10. Blue-Green Dye Laser Seeded Operation of a Terawatt Excimer Amplifier

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With 5 Figures

A tunable, subpicosecond dye laser oscillator–amplifier system, generating 2-mJ pulses in the blue-green spectral region, has been developed. It consists of a hybrid synchronously pumped oscillator followed by a specially designed, high-performance, two-stage amplifier system. The pulse duration is either 800 fs or, by addition of a fiber-prism compressor, 250 fs. The ASE content of the dye oscillator–amplifier system is as low as 0.1%. The system is used to study the short pulse spectral and temporal gain characteristics of the XeF(C → A) amplifier. A saturation energy density of 50 mJ/cm^2 for 250-fs pulses and a gain bandwidth of 60 nm were measured for the XeF(C → A) excimer transition. Generation of terawatt pulses (275 mJ, 250 fs) with the XeF(C → A) excimer amplifier was demonstrated.

10.1 Introduction

In recent years there has been an ever growing interest in the development of laboratory-scale, ultrahigh-power short-pulse laser systems [10.1–10]. However, an optimal solution to some of the fundamental problems encountered in high-power optical amplifiers still needs to be found. In laboratory-scale systems, terawatt levels can only be achieved by using short pulses. In the short-pulse regime, however, the extractable energy density is limited by the saturation energy density of the amplifying medium.

Up to now there are two fundamental approaches in solving this problem: one can either use active media with low saturation density (such as XeCl and KrF excimers with typical saturation densities in the mJ/cm^2 range) but with the potential of building large-cross-section modules (up to 1000 cm^2), or one can choose an active medium with a large saturation density and relatively small cross section.

The first approach is successfully used by several groups [10.1–7], and peak powers as high as 4 TW have been generated [10.7]. For the second approach, the most promising candidates are solid-state media with saturation densities in the J/cm^2 range. This technique, however, originally suffered from the inherent large optical nonlinearities of solid-state media, which can cause catastrophic self-focusing at high intensities. However, a breakthrough was achieved by the

resulting in the generation of 20 TW of output power [10.10].

This paper describes an alternative approach to high-power pulse amplification that employs a XeF(C–A) excimer amplifier. This amplifier has a saturation energy density of more than an order of magnitude larger than conventional UV excimers, but which is also scalable to large amplifier apertures. After 10 years of development, the XeF(C–A) technology has reached maturity, and a Joule-class amplifier module with nanosecond injection control has been demonstrated [10.11]. This makes it possible to study the gain characteristics and the optimization of energy extraction of a subpicosecond XeF(C–A) amplifier [10.12].

The highly repulsive lower state of the XeF(C–A) excimer transition results in two properties which are highly desirable for short-pulse, high-power amplification. The first one is that the saturation energy density, calculated from the stimulated emission cross section reported in [10.13], is expected to be 50 mJ/cm^2. The other, equally important property of the XeF(C–A) transition is that, compared to conventional excimers, it has an extremely broad (60 nm) gain bandwidth which theoretically is sufficient to support pulse durations as short as 10 fs.

Up to now, apart from our recent investigation [10.12], neither the saturation characteristic nor the gain bandwidth has been studied by direct short pulse measurements. Both for saturation studies and for energy extraction measurements, a subpicosecond, blue-green dye laser system is required. The requirements for such a laser system are rather stringent. For XeF(C–A) saturation measurements, because of the expected large saturation values, injection fluences in the range of 150 mJ/cm^2 are needed. This, however, cannot be achieved simply by tight focusing because the Rayleigh length of the probe beam should be larger than the 50-cm-long excimer amplifier cell. Therefore, the dye laser system should be able to generate subpicosecond pulses with energies in the mJ range. For the energy extraction experiments, because of the low single-pass gain, a similarly high-pulse energy is necessary. In addition, the level of the amplified spontaneous emission (ASE) from the injection laser should be extremely low. To meet these requirements a high-power subpicosecond dye laser system has been developed [10.14].

10.2 Dye Laser System

10.2.1 Oscillator Section

The schematic diagram of the dye laser system is depicted in Fig. 10.1. The oscillator was a modified blue-green Coherent 702 mode-locked dye laser, synchronously pumped by the third harmonic of a CW mode-locked Nd:YAG laser (Coherent Antares 76-s). The original pump system used KTP and BBO

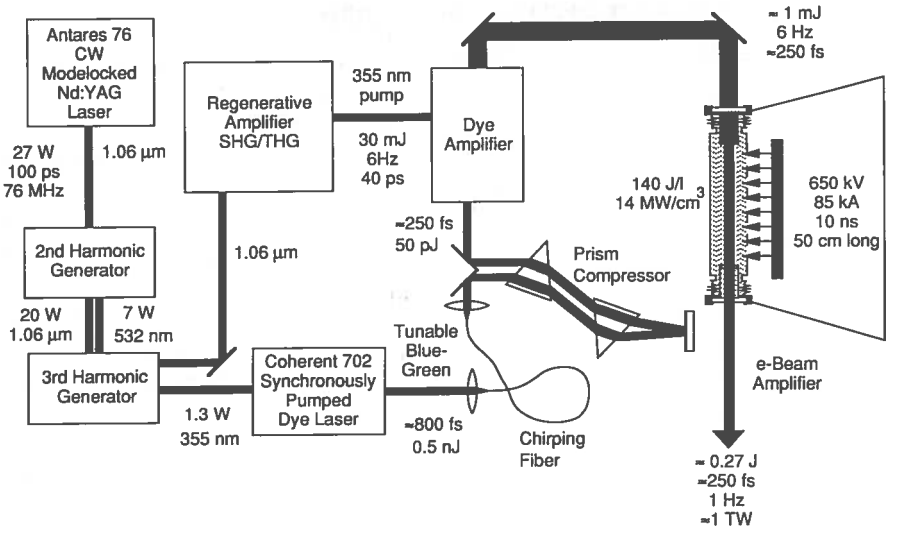

Fig. 10.1. Schematic diagram of the subpicosecond blue-green dye laser system

crystals for frequency doubling and tripling, respectively, and provided 1 W of output power at 355 nm. Since this is only slightly above the threshold (800 mW) of the dye laser, the stability was not satisfactory. By replacing both nonlinear crystals by LBO crystals, the system performance was improved significantly to yield a pump power of 1.3 W at 355 nm. The oscillator then was capable of generating stable 800-fs pulses, tunable from 465 to 515 nm when using coumarin 480 and DOCI as an amplifier and saturable absorber dyes, respectively. The average power of the dye laser oscillator at a repetition rate of 76 MHz was 20–40 mW, depending on wavelength.

The output from the oscillator was amplified either directly, or after being sent through a fiber and prism compressor. In the compressor section, the pulse coming from the oscillator was chirped by a 92.7-cm-long polarization-preserving fiber designed for the blue-green spectral region (Newport F-SPA), and compressed by a two-prism system. The prismatic compressor consisted of two 60° SF-14 prisms used in a double-pass arrangement. The optimum prism separation was found experimentally to be 95 cm. When operating under optimal conditions, the compressor produced 250-fs pulses. It is important to note, however, that the 250 fs should be considered as an upper bound for the pulse duration, because the BBO crystal, used in the multiple-shot background-free autocorrelator (Inrad 514-A), was 0.5 mm thick, which may lead to apparent pulse broadening in the autocorrelation measurements [10.15]. The tuning range was restricted to above 488 nm by the cutoff wavelength of the fiber. The overall transmission of the compressor (fiber + prisms) was 20%.

For pulse amplification to the millijoule level, we have designed a special dye amplifier system. In this system, contrary to previous millijoule subpicosecond amplifiers that have generally relied on a three-stage configuration [10.16], we used a two-stage design. In systems operating in the red, a three-stage design can be afforded because the ASE can be suppressed by insertion of saturable absorbers in the amplifier chain. In the blue-green region, where no good saturable absorbers are available, the only way for ASE suppression is spatial filtering. However, spatial filtering can effectively be used only once, because after the first spatial filter the divergence of the signal and the ASE is the same. Another conceptual point of our design was to use short pump pulses for the amplifiers because in this case the ASE can be further decreased by proper synchronism between the pump and signal pulses.

The two-stage amplifier system presented here can be used in any application provided that the input energy is high enough to drive the first amplifier into saturation. The minimum necessary energy can be estimated as follows: our experimental experience supports the assumption that the smallest practical diameter and highest achievable gain at the first cell is 0.1 mm and 10^5, respectively. Then, allowing for 10 mJ/cm^2 output energy density, which is about 5 times higher than the saturation energy density of typical dyes and represents a good practical compromise between pulse broadening and energy extraction efficiency [10.17], the minimum input energy is 0.01 nJ. This obviously covers all kinds of practical dye lasers.

The dye amplifier was pumped with 40-ps-long, frequency-tripled pulses of a Nd:YAG regenerative amplifier which was seeded by the residual fundamental beam after frequency tripling of the CW mode-locked Nd:YAG laser. The regenerative amplifier (Continuum Corp. RGA-69) was configured with a closed cavity amplifier and one additional single-pass module. This system produced 250-mJ, 1064-nm pulses at 6 Hz repetition rate and pulses of 40 mJ energy at 355 nm after third-harmonic generation.

The experimental layout of the subpicosecond dye laser and amplifier can be seen in Fig. 10.2. The beam waist of the input dye laser was transformed by a 450-mm achromatic lens to match the cross section of the first amplifier stage. The first amplifier stage consisted of an 8-mm-long, dye flow cell pumped by 10% of the total pump energy via an adjustable delay line. The diameter of the dye laser beam at the first cell was 0.2 mm, corresponding to an injected energy density of 2×10^{-7} J/cm^2 (without pulse compressor). The output energy of the first stage was 25 μJ, representing an output energy density of 20 mJ/cm^2. To minimize pulse broadening, care was taken to operate both amplifier stages at about the same level of saturation.

ASE suppression was accomplished by spatially filtering the output of the first stage with an achromatic telescope of 10 times magnification and a 50-μm pinhole as depicted in Fig. 10.2. The telescope expanded the dye beam to fill the 2-mm aperture of the dye flow cell of the second amplifier. The construction of

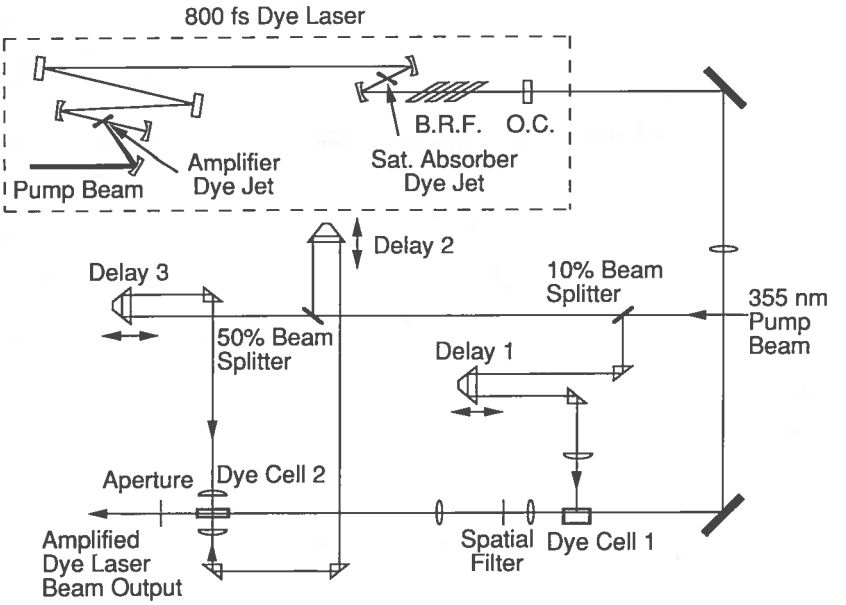

Fig. 10.2. Experimental layout of the dual jet dye laser and its two-stage dye amplifier. BRF: birefringence filter, OC: output coupler

the dye cell allowed symmetrical, double-sided transverse pumping to achieve a uniform energy deposition in the dye. The pump beam for the second stage was divided into two equal parts which could be adjusted by independent delay lines. Two independent delay lines were required, because the pump timing sensitivity was measured to be 10 ps. Hence, in this design the use of Bethune type prismatic dye cells [10.18] should be avoided. At the amplifier output the off-axis ASE was removed by means of an aperture.

The dye oscillator–amplifier system delivered more than 1 mJ pulse energy in the range from 470 to 510 nm, with a maximum of 2 mJ at around 505 nm. The output energy in the 470–480 nm region is due to the lower performance of the coumarin 480 dye, since in this range the oscillator provided 2 times higher input signals to the amplifier than around 500 nm.

While the pulse duration of the oscillator was continuously monitored by a multiple-shot autocorrelator, the amplified output was studied by a single-shot, phase-resolved autocorrelator [10.19]. The pulse duration measurements revealed that no significant pulse broadening in the dye amplifier occurs. The best fit for the autocorrelation trace was achieved by assuming an asymmetric exponential pulse shape with a rise/fall ratio of 1:5 and a pulse duration of 800 fs.

The spatial far field beam profile was close to Gaussian with a slightly elliptical shape in horizontal direction. The divergence was nearly diffraction-limited.

amplifier dropped to about 20%, but since the amplifier system was operating in saturation, this resulted only in a less than 50% decrease in output energy. The highest output energy with 250-fs pulses was 1.2 mJ.

As mentioned earlier, the ASE content of the dye laser pulses is a critical issue for the XeF(C–A) studies. For a measurement of the ASE level, the output of the dye laser was spectrally dispersed so that the spectrum filled the cathode of a photodiode. The injected dye laser beam was then separated from the ASE by a beam stop in front of the diode. Using this method, the ASE was found to be less than 0.1% of the output energy.

In order to measure the XeF(C → A) gain with picosecond pulses, a simple dye laser oscillator was designed that consists of a Littrow grating, a transverse-flow dye cell and an output coupler. The oscillator and a subsequent dye amplifier stage were pumped by the 40-ps-long, third-harmonic output of the regenerative amplifier used for the femtosecond dye laser system described above. This laser produced pulses of ~ 100 ps duration and 2 mJ maximum energy and, by using different dyes, the entire XeF(C → A) spectrum from 450 to 530 nm could be probed.

10.3 XeF(C–A) Experiments

10.3.1 Experimental Arrangement

A detailed description of the XeF(C → A) excimer laser system is given in [10.11]. The XeF(C–A) excimer gas mixture consisted of 12 Torr Xe, 1 Torr F_2, 12 Torr NF_3, 750 Torr Kr and 4100 Torr Ar, which was transversely excited by energetic (650 keV), short duration (10 ns) electron beam pulses. All optical elements were made of fused silica to reduce induced absorption effects [10.13], except for the 0.375″ thick excimer chamber windows, for which MgF_2 was chosen to avoid self-focusing.

For gain measurements, the dye laser injection signal was sent in a single pass through the excimer gas cell, attenuated by neutral density filters to probe different energy densities. The laser input and output energies were monitored simultaneously by vacuum photodiodes, calibrated relative to each other before gain measurements. The photodiodes were also used to monitor the relative timing between the injection signal and the electron-beam excitation. Additionally, the energy of the amplified pulses was observed by a pyroelectric energy meter. The output beam profile at the gas cell window was imaged by a lens onto a CCD array and recorded for each pulse.

10.3.2 Gain Measurements

A spectrum of the free-running XeF(C → A) excimer laser [10.20] is shown in Fig. 10.3. The broadband laser spectrum is interrupted by several narrow-band

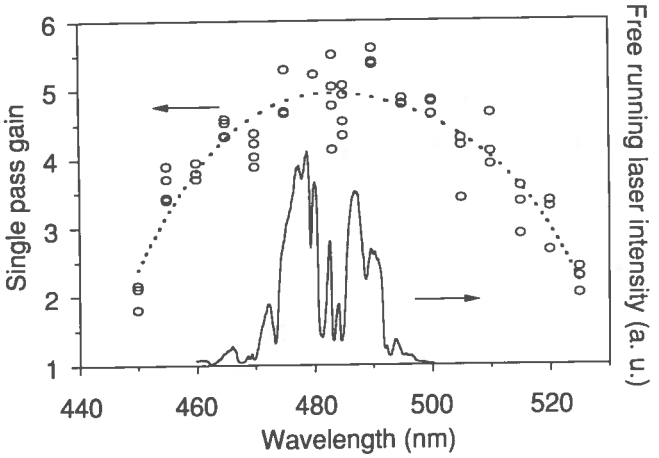

Fig. 10.3. Spectral dependence of the XeF(C → A) excimer single-pass gain for injection pulses of ~ 100 ps duration. The gain length is 50 cm, the injected energy density is 1 mJ/cm^2. The spectrum of the free running, nanosecond laser is shown for comparison. The gain spectrum shows a smooth profile with no apparent influence of narrow-band transient atomic absorbers, visible in the laser spectrum

absorption lines due to excited argon, krypton and xenon atoms created as by-products of the electron beam excitation [10.21]. These absorbers limit the gain bandwidth and therefore restrict amplification of ultrashort pulses. However, if it is possible to saturate these absorbers the full bandwidth is available for amplification. Most of the absorbing species have a larger cross section than the stimulated emission cross section of the XeF(C → A) transition and therefore saturate at smaller energy densities than the gain. This effect has been demonstrated in nanosecond gain measurements at a narrow-band absorber wavelength, where the gain actually increases with higher injected intensities [10.21].

In an attempt to characterize the bandwidth of the XeF(C → A) excimer with short pulse injection, which was investigated earlier for nanosecond pulses [10.20], the gain was measured with 100-ps pulses over a wide spectral region. The single pass gain for 100-ps, 1-mJ/cm^2 pulses, also shown in Fig. 10.3 is characterized by a smooth profile over a bandwidth of 60 nm. Similar to the results obtained with 800-fs pulses, it is also possible with 100-ps pulses to reach an energy density sufficiently high to saturate narrow-band absorbers and to make the entire XeF(C → A) bandwidth accessible to tunable high power generation or ultrafast, large bandwidth amplification.

The dependence of the gain for 250-fs, 800-fs and 100-ps pulses is shown in Fig. 10.4. The gain measurements were performed in a single pass through the 50-cm-long active gain medium at 490 nm, which corresponds to a maximum of the free-running XeF(C → A) laser spectrum. The single-pass gain for the 100-ps and 800-fs pulses was measured to be 5.7, which translates to a gain coefficient of 0.034 cm^{-1}. Using the Frantz–Nodvik model [10.22] the saturation energy

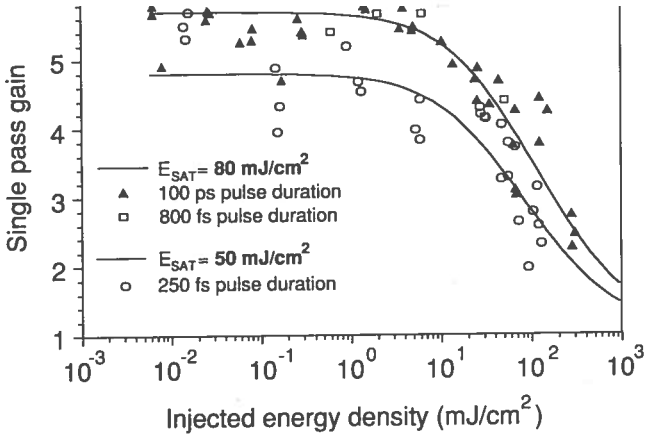

Fig. 10.4. The dependence of single-pass gain of a 50-cm-long XeF(C → A) excimer amplifier for 250 fs, 800 fs and ~ 100 ps pulse durations on the injected energy density at a wavelength of 490.5 nm. The Frantz–Nodvik curves were fitted taking into account beam-broadening effects

density was calculated to be 80 mJ/cm^2 for both pulse durations. The identical saturation behavior for both pulse durations implies that within the experimental accuracy no significant repumping of the gain occurred on a 100 ps time scale.

Measurements with 250 fs probe pulses showed a slightly lower small-signal gain of 4.8, corresponding to a gain coefficient of 0.032 cm^{-1} and a saturation energy density of 50 mJ/cm^2. A decrease of these gain parameters for 250-fs pulses was expected, since the rotational reorientation time of the XeF(C) molecule, estimated to be approximately 0.8 ps prevents complete utilization of the gain medium [10.12].

10.3.3 Energy Extraction

Energy extraction from the XeF(C → A) excimer amplifier for 250-fs, 490-nm pulses was investigated using a specially designed unstable resonator. The unstable resonator was of the positive-branch, confocal type with a magnification of $M = 4$. The injected beam made a total of five passes through the gain medium, resulting in a gain of ~ 1000. The dependence of the output energy of the XeF(C → A) amplifier upon the injected energy is shown in Fig. 10.5. A maximum output energy of 275 mJ was obtained with an injection energy of 0.5 mJ. The highest achieved energy was about one quarter of the maximum energy of 1 J observed for nanosecond injection [10.23], which represents a good efficiency for a femtosecond amplifier. The solid line in Fig. 10.5 depicts the output energy calculated by a numerical model, based on the gain saturation

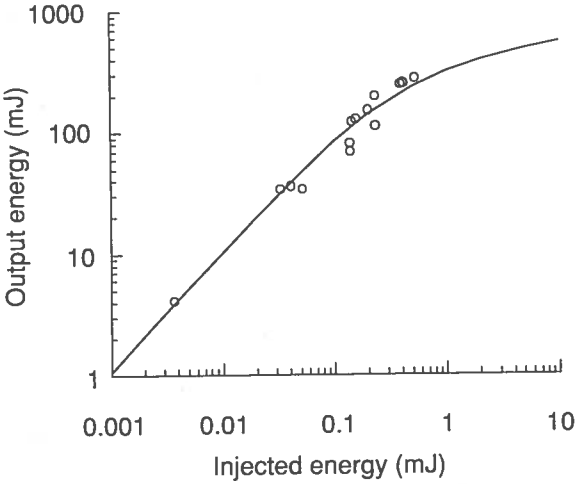

Fig. 10.5. The dependence of the output energy of the XeF(C → A) excimer amplifier on the injected energy for 250-fs pulses at a wavelength of 490.5 nm. The solid line is calculated from gain saturation measurements

and gain lifetime [10.13] measurements. It is apparent that the amplifier is driven into moderate saturation for maximum output, resulting in good energy extraction efficiency. Deep saturation, however, was avoided in order to prevent temporal pulse broadening.

An upper level for the ASE energy was measured at a distance of 4 m by blocking the injection dye laser. The detected energy was smaller than 1 mJ, corresponding to an ASE level of $< 0.4\%$. Further reduction of the ASE level can be achieved by spatial and spectral filtering of the amplified beam and by the use of a saturable absorber. The autocorrelation measurements of the injected and amplified pulses showed large shot-to-shot fluctuations of the pulse duration. Minimum pulse durations of 250 fs after amplification were recorded, indicating no systematic temporal pulse broadening by the amplifier. The 250 fs pulse width should again be considered as an upper bound for the pulse duration because the BBO crystal used in the single-shot autocorrelator was 1 mm thick. Furthermore, contrary to the multiple-shot autocorrelators, the exact effect of the crystal length on the measured pulse duration has not yet been worked out for the single-shot case.

The focusability of the amplified beam was measured by observing the focal spot of a 4-m lens with a CCD camera. A $1/e^2$ spot diameter of 160 μm for the unamplified beam was measured, which corresponds to a diffraction-limited beam taking into account the top hat, torus-shaped beam profile. After amplification the beam focal spot increased to 200 μm or 1.3 times the diffraction limit. This increase is presumably due to the intensity variation in the near-field profile, created by the single-sided transverse electron-beam excitation geometry and the resulting gain gradient. Compared to a Gaussian beam of the same $1/e^2$ beam diameter of 4 cm, the amplified beam exhibits a 3 times larger focal spot diameter. Assuming the measured beam quality and an output power of 1 TW,

more than 10^{18} W/cm^2.

10.4 Conclusions

A 250-fs dye laser-amplifier system was constructed for the blue-green spectral region. A two-stage amplifier design, incorporating a spatial filter, was used to generate tunable 1.2 mJ laser pulses with less than 0.1% of ASE.

The gain of an electron-beam-pumped XeF(C → A) amplifier has been measured for 250-fs, 800-fs and 100-ps pulses. The small signal and saturation behavior for both the 800 fs and the 100 ps measurements was identical to within the measurement accuracy, giving no indication of gain repumping on a 100 ps time scale. The single-pass gain for both pulse durations was 5.7, corresponding to a gain coefficient of 3%/cm. The saturation energy density was 80 mJ/cm^2. For 250-fs pulses the gain coefficient was reduced by 10% and the saturation energy density decreased to 50 mJ/cm^2, believed to be related to the rotational reorientation time of the XeF(C → A) molecule of approximately 0.8 ps. The saturation energy density of 50 mJ/cm^2 for 250-fs pulses is about 30 times larger than for conventional excimer amplifiers, allowing the design of more efficient and compact high-power amplifiers. Narrow-band absorbers in the XeF(C → A) spectrum can be saturated, resulting in a smooth gain profile. A gain bandwidth of 60 nm centered at 480 nm was demonstrated, which can be used either for amplification of extremely short or broadly tunable laser pulses.

Amplification of 490-nm, 250-fs pulses yielded a maximum energy of 275 mJ and laser powers in the terawatt range. An upper limit of 0.4% was found for the ASE energy level. The beam quality of the amplified pulses was determined as 1.3 times diffraction-limited, considering the torus-shaped beam profile.

The amplification of 250-fs pulses with $\sim$ 2 nm bandwidth made use of only a fraction of the XeF(C → A) gain bandwidth of 60 nm. It is expected that pulses of much shorter duration, such as the blue-green 10-fs pulses demonstrated by *Schoenlein* et al. [10.24] or frequency-doubled, mode-locked Ti:sapphire pulses can be amplified in this excimer system, possibly further increasing the peak output power. In fact, modeling of the XeF(C → A) transition suggests that both the gain and the saturation energy density do not change significantly for injection pulse durations as short as 50 fs [10.25]. Scaling of the XeF(C → A) excimer system has been demonstrated successfully for nanosecond systems [10.11], and therefore the design of electron-beam-pumped, large-aperture systems, as demonstrated for the KrF excimer [10.7] should also be applicable to the XeF(C → A) excimer amplifier, increasing the performance of this system even further.

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