

Compact 2.5-W 10-kHz Nd:YLF-pumped dye laser

Andrew J. S. McGonigle, Anthony J. Andrews, David W. Coutts, Geoff P. Hogan, Kristie S. Johnston, Joseph D. Moorhouse, and Colin E. Webb

A 10-kHz pulse repetition frequency dye laser, end pumped by a Nd:YLF laser, is reported. This laser was tunable from 590 to 655 nm, and up to 2.55 W of output power was obtained at the 609-nm peak tuning wavelength. By inserting an etalon into the dye laser cavity and frequency doubling using a β -barium borate crystal, we obtained up to 125 mW of 308-nm single-etalon-mode output, which shows potential for the performance of airborne measurements of tropospheric hydroxyl radical concentrations.

© 2002 Optical Society of America

OCIS codes: 120.0280, 140.2050, 140.3530, 140.3610.

1. Introduction

Ultraviolet frequency-doubled dye lasers that operate with pulse repetition frequencies (PRFs) of ~ 10 kHz, narrow-band spectral output (a few gigahertz), short pulse length (a few tens of nanoseconds), and moderate average powers (hundreds of milliwatts) are important tools for many spectroscopic applications, such as laser-induced fluorescence monitoring of tropospheric hydroxyl (OH) radicals.^{1–5} Until recently this portfolio of specifications was realizable only with copper-vapor-laser- (CVL-) pumped frequency-doubled dye lasers.^{6,7} Indeed, in the past our group has developed a number of lasers based on this scheme, which have been used to make ground-based OH measurements.^{8,9}

To use a 10-kHz PRF diode-pumped solid-state (DPSS) laser as a pump source for these applications, intense diode pumping is required to generate a sufficient population inversion, within the short interpulse duration (~ 100 μ s), to provide output from the DPSS laser of appropriately high average power and

short pulse length. Unfortunately, pumping in this manner also increases the likelihood of unacceptable thermally induced beam quality distortion or even crystal fracture. It has only been recently, with the advent of well-engineered DPSS master oscillator power amplifier architectures, which can manage these thermal loads, that DPSS lasers have been developed that can lead to UV output that satisfies the performance criteria listed above.¹⁰ In this paper we present the performance characteristics of a frequency-doubled dye laser, pumped by such a DPSS laser: a Nd:YLF laser commercially manufactured by Q-Peak.¹⁰

One of the motivations behind the research reported here was to investigate the suitability of this laser for airborne monitoring of OH concentrations. In light of this, we particularly focus on the performance of the dye laser when tuned to 616 nm and that of its 308-nm harmonic (the OH resonance transition wavelength). It is desirable to use a DPSS pump laser, as opposed to a CVL for this application, as the former pump source is lighter, less bulky, and does not require external water cooling and gas handling. Thus a DPSS-pumped dye laser is far more suitable for airborne OH measurements, complementing the capability of the CVL-pumped dye lasers that we previously developed for ground-based OH measurements.^{8,9}

Of course the ideal laser for this application would be an all-solid-state laser, and research has been conducted both in our laboratory and elsewhere in attempts to realize such a system.^{11,12} Indeed, recently it has been demonstrated that a commercially developed Nd:YAG-pumped frequency-tripled Ti:Al₂O₃ laser is capable of laboratory detection of

When this research was performed, A. J. S. McGonigle (ajsm2@cam.ac.uk), A. J. Andrews, D. W. Coutts, G. P. Hogan, K. S. Johnston, J. D. Moorhouse, and C. E. Webb were with the Department of Atomic and Laser Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom. A. J. S. McGonigle is now with the Department of Geography, University of Cambridge, Downing Place, Cambridge CB2 3EN, United Kingdom. J. D. Moorhouse is now with the Department of Atmospheric, Oceanic and Planetary Physics, Clarendon Laboratory, Parks Road, Oxford OX1 3DW, United Kingdom.

Received 29 May 2001; revised manuscript received 16 November 2001.

0003-6935/02/091714-04\$15.00/0

© 2002 Optical Society of America

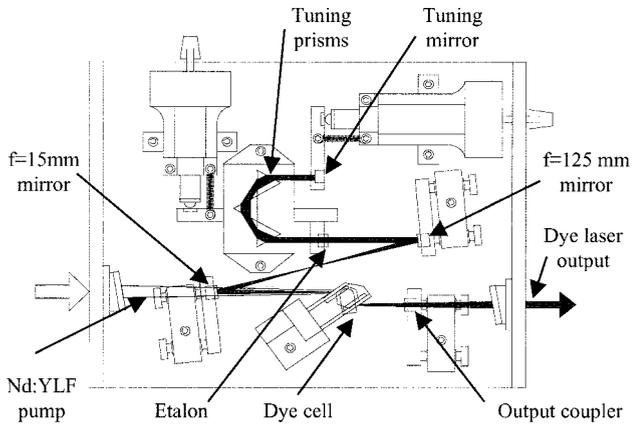


Fig. 1. Experimental configuration of the LAS Intradye II dye laser.

OH,¹³ with a sensitivity similar to that obtainable with the CVL-pumped dye systems mentioned above.

2. Experimental Configuration and Results

The Q-Peak diode-pumped frequency-doubled Nd:YLF laser was operated at a PRF of 10 kHz. This laser contained an acousto-optically *Q*-switched master oscillator and a single-stage power amplifier, which were both based on multipass gain modules.¹⁰ Frequency doubling of the 1047-nm Nd:YLF output was achieved in a noncritically phase-matched temperature-tuned lithium triborate crystal to provide up to 10 W of green (523.5-nm) TEM₀₀ output with a pulse length of 36 ns and a M^2 of less than 1.2.

Figure 1 shows the experimental configuration of the dye laser used in the experiments reported here. The 45-cm-long laser cavity consisted of four mirrors: an $f = 15$ -mm dichroic input coupling mirror, an $f = 100$ -mm output coupler of 20% reflectivity, an $f = 125$ -mm recollimating mirror, and a flat high reflector forming a mode of diameter 0.27 mm in the dye cell. An $f = 200$ -mm lens was used to focus the Nd:YLF 523.5-nm output through the input coupling mirror to form a spot of diameter 0.3 mm within the dye cell. We attained dye laser tunability using three intracavity Brewster prisms that were placed in the collimated section of the intracavity beam. The dye cell itself was 6 mm thick and oriented in a Brewster configuration to favor horizontally polarized oscillation to enhance the transmission of the polarized pump into the cell and, along with the intracavity prisms, to provide horizontally polarized output to enhance the frequency-doubling efficiency. We achieved further line narrowing by inserting an intracavity etalon, which had a finesse of 30 and a free spectral range of 0.25 nm at 616 nm. The dye was circulated with a 5 l/min flow-rate pump, and a 2-mM Rhodamine B dye concentration was used as this was found to optimize the output power at 616 nm. A dye laser having this configuration was first developed by Wennberg *et al.*³ and commercially manufactured by Laser Analytical Systems (LAS) as the Intradye II. The Wennberg *et al.* laser was

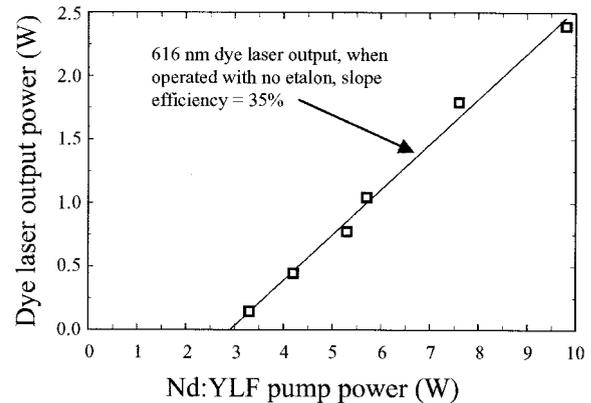


Fig. 2. Dye laser output power versus incident Nd:YLF pump power for operation at 616 nm with no intracavity etalon.

pumped by two Spectra-Physics tightly folded resonator Nd:YLF lasers with a delay between the two lasers corresponding to the buildup time of the dye laser.

Although this laser was originally designed³ for intracavity frequency doubling, at the high pump powers we used extracavity harmonic generation is both simple and efficient. We obtained frequency doubling using an $f = 100$ -mm spherical lens to focus the 616-nm output into a 5 mm × 3 mm × 12 mm β -barium borate crystal ($\phi = 38^\circ$, $\theta = 0^\circ$). The UV output from this crystal was recollimated by use of another $f = 100$ -mm spherical lens and separated from the fundamental by a silica prism. All power measurements were taken with a Coherent DL-20 powermeter, and the temporal pulse shapes were measured with a Thorlabs DET2-SI fast photodiode and a Hewlett-Packard 54111D digitizing oscilloscope.

Figure 2 shows a plot of the output power versus Nd:YLF laser pump power when the dye laser was tuned to 616 nm and operated with no intracavity etalon. The threshold to lasing was 2.9 W, the slope efficiency was 35%, and the maximum output power attained was 2.4 W from 9.8 W of pump power (absolute efficiency of 24%). This laser could be operated for ~40 h before any long-term deterioration in output power was observed.

Tunability of the dye laser, operated both with and without the intracavity etalon, is shown in Fig. 3 when this laser was pumped with 9.8 W of Nd:YLF output. With no etalon the dye laser was tunable from 590 to 655 nm, with a tuning curve peak at 609 nm. The tunability was reduced only slightly (591 to 654 nm) when we inserted the etalon. The powers at 609 nm from the dye laser were 1.3 and 2.55 W, respectively, when the laser was operated with and without the etalon. For operation at 616 nm, up to 1.18 and 2.4 W, respectively, were attained with and without the etalon. A 1-m Spex Czerny–Turner spectrometer was used to determine that the dye laser was indeed oscillating on a single etalon mode for all pump powers; however, the spectrometer had insufficient resolution to determine the exact linewidth

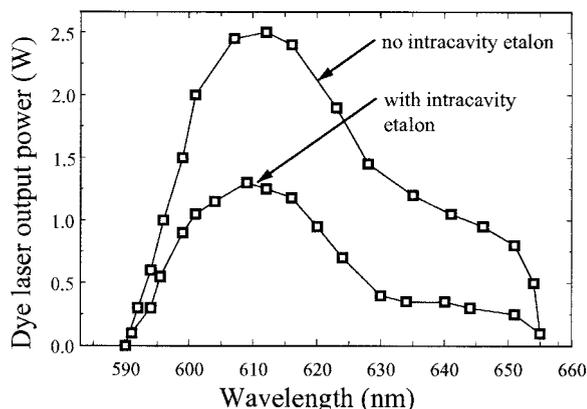


Fig. 3. Tunability of the dye laser, operated both with and without the intracavity etalon, when pumped with 9.8 W from the Nd:YLF laser.

of the single-etalon-mode output. The output linewidth is assumed to be of the order of 5 GHz as this was the single-etalon-mode linewidth obtained when the dye laser was pumped by a CVL during an earlier investigation.⁹

Figure 4 shows temporal pulse shapes of the 616-nm dye laser output, transmitted through the β -barium borate frequency-doubling crystal, both with and without phase matching, when the dye laser was operated with the etalon and pumped with 9.8 W. The maximum depletion of the fundamental dye laser pulse shape at any point in time, which is due to harmonic generation, was 20%, and the FWHM of the undepleted pulse was 28 ns, corresponding to a maximum dye laser peak power of 4.7 kW. Figure 4 also shows the 308-nm harmonic pulse of pulse length 22 ns FWHM. The maximum UV power obtained was 125 mW (correcting for the prism-induced losses) from 1.18 W at 616 nm, corresponding to a harmonic conversion efficiency of 10.5%. The UV pulse-to-pulse amplitude stability was found to be $\pm 3\%$.

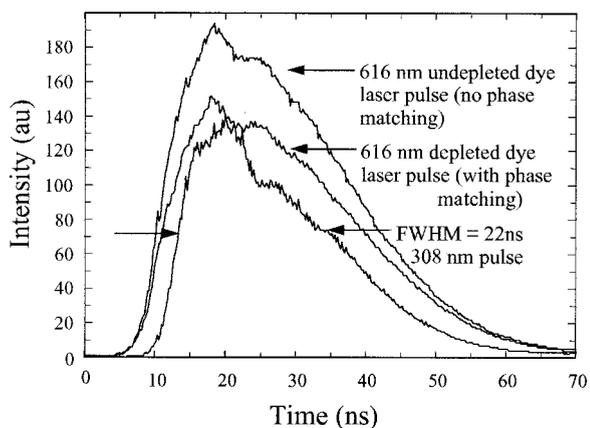


Fig. 4. Temporal pulse shapes from the 616-nm dye laser, operated with the intracavity etalon and pumped with 9.8 W, both with and without phase matching in the β -barium borate crystal (depleted and undepleted pulses, respectively). The 308-nm harmonic pulse shape is also shown.

3. Discussion and Conclusion

In this paper the performance of a 10-kHz PRF Nd:YLF-pumped frequency-doubled LAS dye laser has been documented. The dye laser was found to oscillate on a single etalon mode, even when operated with the maximum pump power (9.8 W). This was due to the constant intrapulse beam quality of the Nd:YLF laser, which provided stable intrapulse mode matching to the dye laser mode. Seeding of a single etalon mode was also favored by the relatively slowly rising leading edge (12 ns) of the pump pulse, which increased the gain gently within the pumped volume. The slow rising edge is analogous to the double-pulse pumping used in Ref. 3.

The performance of a narrow-linewidth, end-pumped laser is particularly sensitive on the pump-laser characteristics, as the focused spot size is a function of the square of the divergence (rather than the first power of divergence for side pumping). For example, during our earlier CVL pumped LAS dye laser investigation, it was found that single-etalon-mode operation was not achievable above pump powers of 4 W.⁹ This was considered to be due to the rapidly improving CVL intrapulse beam quality¹⁴ that resulted in overfilling the cavity mode at the start of the CVL pulse and the fast rising leading edge (4 ns) of the CVL pulse, leading to a sudden increase of gain in the mode volume. Consequently, seeding of multiple etalon modes was favored at elevated pump powers. These dye laser studies serve to highlight the particular suitability that DPSS lasers have to provide high output power by end-pumping narrow-linewidth power oscillators such as the LAS Intradye II. The high laser slope efficiencies observed from the Nd:YLF-pumped LAS dye laser (35% for operation at 616 nm with no etalon) are also thought to be attributable to the constant intrapulse mode matching that the Nd:YLF laser provided (compared with an 18% slope for CVL pumping for the same dye laser cavity conditions).

The broad tunability (590–655 nm) obtained from this Nd:YLF-pumped dye laser (with no etalon) was partly due to the fact that the Nd:YLF pulse length (35 ns) was considerably longer than the excited-state lifetime (a few nanoseconds) of the dye molecules. This provided a relatively long gain duration so that tuning was broadened to wavelengths where the gain was low, and therefore long cavity buildup times were required. In comparison, when the 15-ns pulse-length CVL was used to pump the LAS laser,⁹ tunability was achieved only from 592 to 622 nm (the peak powers of both pump sources were comparable: 28 kW for the Nd:YLF versus 23 kW for the CVL). However, the relatively long Nd:YLF pulse length was disadvantageous in the sense that it led to relatively long dye laser pulse lengths (28 ns) and correspondingly modest conversion efficiencies (10.5%) for UV harmonic generation. In contrast, when the 15-ns pulse-length CVL was used to pump this dye laser, the UV harmonic conversion efficiency was considerably higher (16%).

In conclusion, the performance of a frequency-doubled LAS Intradye II laser, pumped by a Q-Peak 10-kHz PRF Nd:YLF laser, has been presented. When operated without an etalon, broad tunability was obtained from 590 to 655 nm, and as much as 2.55 W of power was generated at the peak tuning wavelength of 609 nm with a slope efficiency of 35%. By inserting an intracavity etalon into the dye laser, we obtained single-mode output at all pump powers, and 1.18 W of output power was generated at 616 nm. When this 616-nm laser was frequency doubled, 125 mW of 308-nm power was obtained, with a conversion efficiency of 10.5%. These results have demonstrated the suitability of DPSS lasers to end pump narrow-linewidth power oscillators such as the LAS Intradye II.

By virtue of its UV tunability, moderate output power, compactness, and relatively light weight, the laser reported here has the potential to be a convenient and useful tool for many spectroscopic applications, in particular airborne laser-induced fluorescence measurements of tropospheric OH radicals—the application that has been one of the motivations behind this research. Although the laser's linewidth has yet to be measured precisely, it was operating on a single etalon mode and is thus likely to be ~ 5 GHz (as discussed above), providing a good fit to the 3-GHz Doppler-broadened 308-nm OH absorption feature. The laser linewidth could be more precisely matched to the OH absorption feature by use of an etalon with a slightly higher finesse, as has been demonstrated with the CVL-pumped LAS laser⁹ where the laser output-power characteristics were negligibly affected when the etalon finesse was varied from 15 to 30.

It should be noted that a number of other research groups are presently using multikilohertz PRF Nd³⁺ laser-pumped frequency-doubled dye lasers to perform airborne OH measurements. However, these lasers have lower average powers or lower PRFs [20 mW at 3 kHz (Ref. 4), 20 mW at 10 kHz (Ref. 5)] than the laser reported here (125 mW at 10 kHz). Consequently, the latter laser shows potential to provide higher temporal resolution OH data because it enables measurement of a particular OH concentration with a given signal-to-noise ratio in shorter integration times than those required with the currently applied lasers.

We acknowledge the European Commission [contracts ENV4 CT95 0003 (with KFA Jülich) and EC DG XII], and Oxford University and National Environment Research Council (Joint Research Equip-

ment Initiative grant GR3/E0058) for funding this research.

References

1. F. Holland, M. Hessling, and A. Hofzumahaus, "In situ measurement of tropospheric OH radicals by laser-induced fluorescence—a description of the KFA instrument," *J. Atmos. Sci.* **52**, 3393–3401 (1995).
2. D. J. Creasey, D. E. Heard, P. A. Halford-Maw, M. J. Pilling, and B. J. Whitaker, "Implementation and initial deployment of a field instrument for measurement of OH and HO₂ in the troposphere by laser-induced fluorescence," *J. Chem. Soc. Faraday Trans.* **93**, 2907–2913 (1997).
3. P. O. Wennberg, R. C. Cohen, N. L. Hazen, L. B. Lapson, N. T. Allen, T. F. Hanisco, J. F. Oliver, N. W. Lanham, J. N. Demusz, and J. G. Anderson, "Aircraft-borne laser induced fluorescence instrument for the in-site detection of hydroxyl and hydroperoxyl radicals," *Rev. Sci. Instrum.* **65**, 1858–1876 (1994).
4. W. H. Brune, I. C. Faloona, D. Tan, A. J. Weinheimer, T. Campos, B. A. Ridley, S. A. Vay, J. E. Collins, G. W. Sachse, L. Jaegle, and D. J. Jacob, "Airborne in-situ OH and HO₂ observations in the cloud-free troposphere and lower stratosphere during SUCCESS," *Geophys. Res. Lett.* **25**, 1701–1704 (1998).
5. E. J. Lanzendorf, T. F. Hanisco, P. O. Wennberg, R. C. Cohen, R. M. Stimpfle, J. G. Anderson, R. S. Gao, J. J. Margitan, and T. P. Bui, "Establishing the dependence of [HO₂]/[OH] on temperature, halogen loading, O-3 and NO_x based on in situ measurements from the NASA ER-2," *J. Phys. Chem. A* **105**, 1535–1542 (2001).
6. M. Broyer, J. Chevalyere, G. Delecrétaz, and L. Wöste, "CVL-pumped dye laser for spectroscopic application," *Appl. Phys. B* **35**, 31–36 (1984).
7. I. J. Evans and C. E. Webb, "Efficient high repetition rate tunable sources for the ultra-violet," *Opt. Commun.* **113**, 72–78 (1994).
8. I. J. Evans and C. E. Webb, "A 10 kHz pulsed tunable laser source at 308 nm for tropospheric OH monitoring," *Chem. Phys. Lett.* **230**, 127–130 (1994).
9. A. J. S. McGonigle, "Tunable UV lasers," D. Phil. dissertation (University of Oxford, Oxford, UK, 2000).
10. K. F. Wall, M. Jaspan, A. Dergachev, A. Szpak, J. H. Flint, and P. F. Moulton, "A 40 W, single frequency, Nd:YLF master oscillator/power amplifier system," in *Advanced Solid State Lasers*, M. M. Fejer, H. Injeyan, and U. Keller, eds., Vol. 26 of OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 1999), pp. 216–221.
11. A. J. S. McGonigle, D. W. Coutts, and C. E. Webb, "530-mW 7-kHz cerium LiCAF laser pumped by the sum-frequency-mixed output of a copper-vapor laser," *Opt. Lett.* **24**, 232–234 (1999).
12. D. J. Binks, P. S. Golding, and T. A. King, "Compact all-solid-state high repetition rate tunable ultraviolet source for airborne atmospheric gas sensing," *J. Mod. Opt.* **47**, 1899–1912 (2000).
13. D. Heard, School of Chemistry, University of Leeds, Leeds, LS2 9JT UK (personal communication, 2001).
14. D. W. Coutts, "Time resolved beam divergence from a copper vapour laser with unstable resonator," *IEEE J. Quantum Electron.* **31**, 330–342 (1995).