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H4.SMR/544 - 4

Winter College on Ultrafast Phenomena

18 February - 8 March 1991

DISTRIBUTED FEEDBACK DYE LASERS

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DISTRIBUTED FEEDBACK DYE LASERS

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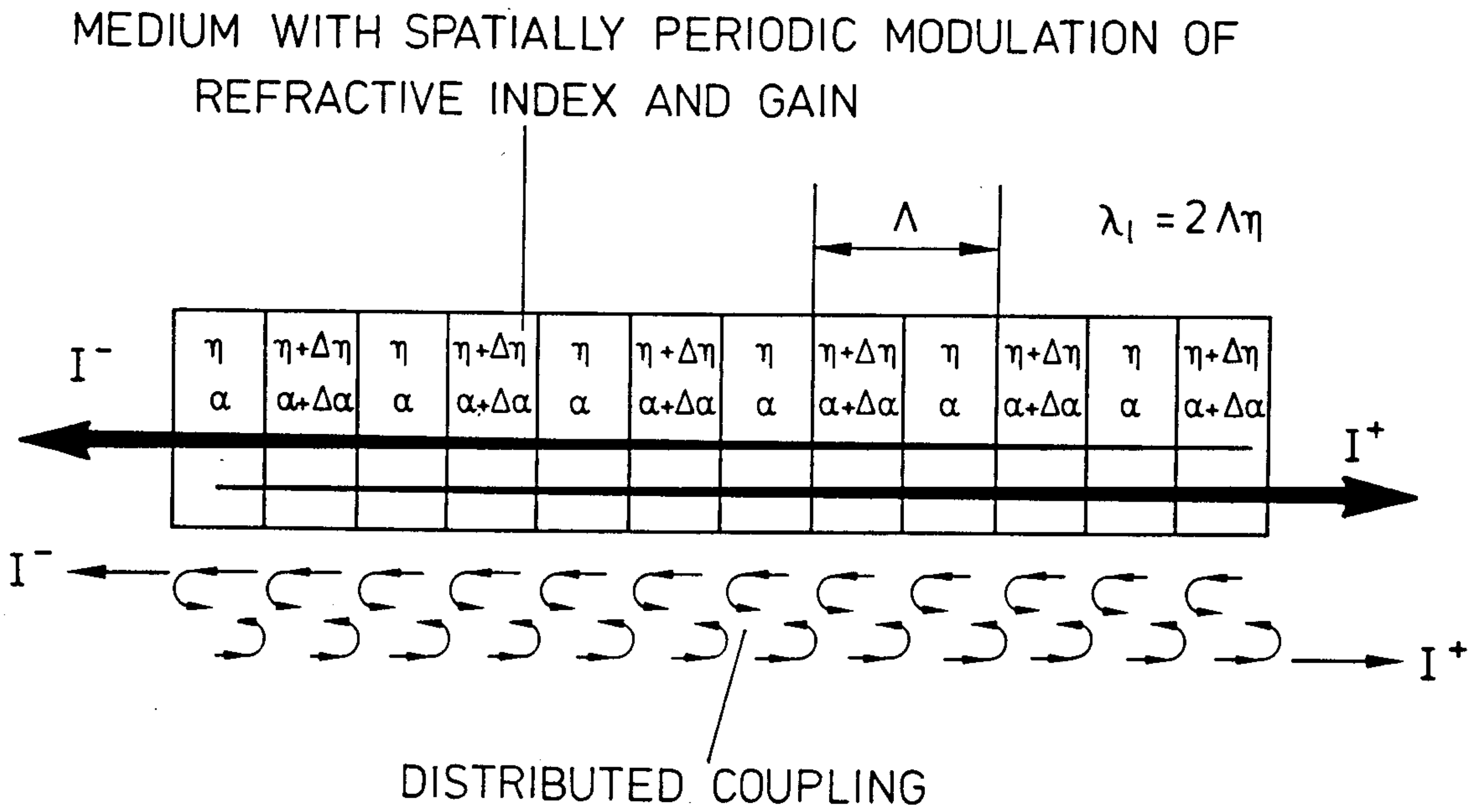
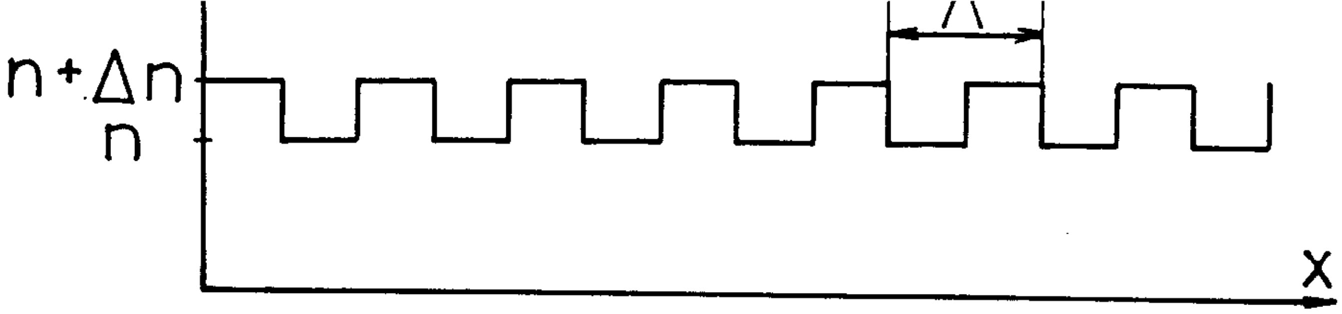
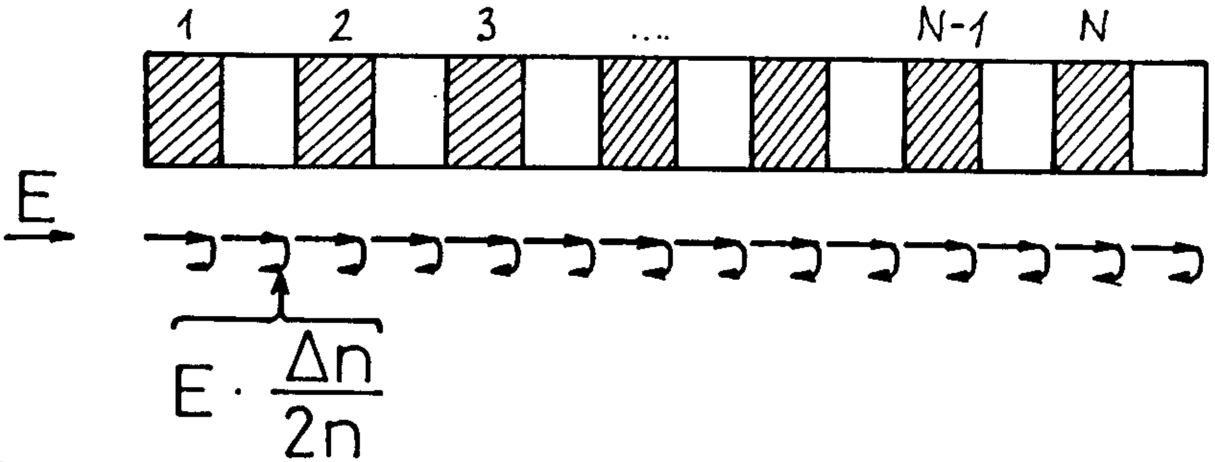
PRINCIPLE OF DISTRIBUTED FEEDBACK LASER







$$\left.\begin{array}{l} L = 1\,\mathrm{cm} \\ \lambda = 0.6\,\mu \end{array}\right\} N = 5 \cdot 10^{4} \qquad \begin{array}{l} \Delta n = 2 \cdot 10^{-5} \ (0.05\,^{\circ}\mathrm{C}) \\ n = 1.5 \end{array}$$

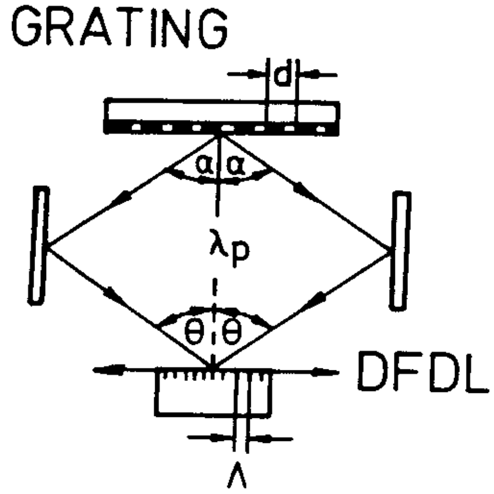
AT BRAGG - CONDITION : $\lambda = 2\Lambda n$

$$R = \left(\frac{\Delta n}{2n} \cdot N\right)^2 = \left(\frac{2 \cdot 10^{-5}}{2 \cdot 1.5} \cdot 5 \cdot 10^{4}\right)^2 = 0.11 = 11\,\%$$

FAR FROM BRAGG-CONDITION : $\lambda \neq 2\Lambda n$

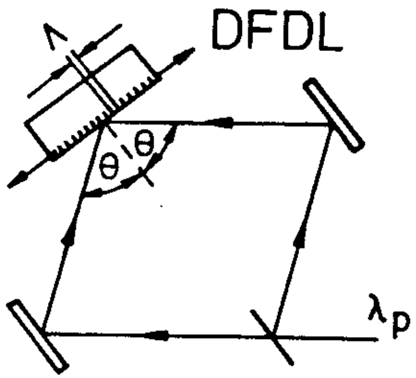
$$R = \left(\frac{\Delta n}{2n}\right)^2 \cdot N = \left(\frac{2 \cdot 10^{-5}}{2 \cdot 1.5}\right)^2 \cdot 5 \cdot 10^{4} = 2.2 \cdot 10^{-6} = \underline{\underline{2.2 \cdot 10^{-4}\,\%}}$$

NEW SETUP



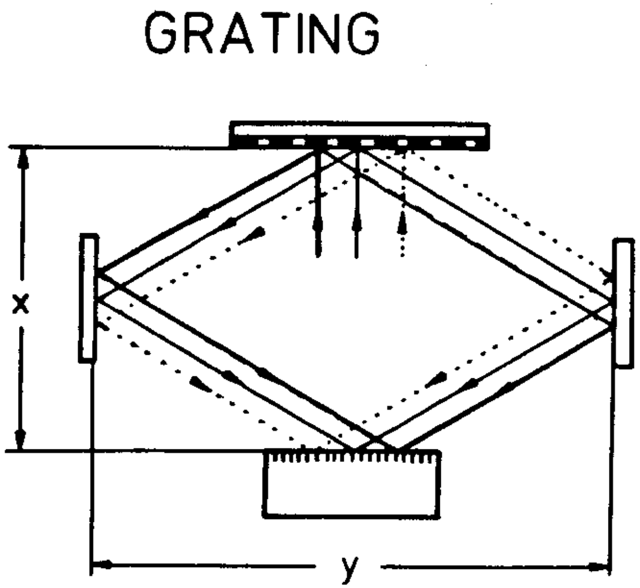
$$\left.\begin{aligned} d\sin\alpha &= \lambda_p \\ \frac{\lambda_p}{2\sin\theta} &= \Lambda \\ \alpha &= \theta \end{aligned}\right\} \Lambda = \frac{d}{2}$$

EARLIER USED SETUP

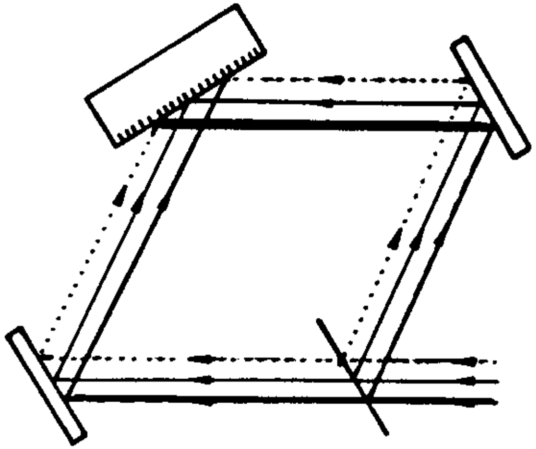


$$\Lambda = \frac{\lambda_p}{2\sin\theta}$$

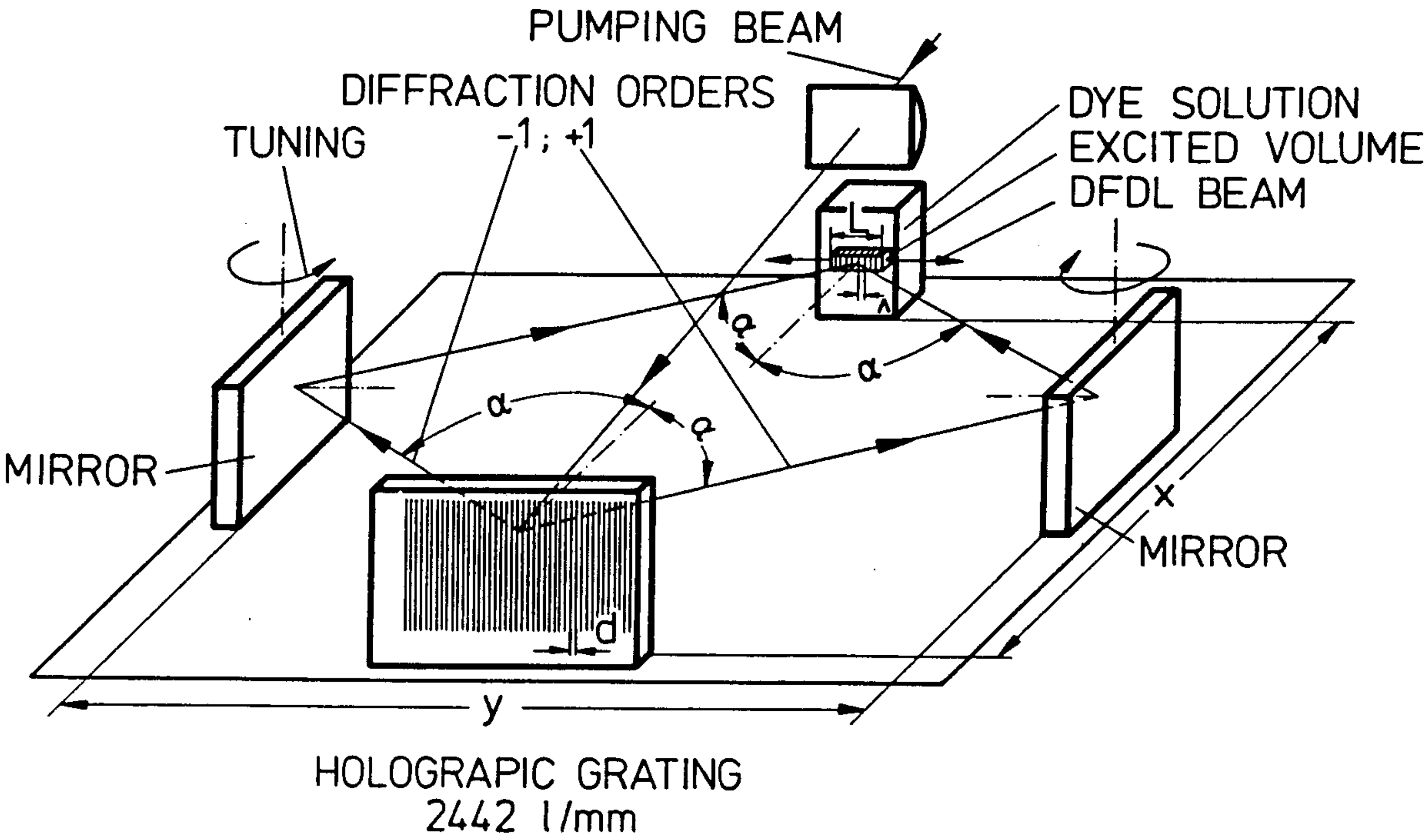
NEW SETUP

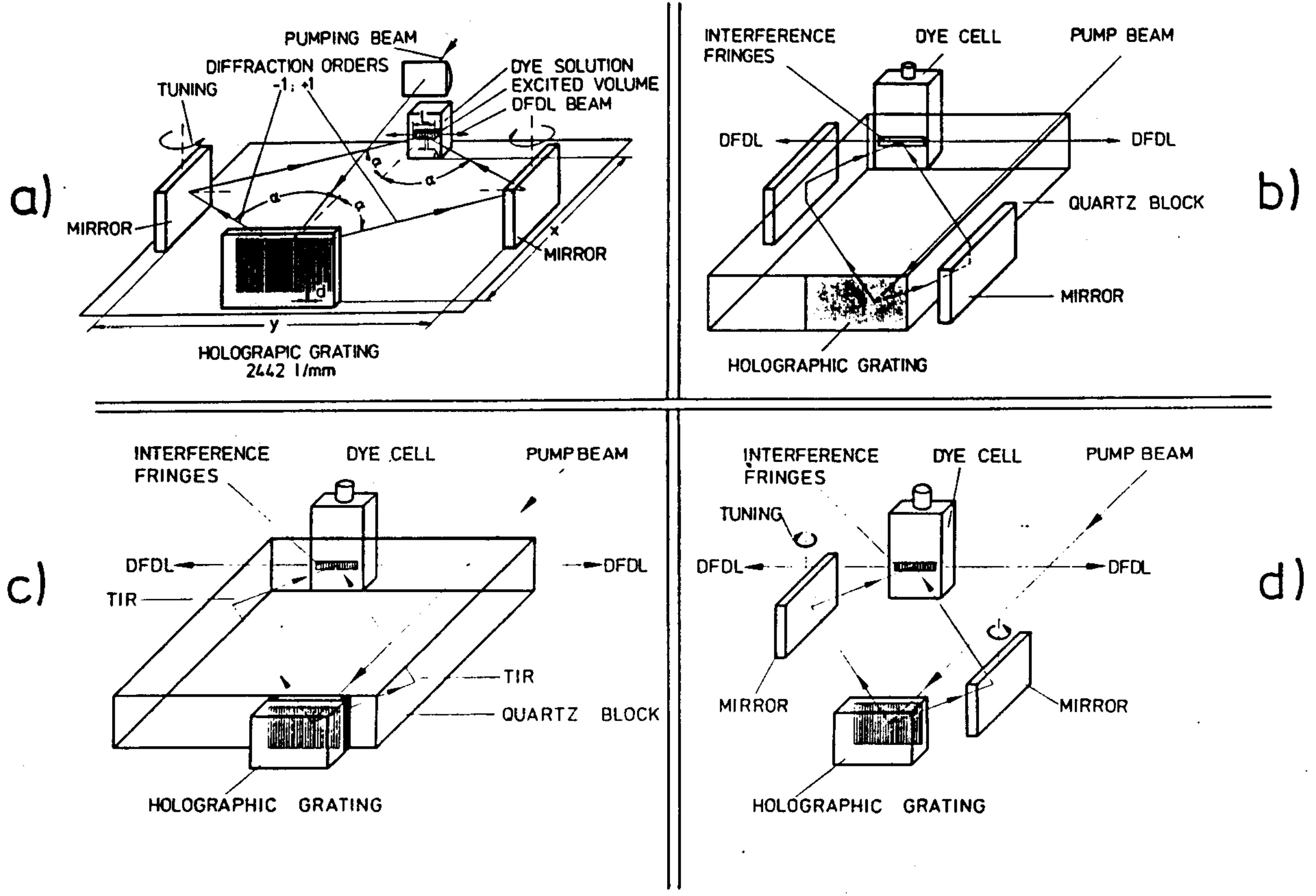


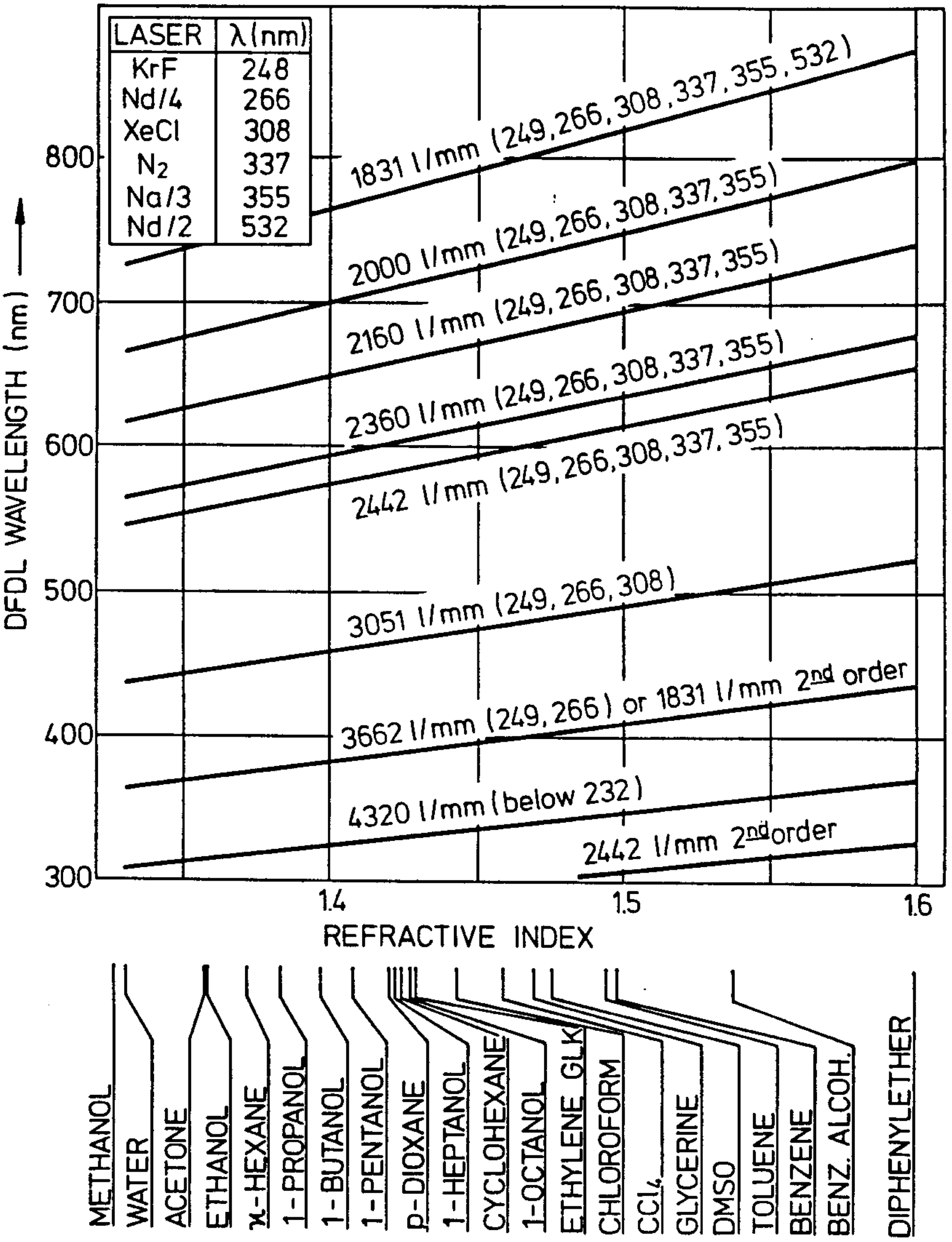
EARLIER USED SETUP

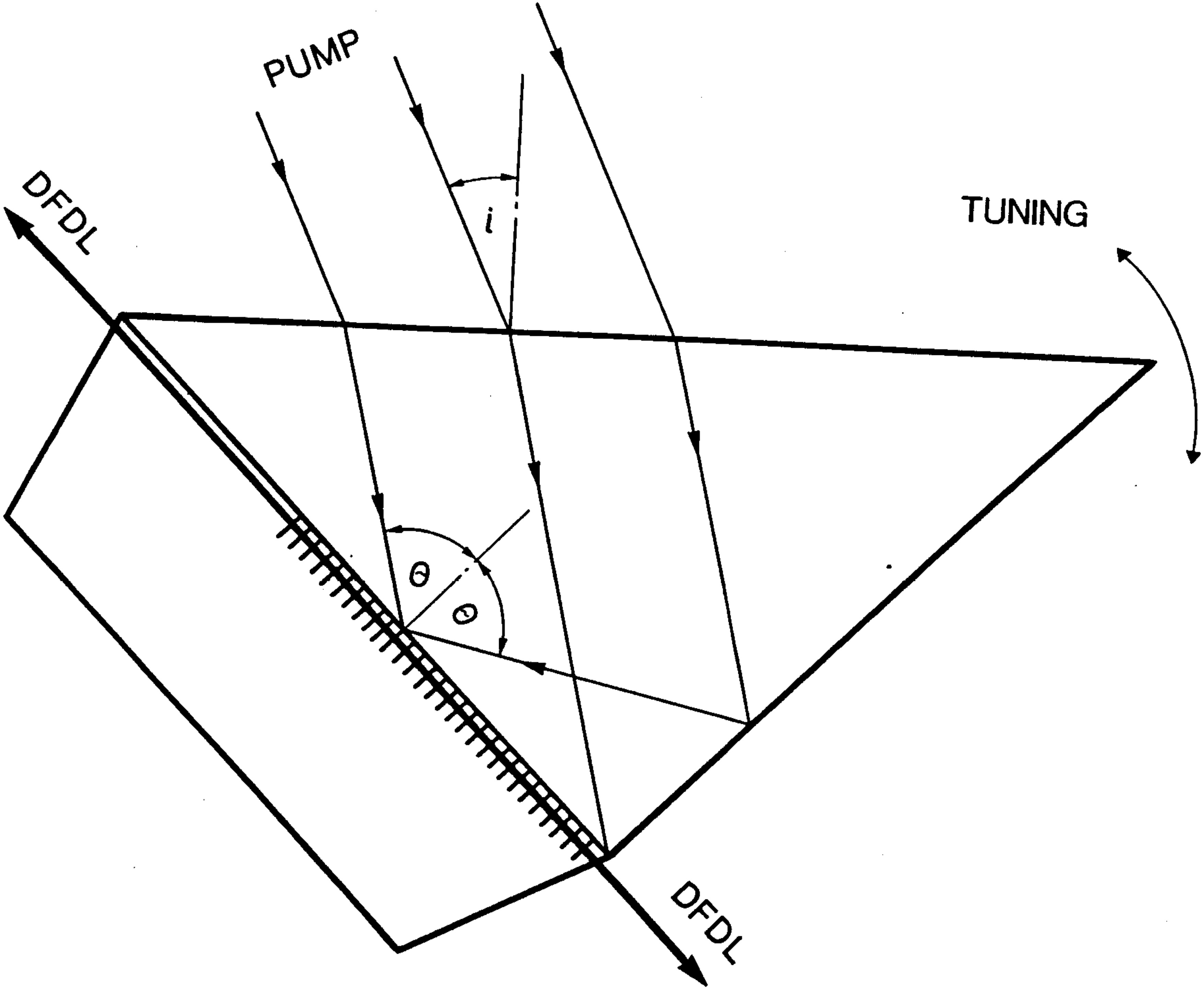


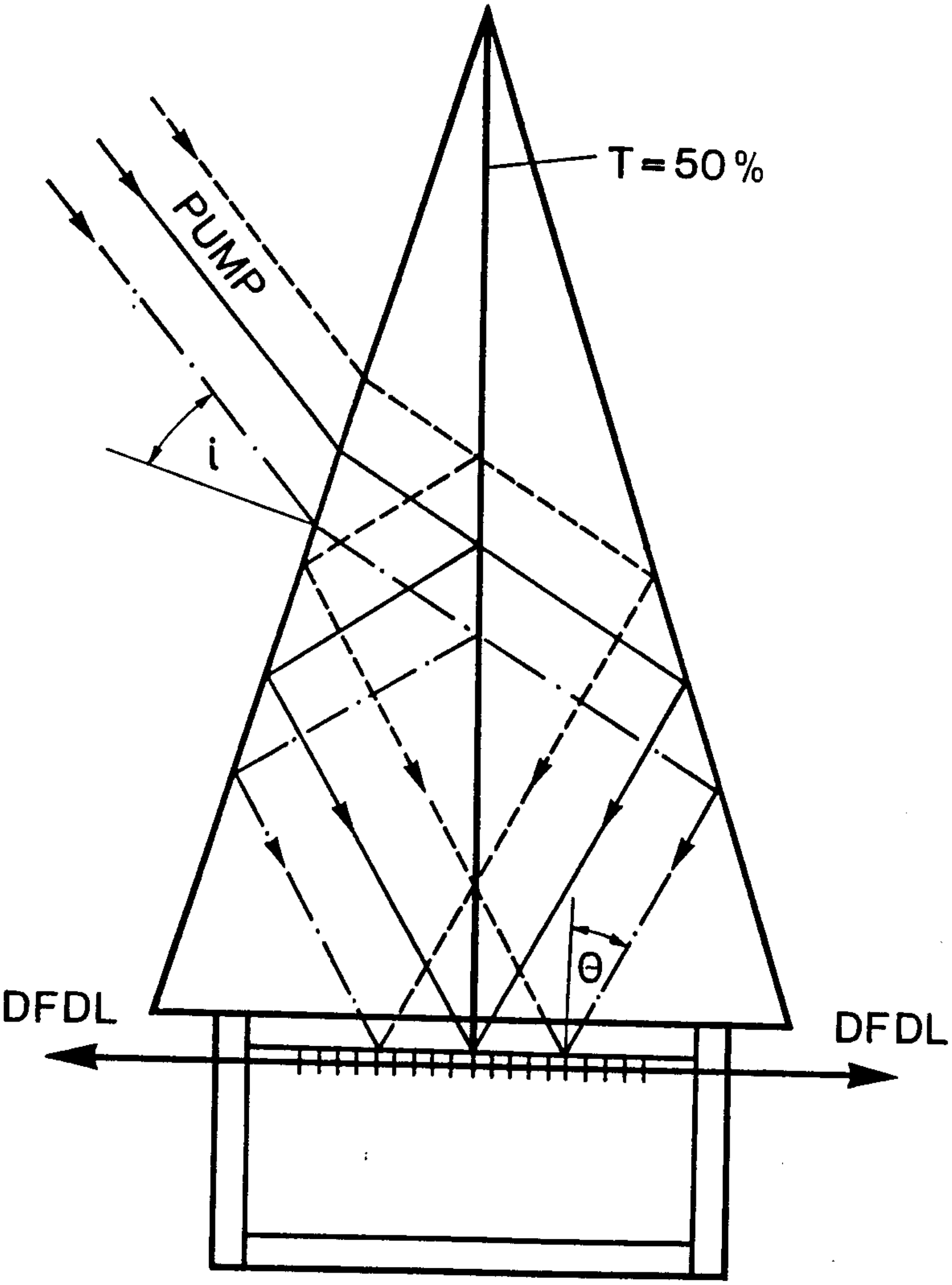
418 38

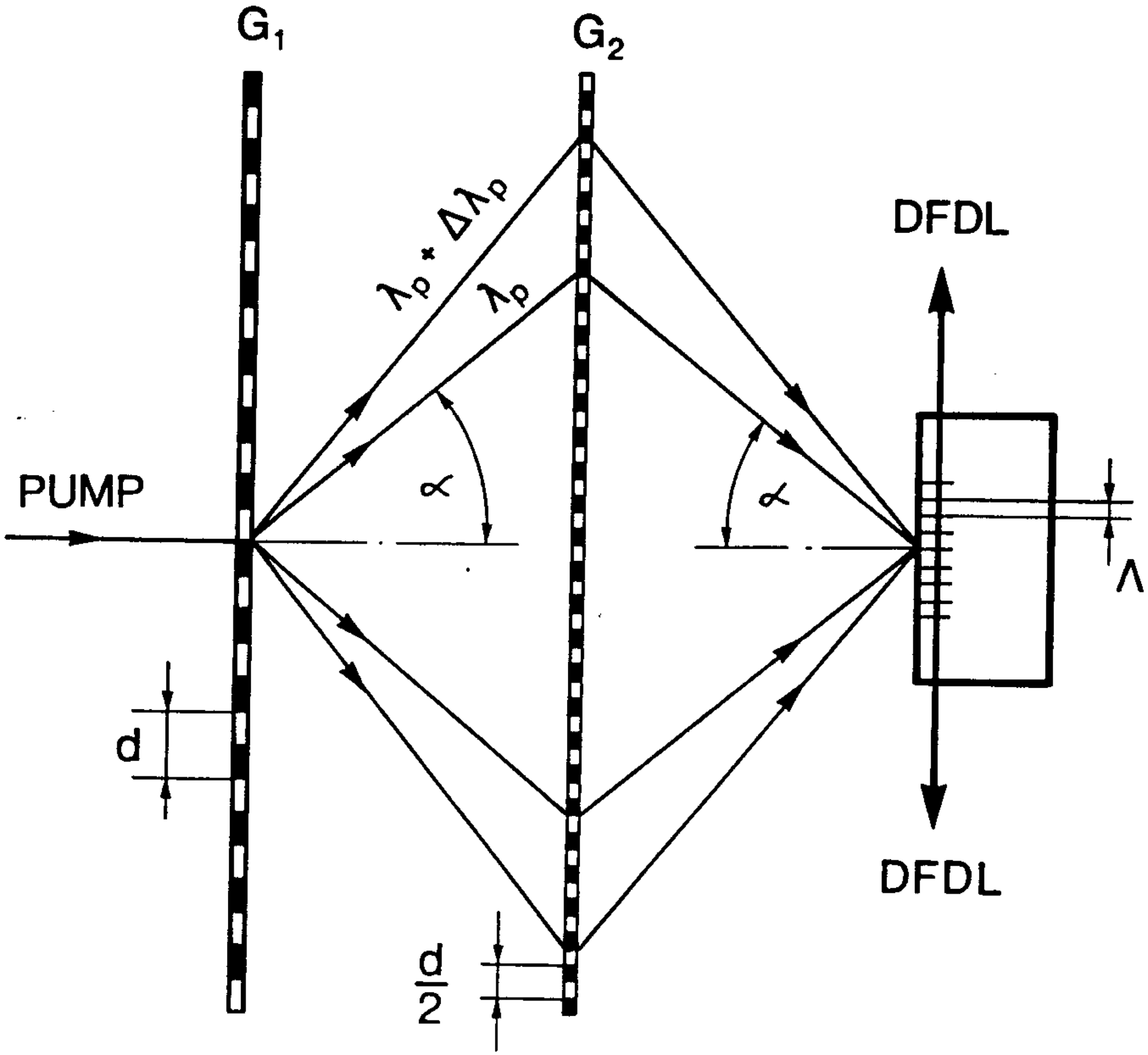

PUMPING BEAM
DIFFRACTION ORDERS
-1 ; +1
TUNING
DYE SOLUTION
EXCITED VOLUME
DFDL BEAM
L
Λ
α
α
α
α
MIRROR
MIRROR
x
d
y
HOLOGRAPIC GRATING
2442 l/mm

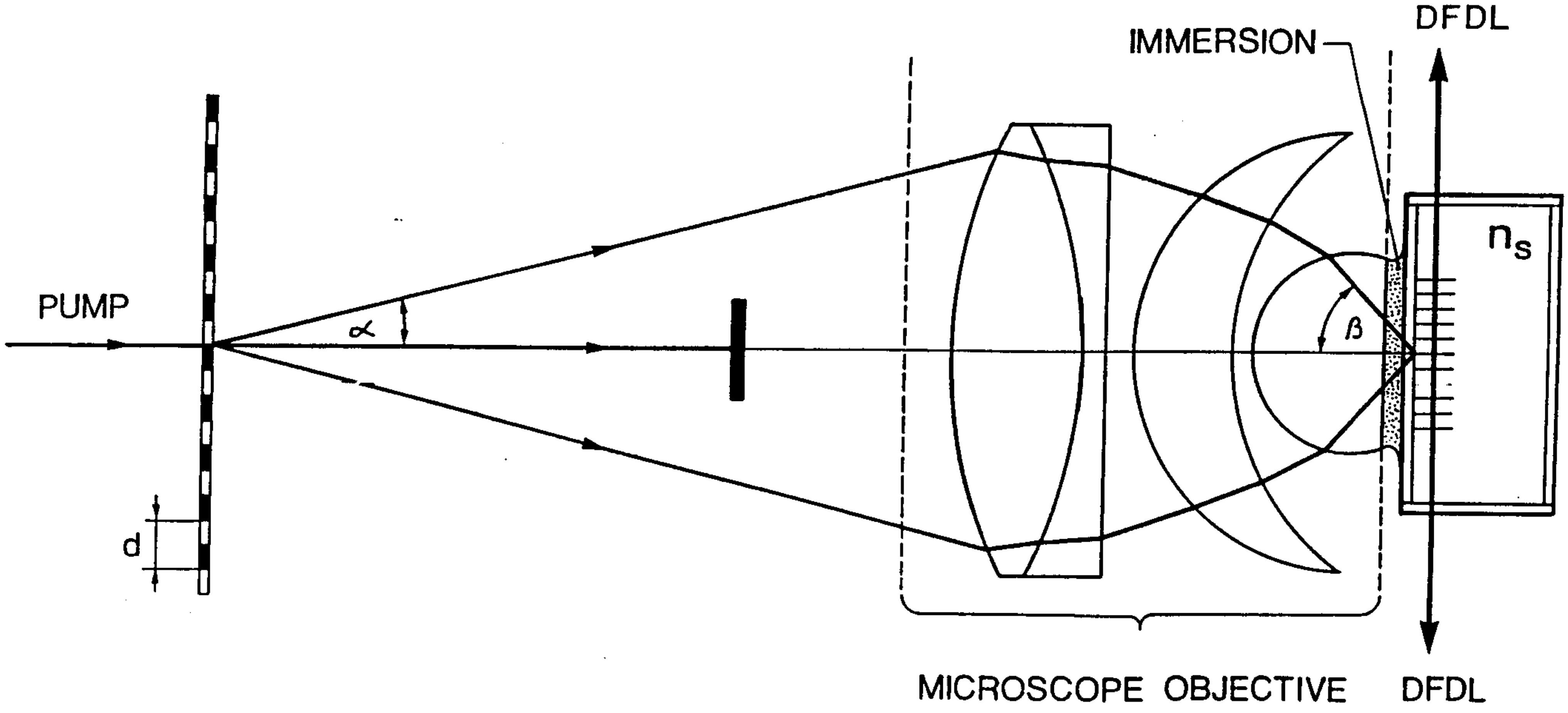

a)
PUMPING BEAM
DIFFRACTION ORDERS
-1; +1
TUNING
DYE SOLUTION
EXCITED VOLUME
DFDL BEAM
MIRROR
MIRROR
α
x
y
d
HOLOGRAPIC GRATING
2442 l/mm
b)
INTERFERENCE
FRINGES
DYE CELL
PUMP BEAM
DFDL
DFDL
QUARTZ BLOCK
MIRROR
HOLOGRAPHIC GRATING
c)
INTERFERENCE
FRINGES
DYE CELL
PUMP BEAM
DFDL
DFDL
TIR
TIR
QUARTZ BLOCK
HOLOGRAPHIC GRATING
d)
INTERFERENCE
FRINGES
DYE CELL
PUMP BEAM
TUNING
DFDL
DFDL
MIRROR
MIRROR
HOLOGRAPHIC GRATING

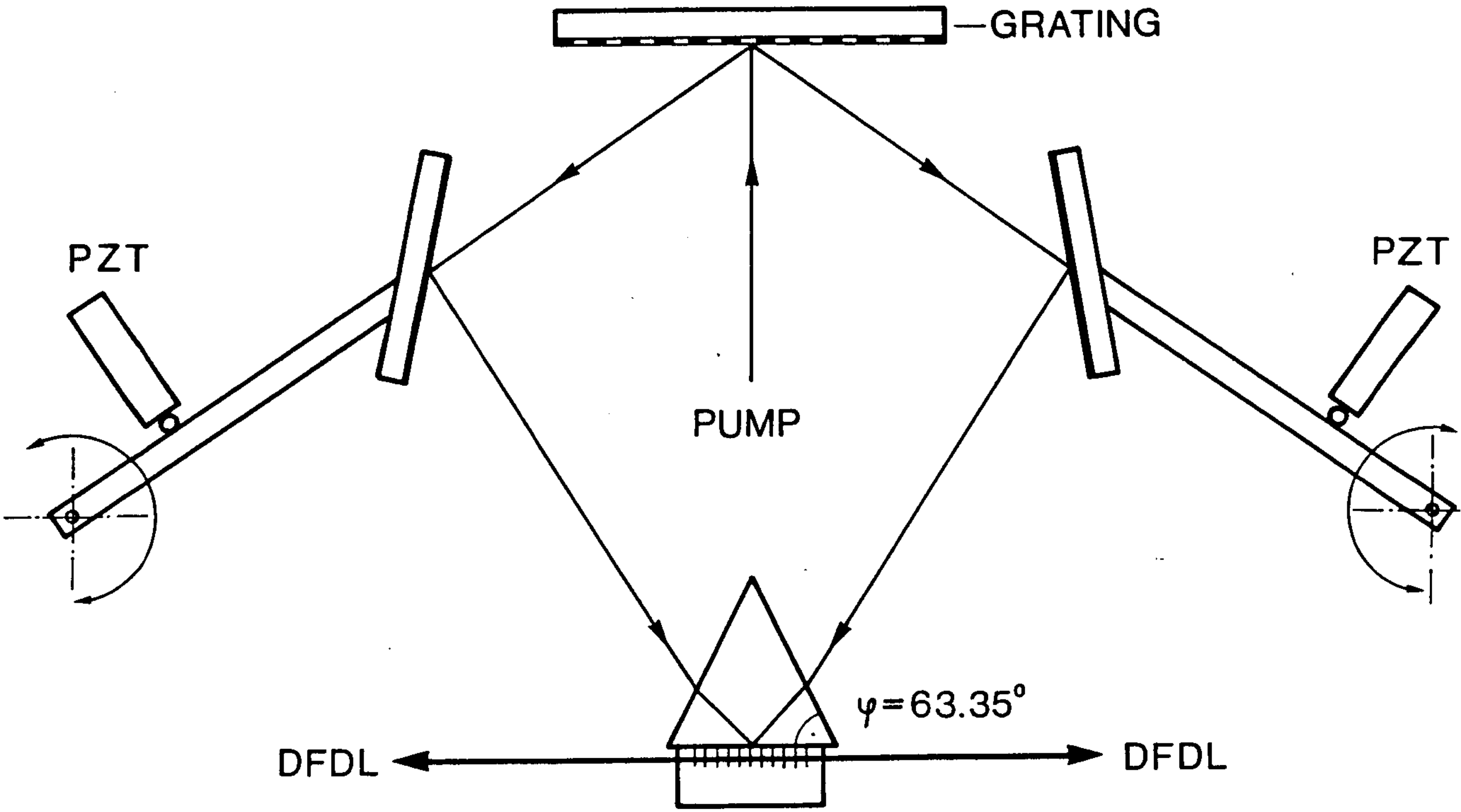

LASER | λ(nm)
KrF 248
Nd/4 266
XeCl 308
N2 337
Na/3 355
Nd/2 532
1831 l/mm (249,266,308,337,355,532)
2000 l/mm (249,266,308,337,355)
2160 l/mm (249,266,308,337,355)
2360 l/mm (249,266,308,337,355)
2442 l/mm (249,266,308,337,355)
3051 l/mm (249,266,308)
3662 l/mm (249,266) or 1831 l/mm 2nd order
4320 l/mm (below 232)
2442 l/mm 2nd order
DFDL WAVELENGTH (nm)
800
700
600
500
400
300
1.4
1.5
1.6
REFRACTIVE INDEX
METHANOL
WATER
ACETONE
ETHANOL
n-HEXANE
1-PROPANOL
1-BUTANOL
1-PENTANOL
p-DIOXANE
1-HEPTANOL
CYCLOHEXANE
1-OCTANOL
ETHYLENE GLK
CHLOROFORM
CCl4
GLYCERINE
DMSO
TOLUENE
BENZENE
BENZ. ALCOH.
DIPHENYLETHER

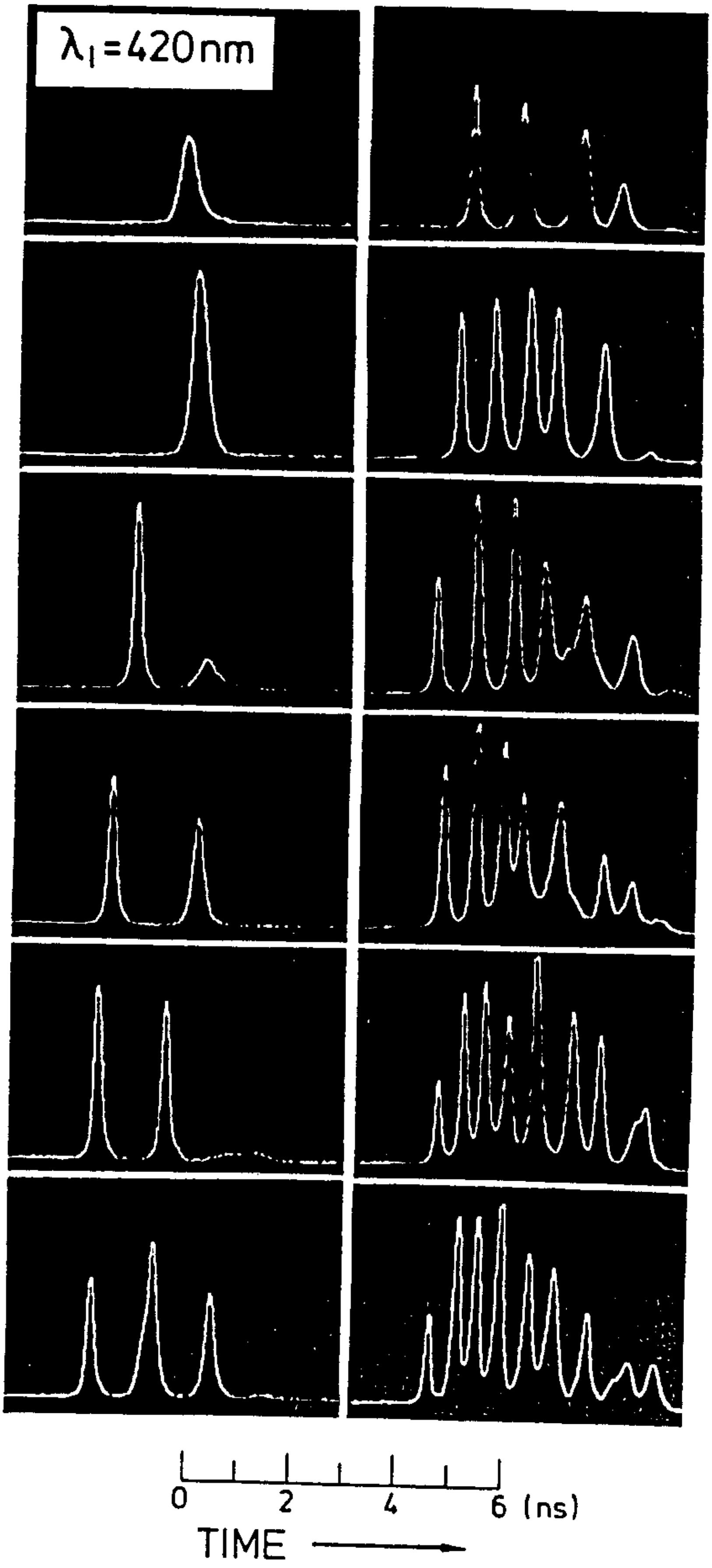

PUMP
i
TUNING
DFDL
θ
θ
DFDL


T = 50 %
PUMP
i
θ
DFDL
DFDL


G$_1$
G$_2$
$\lambda_p + \Delta\lambda_p$
λ_p
DFDL
PUMP
α
α
Λ
d
DFDL
$\frac{d}{2}$

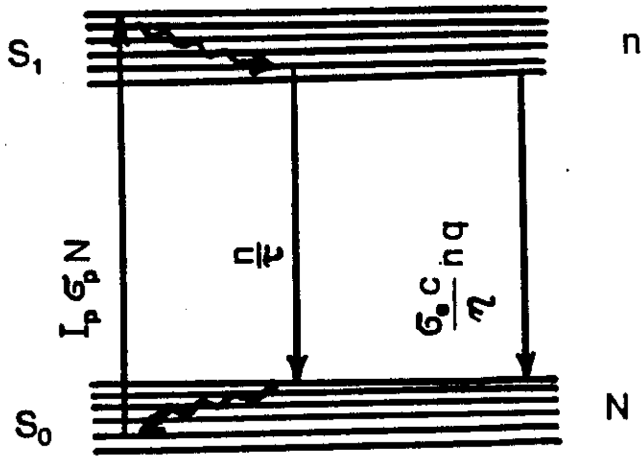

DFDL
IMMERSION
PUMP
α
β
n_s
d
MICROSCOPE OBJECTIVE
DFDL


GRATING
PZT
PUMP
PZT
φ=63.35°
DFDL
DFDL


λ_1=420nm
0
2
4
6 (ns)
TIME →

MODEL OF THE DFDL

(IEEE J. QE-16, 517 (1980)



$$\frac{dn}{dt} = I_p \sigma_p N - \frac{\sigma_e c}{\eta} n q - \frac{n}{\tau}$$

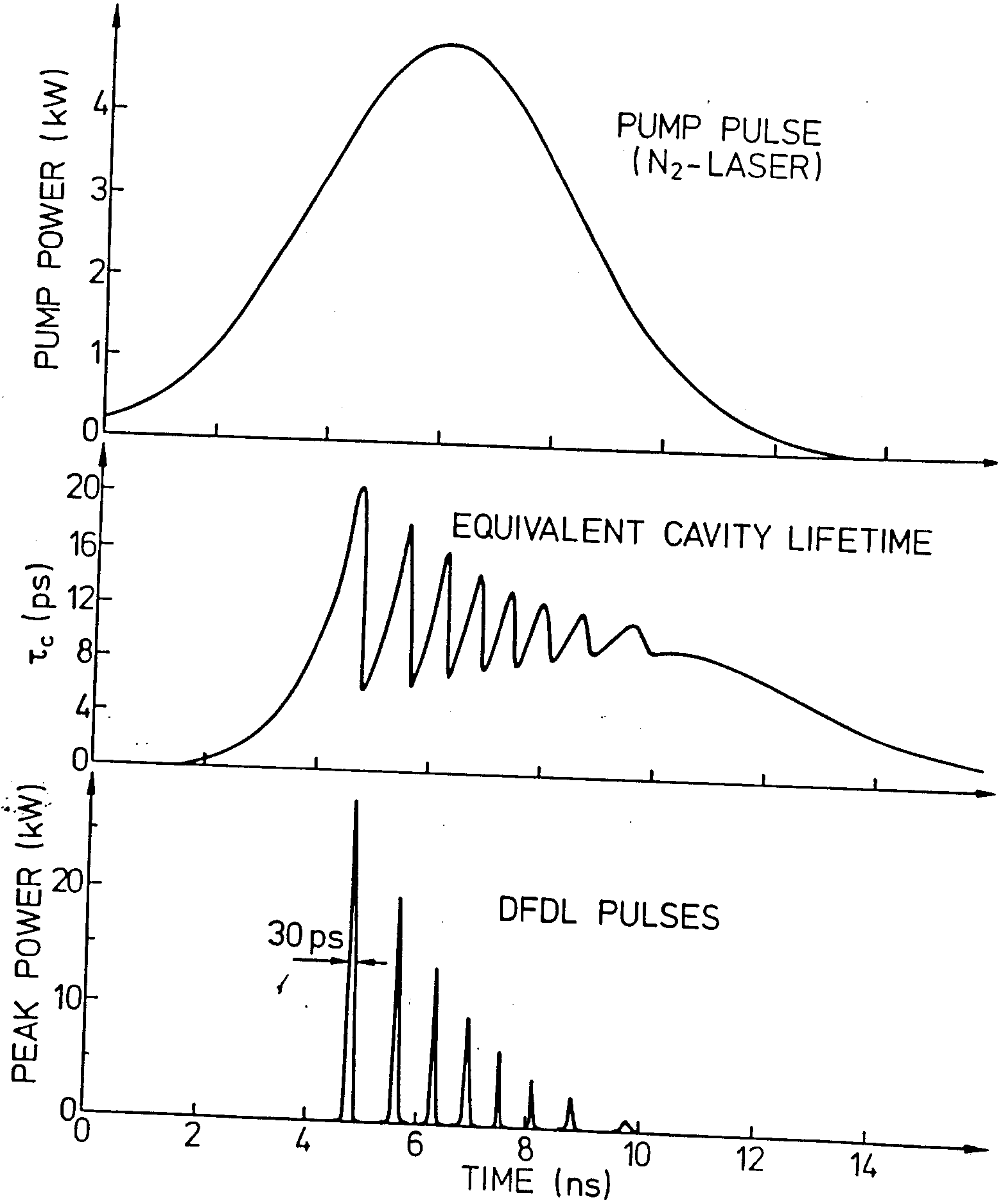
$$\frac{dq}{dt} = \frac{\sigma_e c}{\eta} n q - \frac{q}{\tau_c}$$

equivalent cavity decay time

$$\tau_c = \frac{\text{number of photons in the DFDL}}{\text{photon loss rate}}$$

$$\tau_c = \frac{\eta L^3}{8 c \pi^2} (n \sigma_e V)^2$$

$$\tau_c \sim n^2 \longrightarrow \underline{\tau_c \sim \alpha^2}$$


PUMP POWER (kW)
0
1
2
3
4
PUMP PULSE
(N$_2$-LASER)
τ_c (ps)
0
4
8
12
16
20
EQUIVALENT CAVITY LIFETIME
PEAK POWER (kW)
0
10
20
30 ps
DFDL PULSES
0
2
4
6
8
10
12
14
TIME (ns)

15

Rate equation model of DFDL

$$\dot{n} = I_p \, \sigma_p \, (N-n) - \frac{\sigma_e c}{\eta} n q - \frac{n}{\tau}$$

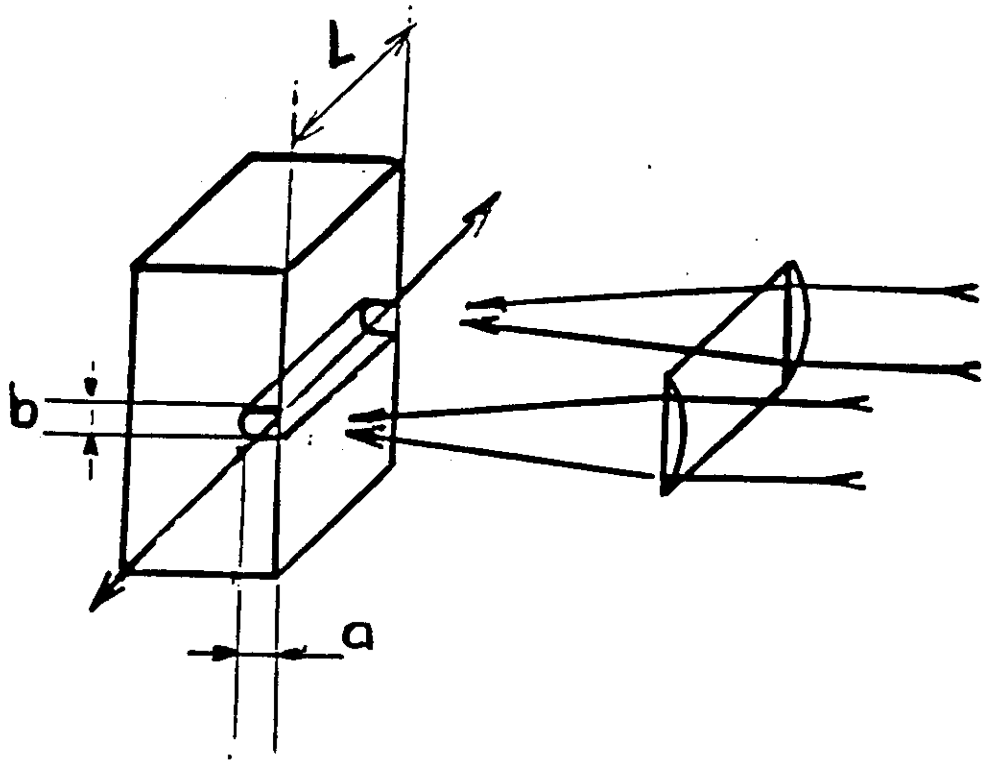
$$\dot{q} = \frac{(\sigma_e - \sigma_a) c}{\eta} n q - \frac{q}{\tau_c} + \frac{\Omega n}{\tau}$$

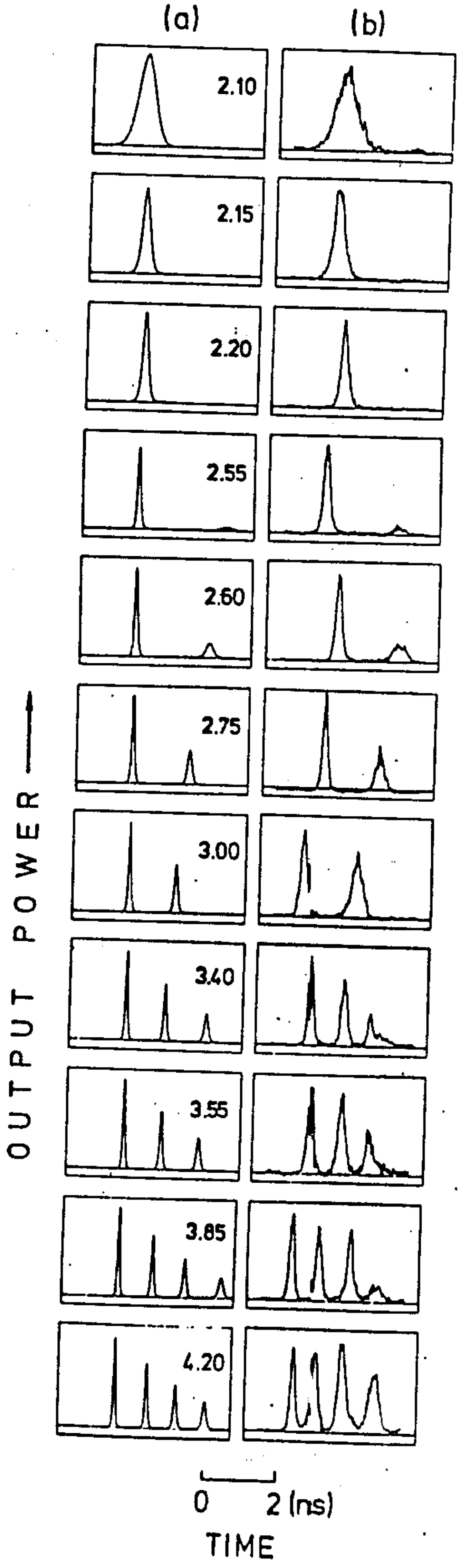
$$\tau_c = \frac{\eta L^3}{8 c \pi^2} \left(n (\sigma_e - \sigma_a) V \right)^2$$

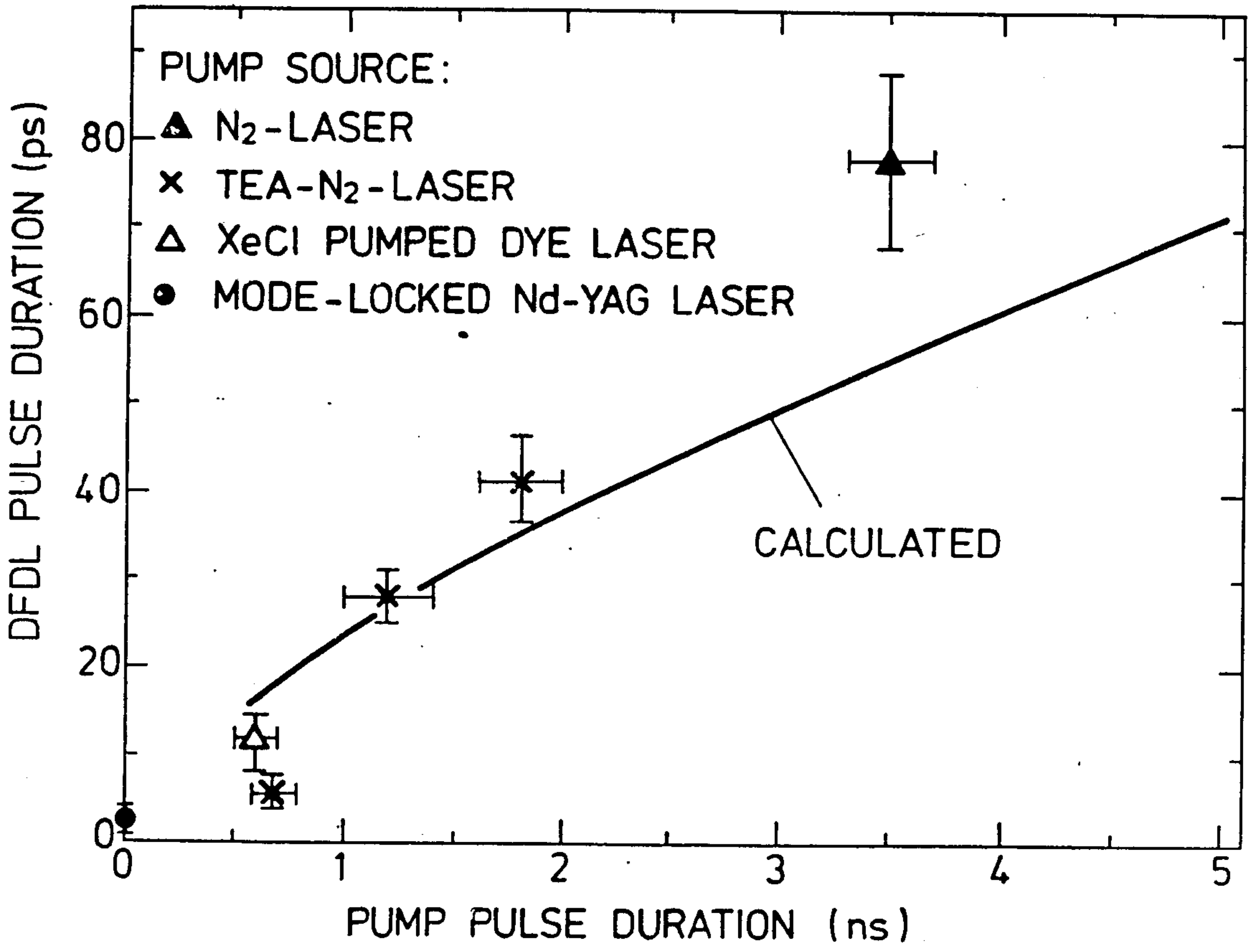
$$P_{out} = \frac{1}{2} \frac{hc}{\lambda_0} \cdot \frac{q L a b}{\tau_c}$$

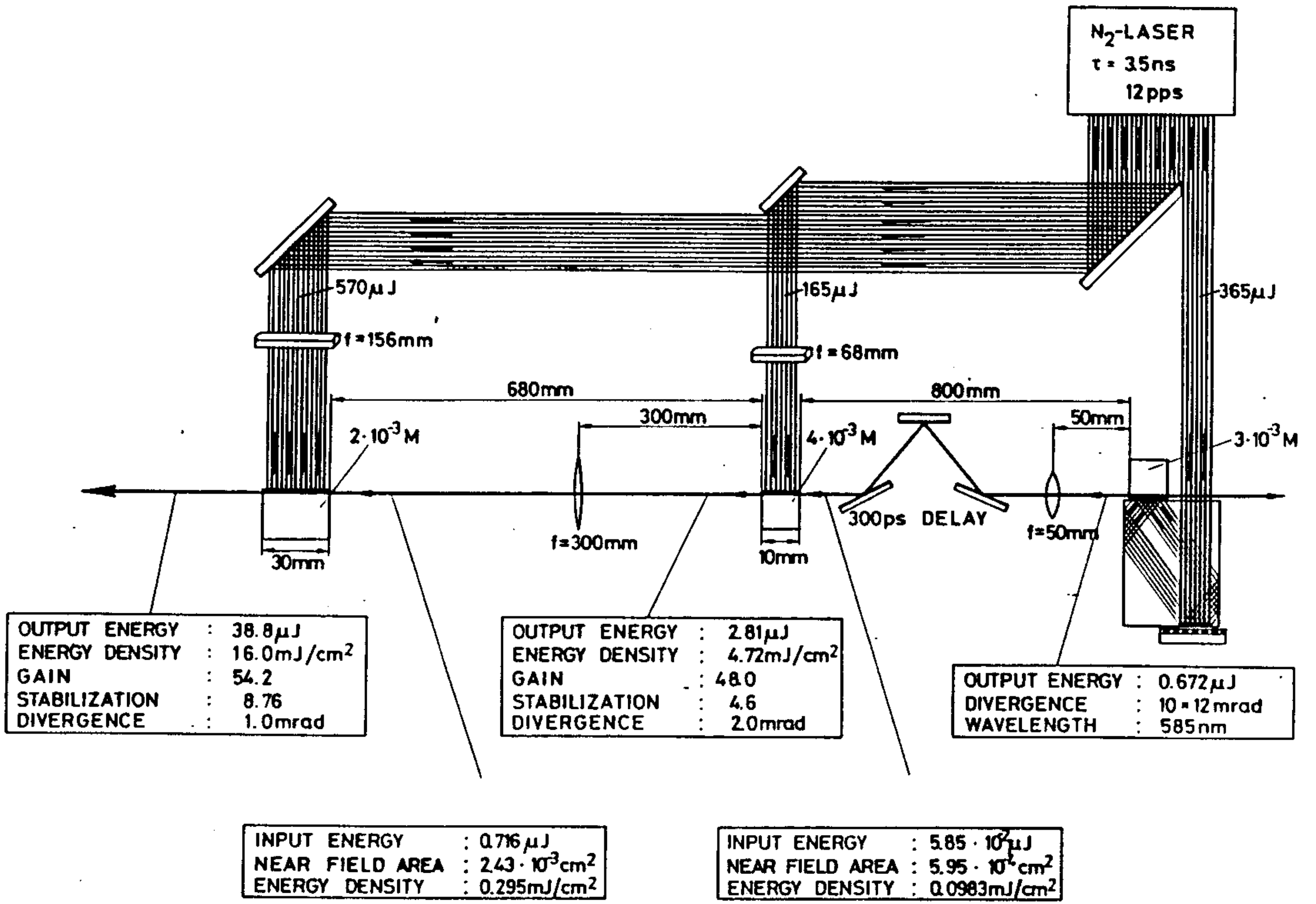
$$a = \frac{1}{N \sigma_p}$$

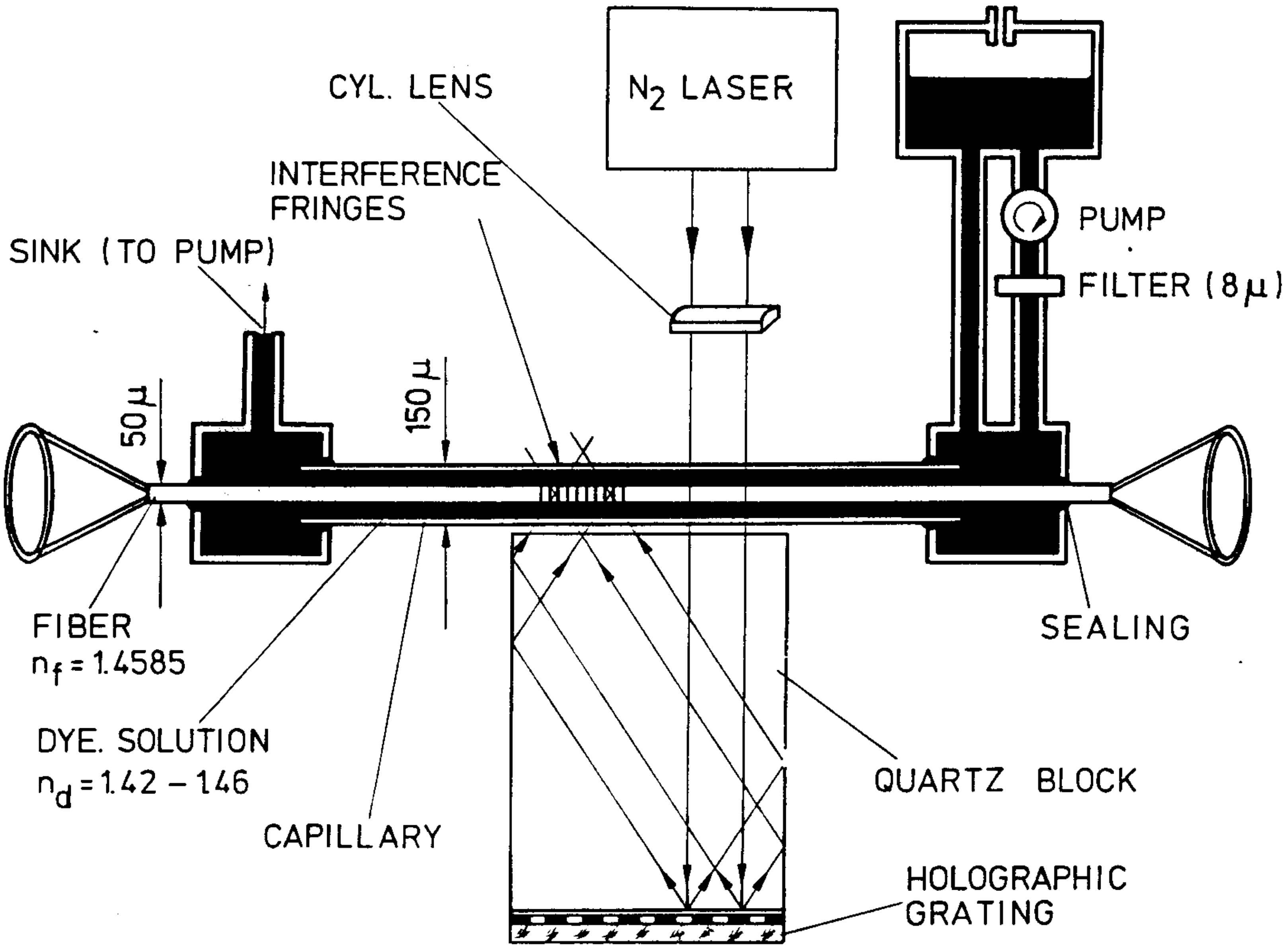
I_p, N, τ, L, a, b

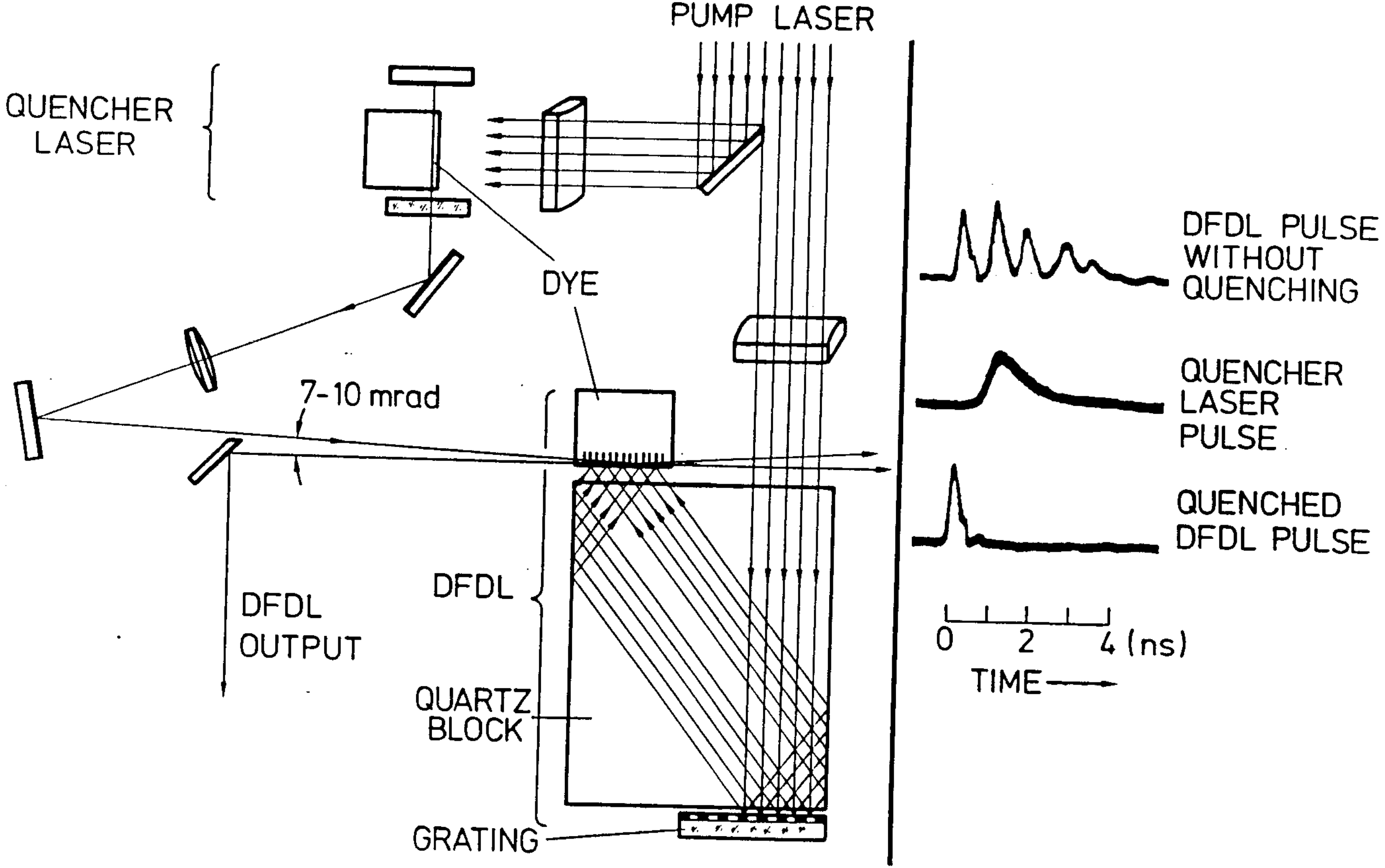


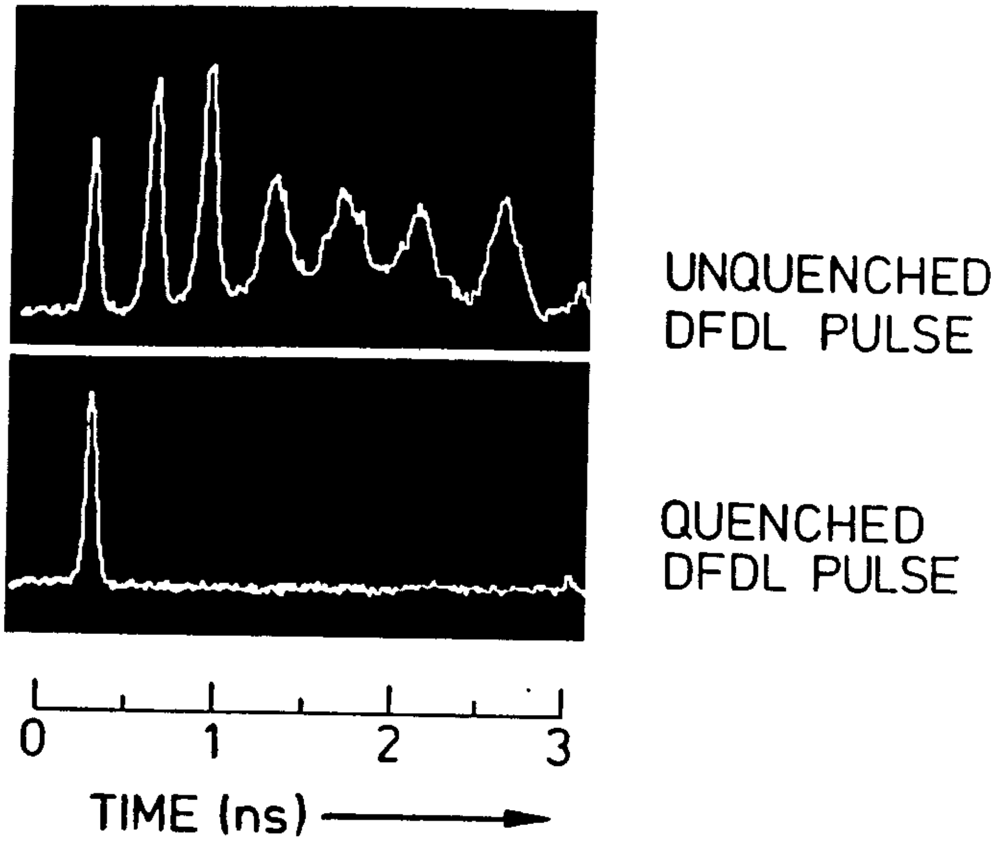

(a)
(b)
2.10
2.15
2.20
2.55
2.60
2.75
3.00
3.40
3.55
3.85
4.20
OUTPUT POWER
0
2 (ns)
TIME

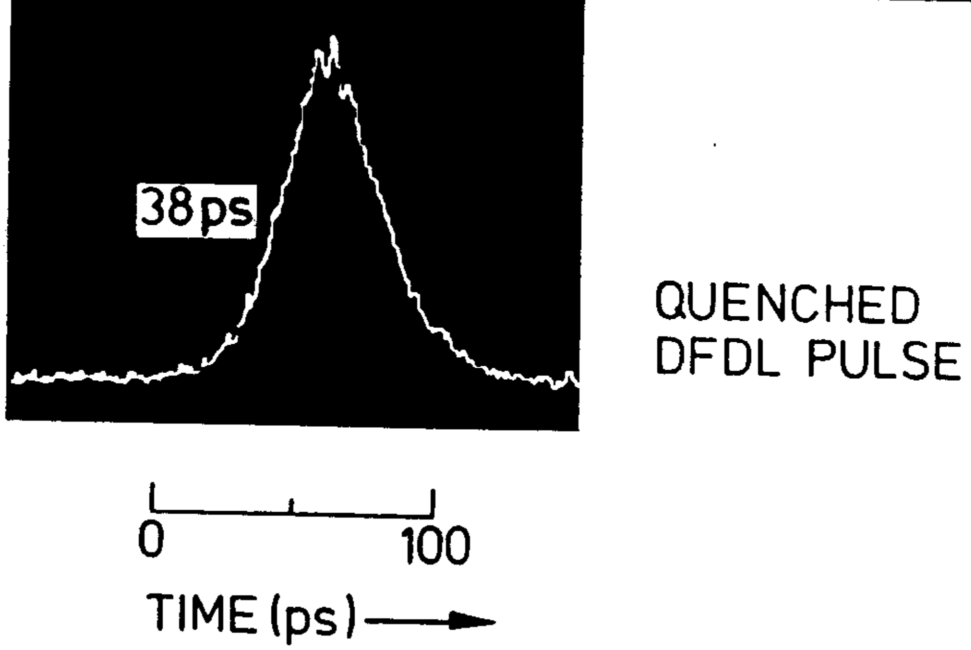

PUMP SOURCE:
▲ N_2-LASER
× TEA-N_2-LASER
△ XeCl PUMPED DYE LASER
● MODE-LOCKED Nd-YAG LASER
CALCULATED
DFDL PULSE DURATION (ps)
0
20
40
60
80
PUMP PULSE DURATION (ns)
0
1
2
3
4
5

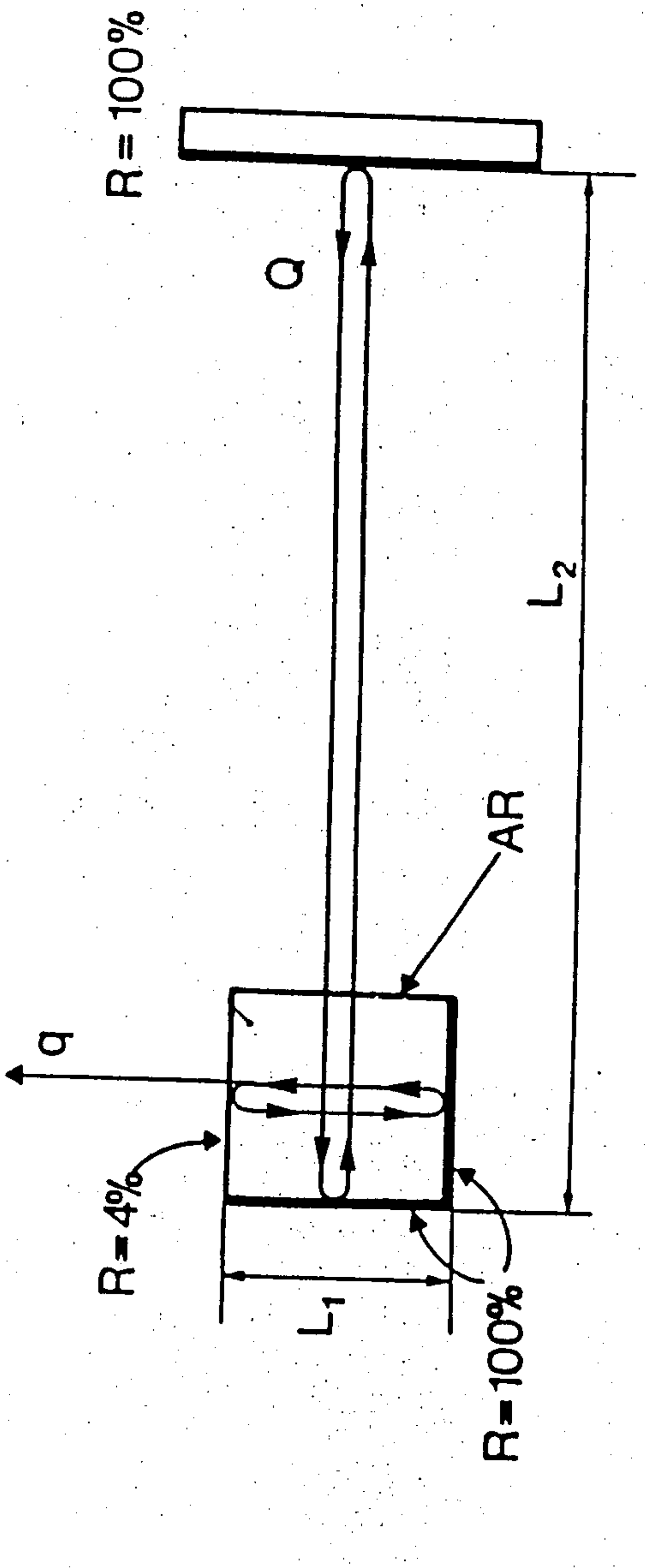

N_2-LASER
τ = 3.5ns
12pps
570µJ
f = 156mm
165µJ
f = 68mm
365µJ
680mm
300mm
800mm
50mm
$2 \cdot 10^{-3}$ M
$4 \cdot 10^{-3}$ M
$3 \cdot 10^{-3}$ M
300ps DELAY
f=300mm
f=50mm
30mm
10mm
OUTPUT ENERGY : 38.8µJ
ENERGY DENSITY : 16.0mJ/cm^2
GAIN : 54.2
STABILIZATION : 8.76
DIVERGENCE : 1.0mrad
OUTPUT ENERGY : 2.81µJ
ENERGY DENSITY : 4.72mJ/cm^2
GAIN : 48.0
STABILIZATION : 4.6
DIVERGENCE : 2.0mrad
OUTPUT ENERGY : 0.672µJ
DIVERGENCE : 10 × 12 mrad
WAVELENGTH : 585nm
INPUT ENERGY : 0.716µJ
NEAR FIELD AREA : $2.43 \cdot 10^{-3}$cm^2
ENERGY DENSITY : 0.295mJ/cm^2
INPUT ENERGY : $5.85 \cdot 10^{-2}$µJ
NEAR FIELD AREA : $5.95 \cdot 10^{-2}$cm^2
ENERGY DENSITY : 0.0983mJ/cm^2

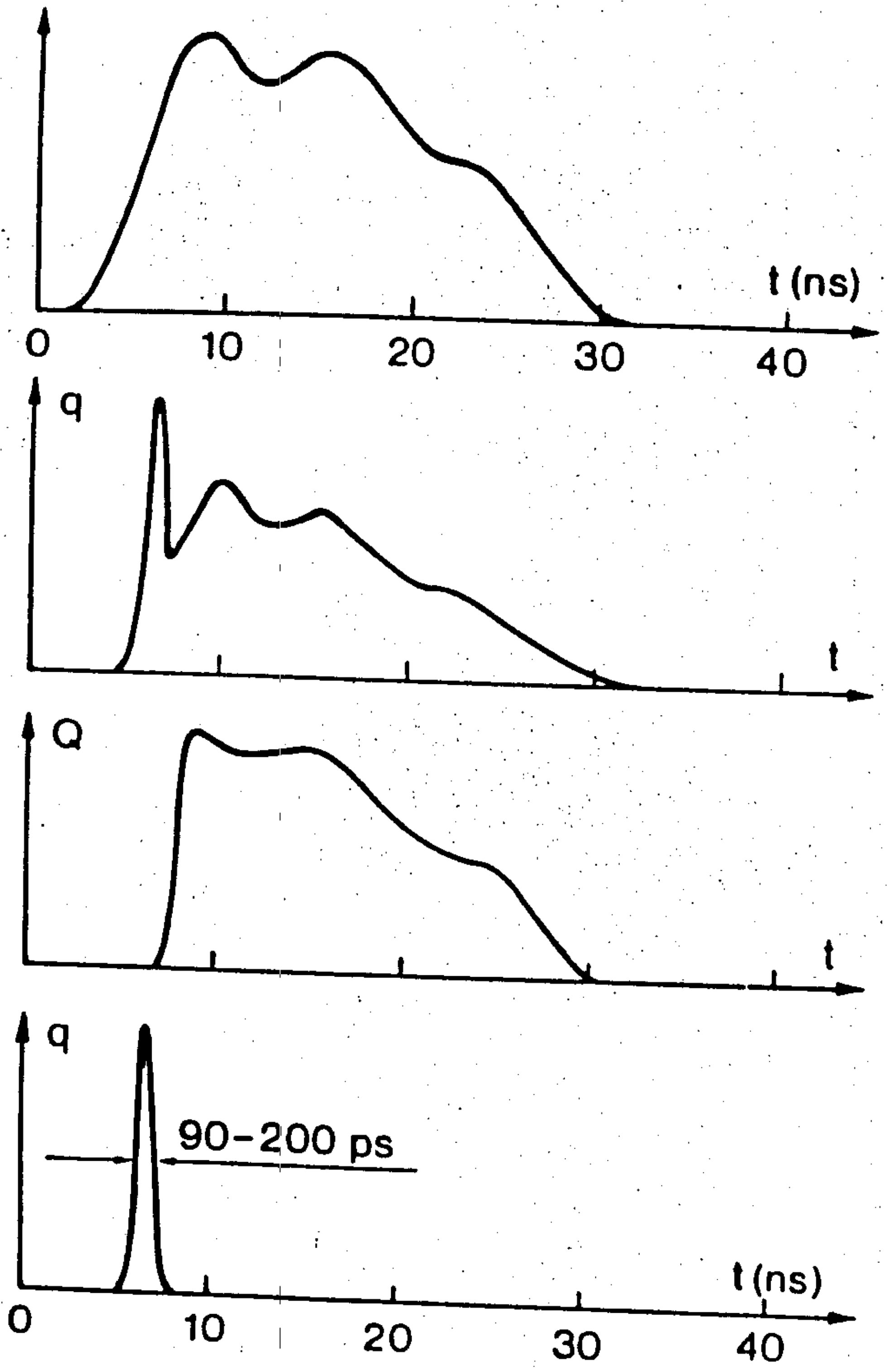

CYL. LENS
N_2 LASER
INTERFERENCE FRINGES
PUMP
FILTER (8μ)
SINK (TO PUMP)
50μ
150μ
FIBER
$n_f = 1.4585$
SEALING
DYE. SOLUTION
$n_d = 1.42 - 146$
QUARTZ BLOCK
CAPILLARY
HOLOGRAPHIC GRATING

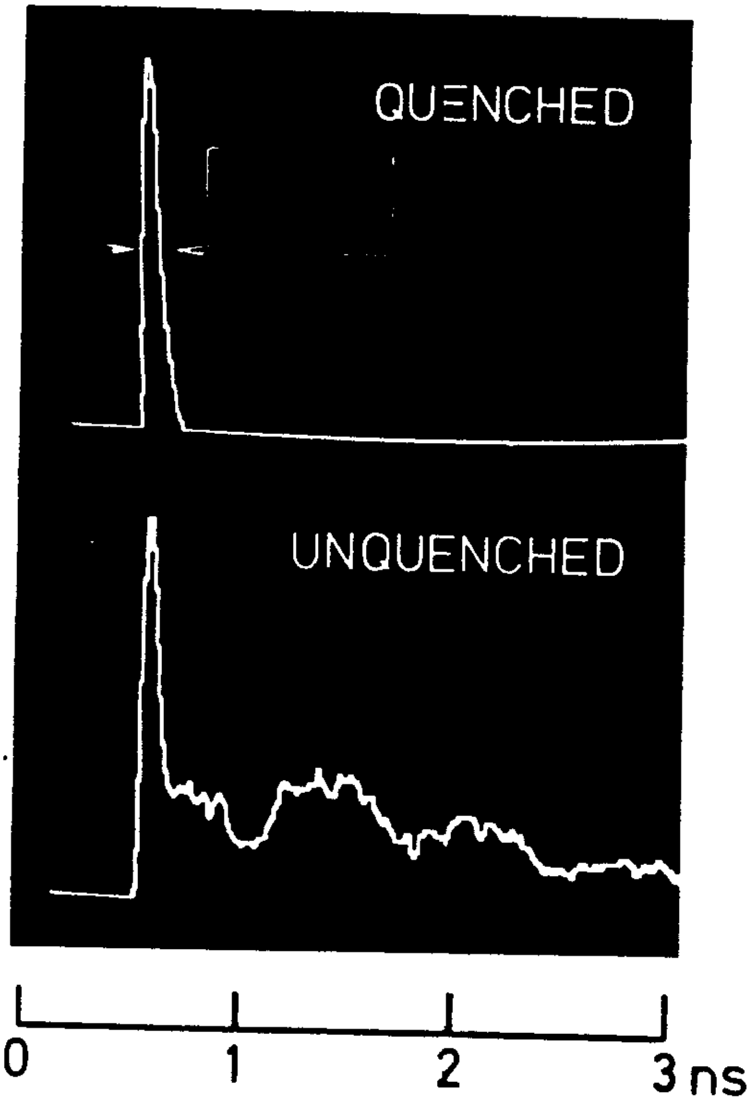

PUMP LASER
QUENCHER LASER
DYE
7-10 mrad
DFDL OUTPUT
DFDL
QUARTZ BLOCK
GRATING
DFDL PULSE WITHOUT QUENCHING
QUENCHER LASER PULSE
QUENCHED DFDL PULSE
0
2
4 (ns)
TIME


UNQUENCHED DFDL PULSE
QUENCHED DFDL PULSE
0
1
2
3
TIME (ns)

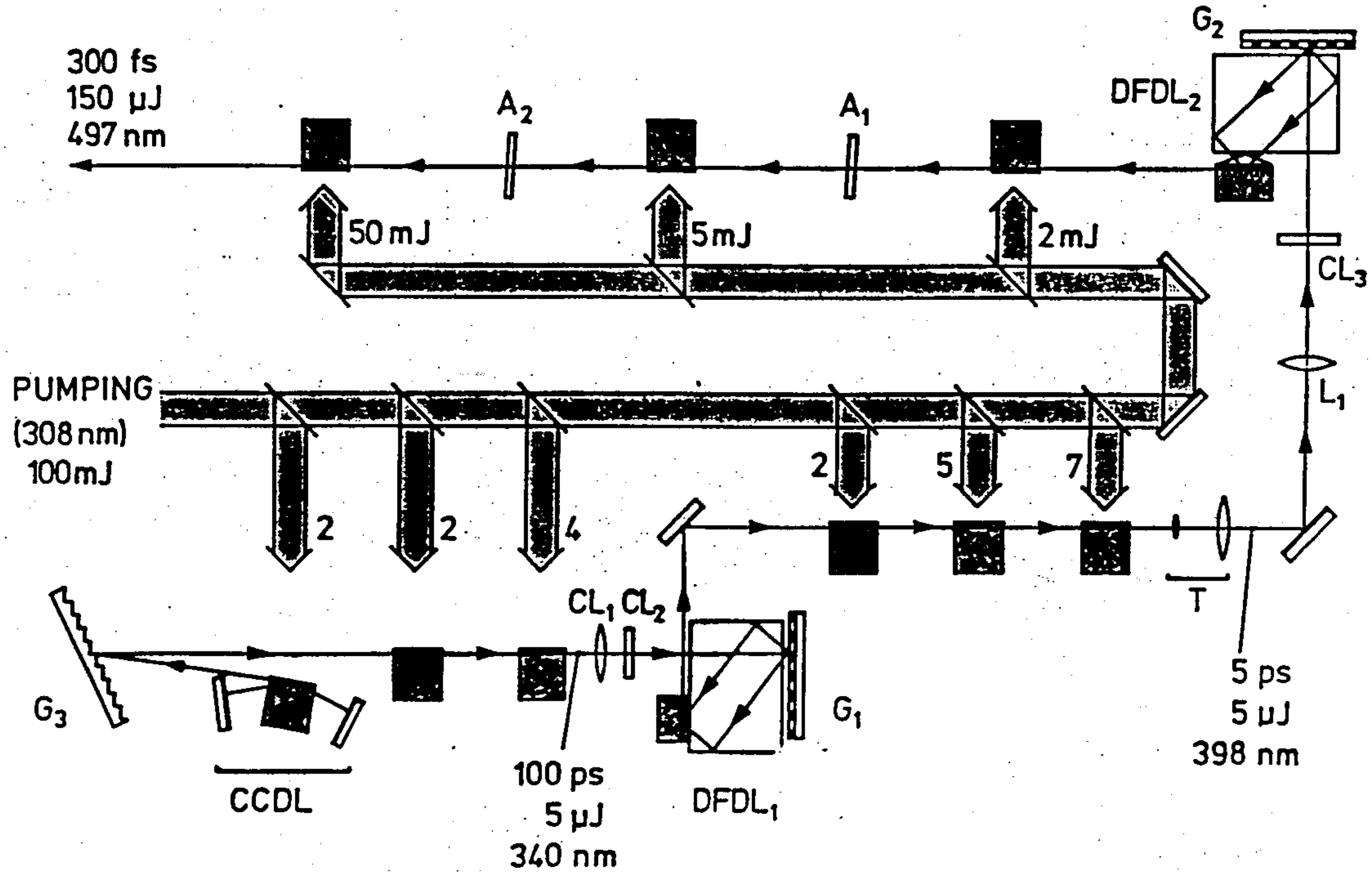

38ps
QUENCHED DFDL PULSE
0
100
TIME (ps)


R = 100%
Q
L_2
AR
q
R = 4%
L_1
R = 100%


t (ns)
0
10
20
30
40
q
t
Q
t
q
90-200 ps
t (ns)
0
10
20
30
40



25


300 fs
150 µJ
497 nm
A$_2$
A$_1$
G$_2$
DFDL$_2$
50 mJ
5 mJ
2 mJ
CL$_3$
L$_1$
PUMPING
(308 nm)
100 mJ
2
5
7
2
2
4
T
CL$_1$ CL$_2$
G$_3$
G$_1$
CCDL
100 ps
5 µJ
340 nm
DFDL$_1$
5 ps
5 µJ
398 nm

Tunable Picosecond Pulse Generation by an N_2 Laser Pumped Self Q-Switched Distributed Feedback Dye Laser

ZSOLT BOR

***Abstract*—The temporal characteristics of a distributed feedback dye laser are investigated both theoretically and experimentally. It is shown that the feedback is provided by the gain modulation and that the effect of refractive index modulation is negligible. The solution of the coupled rate equations predicts the generation of picosecond pulses. The mechanism of short pulse formation is self Q-switching which differs fundamentally from mode locking. A novel experimental arrangement for pumping the distributed feedback laser is described which makes it possible to obtain transform-limited pulses even with a partially coherent pumping source such as the N_2 laser. The experimental results show excellent qualitative and satisfactory quantitative agreement with the computer solutions. A very simple and reliable method for generation of tunable, nearly transform-limited single pulses with 36 ps duration (FWHM) is demonstrated experimentally. The feasibility of picosecond and subpicosecond pulse generation in the entire visible and near IR region is discussed.**

I. INTRODUCTION

THE major distinction between a distributed feedback laser (DFL) and a conventional laser is that the DFL does not use cavity mirrors: instead, feedback is provided via Bragg scattering from spatially periodic perturbations of the optical parameters of the active medium. Such parameters can be the refractive index of the laser medium [1]-[3], the optical gain, or both of these together [4]-[7], or the waveguide cross section in the case of thin film lasers [8]. In this paper we shall concentrate on the first two types of DFL, while a discussion of integrated optics DFL has appeared in a number of review articles [9]-[11].

Since Bragg scattering is highly frequency selective, narrow linewidths are readily achieved. If we consider first-order Bragg scattering the oscillation wavelength λ_0 in vacuo is approximately given by

$$\lambda_0 = 2\Lambda\eta \tag{1}$$

where η is the average refractive index of the active medium and Λ is the period of modulation. If the perturbation period or the refractive index can be varied experimentally then tuning is accomplished.

The periodic modulation of the refractive index or gain is produced either permanently (for example, by means of the technique used for preparing holographic gratings [1]-[3], or by ion milling [17], [18]), or temporarily during the time of the lasing action. The latter is realized by pumping the active medium with two interfering beams [4]-[7], [12]-[16]. In this case tuning is easily achieved by changing the interference angle of the pumping beams.

Threshold conditions, spectral selectivity, and modes of DFL in the linear approximation have been calculated by Kogelnik and Shank [19]. Hill, Watanabe [20], and Haus [21] have predicted the steady-state behavior of DFL taking into account the gain saturation above threshold. Chinn [22] calculated the steady-state behavior and the relaxation oscillations using a rate equation model and small-signal analysis. To our knowledge, an experimental study of the time behavior of DFL pulses has not been previously published.

In this paper we shall treat both theoretically and experimentally the time behavior of DFL. The analysis is confined to those distributed feedback dye lasers (DFDL) in which the periodic modulation is accomplished by the interference of the pumping beams. In Section II the rate equations are presented, and in Section III numerical solutions are given and the short pulse formation mechanism is discussed. Section IV is devoted to a description of a new optical arrangement for pumping the DFDL with an N_2 laser. Experimental results confirming the rate equation analysis are presented, and a new method for single picosecond and subpicosecond pulse generation is proposed.

II. RATE EQUATIONS

Let us assume that the dye laser can be represented by an ideal four-level system, that the emission and the absorption bands of the dye molecules in solution are homogeneously broadened, and we shall neglect the effects of higher excited states and triplet states. The rate equations describing the lasing level population and the photon densities are

$$\frac{dn}{dt} = I_p \sigma_a N - \frac{\sigma_e c}{\eta} nq - \frac{n}{\tau} \tag{2}$$

$$\frac{dq}{dt} = \frac{\sigma_e c}{\eta} nq - \frac{q}{\tau_c} \tag{3}$$

where

N is the concentration of dye molecules (cm^{-3}) in the ground state, which at the pumping intensities used in the experiments is nearly equal to the total concentration of dye molecules.

Manuscript received October 4, 1979; revised November 28, 1979.

The author is with Attila Jozsef University, Szeged, Hungary.

η	is the average concentration of dye molecules in the first excited singlet state (cm^{-3}),
q	is the concentration of DFDL photons (cm^{-3}),
σ_e, σ_a	is the absorption and stimulated emission cross-section of dye molecules at the pumping and lasing wavelength, respectively (cm^{-2}),
τ	is the fluorescence lifetime in the absence of stimulated emission (s),
c	is the speed of light,
η	is the average refractive index of the dye solution,
I_p	is the average pump photon flux per unit area (photon $cm^{-1} \cdot s^{-1}$),
τ_c	is the equivalent cavity decay time.

In this model we assume a uniform transversely excited region of length L, height b, and depth a (Fig. 5). Thus, the exponential decrease of I_p with distance into the dye is approximated by a step function of height I_p and depth a. In order to equalize the number of incident and absorbed pump photons it is necessary to take the e^{-1} penetration depth:

$$a = \frac{1}{N\sigma_a}. \tag{4}$$

The validity of this assumption is consistent with the near field size measurement. n, I_p, and η are given values spatially averaged over one period of the interference pattern. Since the DFDL has no external cavity, the equivalent cavity decay time is defined more generally as the total photon number divided by the rate of photon loss. This approach was confirmed for DFL in [22].

If only output losses are considered, the cavity decay time τ_c for a first-order overcoupled DFDL is given by [22]

$$\tau_c(t) = \frac{\eta L^3}{2c\pi^2}\left[\left(\frac{\pi}{\lambda_0}\eta_1(t)\right)^2 + \left(\frac{1}{2}\alpha_1(t)\right)^2\right] \tag{5}$$

where $\eta_1(t)$ and $\alpha_1(t)$ are the amplitudes of the spatial modulation of the refractive index and of the gain coefficient, respectively [19]. The output power from one end of the DFDL (in watts) is calculated to be

$$P_{\text{output}} = \frac{1}{2}\frac{hc}{\lambda_0}\frac{qLab}{\tau_c}. \tag{6}$$

In the case of DFDL's described in this paper, the spatial modulation is produced by pumping the dye solution (Fig. 5) with an interference pattern

$$I_p(x, t) = I_p(t)\left(1 + V \sin\frac{2\pi}{\Lambda}x\right) \tag{7}$$

where V is the visibility of the interference pattern and $I_p(t)$ is the pumping pulse shape. The latter is assumed to be a Gaussian pulse with peak power I_p.

For pumping intensities and pumping durations which are typically used in the DFDL, the biggest contribution to the refractive index change is due to thermal effects and thus

$$\eta_1(t) = \left(\frac{\partial n}{\partial T}\right)_p \Delta T(t) \tag{8}$$

where $\Delta T(t)$ is the amplitude of temperature grating induced by the sinusoidal pumping (7). The method of calculation of $\Delta T(t)$ is given in the Appendix.

The sinusoidal pumping (7) induces not only a temperature grating but also a gain grating. It is easy to show that the amplitude of the spatial modulation of the gain can be calculated as

$$\alpha_1(t) = \sigma_e V n(t) \tag{9}$$

where $n(t)$ is the average population of the first excited singlet state. An important feature of the DFDL is that the cavity decay time $\tau_c(t)$ is time dependent and, therefore, for solving the coupled rate equations (2), (3), and (5), it is necessary to calculate the time dependent values of $\eta_1(t)$ and $\alpha_1(t)$.

III. Solution of Rate Equations

The rate equations were solved numerically with parameters corresponding to our experimental conditions: $N = 6.6 \cdot 10^{-3}$ M/1, $\sigma_a = 2.4 \cdot 10^{-17}$ cm^2, $\sigma_e = 1.45 \cdot 10^{-16}$ cm^2, $\tau = 5 \cdot 10^{-9}$ s, $\lambda_0 = 590$ nm, $\lambda_p = 337$ nm, $\eta = 1.44$, $V = 1$, $L = 0.9$ cm, $b = 0.025$ cm, $A = 0.5$, $\alpha = 4.4 \cdot 10^{-4}$ cal $\cdot$ cm$^{-1} \cdot$ s$^{-1} \cdot$ °C^{-1}, $c = 0.579$ cal $\cdot$ g$^{-1} \cdot$ °C^{-1}, $\rho = 0.785$ g $\cdot$ cm^{-3}, $(dn/dt)_p = 4 \cdot 10^{-4}$ °C^{-1}. The pumping pulse shape was assumed to be a Gaussian pulse with 5.5 ns FWHM.

Fig. 1 shows the plots of the computer solutions for different pumping rates. Note that the solution for n, q, τ_c depends only on the pumping rate $I_p \sigma_a N$ and not on the individual values of I_p, σ_a, and N. The output power I_{output} [see (6)] is inversely proportional to the concentration N, since so is the near field size a as well [see (4)]. The calculation verified that the DFDL is overcoupled, except for the early part of the pulse when the gain is still below threshold. (Overcoupled for our case means in terms of cavity decay time $\tau_c > 2.35$ ps.)

In Fig. 1 we also indicate the stationary threshold values of the excited singlet state population n_{th} and of the pumping rate $(I_p \sigma_a N)_{\text{th}}$

$$n_{\text{th}} = \frac{2}{\sigma_e L}\left(\frac{\pi}{V}\right)^{2/3} \tag{10}$$

$$(I_p \sigma_a N)_{\text{th}} = \frac{n_{\text{th}}}{\tau}. \tag{11}$$

These values have been calculated from (2), (3), and (5) assuming $dn/dt = 0$, $dq/dt = 0$, and $q = 0$.

The computer solution predicts the generation of a series of picosecond pulses with decreasing amplitude. With increasing pump power the number of pulses increases, and the time separation between them decreases. There is a certain region of pumping rates, where single picosecond pulses are generated. With increasing pumping rates the peak power of single pulses increases and the pulse duration decreases (see Fig. 2). The pulse shape is closer to a Gaussian than a Lorentzian (see Fig. 3).

The short pulse formation mechanism is similar to the formation of relaxation oscillations in ruby laser or in conventional dye lasers with short external cavity [23], namely the interaction between excess population $(n - n_{\text{th}})$ and the number of photons in the cavity. But there is also an important difference between these mechanisms. As it is shown in the

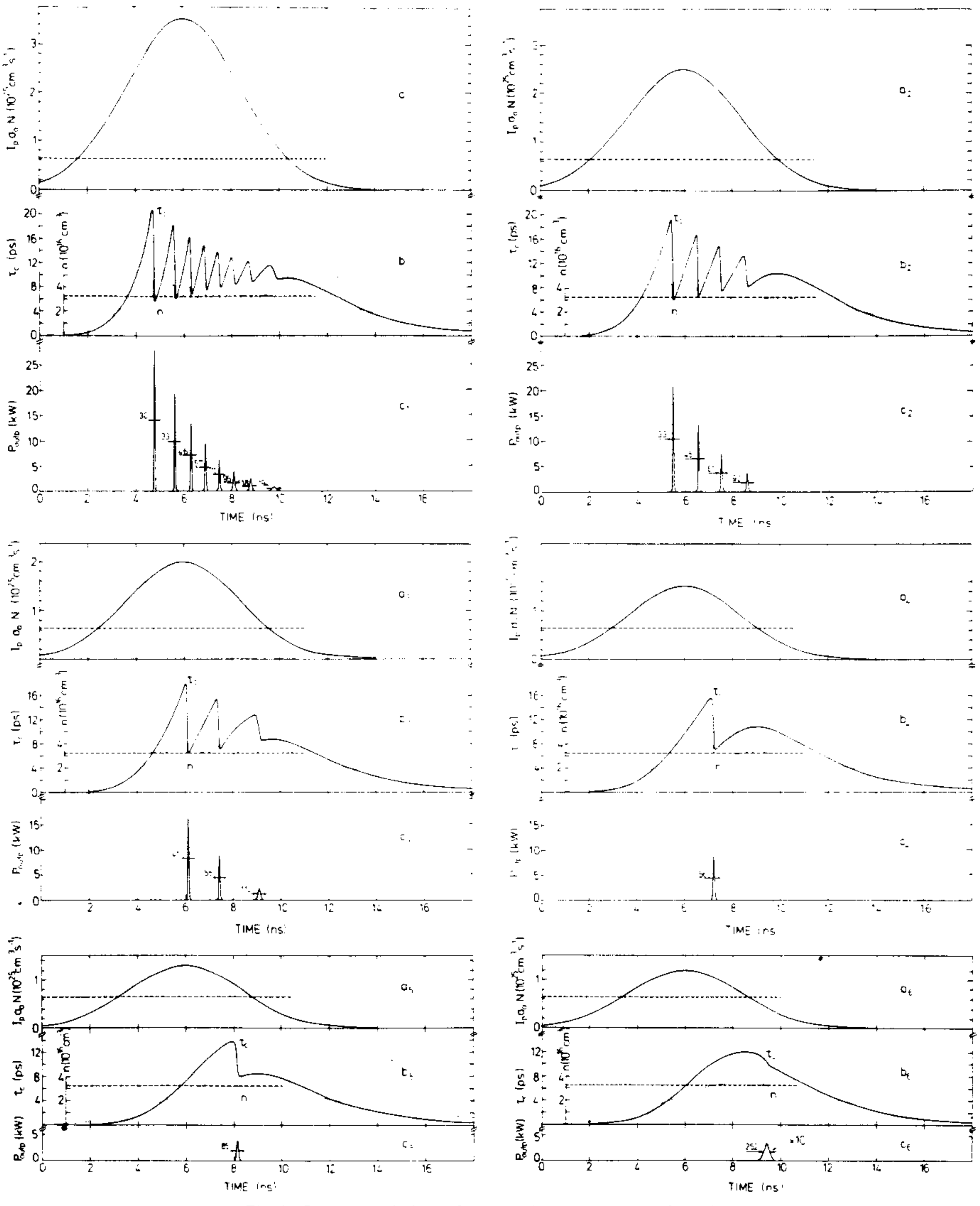


Fig. 1. Computer solutions of the coupled rate equations for a $6.6 \cdot 10^{-3}$ M/l rhodamine 6G solution. (a) Pumping rate ($I_p \sigma_a N$); (b) population of the first excited singlet state (n) and equivalent cavity decay time (τ_c); (c) output power of the distributed feedback dye laser (P_{output}); pulse durations in picoseconds are also indicated. The stationary threshold values of the pumping rate and of the excited state population are shown by dashed lines in (a) and (b), respectively. A 1 kW pumping power corresponds to a pumping rate $I_p \sigma_a N = 0.717 \cdot 10^{25}$ cm$^{-3} \cdot$ s^{-1}.

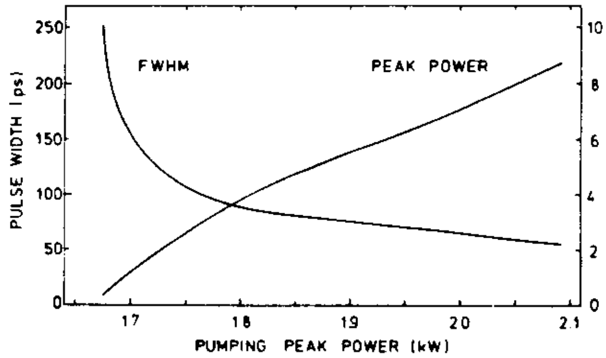


Fig. 2. Computed single pulse peak power and pulse duration (FWHM) as a function of pumping power. The parameters used for computation are the same as in Fig. 1.

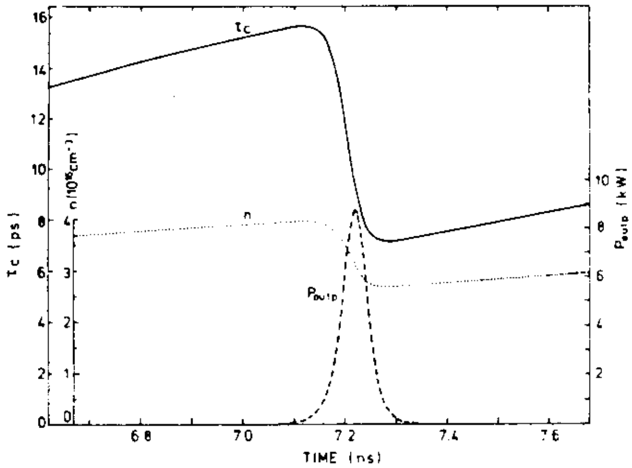


Fig. 3. Comparison of the single pulse shown in Fig. 1(c) with Gaussian and Lorentzian pulses.

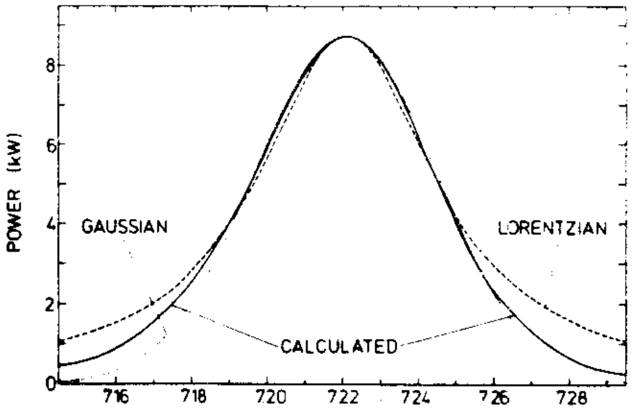


Fig. 4. Curves b_4 and c_4 from Fig. 1 shown on an expanded time scale.

Appendix, the DFDL is gain coupled and the effect of refractive index modulation on the feedback is negligible. Therefore, the equivalent cavity decay time is not constant as for a conventional dye laser, but $\tau_c \sim n^2$ [see (5)]. Thus, as shown in Fig. 4, τ_c has a large value during the rising half of the DFDL pulse and has a low value during the decaying half. In this way the change of τ_c during the pulse increases the absolute value of dq/dt and consequently favors the formation and shortening of the DFDL pulses. Therefore, the short pulse formation mechanism described above can be called self Q-switching.

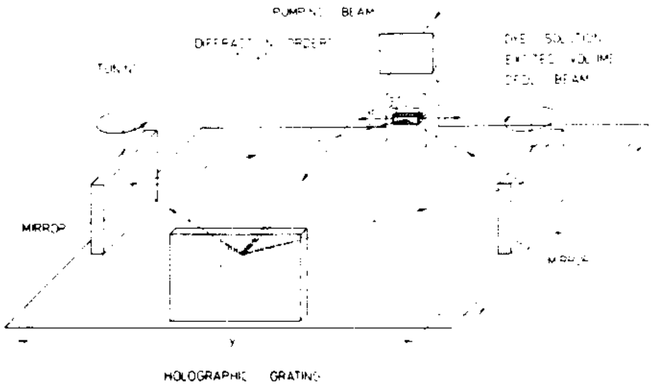


Fig. 5. The experimental arrangement used for pumping the distributed feedback dye laser with a partially coherent N_2 laser beam.

Note that the equivalent cavity decay time corresponding to the threshold population n_{th} is rather low (10.2 ps) and this also favors the short pulse formation.

IV. Experimental Arrangement and Results

A. Experimental Arrangement

The N_2 laser is an inexpensive, reliable, high repetition rate, high power UV laser, and is widely used to pump pulsed dye lasers, which are tunable throughout the visible and near infrared region. In order to demonstrate the practical importance of our method of single picosecond pulse generation we also used this pump source.

The N_2 laser pumped DFDL operation was successfully demonstrated earlier [26], but the linewidth was about 2.5 Å. The reason for this large linewidth is that the N_2 laser linewidth is about 1 Å, and consequently the interference pattern does not have a well defined fringe separation. Moreover, since the spatial coherence of the N_2 laser beam is low, the visibility of the interference fringes is low too, and therefore the threshold gain is high, which causes a high level of amplified spontaneous emission [24]. With the new optical arrangement described here these two disadvantages of N_2 laser pumping have been successfully eliminated.

The key element of our pumping arrangement is a 2442 line/mm holographic grating, which is used as a beam splitter (see Fig. 5). The N_2 laser pump beam is diffracted into the +1 and -1 orders, and then recombined on the dye cell. Higher orders of diffraction for normal incidence do not occur. The zeroth-order is not shown in Fig. 5. The beam is focused into a line on the dye cell with the cylindrical lens. The mirrors are perpendicular to the plane of the grating and parallel to the groves.

It can be easily shown that the fringe separation on the surface of the dye cell is given by

$$\Lambda = \frac{d}{2} \tag{12}$$

where d is the grove separation of the diffraction grating. This means that each spectral component of the N_2 laser beam produces an interference pattern with the same fringe separation.

Another important property of the arrangement is that if the geometrical relation

$$\frac{x}{y} = \sqrt{\left(\frac{d}{\lambda_p}\right)^2 - 1} \tag{13}$$

holds, then for each point on the dye cell the two interfering beams have been diffracted from the same point on the grating. That means that it is possible to get good visibility of the interference fringes with a pump laser having low spatial coherence. If a mirror type beam splitter had been used instead of the grating, these two properties would not occur.

The wavelength of the DFDL in vacuo is given by

$$\lambda_0 = d \cdot \eta \tag{14}$$

where η is the refractive index of the solution. The solvent we used was a mixture of ethanol and dimethylsulfoxid having a refractive index of 1.44. In that case the laser operated at 590 nm. The laser can be tuned in different ways; for example, by simultaneously rotating the mirrors in opposite directions about a vertical axis. A detailed description of the excellent spectral properties of this pumping arrangement is described in [27]-[29].

B. Experimental Results and Their Comparison with Theory

Fig. 6 shows streak camera traces of the DFDL pulses at different pumping intensities using a $6.6 \cdot 10^{-3}$ M/l rhodamine 6*G* solution.[1] The laser was operated at 12.5 Hz repetition rate, but a flowing dye cell was not used. The streak camera was an IMACON 600 type coupled with a PAR 1205 D optical multichannel analyzer. The vertical sensitivity from (a) to (e) was decreased about 10 times, the horizontal scale is unchanged. Since the DFDL and the streak camera were located in different parts of the laboratory and there was a long delay time associated with the streak camera triggering, the pulses were sent through a 90 nm long optical delay line. Under these experimental conditions the value of vertical sensitivity quoted in the caption to Fig. 6 is only approximate. The time resolution of the streak camera at the sweep speed used in Fig. 6 was 41 ps. The pulsewidths indicated in the figure are deconvoluted values assuming Gaussian shapes for both the DFDL pulse and the instrument function of the streak camera.

The shot-to-shot reproducibility of pulses at that concentration was rather poor. The pulses shown in Fig. 6 have been selected as typical for each pumping intensity. The experimental results show excellent qualitative agreement with the computer solution. The quantitative agreement is as follows: the threshold pumping power is 4 times higher than calculated (see Fig. 2), the energy of single pulses (just before the second pulse appears) is 3.5 times lower than calculated, and the pulse separation is about 200-250 ps instead of 600-1500 ps as calculated.

Considering the simplicity of the theory such agreement is

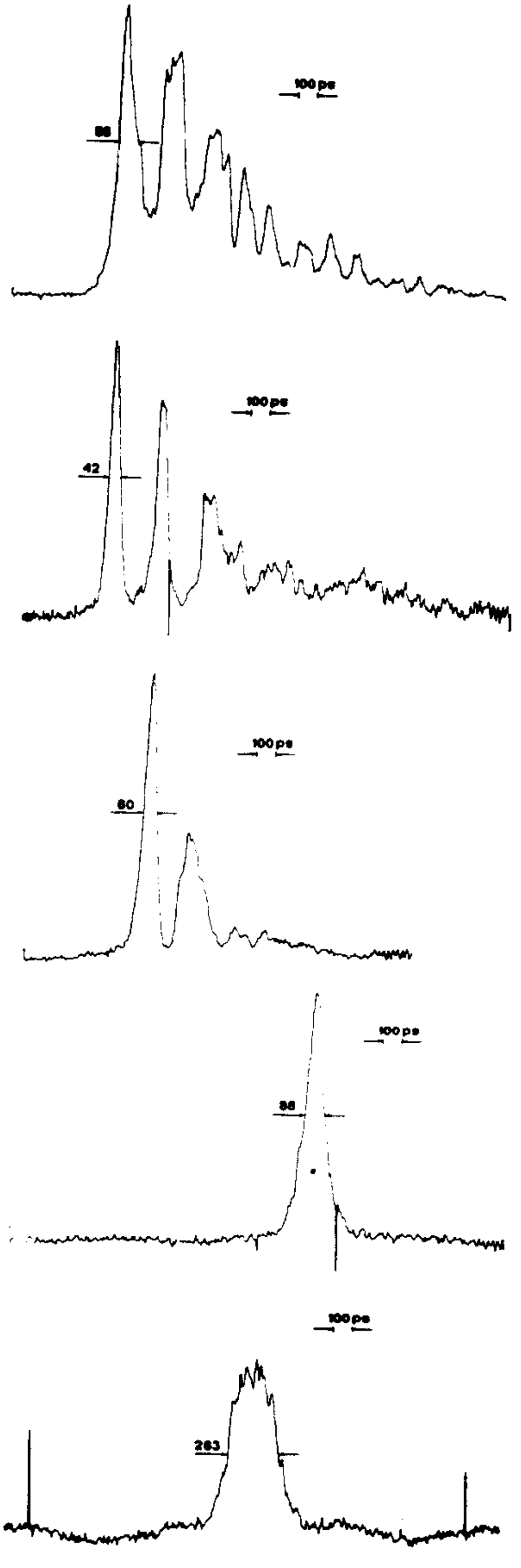


Fig. 6. Streak camera traces of distributed feedback dye laser pulses using a $6.6 \cdot 10^{-3}$ M/l rhodamine 6*G* solution. The pumping intensity was decreased from (a)-(e) about 2.5 times.

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[1] The streak camera measurements were made by Prof. A. Müller.

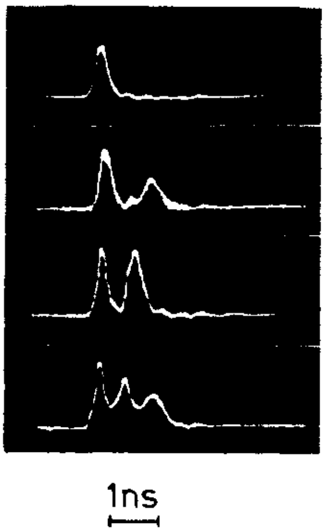

Fig. 7. Temporal behavior of the distributed feedback dye laser pulses using a $3 \cdot 10^{-3}$ M/l rhodamine 6G solution. The pumping intensity was increased in going from (a)-(d).

quite satisfactory, since apart from output losses no other type of loss (diffraction, self absorption by ground state molecules, absorption by higher excited states) are taken into account and no macroscopic dependence of I_p, n, and q is considered, etc. It appears that amplified spontaneous emission (ASE) [24] is the most important effect in determining the pulse buildup behavior not only in the DFDL but also in conventional dye lasers with an external cavity. The exact theoretical description of ASE is mathematically fairly complicated [24], [25] but it is expected that the presence of ASE decreases the DFDL pulse buildup time and the time separation between pulses. In order to get a better quantitative agreement between theory and experiment it is possible to reduce the level of ASE by using lower dye concentrations. The following results have been obtained with a $3 \cdot 10^{-3}$ M/l rhodamine 6G solution. Unfortunately no streak camera measurements are available.

Fig. 7 shows the DFDL pulses measured with a TVHR 13 biplanar photodiode and a SODERN S 501 5 GHz real time oscilloscope. The individual pulses are not completely temporally resolved. The shot-to-shot pulse reproducibility at that concentration was excellent. In this case the time separation between pulses was about 600-900 ps which agrees more closely with the theoretical calculations.

The computer solution of equations (2), (3), and (6) also gives the time variation of the excited state population $n(t)$ [see Fig. 1(b)]. Exact measurement of $n(t)$ is difficult. However, by measuring the intensity of ASE it is possible to make an estimate, since a higher intensity of ASE corresponds to higher $n(t)$. Fig. 8 shows the experimental arrangement for simultaneous measurement of the ASE and DFDL pulses. The DFDL pulse was spectrally separated from the ASE with the help of a dispersion direct vision prism, attenuated 10 times, and the two pulses delayed with respect to each other. The dye solution used was $5 \cdot 10^{-3}$ M/l rhodamine 6G.

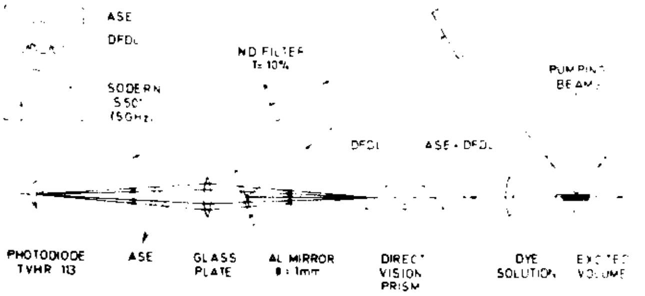

Fig. 8. The experimental arrangement used for simultaneous measurement of amplified spontaneous emission (ASE) and distributed feedback dye laser (DFDL) pulses.

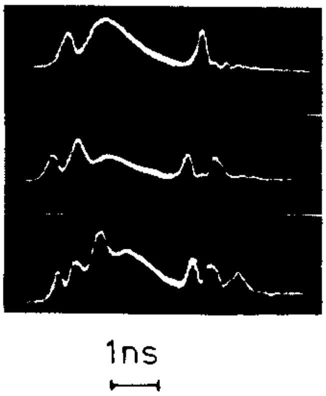

Fig. 9. Temporal behavior of ASE (left-hand part of trace) and DFDL pulses (right hand part of trace) using a $5 \cdot 10^{-3}$ M/l rhodamine 6G solution. The pumping intensity was increased in going from (a)-(e).

Fig. 9 shows the temporal behavior of the ASE and DFDL pulses. As predicted by the computer solution (see Fig. 1), each DFDL pulse causes a sharp decrease of $n(t)$ and consequently of ASE as well. These measurements confirm the validity of the rate equation model.

C. Single Picosecond and Subpicosecond Pulse Generation

From the point of view of practical applications, the most important characteristic of the DFDL is the possibility of generating single picosecond pulses by self Q-switching. We describe below some preliminary experimental results and discuss the feasibility of the method.

Fig. 2, which shows the single pulse peak power versus pumping intensity, predicts that the relative shot-to-shot peak power fluctuation of DFDL pulses is about 5 times greater than that of the pumping pulse. If a good commercial N_2 laser is operated at a gas pressure about 30 percent lower than that which gives the maximum output power, then its pulse-to-pulse stability is about ±2 percent, and therefore we can expect a ±10 percent stability for the picosecond pulses produced by the DFDL, which is reasonably good. A statistical analysis of 72 measured DFDL pulses [one of which is displayed in Fig. 7(a)] shows a ±8 percent peak power fluctuation. Fig. 2 shows the theoretically predicted lengthening of the pulse duration near threshold. This has been confirmed by the experimental result [see Fig. 6(d) and (e)]. The possibility of controlling the pulse duration could be very useful when the linewidth of DFDL pulses is Fourier transform limited. However, it requires an extremely stable pumping source which could be achieved using saturable absorbers.

The computer solutions predict pulse shortening as the length of the DFDL is decreased. For an $L = 3$ mm long rhodamine $6G$ laser we measured a 36 ps duration (FWHM) of single pulses. The most effective way of pulse shortening is to use shorter pumping pulses. Using a TEA N_2 laser as the pump it should be possible to generate 3 ps pulses, or with a mode-locked Nd:YAG pump 0.3 ps pulse generation should be possible. However, in the latter case the validity of the rate equation model should be reexamined. Previously [18] a DFDL was described in which 13 ps pulses were generated when the DFDL was pumped with 30 ps single pulses. However, this laser differed fundamentally from the DFDL which we describe here since the feedback was provided by surface corrugation of the substrate.

Single picosecond pulse generation with a self Q-switched DFDL has many advantages compared with the usual mode-locking techniques.

1) The system is inexpensive and reliable since it does not contain any sophisticated optical or electronic components.

2) The method should be applicable to any dye operating at any wavelength, since the system does not use saturable absorbers.

3) It is possible to obtain single picosecond pulses without pulse selectors. The pulse repetition rate is continuously variable and is determined by the repetition rate of the N_2 laser.

4) With the optical arrangement described above, any pulsed pumping laser, including N_2, excimer, or Cu vapor lasers, can be used.

5) It is easy to build an oscillator-amplifier system, since only a small fraction of the pumping laser power is needed to pump the DFDL, and the peak power of the generated single pulses is high enough for amplification.

6) The DFDL has superior spectral properties.

a) In contrast to the prediction of the linear stationary theory [19], no longitudinal modes are built up, and therefore tunability without mode hopping is very straightforward. Experimentally, continuous tuning over a 50 Å range without mode hopping was demonstrated [27].

b) Simultaneous measurement of the pulse durations and of the linewidths showed that the time-bandwidth product of the single pulses is less than 0.6 [27], [28].

The disadvantage of the method is that the DFDL beam divergence is about 10 mrad, however, it can be dramatically improved by using an amplifier stage.

V. CONCLUSIONS

We have shown that the distributed feedback dye laser pumped by the interference fringes produced by two pumping beams are gain coupled and that the equivalent cavity decay time is proportional to the square of the gain. The latter leads to a self Q-switching effect which is responsible for picosecond pulse generation. It has been demonstrated that short pulse generation by a self Q-switched distributed feedback dye laser has many advantages and potentially could become a competitor or replacement for the mode-locking technique.

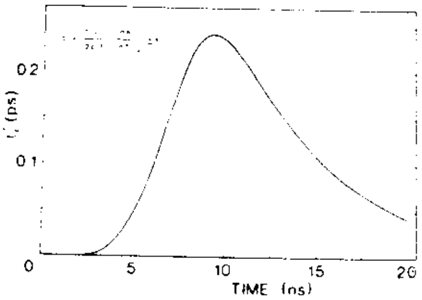

Fig. 10. τ_c^r as a function of time for pumping rate $I_p \sigma_a N = 3.5 \cdot 10^{25}$ $\text{cm}^{-3} \cdot \text{s}^{-1}$. τ_c^r describes that part of the feedback in terms of cavity decay time which is due to the refractive index modulation.

APPENDIX

The sinusoidal pumping produces a sinusoidal temperature grating. During the time of the laser action (~5 ns) no significant heat conduction occurs from the excited volume to the nonexcited part of the solution. Thus, the heat conduction can be treated as a one dimensional problem:

$$\frac{\partial T(t,x)}{\partial t} = \frac{\alpha}{\rho c} \frac{\partial^2 T(t,x)}{\partial x^2} + \frac{h\nu A \sigma_a N}{\rho c} I_p(t) \left(1 + V \sin \frac{2\pi}{\Lambda} x\right) \tag{A1}$$

where $h\nu$ is the photon energy of the pumping source, α is the heat conduction coefficient, ρ and c are the density and the specific heat of the solution. A is that part of the absorbed photon energy which is converted directly to heat and may be approximately calculated from

$$A = 1 - \phi \frac{\lambda_p}{\lambda_f} \tag{A2}$$

where ϕ is the fluorescence quantum yield, and λ_p and λ_f are the pumping and average fluorescence wavelengths.

We seek the solution of (A1) in the form

$$T(t,x) = T_0 + \frac{h\nu A \sigma_a N}{\rho c} \int_{-\infty}^{t} I_p(t)\, dt + \Delta T(t) \sin \frac{2\pi}{\Lambda} x \tag{A3}$$

where T_0 is the initial temperature of the solution and the second term is the spatially averaged temperature change of the medium, which increases linearly with the absorbed energy. The third term gives the temperature grating which is responsible for the refractive index modulation. Inserting (A3) into (A1) we obtain that $\Delta T(t)$ can be calculated from

$$\frac{d\Delta T(t)}{dt} = \frac{h\nu A \sigma_a N V I_p(t)}{\rho c} - \frac{\Delta T(t)}{\tau_t} \tag{A4}$$

where τ_t is the relaxation time of the temperature grating and can be calculated as

$$\tau_t = \frac{\rho c}{\alpha} \left(\frac{\Lambda}{2\pi}\right)^2. \tag{A5}$$

In Fig. 10 the time dependent values of $\tau_c^r(t) = (\eta L^3/2c\lambda_0^2) \cdot (\partial n/\partial T)_p^2 \Delta T^2(t)$ are shown for pumping rate $I_p \sigma_a N = 3.5 \cdot$

10^{25} cm^{-3} · s^{-1}. $\tau_c^r(t)$ describes that part of the feedback (in terms of cavity decay time) which is due to the refractive index modulation [see (5) and (8)]. Comparing $\tau_c^r(t)$ with $\tau_c(t)$, shown in Fig. 1, b_1, and taking into account that $\tau_c^r(t)$ is proportional to the square of the pumping intensity (I_p) [see (5), (8), and (A4)], we can conclude that for the pumping intensities we used, the feedback is provided by the gain modulation and the effect of refractive index modulation on the feedback is negligible.

This is an important conclusion since the question concerning which is the dominant feedback mechanism, the refractive index modulation, or the gain modulation has not been solved up to now.

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Picosecond Distributed Feedback Dye Lasers

ZSOLT BOR AND ALEXANDER MÜLLER

(Invited Paper)

***Abstract*—The distributed feedback dye laser is a source of single picosecond pulses tunable in a very wide wavelength range. Operational principles, experimental arrangements, and recent applications are reviewed in this paper.**

I. Introduction

Unlike other common laser oscillators, distributed feedback lasers possess no external elements providing optical feedback. As the name implies feedback necessary to obtain laser oscillations is provided within the active medium itself by Bragg scattering from spatially periodic perturbations of the optical parameters. Such parameters can be the refractive index of the laser medium [1]-[3], the optical gain, or both of these together [4]-[7]. This principle of feedback can be employed in solid media [1], [3] as well as in liquids [4], gases [9], and semiconductors [10].

Since laser media having a broad emission band are of particular interest with regard to continuous tuning of wavelength and to generation of extremely short light pulses, we intend to discuss in this paper exclusively *distributed feedback dye lasers* (DFDL) in the liquid phase.

II. The Principle of Distributed Feedback

An example of a method to produce distributed feed back in a solution of a laser dye, e.g., Rhodamine 6G in an organic solvent, is shown in Fig. 1 (right-hand side). The coherent light beam of the pump laser is split into two fractional beams by a beamsplitter. After reflection, the two beams interfere in the dye solution, creating an interference pattern of the pump light, which produces a periodic spatial modulation of the gain and the refractive index of the laser medium. As Kogelnik and Shank have shown already in 1971 [1], [4], [5], a light wave undergoes Bragg reflection at this periodical structure, causing optical feedback.

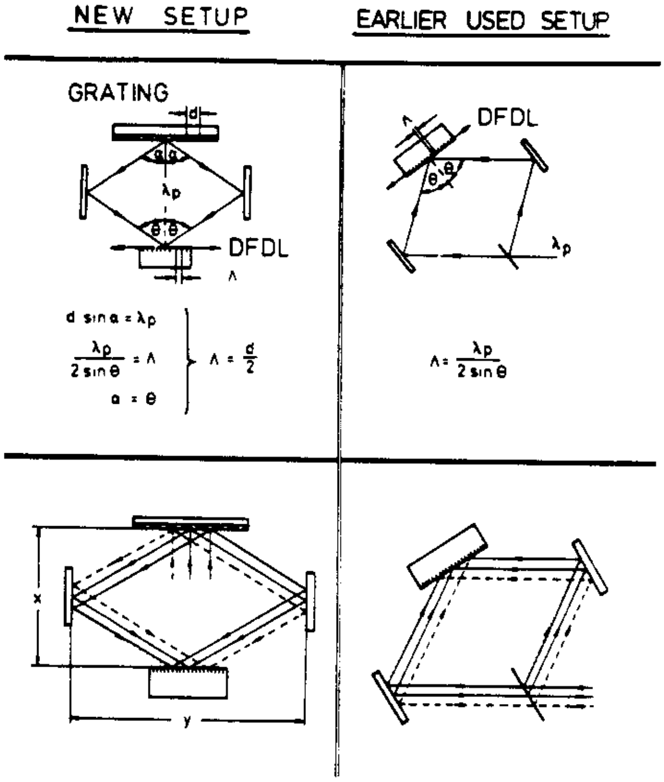

Fig. 1. Arrangements for producing distributed feedback in a cell with a laser dye solution: (right-hand side) with a 50 percent beamsplitting mirror, and (left-hand side) with a reflecting holographic diffraction grating as beamsplitter. λ_p = pump beam at wavelength λ_p, d = groove spacing of the grating, α = angle of diffraction on the grating, θ = interference angle, Λ = interference fringe separation, and DFDL = distributed feedback dye laser beam. The lower part illustrates the generation of the interference pattern by superposition of the fractional beams. From [26]. See text for details.

Spontaneous emission by excited dye molecules gives rise to two weak counterrunning waves. These experience gains in the inverted regions of the interference pattern are mutually coupled by Bragg reflection, thus producing a crescent standing wave by superposition. This process has a pronounced directionality, due to the fact that the pump-light-induced grating extends along the longitudinal axis of the laser medium. Since Bragg scattering is highly frequency selective, narrow linewidths are easily achieved. If we consider first-order Bragg scattering the oscillation wavelength *in vacuo* is approximately given by

$$\lambda_D = 2 \cdot n_L \Lambda \tag{1}$$

where λ_D is the generated laser wavelength, n_L is the re-

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fractive index of the dye solution at λ_D, and Λ is the separation of the interference fringes. Λ is given by

$$\Lambda = \lambda_P/2 \cdot \sin\theta \tag{2}$$

where λ_P is the pump wavelength and θ is the angle of intersection of the two fractional beams.

The steady-state theory of the DFDL has been treated by a number of authors [10]-[17]. We will not consider it further in the present paper.

III. EXPERIMENTAL ARRANGEMENTS

Any visible or ultraviolet pulsed laser producing pulses of at least 0.1 mJ can, in principle, serve as a pump source for the DFDL. Nitrogen and excimer lasers appear to be particularly attractive because of their easy handling properties and robustness in operation. The spatial and temporal coherence of these lasers is, however, relatively low. As one can see in Fig. 1 (right-hand side), different parts of the pump beam are brought to interference when a simple beamsplitter is used, producing fringes of low contrast if the spatial coherence is low. The same is true for arrangements using one prism as a beam divider [7]. One can eliminate this influence by using a holographic grating in place of the beamsplitter (Fig. 1, left-hand side) [18], or by inverting the wavefront with special t prism arrangements [21], [22]. Now, only those parts of the beam which have been diffracted from the same points of the grating are brought to interference.

With a diffraction grating, only those parts of the beam which have been diffracted from the same points of the grating are brought to interference if the following geometrical condition for the distance between the deflecting mirrors y, and between the grating and dye cell x, respectively, is obeyed:

$$x/y = \sqrt{(d/\lambda_P)^2 - 1} \tag{3}$$

where d = distance of the grooves of the grating (millimeters).

When the two deflecting mirrors are oriented parallel to each other and perpendicular with respect to the grating, the angles α and θ are equal. In this case, the arrangement has the additional property of being *achromatic* with respect to pump wavelength since we have now $\Lambda = d/2$. Using the grating equation (for the first order of diffraction)

$$\sin\alpha = \lambda_P/d, \tag{4}$$

together with (2) for the fringe separation, we obtain from (1):

$$\lambda_D = n_L \cdot d. \tag{5}$$

Because the pump wavelength has been eliminated, the effects of a relatively large bandwidth and the presence of several spectral components in the output of the pump laser are removed.

A method to stabilize the DFDL wavelength against a wavelength shift of the pumping laser has been described earlier [20].

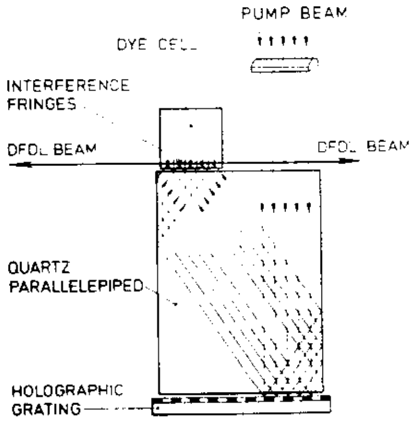

Fig. 2. Pumping arrangement for the distributed feedback dye laser. From [32].

A particularly simple version of DFDL may be set up by replacing the two deflecting mirrors with an optically finished quartz block having the shape of a rectangular parallelepiped. The proper dimensions can be calculated with the aid of (3) and the refractive index of quartz. In this case, internal reflections at the quartz-air interfaces serve to deflect the beams [32]. Fig. 2 shows an outline of this arrangement.

It is possible to coat the diffraction grating serving as beamsplitter directly onto the quartz block. This allows the use of a nitrogen laser or some other laser in the same wavelength region as the pump laser to produce DFDL output at short wavelengths. With an air interface between the diffraction grating and quartz block, no diffraction order would be produced for, e.g., a pump wavelength of 337 nm and $d = 1/3600$ mm since (4) would yield $\sin\alpha > 1$. If the grating is produced directly on the quartz block, however, the limiting pump wavelength will be shifted to longer wavelengths, corresponding to a factor which is equal to the refractive index of quartz [26].

Another achromatic setup in which the holographic grating has been made directly on the surface of the dye cell has been described by Vabishevich *et al.* [33]. This is also a simple arrangement. However, in this setup the visibility of the interference fringes formed by the beams diffracted into the +1 and −1 orders is lowered by the presence of the zero-order beam.

IV. WAVELENGTH TUNING

The possibilities of tuning the wavelength of the DFDL have been discovered and experimentally demonstrated already at an early time [4], [7], [24].

According to (1), the wavelength of the DFDL is proportional to the refractive index n_L and to the fringe separation Λ. A variation of n_L can be accomplished by mixing liquids with different refractive indexes [4]. Linear tuning of the laser wavelength through a wide range is possible, as the example of Fig. 3 shows for three Rhodamine dyes [18].

Continuous fine tuning can be achieved by pressure tuning, which also varies the refractive index of the dye so-

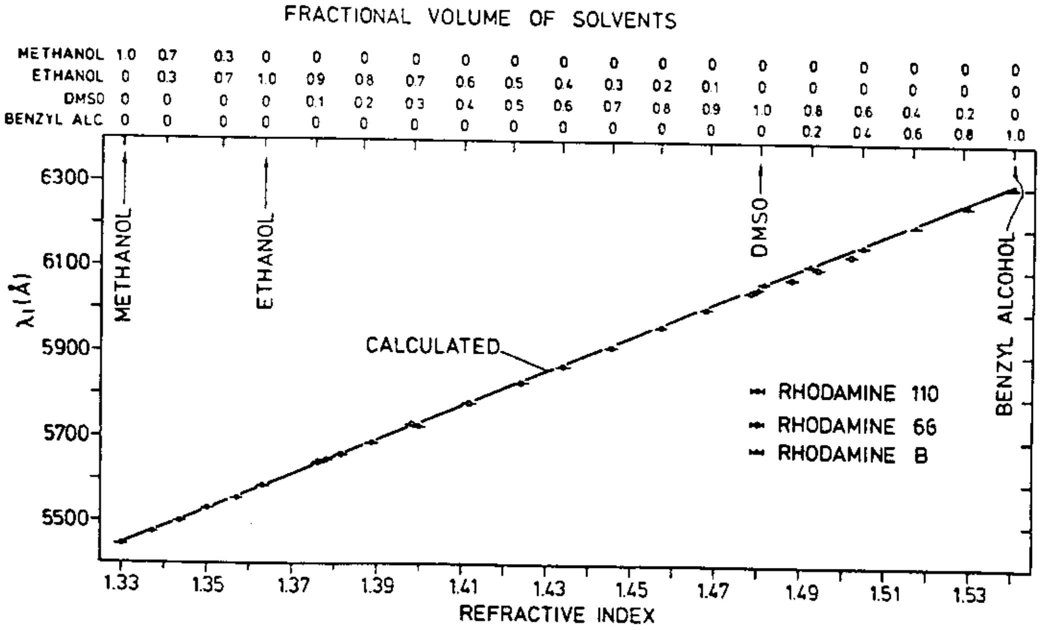

Fig. 3. Wavelength tuning of a DFDL by refractive index variation of the dye solution in solvent mixtures. From [18].

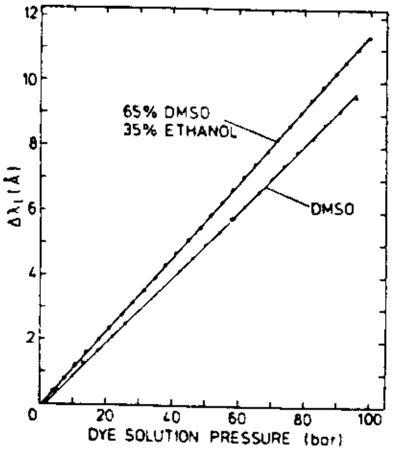

Fig. 4. Continuous tuning of DFDL wavelength by varying the internal pressure of the dye cell, shown for two solvents. Laser dye: Rhodamine 6G (3 mmol/L). DFDL wavelength: 590 nm for DMSO + ethanol, 605 nm for DMSO. From [25].

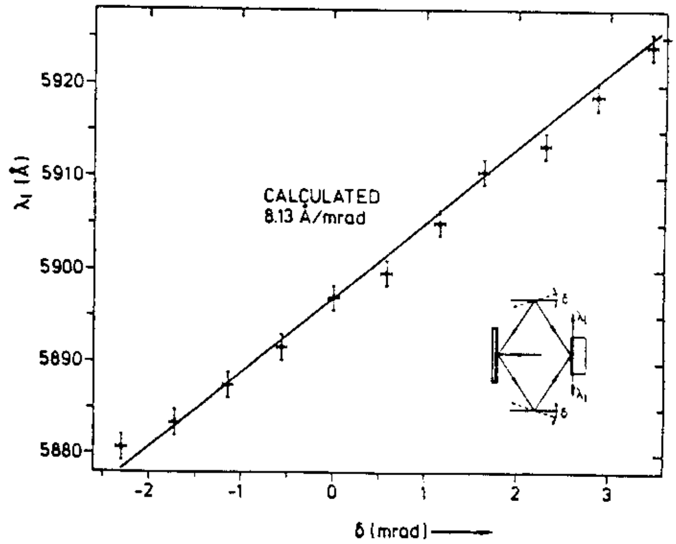

Fig. 5. Continuous tuning of DFDL wavelength by varying the separation of the interference fringes through mirror rotation. From [18].

lution (Fig. 4) [25]. Because of the low compressibility of solvents the tuning range is rather narrow, however.

A larger tuning range is obtained upon variation of the fringe separation, which, according to (2), can be changed by altering the interference angles θ (Fig. 5). This can be accomplished experimentally by rotating the two deflecting mirrors about vertical axes [4], [5], [18], [22], [27], [28]. Based on this principle of tuning, a computer-controlled DFDL has been constructed which achieves a wide tuning range by simultaneous rotation of the deflecting mirrors and variation of the grating to dye cell distance by stepping motors [29]. It has to be noted, however, that the *achromatic* characteristic of the arrangement is lost when the condition $\alpha = \theta$ is no longer satisfied. Two different schemes retaining the achromatic characteristic while the wavelength is tuned have been published recently [30], [31]. Tuning between 400 and 800 nm at transform-limited bandwidths has been demonstrated.

Wavelength tuning into the near-UV region may be accomplished by utilizing second-order Bragg reflection [5], [32]. In this case, the DFDL wavelength is given by

$$\lambda_D = 2 \cdot n_L \cdot \Lambda/\kappa \qquad (6)$$

where $\kappa = 2$.

Linewidth of DFDL emission will be treated in the context of temporal characteristics in a later section.

V. TEMPORAL CHARACTERISTICS OF DFDL EMISSION

Whereas the spectral characteristics of the DFDL have been known since 1971, its temporal behavior has been studied only recently. As a first approach, Chinn [34] presented a linear perturbation analysis of the rate equations.

An experimental observation of the pulsed output was only possible with the application of techniques having extremely high time resolution, which then stimulated a theoretical analysis with the aid of nonlinear rate equations [35]–[37].

The following system of rate equations may be used for modeling the temporal behavior of the DFDL:

$$\frac{dN}{dt} = I_p \cdot \sigma_P \cdot (N_0 - N) - \frac{\sigma_e \cdot c}{n_L} \cdot N \cdot Q - \frac{N}{\tau_f} \tag{7}$$

$$\frac{dQ}{dt} = \frac{(\sigma_e - \sigma_a)c}{n_L} \cdot N \cdot Q - \frac{Q}{\tau_c} + \frac{\Omega \cdot N}{\tau_f}. \tag{8}$$

Reabsorption of laser photons by molecules in their electronic ground state will be neglected.

The following definitions are used:

$N(t)$	spatially averaged density of molecules in the first excited singlet state S_1 (cm^{-3})
$Q(t)$	density of DFDL photons (cm^{-3})
N_0	density of dye molecules (cm^{-3})
$I_P(t)$	spatially averaged pump photon flux per unit area (cm^{-2}/s)
σ_P	absorption cross section from S_0 to higher-lying singlet states at the pumping wavelength (cm^2)
σ_a	excited state absorption cross section from S_1 to S_2 at the lasing wavelength (cm^2)
σ_e	stimulated emission cross section from S_1 to S_0 at the lasing wavelength (cm^2)
τ_f	fluorescence lifetime in the absence of stimulated emission and quenching processes (s)
τ_c	average lifetime of a photon in the DFDL (s)
c	speed of light *in vacuo* (cm/s)
Ω	factor determining that fraction of the spontaneous emission which propagates into the angular and spectral range of the DFDL beam.

The term describing the specific behavior of the DFDL is τ_c. In analogy to the usual description of the losses of a conventional resonator by an average "resonator decay time," τ_c represents an equivalent entity for the DFDL.

According to [35], τ_c can be calculated in the following way:

$$\tau_c = \frac{n_L \cdot L^3}{8 \cdot \pi^2 \cdot c} \cdot [N(\sigma_e - \sigma_a) \cdot V]^2 \tag{9}$$

where L is the length of the pumped volume (centimeters) and V is the visibility of the interference fringes formed by the pump light.

It can be shown that thermal modulation of the refractive index of the dye solution makes a negligible contribution in this case. Essentially, feedback is provided by gain modulation only. This has been taken into account in (9).

In contrast to the case of an external resonator, in the case of distributed feedback the loss term τ_c contains the square of the population of the upper laser level. It is this nonlinearity which is responsible for the typical "self-Q-switching" behavior of the DFDL.

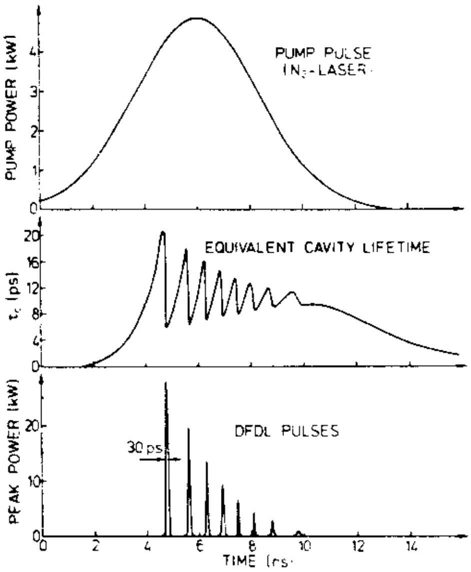

Fig. 6. Time courses of pump pulse, equivalent cavity decay time and DFDL pulses computed with the nonlinear rate equation model. Parameters of a Rhodamine 6G solution were used. From [35].

The system of differential equations (7), (8) can be solved numerically with the aid of a fourth-order Runge-Kutta procedure, and the time sources of $N(t)$, $Q(t)$, and $\tau_c(t)$ may be computed for selected pump pulses. Fig. 6 shows a typical example of the time courses of I_P, τ_c, and P_D, the output power of the DFDL, which is given by

$$P_d = \frac{h \cdot c\, Q}{2 \cdot \lambda_D \cdot \tau_c} \cdot L \cdot a \cdot b. \tag{10}$$

h	Planck's constant
a	penetration depth of the pump light into the dye solution (cm)
b	height of the excited volume (cm)

P_D has been determined in order to compare the results of the model computation to the experimental results.

As one can see, relaxation oscillations arise because of the nonlinear dependence of the "resonator losses" described by τ_c on the population N of the upper laser level. During pumping, τ_c grows at first and finally drops rapidly to a lower level when the gain is depleted at high light intensity. Simultaneously, with the drop of τ_c a short DFDL output pulse is produced as indicated by the time course of P_D (cf. Fig. 6). If pump pulse intensity is still above laser threshold afterwards, then the whole process can repeat.

It is possible to compare time courses of DFDL output pulses obtained with the aid of a high-resolution streak camera to results of the model computations if the latter are based on realistic values of the parameters. As Fig. 7 shows, there is very good agreement between the rate equation model and the experimental data.

One should notice that there exists a range between laser threshold of the second pulse where a single short pulse

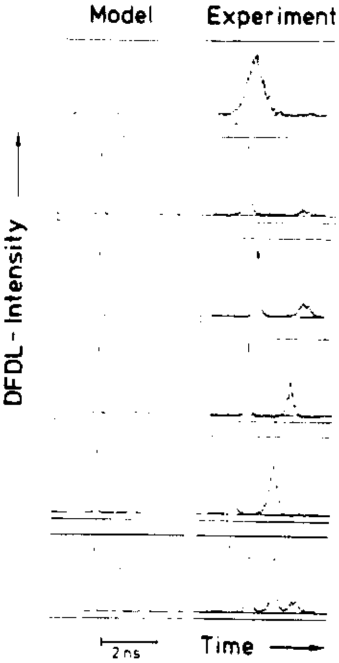

Fig. 7. Computed (left-hand side) and measured (right-hand side) pulses of a Rhodamine 6G DFDL. Pump power increases from top to bottom. The experimental results have been obtained with an 11 ps streak camera system. From [36].

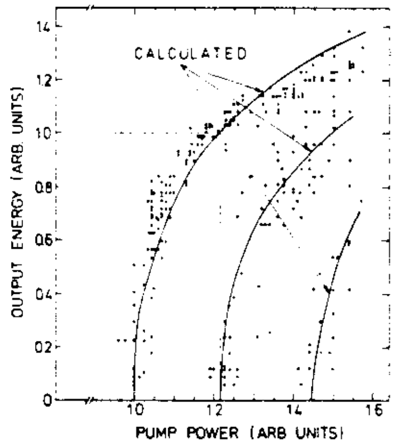

Fig. 8. Dependence of DFDL pulse energy on pump power. The solid lines are results of the nonlinear rate equation model. From [37].

is generated. The DFDL pulse duration is about 50 times shorter than that of the pump pulse if one operates just below the threshold of the second pulse. In this way it is possible to generate single DFDL pulses of 70 ps duration when nitrogen laser pumping of 3.5 ns pulse duration is applied. No additional electronic single-pulse selecting devices are necessary, contrary to the case of solid-state lasers. This property is indeed a very useful one for practical applications; the more since it exists throughout the spectral range between 360 and >700 nm, and pulse shape remains essentially unchanged for all laser dyes.

Calculation of the energy of the DFDL pulses in dependence on pump power using the rate equation model yields the diagram shown in Fig. 8 (solid lines) [37]. The experimental results (data points) exhibit good agreement with the theoretical values in this case too.

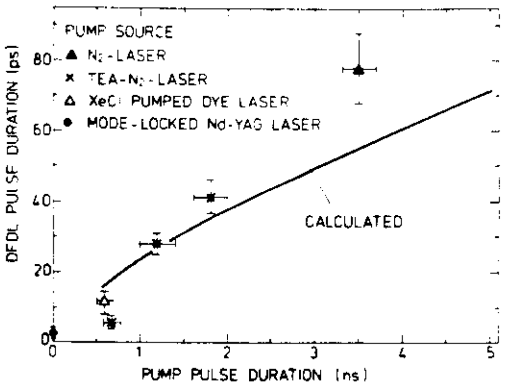

Fig. 9. Reduction of DFDL pulse duration when various pump lasers are used. From [26].

A detailed analysis of the temporal and energetic properties of DFDL output pulses as a function of the various parameters occurring in the system of rate equations has yielded very good agreement between model and experiments. The cited literature should be consulted for further details.

VI. SHORTENING THE PULSE DURATION

Further reduction of the DFDL pulse duration upon decrease of the duration of the exciting pulse $I_P(t)$ as predicted by the rate equations model has been verified experimentally (Fig.9) [38]. The pump source consisted of a TEA-N_2 laser as oscillator, coupled to a normal low-pressure nitrogen laser serving as amplifier. By variation of the length of the TEA laser electrodes and addition of a cuvette containing a saturable absorber between oscillator and amplifier it was possible to vary the duration of the pump pulses. In this way, pulses of 3.5, 1.8, 1.2, and 0.7 ns duration were available.

In order to apply still shorter pump pulses, one may use a mode-locked Nd:YAG laser with frequency doubling or tripling in nonlinear optical crystals. Fig. 10 shows an arrangement [39] in which a DFDL oscillator and one dye amplifier stage are pumped by the third harmonic (355 nm) of a Nd:YAG laser having a pulse duration of 16 ps. The second dye amplifier is pumped by 18 ps pulses of the second harmonic (532 nm).

The DFDL pulse duration was measured by the single-pulse autocorrelator shown in Fig. 11 [39]. Here, the two gratings G_1 and G_2 are used to form a continuous temporal delay across the two output beams [40]. The spatial distribution of the SHG beam displays the background-free second-order autocorrelation function of the input pulse.

The optical spectrum of the DFDL pulse is recorded simultaneously using a high-resolution grating spectrograph. Optical multichannel analyzers are used for recording in both cases.

For a DFDL operated with Rhodamine 6G at 590 nm, experimentally determined reciprocal bandwidths $1/\Delta\nu$ have been plotted in Fig. 12 versus pulse durations ob-

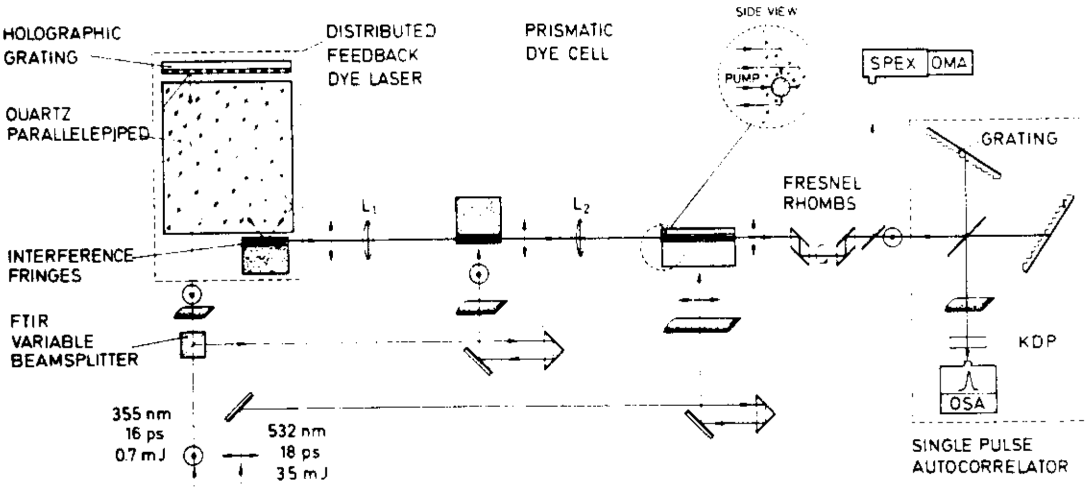

Fig. 10. Experimental arrangement of a DFDL system pumped by the second and third harmonics of a mode-locked Nd:YAG laser system (not shown). Synchronization of the DFDL and the two dye amplifiers is accomplished by optical delay lines. Autocorrelation functions and spectra of generated pulses are registered simultaneously. From [39].

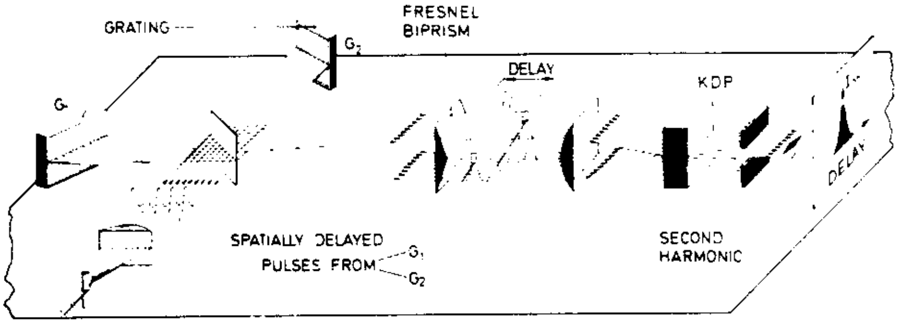

Fig. 11. Setup used for recording the complete background-free autocorrelation function of a single laser pulse. From [39].

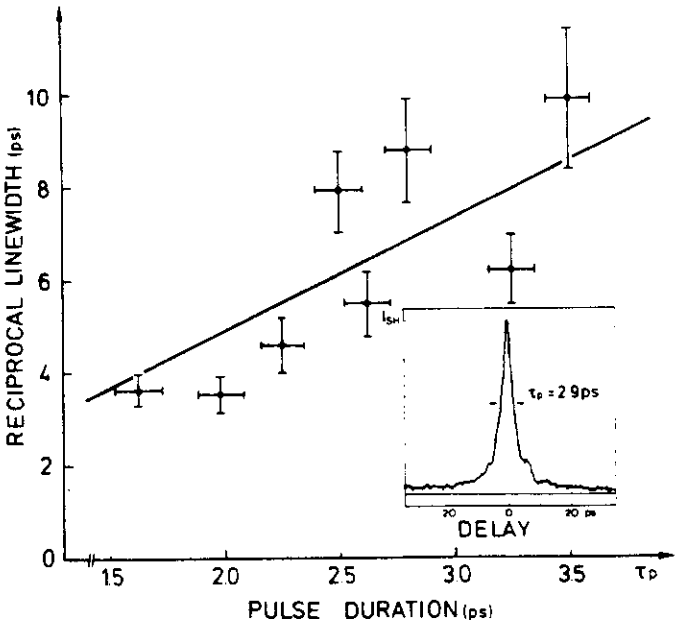

Fig. 12. Relationship between reciprocal linewidth and pulse duration as determined from the autocorrelation functions (cf. inset) of DFDL pulses. The straight line corresponds to bandwidth-limited pulses. From [39].

tained from simultaneously measured correlation functions. The straight line corresponds to a time-bandwidth product $\Delta\nu \cdot \Delta t = 0.41$ (cf. [42]). In plotting Fig. 12 it was assumed for both calculation of the straight line and conversion of measured autocorrelation delays to pulse durations that the DFDL pulses have pulse shapes identical to the calculated ones of [35]. It follows that the DFDL pulses are transform limited. The same is true for DFDL pulses of longer duration.

While earlier papers from other laboratories reported DFDL data which indicated time-bandwidth products being far from the transform limit [45], [46], observation of transform-limited DFDL pulses has been confirmed in recent work [21].

The shortest pulses which have been obtained with picosecond Nd:YAG laser-pumped DFDL's are about 1.5 ps long [39],]29]. Obviously, the relative reduction of the DFDL pulsewidth as compared to the width of the pump pulse is not as big as might be expected according to the rate equation model. One finds also a discrepancy in the dependence of the pulse duration on DFDL length in this limiting case, when the DFDL pulse duration is

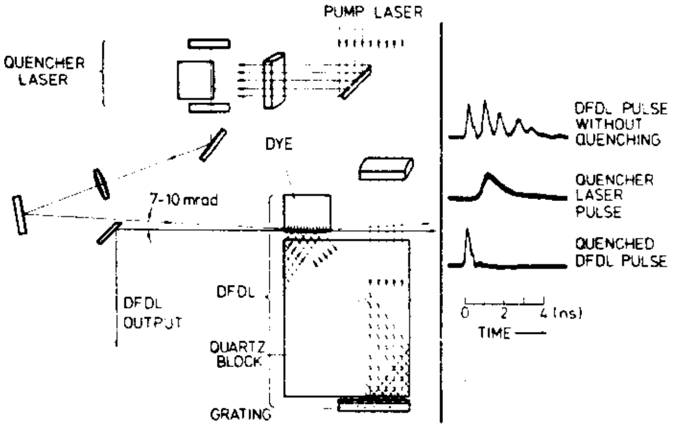

Fig. 13. Experimental arrangement of a DFDL with quencher laser. The various pulse shapes are shown on the right-hand side. From [23].

comparable to the transit time of light through the active medium.

Recently, the time- and space-dependent semiclassical theory of the DFDL has been published [43], [44]. For pulse durations longer than the transit time, it fully confirms the validity of the rate equation model in which the spatial dependences are not considered [35]-[37]. For shorter pulses, for which the validity of the rate equations breaks down, the semiclassical theory predicts that the shortest DFDL pulses will be somewhat shorter than the transit time.

VII. Generation of Stable Single Picosecond Pulses and Pulse Amplification

The method which we have discussed so far achieves generation of single picosecond pulses just by control of pump power. An increase of pump energy results in multiple pulse generation. Furthermore, if the energy of the pump pulses is not constant within narrow limits, then DFDL pulse energy as well as pulse duration will fluctuate. There exists, however, a method to avoid such fluctuations, which we will discuss now.

The principle of the method is depicted in Fig. 13 [47]. One half of the pump beam is used to pump the DFDL, while the other half pumps a second dye laser acting as a "quencher laser." The output of the quencher laser is injected into the DFDL under a small angle via an optical delay. This delay has to be adjusted in such a way that the quenching pulse arrives in the DFDL just after the first DFDL pulse has been generated. The quenching pulse, being also amplified in the DFDL, uses up the stored energy, thus reducing the gain of the DFDL. If the quenching pulse was of sufficient energy, then the gain of the DFDL will remain below threshold for the remaining duration of the pump pulse. In this way only a single DFDL pulse is produced, while all the later pulses are quenched. As can be seen in Fig. 13, the DFDL pulse can be separated spatially from the quenching pulse in a simple way. The shortest pulse that has been obtained using this scheme had a width of 17 ps. The laser dye was BiBuQ (4,4‴-*bis*[2-butyloctyloxy]-*p*-quaterphenyl) and the pumped region was 1.5 mm long.

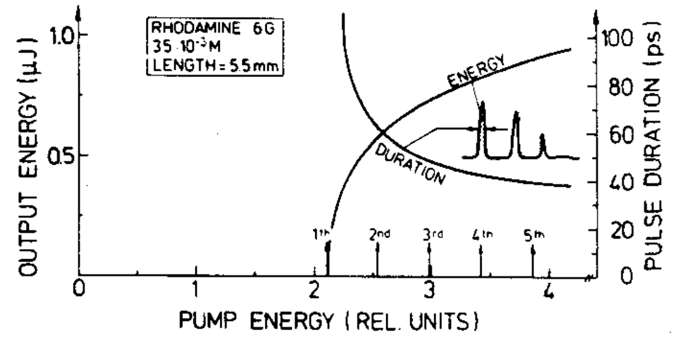

Fig. 14. Computed pulse duration and energy of the first DFDL pulse as a function of pump energy. Thresholds of DFDL pulses are also indicated. From [23].

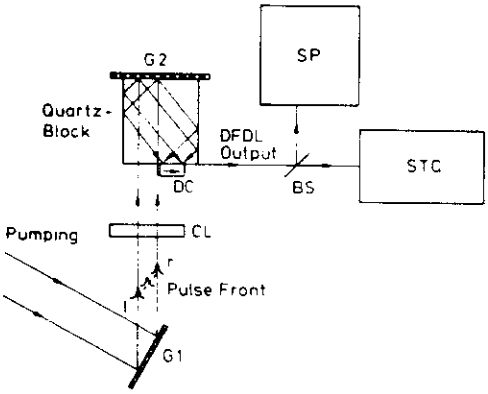

Fig. 15. Scheme of a traveling-wave-pumped DFDL. *G1* = holographic diffraction grating used to produce the skewed pulse front. *G2* = holographic diffraction grating serving as beamsplitter in the DFDL, CL = cylindrical lens, DC = laser dye cell, BS = beamsplitting mirror, SP = spectrograph, and STC = 1.7 ps streak camera system. The angle between the pulse front and the normal to the wave vector is called γ in the text. From [52].

Fig. 14 shows that DFDL output energy is stabilized against fluctuations of pump energy towards higher excitation because of the decreasing slope of the energy characteristic.

Peak powers which can be achieved with this method are in the range of 20 to 40 kW for single pulses, and the beam divergence is about 10 mrad. However, these values can be improved considerably by the use of additional amplifier stages, which can be excited by the same pump laser in a fashion similar to that shown in Fig. 10 (cf. also [47], [48]).

VIII. The Step into the Subpicosecond Region

As we have seen in Section VI, the shortest pulse duration which can be generated is limited by the transit time of light in the DFDL. In order to accomplish a further reduction of pulsewidth a new pumping scheme, namely, *traveling-wave excitation*, has to be applied [49]-[51]. The principle of this method is the formation of a skewed pulse front with the aid of a dispersive element [41].

Fig. 15 shows how this excitation scheme can be ap-

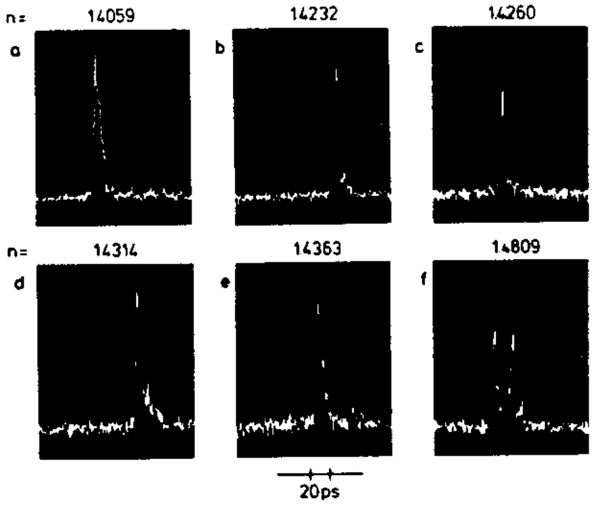

Fig. 16. Variation of pulse duration of a traveling-wave-pumped DFDL as a function of the refractive index of the dye solution. Pulse recording by 1.7 ps streak camera system. Time base: 0.7 ps per OMA channel. Laser medium: 5 mmol/L Rhodamine 6G in a mixture of DMSO and methanol. Refractive indexes (indicated on top) adjusted by variation of the mixing ratio of solvents. From [52].

plied to a DFDL [52]. In this case a grating is used to produce the delay. The pulse front is entering the quartz block DFDL under an angle γ with the normal to the wave vector, creating an induced interference grating in the dye cell. While the position of the interference maxima and minima is stationary, their intensity profile is sweeping from left to right with velocity

$$v = c/\tan \gamma. \quad (11)$$

Traveling-wave pumping is achieved when $v = c/n_L$ where n_L is the refractive index of the dye solution. This can be achieved by either rotating the delay grating ($G1$) or by adjusting the refractive index. Notice, however, that a variation of n_L also tunes the laser wavelength according to (1).

DFDL pulses which have been generated by this method are shown in Fig. 16 [52]. One can see that upon variation of the refractive index of the dye solution, pulse duration is going through a minimum. The shortest pulse is produced when the condition $n_L = \tan \gamma$ is satisfied [cf. Fig. 16(c)]. The streak camera system used for these recordings has a temporal resolution of 1.7 ps, which is insufficient to resolve the shortest pulse. One can estimate a maximum duration of 1 ps [52], but it is likely that the pulses are even shorter if one considers spectral width also, which amounts to 4 Å.

Besides this reduction of pulsewidth, traveling-wave excitation has an additional desirable feature. Under the conditions of the experiment shown in Fig. 16(a) a *series* of pulses would have been generated without traveling-wave excitation because pump power was about 20 times above the threshold for single pulses. As one observes, however, only a *single* pulse is produced when the traveling-wave condition is fulfilled exactly. Thus, traveling-wave pumping allows high pumping powers without need for an extra single-pulse selecting device.

IX. APPLICATIONS

Several papers describing DFDL-dye-amplifier systems suitable for practical applications have appeared recently [47], [53]. Initial charge separation in bacteriorhodopsin occurring with a time constant of about 30 ps was measured with the aid of a DFDL producing single 55 ps pulses of 50 μJ energy at 10 Hz [54]. Picosecond gain dynamics of a KrF-excimer laser have been studied using very-high-power DFDL pulses of less than 10 ps at 248.5 nm [55]. The laser system described in [47] was employed to produce pulses of 0.5 mJ at 620 and 498 nm, respectively, which were used to demonstrate the capabilities of surface-enhanced SHG for a new autocorrelation technique. Autocorrelation functions and streak camera measurements gave consistent pulse durations of 2.5 ps (620 nm) and 6.0 ps (498 nm) [56]. The relaxation of Ag centers in single crystals of KI could be analyzed following optical excitation by 2 ps pulses of 10 mJ at 308 nm which were generated by a DFDL-amplifier system [57]. A DFDL producing transit-time-limited pulses of 1.5 ps at 570 nm when pumped by the third harmonic of a mode-locked Nd:YAG laser has been used to excite Scheibe aggregates of pseudoisocyanine in solution and in molecular monolayers in order to measure the temperature dependence of fluorescence lifetime. In the same investigation a continuously wavelength-tunable DFDL was employed to obtain fluorescence excitation spectra of these dye aggregates [29].

X. CONCLUSION

In this paper, operational principles and experimental arrangements of distributed feedback dye lasers have been reviewed. After presenting various methods of wavelength tuning, we have discussed theoretical approaches for modeling the temporal and energetic characteristics and compared them to experimental results. It has been shown that distributed feedback dye lasers are capable of producing ultrashort light pulses tunable over the full spectral range for which laser dyes exist. It appears that pulse durations well in the subpicosecond range can be realized. A number of papers describing applications of picosecond DFDL's to various spectroscopic problems provide proof that these devices are potent research tools despite their inherent simplicity.

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300 Femtosecond Pulses at 497 Nanometer Generated by an Excimer Laser Pumped Cascade of Distributed Feedback Dye Lasers

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Abstract. The setup is a cascade of 3 lasers: A competing cavity dye laser pumped by a XeCl excimer laser, followed by two distributed feedback dye lasers. The typical durations of the pulses from the lasers are 100 ps, 5 ps, and 300 fs, respectively. The output pulses at 497 nm are amplified up to 500 MW. The shortest pulse duration obtained was 198 fs.

PACS: 42.60, 42.55 M

High-power subpicosecond excimer laser pulses are of great interest for use in various fields of nonlinear optics, plasma physics and X-ray research. Such pulses are generated by multipass amplification of short seed pulses in excimer amplifiers [1–10]. The seed pulses are obtained by frequency doubling or mixing of pulses from mode-locked lasers [1–5, 12], from distributed feedback dye lasers [6–8], or by stimulated Brillouin scattering in liquids [11].

In this paper we describe a XeCl laser pumped cascade distributed feedback dye laser (DFDL) system generating 300 fs long seed pulses for a KrF excimer amplifier.

1. Experimental Arrangement and Results

The cascaded setup is pumped with 100 mJ from a XeCl excimer laser (Lambda Physik EMG 1003i). The system consists of 3 lasers: a Competing Cavity short-pulse Dye Laser (CCDL) and two Distributed Feedback Dye Lasers: $DFDL_1$ and $DFDL_2$ (Fig. 1). The CCDL with two amplifier stages generates 100 ps long pulses at 340 nm, with pulse energy of 5 μJ and linewidth of 0.5 nm. The principles of operation of CCDL's are given in [7, 8, 13–15]. The operation parameters and the technical details of the CCDL used in this experiments can be found in [26]. The cylindrical lenses CL_1 and CL_2 ($f = 66$ mm and 43 mm) are used to focus the beam of the CCDL onto $DFDL_1$.

The amplitude-phase grating necessary for operation of $DFDL_1$ is created by the interference of the pumping beams. The two interfering pumping beams are formed by the holographic grating (G_1) and a silica block (10 mm × 6.45 mm). The properties of such pumping arrangement were described in detail in [16–20]. The grating, the silica block, and the dye cell are glued together with a uv transparent adhesive (Epotek-500). The grating line density (3600 l/mm) and the refractive index of the dye solution ensured DFDL operation at $\lambda_1 = d \cdot n_s = 398$ nm wavelength [18]. The active medium was a $3 \cdot 10^{-3}$ mol/l solution of PBBO in dioxane. The excited volume of $DFDL_1$ had dimensions of 0.5 mm × 0.02 mm × 0.025 mm. The latter value is determined by the penetration depth of the pumping beam into the dye solution. The pulse duration of $DFDL_1$ measured by a streak camera (Hamamatsu C1587) was 3–4 ps, which broadened to 5 ps when amplified up to 5 μJ in the 3 stage amplifier. The amplifiers used $1.5 \cdot 10^{-3}$, $2 \cdot 10^{-4}$, $1 \cdot 10^{-4}$ mol/l solutions of PBBO in cyclohexane, respectively. The spectral width of the amplified radiation was about 0.1 nm and the shot-to-shot wavelength fluctuation

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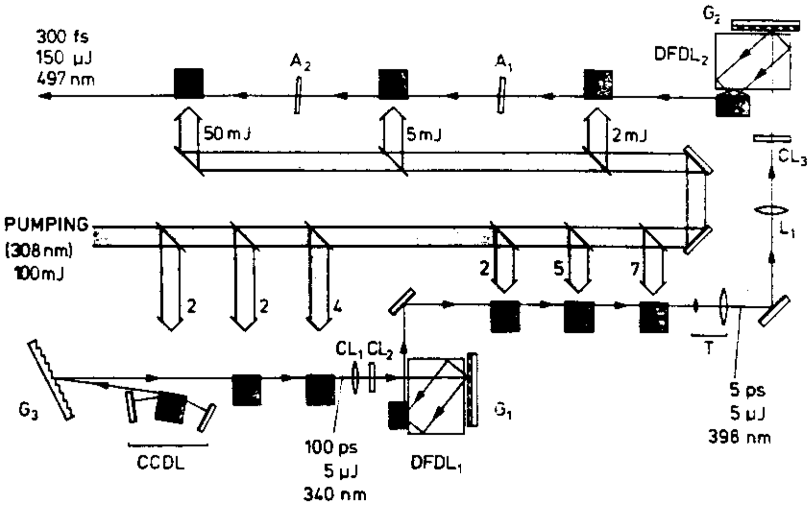

Fig. 1. Experimental arrangement. CCDL: competing cavity short pulse dye laser; DFDL$_1$ and DFDL$_2$: cascade distributed feedback dye lasers. The numbers indicate the approximated pump energies in mJ

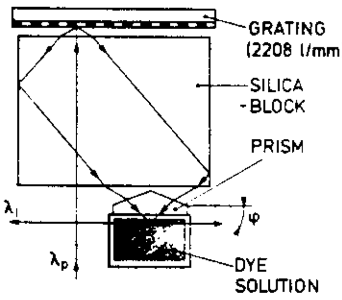

Fig. 2. Optical scheme of DFDL$_2$

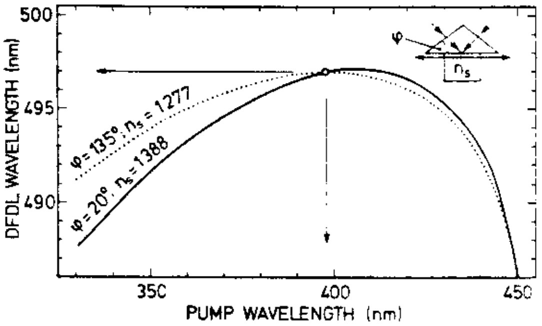

Fig. 3. Wavelength of DFDL$_2$ as a function of the pumping wavelength. At 398 nm the solid curve has a slope of 0.0334. This means that the arrangement is nearly achromatic

was of the same order. The long-term wavelength stability (determined by the temperature dependence of the refractive index of the dye solvent) was about 0.4 nm/K.

The optical arrangement of DFDL$_2$ is shown in Fig. 2. The size of the silica block is 52 mm × 40 mm. The lasing wavelength of DFDL$_2$ λ_1 can be calculated as

$$\lambda_1=\frac{n_s}{n_p}\frac{\lambda_p}{\left[\sin\quad \varphi+\arcsin\dfrac{\sin\left(-\varphi+\arcsin\dfrac{\lambda_p}{d}\right)}{n_p}\right]},\tag{1}$$

where λ_p is the pump wavelength, n_p and n_s are the refractive indices of the prism and of the solvent, respectively, d is the line separation of grating G$_2$, and φ is the angle of the prism, as shown in Fig. 2.

Figure 3 shows the calculated lasing wavelength of DFDL$_2$ as a function of pump wavelength for $n_s=1.338$ (73% hexafluoroisopropanol and 23% benzylalcohol), $\varphi=20°$ (solid line) and $n_s=1.272$ (hexafluoroisopropanol), $\varphi=13.5°$ (dotted line). The grating line density was 2208 l/mm. n_p was calculated using the polynomial given for suprasil [25]. The purpose of the silica prism on the surface of the dye cell is to shift the DFDL wavelength into the blue spectral range [16]. (Using a 2208 l/mm grating and a refractive index of 1.272 without the prism, the DFDL$_2$ would operate at 676 nm).

It is important to notice, that the dotted curve has a maximum at $\lambda_p=398$ nm. It means that around $\lambda_p=398$ nm the DFDL wavelength is independent of the pump wavelength, i.e. each spectral component of the pump beam creates an interference pattern with the same period. Such property of the pump arrangement is called achromatism [16, 19–24].

As a measure of achromatism we may define a stabilization factor

$$S=\frac{\lambda_1}{\lambda_p}\frac{\Delta\lambda_p}{\Delta\lambda_1},\tag{2}$$

where $\Delta\lambda_1$ is the wavelength shift of DFDL$_2$ caused by the shift of the pump wavelength by $\Delta\lambda_p$.

Instead of a prism with the optimal angle of $\varphi=13.5°$ which would give $S=\infty$, a prism with an angle $\varphi = 20°$ was available for our present experiment. From the solid curve in Fig. 3 corresponding to $\varphi = 20°$ we obtain $S = 37$ at $\lambda_p = 398$ nm.

The stabilization factor of 37 is large enough to compensate for possible chirp of the pump wavelength and eliminates the unwanted influence of shot-to-shot and long-term wavelength change of the pump laser.

Instead of the arrangement shown in Fig. 2, the usual arrangement (i.e., rectangular silica block, without the prism on the surface of the DFDL dye cell [16]) could also have been used. In this case, a 3050 l/mm grating and a solvent refractive index of $n_s = 1.516$ would have resulted in a DFDL operation at 497 nm [16]. However, for a pump wavelength of 398 nm diffraction at normal incidence on a 3050 l/mm grating will occur only if an immersion liquid is used between the grating and the silica block. We found that the arrangement incorporating the 20° prism is more convenient in practice than the one with immersion.

The spherical lens L_1 ($f = 500$ mm) and the cylindrical lens CL_3 ($f = 220$ mm) were used to focus the pump beam onto the dye cell of $DFDL_2$ into a narrow line. In order to obtain a small size of the pumped volume, a telescope T with a magnification of 10 was used in front of L_1, CL_3. The length of the second DFDL structure should be chosen carefully. The shortest pulse duration is limited to about one half of the transit time of the light through the DFDL structure [30]. Taking into consideration the refractive index of the solution for 200 fs long pulses, this means that the length of the DFDL should be shorter than 90 µm. On the other hand, short DFDLs require high pump power densities to reach threshold. In this case the required pump power density might exceed the saturation power density and the absorption of the dye solution could be bleached. This would lead to an unwanted degradation of the quality of the interference fringes or could even prevent the operation of the DFDL [29]. The most stable operation was obtained with a 50 µm long and 20 µm wide pumped volume.

The $DFDL_2$ used a solution of Coumarin 307 in 73% hexafluoropropanal and 27% benzylalcohol mixture. This mixture has the necessary refractive index ($n_s = 1.338$) and has high solubility for the dye. The optimal dye concentration was found to be 0.1–0.05 mol/l. At lower concentrations, the threshold could not be reached even with very strong pumping. At higher concentrations, the small penetration depth of the pump beam into the solution increases the diffraction losses. Moreover, at high concentrations strong quenching of the fluorescence occurs. The threshold pump energy (falling onto the dye cell) was about 0.2 µJ. The most stable operation was observed when the pump energy was about 3 times the threshold value. The pump intensity was controlled by an attenuator and by blocking part of the pump beam of the last amplifier in front of the telescope T.

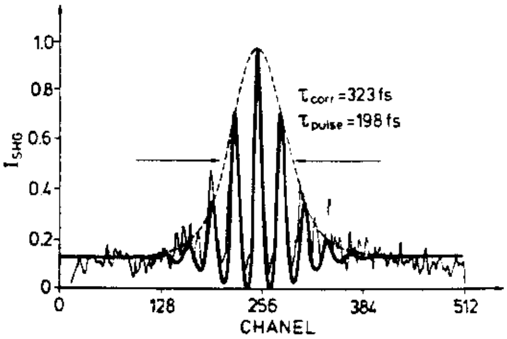

Fig. 4. Phase sensitive intensity autocorrelation of a pulse. (Heavy line: calculated, broken line: envelope, solid line: measurement). The 323 fs autocorrelation corresponds to 198 fs pulse duration assuming a sech^2 pulse shape

Except for the excimer laser which was unpolarized, all beams were polarized with vertical electric field.

The output beam of $DFDL_2$ was amplified in a 3 stage amplifier chain up to 150 µJ. The amplifiers used $1 \cdot 10^{-2}$, $5 \cdot 10^{-3}$, $1.3 \cdot 10^{-3}$ mol/l solutions of Coumarin 307 in ethanol. The divergence of the output beam measured 20 m behind the laser was 0.5 mrad. The pump energy was distributed between the amplifiers by beam splitters and by spatial division of the pump beam. No special efforts were taken to optimize the output energy by a more suitable distribution of the pump energy between the amplifiers and a better choice of dye concentrations in the amplifiers. The saturable absorbers A_1 and A_2 (DASBTI [28], small signal transmission 1%) are used to suppress the amplified spontaneous emission in the amplifier chain. No significant spectral changes of the pulses were observed during amplification.

The duration of the pulses was measured with a new single-shot phase-sensitive autocorrelator [27]. The autocorrelation width of the pulses shown in Fig. 4 is 323 fs, which corresponds to 198 fs pulse duration when a sech^2 pulse shape is assumed. [It can be calculated that the full width of the envelope of the autocorrelation curve at the level of $(1 + 7/2)/8$ of the peak intensity is related to the pulse duration by a factor of 1.63. The indicated level corresponds to the middle between the peak and the background of the autocorrelation curve, having the theoretical 8 : 1 peak to background ratio.] These pulses are somewhat shorter than reported before [32].

The typical pulse duration of 300 fs is 17 times shorter than the duration of the pump pulse. This pulse shortening factor is smaller than the 50–100 fold shortening which is predicted by the theory for nanosecond and subnanosecond pump pulse durations

[18, 31]. The reason for the smaller shortening factor is not clear. However, degradation of the pumping interference pattern caused by the bleaching of the absorption and by spatial migration of excitation by Förster-type energy transfer, reduction of fluorescence quantum yield at high concentrations, diffraction losses in $DFDL_2$ due to a small penetration depth of the pumping are effects which obviously do not favour the operation of $DFDL_2$. All these effects might act together resulting in a smaller pulse shortening ratio than predicted by the theory.

2. Conclusions

The cascade DFDL system presented in this paper can generate 300 fs long pulses with pulse energy of 150 µJ. The shortest pulse obtained was 198 fs which is the shortest pulse obtained from a distributed feedback dye laser so far. After frequency doubling these pulses can be used as seed pulses for a KrF excimer amplifier.

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