New opportunities with intense ultra-short pulse lasers

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ABSTRACT

We review the motivations for the rapid progress in ultra-high intensity, short-pulse lasers. The advantages, advances and limitations of current laser approaches are discussed. We also appraise the potential impact on this field of new laser materials and advanced laser engineering. Some of the new sciences and applications which will benefit from future advances in this field are identified.

1. Introduction.

The interaction of intense laser light with matter has always been a rich field of investigation. Within the last few years one of the most exciting parts of this field has been the interaction of very short laser pulses with matter. From a theoretical standpoint this covers a very broad territory ranging from the behaviour of single atoms in intense laser fields to the interaction of very high-power lasers with dense plasma. When the interacting laser pulse is extremely short, the dominant physical processes occur for only a small number of optical cycles. This permits, in principle, the isolation of high intensity and high field phenomena from other processes that occur on a longer timescale. This field of study has in large measure been driven by the pace of advances in short pulse laser technology. With each advance in focused laser intensity has come the observation of new physical phenomena. In the following, we assess the progress of development of high intensity lasers. In section 2 we identify some the prominent physics issues which will be the focus of attention in the future. We review in section 3 the laser technologies which have lead the advance in progress up to now. Emphasis is given to examining the physical foundations and the technical limitations of extending these technologies to higher intensities. Finally in section 4 we look at some new laser systems and techniques which might impact the direction and of future developments.

2. Some future topics of study with high intensity lasers.

The access to some new sciences with ultra-short lasers is limited either by the maximum available focused intensity, or by the maximum energy deliverable in a short time duration. Currently the highest focused powers are in the range of 10^{19}W/cm^2.[1-3] The highest energies generated in pulses of duration ~ 1ps is in the 10's of joules.[4-6] However most high-intensity phenomena studied so far have been at significantly lower focused intensities and energies than these. As laser developments press the limits in maximum focused intensity and energy deposition, some major new fields of physics come within range of experimental investigation.

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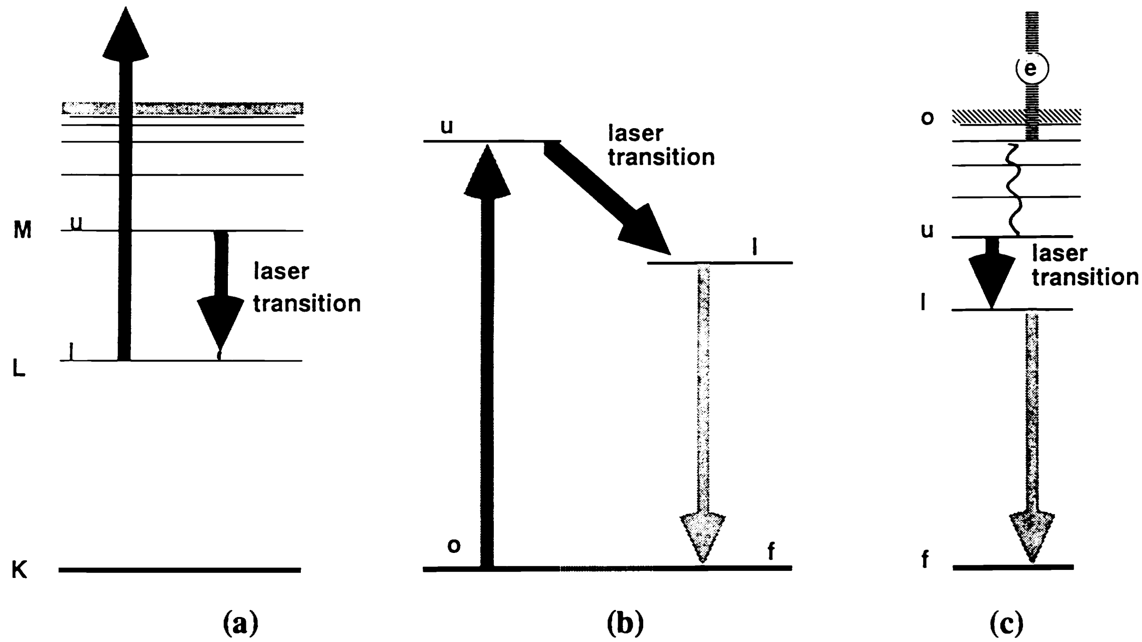

fig.2.1. Primary approaches to ultra-short pulse x-ray lasers, (a) photoionization, (b) photo-excitation, and (c) electron collisional recombination.

2.1. X-ray lasers.

It has long been the hope that the development of efficient picosecond or sub-picosecond lasers would lead to the development of laboratory x-ray lasers. Many proposed x-ray laser schemes depended upon ultra-fast energy deposition along a linear target.

Much of the early interest in x-ray lasers was based on schemes depending on the rapid photoionization of inner-shell transitions, such as the 3s-2p transition in NaII,[7] of the type shown in fig.2.1(a)., or of the 2s electrons in NeI.[8] Although early attempts to demonstrate these systems were unsuccessful, the improvements in laser output energy, and the reduction of the pulse duration of ultra-short lasers to values less than the characteristic de-excitation times of these systems now provides renewed impetus to their investigation.

There is now also increasing interest in schemes involving various forms of selective photo-excitation.[9] Conceptually, fig.2.1(b), this is the x-ray analog of the optically-pumped visible laser. Although in principle this is a self-replenishing scheme because the initial and final states are the same, in order to generate the high gains required for practical laser conditions, very high lasing ion densities are required. The latter implies high photon densities in narrow bandwidths well-matched to the absorbing transitions.[10] Numerous line matches of potential Li-like and H-like pump transitions with possible absorption lines have been identified.[9] Improvements in high-power energy deposition and in the design of efficient energy coupling geometries make selective photo-excitation more promising for the future.

Recombination as a lasing mechanism in a collisionally heated plasma, fig.2.1(c),[11] has been the subject of intense study. Several detailed experimental investigations of this approach have been made British and Japanese groups . One of the latest results has been the observation of gain on the Bα-line of H-like Na.[12] Recombination lasers should be more efficient for shorter pulse target irradiation.

Those that depend on cooling by cylindrical adiabatic expansion, are governed by a characteristic plasma expansion time, t_p, of the form,

$$t_p \sim 5 \times 10^{-7} Z^{-4} \quad s$$

where Z is the nuclear charge. Thus to achieve gain at shorter x-ray wavelengths, (the x-ray laser wavelength scales as Z^{-2} for hydrogenic transitions for example) shorter deposition times are required. Also, as the electron temperature $T_e \sim Z^2$ the laser intensity, I_0, must be increased by $I_0 \sim Z^4$ for shorter wavelength x-ray lasers.[13]

2.2. High density plasmas.

Ultra-short(<1ps) intense laser pulses will interact with solid targets in a distinctly different manner to longer laser pulses. The interaction occurs in a time shorter than hydrodynamics expansion times, the absorbed energy being deposited within a plasma skin depth. This can be as small as 10nm.[14] High absorption[15] can lead to the production of highly ionised, high-density plasmas. At extremely high intensities, relativistic laboratory plasmas are predicted to occur.[16] A number of absorption measurements have been made with ultra-short laser pulses[17-19] and x-ray emission from highly ionised species in the plasma observed.[20,21] This regime poses many interesting questions relating to the coupling and transport of the laser radiation in the surface plasma. The plasma is itself interesting in that, being only a few skin depths thick, it will be optically thin, despite being at plasma densities close to solid density. Many features of the radiation transport and emission remain to be addressed. Experimentally, several challenges must be met. Laser pulses with extremely high contrast ratios must be generated. Since plasma is generated from a surface at intensities of $\sim 10^{10}$W/cm^2, pulse contrast ratios exceeding 10^8 may be required to ensure that low temperature, long scalelength plasma is not created on the target by laser pre-emission, prior to the incidence of the intense pulse. In addition, delineation of the processes occurring in the plasma may be difficult without some high time- and space-descriminating instrumentation.

2.3. Ultra-short x-ray generation.

High-density plasmas created by ultra-short laser pulses can be sources of ultra-short x-rays. A number of potential applications exist for short bursts of picosecond or sub-picosecond x-ray radiation. These range from fast fluorescence studies, x-ray scattering, to ultra-fast radiography. Picosecond x-ray emission from these plasmas has been observed,[22,23] and special targets have been devised for the production of x-ray pulses of even shorter duration.[24]

2.4. Photon-electron interactions.

The prospect of generating extremely high-power densities, in the range of 10^{20}W/cm^2, with electric field strengths of 10^{11}V/cm, gives optimism that fundamental phenomena in nonlinear quantum electrodynamics can now be observed. A subject of intense interest since soon after the invention of the laser,[25] many effects are predicted to occur in the interaction of intense photon beams with free electrons. These include multiphoton effects, non-linear Thomson scattering,[26] beat production,[27] Compton scattering[28] and electron-positron pair production.[29] As progress occurs in the development of intense lasers in the next few years, attempts to observe some of these effects will be made.

3 Recent progress in the development of intense lasers.

High-intensity short-pulse lasers have in the past been developed along three different paths, depending on the type of laser media used. These three types can be categorised as (i) femtosecond dye laser systems, (ii) excimer gas laser systems and (iii) solid-state laser systems incorporating chirped pulse amplification(CPA). Each approach has its own advantages and disadvantages. In the following we briefly review these approaches, emphasising their inherent advantages, and identifying the limitations which may inhibit their development to still higher laser intensities.

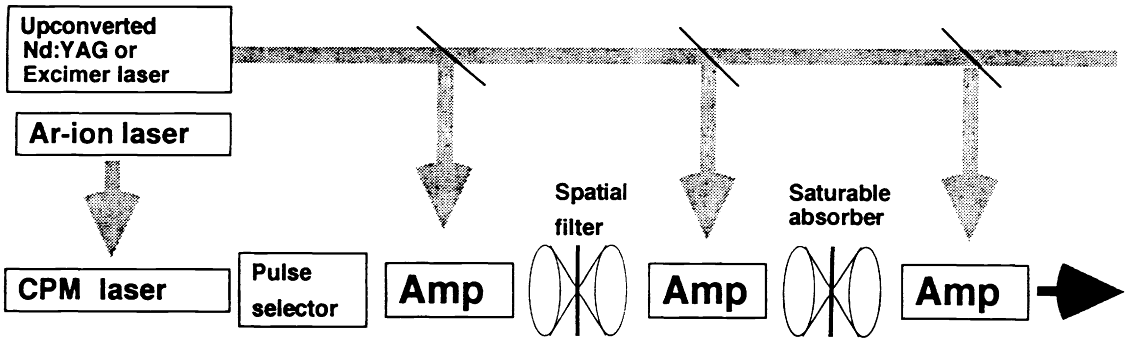

fig 3.1. Typical sytem configuration of a high intensity femtosecond dye laser.

3.1. High-power femtosecond dye lasers.

High-intensity dye lasers provide the shortest available pulses to date with wavelengths in the visible range. Depending on the pumping conditions, they also tend to have the highest repetition rates at the present time, typically >20Hz. This type of laser is particularly useful for experiments in which some form of signal averaging is required.

The inherent broad spectral bandwidth of many dye lasers[30] and the use of c.w. mode-locking techniques such as colliding pulse mode-locking(CPM),[31] have resulted in the generation of the shortest laser pulses to this date(~6ps). Single pulses derived from mode-locked oscillators have been amplified in coherently pumped MOPA(master oscillator-power amplifier) system architectures of the type shown in fig.3.1. A number of such systems have been built.[32-34] The high small-signal gain of dye laser media, such as Rhodamine 6G, can be used to amplify low energy (<1nJ) pulses selected from the output of a CPM oscillator, in a series of flowing dye cell amplifiers progressively larger in aperture. The most effective pumping of these amplifiers is with the use of upconverted Nd: YAG lasers or excimer lasers (flashlamp-pumping of dye laser amplifiers has many disadvantages). This, however increases the system complexity. Moreover, the maximum output energy is ultimately limited by the aperture of the largest dye cell that can be effectively pumped. This aperture must be larger than that required for the amplified laser pulse to remain below the saturation fluence . If this condition is not maintained, then ASE(amplified spontaneous emission) within the amplifier chain will preferentially build up to provide significant pre- and post-pulse laser emission. The latter can be inhibited with the use of saturable absorbers positioned throughout the amplifier chain, but at a cost in the final output energy. Despite these limitations in the ultimate output energy, the amplification of femtosecond pulses in such systems permits the generation of high focal spot intensities. With the use of spatial filters in the amplifier chain(fig 3.1), low divergence output beams are generated. Thus focal spot intensities of ~ 10^{17} W/cm^2 have recently been generated[34] at wavelengths of 620 nm with 2mJ pulses of 70fs duration.

3.2. High intensity excimer laser systems.

Several laboratories have made heavy investments in the development of subpicosecond high-intensity excimer laser systems. These have the advantage of providing very high intensities in the ultraviolet. Short wavelength high-intensity pulses are of advantages for several types of multiphoton ionization studies, and for inner-shell excitation of many atomic transistions.[35]

Although KrF and XeCl excimer laser discharges are not used themselves to generate short pulses because of their short gain duration, they can be used to amplify ultra-short pulses generated from mode-locked dye lasers. High intensity systems based on this approach are in operation at LANL, at the University of Illinios, and at the Max Planck Institute for Chemical Biophysics in Göttingen. In the system at LANL,fig.3.2, a single pulse selected from the output of a Nd:pumped mode-locked dye laser oscillator-amplifier is doubled in frequency and then progressively amplified in XeCl excimer discharges[36] This system is capable of providing output powers of ~1TW at 308nm, which when focussed to spot sizes of ~3.5μm, creates intensities of $6x10^{18}$ W/cm^2. A similar system utilizing KrF amplifiers at 248nm, has been reported to generate focal spot intensities of ~$2x10^{19}$W/cm.[2,36] Longer pulses at 248nm, of duration 3.5ps at about the same power level have also been reported.[37] The Göttingen group also utilize a hybrid dye-plus-excimer design for their TW laser system, but with the inclusion of a distributed feedback dye laser midway in the laser chain.[38] This system exhibited a very high pulse-to-background ratio(~10^9 in focussed energy). Pulses as short as 60fs are generated at 248nm.[39] In the future it is hoped to extend this system to the 100J, 100fs, pulse level with the addition of large apèrture, double-pass E-beam-pumped KrF amplifiers.[40]

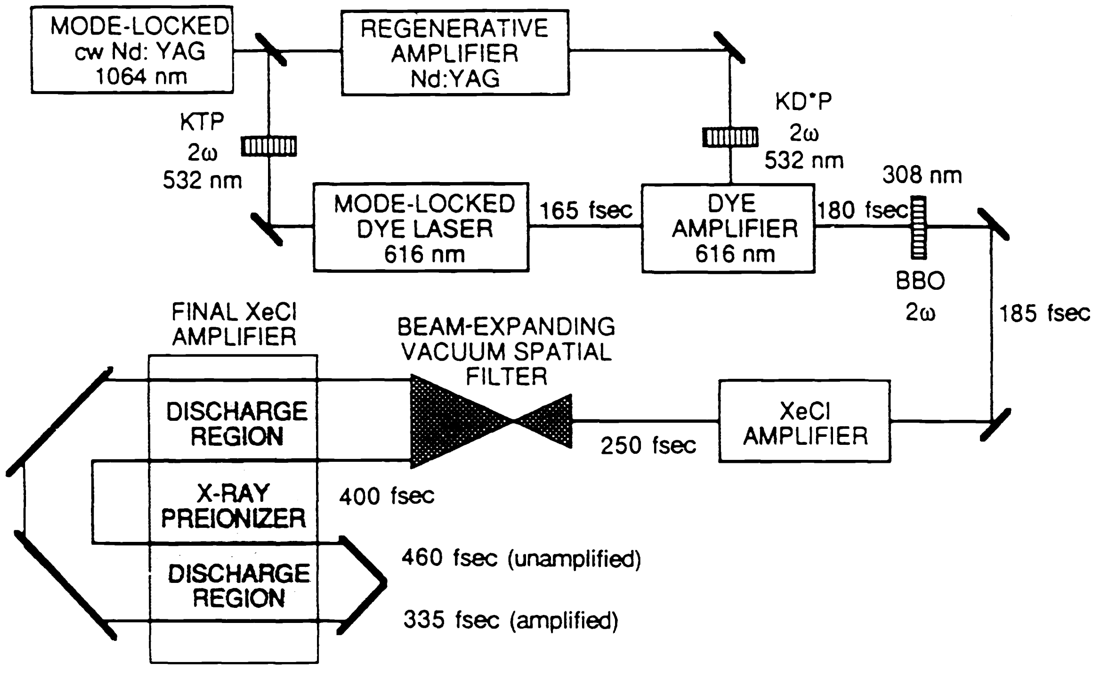

Fig. 3.2. General configuration of the high-intensity XeCl excimer laser at LANL,[1]

3.3. Single picosecond pulse Nd:glass lasers using CPA.

As laser amplifiers, solid state laser media such as those doped with Cr, Nd or Ti, have an intrinsic advantage over dye and excimer lasers in that their saturation fluence is many orders of magnitude higher, up to 5J/cm^2. However, in amplifying directly picosecond or subpicosecond pulses, this high energy storage capabilty cannot be used effectively because of the need to keep the beam intensity below that at which intensity-dependent refractive index effects broaden the pulse. this occurs when the B integral of the medium,[41]

$$B = \frac{2\pi}{\lambda} \int \frac{\Delta n}{n} dl = \frac{2\pi n_2}{\lambda} \int_0^L I(z)dz$$

exceeds a value of ~5. This corresponds to a laser fluence of 10GW/cm^2, well below the saturation fluence for picosecond pulses. This limitation can be avoided by adopting the technique of chirped pulse amplification.[42] In this approach, an initial ultrashort pulse is first stretched out in time by a dispersive delay, and then amplified as a nanosecond pulse at intensitites below that at which B integral effects occur. The amplified pulse is then compressed back to it's original duration by using a spectral dispersive compressor having similar charateristics to that used to stretch the pulse prior to amplification, fig 3.3. Several large Nd:glass high intensity lasers have been constructed using this approach.[43-45] The highest energies generated in ~1ps at a wavelength of 1um at present is ~30J.[45] At present, these systems utilise self-phase modulation(SPM) and group velocity dispersion(GVD) in optical fibers and in some cases a double-grating combination[46] to stretch the pulse injected into the amplifier chain, and a double grating combination to compress the pulse. A limitation that occurs in this approach, to date, has been that a small fraction of the intially chirped pulse has been uncompressed in the output, producing a low intensity (~10^{-4} x I_{max}) pedestal having a duration of up to 1ns, on which the compressed pulse sits. A number of measures have been taken to reduce this

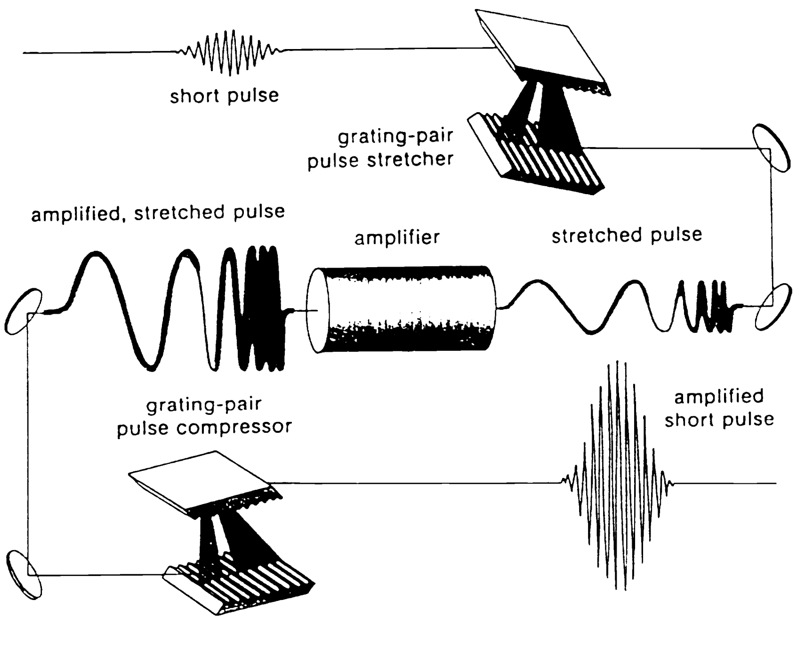

fig 3.3. Principle of Chirped Pulse Amplification(CPA).

pedestal, including the use of spectral or temporal clipping in the expansion stage, harmonic conversion and the use of saturable absorbers. Reductions in the pedestal to levels $\sim 10^{-6}$ of the peak laser power have been observed.[3] However, in the future, higher pulse contrast ratios may be achieved by using an initial 1ps, pulse source at a wavelength of 1um, thus eliminating the the need for using SPM and GVD in an optical fiber. Nd:glass laser oscillators producing pulses shorter than 1ps have been developed using electro-optic feedback controlled active[47] or passive[48] mode-locking (FCM). Their incorporation into a CPA Nd:glass laser system will probably produce higher contrast laser pulses. In a separate direction, Akhmanov's group at Moscow State University have recently used nonlinear optical techniques with a high power picosecond pulse Nd:glass laser to provide the first high intensity (~400GW) sub-picosecond tunable pulses in the infrared.[49]

3.4 . High-intensity femtosecond pulse generation in Ti:sapphire and Alexandrite.

The large spectral gain bandwidth of materials such as Ti:sapphire and alexandrite permit the generation of laser pulses in the 100fs range. A number of modelocking techniques have been used to mode-lock Ti:sapphire, and last year, two of these methods succeeded in generating pulses of 100ps or shorter. These were with the use of coupled-cavity techniques[50] and the use of active mode-locking.[51] Recently self mode-locking in Ti: sapphire has been observed,[52,53] and with the use of intra-cavity dispersive techniques, pulses as short as 60fs observed.[52] These pulses are well suited for injection into chirped pulse amplifier systems employing broad bandwidth solid-state amplifying media such as Ti:sapphire or alexandrite. Millijoule, 100fs pulses have been amplified by CPA in Ti:sapphire,[53] and there are now several groups working towards energies of ~1J. In addition, 100fs pulses have been amplified in in alexandrite.[54]

4. Potential impact of new technologies on high-intensity laser development.

Notwithstanding the immense progress made in high intensity lasers in the last few years, it is worth considering those technologies which could impact this field and give it new thrust in the future. Clearly their are many directions in which high intensity laser development can go; towards longer or shorter wavelength, greater tunability, shorter pulse durations, higher contrast and better focusability. We shall not discuss here all these directions. Instead, we will consider the impact of a few significant technologies, technologies which, in the view of this author, have the capacity to exert a strong influence on the direction of ultra-short laser system development, and on their potential applications.

4.1. New laser materials for ultrashort laser pulses.

The trend towards shorter laser pulses in the pursuit of higher focussed spot intensities meets several criteria. Firstly, shorter pulses are of greater interest for the study of high-density laser-matter interactions, because the effects of hydrodynamic phenomena are avoided. In the achievement of higher focal spot intensities, the use of shorter pulses, without the need for greater pulse energy, alleviates the need to employ larger aperture final stage amplifiers, at least for solid-state media. With the use of techniques such as chirped pulse amplification, which circumvent high-power pulse propagation limitations, solid-state laser materials are attractive media because of their high saturation fluence. Physical requirements that the media must have include good optical properties, a high damage threshold and the capability to be fabricated in large sizes. To be efficient as a laser amplifying medium for ultrashort laser pulses or chirped amplifier pulses, they must have broad gain bandwidth, and be efficiently pumped by flashlamps over large apertures. For pulses of 100fs duration, two media have so far been used, Ti:sapphire and alexandrite. However, although these materials have broad gain bandwidths, (Table 4.1.), they both have some drawbacks for the longterm development of high intensity lasers. The short 3.2µs fluorescence decay time of Ti:sapphire forces the need for coherent optical pumping by an auxiliary high power laser (upconverted Nd: glass or YAG, or an excimer laser). Although alexandrite is flashlamp-pumped, its optical quality and the need to operate the medium at hgih temperatures increase its complexity as an amplifying medium.

Table 4.1. Properties of some solid state laser materials suitable for ultrashort pulse generation.

Material	Wavelength (nm)	Linewidth (nm)	Cross-sect σ (cm 2)	Lifetime τ (μs)	$\sigma\tau$ product (cm 2 sec)
Cr:LiSAF	850	220	$5.0 x10^{-20}$	67	$3.4x10^{-24}$
Cr:LiCAF	770	120	$1.3x10^{-20}$	175	$2.3x10^{-24}$
Ti:sapphire	735	122	$4.1x10^{-19}$	3.2	$1.3x10^{-24}$
Alexandrite	760	100	$8.5x10^{-19}$	260	$2.2x10^{-22}$
ruby	694	0.5	$2.5x10^{-20}$	3000	$7,5x10^{-23}$
Nd:YAG	1064	0.5	$6.5x10^{-19}$	230	$1.5x10^{-22}$
Nd:glass	1053	22	$4.0x10^{-20}$	330	$1.3x10^{-23}$

There are several new laser materials which appear to overcome these limitations. The properties of two of them, Chromium-doped $LiSrAlF_6$,[55] and $LiCaAlF_6$.[56] are listed in Table 4.1. Significant developments of these materials, particularly, their fabrication in large sizes with good optical quality have been made by B. Chai at CREOL[57].

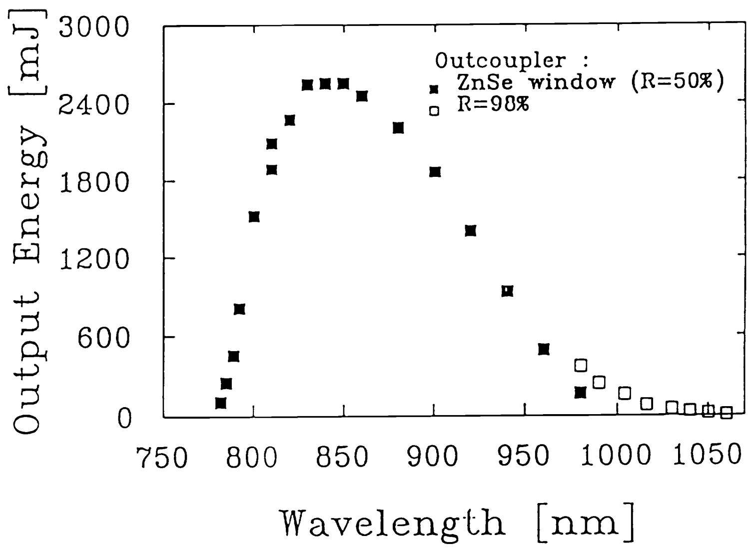

fig.4.1. Tuning range of $LiSrAlF_6:Cr^{+3}$. (Courtesy of M. Stalder & M.Bass).

With a fluorescence decay time of 67µs, $LiSrAlF6:Cr^{+3}$, can be configiured as a conventional , flashlamp-pumped amplifier, where the Cr concentration is adjusted to provide uniform pumping for a given pumping fluence and geometry. Efficient laser operation has been demonstrated over a significant portion it's fluorescence bandwidth.[58] Recently this range has been extended on the long wavelength side to beyond 1.06µm, fig.4.1. It is now possible to consider the development of a 1ps mode-locked $LiSrAlF6:Cr^{+3}$ laser operating at 1.06µm as an initial pulse source for injecting into high intensity Nd:glass CPA laser systems. In addition, the broad bandwidth of these new materials, there optimisaton to flashlamp pumping, their optical quality and manufacturability, make them attractive media for future high power femtosecond laser systems. Their gain bandwidth overlaps well that of

Ti:sapphire and alexandrite. Thus the development of multi-media high-intensity systems, incorporating techniques such as CPA can be envisaged in the future.

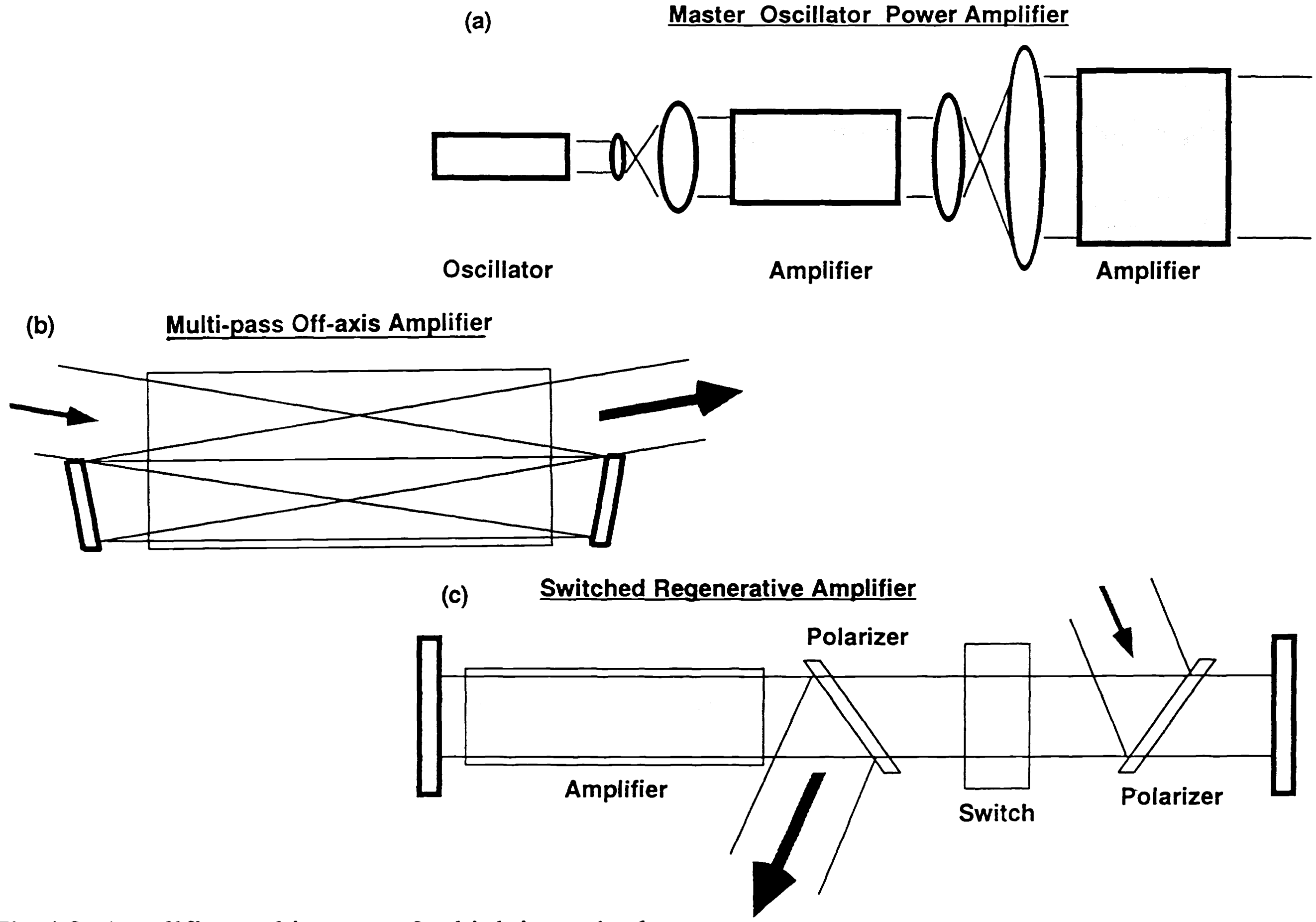

Fig 4.2. Amplifier architectures for high intensity lasers.

4.2. New amplifier technologies.

Most high-power laser systems, solid-state, gas, and liquid have employed the MOPA system of laser architecture. Although this is the simplest in concept, and effective in operation, it is theoretically not the most efficient or the least costly approach. More efficient architectures use multi-pass amplifier configurations. Fig 4.2. shows a selection of different amplifying configurations [59] A varaint of the version shown in fig 4.2(c). forms the subject of a detailed paper by Hunt,[60] a companion to this paper. The incorporation of this type of architecture with chirped pulse amplification will increase considerably the short pulse energy extraction efficiency for solid state laser systems. Considerable cost savings compared to the standard MOPA design will also occur as a consequence of the reduced use of secondary optics, spatial filters and the like.

4.3. High-intensity short-pulse focusing optics

Several authors have recognised the limitations of conventional refractive optics for focussing compressed ultrashort laser pulses. apart from the efects of fabrication imperfections in large aperure optics, two major limitations have been identified with the use of refractive optics. Firstly, nonlinear optical effects occuring in the focusing lens and subsidiary optics would induce temporal dispersion on the beam. Secondly, the effects of pulse distortion of the ultrashort pulses in refractive focussing optics cannot be ignored.[61] Many of these problems can be avoided by utilising reflective optics. This may involve considerable complexity, since part, if not all, ofthe compression optics may well have to be maintained within vacuum.

5. Summary

To the present time progress in high-intensity laser capabilities have taken place, largely, as a result of extensions of existing technologies. new laser materials and new laser architectures will significantly affect the direction of future progress. Efficient broad bandwidth solid-state laser materials in combination with nonlinear optical techniques will lead to tunable high intensity sources across wide ranges of the visible and infrared spectrum. In addition, new amplifier technologies combined with these new laser materials point the way toward the design of systems having efficient energy extraction for pulses in the 100fs range. The generation of single pulse energies inthe 10-100J range should soon be realizable. With the assumption that these petawatt(PW) powers can be focussed to spot sizes of a few tens of microns, permits the generation of pulse intensities and elelctric field densities at which many new physical phenomena should be observable.

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