A High-Power Synchronously Pumped Dye Laser System with Diffraction and Transform-Limited Performance

DAVID C. HANNA, DAVID J. POINTER, AND KENNETH A. URE

Abstract—**The design and performance of a high-power synchronously pumped dye laser is described. Emphasis has been given to achieving bandwidth-limited and diffraction-limited performance so that efficient, reproducible nonlinear frequency conversion can be achieved. Pulses of 1 mJ from Rhodamine 6G and 250 μJ from DCM with tuning ranges of 557–584 nm and 609–676 nm, respectively, have been obtained. Second harmonic generation with 40 percent efficiency to ~280 nm and efficient multiple Stokes Raman shifting to cover the range 1.1–1.2 μm has been demonstrated. Blue dyes, pumped by third harmonic of Nd:YAG, have been operated. Typically, for Stilbene 3 (peak at 420 nm), pulses of 160 μJ were obtained.**

Introduction

THE development of tunable light sources of picosecond duration is currently being pursued through a variety of approaches. Given the demand for tunability over a very wide spectral range, in the UV, visible, and infrared, there are obvious attractions in a system based on a tunable visible source with sufficient power to allow efficient nonlinear frequency conversion. A dye laser synchronously pumped by a mode-locked, Q-switched, pulsed Nd:YAG laser provides a convenient way of meeting the high power requirement [1], [2]. This approach was favored rather than, for example, the short cavity dye laser [3] because of a) the design simplicity (in particular, cavity length control and amplifier design) and b) the ability to generate spatially coherent light pulses necessary for nonlinear techniques of frequency extension. In this paper we discuss the design and performance of such a laser based on a commercially available actively mode-locked Nd:YAG laser. Emphasis has been given to simplicity of design and, with a view to extending the laser's tuning range by nonlinear optical techniques, we have paid careful attention to features such as diffraction-limited performance, bandwidth-limited performance, efficiency, and amplitude stability. With Rhodamine 6G as the dye medium, the output was ~1 mJ in ~70 ps. DCM dye proved to be less efficient, giving 250 μJ, but had wider tunability, 609–676 nm. We have confirmed that these power levels, combined with the good spatial coherence, allow efficient frequency conversion from this laser. An example of this is third Stokes generation in CH_4 gas with up to 14 percent photon conversion efficiency and greater than 10 μJ pulse energy over the range 1.1–1.18 μm as the Rhodamine 6G tuning range is spanned. This result suggests that with such efficient conversion to the near infrared, the use of nonlinear techniques may offer an attractive alternative to direct pumping of near infrared dyes.

Efficient harmonic generation has also been demonstrated, with 40 percent conversion from Rhodamine 6G to the 280 nm region. Furthermore, to demonstrate the versatility and full tuning capability of this laser, we report here initial results for dyes operating in the blue to green region pumped by the third harmonic of the Nd:YAG laser. For example, using Stilbene 3 (peak at 420 nm) we have produced single pulses of 160 μJ.

Laser Design

The basic design of this dye laser is similar to the design of Wokaun *et al.* [2] in which a mode-locked, Q-switched Nd:YAG laser system provides two outputs, one being a train of mode-locked pulses, and the other being an amplified single pulse derived from the train. After frequency doubling, the train is used to synchronously pump a dye laser while the single pulse pumps a dye amplifier. The single pulse is timed to arrive at the amplifier in coincidence with a pulse in the dye oscillator output, thus leading to a single amplified dye laser pulse. In this way Wokaun *et al.* produced a single pulse of 1.5 mJ and 20 ps duration from Rhodamine 6G. Our aim has been to achieve a similar output energy from a smaller Nd:YAG laser using just one dye amplifier, rather than the three used by Wokaun. The overall efficiency has been achieved by efficient design in both the dye oscillator and amplifier. In the case of the oscillator this has led to a stable resonator design (by placing a lens inside the resonator [4], [5]) instead of the usual plane/plane resonator used with most high power lasers pumped by nanosecond pulses. In the latter case the resonator can be kept short, but the synchronously pumped dye laser resonator is necessarily long and for a plane/plane resonator this would result in

Manuscript received December 3, 1985; revised March 10, 1986. This work was supported in part by the U. K. Science and Engineering Research Council (SERC), by a Case Studentship from SERC and Culham Laboratory, and by the British Department of Industry.

D. C. Hanna and D. J. Pointer are with the Department of Physics, University of Southampton, Southampton, SO9 5NH, England.

K. A. Ure is with J K Lasers Ltd., Rugby, CV21 1QN, England.

IEEE Log Number 9608788.

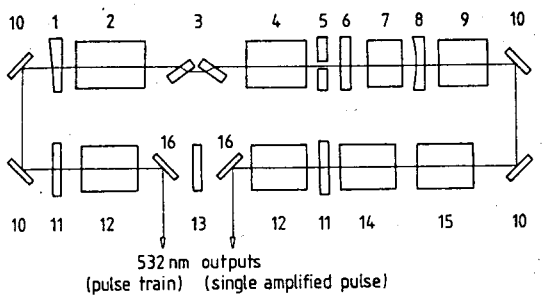


Fig. 1. Schematic layout of the J K Lasers System 2000 AML. 1: Output mirror $R = 50$ percent. 2: Pumping chamber (oscillator). 3: Cavity length adjustment. 4: Pockels cell Q-switch. 5: Aperture. 6: Shutter. 7: Acoustooptic loss modulator. 8: Output mirror $R = 70$ percent. 9: Single pulse selector Pockels cell. 10: Beam steering mirror. 11: $\lambda/2$ plate. 12: Harmonic generator. 13: Beam dump. 14: Pumping chamber (amplifier). 15: Beam expanding telescope. 16: Dichroic steering mirror.

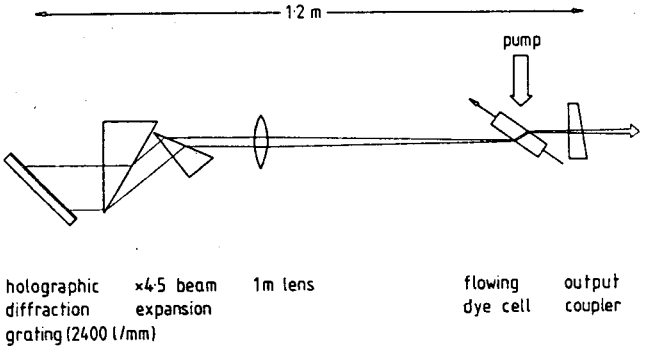


Fig. 2. Schematic layout of the dye oscillator.

high diffraction loss and low efficiency, as evidenced by Wokaun's laser. On the other hand, the higher efficiency, in the oscillator and the amplifier, implies strong saturation of the dye with consequent pulse broadening [6], [7]. Thus, our output pulse from the dye amplifier, while having a comparable energy (~ 1 mJ) to Wokaun's laser, has a duration ~ 70 ps, which is around three times longer.

We have tried two dye oscillators of quite different configuration. One is longitudinally pumped and uses a grazing incidence grating [8], and the other is transversely pumped and uses prism beam expanders and a grating in the Littrow configuration [9]. Despite these differences we have found that both have similar performance with Rhodamine 6G and DCM. The transversely pumped configuration will be discussed first and in greater detail.

The Nd:YAG laser we have used is a J K Lasers System 2000 AML, consisting of an actively mode-locked, Q-switched oscillator (TEM_{00} output) with a single pulse selector and a single-stage Nd:YAG amplifier (see Fig. 1). The Nd:YAG oscillator is operated, at 10 Hz repetition rate, with a long pumping pulse of 5 ms duration to provide the long prelase needed for bandwidth-limited operation, as discussed by Kuizenga [10]. Outputs are taken from each end of the oscillator (one mirror has 50 percent reflectivity, the other 70 percent) and have the form of a train of mode-locked pulses of 8 ns separation in an envelope of 50 ns duration (FWHM). The total envelope energies are typically 3.3 mJ from the 50 percent mirror and 1.7 mJ from the 70 percent mirror. The duration of the mode-locked pulses, measured using a background-free autocorrelation technique [11], was found to be ~ 100 ps, in agreement with calculation based on Kuizenga's analysis [10]. The entire pulse train from the 50 percent reflector was frequency doubled, either in a 4 cm KDP crystal (type II phase matching) or 2 cm CDA crystal, giving 1 or 1.5 mJ at 0.53 μm in a pulse train of 40 ns duration (FWHM), containing ~ 5 pulses. This was used to synchronously pump the dye oscillator. The output from the 70 percent reflector of the Nd:YAG oscillator passed through a Pockels cell single pulse selector, and then, after a $\times 2$ beam expansion, the single pulse was amplified by a single pass through an Nd:YAG amplifier to 8 mJ. This pulse was frequency doubled in another KDP crystal to give 4 mJ at 0.53 μm. The duration of the pulse was confirmed to be $\sim \sqrt{2}$ shorter than the fundamental pulse, i.e., 70 ps. This pulse was used to longitudinally pump the dye amplifier stage.

In choosing the layout of the dye oscillator (see Fig. 2), consideration was first given to a number of polarization requirements. It has been shown [2] that the shortest pulse duration and highest efficiency is achieved when the dye oscillator and amplifier emit with polarization parallel to that of the pump. Due to rotational diffusion of the dye molecules, the gain for parallel polarization initially decays with a time constant on the order of 100 ps, so by placing the dye cell close to the output mirror (an uncoated plate with 30′ wedge), a significantly higher gain could be achieved for light which initially traveled towards the adjacent mirror. This effect contributes towards achieving unidirectional operation. Unidirectional operation was additionally favored by the fact that the fluorescence lifetime is comparable to the cavity round trip time, thus resulting in an asymmetry in the gain with propagation direction. The cell (a 1 cm long spectrophotometer cell) was oriented at Brewster's angle and placed 1 cm from the output mirror. As expected, no evidence was found for double pulsing in the output, confirming that unidirectional operation was taking place. Two prism beam expanders with near Brewster-angled expansion faces were introduced to provide sufficient bandwidth reduction to ensure bandwidth-limited operation. The holographic grating (2400 l/mm) was found to have greater efficiency for p polarization (i.e., perpendicular to the grating grooves, but lying in the plane of incidence) over the wavelength range of the DCM laser but greater efficiency for s polarization for the Rhodamine 6G wavelengths. Similar polarization effects have been shown by Hutley [12]. Operation of both lasers was on p polarization.

To set up the dye oscillator, the laser was first operated without the intracavity lens. The width of the pump beam at the cell (formed to a line by a cylindrical lens) was chosen to be 0.3 mm, with the dye concentration set at 2.5×10^{-4} M (for Rhodamine 6G), so as to give a reasonably symmetrical spot in the far field, implying that the depth of pump penetration into the dye was comparable to the transverse width of the pumped region. The

far field spot size was consistent with diffraction limited operation. The grating was mounted on a micrometer translation stage to provide cavity length adjustment, and synchronous pumping was roughly achieved by adjusting the cavity length for maximum output energy, at which condition an oscilloscope trace displayed a stable mode-locked train from the oscillator. Fine adjustment of cavity length for exact synchronism was made by observing either a minimum in the pulse duration (measured by background-free second harmonic autocorrelation) or, more conveniently, a minimum output energy fluctuation. It was found that the minimum fluctuation coincided with the minimum pulse duration. To monitor the energy fluctuation, typically 600 shots were logged and the stability calculated by an on-line microcomputer. At the exact synchronism setting, 95 percent of all pulses lay within ± 4 percent of the mean energy and a cavity length change of ~ 3 mm would be sufficient to worsen the stability by a factor of 2. A minimum in the pulse duration could be located to within 0.5 mm of the exact cavity length.

Having set the resonator length, the optimization of the pumped volume in the dye was carried out by changing the pump beam width at the dye cell (by translating the cylindrical lens), readjusting the dye concentration for a symmetrical far field spot in each case, and monitoring the output energy. The pumped volume which gave maximum energy was also found to give maximum tuning range. Finally, an intracavity lens of focal length 1 m was inserted, set at a nominal 1 m distance from the output mirror. As the lens was moved along the beam axis away from the output mirror, a point was reached at which the output beam divergence increased. This corresponds to a hemispherical resonator with a very small mode spot in the dye cell, hence resulting in a multimode, highly divergent output. Relative to this point, the lens was then repositioned towards the output mirror until it gave a far-field spot size commensurate with diffraction-limited operation. The inclusion of the lens was found to increase both the output energy and tuning range available. Another advantage of the lens is that it produces a collimated beam incident on the grating. With the Littrow configuration, the oscillation bandwidth $\Delta\nu$ selected by the grating is

$$\Delta\nu = \frac{2c}{\pi D \sin\theta}$$

where θ is the angle of incidence and D is the waist diameter of the beam incident on the grating [9]. Without the lens D would correspond to the size of the pumped region in the dye; thus, the inclusion of the lens helps to narrow the oscillation bandwidth. In fact, to achieve bandwidth-limited operation we found it necessary to increase D further by introducing two beam-expanding prisms between the lens and grating. Without these prisms the bandwidth for Rhodamine 6G was ~ 1.4 cm^{-1}, giving a time–bandwidth product of 5.25 for the measured ~ 70 ps pulses. With the two prisms (fused quartz, near Brewster's angle expansion face, giving a combined expansion

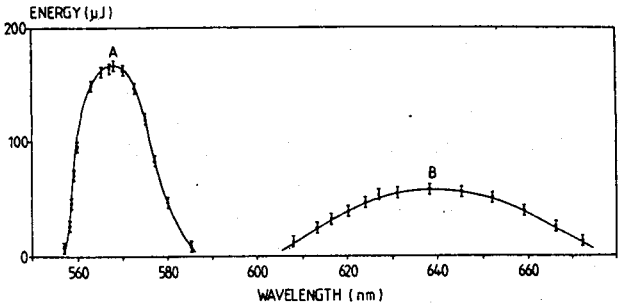


Fig. 3. Dye oscillator tuning behavior. A: Rhodamine 6G; B: DCM.

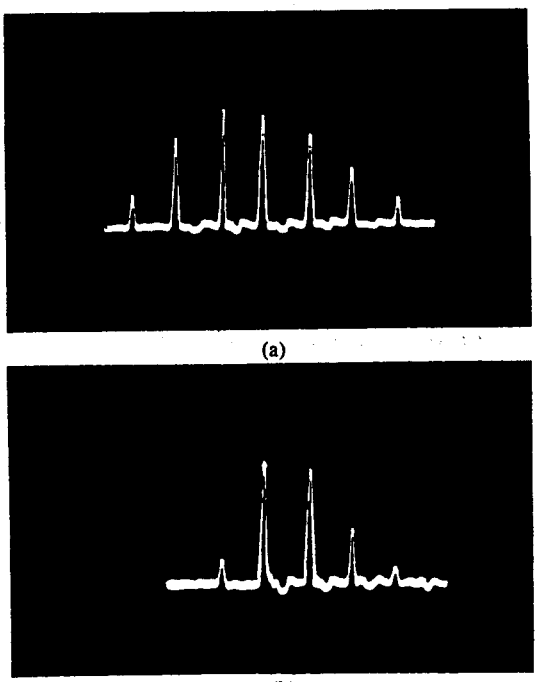


Fig. 4. Mode-locked pulse train. (a) 532 nm pump. (b) Dye oscillator output.

of 4.5) the bandwidth reduced to 0.3 cm^{-1}. The pulse duration was unchanged at 70 ps, thus giving a time–bandwidth product of 0.63. Both the pulse duration and bandwidth measurements were made in the entire pulse train, thus giving average values. Bandwidth measurements were made using a scanning Fabry–Perot interferometer (free spectral range of 2 cm^{-1} and finesse of 30).

Fig. 3(a) and (b) respectively show the tuning behavior for Rhodamine 6G in methanol and DCM in methanol when set up with the procedure outlined above. The total energy in the 0.53 μm pump train [Fig. 4(a)] was 700 μJ. A typical dye oscillator pulse train is shown in Fig. 4(b).

The longitudinally pumped resonator employed a wedged folded dye cell (Fig. 5), which incorporates a broad-band high-reflectivity mirror and an uncoated window output coupler. This design minimizes the losses associated with parasitic oscillations between the dye cell windows, and since the output coupler is in contact with the dye, the maximum possible gain is achieved on the second pass, resulting in efficient and unidirectional operation. Tuning is achieved by a near grazing incidence diffraction grating and mirror arrangement. Using this

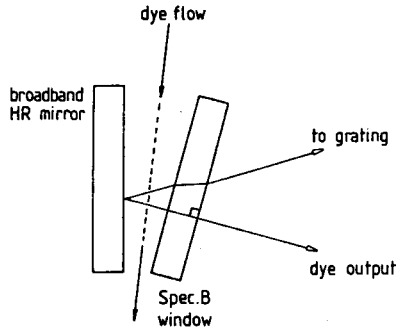


Fig. 5. Wedged folded dye cell in the longitudinally pumped oscillator.

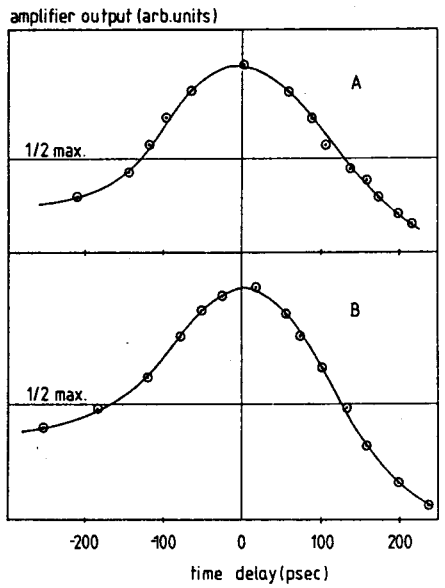


Fig. 6. Amplifier efficiency versus relative delay between the pump and dye oscillator pulses. A: Rhodamine 6G; B: DCM.

configuration provides sufficient dispersion to achieve bandwidth-limited operation without the need to use beam expansion, and the preferred polarization orientation remains the same over the entire grating tuning range. To minimize diffraction losses in the oscillator, an intracavity lens of 3 m focal length is placed ~1 m from the dye cell. This was chosen to efficiently couple back into the gain region (spot size ~0.5 mm) while only allowing single transverse mode operation. The tuning behavior and efficiency for this design was similar to the transversely pumped oscillator, both for Rhodamine 6G and DCM dyes.

The dye amplifier is quasi-longitudinally pumped by 2.7 mJ of energy in a single pulse. Timing of the arrival of the pump pulse needs to be accurate to within ~30 ps for maximum gain. Fig. 6(a) and (b) show the dependence of amplifier output on relative timing of the arrival of oscillator pulse and pumping pulse at the amplifier. Both Rhodamine 6G [Fig. 6(a)] and DCM [Fig. 6(b)] show similar rotational diffusion timescales of ~150 ps. The pump beam was focused to a diameter of 0.8 mm in the 1 cm long amplifier cell and the oscillator beam size was matched to this. Dye concentration was varied to achieve maximum output energy. In the case of Rhodamine 6G this was 0.8×10^{-4} M and for DCM, 2×10^{-4} M. Energy gains from the single amplifier of ~15 (70 μJ input, ~1 mJ output) for Rhodamine 6G and ~22 (10 μJ input, ~0.22 mJ output) for DCM were obtained. The tuning curves for amplifier output for both dyes are shown in Fig. 7. The conversion efficiency in the amplifier reached a maximum of 33 percent for Rhodamine 6G and 8.5 percent for DCM. The lower efficiency for DCM is not at present understood. It is significantly lower than values quoted for pumping with longer (~10 ns) pulses. Reduced efficiency for longer wavelength near IR dyes has previously been noted by Kobayashi *et al.* [13].

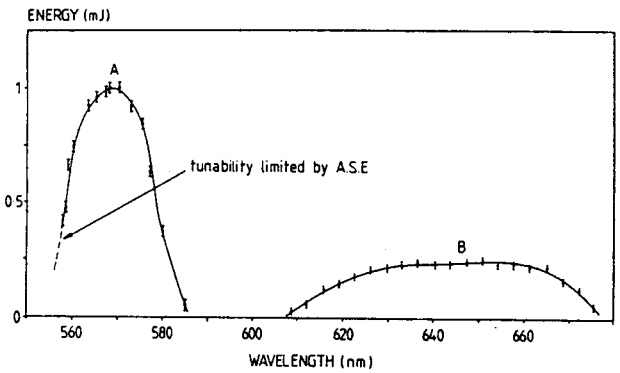


Fig. 7. Dye amplifier tuning behavior. A: Rhodamine 6G; B: DCM.

Measurement of the pulse duration for the amplifier output yielded ~90 ps for both Rhodamine 6G and DCM, indicating that any pulse lengthening introduced by the highly saturated amplifier was quite modest. The output beam remained diffraction limited, taking on a cleaner, more circular shape as a result of the longitudinal pumping. The amplitude stability of the amplified pulse was measured to be typically ±7 percent.

We have demonstrated the potential of this system by operating dyes in the blue to green region pumped by the third harmonic of the Nd:YAG laser. We report here a brief outline of the results and will report fully in a further publication. A conversion efficiency to 354 nm of 26 percent was obtained from the Nd:YAG using a 3 cm KD*P crystal for second harmonic and 2 cm KD*P crystal for the sum frequency generation. Using a transversely pumped oscillator, Stilbene 3 produced a train of 11 pulses of 44 μJ total energy at the peak of the tuning curve (420 nm) which extended from 412 to 440 nm. Single-stage amplification, using a transverse pumping configuration, yielded single pulses of 160 μJ. Other dyes have been operated (e.g., Coumarin 500 and 440). The performance of these dyes will be reported later.

To test the suitability of the dye laser for further frequency extension, we have looked at second harmonic generation and stimulated Raman scattering. Second harmonic generation of the Rhodamine 6G output at 560 nm readily gave 40 percent conversion efficiency to 280 nm, using a 1 cm crystal of ADP. Stimulated Raman scattering has been examined both in CH_4 gas and in liquid acetone. The latter provided a particularly simple and convenient check on the beam quality of the dye output since the expected threshold for a diffraction limited beam could be

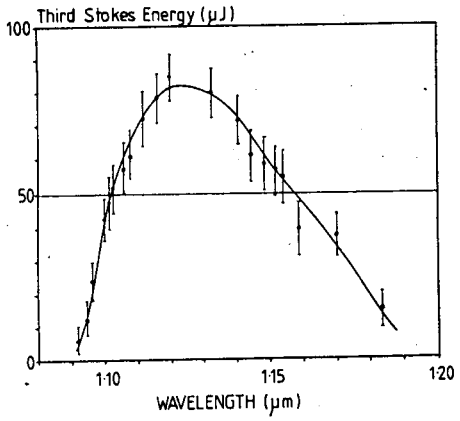


Fig. 8. Tuning behavior for third Stokes generated in CH_4 gas: double cell arrangement.

calculated with accuracy, and the ratio of measured to calculated threshold then indicates the factor by which the beam divergence exceeds the diffraction limit. Comparison with the threshold obtained with 0.53 μm light, whose beam was known to be TEM_{00}, also gave a good check on the dye beam quality. Using an acetone cell of length $l = 15$ cm, with tightly focused pump ($l/b \sim 2.6$ where b is the confocal parameter), the observed threshold was 30 μJ for a 70 ps, 0.53 μm pulse. This compares well with the predicted value of 33 μJ (see [14] for the analysis) based on a Raman gain coefficient $g_R = 1.17 \times 10^{-11}$ m/W [15]. The threshold with a 70 ps, 0.57 μm pulse from the Rhodamine 6G laser was 32 μJ, in agreement with the calculated value of 36 μJ, confirming the diffraction limited beam quality. It is interesting to note that this Raman scattering in acetone was extremely efficient, giving 80 percent energy conversion to 630 nm for a 0.3 mJ input at 0.53 μm. Results like this suggest that Raman shifting may prove a more convenient way of achieving long wavelength operation rather than using direct pumping of near infrared dyes, although it has to be noted that the generated 0.63 μm radiation had a bandwidth of ~ 1.6 cm^{-1} (the spontaneous Raman linewidth in acetone is ~ 17 cm^{-1}) and, hence, was not bandwidth limited. Raman shifting in CH_4 (spontaneous Raman linewidth ~ 0.68 cm^{-1} at 30 atm [16]) provides a narrow enough linewidth for bandwidth-limited Stokes pulses and furthermore, with its good infrared transmission, offers the possibility of efficient multiple Stokes shifts into the infrared. As an example of this we quote the results obtained using a 1.5 m long gas cell containing CH_4 at 30 atm. The Rhodamine 6G dye beam was focused to a spot size of ~ 100 μm ($b = 11$ cm) in the first half of the cell, and a 20 cm lens inside the cell produced a second focus for the pump and Stokes light generated at first focus. With this arrangement we have obtained 14 percent photon conversion to third Stokes around 1.15 μm for 0.5 mJ input. The energy tuning curve for third Stokes is shown in Fig. 8, indicating that greater than 10 μJ of third Stokes could be generated over the range 1.1–1.18 μm as the Rhodamine laser was tuned.

Conclusion

We have described the design and operation of a synchronously pumped dye laser where the aim has been to achieve a high-power, bandwidth-limited, and diffraction-limited output suitable for further frequency extension via nonlinear techniques. Simplicity of design has been a further aim, although to achieve this we have compromised on the ability to generate the shortest possible pulses and have typically produced ~ 70 ps pulses. However, there now exist well-established techniques of pulse compression which could be incorporated into this laser. With the high power available at the third harmonic of the Nd:YAG laser, we have successfully demonstrated the ability of this laser to operate with dyes in the blue to green region. Given also the demonstrated capability of this laser for efficient harmonic generation and Raman shifting, it appears that the basis for a very widely tunable high power picosecond light source now exists.

Acknowledgment

This work has been supported in part by the SERC, and the second author acknowledges support from the SERC and Culham Laboratory in the form of a Case Studentship. The development of a tunable short pulse laser by J K Lasers Ltd. was financed in part by the British Department of Industry.

References

[1] L. S. Goldberg and C. A. Moore, "Transform-limited picosecond pulse generation in a synchronous mode-locked dye laser pumped by doubled NdYAG," *Appl. Phys. Lett.*, vol. 27, pp. 217–219, 1975.
[2] A. Wokaun, P. F. Liao, R. R. Freeman, and R. H. Storz, "High energy picosecond pulses: Design of a dye laser amplifier system," *Opt. Lett.*, vol 7, pp. 13–15, 1982.
[3] H. P. Kortz, P. Pax, and R. Aubert, "Generation of high power and tunable picosecond pulses," *SPIE Proc.*, vol. 533, Jan. 1985.
[4] J. A. Lisboa, S. Ribeiro Teixeira, S. L. S. Gunha, and R. E. Francke, "A grazing incidence dye laser with an intracavity lens," *Opt. Commun.*, vol. 44, pp. 393–395, 1983.
[5] A. G. Yodh, Y. Bai, J. E. Golub, and T. W. Mossberg, "Grazing incidence dye lasers with and without intracavity lenses; a comparative study," *Appl. Opt.*, vol. 23, pp. 2040–2042, 1984.
[6] E. P. Ippen and C. V. Shank, *Picosecond Phenomena.* Berlin, Germany: Springer-Verlag, 1978, p. 103.
[7] P. J. K. Wisoff and R. G. Caro, "A high power picosecond dye oscillator synchronously pumped by a *Q*-switched, mode-locked NdYAG laser," *Opt. Commun.*, vol. 54, pp. 353–357, 1985.
[8] M. G. Littman and H. J. Metcalf, "Spectrally narrow pulsed dye laser without beam expander," *Appl. Opt.*, vol. 17, pp. 2224–2226, 1978.
[9] D. C. Hanna, P. A. Karkkainen, and R. Wyatt, "A simple beam expander for frequency narrowing of dye lasers," *Opt. Quantum Electron.*, vol. 7, pp. 115–119, 1975.
[10] D. J. Kuizenga, "FM and AM modelocking of the homogeneous laser—Part 1: Theory," *IEEE J. Quantum Electron.*, vol. QE-6, pp. 694–708, 1970.
[11] E. P. Ippen and C. V. Shank, *Ultrashort Light Pulses.* Heidelberg, West Germany: Spinger-Verlag, 1977.
[12] M. C. Hutley, "Interference diffraction gratings," *Sci. Progr. Oxford*, vol. 61, pp. 301–321, 1974.
[13] H. Kobayashi, S. Tanaka, A. Watanabe, T. Kuwata, H. Tabuchi, and M. Veoka, "Tunable near-infrared picosecond pulses from a mode-locked dye laser synchronously pumped by second harmonic of a NdYAG laser," *Japan. J. Appl. Phys.*, vol. 23, pp. L40–L42, 1984.
[14] D. C. Hanna, D. J. Pointer, and D. J. Pratt, "Stimulated Raman

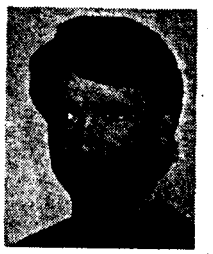
scattering of picosecond light pulses in hydrogen, deuterium, and methane," *IEEE J. Quantum Electron.*, vol. QE-22, pp. 332–336, Feb. 1986.

[15] M. J. Colles, "Efficient stimulated Raman scattering from picosecond pulses," *Opt. Commun.*, vol. 1, pp. 169–172, 1969.

[16] Y. Taira, K. Ide, and H. Takuma, "Accurate measurement of the pressure broadening of the v_1 Raman line of CH_4 in the 1–50 atm region by inverse Raman spectroscopy," *Chem. Phys. Lett.*, vol. 91, pp. 299–302, 1982.

David C. Hanna, for a biography, see p. 336 of the February 1986 issue of this TRANSACTIONS.

David J. Pointer, for a photograph and biography, see p. 336 of the February 1986 issue of this TRANSACTIONS.



Kenneth A. Ure was born in Bathgate, Scotland, on August 24, 1960. He received the B.Sc. degree in physics from Edinburgh University, Edinburgh, Scotland, in 1982.

In 1982 he joined J K Lasers Ltd., Rugby, England, where he has worked on the development of short pulse solid-state and dye lasers. He is also a part-time research student at the University of Southampton, Southhampton, England, where he continues to study for the Ph.D. degree in the generation of short, high-power, optical pulses.