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PICOSECOND PULSE GENERATION BY DISTRIBUTED FEEDBACK DYE LASERS

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Many primary physical, chemical and biological processes occur on the 0.1 to 100 ps time scale. Until quite recently, their experimental study was strongly impeded by the absence of electronic instruments being capable of resolving such short time intervals.

Since these processes can often be stimulated by optical excitation and are also frequently accompanied by some change of an optical property of the sample, such as absorption or reflection coefficient, refractive index, birefringence, fluorescence etc., optical methods can be applied to investigate them. The common feature of all these methods [1-4] is the use of high power ultrashort (0.1 to 100 ps) light pulses. Such pulses have usually been generated by mode-locked lasers.

The recently observed self-Q-switching effect occurring in distributed feedback dye lasers (DFDL) [5-7] now opens an alternative way to ultrashort pulse generation. This new technique has many advantages over the mode-locking technique (cf. section 8).

1. Principle of Operation of a Distributed Feedback Dye Laser

A distributed feedback dye laser has no external cavity (Fig.1). Instead, the optical feedback necessary for laser oscillation is provided within the active medium itself by a spatially periodic modulation of either the gain coefficient or the refractive index [8-11], or both. The lasing wavelength is determined by the Bragg condition. DFDLs as tunable narrow linewidth light sources have been studied extensively [8-12].

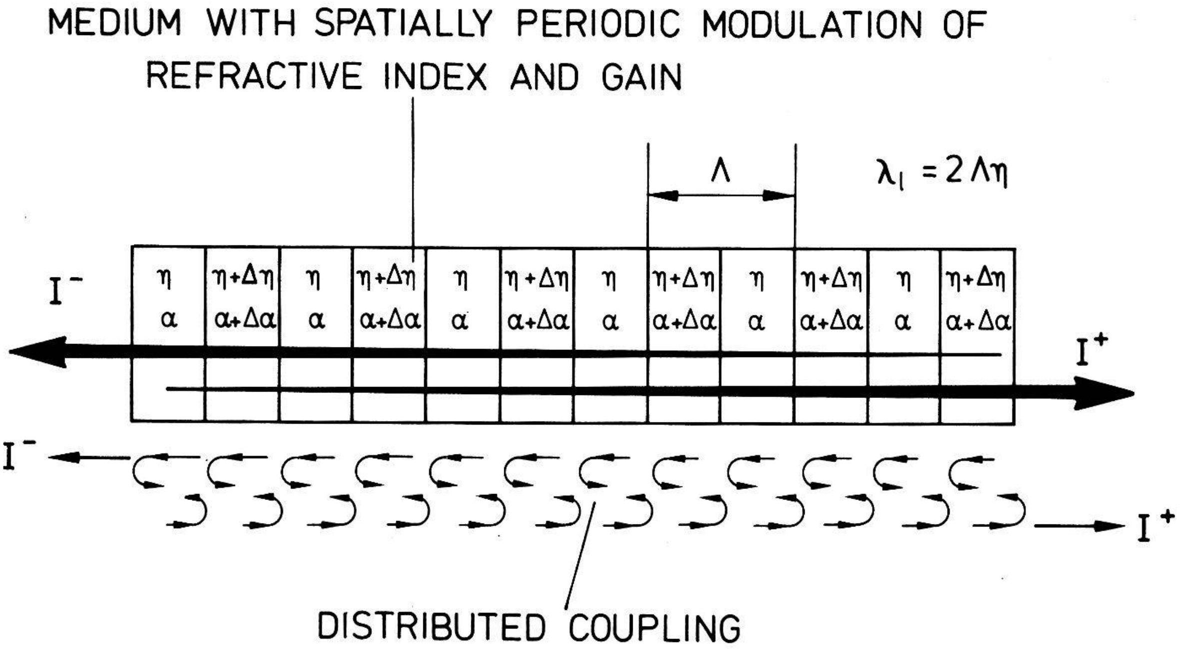


Fig.1. Principle of distributed feedback. η = refractive index, α = gain coefficient, Λ = spacing of interference fringes, λ_1 = laser wavelength.

2. The Self-Q-switching Effect: Physical Principle of Short Pulse Generation

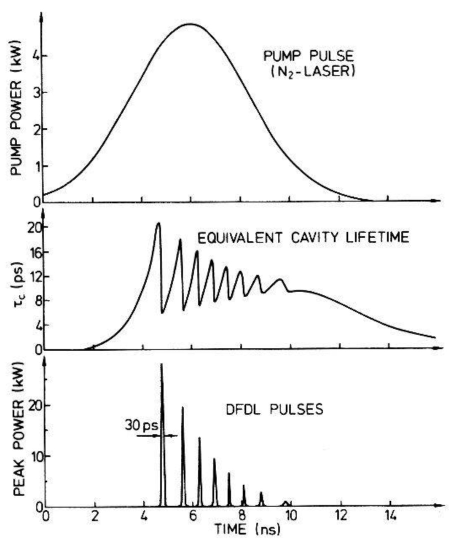


Fig.2. Time courses of equivalent cavity lifetime and DFDL output power obtained with the rate equation model [5] for N_2-laser pump pulse. Dye solution parameters were those of rhodamine 6G.

The temporal behaviour of a pulsed laser (e.g. N_2-laser) pumped DFDL has been described by a rate equation model [5-7]. A typical result obtained with this model is shown in Fig.2. We see that the DFDL generates a train of picosecond pulses. After each pulse the equivalent cavity decay time drops to a lower value. The equivalent cavity decay time τ_c is the average lifetime of a photon in the DFDL and is a measure of feedback efficiency. Such change of the cavity lifetime was called self-Q-switching [5]. It is responsible for the generation of ultrashort pulses.

According to the model [5-6] the number of DFDL pulses increases

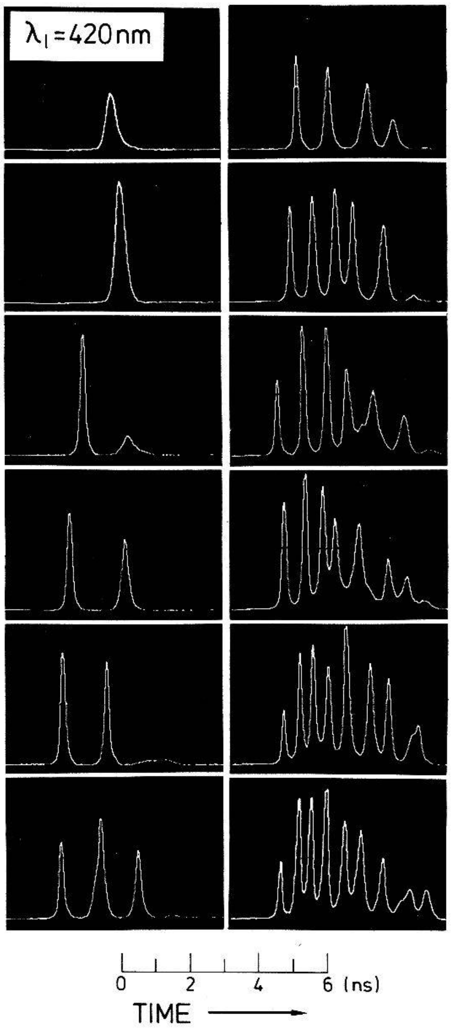


Fig.3. Streak camera recordings of DFDL output pulses as a function of pump power. Pump power starts with low level at upper left and ends with high level at lower right corner of the figure. 10^{-3} mol/l bis-MSB in diphenylether/dioxane 1:1.

with increasing pump power. There exists a certain pump intensity range where single ultrashort pulses are generated.

Figure 3 shows the measured pulse shapes produced by a N_2-laser pumped DFDL [13]. Pulses like these are generated at any wavelength in the 360 to 700 nm spectral region. The temporal shapes of the pulses are essentially the same for all dyes [5-7,13,14].

3. Optical Schemes for Distributed Feedback Dye Lasers

The periodic structure (Fig.1) required for distributed feedback in a DFDL is usually induced by pumping the dye solution with a pattern of interference fringes formed by the pumping beam [9,11,12]. The conventionally used pump schemes require pump lasers with a high degree of spatial and temporal coherence. The fact that such lasers are often very sophisticated and unreliable, and also have lower efficiency, has prevented the widespread use of DFDLs.

Recently developed pumping schemes (Fig.4) allow to produce perfect pumping interference fringes even with lasers having low coherence. This is illustrated in Fig.5 which shows that the interference fringe separation Λ is equal to one-half of the grating constant (d) for each spectral component of the pump pulse. In other terms, this implies reduced requirements for tem-

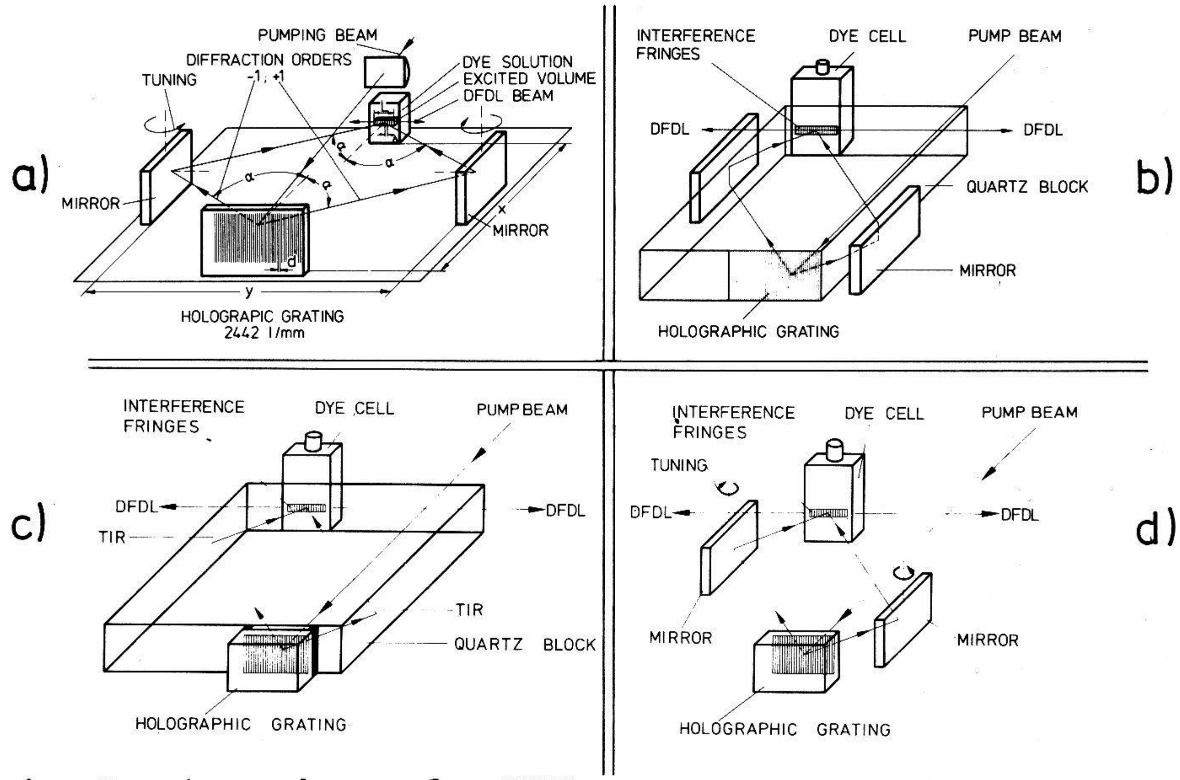


Fig.4. Pumping schemes for DFDLs.

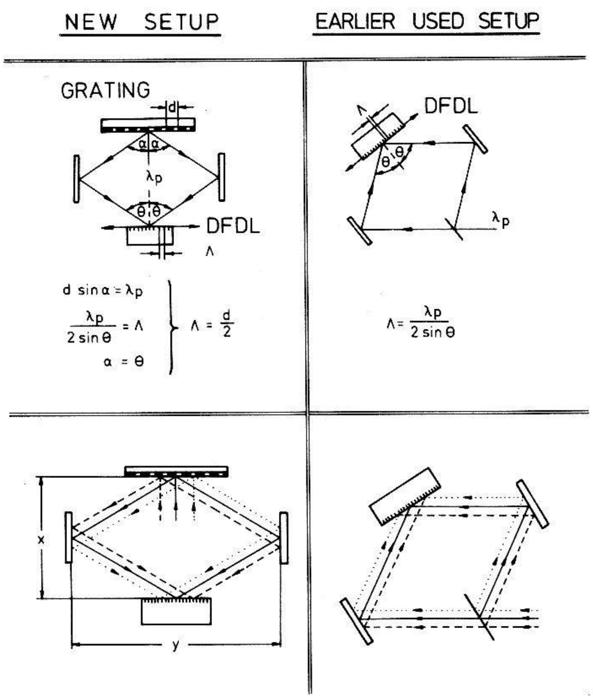


Fig.5. Comparison of two arrangements to obtain laser pumped distributed feedback structure in a dye cell.

poral coherence. Figure 5 also shows that with the new pump scheme for each point on the dye cell the two interfering beams have been diffracted from the same point on the grating. This means reduced demands for the spatial coherence of the pumping beam.

4. Tuning Methods

The lasing wavelength of the DFDL is given by $\lambda_L = 2\eta\Lambda$. Comparing the Bragg condition (Fig.1) with the relationship for the period of the interference fringes (Fig.5) reveals that tuning can be accomplished either by change of the diffraction grating period or the refractive index of the dye solution. - It is obvious that the dye must have optical gain at the desired lasing wavelength. -

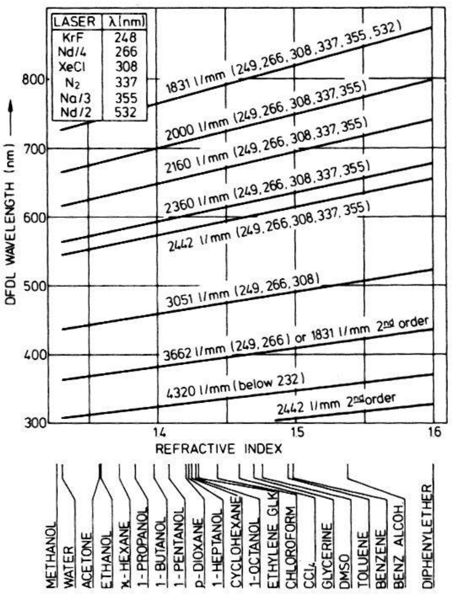


Fig.6. DFDL output wavelength as a function of the refractive index of the dye solution. Some values for commonly used pure solvents are given under the abscissa. The straight lines were computed for commercial diffraction gratings. Numbers in brackets indicate which of the pump lasers given in the box may be used with the respective grating.

Lasing wavelength as a function of refractive index of the dye solution is plotted for various diffraction gratings in Fig.6. The numbers in brackets indicate pump wavelengths which can be used with any given grating. An upper limit for the pump wavelength is set by the absence of diffraction orders when the wavelength exceeds the grating constant (d). For the pump scheme shown in Fig.4b, employing a diffraction grating in direct contact with the quartz parallelepiped, the upper limit of the pump wavelength is extended by a factor equal to the refractive index of fused quartz.

Other tuning methods based on the temperature and pressure dependence of the refractive index are described in [15,16].

Tuning may also be achieved by rotating the two mirrors (Fig.4a,b,d) in opposite directions about vertical axes. This changes the period of the interference fringes and with it the laser wavelength. The tuning sensitivity is about 5-10 Å/mrad [15]. A computer controlled tunable DFDL using this technique is presently used in our laboratory for fluorescence lifetime measurements of dyes in monomolecular layers [17].

5. Single Pulse Generation by DFDL

For practical applications single DFDL pulses are needed. Single pulses are generated if the pump power does not exceed the threshold value for more than the following factors: 0.2, 0.4 and 0.8 for 3.5 ns (typical N_2-laser), 1 ns (TEA-N_2-laser) and 16 ps (mode-locked Nd:YAG-laser) pump pulse durations, respectively [18].

By careful control of the pump power single pulses with ±7 % shot-to-shot stability have been generated [7].

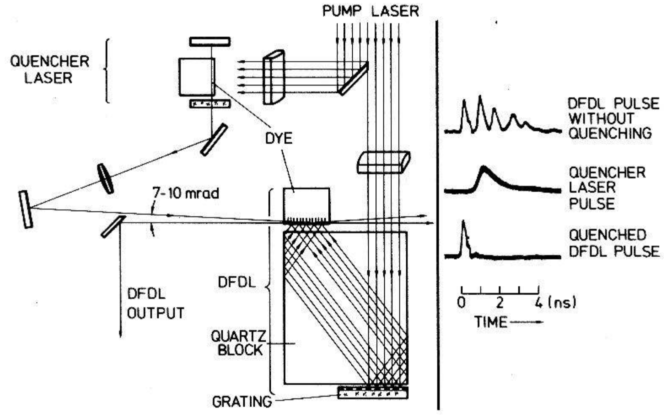
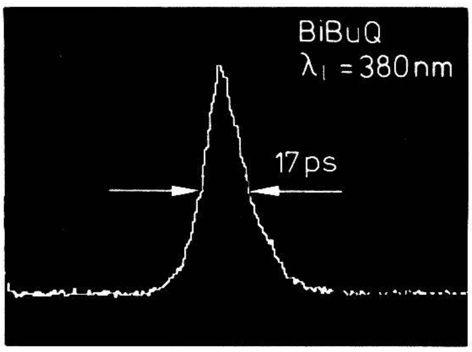


Fig.7. Experimental arrangement of a quenched DFDL.

A more advanced method for single pulse generation is shown in Fig.7. The DFDL which is pumped by about half the N_2-laser beam cross section generates about 6 pulses (Fig.7). The other half of the beam pumps a conventional dye laser with external cavity. The pulse of this laser is injected into the DFDL under an angle of about 7-10 mrad just after the first DFDL pulse has been generated. This injected pulse lowers the gain of the distributed feedback structure below the lasing threshold, and thus all later pulses of the DFDL are quenched. The DFDL beam is spatially separated from the beam of the quenching laser.



The shortest single pulse generated in this way was 17 ps (Fig.8). A detailed description and the characteristics of this setup (Fig.7) will be presented elsewhere [13].

Fig.8. Single pulse output of a quenched DFDL recorded with streak camera. 10^{-3} mol/l BiBuQ in dioxane/ methanol 3:2.

6. Duration of the DFDL Pulses

Calculations and experiments have shown [19] that the duration of single DFDL pulses is about 50 to 100 times shorter than the duration of the pump pulse (Fig.9). In these experiments

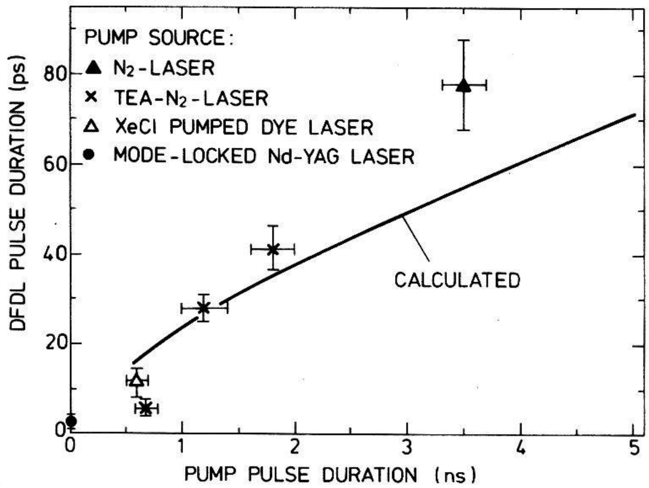


Fig.9. Duration of output pulses of a rhodamine 6G DFDL as a function of pump pulse duration. Solid line was computed using rate equation model.

single pulse generation was accomplished by controlling the pump energy. Further pulse shortening into the subpicosecond range seems also feasible (cf. section 9).

7. Amplification of the DFDL Pulses

The energy needed to pump a DFDL above threshold amounts to about 0.5 mJ. The remaining 2-4 mJ of the pumping N_2-laser are available for pumping of a multistage amplifier chain (Fig.10). In this way DFDL pulses can be easily amplified into the megawatts power range [20].

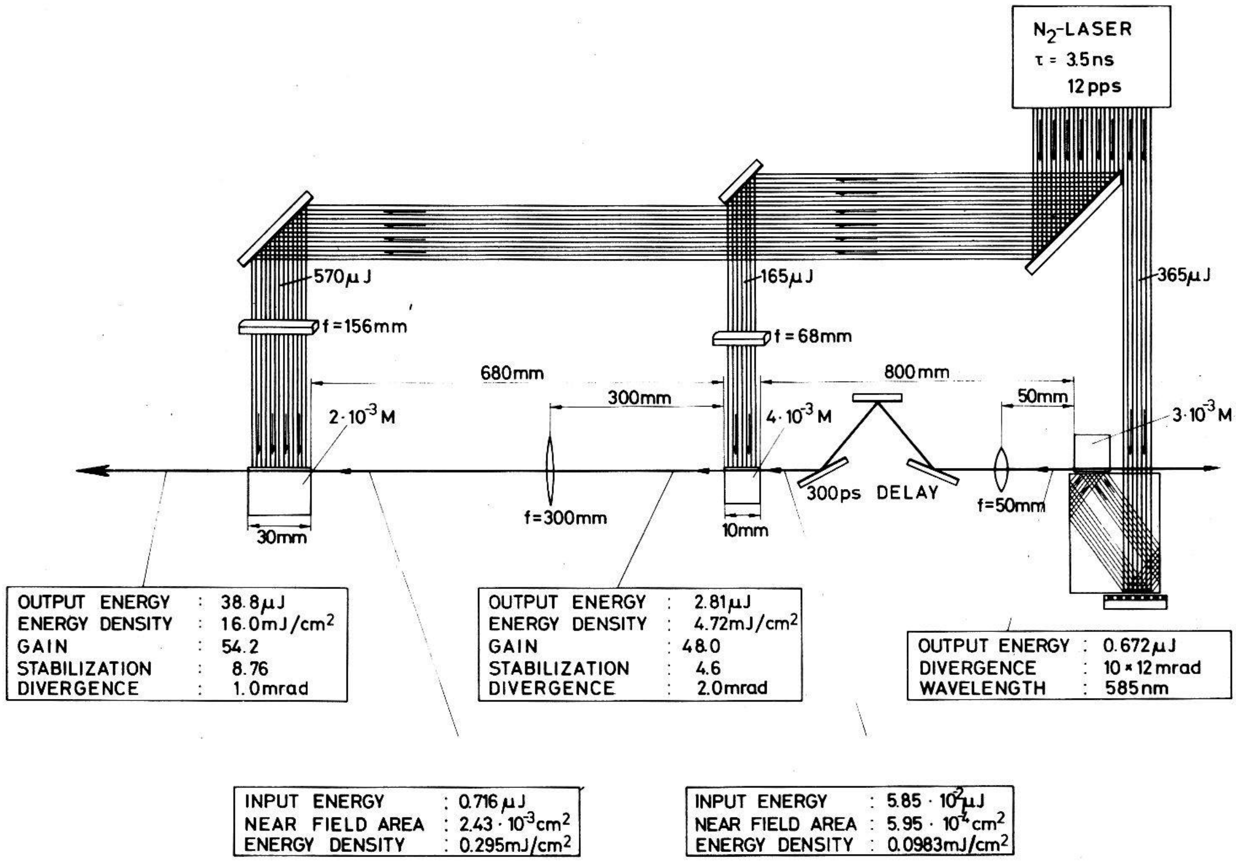


Fig.10. Nitrogen laser pumped DFDL-oscillator-amplifier system [20].

8. Comparison of Picosecond Pulse Generation by Mode-locking and by the Distributed Feedback Method

8.1 Synchronously pumped mode-locked dye lasers have been refined to a high state of technical perfection by persistent efforts of many laboratories. Pulses as short as 65-100 fs can now be generated [4]. The high cost of mode-locked laser systems is, however, a serious obstacle for their widespread application.

Nitrogen laser pumped DFDLs, in contrast, are simple in construction, they do not contain complicated or expensive components. Both the pump laser and the DFDL, can be easily constructed even by laboratories which have only moderate workshop facilities. Their only drawback being the somewhat longer pulse duration (cf. Fig.9). Thus DFDLs can be conveniently used in experiments where temporal resolution of a few picoseconds is satisfactory.

8.2 The operation of synchronously pumped mode-locked dye lasers in the UV part of the spectrum has been prevented by the absence of suitable pump sources and saturable absorbers for mode-locking. To our knowledge the shortest wavelength reported so far is 420 nm. DFDLs are capable to operate at any wavelength for which laser dyes exist, including the 420-360 nm range (cf. Figs.3 and 8) [14]. Thus, DFDLs are well suited to study ultrashort phenomena requiring UV and blue pulses.

8.3 Mode-locked dye lasers generate a train of closely separated (~10 ns) pulses, but besides single photon counting experiments most of the applications require single pulses. Such pulses are obtained by using special pulse selecting devices. In contrast, a DFDL can generate single pulses directly, without a pulse selector.

8.4 For most of the applications the energy of the mode-locked laser pulses is boosted by a pulsed Nd:YAG or excimer laser amplifier. Stable long term synchronization of the latter to the mode-locked laser is an intricate electronic problem. For the ultrashort pulse amplifier coupled to a DFDL the synchronisation problem can be solved easily by choosing the proper light delays (Fig.10).

9. Outlook

We infer from our experiments that the duration of the DFDL pulses is presently limited by the transit time of light through the DFDL structure. Pumping the DFDL in a travelling wave arrangement will, perhaps, eliminate this effect. In this way, we think to be able to generate subpicosecond pulses.

A XeCl-excimer laser pumped DFDL system is under construction and test. About 20 % of the energy of the XeCl-laser is used to pump a dye laser delivering 300 ps pulses of 0.3 mJ energy. Pumping the DFDL with these pulses we obtain 5 ps pulses in the 360-700 nm spectral range. The energy of these pulses is boosted to 500 MW in a 4 stage amplifier pumped by the remaining 80 % of the XeCl-laser energy. We regard this laser to be the prototype of a future picosecond spectroscopy "workhorse".

Acknowledgements

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