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Tunable High Power Excimer Lasers Physics and Technology

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– ABSTRACT –

The development of scalable high power lasers in the UV-visible range and ultrashort high brightness laser sources impact a number of key technologies. Experiments of scaling the e-beam pumped XeF (C → A) laser system to the 1 Joule / pulse output level at a 1 Hz repetition rate are described. Recent progress in the amplification of tunable ultrashort laser pulses in the visible spectrum, utilizing the broadband XeF (C → A) excimer transition, is also reported.

1. SCALING CHARACTERISTICS OF THE XeF(C → A) EXCIMER LASER

Excimer lasers play an important role in specific applications that utilize their unique characteristics such as high average power for material processing, and high spectral purity and broad band tunability for remote sensing, optical communication and basic physics research. There is considerable interest in the XeF (C → A) excimer laser as an efficient, tunable source of radiation in the blue-green region of the spectrum. Using an electron beam as an excitation source and a kinetically tailored five component gas mixture,[1] efficient operation of this laser system has been demonstrated for short pulse 10 ns (10 MW /㎤),[2] intermediate pulse 250 ns (~1 MW /㎤),[3] and long pulse 700 ns (~250 kW /㎤)[4] electron beam pumping durations. When the short pulse, high current density electron beam excitation technique is employed, peak values of small signal gain exceed 3% cm^{-1}, permitting efficient injection controlled operation. This results in a very effective method for wavelength tuning. Continuous tuning between 450 and 530 nm with a linewidth as narrow as 0.001 nm and good spatial beam quality (3× diffraction limit) have been demonstrated. An output energy density of ~1.7 J / l with an intrinsic efficiency of 1.3% has been achieved for wavelengths between 470 and 510 nm.[5]

With its gaseous active medium, the XeF (C → A) excimer laser is readily scalable to the high energy and power required for numerous potential applications. Experiments have been performed that have led to the successful scaling of an electron beam pumped XeF (C → A) laser from an active laser volume of ~0.02 l to one of ~0.51 l with a pumped length of 50 cm^6. An improved output energy of 1.2 J / pulse in repetitive operation of up to 1 Hz, was achieved with a large aperture square unstable resonator geometry, using a compact, halogen compatible, closed flow loop incorporating a transverse in-line fan for gas circulation.

The electron beam generator, described in detail elsewhere,[6,8] produces a 650 keV, 90 kA pulse in 10 ns (FWHM) with a current density of 250 A / ㎠. The generator was specifically designed for this application and is capable of repetitive operation at up to 1 Hz. The electron beem enters the laser chamber transversly through a 25 um titanium foil anode coated with a 5 um layer of ion vapor deposited aluminum to prevent its interaction with the reactive fluorine in the laser gas mixtures. A 0.2 Tesla magnetic guide field provides a three-fold increase in the electron beem current density that is delivered at the optical axis of the laser chamber. The electron beem energy deposition into the active medium was measured by using chlorostyrene film dosimetry. The averaged spatial energy density for the cross-section of the resonator volume was measured to be ~110 J / l.

The optical cavities investigated in these experiments were positive branch confocal unstable resonators. A resonator magnification of 1.67 was chosen for optimized performance in a mixture consisting of 1.3 mbar F_2, 16 mbar NF_3, 16 mbar Xe, and 1 bar Kr and completed to a total pressure of 6.5 bar with Ar as the buffer.[6] The plano concave total reflector (radius of curvature of 3.0 m) had a highly reflective coating (R>99.9%) and an AR coating deposited on the concave and the flat surface, respectively. An injection seed laser beem entered the resonator through a 1.5㎜ diameter hole in the high reflectivity coating located at the center of this reflector on the concave surface. The output reflector consisted of a meniscus lens with radii of curvature ±1.8 m and a 2.8㎝ × 2.8㎝ square reflective coating centered on the convex surface. These mirrors also had fluorine protective coatings and were mounted directly on the ends of the laser cell in contact with the laser gas mixture spaced 60 ㎝ apart. A XeCl excimer laser pumped dye laser provided a 40 ns (FWHM) long injection pulse which resulted in good temporal overlap with the gain duration (~20 ns) in the electron beam pumped amplifier. The seed beam diameter was adjusted with a telescope to achieve uniform injection and the typical peak injection intensity coupled through the hole corresponded to ~3 MW / ㎠. In principal, it is possible to use other more compact external or self injection schemes. One such proposed scheme that is being presently investigated is the use of on appropriately tailored gas mixture to generate both KrF (B → X) and XeF (C → A) emission and the generation of the fifth Stokes component of KrF (B → X) at 514 nm in H_2, which can then control the XeF (C → A) laser output.

The output energy of the injection controlled XeF (C → A) amplifier was monitored by a pyroelectric detector, and the temporal pulse duration, typically 10 ns in (FWHM), was detected by a fast vacuum photodiode. The spectral characteristics were measured by a 0.25 m spectrometer with an optical multi-channel analyzer (OMA). In single laser short operation, the timing between the electron beam and injection dye laser was carefully adjusted to obtain optimum laser pulse energy stability. A maximum energy per pulse of 1.2 J was extracted at an injection wavelength of 486.8 nm using the square unstable resonator geometry. Repetitive laser performance is shown in Fig.1 which depicts the signal from a pyroelectric energy monitor for 10 shots in a 1 Hz sequence. The output energy remained constant throughout the 1 Hz series at the same output level as observed under single shot operation, without shot-to-shot energy degradation due to thermal turbulence. Thus, both the high laser energy output and repetitively pulsed operation were demonstrated for a scaled electron beam pumped XeF (C → A) laser amplifier system.

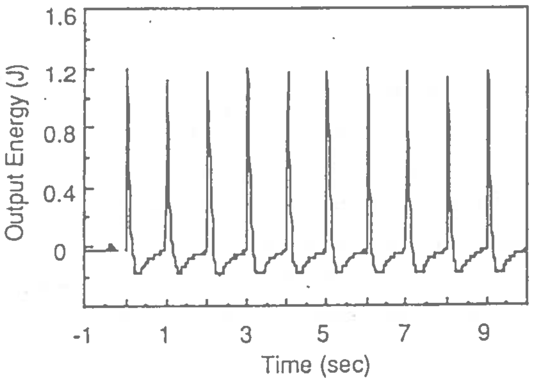

Fig. 1 *XeF(C→A)* laser output energy. The trace depicts 10 individual shots obtained in a 1 Hz sequence. The wavelength and spectral width are 486.8 nm and 0.005 nm, respectively.

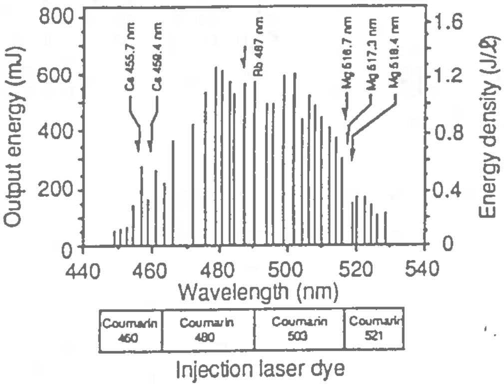

Fig. 2. Wavelength tuning characteristics of the injection controlled *XeF(C→A)* laser.

Shown in Fig.2 is the output energy of the XeF (C → A) injection controlled laser of wavelengths between 450 and 530 nm chosen not to coincide with known narrow band atomic absorbers. The injection dye laser had a linewidth of 0.005 nm, and the data were normalized to an injection intensity of 2 MW/cm^2. A tuning bandwidth of ~60 nm (FWHM), centered at 490 nm, was observed for this injection intensity with a peak specific energy density of 1.3 J/l. A maximum laser output of 0.7 J was measured at 486.8 nm with an intrinsic efficiency of 1.2 %. Even in the wings of the gain profile, at 450 and 530 nm, an energy density of 0.2 J/l was obtained with complete spectral control of the laser output. Also indicated in Fig.2 are the four injection dyes used to span the XeF (C → A) spectral region. The spectral bandwidth of the output of both the injection laser and the XeF (C → A) amplified laser output were measured with an air spaced etalon. The spectral bandwidth of the tuned output from the XeF (C → A) laser was found to completely preserve that of the injection dye laser for linewidth as narrow as 0.001 nm. As illustrated in Fig.2, the accessible spectral regions includes the wavelength of various atomic resonance filters. These results make the XeF (C → A) laser an attractive candidate for applications requiring high-power, wavelength tunability and spectral bandwidth control such as optical communications and remote sensing.

2. ULTRASHORT LASER PULSE AMPLIFICATION IN THE XeF (C → A) GAIN MEDIUM

In recent years there has been an ever growing interest in the developement of laboratory-scale, ultrahigh-power short-pulse laser systems [9-15]. In laboratory scale systems, terawatt levels can only be achieved by using ultrashort pulses. In the ultrashort pulse regime, however, the extractable energy density is limited by the saturation energy density of the amplifying medium.

Up to there are two fundamental approaches in solving this problem : one can either use active media with low saturation density (such as XeCl and XeF excimers with typical saturation densities in the mJ/cm² range) but with the potential of building large cross-section modules (up to 1000 cm²), 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 [9-13], and peak powers as high as 4 TW have been generated [16]. For the second approach, the most promising candidates are solid-state media with saturation densities in the J/cm² range. This technique, however originally suffered by inherent large optical non-linearities of solid-state media, which cause self-focusing at high intensities. However, a breakthrough was achieved by the introduction of chirped pulse amplification (CPA) to these systems [14], resulting in the generation of 20 TW of output power [15]. This paper describes an alternative approach to high power pulse amplification that employs a XeF (C → A) excimer amplifier.

The highly repulsive lower state of the XeF (C → A) excimer transition results in two properties which are highly desirable for ultrashort-pulse, high power amplification. The first one is that the saturation density, calculated from the stimulated emission cross-section, is expected to be 50 mJ/cm². The other, equally important property of the XeF (C → A) tansition 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 [16].

Both, for saturation studies and for energy extraction measurements, a subpicosecond, blue-green injection laser is required. For XeF (C → A) saturation measurements, because of the expected largre saturation values, injection fluences in the range of 150 mJ/cm² are needed. This, however, cannot be achieved simply by tight focussing because the Rayleigh length of the probe beam should be larger than the 50 cm long excimer amplifier cell. Therefore, the injection laser system should be able to generate subpicosecond pulses with energies in the mJ range.

The schematic diagram of the experimental set-up consisting of a dye laser and XeF (C → A) amplifier system is depicted in Fig. 3. The oscillator was a modified blue-green Coherent 702 mode-rocked dye laser, synchronously pumped by the third harmonic of a CW mode-rocked Nd-YAG laser (Coherent Antares 76-S). The oscillator 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 [17]. 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 92.7cm 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 seperation was found experimentally to be 95cm. When operating under optimal conditions, the compressor produced 200 fs pulses. The tuning range was restricted to above 488 nm by the cutoff wavelength of the fiber. The overall transmission of the cmpressor (fiber + prisms) was 20 %.

For pulse amplification to the milijoule level, a special dye amplifier system was designed. In this dye amplifier system, contrary to previous milijoule subpicosecond amplifiers that have generally relied on a three stage configuration [18], a two stage design was used. In system operating in the red, a three stage design can be used 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. 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 the CW mode - locked Nd : YAG laser. The regenerative amplifier 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 dye oscillator - amplifier system delivered more than 1 mJ pulse energy in the range from 470 to 570 nm, with maximum of 2 mJ at around 505 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 [19]. The pulse duration measurements revealed 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 :

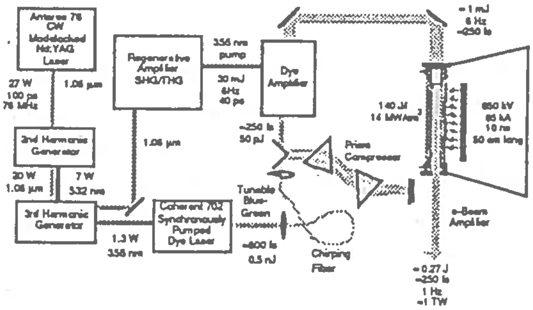

Fig. 3. Schematic diagram of the subpicosecond blue-green dye laser and electrically excited excimer amplifier system.

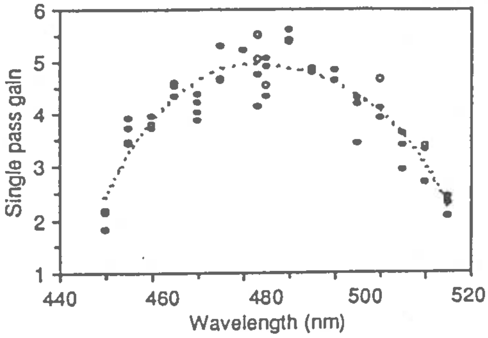

Fig. 4. Spectral dependence of the *XeF(C→A)* excimer single pass gain for injection pulses of ~100 ps duration. The gain length is 50 cm, the injected energy density is 1 mJ/cm^2. The gain spectrum shows a smooth profile with no apparent influence of narrowband transient atomic absorbers.

By insertion of the fiber and prism compressor stage, the input energy to the 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.

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 nm to 530 nm could be probed.

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 photodiode, 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.

In an attempt to characterize the bandwidth of the XeF (C → A) excimer with short pulse injection, which was investigated earlier for nanosecond pulses [5], the gain was measured with 100 ps pulses over a wide spectral region. The single pass gain for 100 ps, 1 mJ/cm² pulses, shown in Fig. 4 is characterizedby a smooth profile over a bandwidth of 60 nm.

The dependence of the gain for 250 fs, 800 fs and 100 ps pulses is shown in Fig. 5. The gain measurements were performed in a single pass through the 50cm 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 $0.034\mathrm{cm}^{-1}$. Using the Frantz - Nodvik model 20, the saturation energy density was calculated to be 80 mJ/cm² for both pulse durations. The identical saturation behavior for both pulse durations implies that within the experimental accuracy no significant repumping of the gain occured 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 $0.032\mathrm{cm}^{-1}$ and a saturation energy density of 50 mJ/cm². A decrease of the saturation energy density 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 [21].

Energy extraction from the XeF (C → A) excimer amplifier for 250 fs, 490 nm pulses was investigated a specially designed unstable resonator. The unstable resonator was of the positive - branch, confocal type having magnification of M = 4. The injected beam made a total of five pass 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. 6. A maximum output energy 0f 275 mJ was obtained with an

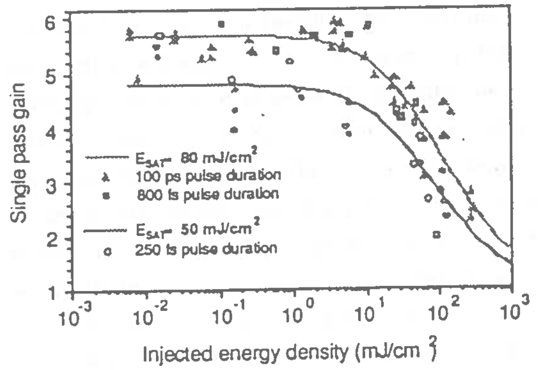

Fig. 5. The dependance of single pass gain of a 50 cm long *XeF(C→A)* excimer amplifier for 250 fs, 800 fs and ~100 ps pulse durations on the injected energy density at a wavelength of 490.5 nm. The Frantz-Nodvik curves were fitted taking into account beam-broadening effects.

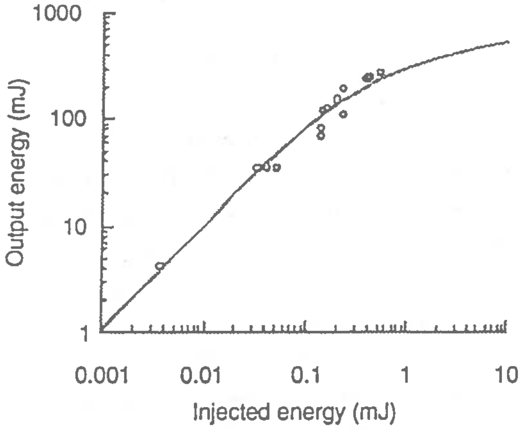

Fig. 6. The dependence of the output energy of the *XeF(C→A)* excimer amplifier on the injected energy for 250 fs pulses at a wavelength of 490.5 nm. The solid line is calculated from gain saturation measurements.

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[7], which represents a good efficiency for a femtosecond amplifier. The solid line in Fig. 6 depicts the output energy calculated by a numerical model, based on the gain saturation and gain lifetime[16] 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 tha 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 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㎜ 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 2000㎜ 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㎝ the amplified beam exhibits a three times larger focal spot diameter. Assuming the measured beam quality and an output power of 1 TW, the use of f/1 parabolic reflector would result in an intensity at the focal point of more than 10^{18} W/㎠.

SUMMARY

Scaling experiments of an injection controlled XeF (C → A) laser pumped by an intense short pulse electron beam have been performed. Using a multi-component high pressure gas mixture comprised of F_2, N_3F, Xe, Kr, and Ar, a specific laser energy output density of 1.7 J/l with an intrinsic efficiency of 1.3% were achieved at 486.8 nm, corresponding to an output energy of 0.8 J. Furthermore, the short pulse electron beam excited XeF (C → A) excimer laser was demonstrated to be continuously tunable between 450 nm and 530 nm with an output linewidth as narrow as 0.001 nm. Improved laser performance of 1.2 J per pulse and, in particular, repetitive operation of up to 1 Hz using a compact, gas fiow (~5.7m/s) system has been achieved.

In addition, the broad gain bandwidth, which is significantly larger than those available from organic dyes and UV excimer lasers, makes the XeF (C → A) system a suitable medium for amplfication of ultrashort laser pulses to the terawatt level in the blue green. The feasibility of developing an ultrahigh brightness laser source in the visible based on the XeF (C → A) transition has been established. Amplification of 490 nm, 250 fs pulses yielded a maximum energy of 275 mJ and laser power in the terrawatt 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 ~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 Schoenlen et al. or frequency doubled, modelocked Ti : sapphire pulses can be amplified in this excimer system, possibly further increasing the peck 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 duration as short as 50 fs. Scaling of the XeF (C → A) excimer system has been demonstrated successfully for nanosecomd system, and therefore the design of electron-beam pumped, large aperture systems, as demonstated for the KrF excimer should also be applicapable to the XeF (C → A) excimer amplifier, increasing the performance of this system even further.

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