Wideband-Tunable High-Power Radiation by SRS of a XeF($C \rightarrow A$) Excimer Laser

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Abstract—**Efficient wavelength shifting by means of stimulated Raman scattering (SRS) in hydrogen and liquid nitrogen of the blue-green XeF($C \rightarrow A$) excimer laser was demonstrated. Energy conversion into the first Stokes line with an efficiency of 38% was achieved. Continuously tunable radiation from 523–579 nm and from 578–650 nm with pulse energies ranging from 100 to 210 mJ was generated in liquid nitrogen and hydrogen, respectively. A peak power of 35 MW at 549.0 nm was obtained.**

I. Introduction

TUNABLE high-power laser sources in the visible spectral region are useful for a number of applications. For a long time, dye lasers were the only lasers available for such applications [1]. The development of tunable solid-state lasers, such as the Ti:sapphire laser in recent years has been a significant advance in this direction. Pulse energies in excess of 200 mJ are now available [2], tunable over a broad spectral range in the near infrared and in the blue–green visible spectrum from Ti:sapphire lasers operating in the fundamental and in second harmonic generation mode, respectively. However, a large portion of the visible spectrum from 530–660 nm remains inaccessible by solid-state lasers. The wavelength region above ~ 570 nm may be accessible in the future by a frequency doubled forsterite laser [3].

In this letter we report on Raman shifting of a tunable (460–525 nm), scalable XeF($C \rightarrow A$) excimer laser [4] to effectively close this gap in wavelength coverage. Using liquid nitrogen and gaseous hydrogen as Raman-active media the first Stokes lines combined cover the wavelength range from 525 to 650 nm.

Although the Raman gain coefficient for nonresonant vibrational scattering is smaller at visible wavelengths than at UV excimer laser wavelengths [5], we demonstrate high Raman gain and efficient wavelength conversion of a blue–green XeF($C \rightarrow A$) laser by utilizing injection-controlled laser operation. Injection control by dye laser results in good spatial beam quality and high intensities in the focused beam [6], which is essential for obtaining a high Raman conversion efficiency.

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Frequency conversion by means of stimulated Raman scattering has a number of advantages over other conversion schemes. High conversion efficiencies of $\sim 40\%$ have been demonstrated with superfluorescent Raman lasers pumped by UV excimer [7]–[9] and frequency doubled Nd:YAG lasers [10]. Dye laser radiation was shifted with 80% energy conversion efficiency in liquid nitrogen [11]. Raman converters are readily scalable and problems with permanent optical damage are avoided due to the use of a gaseous or liquid medium. Wavelength tuning can be accomplished without the need for any adjustment of the Raman-active medium, as compared to phase-matching restrictions in nonlinear optical crystal converters. Therefore, Raman conversion allows direct and efficient generation of tunable radiation.

II. Experimental Arrangement

A superfluorescent Raman laser scheme was chosen for wavelength conversion experiments. A Raman amplifier, using an external Stokes seed signal would yield higher efficiency [12], but at a cost of a more complex laser system.

The experimental arrangement is shown in Fig. 1. The single-shot XeF($C \rightarrow A$) laser was injection controlled by an excimer-pumped dye laser. In order to cover the tuning range of the XeF($C \rightarrow A$) transition, two dyes, Coumarin 480 and Coumarin 503, were used. The dye laser injection energy was typically 2 mJ. The XeF ($C \rightarrow A$) excimer laser was transversely excited by an intense (~ 10 MW/cm^3) electron beam of 12 ns duration [13]. A confocal unstable resonator was used as the laser cavity. Two different laser resonator magnifications were used: $M = 1.33$ and $M = 1.7$. The laser output energy varied between 450 and 750 mJ depending on wavelength. The pump energy in the Raman cells was 78% of the laser energy due to surface losses on windows and lenses in the beam path. The spatial beam profile of the XeF($C \rightarrow A$) laser, equipped with an unstable resonator, has an annular shape. The intensity distribution is graded with a peak to average ratio ~ 2 due to the single-sided transverse electron-beam excitation. The beam quality of the injection-controlled XeF($C \rightarrow A$) laser was determined previously as three times diffraction limited [6]. The linewidth of the XeF laser is 0.2 cm^{-1} [4] which is considerably broader than the vibrational linewidths of hydrogen (0.02 cm^{-1} at 10 bar) [12] and liquid nitrogen (0.07 cm^{-1}) [14].

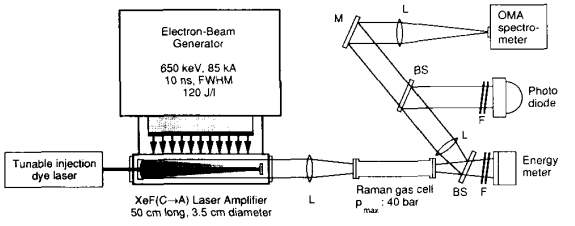

Fig. 1. Experimental arrangement of the XeF($C \rightarrow A$) excimer laser, the Raman gas cell and the beam diagnostics. *L* convex lens, *BS* fused silica beam splitter, *M* mirror, *F* neutral density, and color glass filter.

The energy and the temporal pulse shape of the Raman cell output were measured with a pyroelectric energy meter and a vacuum photodiode, respectively. Bandpass and cutoff filters were used to differentiate between the fundamental and the various Stokes lines. All energy values for conditions of optimized conversion have an accuracy of $\pm 10\%$ and are corrected for losses in beam-steering optics and filters. The spectral profile of the pump beam was recorded by an optical multichannel analyzer (OMA) spectrometer, incorporating a grating with 1800 grooves/mm and having a resolution of 0.2 nm. The same spectrometer equipped with a 300 grooves/mm grating was used to observe the output spectrum of the Raman cells with 1 nm resolution.

Hydrogen and liquid nitrogen were selected for wavelength conversion because of their large Raman gain coefficients ($4 \cdot 10^9$ cm/W for H_2, $2 \cdot 10^{10}$ cm/W for LN_2 [12]). Their vibrational shifts of 4155 cm^{-1} for hydrogen and 2326 cm^{-1} for liquid nitrogen compliment each other to continuously extend the tuning range of the XeF($C \rightarrow A$) excimer laser (~ 2500 cm^{-1}) from 525 to 650 nm. Other Raman-active media yielded conversion efficiencies of less than 15% (methane, deuterium) and 1% (gaseous nitrogen) in our experiments due to their comparably small gain coefficients [5].

III. Conversion in Hydrogen

For wavelength conversion in hydrogen, a gas cell of 62 cm length, 4 cm diameter and outfitted with two 12.7 mm thick fused-silica windows was used. The maximum pressure rating of the cell was 40 bar. The pump beam was focused into the Raman gas cell with a convex lens. Using the $M = 1.33$ resonator for the XeF($C \rightarrow A$) laser, a focal length of 50 cm could be used without causing breakdown of the laser radiation in the gas cell. The laser resonator with magnification $M = 1.7$ provided better beam quality and higher intensities in the focused beam, and the focal length of the lens had to be restricted to a minimum value of 70 cm in order to prevent breakdown.

The dependence of the Raman conversion on the gas pressure for hydrogen is displayed in Fig. 2. The pump beam was focused into the gas cell with a 50 cm focal length lens. The wavelength of the pump beam was 486.8 nm. The energy of the transmitted fundamental beam decreases monotonically with increasing hydrogen pressure due to energy conversion into Raman lines. Generation of the first Stokes line at 610 nm was observed above a pressure threshold of 4 bar. At a pressure of 10 bar a maximum first Stokes energy of 170 mJ is obtained for a pump energy of 440 mJ, representing an energy conversion efficiency of 38% and a quantum efficiency of 47%. The first Stokes energy decreases at higher pressures due to buildup of the second and higher-order Stokes lines. At a hydrogen pressure of 28 bar a maximum energy of ~ 70 mJ was contained in the second Stokes line at 817.5 nm and higher-order Stokes lines.

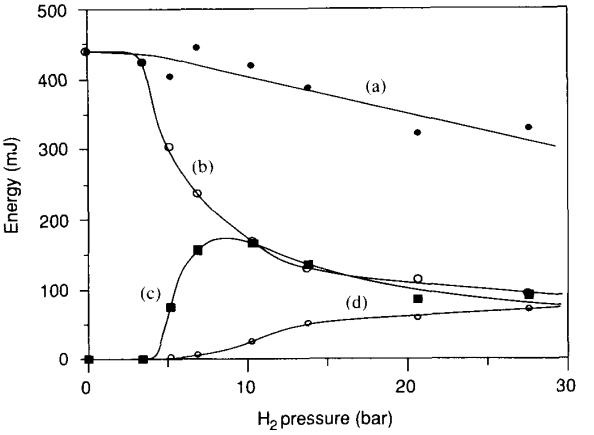

Fig. 2. Dependance of the Raman conversion in hydrogen on gas pressure. The pump beam was focused into the gas cell with a 50 cm lens. (a): total output energy, corrected for the quantum loss due to Raman process. (b): residual fundamental energy, 486.8 nm. (c): energy in the first Stokes line, 610.2 nm. (d): energy in second (817.5 nm) and higher-order Stokes lines.

The first anti-Stokes line at 404.9 nm was observed. A maximum energy of 7 mJ was obtained in the anti-Stokes line for a hydrogen pressure of 20 bar. Furthermore, the first Stokes and anti-Stokes lines of the rotational Raman shift of orthohydrogen (587 cm^{-1}) were detected. The energy contained in these lines was not resolved and is included in the reported energy values for the residual pump beam. However, spectrometer recordings indicate a conversion efficiency for the first rotational Stokes line of less than 10%.

Also shown in Fig. 2 is the total output energy corrected for the quantum loss of the Raman conversion. The total transmitted energy decreases with increasing pressure, which is believed to reflect losses due to the onset of optical breakdown in the gas. Backward Raman scattering is unlikely to account for losses because of the broadband nature of the pump beam relative to the vibrational linewidth of hydrogen. With the magnification $M = 1.7$ laser resonator and 70 cm focal length a similar pressure dependence of the conversion efficiency was observed. However, optimal conversion into the first Stokes line was obtained at a lower hydrogen pressure (7 bar) due to the increased spatial beam quality yielded by the $M = 1.7$ laser resonator.

IV. Conversion in Liquid Nitrogen

For measurements in liquid nitrogen, a Pyrex glass cell of 15 cm length and 5 cm diameter was designed. The nitrogen cell was contained in high vacuum to prevent condensation on the cell windows. A commercially available glass to Kovar metal tube provided both mounting and external access to the glass cell. The use of this tube resulted in high mechanical stability against temperature stress and excellent vacuum conditions. When filling the cell, the liquid nitrogen was filtered through cotton wool to remove any ice crystals, which would cause strong light scattering. Prior to experiments, the nitrogen in the cell was cooled below the atmospheric boiling point by pumping. This technique prevents the occurrence of gas bubbles and ensures good optical quality of the liquid. However, excessive cooling can lead to temperature and refractive index gradients and hence to distortion of the pump beam.

In a first experiment, the liquid nitrogen cell was operated under the same conditions as for hydrogen. The XeF($C \rightarrow A$) laser was equipped with an $M = 1.33$ resonator and the laser beam was focused into the cell with a lens of 70 cm focal length. The output spectrum of the liquid nitrogen cell (Fig. 3) exhibits the first three Stokes and the first anti-Stokes line. Detection of higher-order Stokes lines was prevented by the OMA spectrometer sensitivity. A large number of wavelengths can be generated in this way, however, the efficiency for energy conversion into a single line is limited.

Since the Raman gain coefficient for liquid nitrogen is much larger than for gases, a smaller intensity of the pump beam is required to obtain a high conversion efficiency [11], [14]. For this purpose, the XeF($C \rightarrow A$) laser beam was focused by a 1.5 m lens and the nitrogen cell was placed between the lens and the focal point. The intensity in the liquid nitrogen cell was varied by moving the cell relative to the focal position.

For liquid nitrogen, optimal energy conversion into the first Stokes line was achieved for an average beam diameter of 1.5 cm and a spatially averaged peak intensity in the cell of ~ 60 MW/cm^2. Depicted in Fig. 4 are the temporal profiles of the pump beam and of the generated first and second Stokes beams. The pump beam had a peak power of 72 MW and a full width at half maximum duration of 6.5 ns. The depletion of the pump beam due to energy conversion into Stokes lines is clearly visible. A peak power of 35 MW was generated in the first Stokes line.

The pump energy at a wavelength of 486.8 nm was 550 mJ. The first three Stokes lines at 549.0, 629.3, and 737.2 nm had pulse energies of 210, 20, and 5 mJ, respectively. The energy conversion efficiency into the first Stokes line was 38% and the quantum efficiency 43%. At an average pump intensity of 125 MW/cm^2 an energy of 140 mJ in the first, 100 mJ in the second, and 50 mJ in the third Stokes line were generated.

The spatial profiles of the liquid nitrogen cell output beams were observed with a CCD camera at a distance of 50 cm. While the annular-shaped beam profile of the XeF($C \rightarrow A$) laser is preserved after the Raman cell, the first and second Stokes beams display a more uniform profile. The Raman beams are deflected relative to the fundamental beam by approximately 5 mrad, which probably is caused by a vertical refractive index gradient and associated dispersion gradient in the liquid nitrogen.

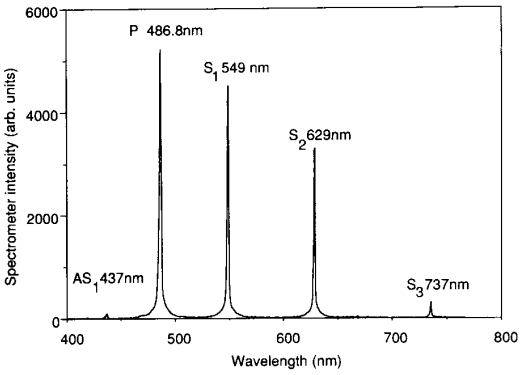

Fig. 3. Spectrum of the liquid nitrogen cell output showing the first three Stokes and the first anti-Stokes lines. The pump beam was focused into the cell with a 70 cm lens. Intensities are not corrected for spectral sensitivity.

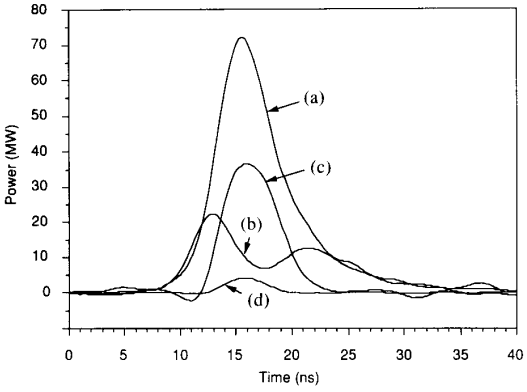

Fig. 4. Temporal profiles of the pump and converted laser pulses for the liquid nitrogen cell. (a): pump laser pulse at 486.8 nm. (b): residual 486.8 nm radiation in the liquid nitrogen cell output. (c): generated first Stokes line at 549.0 nm. (d): generated second Stokes line at 629.3 nm.

V. Wideband Tuning

The tuning range of the first Stokes line of both hydrogen and liquid nitrogen is displayed in Fig. 5. The XeF($C \rightarrow A$) laser was tuned from 466 to 514 nm in ~ 4 nm steps. Wavelengths of narrow-band absorbers in the XeF($C \rightarrow A$) laser gas [4], [15], which yield reduced laser output were specifically probed to demonstrate continuous tunability. These wavelengths are apparent in the Stokes spectrum near 545 nm for liquid nitrogen and near 605 nm for hydrogen.

The Raman cells were operated in a saturated mode, i.e., the increased generation of first Stokes energy with

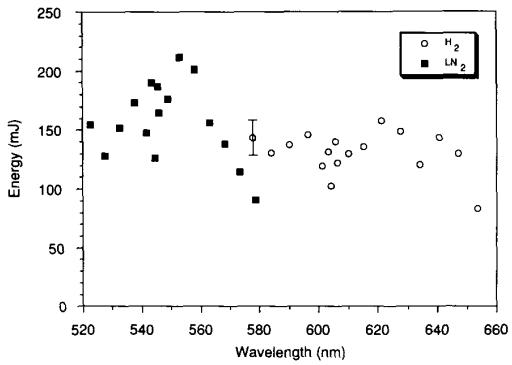

Fig. 5. Spectrum of the first Stokes lines showing the energies obtained with hydrogen and liquid nitrogen as the Raman-active media.

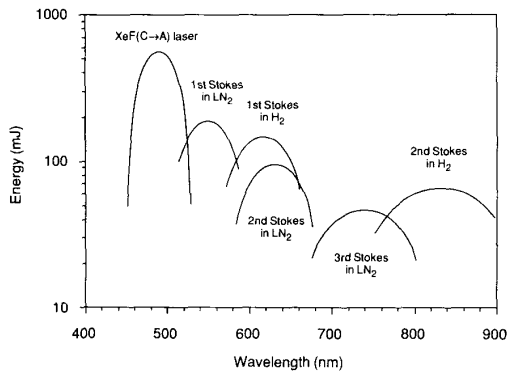

Fig. 6. Spectral range accessible by the XeF($C \rightarrow A$) excimer laser and Stokes lines in liquid nitrogen and hydrogen. The profiles of the second and third Stokes lines are extrapolated from energy measurements at a single laser wavelength.

stronger pumping is offset by losses due to second Stokes energy buildup. This has the effect of reducing the sensitivity of the first Stokes energy to variations in the excimer laser energy. An optimization for the intensity at every wavelength was not performed since this would limit the ease of tunability of the conversion system. The hydrogen gas pressure was fixed at 7 bar and the average beam diameter in the liquid nitrogen cell was set to 1.3 cm. With liquid nitrogen as the active media a wavelength range from 523 to 579 nm is accessible, whereas hydrogen covers the range from 578 to 650 nm. Stimulated Raman scattering was found to expand the tuning range of the XeF($C \rightarrow A$) excimer to cover the spectral region from 455 to 650 nm with energies in excess of 100 mJ.

The wavelength range accessible by the XeF($C \rightarrow A$) excimer laser operating in fundamental mode and Raman shifted in liquid nitrogen and hydrogen is plotted schematically in Fig. 6. The profiles of the second and third Stokes lines were extrapolated from the energy obtained at a single laser wavelength for optimized conditions. Pulse energies of several tens of millijoules are available over a spectral range from 450 to 900 nm.

VI. Conclusion

Raman shifting of the XeF($C \rightarrow A$) excimer laser in hydrogen and liquid nitrogen resulted in continuous tunability from 523 to 650 nm with output energies ranging from 100 to 210 mJ and output powers between 15 and 35 MW. The described system therefore is useful for those applications requiring green to red, high-power radiation. Further increase in first Stokes energy can be expected from a Raman amplifier with external seeding [12] and from the use of a 1.2 J XeF($C \rightarrow A$) laser system [16]. The absence of phase-matching requirements of the Raman conversion process should make this scheme also applicable to a wavelength-agile XeF($C \rightarrow A$) laser [17]. This would allow arbitrarily controlled tuning of the Raman shifted output at a 1 Hz pulse repetition frequency while maintaining 100 mJ pulse energies. Furthermore, the XeF($C \rightarrow A$) excimer laser system in combination with higher-order Stokes generation is capable of covering the blue to red spectral region with pulse energies on the tens of millijoule level.

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