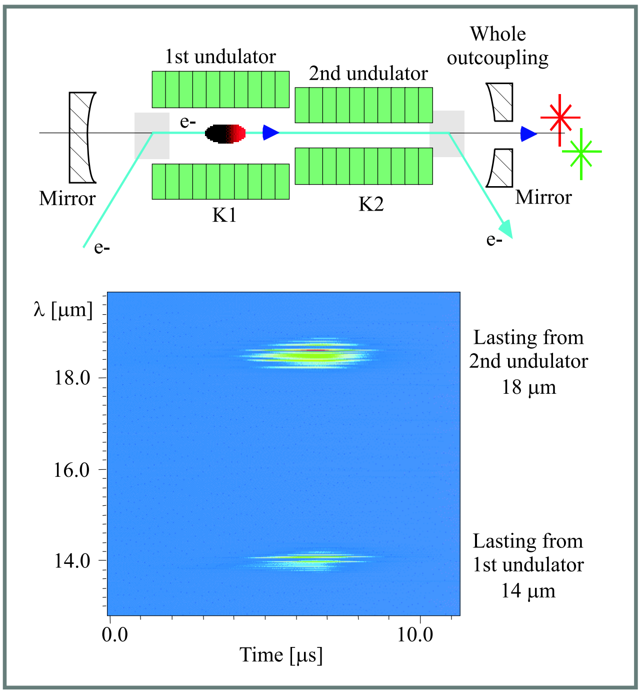

Fig. 8. IR-FEL cavity design, and operation in the „two-colour" mode (*by courtesy of J. M. Ortega*)

FEL can also operate in the visible-UV part of the spectrum [7], for beam energies in the GeV range. Such FEL are usually installed on storage rings. At present, their cost/performance ratio is not so impressive compared to other systems in the same wavelength range, and their essential interest is that they are synchronized by nature with Synchrotron Radiation which allow to consider a number of innovative pump probe experiments. However, the recent appearance of third generation rings should lead to a dramatic improvement of the UV-FEL characteristics. Pulse durations of the order of a few picoseconds should be available, and the peak power is expected to reach beyond the MW. Intensities of the order of 10 TW/cm^2 can reasonably be expected, at a repetition rate of tens of MHz. Besides, an increased harmonic content is expected, so that using the fifth harmonic of the laser, intensities close to the MW/cm^2 should be obtained for photon energies of 20 to 80 eV so that, given the high repetition rate, it should be possible to observe nonlinear effects in the XUV wavelength range. This has been one of the aims of the recent development of intense XUV sources, that we now comment.

Short–pulse, high–intensity XUV sources

The important progresses discussed above on the lasers gave birth to a number of schemes for producing high intensity, short pulses of XUV light through the interaction of high power laser pulses with matter in its different states. Attempts to obtain population inversion between states of highly ionised atoms, as they can be obtained in the plasmas generated in the interaction of high energy (hundreds of joules) laser pulses with metals have an already long story. A number of schemes have been proposed to realise this way a „X-ray laser". In particular, several successes were obtained using the „collisional pumping" scheme, particularly in the case of a number of Ni-like and Ne-like metal ions. Among the lasing transitions observed, one finds Ne-like Fe at 21.2 and 25.5 nm [8], Ti at 32.6 [9] and Ge at 19.6 nm [10], and also, more recently, Ni-like Ag at 13.9 nm and Sn at 11.9 nm. Some laser schemes using a half cavity to increase the gain length have been realised, and some of these laser are presently operated as user facility providing pulses for applications. The essential difficulty in achieving such a goal was that the gain medium is essentially very inhomogeneous and nonstationary, since it is composed of an expanding plasma in front of the metallic target, so that it is necessary first to optimise the plasma conditions through the definition of a sequence of pumping pulses in order to control both the density gradient in the plasma and the electron temperature, which requires a significant amount of know-how. Such X-ray lasers are generally pumped by multibeam giant lasers with energies per (nanosecond) pulse in the 10 to 100's of Joules. They deliver pulses with durations of tens of picoseconds, and are presently the most intense X-Ray sources. In a recent past, the possibility of obtaining very high transient gains by pico or subpicosecond heating of a preformed plasma has resulted in the first observations of very short X-ray laser pulses (whose duration, though not yet precisely measured is most likely of the order or below 1 ps).

In the X-ray laser case, as already mentioned, the laser induced plasma which is the emitting medium has been allowed to expand, which means that its density, as well as its temperature have significantly decreased. With the appearance of subpicosecond lasers able to produce intensities in excess of 10^{15} W/cm^2, one obtained also the possibility of creating very hot (temperatures of tens of keV) and dense (solid density) plasmas since the dense matter can be heated in a time much shorter than the „hydrodynamical time constants". This was at the origin of a number of new sources of hard X-ray radiation (keV and more) [11]. The emission spectrum strongly depends on the nature of the metallic target, and can be dominated by the K lines of the used element (in the case of light species, e.g. : Al), but can also contain a significant continuous contribution when heavy (e.g.: Au) elements, in which Bremsstrahlung dominates the emission mechanisms, are used. Let us note that despite a high number of emitted photons per pulse, the emission occurs in 2π steradians, so that it is not easy to obtain high intensities on target with the help of such sources, contrary to the case of X ray lasers whose spatial coherence is much better.

Finally, in this category of laser-driven plasma sources, one should mention an already ancient observation [12] that interaction of moderately intense picosecond laser pulses (in the TW/cm^2 intensity range) with metal surfaces can produce hard X-ray radiation when the surface is placed under high cw polarization (MV/cm) so as to block the electron emission. Despite an only very limited characterisation, its seems that under such conditions, intense emission extending into the 10's of keV range is observed [13]. There is so far no interpretation of this effect, but it could clearly constitute an easy alternative to the use of very sophisticated laser sources (the necessary conditions can be achieved with a moderate size table top system) to produce high energy X ray.

Besides these laser-driven plasma sources, there is another important class of XUV sources based on high order harmonic generation of intense laser pulses [14]. Their appearance dates from more than ten years ago, at that time using Nd:Glass lasers as a pump source. They have also immensely benefited from the progresses in laser sources, and can now be found in almost every laboratory operating a high intensity Ti:Sa system. They are generally based on high order (up to 150 or more) harmonic generation in high density jets of rare gases. They present a very characteristic spectrum (Fig. 9): after a rapid decrease for the first few orders, the harmonic yield stabilizes through a region known as the „plateau", up to a cut-off frequency. Both the extension of the plateau as well as the yield depend on the type or rare gas used : the lighter elements provide the highest harmonics, but a better yield is obtained using heavier elements. In the general case, because of symmetry reasons, only the odd harmonics are produced. However, by adding fundamental and second harmonic frequencies [15], some even harmonics can be obtained (top of Fig. 9).

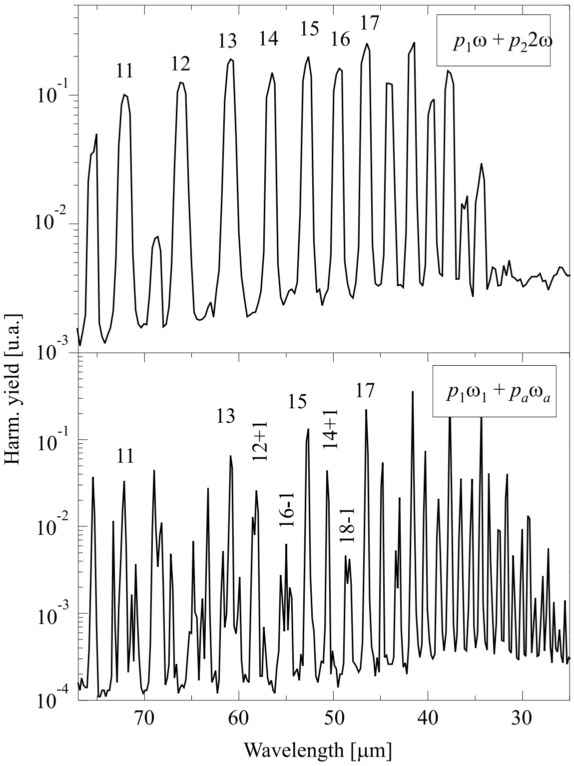

Fig. 9. High order harmonic spectra of the Ti:Sa laser obtained in Argon: top: fundamental and second harmonic of the pump laser are added to obtain even harmonics; bottom: the Ti:Sa fundamental is added to an OPA to obtain fine tunability (*by courtesy of B. Carré and F. Salieres*)

The number of harmonic photons in a given harmonic is of course the result of a combination of the single atom response and of the phase matching conditions in the pumping laser focal region. It is now well established that the important phase slips resulting from ionisation of the gas in the focal region limits the useable intensity range to less than the saturation intensity for a given element. Since the intensity also determines the cut-off frequency (ω_{co}), following a law of the type : $h\omega_{co} = I.P. + nU_p$, where $I.P.$ is the element ionisation potential, n a model dependent quantity usually between 2 and 3 and U_p the light „ponderomotive potential" (proportional to the intensity), it is easy to understand its increase for lighter element which are harder to ionise. Otherwise, the overall characteristics of the source depend on the focusing conditions, but they are now rather well under control, and high order harmonic sources are now reliable and can be used for applications. let us note in particular that their pulse duration has been measured using cross-correlation experiments with subpicosecond laser pulses, and they were shown to be even shorter in time than the pumping laser pulse (which is expected due to the nonlinear character of the interaction), so that they represent to date the shortest type of XUV pulses (40 fs has already been obtained, and 20 fs is expected for a near future. Their tunability is large and can be quite precise. It is first possible to select a given harmonic either with use of a spectrometer (which still causes an appreciable amount of lengthening of the pulse) or even using a selective multilayer mirror (which conserves the pulse duration, but is not by far as selective). Fine tuning of the harmonic frequency can even be obtained (Fig. 9) by combining in the generation processes photons of fixed frequency (usually a Ti:Sa laser) and tunable ones (from an OPA system) [15]. Any even combination of frequency can be obtained this way. Note in Fig. 9 that despite the fact that the pulse energy in the OPA system was quite small (about 50 μJ only) this does not affect the overall harmonic yield on combined-frequency peaks which are less than one order of magnitude below the pure laser harmonics ones.

It is instructing to compare the characteristics of these different sources, since they operate on a quite broad variety of mechanisms (Fig. 10). It should be noted that the result of the comparison may apparently depend on the quantity used to qualify a source. Here, we use the instantaneous brilliance, since it is the useful characteristics for nonlinear application: a source is efficient if it is energetic, point-like, delivers short pulses and has a low divergence. We note that in this respect, all the sources we have discussed are very competitive, even compared with future third generation undulators. However, if one cares about average power (as in linear applications), the picture would be completely reversed. In this respect, it is clear that FEL on third generation rings, operating at the five harmonics will represent a very competitive solution when it will be available, if they hold their promises, even if their pulse duration is not expected to fall down to what is now obtained with harmonics.

Applications

Nonlinear optics is of course deeply involved in the physics underlying the production of short intense pulses through the whole wavelength range discussed above. This is quite obvious in the case of OPO/OPAs, Kerr Lens Mode Locking or High Order Harmonics. It is also true in the case of plasma based sources since the plasma ionisation and heating mechanisms are in general highly nonlinear (multiphoton ionisation or resonant absorption come into play). We will not discussed here these aspects. Applications both concern the linear and nonlinear domain. We will discuss here three examples pertaining to the infra-red, the visible and the XUV range.

Infrared vibrational spectroscopy is one of the most important tools of physico-chemistry to characterize the chemical bonds in even complicated compounds (polymers for instance). On the other hand, the physico-chemistry of interfaces has become increasingly important with the rapid development of thin layers science, with applications to electrochemistry or catalysis for instance. One therefore needs methods for characterising the interfaces which are frequently buried under some amount of bulk material. In this respect, electronic spectroscopies (such as XPS)

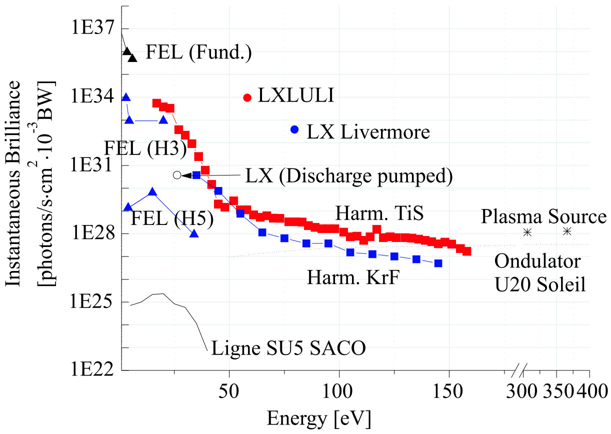

Fig. 10. Comparison of the instantaneous brilliance of different XUV sources (*by courtesy of P. D'Oliveira*)
Typical repetition rates for (LX): a few shots per hour Harm.: 10–20 Hz, FEL and Synchrotron radiation operate at MHz rep rates, but represent extrapolation to future third generation rings (except SACO).

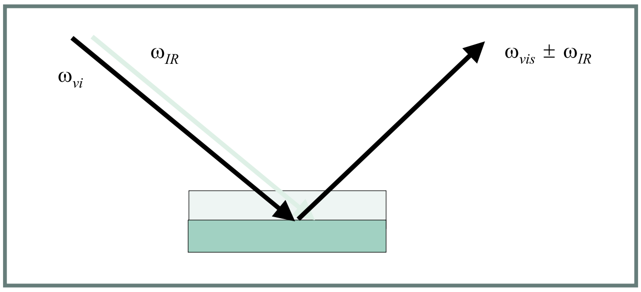

Fig. 11. Principle of the Sum (or Difference) Frequency Generation at interfaces

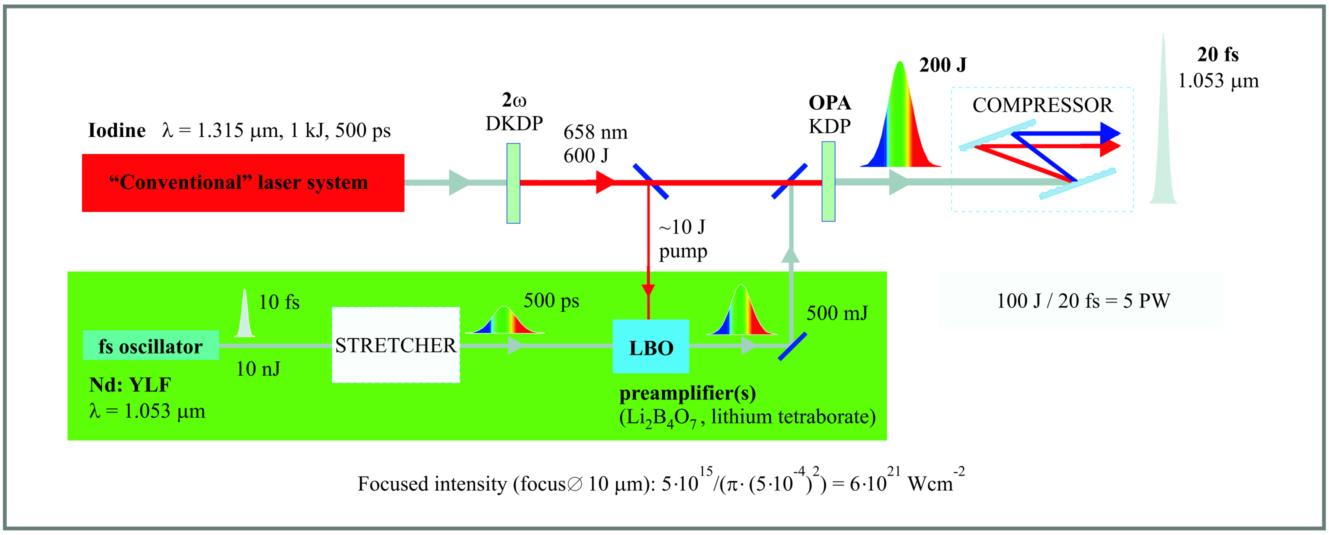

Fig. 12. Proposed scheme „Optical Prarametric Chirped Pulse Amplification" of an iodine laser at PALS (Prague Asterix Laser System) (*by courtesy of Bedrich Rus*)

rapidly reach their limits because the interface shift of a core level can sometimes be very small, and is rapidly masked by the increasing contribution of bulk material. In this respect Sum and Difference Frequency Generation (SFG/DFG)[16], whose principle is shown in Fig. 11, are of paramount importance since they are interface-selective. Indeed sum frequency generation, just as second harmonic generation, is forbidden in the case of centro-symmetric media, which is almost always the case of the overlaying material so that only the interface (where centro-symmetry is broken) contributes to the signal. By tuning the wavelength of the IR radiation, it is then possible to study the Infra Red spectroscopy of the interface. More sophisticated applications can be considered whith ultrashort pulses since this methods gives access to the dynamics of the interface vibrations (measure of the coherence times). An essential advantage of this method, besides its interface selectivity, is the fact that the measurements are made in the visible, where the detectors are much more efficient. SFG/DFG has become one of the most important applications of short intense IR pulse generated by OPOs and FELs.

We have mentioned the important applications of nonlinear optics to parametric generation, it can also be used in combination with CPA to improve the performances of laser systems which do not possess the short pulse capacity because of their limited bandwidth. This is for instance the case of Iodine lasers which can deliver high energies, but have an emission width of less than 2 Å. Figure 12 presents a proposed scheme for producing short pulses with help of an iodine laser, whose implementation is proposed on PALS (for Prague Asterix Laser System, a facility under construction in order to move the MPI-Garching Iodine laser). This scheme combines Optical parametric Amplification and Chirped Pulse Amplification, hence his appellation of OPCPA.

In this scheme, the iodine laser is used to amplify in nonlinear crystals an Nd:Ylf oscillator whose spectral width has been increased through self-phase modulation (another widely used nonlinear mechanism in the production of short laser pulses), so as to be able to produce 10 fs pulses, and then stretched to 500 ps to match the Iodine laser pulse duration. Amplification is then realised in standard OPA with pumping by the iodine second harmonic. The pulse is then recompressed so as to obtain 20 fs pulses of 100 J energy.

Finally, we would like to finish with a linear application in development on XUV sources, that is to say X-Ray Wavefront Division Interferometry applied to surface imaging [17]. In Wavefront Division Interferometry, spatial coherence is of paramount importance since it is different zones of a single input beam which are made to interfere through the use of a Fresnel bi-mirror (instead of the more traditional interferometry where two coherent identical beams are interfering, generated with use of beam splitters which do not exist in the XUV). The principle of this experiment is shown in Fig. 13. impinges on a Nb mirror whose left half is placed in front of an anode biased to an adjustable high voltage (so as to submit the Nb surface to fields in the 10 s of MV/m range). Then the beam is reflected by a Fresnel bi-mirror so as to make the part of the beam reflected on the biased area of the Nb mirror to interfere with the other part (reference wave). The bias voltage is progressively increased between the laser shots until changes in the interferograms, signalling the appearance of a field-induced surface defect are visible. This surface defect will upon further increase of the bias voltage cause a breakdown of the surface. This type of experiments can be of paramount importance to understand the basic effects at the origin of the gain limitation in superconducting cavities for future accelerator. Wavefront division interferometry requires highly spatially coherent intense sources. Originally demonstrated in the XUV range on synchrotron radiation, it has now been implemented on both X ray lasers and high order harmonic sources. One can thus consider high

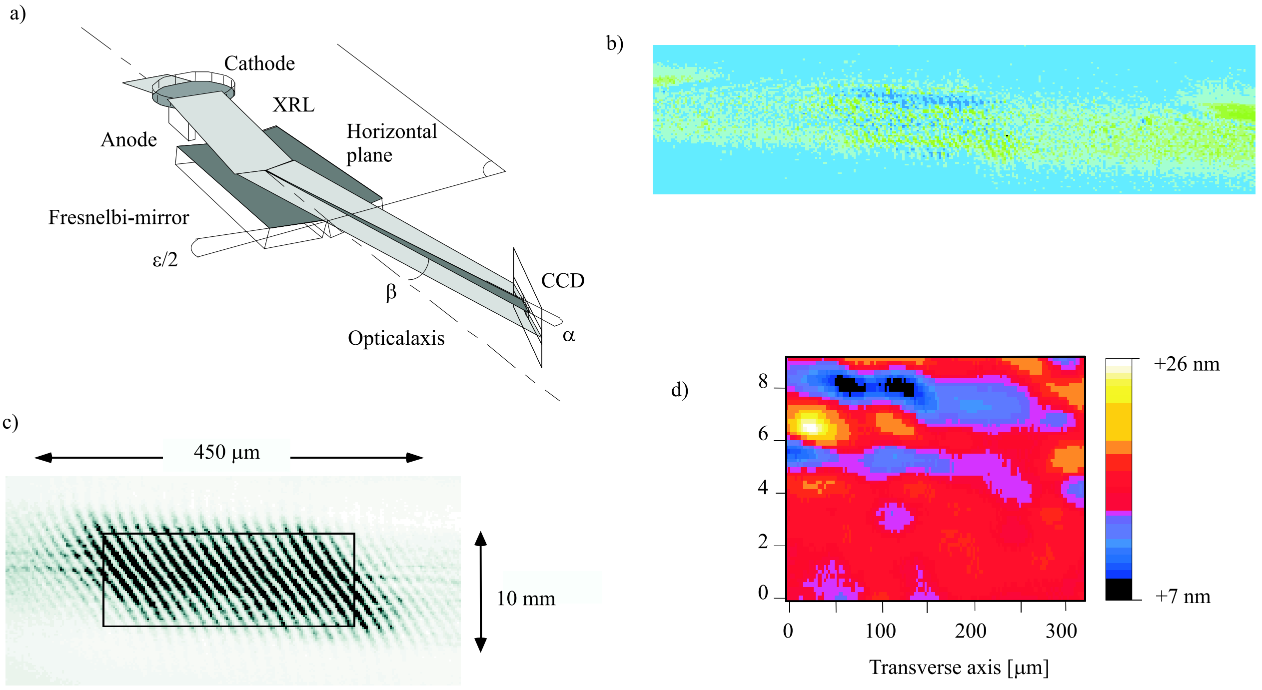

Fig. 13. Wavefront-division interferometric imaging of field-induced surface defects with X-ray lasers (*by courtesy of F. Albert*): a – eksperimental set up; b – interferogram substraction between 14 MV/m and 0 MV/m (reference); c – example of experimental interferogram; d – height variation of the surface between 27 and 37 MV/m obtained by phase reconstruction.

time resolution pump-probe types of experiments using this time-resolved imaging technique, with applications to fast optically driven processes such as laser breakdown for instance.

Other applications of short pulse intense XUV sources include for instance time-resolved photelectron spectroscopies, and of course X-ray microlithography which can be of extreme importance as a future technology for submicron integrated electronics. Let us also note that mixed nonlinear processes involving one XUV photon and one laser photon have already been observed (laser-assisted Auger decay [18]), but that so far there is no experimental evidence of a purely XUV nonlinear process.

Perspectives and problems to be solved

As explained above, there has been huge progresses in the recent past on all the fronts concerning the production and the use of short and intense optical pulses. However, some progresses can still be expected in the near future, not all of them requiring technological breakthroughs.

Some prospective aspects mentioned above are of course linked to progresses in other domains such as for instance FELs which requires the construction of third generation rings (with some contradictory aspects since the FEL prefers low beam energies, which is generally not the case of other synchrotron radiation users).

Concerning the lasers in the optical and near IR domains, progresses are still ongoing. One should not expect a significant decrease of the available pulse duration (already down to 20 fs for high intensity Ti:Sa systems). Gain narrowing due to the amplification, as well as the extreme precision required in the design and alignment of the compressor (geometrical aberrations already have to be compensated up to the fifth order) seriously precludes such a possibility. Moreover, even keeping the pulse duration at this level through the optical system required by the application is already hard enough, and going further could make the experiment simply intractable. The essential progresses are expected on the repetition rates. Most of today's systems are operated at 10 or 20 Hz, but KHz systems with outputs in the mJ/pulse range are already available. High intensity kHz systems under construction now should deliver energies in the 20 mJ/pulse range. Progresses along this line are essentially expected to stem from progresses in diode pumped Neodymium lasers. Of course, high intensity diodes able to efficiently pump directly the Ti:Sa would be in this respect a major improvement, but they are not in view yet.

Any progress in terms of driving laser readily transpose to progresses in the derived XUV sources. Indeed, the X-ray laser greatly suffers from its low repetition rate (limited to a few shots per hour so far). Improvement can be expected from ongoing projects of diode pumped high energy Neodymium lasers (such as the Mercury project in

LLL), but also from an improvement of the X-ray laser's pumping scheme. In this respect, short pulse operation using transient pumping is obtained at pump level below 10 J per pulse, which is a significant progress. Concerning the X-ray lasers, it is also important to make the new experimental schemes available for application experiments, which is not a simple matter since X-ray lasers are still pumped by „giant" laser systems which are in very small number. Concerning the harmonic generation, reaching the kHz repetition rate is possible, but so far impeded by simple technical problems as the pumping speed in the vacuum system they require. Recent experiments based on guided propagation of the pump laser together with the harmonics in hollow fibres seem to offer an effective alternative to production in gas jets, but the important question of phase matching in such conditions have to be revisited to obtain an optimized efficiency.

An extremely serious bottleneck concerning the wide use of XUV radiation (which is common to all sources of this type) is the limited performances of X-ray optics. Except for a very limited wavelength range (around 14 nm) where a good solution exists (B-Si), multilayer mirrors achieve at best a 25% reflection efficiency, and the limitation of the number of useable layers precludes a good wavelength selectivity. Problems to be solved concern the choice of materials, as well as the roughness of the interfaces. This is an extremely serious problem since applications concerning BECU-size programs (such as microlithography in the USA) may turn out to be useless if it is not solved. Another problem concerns harmonic's selection with use of gratings. As mentioned above, using a single grating stretches the pulse duration to several picosecond, but it should be in principle possible to use a double grating system, in the same way it is used in the optical domain for pulse compression. We are only aware of unsuccessful attempts in this area, for reasons which are not quite clear.

Finally, the nonlinear crystals used for both low order harmonic generation and OPO/OPA have reached a satisfactory level. Their operation in this context is essentially limited by group velocity dispersion which precludes the use of long crystals, and thus limits their overall efficiency. This is especially true in the UV. It is clear that any improvements on this side would greatly benefit to laser systems.

Likewise, OPAs in the IR are already very exciting sources, but broadband tunability is here of paramount importance since the essential application concern vibrational spectroscopy. The 1–10 μm wavelength range achieved with Nd pumped OPAs, with hopes to extend it to more than 12 μm in a near future, if it could be achieved with Ti:Sa pumped systems would represent a major improvement since more than one order of magnitude would be gained in terms of pulse duration.

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