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# A High-Power Subpicosecond Distributed Feedback Dye Laser System Pumped by a Mode-Locked Nd:YAG Laser

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**Abstract.** The design and operation characteristics of a distributed feedback dye laser (DFDL) system pumped by the second harmonic of a flashlamp pumped mode-locked Nd:YAG laser are described. The DFDL oscillator facilitates a large tuning range with nearly Fourier limited pulse durations of about 1.6 ps. The combined action of saturated absorption and amplification results in a pulse shortening to about 600 fs, with small fluctuations in the pulse duration. Output pulse energies of more than 400 μJ are achieved, corresponding to a peak power of more than 650 MW. Since the dye amplifiers are pumped by pulses of only 25 ps duration the amplified spontaneous emission (ASE) is very low, typically less than $10^{-4}$.

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In recent years the growing interest in studying nonlinear optical phenomena with high time resolution has spurred the development of high peak power, subpicosecond laser sources. Usually, such systems rely on the multistage amplification of a separate low intensity laser, which generates the desired ultrashort pulses, e.g. colliding pulse mode-locked (CPM) or hybrid mode-locked dye lasers [1]. Although such laser systems deliver high peak power, they generally lack tunability over a large spectral range. This tunability together with a short pulse duration can be achieved with a distributed feedback dye laser (DFDL) [2–4]. Pulse durations of less than a picosecond [5, 6] and tunability across the entire visible spectrum [7, 8] have been demonstrated. The principle of operation of a DFDL has been described in detail previously [2–9]. The important feature is the creation of a high visibility gain grating in the active medium, which satisfies the Bragg condition, $\lambda_L = 2n\,\Lambda$, where $\lambda_L$ denotes the lasing wavelength, $n$ the refractive index of the medium, and $\Lambda$ the grating period. Due to the rapid increase of the gain grating feedback during pumping and its decrease after onset of laser action, the DFDL displays a self-$Q$-switching effect, which results in an output pulse with considerably shorter duration than the pump pulse; see e.g. [9]. A major refinement of this laser source for practical applications was achieved by Szatmari and Schäfer, by introducing a combination of transmission grating and a microscope objective to form the gain grating in the active medium [8]. A simple translation of the transmission grating along the pumping beam leads to a change of the grating period, and thus to a tuning of the laser wavelength $\lambda_L$.

Stable, single-pulse operation of the DFDL is achieved only close to the threshold. The resulting low intensity output therefore has to be amplified in several stages for high power applications. Unlike conventional systems a single pump laser source can be employed to create the short pulses and provide the amplification to high power levels [6, 10, 11]. In this paper we report the performance of a comparatively simple laser system. A mode-locked Nd:YAG laser is used as the pump light source [4] for the tunable, amplified DFDL operating in the red spectral region. We employ difference frequency mixing between this radiation and the residual fundamental of the Nd:YAG laser for the generation of tunable, picosecond IR radiation.

## 1. Experimental Arrangement

The system consists of a commercial flashlamp pumped, active/passive mode-locked Nd:YAG laser oscillator (Quantel YG 501) as primary laser source. The oscillator

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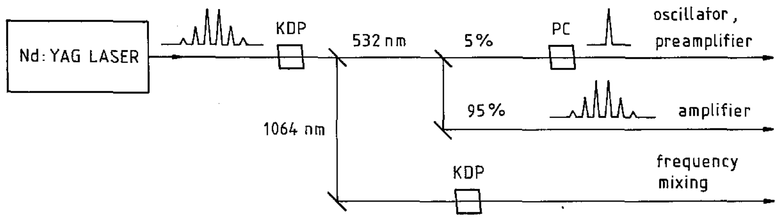


**Fig. 1.** Schematic diagram of the pump laser beams derived from the mode-locked Nd:YAG laser

is operated with Kodak 9740 saturable absorber, and delivers a train of six pulses separated by about 6.7 ns at 10 Hz repetition rate. This pulse train is amplified in a double pass amplifier (115 mm × 7 mm diameter) yielding a pulse energy in the infrared of about 45 mJ for the whole train. After frequency doubling this radiation in a KD*P crystal, about 20 mJ at 532 nm is available, with an energy distribution in the six pulses of about (1.6:4.1:5.7:4.9:2.8:0.8) mJ. A pulse duration of about 25 ps is measured for a single green pulse. As shown schematically in Fig. 1, about 5% of this green pulse train is sent through a Pockels cell to select a single pulse. This single pulse, which is the third pulse of the train, is used to pump the distributed feedback dye laser oscillator and the preamplifier. The pulse train provides the pump energy for the main amplifier. The fundamental radiation remaining after frequency doubling is coupled out by a dichroic mirror and is again frequency doubled in a second KD*P crystal. The resulting 1 mJ in the most intense pulse of the train is used for difference frequency mixing with the dye laser to yield tunable, short-pulse IR radiation.

### *Distributed Feedback Laser Oscillator*

Tunable picosecond laser radiation is generated in a DFDL oscillator by the single pulse selected by the electro-optic switch, of which 20% is used to pump the dye oscillator. After being passed through a variable attenuator, an adjustable slit provides the desired beam dimensions to create an active volume of about 100 μm × 10 μm × 10 μm in the gain medium. This pump laser beam is split by a transmission grating (55 lines/mm) into two coherent beams. Both beams, the +1 and −1 diffraction order of the grating, are imaged with a lens $L_1$ and a microscope objective (Leitz, 63/1.30) into the dye cell to produce an inversion grating.

This arrangement was adapted from Szatmári and Schäfer [8], and provides a simple means to tune the emission wavelength. By changing the distance between the transmission grating and the dye cell the interference angle of the two pumping beams is varied and thus the period $\Lambda$ of the inversion grating is changed in the dye cell. The emission wavelength $\lambda_L$ of the laser is connected with this grating period by

$$\lambda_L = 2n\Lambda, \quad (1)$$

where $n$ denotes the refractive index of the active medium relative to air. This grating period in turn is given by

$$\Lambda = \lambda_p (2n \sin\alpha)^{-1}, \quad (2)$$

where $\lambda_p$ is the wavelength of the pump laser and $\alpha$ the angle of the interfering pump laser beams.

For focussing the pumping radiation at 532 nm with the mentioned microscope objective, the transmission grating should have 36.6 lines/mm to create a gain grating period which permits laser action in the red spectral region. Since we used a 55 lines/mm transmission grating, the grating has to be imaged by a lens L ($f_L = 200$ mm) to yield the desired grating period. The emission wavelength $\lambda_L$ of the distributed feedback laser is then given by

$$\lambda_L = nd \frac{f_L}{f_L - l_1} \cdot f_\mu \left( \frac{f_L l_1}{f_L - l_1} + l_2 - f_\mu \right)^{-1}, \quad (3)$$

where $n$ denotes the refractive index of the dye solution, $d$ is the period of the transmission grating, $f_L$ and $f_\mu$ represent the focal lengths of the imaging lens L and the microscope objective, respectively. The distance between the transmission grating and the lens L is given by $l_1$ and that between this lens and the principal plane of the microscope objective by $l_2$.

### *Dye Laser Amplifiers*

The output of the DFDL oscillator is imaged by a planoconvex lens ($f = 25$ mm) into the preamplifier operating with a solution of $2 \times 10^{-3}$ mol/l DCM in methanol. This active medium with a length of 9 mm is transversely pumped by the vertically polarized single pulse with about 45 μJ pulse energy. After preamplification the dye laser radiation is sent through a saturable absorber cell (dye: nile blue in methanol, small signal transmission about $10^{-3}$). The saturable absorber not only reduces possible amplified spontaneous emission but also produces a steep leading edge on the transmitted pulse.

The radiation is then directed anti-collinearly through the main amplifier cell (length: 10 mm with $1.8 \times 10^{-4}$ mol/l cresyl violet dissolved in methanol as the active medium. This dye cell is pumped longitudinally by the whole pulse train, as shown in Fig. 2. The pumping laser pulse train is optically delayed to facilitate the overlap of the first pulse of the train with the preamplified dye laser pulse, which is pumped by the third Nd:YAG laser pulse. Only the four largest pulses of the train are used for consecutive amplification. Time synchronization of the following pulses is now achieved by a corresponding delay of the dye laser pulse. After the second pass through the main amplifier another saturable absorber cell (length: 8 mm, dye: nile blue in methanol, small signal transmission about $10^{-4}$) reduces possible amplified spontaneous emission.

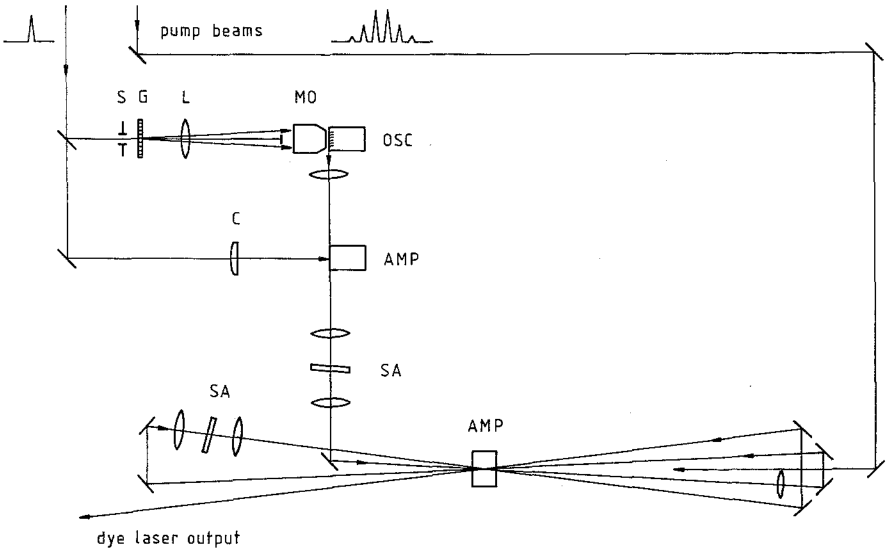


**Fig. 2.** Schematic diagram of the set-up for the oscillator, preamplifier, and multiple path main amplifier

*Infrared Generation*

The amplified dye laser pulse and the 532 nm radiation derived from the remaining Nd:YAG laser fundamental (Fig. 1) as directed through a 15 mm long $LiIO_3$ crystal to produce IR radiation by type I difference frequency mixing. The pulse energy of the generated IR radiation is measured by a calibrated pyroelectric detector (Eltec 420).

## 2. Results and Discussion

Figure 3 shows the tuning behaviour of the distributed feedback dye laser oscillator, as the transmission grating for the pumping radiation is moved along the pump laser beam direction. At a distance of the grating of 163 mm from the dye cell the emission wavelength is $\lambda_L = 635$ nm. Decreasing this distance by 13 mm leads to an increase of the emission wavelength to $\lambda_L = 670$ nm, resulting in a tuning constant of 2.69 nm/mm. The dashed line in Fig. 3 represents the tuning behaviour expected from (1), when a refractive index of $n = 1.43$ for the dimethylformamide (DMF) solvent is used. The figure clearly shows that the

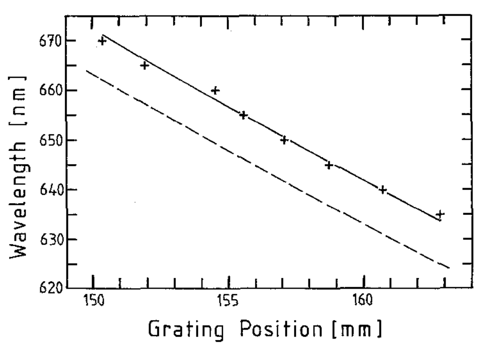


**Fig. 3.** Tuning behaviour of the distributed feedback dye laser oscillator for DCM dissolved in DMF. The dashed line shows the tuning behaviour as expected from (3) when the refractive index $n$ of the pure solvent is used, the full line accounts for a correction of $n$ by 1%

measured data are consistently shifted to longer wavelengths. Such a shift can be expected, since a saturated solution ($\sim 5 \times 10^{-2}$ mol/l) of DCM in DMF is used. Increasing the refractive index of the dye solution by only 1% leads to good agreement between the theoretically calculated emission wavelength and the experimental data (full line in Fig. 3).

A stable operation of the DFDL oscillator with single pulse emission is obtained when pumping it at about 1.2 times above the lasing threshold. In our case this condition is met at a pump pulse energy of about 80 nJ. The oscillator then emits pulses with about 1.5 nJ energy. These pulses are amplified in the preamplifier up to 1.4 µJ pulse energy. At this energy level the pulse width could be measured by an autocorrelator which yielded a pulse duration of 1.6 ps. If no pulse broadening occurs in the preamplifier, then this should also represent the pulse duration of the oscillator. The self-$Q$-switching effect thus leads to a pulse shortening with respect to the pump laser pulse of about a factor of 16. This pulse shortening is of the same magnitude as observed by others [8, 9].

The saturable absorber behind the preamplifier reduces the pulse energy to 0.25 µJ. By using a cell of 1 mm thickness for the saturable absorber, an asymmetric pulse shape with a steep leading edge accompanied by a corresponding spectral broadening is created. This results in a shortening of the pulse duration as the pulse is further amplified, because the diameter of the dye laser beam in the main amplifier is adjusted so that the amplification is saturated in all four passes. When the amplification is not saturated or thinner dye solutions are used in the saturable absorbers, a pulse lengthening occurs in the amplifier chain, as expected. The pulse duration of the dye laser is measured with both a conventional background free multishot and a background free single shot autocorrelator [4, 12]. Figure 4 shows a typical autocorrelation trace of the final pulse obtained with the single shot device. The width of the autocorrelation curve shown is 900 fs (FWHM). Assuming a Gaussian pulse shape a pulse duration of 636 fs is derived, and 583 fs for a $sech^2$-pulse. The laser produces clean pulses as shown by the absence

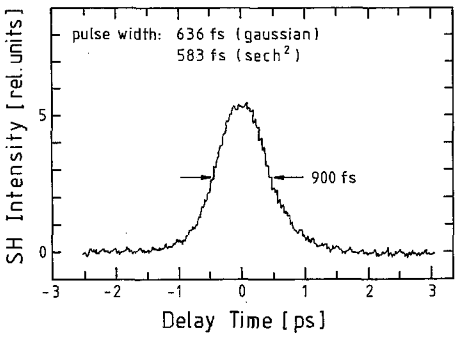


**Fig. 4.** Typical background free autocorrelation signal measured with the single pulse autocorrelator. (The apparent noise on the trace results from electrical overshoots between adjacent diodes in a 1024-element diode array)

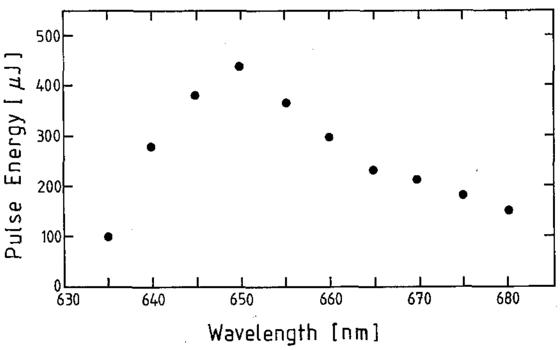


**Fig. 5.** Output pulse energy of the laser system at different wavelengths

of wings in the autocorrelation trace. This is a typical feature of this type of laser. The single shot autocorrelator enables us to perform a statistical characterization of the fluctuations in pulse duration and amplitude. Relatively large amplitude fluctuations of 33% (rms) are found, whereas the fluctuation of the duration is less than 10% (rms).

In Fig. 5 a tuning curve of the laser system is shown without changing the concentration in the dye solution for the amplifiers or the saturable absorbers. The system is obviously optimized at $\lambda = 650$ nm with a pulse energy of more than 400 μJ. This corresponds to a pump laser energy conversion efficiency of more than 8% for the last stage. It should be noted that the laser shows a very low level of amplified spontaneous emission of about $10^{-4}$. The pulse width remains constant in a spectral range of about 10 nm around the maximum. It lengthened up to 1.5 ps at the ends of the tuning range. This increase in pulse duration is caused by the lower gain in the main amplifier which leads to incomplete saturation of the amplification, so that the pulse chirp introduced by the saturable absorbers cannot be compensated completely. Table 1 summarizes the characteristics of the laser system at various stages of amplification. The time-bandwidth product $(\Delta t \Delta \nu)$ is also given. Its variation at various amplification stages indicates the presence of phase modulation on the pulse. The $(\Delta t \Delta \nu)$ value of 0.68 for the final pulse is about twice the Fourier limit, assuming a Gaussian pulse shape.

Table 1. Characteristics of the dye laser system [$\lambda_L = 645$ nm, $\lambda_P = 532$ nm (25 ps)]

| Stage | Pump pulse energy [mJ] | Pulse energy [μJ] | Efficiency [%] | Gain | Pulse duration [ps] | $(\Delta t \Delta \nu)$ |
|---|---|---|---|---|---|---|
| Oscillator | $80 \times 10^{-6}$ | 0.002 | 2.5 | – | – | – |
| Preamplifier | 0.045 | 1.35 | 3.0 | 675 | 1.6 | 0.81 |
| 1st absorber | – | 0.25 | – | 0.17 | 1.3 | 0.94 |
| Main amplifier: | | | | | | |
| 1st pass | 1.6 | 6.75 | 0.4 | 30 | 1.4 | 1.31 |
| 2nd pass | 4.1 | 90 | 2.2 | 13 | 1.2 | 0.91 |
| 2nd absorber | – | 70 | – | 0.77 | 0.93 | 0.74 |
| 3rd pass | 5.7 | 170 | 3.0 | 2.4 | 1.0 | 0.43 |
| 4th pass | 4.9 | 390 | 8.0 | 2.3 | 0.62 | 0.68 |

The laser system described facilitates a comparatively simple generation of tunable, ultrashort IR pulses by frequency mixing the dye laser with the mode-locked Nd:YAG laser radiation or its harmonics. Using a single pulse of 532 nm radiation (pulse duration 25 ps) with about 1.6 mJ pulse energy, tunable IR radiation around $\lambda = 2.7$ μm can be obtained from a dye laser operating at 660 nm. Optimizing the laser for this wavelength gives an IR pulse energy of about 30 μJ after mixing in a 15 mm long $LiIO_3$ crystal. This corresponds to a photon conversion efficiency of about 30%. The pulse energy is limited by the power available in the green pulse, which is depleted by the dye laser pulse. With such a long mixing crystal a broadening of the IR pulse to a length of 2.2 ps due to group velocity dispersion occurs, as is measured by a cross correlation with the visible pulse on GaAs [13] and Pd (111) [14] surfaces.

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