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#### KILOHERTZ PICOSECOND DISTRIBUTED-FEEDBACK DYE LASER SYSTEM

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Dual distributed-feedback oscillator-amplifier systems, pumped by a *Q*-switched, mode-locked, cavity-dumped Nd: YAG laser, provide independently tunable, Fourier-transform-limited, synchronized picosecond pulses at megawatt power levels with a repetition rate in kilohertz range.

#### 1. Introduction

The requirement for kilohertz repetition rate, microjoule energy range, tunable picosecond pulses has led to the development of correspondingly high repetition rate pump sources for dye amplification of cw mode-locked dye laser output. These pump sources include *Q*-switched, mode-locked, and cavity-dumped Nd: YAG or Nd: YLF lasers [1–4].

However, these systems are inherently complex as they need two pump lasers (one for the cw dye oscillator and another for the amplifiers) and, in the optimal case of short amplifier pump pulse width, necessitate the precise synchronization of the amplifier pump and the dye oscillator pulses. Further difficulties may arise if two independently tunable amplified pulse trains are required, as temporal jitter [4] between the two trains may cause problems in certain experiments.

We present here a novel system providing two independent, one kilohertz repetition rate, amplified, tunable, picosecond pulse trains, which are essentially Fourier-transform limited and jitter free. This system is based on a single *Q*-switched, mode-locked and cavity-dumped Nd: YAG laser which drives twin distributed-feedback dye laser (DFDL) oscillator-amplifier chains. Because the DFDL requires no ex-

#### 2. The pump source

The Nd:YAG laser system is drawn schematically in fig. 1. The double lamp continuously-pumped YAG head (Microcontrôle) is placed between two

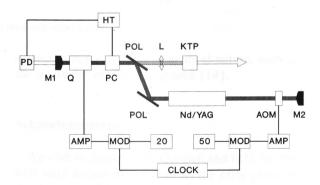


Fig. 1. Schematic of the cavity-dumped, mode-locked, Q-switched, continuously-pumped Nd:YAG laser. PD: photodiode. HT: high-voltage triggered Pockels driver.  $M_1$ : 800 mm radius 100% convex mirror. Q: acoustic-optic Ar-coated Q-switch. PC: lithium niobate AR coated Pockels cell. POL: dielectric polarization-sensitive mirrors. L: lens. KTP: frequency doubling crystal. Nd:YAG:  $4\times100$  mm YAG rod. AOM: Brewster angle acousto-optic mode-locking crystal.  $M_2$ : 1200 mm radius 100% convex mirror. AMP: rf amplifier. MOD: rf modulator. CLOCK: 1kHz 8 and 80  $\mu$ s synchronization pulses for the rf drivers. The numbers in boxes give the rf frequencies employed in MHz.

ternal mirrors the experimental set-up is also spatially compact.

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divergent reflectors of 800 and 1200 mm radius of curvature, at a distance of 600 mm left and 900 mm right respectively. This leads to a moderately large mode volume inside the YAG rod mode diameter decreasing towards each reflector. Laser emission is polarized by two dielectric polarizers in reflection mode. With a right mirror transmission of 10% and no other elements in the cavity, polarized laser output is 16 W TEM $_{00}$ .

The laser is Q-switched and mode-locked at 50 MHz by quartz acousto-optic elements. The Q-switch is operated at a frequency of between 1 and 2 kHz with an open time of 8  $\mu$ s. The mode-locking rf of 3 W power is pulsed on during 80  $\mu$ s in end-synchronism with the Q-switch. Good quality mode-locking and pulse-train energy depend critically on the level of prelasing (intertrain lasing) which may either be adjusted electronically or by orienting the Q-switch.

A single pulse is switched out of the Q-switched, mode-locked pulse train using an antireflection coated lithium Pockels cell. The Pockels cell is driven to quarterwave voltage in 5 ns, just after the passage (from left to right) of a laser pulse, using a high voltage triode triggered by a photodiode detecting end-leakage from the cavity. The switched-out pulse exists the cavity through the dielectric polarizer.

With 100% end-reflectors, the single pulse energy is 800  $\mu$ J at 1 kHz (0.8 W) and 500  $\mu$ J at 2 kHz (1.0 W). Infrared pulse duration is 70 ps due to the high peak rf power in the mode-locking crystal. The TEM<sub>00</sub> output is frequency doubled to 532 nm by slightly focussing into a KTP crystal to obtain second harmonic energies of 450  $\mu$ J (0.45 W) at 1 kHz and 300  $\mu$ J (0.6 W) at 2 kHz. Second harmonic duration is 65 ps in a clean pulse with no detectable leading or trailing satellites. Output energy is remarkably stable, exhibiting short-term fluctuations of the order of  $\pm$ 2%. The laser is normally operated at 1 kHz.

The energies cited above are limited by two factors. The first is the fragility of the antireflection coatings of the lithium niobate crystal which in our system will damage at power levels more than 50% over the stated operating point. Interruption of cavity dumping will also damage the Pockels cell in a few seconds. The second limitation is caused by pump-induced birefringence in the YAG rod which constrains the usable mode volume. Both these restrictions could be somewhat eased by employing

Nd:YLF as the active medium, as the lower straininduced birefringence of this material allows larger beam diameters.

#### 3. The dye laser system

The picosecond dye lasers are based on a modified version of the distributed feedback oscillator described in ref. [5]. The oscillator is schematically drawn in fig. 2. The microjoule energy pump beam at 532 nm, with a duration of 65 ps, is divided into two equal-delay equal-intensity beams which interfere in the dve cell after deviation by a prism and reflection from a rotatable mirror. The rotation axes of the mirrors chosen to minimize beam movement at the dye cell while tuning the laser. A 60° quartz coupling prism is fixed to the quartz dye cell using Canada balsam. The beams are focussed into the dye using either a cylindrical or spherical lens, depending on the required excitation length. Production of short pulses demand short pumped lengths and we employ a spherical lens with the focus on the dye cell wall. In this case the transverse dimension may be adjusted by rotating the two prisms away from the minimum deviation position.

The principle of operation of the DFDL has already been described in detail elsewhere [5–7] and we simply recall here that the total gain of the system

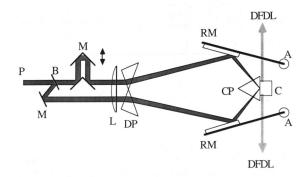


Fig. 2. Outline of the distributed-feedback dye oscillator. P: microjoule energy 65 ps 1 kHz pump at 532 nm. M: 100% plane reflector. B: 50% beam splitter. L: cylindrical or spherical focussing lens. DP: orientable beam-deviation prisms. RM: co-rotating 100% tuning mirrors. CP: coupling prism. C: dye circulation cell. A: rotation axes of co-rotating mirrors. DFDL: twin dye laser output beams.

and the transient induced distributed grating reflectivity vary in a very non-linear fashion with the inversion of the dye medium. Depending on pumping intensity, this leads to the emission of one or more dye laser pulses, which in general are significantly shorter than the pumping pulse width. Because the DFDL pulse evolves inside a high-visibility distributed reflection grating, this pulse is usually near Fourier-transform limited, regardless of its temporal width [5].

The duration of the emitted tunable DFDL pulse depends strongly on the length of the interference fringe system inside the dye cell (and implicity on dye concentration which must be adapted to the pumping level [5]). With a pumped length of 8 mm, obtained using a cylindrical lens, and an inversion close to first threshold, we obtain 25 ps long pulses. These long pulses may be amplified up to energies in excess of 10 µJ by a two stage amplifier (fig. 3, without saturable absorber) pumped by 10% (pre-amplifier) and 35% (main amplifier) respectively of the single pulse doubled YAG output. Small signal gain of the amplifier chain is of the order of 104. To reduce thermal effects in the amplifiers and the DFDL oscillators we normally use a 70/30% water/ethanol mixture as a solvent for the dyes. Long pulse tunability from 550 to 650 nm can be readily achieved

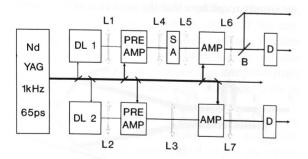


Fig. 3. Simplified schema of the complete dual dye laser system. Nd: YAG: 1 kHz 65 ps 450  $\mu$ J pump at 532 nm. DL 1, 2: independent distributed-feedback dye oscillators. L1, L2: 70 mm focal length DFDL beam collector lenses. PRE-AMP: 10 mm long dye pre-amplifiers pumped by 10% of the YAG energy. L3: 1000 mm coupling lens. L4, L5: 50 mm focussing lenses for the saturable absorber. SA: 1 mm long saturable absorber cell. AMP: 20 mm long dye amplifiers pumped by 35% of the YAG energy. L6, L7: 1000 mm collimating lenses, B: beam-splitter. D: adjustable delay lines. In the real system the amplifiers are forward pumped at a small angle to the dye laser beam.

using appropriate dyes.

For a DFDL pumped length of 0.5 mm excited to just below the threshold for second spike generation the output pulse width is about 10 times shorter than above. With a high-gain amplifier system and in the absence of a saturable absorber this pulse is broadened to approximately 4 ps after amplification. The introduction of a saturable absorber between the preamplifier and the main amplifier (fig. 3) and avoidance of excessive gain saturation allows the production of amplified pulses less than 2 ps in duration with an energy greater than 2  $\mu$ J (peak power>1 MW).

Fig. 4 shows the autocorrelation trace, measured by non-linear second harmonic generation, of such an amplified pulse, where we plot the logarithm of the autocorrelation signal  $S_A(t_d)$  as a function of relative delay  $t_d$  in picoseconds. Laser wavelength was 572 nm, obtained using rhodamine 6G in the oscillator and amplifiers and DODCI in ethanol as saturable absorber. The autocorrelation curve yields a pulse duration of 1.8 ps, assuming a sech pulse shape [5], as suggested by the observed shape and exponential wings of the autocorrelation trace. Any slight asymmetry of the pulse which might be introduced by the saturable absorber could only be detected in a higher order correlation experiment (see below) and should have a negligible influence on the deduced pulse-width. Because dye concentrations are required to be high for short pulse generation in or-

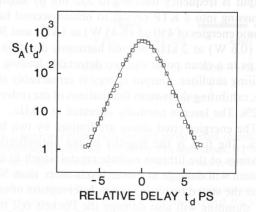


Fig. 4. Auto-correlation trace of amplified dye laser 1 for a 0.5 mm DFDL pumped length. The auto-correlation signal is plotted on a  $\log_{10}$  scale against relative delay in picoseconds. The full line is calculated for a pulse width of 1.8 ps assuming a pulse shape of the form  $\operatorname{sech}(t/t_{\nu})$ .

der to support adequate gain over small pumped length, self-absorption of the laser wavelength becomes an important factor and it is necessary to pump the dye close to the edge of the dye cell to minimize the length of unpumped dye between the lasing region and the cell wall. For this reason it is also difficult to obtain short pulses at a wavelength of less than 565 nm using rhodamine 6G.

With DFDL compression ratios of more than 30, it is important to address the problem of the relative jitter between two independent DFDL oscillators, as this can be a determining factor for time-resolution in many experiments. We may readily calculate the relative movement of a DFDL spike inside the pumping pulse envelope, as a function of pump intensity, using the formalism described in ref. [5]. For short pulse generation this movement is insensitive to pulse duration, a typical value being -0.7 ps/percent increase of pump power. Moreover this value only varies slowly with absolute pump intensity, by about 10 fs/percent change of pump level. This means that two DFDL oscillators with the same operating point above threshold, within 10%, would have a relative jitter < 100 fs/percent of pump power. For the  $\pm 2\%$  fluctuation, typical of our doubled YAG laser, this leads to a calculated jitter  $< \pm 200$  fs, fairly independent of pulse duration.

This prediction is borne out by experimental measurement of auto- and cross-correlation curves for the two amplified DFDL systems, without any saturable absorber. In short-pulse generation conditions, when both oscillators are adjusted to just below second-spike threshold, we are unable to distinguish autocorrelation and cross-correlation traces and we conclude that any jitter must be negligible compared to the 4 ps pulse width. Note that this result represents a significant improvement over that obtained for synchronously-pumped dye laser systems, where relative jitter may easily be several picoseconds [4].

#### 4. Improving time-resolution

One obvious way to improve the time-resolution of the present system is to decrease the duration of the pumping pulse. A factor of two overall decrease in pulse width might be achieved by replacing the YAG rod with a YLF rod. A similar improvement can be obtained at the expense of increased jitter between the dye lasers. This method involves pumping the DFDL oscillators above the threshold for generation of secondary pulses, but with different pumping levels so that the secondary pulses are not time-coincident. Higher pumping level increases the rate of rise of the inversion in the dye medium and is initially equivalent to a shorter pumping pulse.

Fig. 5 shows the result of a non-resonant time-resolved CARS experiment using the set-up of fig. 3 and differentially over-pumping the DFDL oscillators beyond second-spike threshold. In this experiment the shorter amplified dye laser pulse  $(I_1)$  is divided into two, to provide both excitation and probe beams, leading to the following correlation function,

$$S(t_{\rm d}) = \int_{-\infty}^{+\infty} {\rm d}t \, I_1(t) I_2(t) I_1(t-t_{\rm d}) .$$

The observed signal  $S(t_d)$ , which is plotted on a logarithmic scale as a function of  $t_d$  in picoseconds, exhibits a decay time of 490 fs over four orders of signal magnitude and is subsequently limited by system noise out to at least 25 ps delay. The slight asymmetry of the above curve may be fitted (full line in fig. 5) by assuming that the  $I_1$  pulse, which traverses a saturable absorber, rises about 30% faster than it

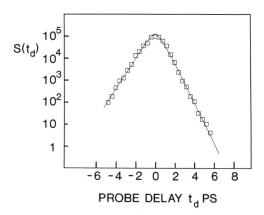


Fig. 5. Three-pulse cross-correlation trace obtained by non-resonant time-resolved CARS and using differentially over-pumped DFDL oscillators. The CARS signal decays over four decades with a time-constant of 490 fs. Each point represents the average of 1000 laser shots or one second of experimental time. The full line is calculated (see text).

falls. The pulse widths used in the fit are 0.7 ps  $(I_1)$  and 1.3 ps  $(I_2)$ . This particular correction function is insensitive to jitter between  $I_1$  and  $I_2$  and jitter manifests itself mainly through increased shot-to-shot fluctuation of signal intensity. Further improvement in time resolution can be obtained if spurious signal structure at large delay, due to secondary pulse overlap, is acceptable.

#### 5. Conclusions

We have developed a dual distributed-feedback dye laser system, pumped by a single 65 ps, 532 nm pulse from a stand-alone YAG laser, and providing independently tunable, synchronized, megawatt power level, kilohertz repetition-rate pulse trains. Single pulse widths of less than 2 ps and CARS time-res-

olution better than 0.5 ps have been obtained. We believe that his system represents an attactive alternative to existing technology for non-linear picosecond spectroscopy in the time and frequency domain.

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