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High Power Ultrashort Pulse Lasers

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Small scale terawatt and soon even petawatt (1000 TW) class laser systems are made possible by application of the chirped-pulse amplification technique to solid-state lasers combined with the availability of broad bandwidth materials. These lasers make possible a new class of high gradient accelerators based on the large electric fields associated with intense laser-plasma interactions or from the intense laser field directly. Here, we concentrate on the laser technology to produce these intense pulses. Application of the smallest of these systems to the production of high brightness electron sources is also introduced.

Introduction

The application of chirped pulse amplification (CPA) to solid-state lasers[1,2] has made it possible to produce pulses exhibiting terawatt peak power from tabletop systems. The CPA technique makes possible efficient energy extraction from good energy storage materials by short pulses, without incurring nonlinear effects associated with high intensity. The rapid progress in peak power has been due largely to our ability to generate and manipulate these short pulses and amplify them in a wide variety of broadband solid-state materials: Nd:glass,[1-4] alexandrite,[5] Ti:sapphire,[6,7] Cr:Li:SAF[8,9] and combinations of these materials.[10] The CPA technique has been also been applied to existing, large-scale Nd:glass systems at the CEA Limeil in France,[11,12] the Rutherford-Appleton Laboratory in Great Britain and at Osaka in Japan. To date, pulses with peak power exceeding 55 TW (25 joules, 400 fs) have been produced.[12] The construction of a number of 100 TW systems is underway at several laboratories and the first 1000 TW (petawatt) laser is under development at Lawrence Livermore National Laboratory.[13]

The production of a high-peak-power pulse is necessary but not sufficient to produce a high-irradiance pulse on target. The brightness and ultimate focused irradiance achievable with a laser pulse is determined by both the peak power and the spatial quality (divergence) of the pulse. By paying careful attention to both linear and nonlinear aberrations in the laser and beam-transport systems, the pulses produced by CPA systems can be nearly diffraction-limited. Irradiance as high as 10^{19} W/cm^2 has been achieved on existing multiterawatt systems. Scaling these CPA based systems up to the 100-TW or even petawatt (1000-TW) level will enable the study of laser-matter interaction at 10^{21} W/cm^2 and beyond.

The availability of these intense pulses opens a new regime of laser-matter interaction to study. Applications stem from four fundamental features of such a pulse. The first is the short duration, which virtually eliminates hydrodynamic motion on the time scale of the pulse. This short duration, combined with a high pulse contrast makes possible the generation of ultrashort x-ray pulses[14] and solid-density plasmas.[15] Second are the large electric and magnetic fields associated with an intense pulse. At 10^{21} W/cm^2, the resulting electric field is 10^{14} V/m, over 100 times the Coulomb field which binds atomic electrons. This field strength is sufficient to ionize heavy atoms such as uranium to U^{82+} within only the first few femtoseconds of the pulse [Figure 1]. Third is simply the energy density and resulting light

pressure of the pulse. At 10^{21} W/cm^2, the energy density of the pulse is over 3 x 10^{10} J/cm^3, which corresponds to a 10-keV blackbody and an equivalent light pressure of 300 gigabars. Finally, the free electrons produced by these intense pulses are driven by the field to a cycle-averaged oscillatory energy given by

$$E_{osc} = mc^2 \{ \sqrt{1 + 2U_p/mc^2} - 1 \} \qquad [1]$$

where U_p (eV) = $e^2E^2/4m\omega^2 = 9.33 \times 10^{-14}\, I$ (W/cm^2) λ^2 (μm). The electron quiver energy and resulting phenomena accessible are shown in **Figure 2**. For a Nd:Glass laser producing 1.05 μm laser pulses, the electron quiver energy is nearly 10 MeV for a laser intensity of 10^{21} W/cm^2. Relativistic effects in such a plasma become dominant and can result in new absorption mechanisms,[16,17] including absorption past the critical surface[18] and self-focusing[19] not observed in conventional laser-plasma experiments.

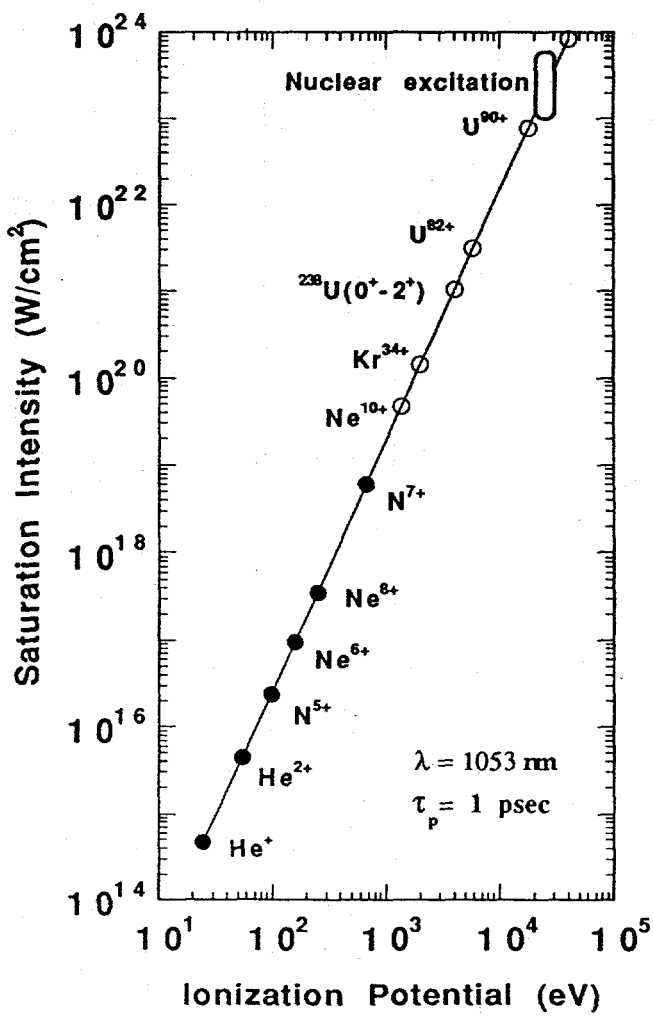

Figure 1

Intensity required to produce a given ionization state by tunneling. Filled circles have been observed to date in experiment.

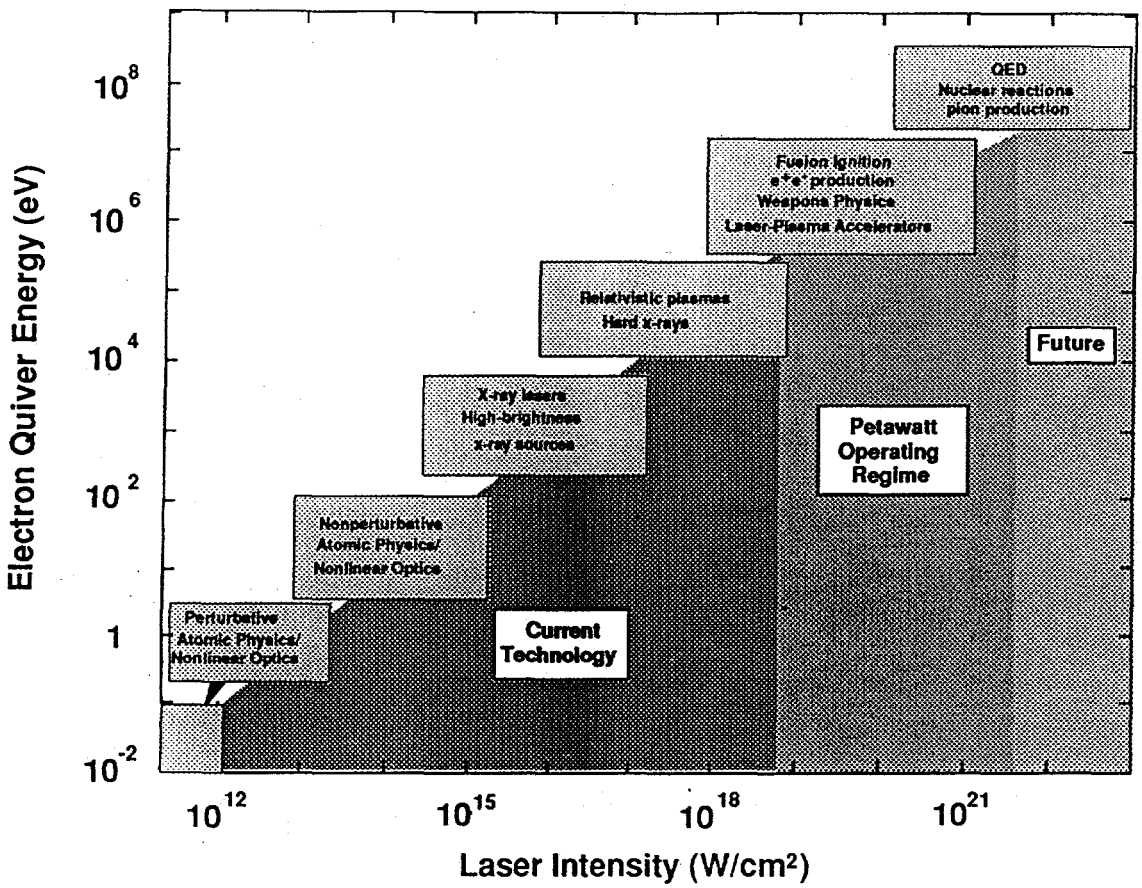

Figure 2. Free electron quiver energy and accessible phenomena as a function of laser irradiance (λ=1053 nm)

Laser Materials

The design of any laser system is determined by the optical and thermo-mechanical properties of the laser material. Production of ultrashort terawatt or petawatt pulses severely limits the choice of laser materials. First, the material must have sufficient bandwidth to support the bandwidth of the final pulse duration. The production of a short pulse operating at the fundamental limit, $\Delta\upsilon\Delta\tau \approx 0.5$, requires materials which exhibit a very broad gain bandwidth. For the production of 100 fsec pulses at 800 nm, the bandwidth of the pulse must be approximately 5 THz ($\Delta\lambda \approx 11$ nm). The handful of laser materials which exhibit a gain bandwidth sufficient to support the bandwidth of these short pulses is shown in Table I.

Table I: Laser Properties of Broadband Materials

Laser Material	Cross Section, σ (10^{-20} cm^2)	$\Delta\lambda$ (nm)	τ (fs)	F_{sat} (J/cm^2)
Rhodamine	20,000	80	8	.0016
Nd:Glass Phosphate	4	22	80	4.8
Nd:Glass Silicate	2.3	28	60	8.0
Nd:Glass Combination	1.5	60	30	12.5
Ti:Sapphire	30	120	8	.8
Alexandrite	1	100	10	25.5
Cr:LiSAF	3	50	15	8.0

A second important property of laser material, the saturation fluence, F_{sat}, is also shown in Table I. The saturation fluence, $F_{sat} = h\,\upsilon/\sigma$, where h is Planck's constant, υ is the laser frequency, and σ the stimulated emission cross section, is a measure of the extractable energy from a laser amplifier. The saturation fluence of most solid-state laser materials is between 1 J/cm^2 (Ti:Sapphire) and 6 J/cm^2 (Nd:Glass), over a thousand times larger than that of organic dyes (Rhodamine) or excimers. It is for this reason that nearly all high pulse energy laser systems are based on solid-state materials.

Unfortunately, the large stored energy of solid-state materials cannot be accessed directly with femtosecond or even picosecond pulses. This is a result of the intensity-dependent index of refraction $n = n_o + n_2 I$, where I is the intensity of the pulse. This intensity-dependent refractive index produces a nonlinear phase retardation given by,

$$B = \frac{2\pi}{\lambda} \int_o^l n_2 I(z)\, dz \qquad [2]$$

This nonlinear phase retardation results in wavefront distortion and eventually catastrophic filamentation[20] resulting in damage to the amplifier. For example, a picosecond pulse amplified to 1 J/cm^2 could travel only 1 mm through a typical solid-state laser material without suffering severe degradation of beam quality. The effects of the nonlinear refractive index are not limiting in dye or excimer systems since their low saturation fluence limits the fluence of amplified pulses to only a few millijoules per square centimeter.

Chirped-Pulse Amplification

The several joule/cm^2 capability of solid-state materials is easily accessed with nanosecond pulses where systems are commonly designed to produce pulses up to the limit imposed by nonlinear beam break-up. This peak power limit in solid-state systems is approximately 2-4 GW/cm^2. Thus, operation of a Nd:Phosphate glass laser at the saturation fluence of 4.8 J/cm^2 would require pulses of greater than 1 nsec to remain below the peak power limit. The technique of chirped-pulse amplification overcomes this limit by making it possible to access the high saturation fluence of solid-state materials with femtosecond pulses. The essential idea of CPA is to stretch the initial femtosecond pulse by a factor of several thousand prior to amplification. The pulse is then amplified by a factor of 10^6–10^{11} and recompressed ideally to its initial value (Figure 3). CPA was developed initially to overcome the peak power limitations in radar.[21] By stretching the pulse before amplification they could achieve high pulse energy for long-distance ranging. Recompression of the echo produced the short pulses necessary for accuracy. In most CPA laser systems, the pulse is recompressed directly after amplification.

1. *Generation of Short and Ultraclean Pulses, E(t)*

The first step in a CPA laser system is the production of the low energy seed pulse. These pulses need to have a duration as short or shorter than the final desired pulse duration (or contain sufficient bandwidth to produce the final pulse). Short pulses are always produced by mode-locking in a laser oscillator. In most laser oscillators, there are many longitudinal modes which can oscillate within the gain bandwidth of the laser material. By locking these various longitudinal modes in phase (mode-locking) short pulses are produced by the linear superposition of the various longitudinal modes. Active modelocking is achieved typically by placing an acousto-optic or electro-optic device within the laser cavity to modulate the loss of the cavity. Only those modes oscillating in phase with the modulator experience a sufficiently low loss to lase. Active mode-locking with solid-state materials typically produces pulses as short as 10-100 psec. Passive mode-locking is often achieved by placing a saturable absorber within a laser cavity. In this case, only a high intensity mode-locked pulse can saturate the absorber and oscillate wihtin the cavity. Passive mode-locking can typically achieve pulses as short as a few picoseconds in broad bandwidth solid-state materials.

Although still commonly used, conventional active and passive mode-locking have been supplanted by a new technique, Kerr-lens mode-locking, for the generation of very short pulses. The Kerr-lens effect is a result of the nonlinear refractive index discussed earlier. As the pulse propagates in the nonlinear material, various locations within the pulse will experience a nonlinear phase retardation proportional to their intensity (eq. 2). Just as a lens focuses light by retarding the phase more at the center than on the edge, those regions with the greatest nonlinear phase (highest intensity) will tend to focus creating a higher local intensity. Normally detrimental, this self-focusing effect can be used to an advantage within a laser cavity for mode-locking. If the cavity is designed as a TEM_{00} (Gaussian) resonator, the intense, mode-locked pulse will self-focus as it propagates through the nonlinear medium. By designing the cavity such that either higher gain or lower loss is associated with the self-focused beam, the intense mode-locked pulse will dominate over continuous wave operation. The self-focusing acts like a very fast saturable absorber within the cavity. Pulses as short as 10 fsec[22] have been produced using the Kerr-lens effect in Ti:sapphire oscillators.[23] Commercial systems producing 50-1000 fsec pulses are now common.

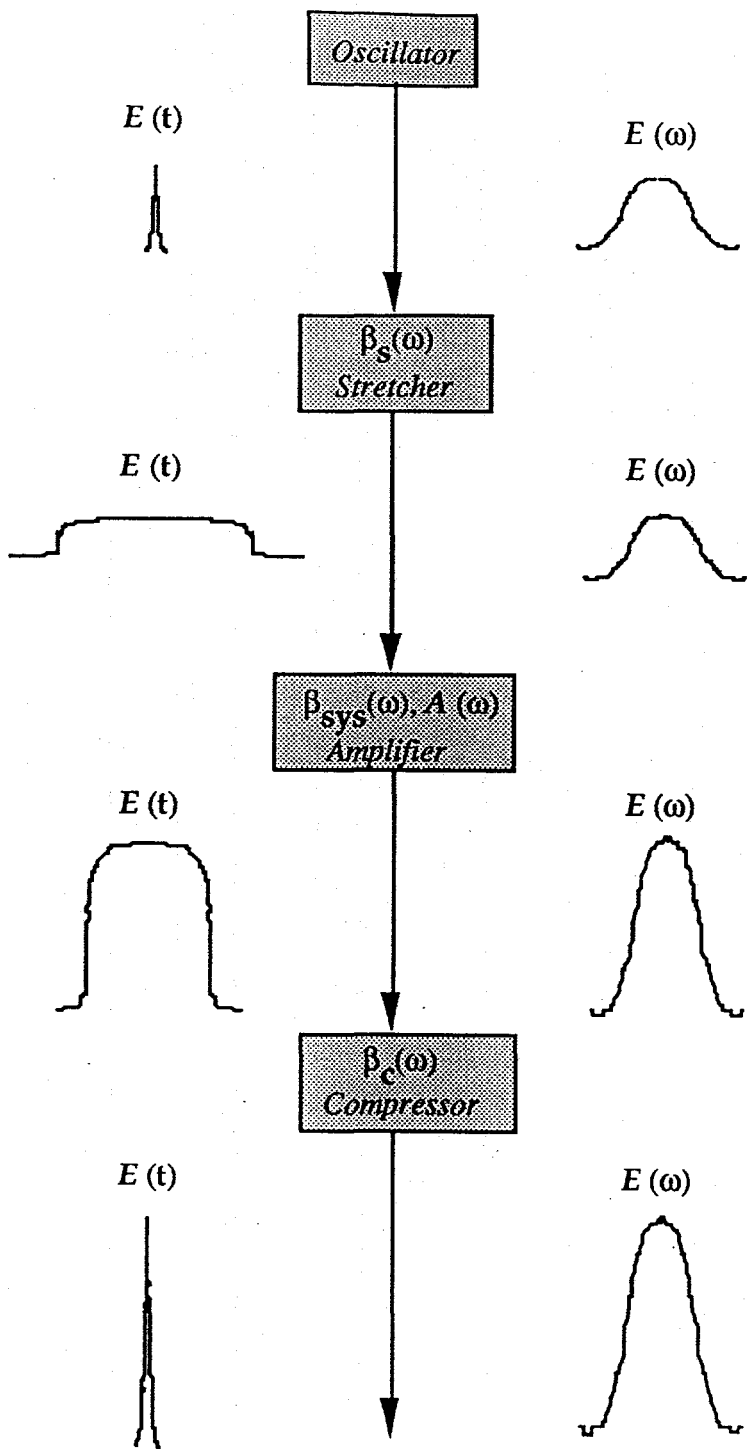

Figure 3. Chirped pulse amplification concept. E (t) and E (ω) represent the time and frequency-dependent field distribution, β(ω) and A (ω) are the frequency-dependent phase and amplitude transfer functions of a given component.

2. *Stretching , $exp[j\beta_S(\omega)]$*

After generation, the short pulse is passed through a device exhibiting a frequency dependent phase function, $\beta(\omega)$, producing a chirped and stretched pulse. A chirped pulse is one exhibiting a frequency-dependent temporal distribution. The first CPA demonstration[1] used group velocity dispersion in fiber to stretch the pulse. The lower frequency (red) components of the pulse propagate faster through the fiber than the higher frequency (blue) components resulting in a stretched and positively chirped (red earlier than blue) pulse exiting the fiber. The recompression was performed by using the negative group velocity dispersion provided by a diffraction grating pair.[24] In fiber-grating based CPA systems, the maximum stretching/compression ratio achievable is of the order of $\Delta\lambda/\lambda$. This is a result of the fact that the frequency dependent phase functions, $\beta(\omega)$ are different between the fiber used for stretching and the grating pair used for compression. The mismatch is tolerable when the stretching ratio, R, is less than approximately $\Delta\lambda/\lambda$. When R exceeds this value, incomplete recompression is performed, producing unwanted ripples before or after the main pulse. As an example, a picosecond pulse at 1.06 μm with a $\Delta\lambda$ of 20 Å cannot be stretched and compressed by more than a factor of 500 with fiber-grating systems. For a 100 fsec pulse, fiber-grating systems are limited to $R \approx 50$.

In 1987, Martinez proposed a compressor with positive group velocity dispersion to compress pulses at 1.5 μm.[25] Martinez' compressor is composed of a telescope with magnification of one between two anti-parallel diffraction gratings. It was immediately recognized that, unlike the fiber this positive dispersion "compressor" would act as a pulse stretcher nearly exactly matched to the negative dispersion grating pair used for compression. M. Pessot, *et al.*[26] first demonstrated this all grating stretching/compression system by stretching an 80-fsec pulse to over 80 psec ($R > 1000$) and then recompressing the pulse without introducing any temporal distortion. The matched stretcher/compressor was a breakthrough. Stretching and compression ratios of over 10^4 were rapidly achieved.[27] A multitude of stretcher designs are now being pursued to further improve the stretching/compression ratio to over 10^5 for pulses below 50 fsec.[28-30]

3. *Amplification and Gain Narrowing, $A(\omega)$*

Short pulses have a large Fourier spectrum which is modified by the amplitude transfer function, $A(\omega)$ of the amplifier system. Because of the large overall gain involved in CPA, typically 10^{10}, the spectrum of the amplified pulse is significantly narrower than the material bandwidth.[4] If we approximate the gain spectrum by a Gaussian distribution, the spectrum following amplification at line center by an amount, G_O, is reduced to a full width at half-maximum, $\Delta\upsilon_{out}$, given by,

$$\frac{\Delta\nu_{out}}{\Delta\nu_a} = \frac{\ln\{\ln G_o/(\ln G_o - \ln 2)\}}{\ln 2}$$

where $\Delta\upsilon_a$ is the material gain bandwidth. For example, the maximum bandwidth achievable following amplification of 10^8 in a material such as Nd:Glass with an effective Gaussian

lineshape of 12 nm would be 2.82 nm resulting in a minimum pulse duration of 650 fsec. Amplification near 800 nm of the same amount in a material with a broader gain bandwidth such as Ti:Sapphire or Cr:LiSAF would produce a pulse with a bandwidth near 26 nm capable of supporting pulses as short as 40 fsec. For these extremely broad bandwidth materials, the ouput bandwidth will often be limited to less than the gain narrowed bandwidth due to limitations in other components such as polarizers, mirrors, etc.

4. *Compression, $\exp[j\beta_c(\omega)]$*

A perfectly matched stretcher/compressor will have conjugate phase functions, $\beta_s(\omega) = -\beta_{compressor}(\omega)$. However, for the very large stretching and amplification required to produce terawatt class pulses, the phase function of the compressor must account for the sum of the phase functions of the stretcher *and* the remainder of the laser system, $\beta_{system}(\omega)$, including amplifiers, mirrors, waveplates, etc. Defining $E_{in}(\omega)$ as the field entering the compressor and $A(\omega)$ as the amplitude transfer function of the compressor, the field exiting the compressor can be written as,

$$E_{out}(\omega) = E_{in}(\omega)A(\omega)\exp\{ j\,[\beta_s(\omega) + \beta_{system}(\omega) + \beta_{compressor}(\omega)]\} \qquad [4]$$

we see that a transform-limited pulse is achieved after compression only if $\delta = [\beta_{compressor}(\omega) + \beta_s(\omega) + \beta_{system}(\omega)] = 0$. In this case, the amplified and compressed pulse has no residual chirp, $\delta = 0$, and the temporal distribution of the field is just the Fourier transform of the spectrum of the field. In practice, the grating separation in the compressor is adjusted until the error, δ is a minimum.

Multiterawatt to Petawatt Pulses

An example of a multiterawatt CPA laser system is shown in figure 4.[31] Although specific systems will differ in the details of the design and/or laser material chosen (Cr:LiSAF, Ti:Sapphire, Nd:Glass), all systems will contain the basic elements to achieve the four basic steps described above and illustrated in figure 4. The laser begins with a Kerr-lens mode-locked Ti:sapphire oscillator producing 100-fs pulses at 825 nm. These low energy (≈ 5 nJ), transform-limited pulses are stretched by a factor of 5000 to 500 psec in a Martinez type, double-grating pulse stretcher employing achromatic lenses. After stretching, the pulse energy is reduced to 1 nJ. This low-energy pulse is injected into a Cr:LiSAF ring regenerative amplifier.[32] For operation in the 800-950 nm range, either Ti:Sapphire or Cr:LiSAF may be used as the gain medium at this stage. When Ti:Sapphire is used, laser pumping of the amplifier is required due to the short (3.2 μsec) lifetime of the upper laser level. On the other hand, Cr:LiSAF exhibits a 67 μsec upper state lifetime,[33] sufficiently long for conventional flashlamp-pumping. The TEM_{00} regenerative amplifier serves as a multipass amplifier while ensuring diffraction-limited beam quality. Thirty-three passes through the gain medium are required to achieve an amplification of over 10^7 at 825 nm. The regenerative amplifier routinely produces 10-mJ pulses. The beam exiting the regenerative amplifier exhibits a diffraction-limited Gaussian profile characteristic of the TEM_{00} regenerative amplifier cavity.

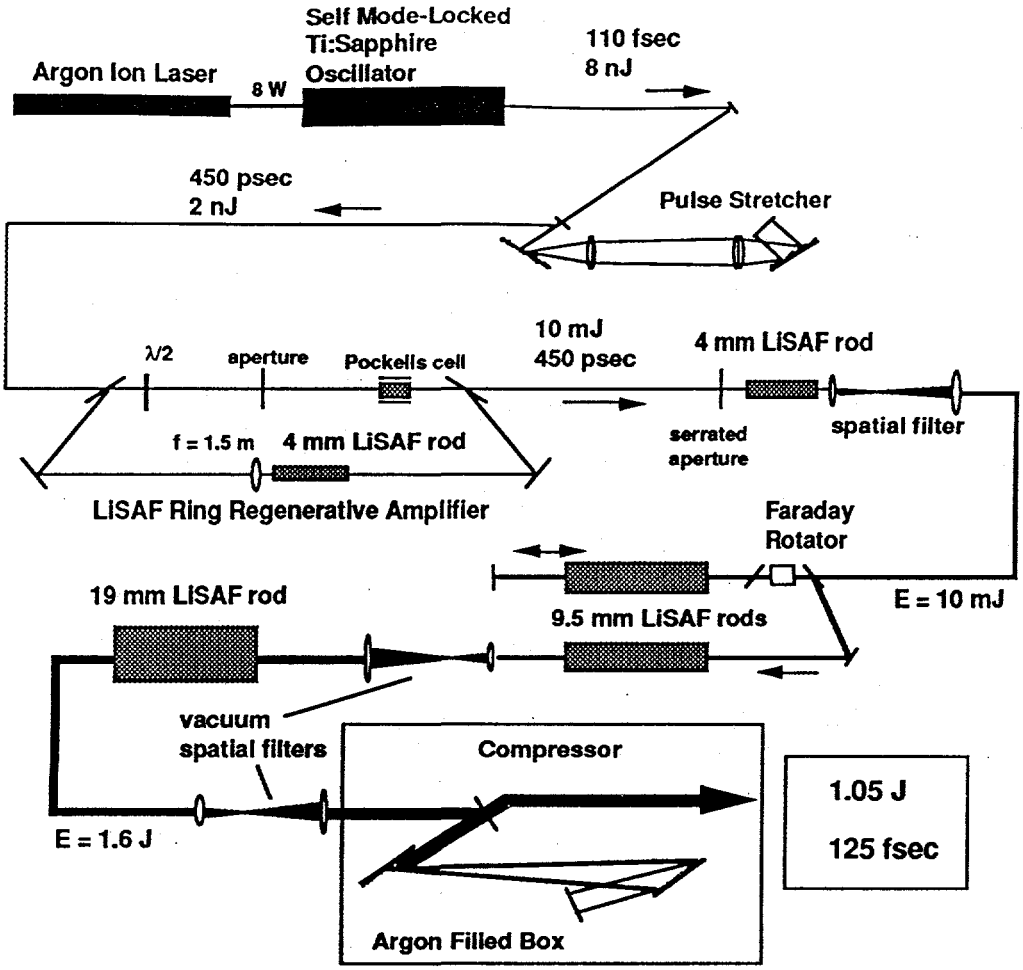

Figure 4: Optical schematic of 8 TW Cr:LiSAF laser system (from ref. 31).

After the regenerative amplifier, the beam profile is transformed from a Gaussian distribution to a near flat-top. Upon further amplification, the flat-top beam can produce greater pulse energy and superior near field beam quality than a corresponding Gaussian. An approximately 60% loss in energy is associated with the transformation to a flat-top. This energy loss is more than recovered in the 4x65 mm Cr:LiSAF amplifier following the aperture. The beam is expanded in diameter with a Galilean telescope and further amplified to 400 mJ in a pair of 9 x 100 mm Cr:LiSAF flashlamp-pumped amplifiers. A second telescope expands the beam to 16 mm in diameter for final amplification to the 1.5 Joule level in a 19x100 mm Cr:LiSAF amplifier.

Following amplification, the beam is expanded in a final telescope and relay imaged to the pulse compressor. The compressor consists of a pair of six inch diameter, 1800 l/mm diffraction gratings. The beam is expanded to 50 mm in diameter before the pulse compressor to remain safely below the 300 mJ/cm^2 femtosecond damage threshold of the gold coated gratings.[34] The gratings are used near the Littrow angle of 48 degrees to minimize astigmatism. The entire pulse compressor resides in an argon filled chamber in order to eliminate the possibility of self-focusing and self-phase modulation in air following compression. The pulse from the amplifier system enters the compressor through a thin MgF_2 window. Diffraction from the first grating produces the negative (blue faster than red) temporal dispersion necessary to compress the pulse. The second grating stops the dispersion induced by the first. The diffracted beam from the second grating strikes a retroreflecting

mirror within the compressor chamber which reflects the beam back through a second pass of the grating pair. The grating separation is adjusted to cancel both the pulse stretching from the initial grating stretcher and temporal dispersion from propagation through the laser system. The gratings exhibit 89 and 91% diffraction efficiency resulting in an overall compressor efficiency of 65 % producing a 1.0 Joule, 125 fsec compressed pulse.[31]

Further developments in CPA based systems will include both higher repetition rate and higher peak power. The peak power will be increased both by decreasing the pulse duration and increasing the pulse energy. As mentioned previously, there are already Kerr-lens mode-locked osillators producing 10 fsec pulses[22] and such oscillators have already now been incorporated into terawatt class systems.[35] The highest pulse energy system currently under development is an Nd:Glass system designed to produce 1 kilojoule pulse in a duration as short as 500 fsec. Further increase in the available power from CPA systems will soon be limited by optical damage to the diffraction gratings used for pulse compression and the optics used to transport and focus the intense pulse. In figure 5, we show the surface damage threshold of the common dielectric materials fused silica and calcium fluoride.[36] The conventional $\tau^{1/2}$ scaling of the damage threshold with pulsewidth breaks down below approximately 10 psec. For these short pulses, there is insufficient time for significant for energy transfer from the electrons to the lattice and damage becomes dominated by plasma formation rather than thermal boiling or melting. In the 0.1 to 10 psec regime, plasma formation is dominated by collisional (avalanche) ionization. As the pulse duration is decreased further, multiphoton ionization plays an increasingly dominate role further decreasing the fluence at which damage occurs (figure 6).

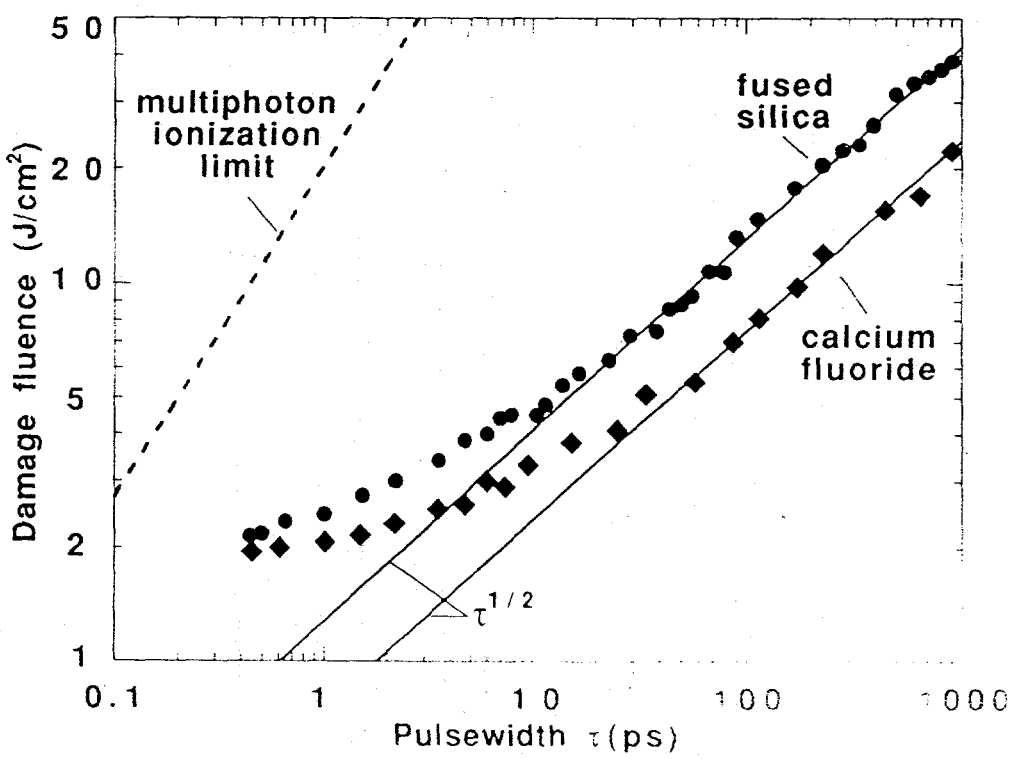

Figure 5: Measured front surface damage threshold of fused silica and calcium fluroide as a function of laser pulse length at 1053 nm (from ref. 36).

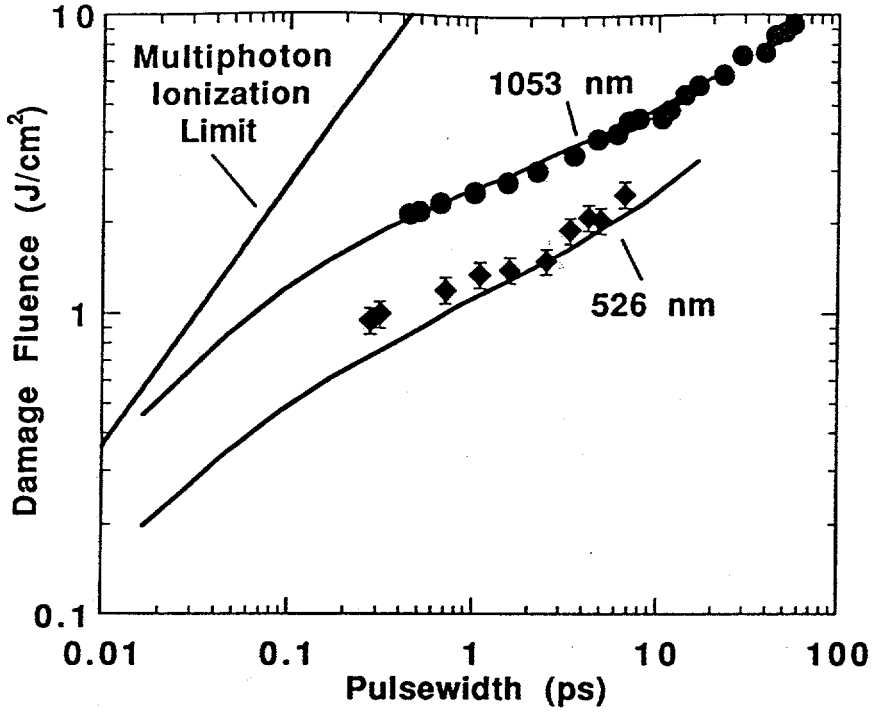

Figure 6: Measured and calculated front surface damage thresholds of fused silica as a function of laser pulse length (from ref. 36).

As a result of the inherently low damage threshold of materials (both metals and dielectrics) in the femtosecond regime, petawatt class lasers will require diffraction gratings which are currently beyond the state of the art. For example, a metallic grating with a damage threshold of 200 mJ/cm^2 for 100 fsec pulses would require 25x50 cm high efficiency diffraction gratings to produce petawatt pulses upon compression. For picosecond pulses 60x94 cm gratings are required to produce petawatt pulses even utilizing recently introduced dielectric reflection gratings.[37] The largest high-efficiency diffraction gratings commercially available are 25x40–cm metallic gratings with a diffraction efficiency as high as 92%.

Equally important to obtaining the highest possible peak power from future CPA systems will be increased average power (repetition rate). Current terawatt class systems exhibit repetition rates from 1-10 Hz with an average power on the order of 1 Watt, approaching that of conventional longer pulse solid-state systems. This average power can be expected to increase for terawatt class systems by perhaps an order of magnitude to the 10 Watt level over the next few years as the cost and complexity of diode-pumping become tolerable. Ti:Sapphire is readily pumped by the second harmonic output of diode-pumped Nd:YAG lasers while Cr:LiSAF can be directly diode pumped in the 680 nm range by AlGaInP diodes. Systems exhibiting a peak power on the order of a few tens of Gigawatts are currently available at repetition rates from 1 to 50 kHz. These systems are also at the 1 Watt average power level increasing to the 10 Watt level over the next few years. Petawatt class systems will likely be limited to average power levels on the order of one watt for several years due to the requirement of cooling large volumes of laser material.

Applications

The applications of these high power, short pulse lasers arise from the basic features of the field described earlier. At the irradiance available with petawatt lasers (>10^{21} W/cm^2) the field strength will be greater than 10^{14} V/m enabling a host of new phenomena including new approaches to direct laser acceleration of light particles (electrons /positrons). As an example, the enormous light pressure (>100 Gbars) no longer restricts laser-plasma interactions to densities below the critical density. The light pulse will penetrate to a density where pressure balance between the light wave and the supercritical density plasma is achieved. This effect was recently inferred by observing a reduction of the plasma thermal expansion with increasing laser intensity.[38] CPA laser systems are being applied to problems ranging from time resolved chemistry to inertial confinement fusion.[39] Concentrating on their application to accelerators, these systems will find application as the driver for laser-plasma based accelerators (beat wave, laser wakefield), direct laser field acceleration and as sources for driving photocathodes as the source in conventional accelerators. The laser acceleration schemes are covered in numerous papers throughout these proceedings. Here, we consider briefly the use of CPA lasers as drivers for photocathode injectors.

Laser driven photocathodes have been introduced as an electron source for accelerators offering substantially improved brightness over conventional thermionic sources.[40,41] Metal photocathodes have been shown to be robust sources providing high current and low emittance.[42,43] Being conventional photoemissive sources, these cathodes require a laser source operating in the ultraviolet providing photons at an energy above the work function of the cathode. Copper cathodes for example require laser sources operating below 268 nm. Operation in the ultraviolet requires careful attention to laser damage and several nonlinear steps to convert the infrared radiation produced by solid-state lasers. Here, we introduce a new type of metal photocathode injector driven by a small scale, high repetition rate CPA system which eliminates the necessity of working in the ultraviolet.

By utilizing pulses with a duration less than approximately 500 fsec, it is possible to saturate the multiphoton emission probability at laser fluences below the damage threshold of standard reflective metals.[44] A typical copper (ϕ=4.65 eV) or aluminum (ϕ=4.30 eV) photocathode requires the absorption of three photons of 800 nm radiation to overcome the work function of the material, ϕ. Estimation of the three photon absorption cross section of aluminum suggests that an intensity on the order of 10^{11} W/cm^2 is required to have an appreciable ionization probability for three photon emission at this wavelength. For 100 fsec pulses, this translates into a pulse fluence of only 10 mJ/cm^2. This fluence is over an order of magnitude below the 400 mJ/cm^2 damage fluence of aluminum for 100 fsec pulses. As a result, space-charge limited flow equal to that achieved with ultraviolet (single-photon absorption) laser light should be achieved from an aluminum photocathode irradiated with 800 nm, 100 fsec laser pulses without damaging the photocathode.

Demonstration of the multiphoton photocathode is shown in figure 7. The photocurrent resulting from 825 nm and 413 nm irradiation of an aluminum photocathode biased with 2-20 kV over a 2 cm gap is shown as a function of incident laser intensity. Space charge limited flow is achieved for both two-photon (λ=413 nm) and three-photon (λ=825 nm) emission.[44] No damage could be observed to the photocathode in either case after days of irradiation.

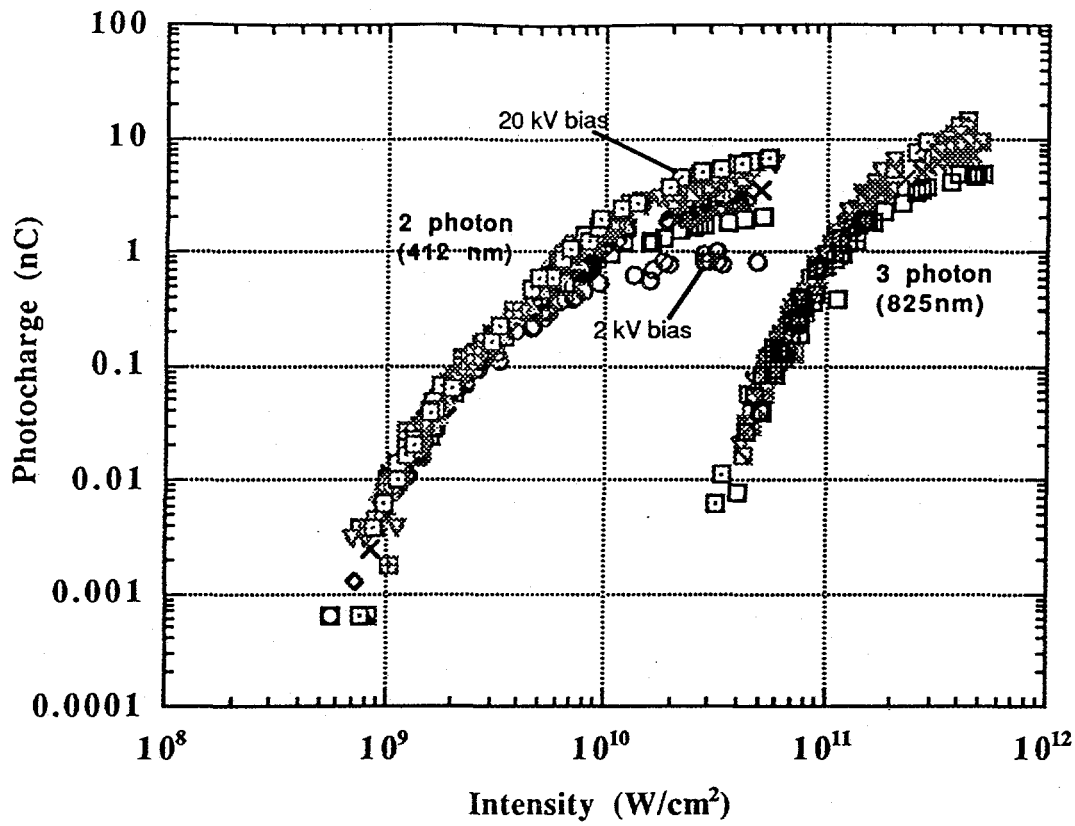

Figure 7: Photocurrent produced by two and three photon ionization with 100 fsec pulses of an aluminum cathode biased at 2 to 20 kV over a 2 cm gap. (from ref. 44)

Conclusion

The development of terawatt- and soon petawatt-class lasers based on chirped-pulse amplification has led to a revolution in the interaction of intense laser radiation with matter. In this article, we have focused on the enabling laser technology and introduced the application of the smallest of these these sources to photocathode injectors. Continuing laser development to produce both high-energy (multikilojoule), low-repetition-rate systems and low-energy (millijoule) but high-repetition-rate (kHz)[45-47] systems will further expand the applications of these powerful new laser sources.

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