Generation and Amplification of Subpicosecond ArF Radiation

Th. Hofmann, K. Mossavi, G. Szabó, and F. K. Tittel Department of Electrical and Computer Engineering, Rice University, Houston, TX 77251-1892

ABSTRACT

A dispersively compensated scheme for sum-frequency mixing has been developed to generate subpicosecond injection pulses at 193 nm for subsequent amplification in ArF excimer amplifier modules. Such a scheme is capable of generating 12 μ J at 193 nm with a spectral bandwidth of 0.22 nm corresponding to 250 fs pulse duration by mixing short pulse radiation at 266 nm and 707 nm in a 1 mm BBO crystal. This VUV source has been used to characterize the small-signal gain and the saturation energy density of a discharge pumped ArF excimer amplifier.

1. INTRODUCTION

Currently, there exists considerable interest in the development of high power ArF excimer lasers. Intense coherent radiation at 193 nm is useful in various potential applications, such as photochemistry, lithography and the generation of XUV radiation by nonlinear optical interactions.

Different methods of generating short pulse 193 nm radiation have been reported. This include sum-frequency mixing (SFM) of UV and IR wavelengths in beta-barium-metaborate (BBO)¹⁻³ and potassium pentaborate,⁴ third harmonic generation in Sr vapor,⁵ and nonresonant,⁶ and near resonant⁷ difference-frequency mixing in xenon. Frequency mixing in gases generally requires very high pump intensities to achieve output energies sufficient for practical applications.

For short pulses, the high dispersion of BBO makes it difficult to achieve efficient SFM in the VUV range. The spectral bandwidth for SFM at 193 nm using input wavelengths of about 266 and 707 nm for a 1 mm thick BBO crystal can be calculated to be about 0.05 nm, by applying the Sellmeier constants reported by Kato.⁸ This bandwidth corresponds to approximately 1 ps minimum pulse duration assuming a Gaussian pulse shape. Thus, to generate 193 nm radiation with pulse durations in the subpicosecond range the crystal thickness should be well below 1 mm, which in turn would result in a significantly decrease in conversion efficiency.¹

Several schemes for efficient frequency doubling ⁹⁻¹² and tripling ¹³ of broadband ultrashort pulses by compensating the chromatic dispersion of the nonlinear crystal by angular spectral dispersion have been reported. The methods described in Ref. 9-12 were developed for frequency doubling, consequently they can not be used directly for SFM. The tripling scheme of Ref. 13 could be used for SFM as well, but its single-beam input design would considerably increase the risk of damage of the dispersive element, and restricting the input energy would limit the nonlinear conversion efficiency.

In this work we present a dispersive scheme especially designed for sum-frequency mixing that increases the spectral acceptance of a given crystal by more than one order of magnitude. This scheme has been used to generate ultrashort seed pulses for ArF excimer amplifier.

2. DISPERSIVELY COMPENSATED SFM

All experimental details discussed below refer to a system that is designed to generate coherent ultrashort radiation at 193 nm by mixing picosecond fourth harmonic pulses (266 nm) from a Nd-YAG laser with femtosecond pulses (707 nm) from a dye laser, (i.e. the UV input pulses are nearly monochromatic while the red input pulses are broadband). This arrangement can be adopted to other combination of wavelengths and pulse durations by using the same design considerations.

The experimental arrangement is depicted in Fig 1. The 266 nm laser beam enters the 1 mm thick BBO crystal (cut for 76° phase matching) directly while the 707 nm beam is directed onto the reflection grating, where it is spectrally dispersed.

400 / SPIE Vol. 1810 Gas Flow and Chemical Lasers (1992)

0-8194-1010-1/93/\$4.00

The surface of the grating is imaged onto the crystal by a lens (L1, f=100 mm) with a magnification that is chosen so that each of the spectral components of the red beam falls on the crystal at an angle that phase matches with the 266 nm beam. The necessary magnification can be calculated by comparing the angular dispersion required by the crystal with that of the grating. By using the Sellmeier constants from Ref. 8 we obtain a crystal dispersion of 9.16 mrad/nm, while the dispersion of the 1200 lines/mm grating used in the experiment is 1.672 mrad/nm, assuming an angle of incidence of 8.75°. (The incidence angle was chosen to achieve the highest efficiency with the given grating.) From these values we obtain 5.5 for the necessary magnification. As a result of the angular dispersion at the input of the crystal, the 193 nm sum-frequency output beam is also dispersed. It is recollimated by means of a lens (L2, f=50 mm) and a 60° fused silica prism set to minimum deviation. The lens is positioned so that the crystal plane is imaged onto the prism with the appropriate magnification to match dispersion. From the angular dispersion of 33.57 mrad/nm at 193 nm and a prism dispersion of 2.707 mrad/nm the required magnification is 12.4.

Imaging the grating onto the crystal and the crystal onto the prism, ensures (as it can be seen by using Fermat's principle) that the optical path is equal for any of the red and VUV spectral components, provided that the lens aberrations can be neglected. This also means that the output pulse duration is expected to be the same as that of the red input pulse. This is, in fact, exactly what is needed in the present experiments, since the system is designed to mix picosecond fourth harmonic pulses from a Nd-YAG laser with femtosecond red pulses from a dye laser system. If, however, the UV input pulse duration were shorter, the scheme could be modified dispersing the UV beam instead of the red beam.

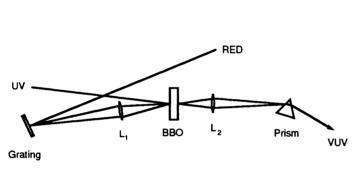


Fig. 1. Experimental arrangement for dispersively compensated sum-frequency mixing.

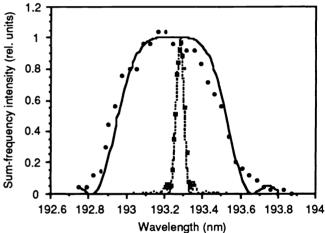


Fig. 2. Measured and calculated SFM spectral acceptance of a 1 mm thick BBO crystal, uncompensated (squares and dashed line) and compensated (dots and solid line).

The performance of the dispersively compensated SFM scheme was tested by simulating the broad bandwidth of the short input pulse with a tunable, narrowband laser. In this case the SFM intensity is measured as function of the (VUV) wavelength while the red laser is tuned, keeping the energy constant. The advantage of this method is that the output bandwidth can directly be measured even if it is larger than that of the presently available shortest pulses at the given wavelength.¹⁴

The measurements have been performed by using two nanosecond dye lasers pumped by the same XeCl excimer laser. One of the dye lasers operated at a constant wavelength of 532 nm with subsequent frequency doubling, while the other was tuned around 707 nm.

The SFM bandwidths are shown in Fig. 2. 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. 2. also indicates

the corresponding calculated SFM bandwidths by dashed and solid lines respectively. The calculations, are in good agreement with the measurements.

From Fig. 2. 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.

3. GENERATION OF SUBPICOSECOND PULSES AT 193 NM

For the generation of ultrashort injection pulses a system as depicted in Fig. 3. has been used. The system consists of a hybridly modelocked dye laser pumped by the second harmonic output of a cw modelocked Nd:YAG laser, generating output pulses at 707 nm with a pulse duration of 800 fs. After pulse compression in a fiber-prism compressor and amplification in a separate dye amplifier we obtained 300 fs input pulses for the dispersively compensated SFM mixer with 1 mJ energy. A fraction of the regenerative amplifier output at 532 nm has been frequency doubled to 266 nm in a KDP crystal and used as the second input beam. 15

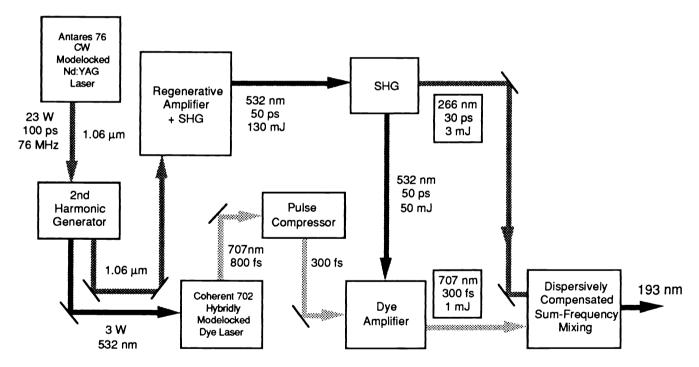
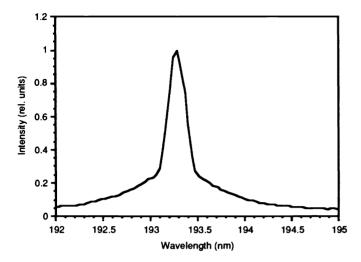


Fig. 3. Schematic layout for the generation of subpicosecond radiation at 193 nm.

Figure 4. shows a typical 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. The pulse energy was typically 12 μJ.

4. AMPLIFICATION OF SUBPICOSECOND PULSES AT 193 NM

We have studied the small-signal gain and the saturation energy density of a discharge pumped ArF excimer amplifier since, to the best of our knowledge, no values for the small-signal gain and the saturation energy density of an ArF excimer amplifier for picosecond and subpicosecond pulses have been published.



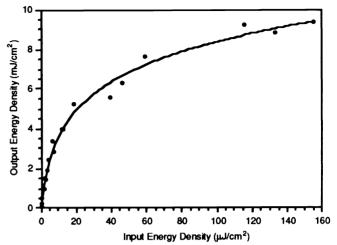


Fig. 4. Spectrum of a subpicosecond pulse at 193 nm.

Fig. 5. Input-output energy density characteristics of a commercial ArF excimer amplifier in the subpicosecond regime. The solid curve represents a Frantz-Nodvik fit for a saturation energy density of 2.3 mJ/cm² and a small-signal gain of 0.15 cm⁻¹.

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 45 cm. After single-pass amplification the output pulses have been reinjected into the amplifier for the measurement of the gain characteristics. The result is schown in Fig. 5. The solid curve indicates a best fit to the experimental data of the Frantz-Nodvik equation for a small-signal gain of 0.15 cm⁻¹ and a saturation energy density of 2.3 mJ/cm². An output energy of 4 mJ was measured after double-pass amplification in an off-axis amplification scheme. However, higher output energies can be expected since only a part of the amplifier aperture has been used in our amplification experiments.

5. SUMMARY

In summary, we have developed a dispersively compensated scheme for sum-frequency mixing which increases the bandwidth of the SFM process more than 10 times compared to the uncompensated bandwidth of the given crystal. By using a BBO crystal we could demonstrate the generation of subpicosecond 193 nm radiation with 12 μ J output energy. A small-signal gain of 0.15 cm⁻¹ and a saturation energy density of 2.3 mJ/cm². has been measured for an discharge pumped ArF excimer amplifier.

In a future experiment the short pulse radiation at 193 nm preamplified in a discharge excited ArF amplifier module will be used as an injection source for an electron-beam pumped ArF excimer amplifier. Such a laser system is expected to yield output energies of 50 mJ when utilizing an amplifier aperture of 15 cm².¹⁷ This would allow the generation of intensities in a focused beam of more than 10¹⁶ W/cm², which would provide the possibility of studying intense laser-matter interactions at 193 nm.

6. ACKNOWLEDGEMENTS

The support of the Robert Welch Foundation grant C-0586 and the Office of Naval Research is gratefully acknowledged. G. S. also acknowledges support from the OTKA Foundation of the Hungarian Academy of Sciences (grant no. 3056).

7. REFERENCES

- 1. S. Szatmári and F.P. Schäfer, "Generation of input signals for ArF amplifiers," J. Opt. Soc. Am. B, vol. 6, pp. 1877-1883, 1989.
- 2. W. Mückenheim, P. Lokai, B. Burghardt, and D. Basting, "Attaining the Wavelength Range 189-197 nm by Frequency Mixing in \(\mathbb{B}\)-BaB2O4," Appl. Phys. B, vol. 45, pp. 259-261, 1988.
- 3. I.V. Tomov, T. Anderson, and P.M. Rentzepis, "High repetition rate picosecond laser system at 193 nm," Appl. Phys. Lett., vol. 61, pp. 1157-1159, 1992.
- 4. R.E. Stickel and F.B. Dunning, "Generation of tunable coherent vacuum uv radiation in KB₅," Appl. Opt., vol. 17, pp. 981-982, 1978.
- 5. H. Egger, T.S. Luk, K. Boyer, D.F. Muller, H. Pummer, T. Srinivasan, and C.K. Rhodes, "Picosecond, tunable ArF* excimer laser source," Appl. Phys. Lett., vol. 41, pp. 1032-1033, 1982.
- 6. J.H. Glownia, M. Kaschke, and P.P. Sorokin, "Amplification of 193 nm femtosecond seed pulses generated by third-order, nonresonant, difference-frequency mixing in xenon," Opt. Lett., vol. 17, pp. 337-339, 1992.
- 7. A. Tünnermann, K. Mossavi, and B. Wellegehausen, "Nonlinear Optical Processes in the Near Resonant Two-Photon Excitation of Xenon by Femtosecond KrF-Excimer Laser Pulses," to appear in Phys. Rev. A, vol. 46, 1992.
- 8. K. Kato, "Second-Harmonic Generation to 2048 Å in β-BaB₂O₄," IEEE J. Quantum Electron., vol. QE-22, pp. 1013-1014, 1986.
- 9. G. Szabó and Z. Bor, "Broadband Frequency Doubler for Femtosecond Pulses," Appl. Phys. B, vol. 50, pp. 51-54, 1990.
- 10. O.E. Martinez, "Achromatic Phase Matching for Second Harmonic Generation of Femtosecond Pulses," IEEE J. Quantum Electron., vol. QE-25, pp. 2464-2468, 1989.
- 11. T.R. Zhang, H.R. Choo, and M.C. Downer, "Phase and group velocity matching for second harmonic generation of femtosecond pulses," Appl. Opt., vol. 29, pp. 3927-3933, 1990.
- 12. R.A. Cheville, M.T. Reiten, and N.J. Halas, "Wide Bandwidth Frequency Doubling with High Conversion Efficiency," accepted for publication in Opt. Lett.
- 13. M.D. Skeldon, R.S. Craxton, T.J. Kessler, W. Seka, R.W. Short, S. Skrupsky and J.M. Soures, "Efficient Harmonic Generation with a Broad-Band Laser," IEEE J. Quantum Electron., vol. QE-28, pp. 1389-1399, 1992.
- 14. Th. Hofmann, K. Mossavi, G. Szabó, and F.K. Tittel, "Spectrally Compensated Sum-Frequency Mixing Scheme for Generation of Broadband Radiation at 193 nm," accepted for publication in Optics Letters.
- 15. T.E. Sharp, C.B. Dane, D. Barber, F.K. Tittel, P.J. Wisoff, and G. Szabó, "Tunable, High-Power, Subpicosecond Blue-Green Dye Laser System with a Two-Stage Dye Amplifier," IEEE J. Quantum Electron., vol. QE-27, pp. 1221-1227, 1001
- 16. S. Szatmári, G. Almási, and P. Simon, "Off-Axis Amplification Scheme for Short-Pulse Amplifiers," Appl. Phys. B, vol. 53, pp. 82-87, 1991.
- 17. C.B. Dane, G.J. Hirst, S. Yamaguchi, T. Hofmann, W.L. Wilson, R. Sauerbrey, F.K. Tittel, W.L. Nighan, and M.C. Fowler, "Scaling Characteristics of the XeF(C→A) Excimer Laser," IEEE J. Quantum Electron., vol. QE-26, pp. 1559-1568, 1990.