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Publication Date

1978-11-01

Submitted to Optics Communications

LBL-8314
Preprint c.2

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A Megawatt Dye Laser Oscillator-Amplifier System
for High Resolution Spectroscopy

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ABSTRACT

Peak powers in excess of 4 MW in the visible and 1.3 MW in the UV with linewidths as narrow as 80 MHz are generated with three YAG pumped amplifier stages following a cw dye oscillator. The laser pulses are nearly Fourier transform limited in frequency space and diffraction limited in coordinate space.

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Narrowband, high powered pulsed dye lasers are finding many uses in multiphoton high resolution spectroscopy, Raman scattering, coherent optics and other rapidly growing fields in non-linear spectroscopy, and high powered Nd:YAG lasers with unstable resonators [1] are attractive pump sources for such dye lasers. We have constructed a dye laser oscillator-amplifier using the doubled output of a Q-switched Nd:YAG laser (Quanta Ray DCR oscillator) to pump a system that amplifies the output of a frequency stabilized cw dye laser (Coherent Inc. Model 599-21). The basic design is very similar to the Hänsch-Salour type [2] with modifications that the increased pumping power necessitated. The high conversion efficiency attainable from a YAG pump combined with the narrow linewidth from the oscillator have enabled us to construct a dye laser with near Fourier transform limited pulses and peak intensities of several megawatts.

The higher harmonics of a Q-switched Nd:YAG provide certain advantages over N_2 or eximer laser pump sources. High peak powers of tens of megawatts or more at 532 nm, 355 nm and 266 nm wavelengths are now commercially available, and one can select the harmonic that best matches the absorption bands of the dye to be used. In particular the second-harmonic of YAG at 532 nm is an excellent pump wavelength for the Rhodamine dyes.

Concentrations of approximately 120 mg/l of R6G (2.5×10^{-4} M/l) are appropriate for transversely pumped dye cells, and are roughly an order of magnitude smaller than the concentrations needed for N_2 pumped dye lasers. The combination of reduced self-absorption in the dye cells and better pump laser beam characteristics allows much more efficient conversion from pump energy input to dye laser output. We have seen as much as

40-45% conversion efficiency from the 532 nm pump to the output of a single-pass transversely pumped R6G amplifier stage.

The expected quantum efficiency can easily be estimated for a traveling-wave dye laser amplifier [2], [3]. The spontaneous and stimulated emission compete for excited dye molecules, and are equally probable at the input laser intensity $I = I_{\text{sat}}$, where I_{sat} is the saturation intensity, a characteristic of the dye medium and operating wavelength. The local quantum efficiency for the conversion of pump photons into laser photons is approximately $\eta = 1/(1 + I_{\text{sat}}/I)$, and when averaged over the length of the dye cell,

$$\langle \eta \rangle \approx 1 - \ln g / \ln g_0$$

where g_0 is the small signal gain, and g is the gain of the amplifier stage. The small signal gain can be measured using the cw dye laser and is roughly 5×10^4 , while the gain in the second stage is measured to be on the order of 500. This yields a quantum efficiency of $\langle \eta \rangle \sim 0.43$. Correcting for the wavelength difference between the pump light and amplified dye laser light we would expect an energy conversion efficiency of roughly 40%, in reasonable agreement with our measured quantum efficiency.

Another attractive feature of the YAG pump is the excellent spatial quality of the beam. In the final amplifier stage pump powers in the tens of megawatts range make transverse pumping impractical since large amplifier beam cross sections are needed to avoid amplified spontaneous emission. However, one can longitudinally pump without focussing tightly, and the dye laser beam will take on the spatial characteristics of the near-diffraction limited pump beam.

Figure 1 shows a schematic of the laser system. Most lenses are of Edmund Scientific quality and the choice of many of the focal lengths was dictated by what happened to be on the shelf. The Faraday optical isolator is needed to reject the amplified spontaneous emission that otherwise gets back into the reference cavity of the feedback system for the cw dye laser. An extinction of roughly 200:1 is adequate to keep the back traveling pulse intensity smaller than the dc light levels in the reference cavity so that it does not disturb the frequency locking mechanism.

Because of the high gains found in a YAG pumped dye laser, amplified spontaneous emission can be a problem and some precautions have to be taken. All dye cells are set at Brewster's angle with respect to the vertical polarization of the cw laser beam. Instead of the conventional wedged cells with antireflection surfaces, our transverse pumped dye cells are made from a standard quartz spectrophotometer cell (from Precision Cells, 10 mm $\times$ 20 mm cross-section) cut in half and glued with RTV to a stainless-steel body. (Fig. 2). This construction avoids the delicate glue joints at the front surface of the dye cuvette that could scatter some of the amplified cw beam. Magnetically stirred cells were tried briefly but were found to be inferior to a flowing dye cell. Also, the pinhole between the first and second amplifier stages and some of the lenses are tilted with respect to the beam axis to prevent the amplification of spontaneous emission.

A 33 cm f.l. lens focuses the 18 mw cw beam to a .14 mm diameter waist in the first amplifier cell. Four percent of the YAG pump beam is picked off by mirror substrate and focussed into the first amplifier cell with a

20 cm f.l. cylindrical lens set approximately 21.5 cm from the dye cell. The cell, sharing a common dye reservoir with the second stage amplifier, contains a 120 mg/l solution of R6G. The single pass gain from the first stage is typically 5×10^4 . Conventional spectral and spatial filtering follows the first stage amplifier [2], and ratios of amplified cw light to background fluorescence measured by blocking and unblocking the Argon ion laser, range from 200 to 1000 to 1. Without a direct vision prism and pinhole the ratio of amplified cw light to fluorescence is roughly 10 to 1.

The lenses between first and second stages are adjusted to give the best possible mode structure after the second stage. The second stage cell is placed about 2 cm after the waist formed by the 5 cm lens after the pinhole, and the cell is transversely pumped with 16% (10mj) of the YAG that is beam split off using a microscope slide. A path delay is chosen for the pump light into the second amplifier stage to assure that the pump pulse arrives slightly after the pulse from the first stage. The pump light is focussed by a 25 cm focal length lens 23.5 cm from the cell. The focal lengths of the cylindrical lenses are not critical since one typically works in a slightly defocused arrangement. The output of the second stage is about 3mj in a beam with circular, uniform cross-section and the ratio of amplified cw power to fluorescence background, measured with and without the Argon laser blocked, varies from 10:1 to 50:1. Higher powers can be obtained by using a larger diameter beam, but diffraction of the beam by the dye cell gives much poorer mode quality.

The second stage output is beam expanded and collimated with a telescope to produce a 2.5 mm diameter parallel beam in the third stage. The longitudinally pumped cell is a 3/8" thick stainless steel plate with a

hole milled in it for the beam to pass with two high quality quartz flats glued on with RTV. The cell is held at Brewster's angle for the vertical polarizations of both the pump and the amplified cw beam. The concentration in the third stage is about 25 mg/l and a filter is placed between the cell and the reservoir to eliminate air bubbles in the flow. The pump beam is focussed with a 1.5 m focal length lens 1.2m from the cell in order to match the diameter of the beam from the second stage. Tighter beam diameters in the third amplifier do not give higher output powers and breakdown of the quartz windows due to the intense radiation at 532 nm becomes a problem. The output of the third stage is about 20mj for a total pump power out of the YAG laser of 60mj.

The linewidth of the laser is measured by scanning the cw oscillator over several GHz and analyzing the dye laser output using a Tropol Spectrum Analyzer (Model 240) with a 1.5 GHz free spectral range. The output of the Tropol goes into a boxcar integrator followed by a chart recorder. The scan is shown in Fig. 3 and indicates a dye laser bandwidth of about 80 MHz.

The temporal character of the 532 nm pump pulse is shown in Fig. 4a, and the dye laser pulse is shown in Fig. 4b. The pulse is measured using an ITT F4018 vacuum photodiode and a Textronix 519 oscilloscope with a combined rise time less than 0.5 nsec. The fwhm spread of 4.5 nsec indicates that the pulse is close to the Fourier transform limit. Although the pulse shape of the dye laser amplifier is irregular and fluctuates from pulse to pulse, the RMS energy jitter from pulse to pulse is about 5%, fwhm.

High doubling efficiency of the laser output is possible because of the high power, narrow linewidth, and near diffraction limited divergence. Using an angle tuned KDP crystal, (Cleveland Crystals 12mm x 12mm x 30mm) conversion efficiencies of 30% - 40% are obtained in doubling the 570nm output. Doubling in the far field region of the unfocussed dye laser beam, we typically get 6mj/pulse of UV light from 15mj/pulse of visible light.

Although the laser system will be used in an experiment to search for parity non-conservation in atomic thallium [3], some of its characteristics can be demonstrated using standard laser spectroscopic techniques. Transitions between the $6^2P_{1/2}$ ground state and $7^2P_{1/2}$ excited state in thallium are possible by two photon transitions, or by single photon transitions due to a highly forbidden M1 amplitude or possibly by weak neutral interactions mixing states of opposite parity. Using an electric field of 400 V/cm to mix opposite parity states, single photon transitions, although still 10^8 times weaker than normal electric dipole transitions, are easily seen in an experimental arrangement similar to one previously described [4]. Selection rules allow $F=0 \rightarrow F'=1$ and $F=1 \rightarrow F'=1$ and 0 transitions between the hyperfine levels in thallium for linearly polarized light perpendicular to the static electric field, and Fig. 5a shows a scan of the dye laser revealing the 21 GHz hyperfine splitting of the $6^2P_{1/2}$ state and the 2 GHz hyperfine splitting of the $7^2P_{1/2}$ state. At cell temperatures of 1100°K, the doppler width is about 1.7 GHz and the pressure broadening is estimated to be less than 10 MHz. For the scan shown, Rhodamine 610 dissolved in methanol is used and the laser is operated at the somewhat reduced power

outputs of 1.5 mj/pulse in the doubled frequency. The theoretical intensity ratios of 2:1:1 for the $F=1 \rightarrow 1$, $1 \rightarrow 0$, and $0 \rightarrow 1$ transitions, [4], are in only rough agreement, primarily because the doubling efficiency as we scan to the $F=0 \rightarrow 1$ transition is not constant. (The background is proportional to the amount of UV light incident on the thallium cell and provides a convenient indication of the total UV light going into the cell.) Higher resolution is possible if we use doppler free two-photon techniques, [5], and Fig. 5b shows the result of a scan of the laser around the $F=0 \rightarrow 0$ transition clearly revealing an isotope shift between Tl^{205} (70.5% abundant) and Tl^{203} (29.5% abundant) of approximately 1.27 GHz, and in good agreement with a previous measurement of 1.266 GHz using a cw dye laser [6]. Note that the linewidth of the doppler free transition is roughly 200 MHz; considerably larger than the measured linewidth using a spectrum analyzer.

The increased linewidth is due to the incomplete overlap of the 5 nsec pulse going into the cell and the counter propagating pulse reflecting from a mirror placed 40cm from the thallium cell. Approximating the temporal shape of the laser pulse as a gaussian $e^{-t^2/2\sigma^2}$, the two photon signal will be proportional to

$$S \sim I_1 I_2 \sim \exp[-t^2/2\sigma^2] \cdot \exp[-(t+\Delta t)^2/2\sigma^2]$$

where Δt is the round trip delay of the light from the thallium cell to the mirror and back to the cell. Rewriting the above expressions,

$$S \sim \exp[-(t + \Delta t/2)^2/\sigma^2] \cdot \exp[-\Delta t^2/4\sigma^2].$$

The magnitude of the signal is suppressed by the second factor and the effective width of the overlapping pulses, and hence the time taken to

complete the measurement, is decreased by $\sqrt{2}$. From the uncertainty principle we would expect the bandwidth to increase by $\sqrt{2}$. An additional $\sqrt{2}$ should come from the fact that we are using two photons in our measurement, each with an energy spread of $\hbar\Delta\omega$, so that one can interpret the 200 MHz linewidth in Fig. 5b as indicative of a roughly 100 MHz bandwidth.

In summary, visible peak powers obtained with our YAG pump are approximately a factor of 40 higher than the results previously reported using a 1 MW N_2 laser, and UV power output is increased by 2-3 orders of magnitude [2]. Furthermore we believe that these results can be easily scaled to higher peak powers and narrower linewidths. Pump energies in excess of 200mj/pulse are commercially available with YAG oscillator-amplifier systems, and trivial modifications of our system should allow over 60mj/pulse in a Fourier transform limited pulse in the visible, and over 15mj/pulse in the UV. Also, by extending the cavity length of the YAG oscillator or pumping closer to threshold, the pump pulse can be extended to 20nsec or more and linewidths 20 MHz or less should be generated.

We wish to acknowledge helpful discussions with Dick Herbst, Carl Weiman, Bob Byer, Ted Hänsch, Alex Jacobson, and Gene Commins, and thank Dave Dolfi for the loan and help with the fast oscilloscope and photodiode. This work was supported in part by the Division of Chemical Sciences, Office of Basic Energy Sciences, U.S. Department of Energy and in part by an N.S.F. predoctoral fellowship (P.D.) and an N.S.F. postdoctoral fellowship (S.C.).

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FIGURE CAPTIONS

Fig. 1. Laser schematic.

Fig. 2. Detail of the transversely pumped flowing dye cell.

Fig. 3. A portion of a scan of the cw oscillator-amplifier system as analyzed by a Tropel Model 240 Spectrum Analyzer (free spectral range 1.5 GHz) with no ramp voltage applied to the spectrum analyzer.

Fig. 4a. Pulse shape of the doubled YAG pump at 532 nm. The sweep speed is 5 nsec/div.

Fig. 4b. Pulse shape out of the dye laser amplifier with the same sweep speed.

Fig. 5a. A scan of the laser oscillator-amplifier system at 292.7 nm showing the 21 GHz $6^2P_{1/2}$ ground state hyperfine splitting and the $7^2P_{1/2}$ upperstate hyperfine splitting. The calibration is provided by a Tropel 240 Spectrum Analyzer. The signal is the 535 nm fluorescence from the $7^2S_{1/2} \rightarrow 6^2P_{3/2}$ transition in thallium that follows the $6^2P_{1/2} \rightarrow 7^2P_{1/2}$ transition.

Fig. 5b. The resolved isotope shift of the $6^2P_{1/2} \rightarrow 7^2P_{1/2}$, $F=0 \rightarrow 0$, transition in thallium using 2 photon spectroscopy. The relative intensities are in good agreement with the relative abundances of Tl^{205} and Tl^{203} .

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Figure 1

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Figure 2

Figure 3

Figure 4a

Figure 4b

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Figure 5a

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Figure 5b

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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