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GENERATION OF ULTRASHORT PULSES WITH DISTRIBUTED FEEDBACK DYE LASERS.

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GENERATION OF ULTRASHORT PULSES WITH DISTRIBUTED FEEDBACK DYE LASERS

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The conventional laser arrangement consists of three main elements:

(1) active medium, (2) optical resonator and (3) pump source.

Commonly the resonator is formed by two (or more) end mirrors providing the feedback necessary for oscillation.

In 1971 Koselnik and Shank proposed a different arransement in which the feedback mechanism is distributed throushout the sain medium and integrated with it. In this arransement the feedback mechanism is provided by Brassscatterins from a periodic spatial variation of the refractive index of the sain medium, or of the sain itself.

The term "Brass scatterins" or "Brass reflection" orisinates from the field of X-ray diffraction but the phenomenon is not restricted to this wavelength range. It can be observed as well with visible light or even with sound waves. We consider a 1 a m i n a t e d s r a t i n s which consists of equidistant, partially transmitting reflective layers. A coherent light beam is coming from the left side and is partially reflected from each layer. In order to determine the directions into which maximum intensity is reflected, we can estimate the path difference for two rays. Maxima (or minima) in the intensity of the reflected (or transmitted) wave occur when the

condition 2.1. n. sin 2 = m. \

is satisfied. This relationship is called B r a 9 9 $^{\prime}$ s l a w. Were m = interference order.

For normal incidence (= 98°) we have:

2n1 = m 7

In order to make a DFBL we have to induce a periodic spatial variation of the refractive index n or the sain constant of the laser medium: n(2) = n + n . COS K2

0((7) = 0(+ 0/4 · CO) KZ

z is measured along the optic axis and $k=2\pi/\Lambda$

 Λ is the period or frinse spacing of the spatial modulation, and M_4 and M_4 are its amplitudes. A DFDL structure of this kind will then oscillate at a wavelensth $\lambda_{\rm L}$ determined by the relationship:

λ, = 2 n 1

There are several ways to prepare a laser medium of this sort. The first experiments of Koselnik and Shank used a selatin film on a slass substrate. The selatin was dichromated and exposed to the interference pattern produced by two coherent UV beams of a helium-cadmium laser. The fringe spacins in the selatin was

about 1 = 0.3 hm

After exposure the selatin was developed (using techniques well-known in holography) resulting in a spatial modulation of the substrate density. The developed selatin was then immersed in a solution of r h o d a m i n e 6 G to make the dye penetrate into the porous selatin layer. After drying the resulting DFDL structure was pumped with UV radiation from a nitrogen laser. At pump power densities $> 40^6 \, \text{W/cm}^2$ laser oscillation was observed at a wavelength $\lambda_{\perp} 630 \, \text{nm}$. The linewidth was $4\lambda < 0.5 \, \text{A}$ (instrument limited).

- 3 -

When a uniform selatin layer dyed with rhodamine 6G was rumped in the same way, stimulated emission was observed at $\lambda = 590$ nm with a line width of $\Delta\lambda \approx 50$ Å. Obviously, in the first case there is considerable line narrowing due to the distributed feedback effect.

The observed behavior is due to the fact that two counterrunnins waves are propagating in the periodic structure which are coupled to each other (Fig.1). The electric field can be expressed in the form:

where z is the direction of propagation, and R and S are complex amplitudes.

The boundary conditions are: R(-L/2) = S(L/2) = 0

At the endfaces of the device a wave starts with zero amplitude. It receives its initial energy through feedback from the other wave. The waves grow in the presence of gain and feed energy into each other due to the spatial modulation of n or &.

You can als look at this in the followins way: the two counterpropasatins waves of equal wavelength superimpose in the region
of the interference pattern to form a standing wave which has
its maxima coincident with the interference maxima. There the
inversion of the active medium has a high value and the sain is
correspondingly high. The knots of the standing wave are, on the
other hand, coincident with the interference minima. A standing
wave of different wavelength would not be amplified in an optimal way
because its maxima would not always coincide with the regions of
highest inversion. Thus, a monochromatic wave is produced
in this structure.

A DFBL structure consisting of a laser dye embedded in some polymeric matrix has the great disadvantage that it does not function for a very long period of time because all organic dyes are more or less quickly degraded by photochemical processes and in the risid matrix they cannot be replenished. It is rather much more practical to use instead a dye in solution as the laser medium and to induce the periodic gain and refractive index variation by forming an interference pattern of the pump light on the surface of the dye cell (Fig.2).

The separation Λ of two interference maxima in this case is:

$$\Lambda = \frac{\lambda \rho}{2 \cdot s = \Theta}$$

Ap = rumr wavelensth

If we combine this with the Brass condition for feedback: $\lambda_L = 2n\Lambda$ we obtain:

 $\lambda_L = \frac{n \cdot \lambda p}{\sin \Theta}$

The DFDL output wavelensth depends on the refractive index of the dye solution n, the angle of incidence Θ of the pump light and the pumping wavelensth λ_{μ} .

Since for any chosen numering source Apaconst., one could either vary n or \bigcirc for tuning the wavelength of the DFDL. But before we have a closer look at tuning let us consider still another arrangement.

When you look at the next figure (Fig.3;left), you will observe that rays from different parts of the pump beam are not combining with themselves to produce the interference pattern on the dye cell. That means low spatial coherence of the pump beam will result in bad fringe visibility V:

 $V = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}}$

In order to improve this situation (which is necessary particularly in the case of nitrogen laser pumping) Zs. Bor (1979) proposed a scheme of the following kind (Fig. 3, right), in which a holographic grating is used in place of the beam splitter.

From the grating equation we have for & (Fis.4):

d = spacing of the grating (mm/groove)

m = 1 (first order of diffraction assumed)

For the separation of the interference fringes we had obtained earlier:

Because the fringe separation turns out to be independent on the pump wavelength in this case each spectral component of the pump N laser creates an interference pattern with the same Λ . Therefore, we call this an achromatic arrangement.

Additionally, if the following seometric relationship is satisfied:

$$\frac{1}{\sqrt{1 + \frac{1}{2}}} \frac{1}{\sqrt{1 + \frac{1}{2}}} \frac{1}{\sqrt{$$

for each point of the dye cell surface the two interfering beams have been diffracted from the same point on the grating. Thus it is possible with this arrangement to obtain good visibility of the

fringes even with a nitrogen laser which has low spatial coherence.

Note that neither of this two properties could have been obtained with the arrangement using a beam splitter in place of the grating!

The grating arrangement with parallel mirrors can be set up in a particularly compact form, which has the advantages of simplicity, stability and ease of alignment (Fig. 5). The mirrors are replaced by total internal reflections from the quartz-air interfaces of the quartz block. The next three slides show this device in operation.

I will now turn to the methods which can be used to tune the wavelensth of the DFDL:

- (1) Variation of the refractive index n of the dre solution:
- (1.1) Various solvents with different indices n or mixtures of solvents (Fig. 6 and 7).
- (1.2) Pressure dependence of index n (Fig. 8):
 The pressure dependence of n is contained in the Lorentz-Lorentz formula for the molar refraction:

$$P_{M} = \frac{n^{2}-1}{n^{2}+2} \cdot \frac{H}{S}$$
, $S = S(P)$, $Se = \frac{1}{S}(\frac{\partial S}{\partial P})_{T=coust}$

Due to the low compressibility of liquids the tuning range is fairly small. For Pmax ~100 bar the wavelength shift is only $\Delta\lambda_L\sim$ 10 - 20 Å

(2) Another way to tune the DFDL is by changing the angle of incidence which can be achieved by rotating the two mirrors in opposite directions by an angle of about a vertical axis (Fig.9). In this way the fringe separation is changed:

$$\Lambda = \frac{\lambda p}{2 \cdot \sin(\theta \pm \delta)}$$

Of course the achromatic property is lost in this case. The lasing wavelength as a function of d is given by:

On this basis we have recently constructed a DFDL which is continuously tunable by a computer (Fig. 18). The dye cell is kept stationary.

The rotating mirrors and the grating are on the translation table which assures that the interference pattern remains on the dye cell surface when the angle of incidence is changed.

The next figure (Fig.11) shows tuning curves obtained with four different dyes.

(3) It is even possible to operate the DFDL in the UV and blue spectral range where the synchronously mode-locked cw dye laser does not yet work (Fig. 12). For this purpose one has to use second order Brass reflection:

$$\lambda_L = \frac{2n\Lambda}{m}$$
, for $m=2$: $\lambda_L = n \cdot \Lambda$

for parallel mirrors:

$$\Lambda = \frac{d}{2}$$
, $\lambda_L = \frac{n \cdot d}{2}$

Let us now have a look at the temporal behavior of the DFDL. We write down the following set of rate equations:

1) Rate of population of the upper laser level:

$$n = n(t)$$
, $q = q(t)$, Ir = Ir(t) here: $p = refrective lu dex!!$

$$\frac{dn}{dt} = \frac{\text{Tp} \cdot \text{Sp}(N-n)}{T} - \frac{\text{Se} \cdot \text{Co}}{T} - \frac{n}{T}$$
Pump light absorption stimulated emission spontageous emission.

2) Photon flux: $T_{c} = T_{c(t)}$

$$\frac{dq}{dt} = \frac{(6e-6e)C_0}{7} \cdot n \cdot q - \frac{q}{T_c} + \frac{\Omega \cdot n}{T}$$
stimulated emission "resonator loss" amplified spont. emission

(ASE)

Absorption from the upper laser level (S1) to higher excited singlet states (S1 -> Sn) is an important loss process in dye lasers that are pumped by short laser pulses. Its contribution is taken into account by including $G_{\rm L}$, which is implicitely also contained in $G_{\rm L}$ (see below!).

The special features of distributed feedback are introduced with the term \mathcal{T}_{c} . In conventional lasers it describes the (constant) resonator losses. In our case a more general definition must be used since there is no external resonator in a DFDL:

The equivalent cavity decay time can be shown to be:

we consider only the sain modulation by rumpins, because it was found that the refractive index modulation makes a neslisible contribution. The amplitude of the spatial modulation of the sain is:

V = Visibility of fringes n = Augrase population of S1 So we obtain finally

$$T_c(t) = \frac{\eta \cdot L^3}{8c_0\pi^2} \cdot \left[n(t) \cdot (6e - 6a) \cdot V\right]^2$$

This system of coupled rate equations can be solved on a small disital computer using a Runge-Kutta procedure of fourth order.

The quantity which is usually most interesting is the output power of the DFDL: $Q \in Q(\Theta)$

Pour = 1/2. A.c. . 9(6) . L. Q. 6

 $a = 1/(N \cdot S_p)$ renetration depth of the pump light

Now we can compare the time courses of Ip, Tc and Pout (Fis.13). The diagram shows that Tc has a strongly non-linear time dependence which gives rise to relaxation oscillations of the output. Their number depends on the symping rate. As I will show, the pulses which are generated are of extremely short duration.

If Pout is plotted on a logarithmical scale against time one can see nicely that the intensity is changing over many orders of magnitude between the individual pulses (Fig. 14).

In order to find out whether the system of coupled rate equations is a sood model of the real DFDL we have compared results of the model computations with output pulses recorded with a streak camera system (Fig. 15). The agreement is indeed remarkably good.

A particular condition exists just below the threshold of the second pulse. Here we have a stable s in sle ultrashort pulse - a property of the DFDL which is very useful, and quite an advantage in comparison to mode-locked lasers where single pulse selection has to be employed using electrooptic methods.

One can now proceed to vary the various parameters of the equations and compare theoretical and experimental results quantitatively.

Some examples are shown in the following figures (Fig. 16, 17).

Instead of temporal relationships one can just as well compare the energies of the s i n s l e pulses (cf. definition given earlier!) (Fig. 18) with the computational results.

Since the fluorescence lifetime of the laser dye occurs in the rate equations one can try to change it by adding quenchers to the dye solution; e.s. potassium iodide KI (Fis.19). According to the Stern-Volmer law:

4 = fluorescence quantum yield without quencher

Q_= " " with quencher

To = fluorescence lifetime without quencher

ጂ = " with swencher

Kg = 68 l/mol = quenching constant

C = quencher concentration

Finally one can measure the output energy of the DFDL as a function of pump power and compare it with the computed results (Fig.28); again noting that the agreement is very good.

The single pulse condition is satisfied when the DFDL is pumped about 15% above its threshold. For the computer controlled DFDL, which I presented earlier in this lecture, we have measured the single pulse condition as a function of output wavelength (Fig.21). As a practical way to obtain single pulses we suggest the following procedure: First insert a neutral density filter with a transmission of 85% into the pump beam. Reduce the pump intensity until lasing

of the DFDL stors and then remove the filter. Under these conditions single pulses will be generated most of the time.

As the figures show the duration of the single pulses is about 50 - 100 ps when a nitrosen laser having a pulse duration of 3 ns is used as the sums source. For many applications it would be desirable to senerate pulses of shorter duration. If we look again at the rate equations, we see that they contain the time-dependent pumping term Ip(t). Solving them for pump pulses of shorter duration they predict a shortening of the output pulses. So we tried to construct rump lasers (Fig.22) which would produce shorter rulses than the ones senerated by the low pressure nitrosen laser used so far. The electrodes of the TEA-N2-lasers were connected to folded parallel plate Bluemlein lines which were switched by a hydrogen thyratron. A telescope (M = 9) inserted between oscillator and amplifier reduced the horizontal diversence of the TEA-laser to about 1.5 mrad. The dye bis-MSB was used as saturable absorber in the focal plane of the telescope. Repetition rate was 12 pps and 0.5 mJ were needed to rums the DFDL.

The relaxation oscillations obtained with the rate equation model are again observed experimentally. Upon lowering of the pump intensity one gets single pulses as before (Fig.23). So, the model is well suited to describe DFDL behavior even under the conditions of variable pulse duration.

The next slide summarizes our results (Fig. 24).

Each point is an average of 20 laser shots. The prediction of the rate equation model is obviously correct: Shorter pump pulses result in shorter output pulses!

From this diagram one would expect that the DFDL is capable of

producing pulses as short as a few picoseconds. It is, however, obvious that the ultimate limit of pulse duration will be about one half of the transit time of light through the DFDL. Therefore, we decreased the length of the DFDL to 2mm when we used the 8.7 ns pumping pulse. This figure (Fig.25) shows the measured pulse shape. The pulse has, indeed, a duration of only 6 ps (FWHM).

The energy of the single pulse was about 40 nJ, corresponding to a

The energy of the single pulse was about 40 nJ, corresponding to a peak power of about 7 kW.

Our results show that the DFDL rulses are about 50 times shorter than the rume rulses. So, in order to find out how short are the shortest rulses we can produce, we have used rulses of 16 ps duration from a mode-locked Nd:YAG laser for rumping the DFDL.

This figure presents an outline of our setup (Fig.26). The DFDL oscillator and the first amplifier stage are pumped with the third harmonic (353 nm) while the second amplifier stage is pumped with the second harmonic (536 nm) of the Nd:YAG laser. Since the pulses generated are too short to be measured with our streak camera; we have used a second order autocorrelation method to determine the pulse duration. The spectra of the pulses were measured simultaneously using a grating spectrograph.

We have plotted here (Fis.27) the reciprocals of the measured spectral widths (1/ ΔV) assinst the measured pulse durations. The experimental points lie quite well on a straight line drawn for $\Delta t \Delta V = 0.44$ which is the time-bandwidth product of transform-limited pulses having a near-Gaussian shape as given by our model.

These are obviously the shortest pulses one can senerate with a DFDL of the kind which we have used so far, their duration being limited by the transit time of light through the DFDL.

Before I proceed to a novel experimental scheme allowing to shorten the output pulses of a DFDL even further, I should like to discuss a method of producing picosecond pulses of h i s h e r power. As you have seen, the DFDL is capable of producing single pulses as long as one limits the pump power to a range which is below the threshold of the second pulse. It follows necessarily that the output energy cannot exceed a certain maximum value. Furthermore, output energy and pulse duration are subject to fluctuations if the pump laser is not working with constant output within narrow limits.

The next figure (Fig. 28) presents a way to elude this limitation.

About 50% of the pumping beam are used to excite the DFDL high above threshold so that multiple pulses are senerated. The remaining 50% are utilized to pump a second dye laser which is not a DFDL and produces a long pulse. This laser serves as a "quenching laser". The output of the quenching laser is injected into the DFDL under a small angle of 7-10 mrad via an optical delay line. The delay has to be adjusted in such a way that the quenchins pulse will arrive in the active region of the BFDL just after the first DFDL pulse has been formed. The quenchins pulse which is also amplified reduces the sain in the DFDL by using up the stored enersy. If the enersy of the suenchins rulse was sufficiently high it will suppress further DFDL output for the remaining duration of the rums rulse. Thus, even though the DFDL was rumped in the multiple pulse resime, only on e DFDL output pulse is senerated and all the other sulses are quenched. As you can see on the figure, the DFDL and the quenching pulses can be separated easily in space optically. The next figure (Fig.29) shows what is sained by using this method. The output energy of the single DFDL pulse can be increased by more than a factor of two and the pulse duration

can be decreased to less than half as compared to a DFDL without avenchins workins in the sinsle pulse resime. There is an additional advantage in usins this method. Because the slopes of the energy and the temporal characteristics are both decreasing for higher pump energies this will serve to stabilize the output power of the DFDL.

Peak power of pulses generated in this way was of the order of 36 kW. The beam divergence was about 16 mrad and a typical pulse duration was 17 ps at a wavelength of 386 nm using a nitrogen laser as pump source and BiBuQ as laser dye (Fig.36).

The principle of suenched resonator transients which I have just described is not limited to DFDLs but, rather, can be used with normal dye lasers as well. Here, it can be utilized for pulse tailoring in order to make the rather long output pulses of an excimer laser suitable for use as a DFDL pumping source. Examples for this kind of application can be found in the literature cited at the end of these lecture notes.

For the rest of this lecture I will return to the problem of producing the shortest pulses possible with a DFDL. For this purpose I have to introduce the concept of travelling wave pumping of a dye laser.

When we take an ultraviolet laser pulse of, say, 5 ps duration and spread it optically in one dimension with a cylindrical telescope (Fig.31) then this pulse will travel as a narrow patch of light which is $\Delta t_p \cdot C_o \approx 4.5$ mm thick. When we now put a diffraction grating into this light path it will diffract the beam into a new direction. Thereby, each partial beam suffers a certain delay with respect to its neighbor. If we illuminate N grooves of the

edse of the beam will amount to $N\cdot\lambda\rho$ because each should produce a delay of $\lambda\rho$.

As you can see, the patch of light is now tilted at an angle against the direction in which the beam proceeds. Using a Cylindrical lens set perpendicular to the beam we can now focus the Pulse onto a dye cell. The right edge of the beam will reach the cell first at its rear part and excite spontaneous fluorescence, while the left edge arrives with a time delay $\Delta t = N \cdot \lambda_P / c_0$ at the front part of the cell. In this way the point of highest excitation is moving with a speed $2 - L/\Delta t$ in the cell. L beins the lensth of the cell. A fraction of the initially excited spontaneous fluorescence moving in the direction of the travelling excitation will underso optimal amplification along the excited volume if its speed of propagation of matches the speed u of the travelling exciting wave, so it will pass through a volume element at the moment of maximum excitation. The speed of light in the dre solution is $v' = C_0/n$ when it has a refractive index n.

Thus, the two speeds will be matched, i.e. v = v', if $n = \frac{1}{L} v = \frac{1}{L} v^2$. This condition can be satisfied easily by proper choice of the grating and the angle of incidence of the pumping beam on it. The weak spontaneous emission starting at the end of the cell will emerge at the front end as a strong, well collimated beam. As one can show, the gain over the path length L can reach very high values of the order of $10^{\frac{10}{2}}$.

The time duration of the pulses senerated in this way depends on the rise time of the pump pulse and on the pumping intensity

(Fig. 32). We could even achieve a shortening of the output pulse to about one half the duration of the pump pulse, which was 12 ps in the example shown.

It is now possible to apply the same excitation scheme to a DFDL. The experimental arrangement is shown in the next slide (Fig. 33). An excimer-dye-laser-oscillator-amplifier system which is not shown serves to generate pump pulses of 5ps duration at 308 nm. A holographic grating (G1) of 2442 lines/mm working in first order of diffraction is used to produce a continuous spatial delay across the diffracted beam, analogous to the previous arrangement. This beam enters the quartz block DFDL, where it is selit into two beams corresponding to the +1 and -1 orders of the second holographic grating (G2) (2442 lines/mm). After total internal reflection at the sides of the quartz block the two beams are recombined on the surface of the dye cell where a diffraction pattern is formed such that the position of the fringes is stationary, while their envelope moves from left to right with a velocity 70 = Co/fg &

where c_{α} is the speed of light in vacuum and δ' is the angle between the pulse front and the normal to the wave-vector. In order to test the velocity matching condition

we varied the refractive index of the solvent mixture rather than changing of which would have required a total realignment of the DFDL. As the next slide (Fig. 34) shows, the width of the DFDL output pulse decreases at first from about 5 ps to some minimum width and then increases again to about 6 ps as the refractive index is varied by changing the proportion of

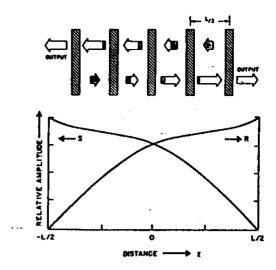
methanol to DMSO. The refractive index for which the shortest pulse is observed corresponds to the condition $\frac{1}{12}\frac{1}{12}=n$, while for $\frac{1}{12}\frac{1}{12}\neq n$ the pulses are broadened. The duration of the shortest pulse is not resolved by our streak camera. On account of the observed spectral width of about 4 Å we concluded, however, that the pulse duration is less than 1 ps.

The nume energy was kept constant during the whole experiment and corresponded to twenty times the threshold nume energy of the DFDL. A standing wave DFDL numbed by the same pulses showed multiple-pulsing already at a nume energy exceeding the DFDL threshold only by four times, while the travelling wave DFDL shows this behavior only if $\frac{1}{16}$ \frac

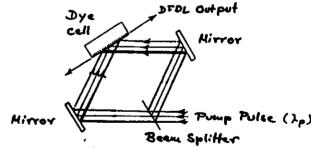
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KOGELNIK & SHANK 1972



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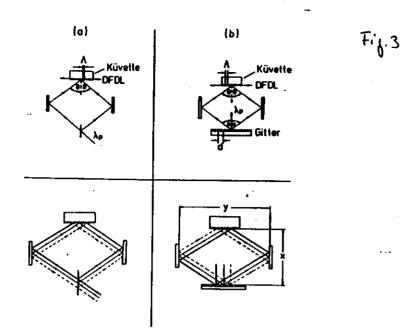
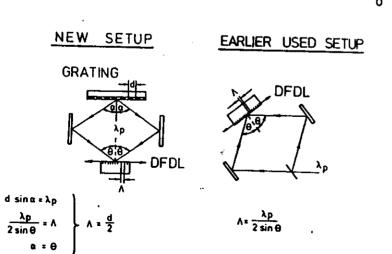
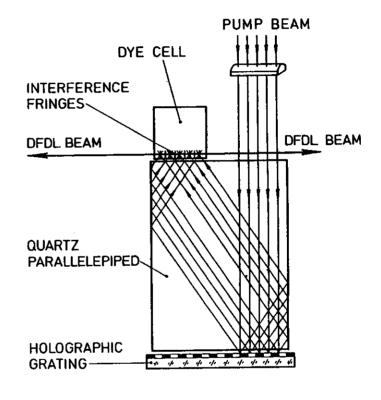
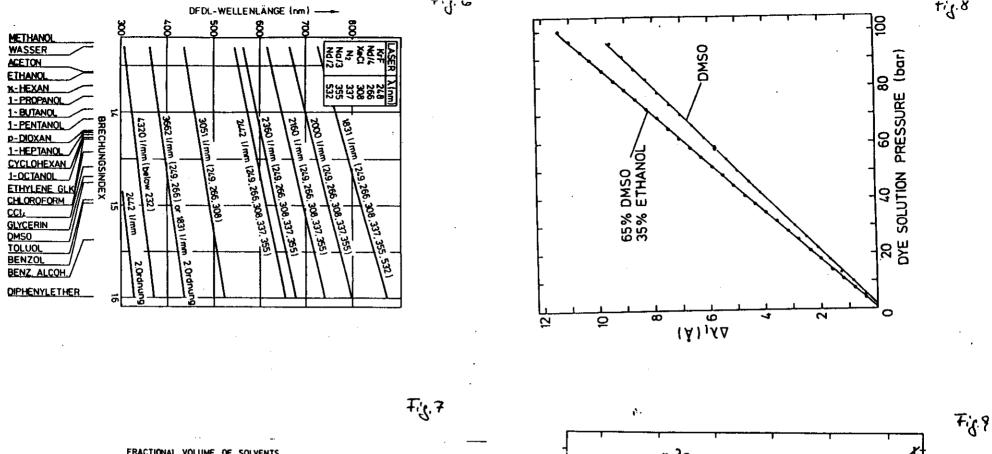
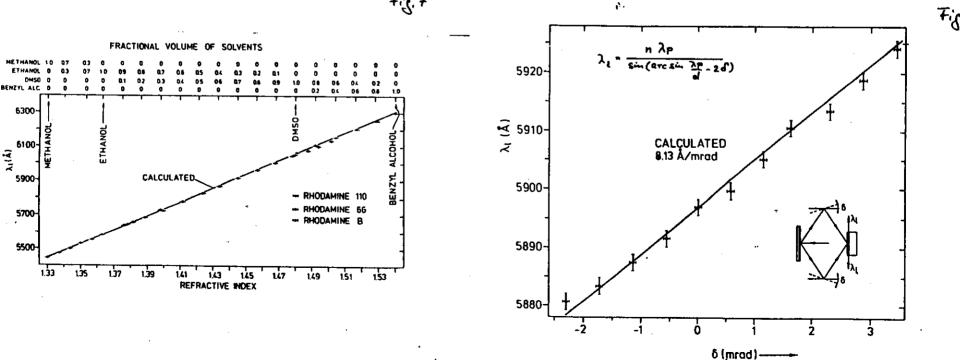


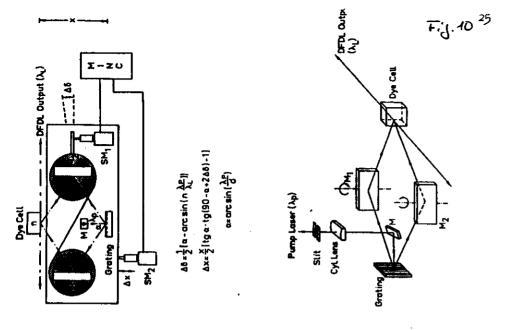
Fig. 4











Computer Controlled Distributed Feedback Dye Loser

Tuning curves of computer controlled DFDL (Single picosecond pulses)

Coumarin 153

Rhodamine 6G

Furan 6

DCM

x0,25

0.5

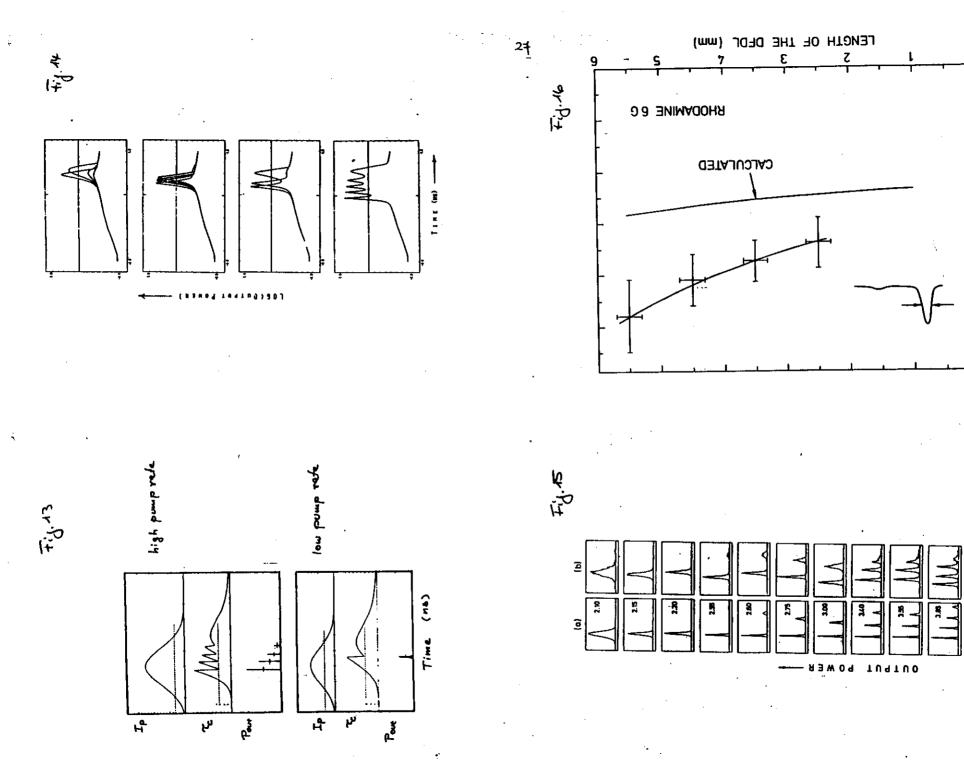
Laser Wavelength

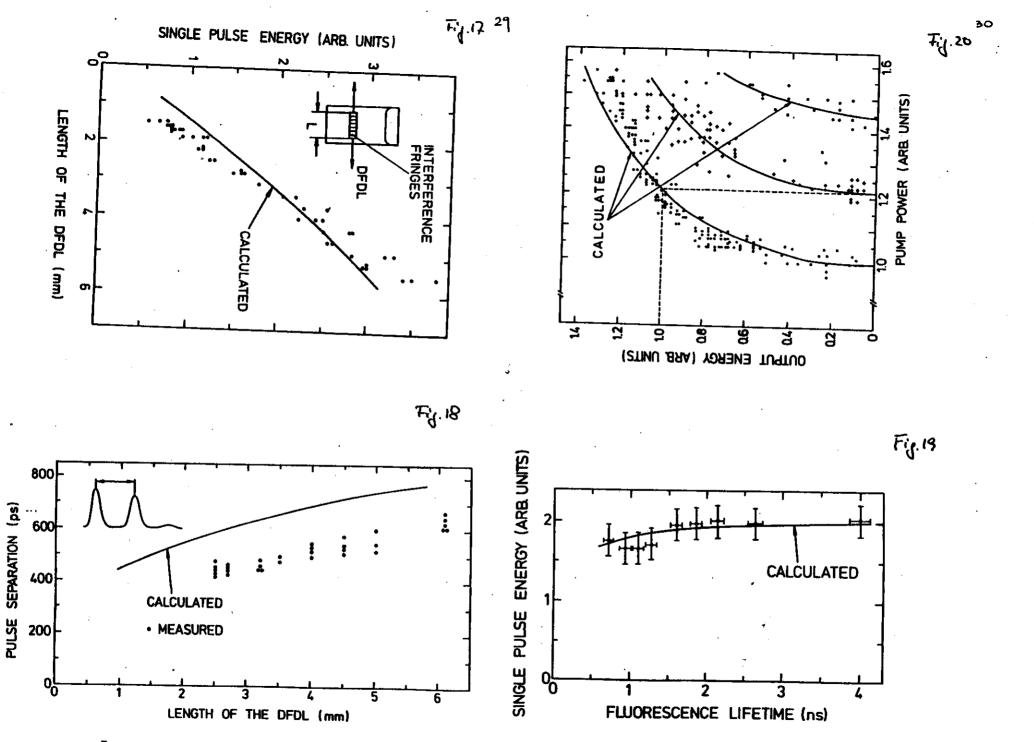
Fig	٦,	۲
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JG WAVELENGTH NM	363	383	388	404	413	421
SOLVENT	38% ETHANC. 62% METHANOL	80% DIOXANE 20% ETHANOL	100% DIOXANE	90% TOLUENE 10% ETHANOL	50% DIMETHYLSULFOXIDE 50% DIPHENYLETHER	25% DIOXANE 75% DIPHENY ETHER
CONCENTRATION MOL/L	5.10-3	1.10³	1,5 · 10 ⁻³	1.10³	SATURATED	1.10 ⁻³
DYE	PBD	BIBUQ	BIBUO	DPS	STILBEN 1	BIS-MSB

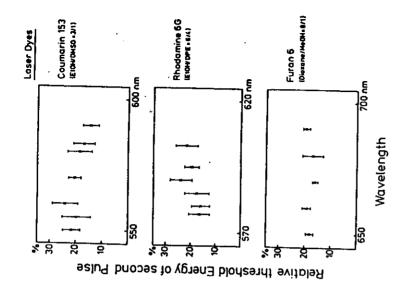
PULSE DURATION (ps)

-100r



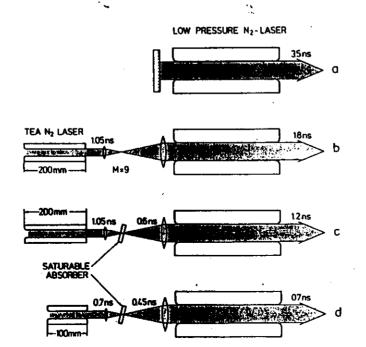


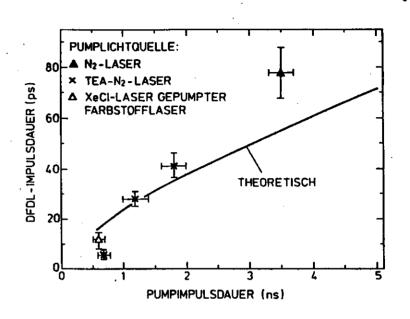
31

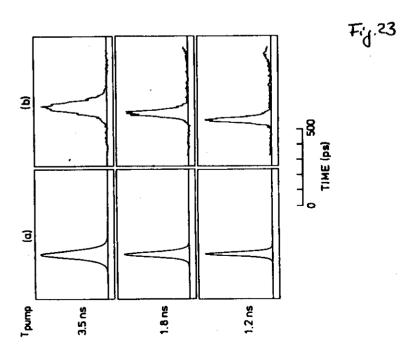


₹

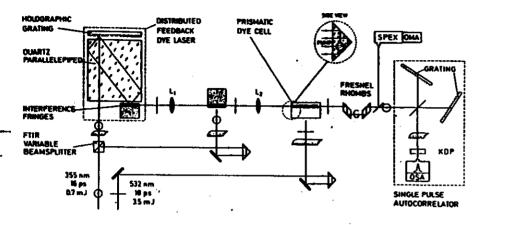
Fig. 22











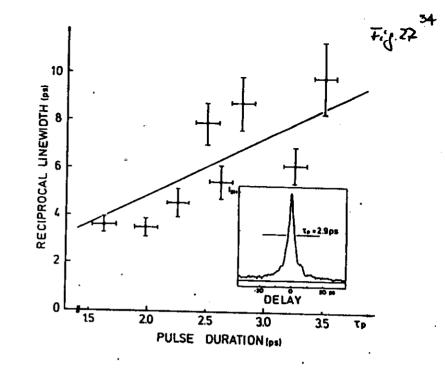
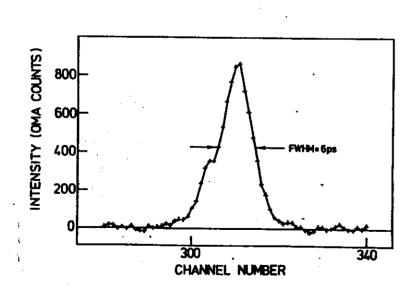
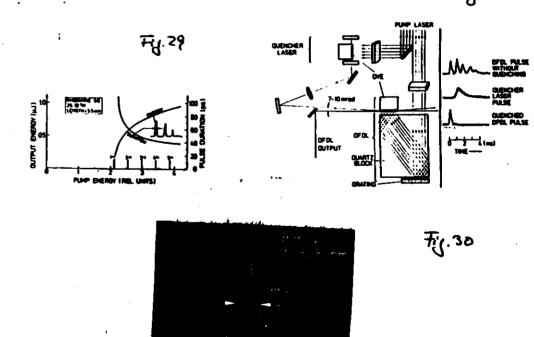
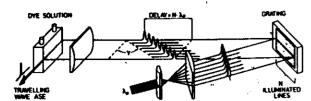


Fig. 28







Arrangement for transversal exci-

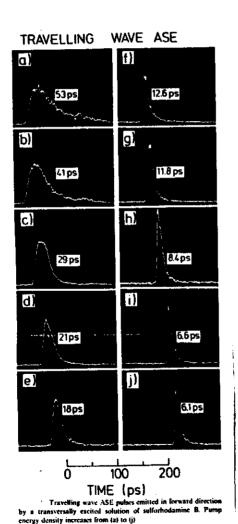
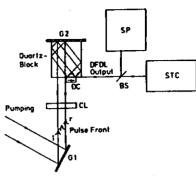
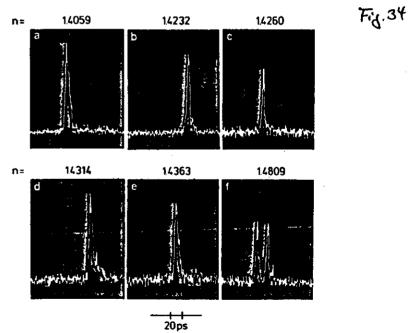


Fig. 32



Experimental arrangement of the travelling wave pumped DFDL. (GI; holographic diffraction grating for producing the delayed pulse front, G2: holographic diffraction grating serving as beamsplitter in the DFDL, CL: cylindrical lens, DC: dye cell, BS: beam splitter, SP: spectrographic recording system, STC: streak camera system).



Variation of the duration of the travelling-wave pumped DFDL output, as measured with a streak camera. Time base: 0.7 ps/channel, 5 × 10⁻³ mol/l shodamine 6G in a mixture of DMSO and methanol. The refractive index of the solution indicated above the traces was adjusted by varying the mixing ratio

