Recent Progress of Terawatt Excimer Laser Sources

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Abstract

Subpicosecond pulses first in the visible and more recently at 193 nm have been amplified in an electron-beam excited XeF(C→A) and ArF(B→X) excimer laser amplifier to pulse energies in the hundred millijoule range. Details of the current experimental program and future prospects will be described.

Introduction

In recent years there has been a rapid development of laboratory-scale, ultrahigh peak power, short-pulse laser systems due to their various potential applications such as plasma and x-ray generation on a subpicosecond time scale [1,2]. Current high-power amplifiers are based on ultraviolet excimer or infrared solid state systems, each of them having distinct advantages and limitations. Excimer laser systems have been succesfully used to generate output powers as high as 4 TW in a 390 fs pulse by a KrF excimer amplifier chain [3].

This paper describes recent progress in the amplification of ultrashort laser pulses to the terawatt power level in the visible (490 nm) and VUV region (193 nm) using the XeF(C→A) and ArF(B→X) transitions, respectively.

XeF(C→A) Terawatt Excimer Laser

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 energy density, calculated from the stimulated emission cross-section, 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 [4].

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 large saturation values, injection fluences in the range of 150 mJ/cm^2 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.

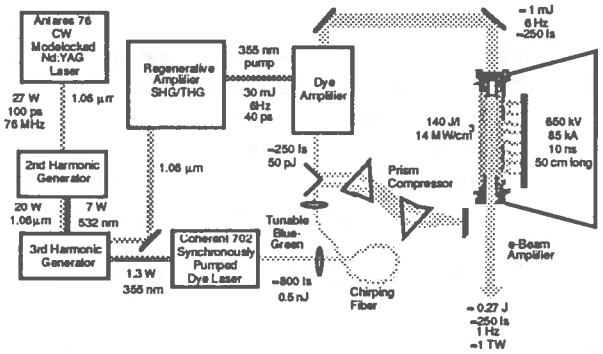

Figure 1. Schematic diagram of the subpicosecond blue-green dye laser and e-beam excited excimer amplifier system.

The schematic diagram of the subpicosecond blue-green dye laser and XeF(C→A) amplifier system is

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depicted in Fig. 1. The primary 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). This 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 [5]. 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. 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 compressor (fiber + prisms) was 20%.

For pulse amplification to the millijoule level, a special dye amplifier system was designed. 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 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 510 nm, with a 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 [6]. 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.

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.

Energy extraction from the XeF(C→A) excimer amplifier for 250 fs, 490 nm pulses was investigated using a specially designed unstable resonator [7]. 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. 2. A maximum output energy of 275 mJ was obtained with an injection energy of 0.5 mJ. A typical autocorrelation trace is shown in Fig. 3. The highest achieved energy was about one quarter of the maximum energy of 1 J observed for nanosecond injection [8], which represents a good efficiency for a femtosecond amplifier. The solid line in Fig. 2 depicts the output energy calculated by a numerical model, based on the gain saturation and gain lifetime [4,7] 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.

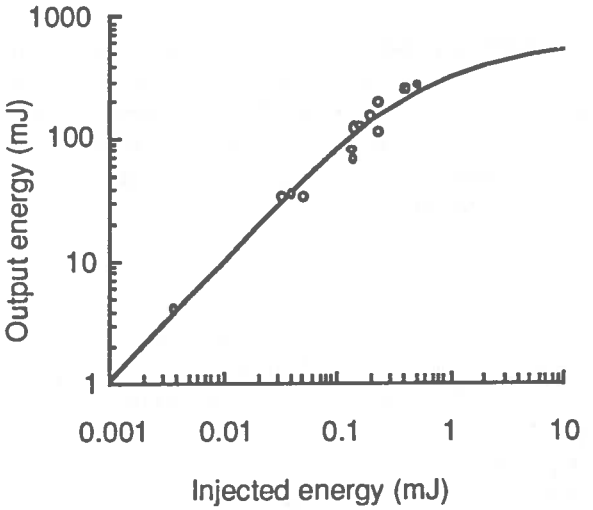

Figure 2. The dependence of the output energy of the XeF(C→A) excimer amplifier on the injected energy for 250 fs pulses at a wavelength of 490.5 nm. The solid line is calculated from gain saturation measurements.

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.

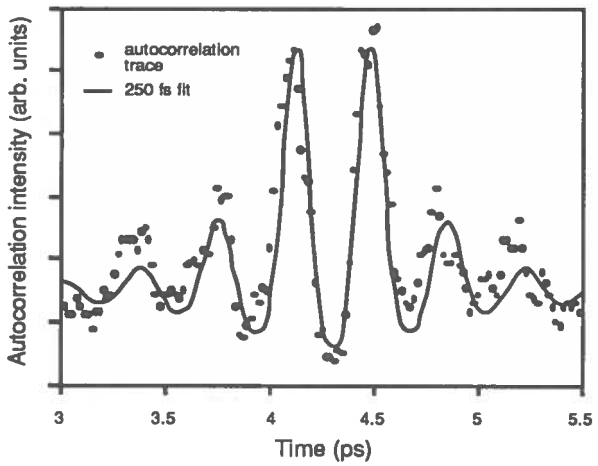

Figure 3. Phase-sensitive single-shot autocorrelation trace of an amplified pulse. The output pulse energy is 200 mJ. The fitted curve represents a 250 fs pulse of double-sided exponential shape.

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 three times larger focal spot diameter. Assuming the measured beam quality and an output power of 1 TW, the use of an f/1 parabolic reflector would result in an intensity at the focal point of more than 10^{18} W/cm^2.

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 (50 fs) from a frequency-doubled, modelocked Ti:sapphire system can be amplified in this excimer system, possibly further increasing the peak output power to the 10 TW level. 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 [9].

Ultrahigh-brightness ArF excimer system

More recently, the feasibility of an ultrahigh-brightness ArF excimer system operating at the 0.1 TW level, based on a discharge pumped preamplifier and an electron-beam pumped power amplifier has been demonstrated. For the generation of subpicosecond injection pulses at 193 nm a spectrally compensated sum-frequency mixing scheme using β-barium borate (BBO) as a nonlinear crystal is utilized [10].

A narrow linewidth UV input pulse at 266 nm and broadband laser radiation at 707 nm are used as input wavelengths for the BBO mixing crystal. The red laser beam is angularly dispersed by means of a grating-lens combination in such a way that every spectral component enters the BBO crystal at an angle that phase matches the 266 nm beam (Fig. 4). Two nanosecond dye lasers, one operating at 707 nm and the other at 532 nm with subsequent frequency doubling have been used in initial experiments.

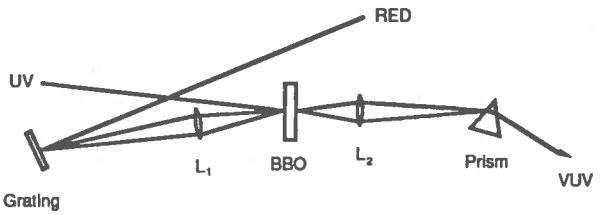

Figure 4. Experimental arrangement for dispersively compensated sum-frequency mixing.

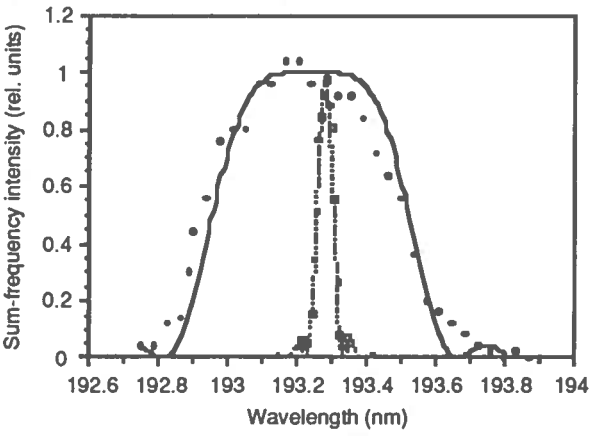

Figure 5. Measured and calculated SFM spectral acceptance of a 1 mm thick BBO crystal, uncompensated (squares and dashed line) and compensated (dots and solid line).

The spectral acceptance of the mixing set-up was measured by tuning the red dye laser. SFM bandwidths are shown in Fig. 5. For comparison the squares show the output of an uncompensated 1 mm thick BBO crystal, while the dots represent the output of the dispersive scheme using the same crystal. Fig. 5 also indicates the corresponding calculated SFM bandwidths by dashed and solid lines respectively. The calculations are in good agreement with the measurements. From

Fig. 5 it is evident that the dispersively compensated scheme increases the uncompensated SFM bandwidth of 0.05 nm to 0.6 nm i.e. by a factor of 12 corresponding approximately to 90 fs minimum pulse duration. This also means that for a given output bandwidth (or pulse duration) the maximum allowable crystal length is 12 times longer than that without compensation. This increase in the useful crystal length is of fundamental importance, since the SFM energy in the low conversion efficiency limit, which is generally valid for ultrashort pulses, is proportional to the square of the crystal length.

For the generation of ultrashort pulses at 193 nm the input signals to the sum-frequency mixer are provided by an amplified hybridly mode-locked dye laser at 707 nm (300 fs) and by fourth harmonic radiation of a Nd:YAG regenerative amplifier with a pulse duration of 50 ps. By using 0.8 mJ and 3 mJ pulse energies at 707 nm and 266 nm, respectively, the pulse energy at 193 nm is 12 μJ.

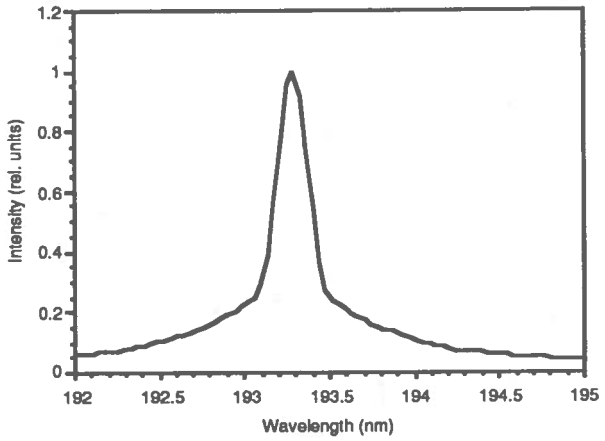

Figure. 6. Spectrum of a subpicosecond pulse at 193 nm.

Fig. 6 shows a spectrum of the generated radiation at 193 nm. The maximum spectral bandwidth of 0.22 nm (FWHM) corresponds to a pulse duration of 250 fs for a Gaussian pulse shape. The substructure in the wings of this spectrum is caused by the OMA used for the measurement of the spectral bandwidth.

This injection source has been used to study the small-signal gain and the saturation energy density of a discharge pumped ArF excimer amplifier module in the subpicosecond regime [11]. The measurement of the ArF excimer gain saturation was performed by injecting the femtosecond seed pulses into a Lambda Physik EMG 101 excimer laser with a gain length of 40 cm. The seed pulses were first preamplified in a single pass and then reinjected into the same excimer laser for the measurement of the gain characteristics. A Frantz-Nodvik fit to the experimental data is shown in Fig. 7. Since nonlinear losses in the window material may distort the measured gain saturation curves, both the input and output energy values are corrected for window transmission. The average small signal gain and saturation energy density is found to be 0.17 cm^{-1} and 3.65 mJ/cm^2. Neglecting the window losses the same data would provide a saturation energy density of 2.3 mJ/cm^2.

An output energy of 8 mJ (ASE < 3%) was measured after double-pass amplification in an off-axis amplification scheme.

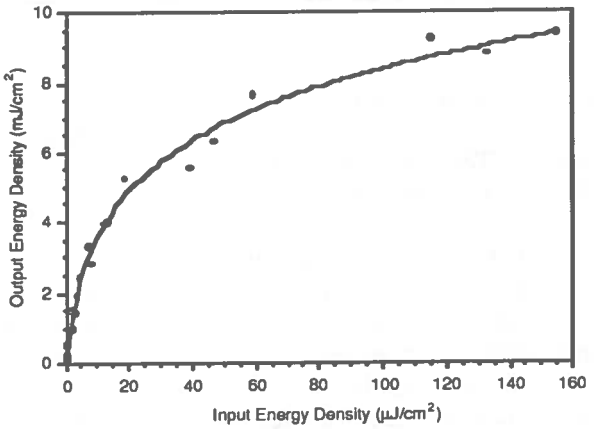

Figure 7. Input-output energy density of an ArF excimer amplifier in the subpicosecond pulse regime.

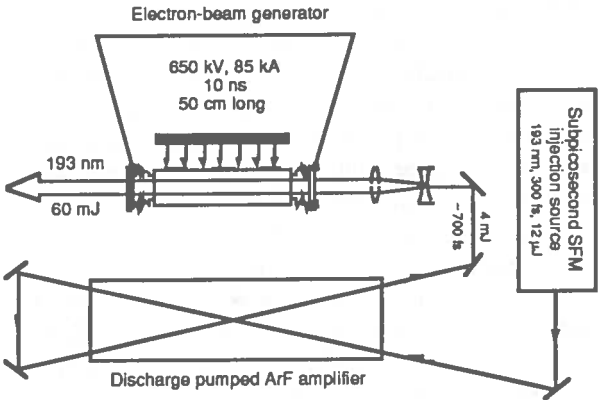

Figure 8. Schematic layout of the ultrahigh peak power ArF excimer laser.

The layout of the high-power laser system utilizing an electron beam pumped ArF excimer amplifier as a power amplifier stage is shown in Fig. 8. The system generates pulse energies of up to 60 mJ at a pulse duration of ~700 fs, with less than 10 % ASE background [12]. Since both the seed pulse generation scheme and the gain bandwidth of the ArF is expected to maintain pulse durations as short as 50 fs, assuming the same output energy density it seems

to be feasible to increase the power up to above 1 TW in future experiments.

Future prospects

Various potential applications such as photochemistry, laser ablation and the generation of powerful XUV and soft x-ray radiation have in particular motivated the development of an ultrahigh-brightness ArF(B→X) (193 nm) excimer system operating at the 0.1 TW level. However, several applications would benefit from short-pulse radiation at even shorter wavelengths. The generation of harmonics [2, 13] as well as four-wave mixing [14] of high-intensity laser radiation in rare gases and hydrogen has been shown to be an effective mechanism for the generation of coherent XUV and soft x-ray radiation. Currently, high-power laser systems in the IR, visible and UV spectral range provide the pump laser radiation for these nonlinear interactions. Due to the low pump-photon energy, the XUV and soft x-ray spectral range is covered only by high-order harmonics with a low conversion efficiency. Consequently, considerable improvement could be introduced by high-power laser systems operating at VUV wavelengths, since the generation of coherent radiation below 100 nm would be possible with low-order nonlinear optical interactions such as third- and fifth-harmonic generation and high conversion efficiency. Suitable gain media for the amplification of picosecond and subpicosecond pulses at VUV wavelengths are electron-beam excited rare gas excimers (Xe_2 (172 nm), Kr_2 (146 nm), Ar_2 (126 nm)) and the F_2 laser transition at 157 nm. As an example, the gain-bandwidth for F_2 is sufficient to support amplification of pulses in the 100-fsec range [15].

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