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Subpicosecond, Widely Tunable Distributed Feedback Dye Laser

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Abstract. A simple, achromatic, widely tunable distributed feedback dye laser arrangement is described. It makes use of a microscope objective, which images a transmission grating into the active medium. With this arrangement subpicosecond operation and broad tunability (400–760 nm) is reported.

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Distributed feedback dye lasers (DFDLs) [1] are convenient tools for generating ultrashort laser pulses in a wide spectral range [2]. For their operation they need a perfect, high-visibility interference pattern to be created in the dye solution. In the earliest DFDLs this was achieved by using a beam splitter in a laser beam to obtain two interfering partial beams, a method which necessitates a laser beam of high spatial and temporal coherence [3, 4]. In a later version a grating was used as a beam splitter [5, 6]. Here the period of the interference pattern can be made independent of the pump wavelength, decreasing considerably the requirements for the temporal and spatial coherence but the position of the interference pattern is still wavelength-dependent. It was shown recently that this drawback can be avoided and perfect achromatism achieved, when a second grating is introduced to create an image of the first grating [7]. With the latter arrangement subpicosecond pulses could be generated [8–10].

The New Arrangement

In the novel type of DFDL presented here a microscope objective is used to create an image of an amplitude transmission grating in the dye solution. The simplest implementation of this idea is shown in Fig. 1. The pump beam is expanded in the plane of the figure by a cylindrical lens telescope for a pencil-like illumination of the grating. The distance between the lenses is set so that the different orders of the diffracted beam are focussed in the nearest focal plane of the microscope objective. In normal operation, the zeroeth order is blocked by a stop and the plus and minus first orders are transmitted by the objective and emerge as collimated beams under a certain angle to create an interference pattern (image of the grating) in the dye solution. Since modern microscope objectives are highly corrected for achromatism over a very wide spectral range and the pump beam for the DFDL usually has a spectral bandwidth of at most several nanometers, no problem arises here. The same is true for other optical aberrations. The only stringent requirement is the numerical aperture NA of the microscope objective, for which a lower limit is derived in the following.

It is known that the period Λ of the interference pattern of a DFDL and its laser wavelength λ are

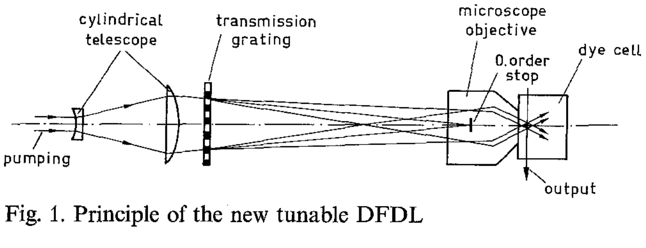

Fig. 1. Principle of the new tunable DFDL

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connected by

$$\lambda = 2 \cdot n \cdot \Lambda ,$$

where n is the refractive index of the dye solution. On the other hand, when the two interfering partial beams of the pump wavelength λ_p enclose an angle 2α the fringe period is given by

$$\Lambda = \lambda_p / (2 \cdot n \cdot \sin\alpha) .$$

If a microscope objective is used to produce the image grating with period Λ, it has to accept the rays making an angle α with the optical axis, i.e. it has to have a numerical aperture

$$\mathrm{NA} = n \cdot \sin\alpha .$$

Combining these equations, one sees that for the shortest wavelength $\lambda_{\min}$ to be generated by the DFDL using a pump wavelength λ_p, the numerical aperture has at least to be

$$\mathrm{NA} = n \cdot \lambda_p / \lambda_{\min} . \tag{1}$$

For the conventionally used glass objectives the shortest pump wavelength is approximately $\lambda_p = 360$ nm because of the absorption edge, which for most glasses lies near 350 nm. Assuming a 20 nm Stokes-shift, the shortest DFDL emission would be at $\lambda_{\min} = 380$ nm. Using the smallest value for n, namely $n = 1.3$, then (1) yields the necessary numerical aperture NA = 1.23.

Most microscope objectives are corrected for a length of the microscope tube of 160–200 mm. If the distance between the grating and the objective is set accordingly, the laser wavelength can be calculated using the nominal magnification factor M of the objective and the grating period d as

$$\lambda = n \cdot d / M .$$

For objectives with M in the range between 40 and 60 only coarse gratings of 20–60 lines/mm are needed. Furthermore an illuminated area of, say, $0.3 \times 4\ \mathrm{mm}^2$ is reduced to the sufficiently small size of $6 \times 80\ \mu\mathrm{m}^2$, necessary for subpicosecond pulse generation. The size of the illuminated area can conveniently be controlled by a mask or slits.

Tuning of the DFDL

Even though most objectives are corrected for a certain object-image distance, a small change of up to a few centimeters will not deteriorate the performance of the objective too much. In our application this means that the laser wavelength can be tuned by this shift in the demagnification by up to a factor of 1.5, which is much more than the tuning range of a single dye. For further extension of the tuning range one can make use of the variability of the refractive index with different solvents (in the range $n = 1.27$–1.60). This means an overall tuning range of this type of DFDL of 1 : 2, i.e. starting at 380 nm, tuning through the whole visible region into the near infrared by a proper choice of solvents and a limited shift of the grating is possible. The tuning range can be extended even further into the infrared by removal of the stop to admit the zeroeth order. This forms a grating image of halved period in the dye solution, so that the operation of the DFDL can be extended up to 1500 nm in principle.

Experimental Results

The performance of the new type of DFDL was tested with two different pump sources, shown in Fig. 2. Both pump sources are excimer-laser-pumped broad-band dye lasers, differing mainly in the pulse duration.

The first arrangement (Fig. 2a) is pumped by 10% of the energy of an excimer laser (Lambda Physik, 80 mJ in 15 ns at 308 nm), of which one third is used to pump a quenched dye laser of the same construction as in [11]. Its output is spatially filtered and then amplified in a dye cell pumped by the other two thirds of the pump pulse. Using a 5×10^{-4} M solution of butyl-PBD in cyclohexane in both dye cells, a 300 ps

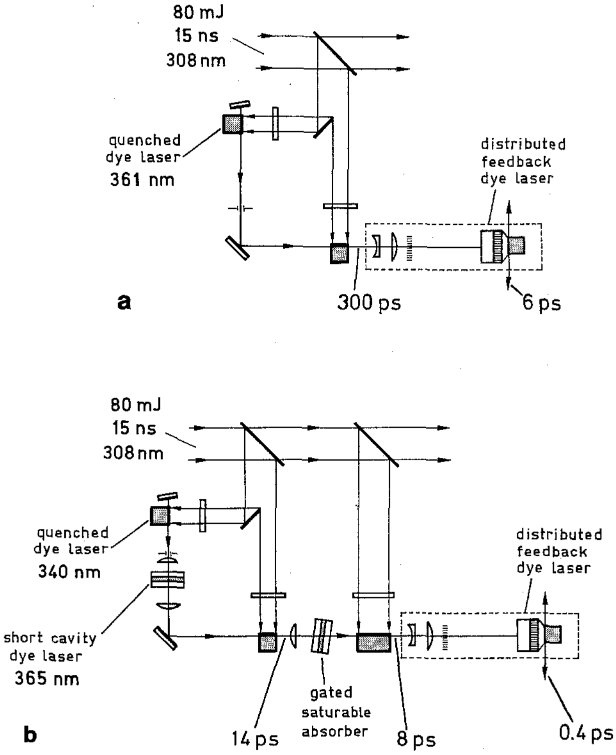

Fig. 2. Experimental arrangements used for pumping the new DFDL

long output pulse of 30 μJ energy is obtained at 361 nm. It is known from earlier observations that the spatial distribution of the output of a transversally pumped amplifier in the far field has no rotational symmetry, but is strongly modulated by diffraction at the edge of the cell window along the horizontal axis. The distribution is shown schematically in Fig. 3a, where the dashed line represents the vertical amplifier cell window. Since in our application a pencil-like illumination of the transmission grating with the longer axis lying horizontally is needed, in both experimental arrangements the last amplifier stage is turned by 90° to turn the diffraction pattern by the same amount. Only the first elliptical lobe of the diffraction pattern, which carries the main part of the energy, is used for the illumination of the grating, while the others are blocked. The horizontal dimension of this beam is then further increased by a beam expanding telescope consisting of two cylindrical lenses (planoconcave, $f=-20$ mm and planoconvex, $f=40$ mm) to about 4–6 mm. The typical vertical dimension at this point is roughly 0.5 mm.

The second experimental (Fig. 2b) arrangement is a modified version introduced first in [12] and will be published elsewhere in detail [13]. It makes use of a cascade of a quenched dye laser, a short cavity dye laser, and a gated saturable absorber, resulting in a stable pulse duration of 8 ps at 365 nm and typically 4 μJ pulse energy.

Both pumping arrangements illuminated a grating of 55 lines/mm (Dr. Johannes Heidenhain GmbH, Traunreut) consisting of evaporated metal stripes on a quartzglass substrate. Each of the two first orders contained 10% of the incident energy.

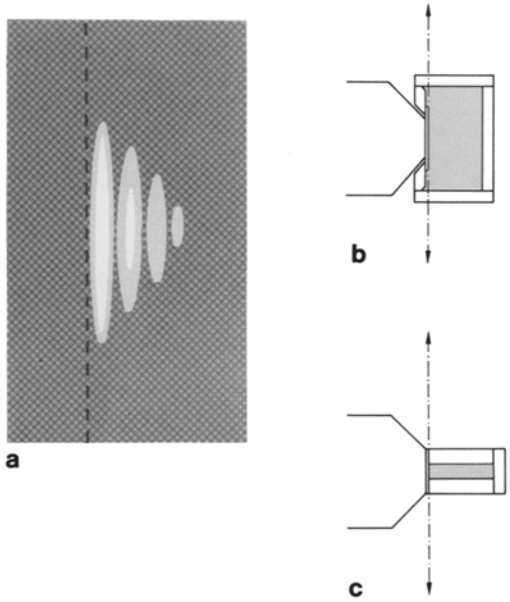

Fig. 3. (a) Spatial distribution of the output of a transversally pumped amplifier. (b) and (c) Cross-sections of the dye cells used in the experiments

Two microscope objectives were used, namely an Olympus 40/1.00 F, and a Zeiss 63/1.20 W Korr. The latter was specially for the shorter wavelength region.

Two kinds of dye cells for the DFDL were used in these experiments (Fig. 3b and c). The first type is made by drilling a hole in a standard dye laser cell and cement a cover glass over the hole from the inside of the cell, as shown in Fig. 3b. This design results in relatively long unpumped regions, so it is more useful with dyes having a large Stokes-shift and consequently little ground-state reabsorption even for concentrated solutions. The other type of cell, shown in Fig. 3c is specially constructed and universally applicable. It consists of 2 mm thick side and bottom windows and a 0.17 mm coverglass, all connected by a sintering process. In both cells the optical contact between the microscope objective and the cover glass was made with glycerole as immersion liquid.

Temporal Behavior

Figure 4 shows streak-camera measurements (Hamamatsu streak-camera C1370-01) of the temporal behavior of the arrangement according to Fig. 2a, using a 2×10^{-1} M solution of Coumarin 307 in hexafluoro-isopropanole (HFP) and the DFDL tuned to 530 nm. When pumped high above threshold, the DFDL output consists of several pulses, as is well known [2] and can be seen in Fig. 4a. The first part of the pulse train is shown with higher sweep speed in Fig. 4b. The width of the first pulse is 6 ps, which means a pulse shortening ratio of 50 with regard to the pump pulse duration of 300 ps.

The pulse duration measurements for the arrangement shown in Fig. 2b could only be done with an autocorrelator because of the shorter pulses. The

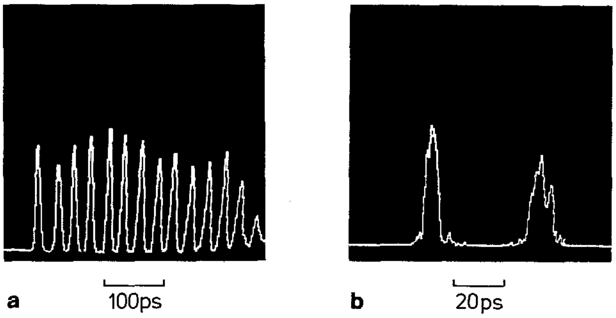

Fig. 4. (a) Streak-camera record of the DFDL pulse train pumped by a 300 ps long pulse. (b) Streak-camera record of the first two pulses with higher temporal resolution

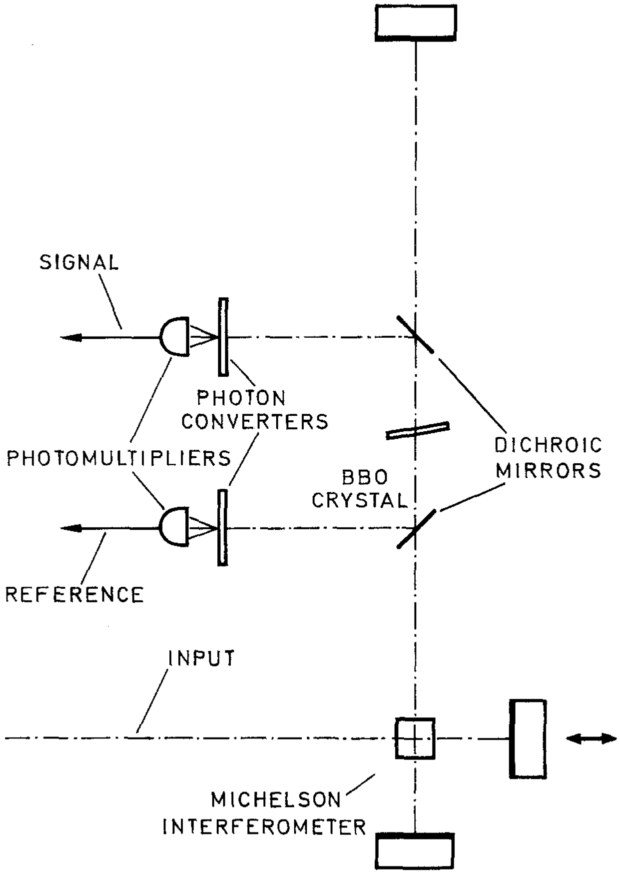

Fig. 5. Principle of the autocorrelator

autocorrelator used for this purpose is shown in Fig. 5. It consists of a small Michelson interferometer, a frequency-doubling crystal (BBO, 1 mm thick, cut 52°) and a detection system. In this arrangement the crystal is used twice. In the first pass the second-harmonic signal is proportional to the autocorrelation function and is coupled out by a dichroic mirror to be measured by the signal photomultiplier. The fundamental is transmitted and reflected by a mirror positioned in half a meter distance. Since one of the mirrors of the Michelson interferometer had to be slightly misaligned in order to measure the slow autocorrelation function (averaging over several fringes in the field of view), the two partial beams returning from the distant mirror do not overlap any more when passing the crystal for the second time. Consequently they generate a second-harmonic signal which is only proportional to the square of the intensity of the input pulse, so that it can be used as a normalization reference signal. The ratio of the autocorrelation and the reference signal was formed and then averaged in a Lambda-Physik laser photometer (LF300).

Figure 6 shows autocorrelation curves obtained with the arrangement according to Fig. 2b after amplification in a single stage amplifier. The Zeiss objective was used and a 1×10^{-1} M solution of Coumarin 307 in HFP. The DFDL was tuned to 500 nm and pumped two times (Fig. 6a) or three times (Fig. 6b) above threshold. In the first case the autocorrelation width is

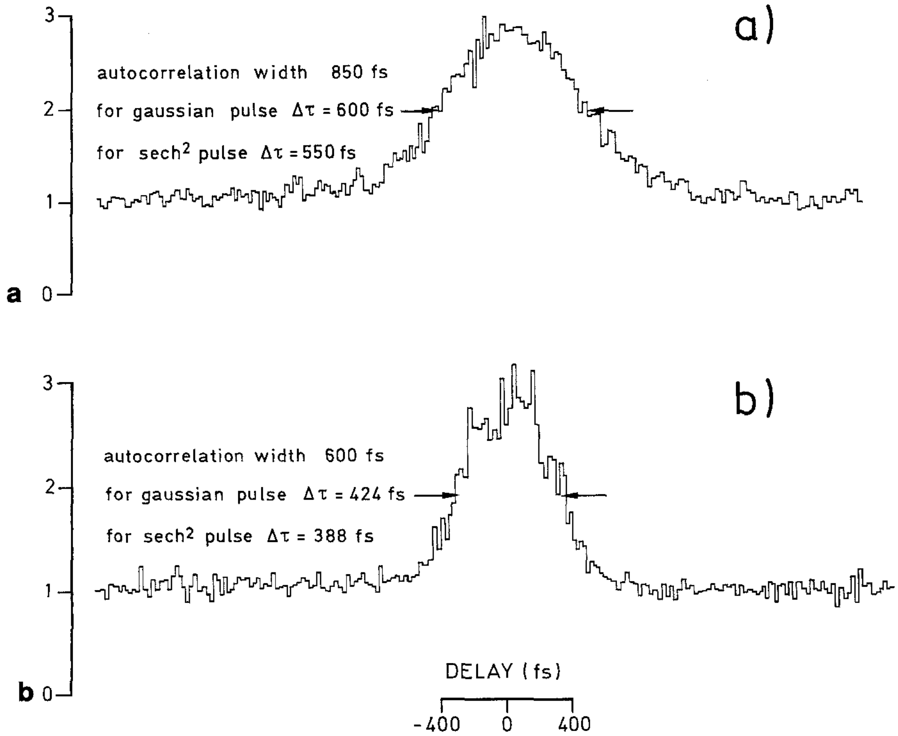

Fig. 6a, b. Autocorrelation curve of the amplified DFDL pulses when the DFDL is (a) two times, (b) three times above threshold

850 fs, corresponding to 550 (600) fs pulse duration assuming a sech^2 (Gaussian) pulse form. With stronger pumping the pulse width decreases, but the fluctuations increase, as can be seen in Fig. 6b. Further amplification of these pulses led to less fluctuation and further shortening of the pulse duration.

Spectral Behavior

The shape of the spectral band of the DFDL is strongly dependent on the pump level. Figure 7 shows a series of spectra obtained with increasing pump level. Figure 7a shows the spectrum slightly above threshold, consisting of a single line of 0.2–0.3 nm width. It broadens to 0.6 nm when pumped two times above threshold, corresponding to shorter pulses. At still higher pump levels the spectrum becomes modulated with statistically fluctuating separations of the lines (Fig. 7c and d). This was previously thought to be a superposition of the spectral lines of the numerous pulses of a strongly pumped DFDL [14, 15]. However, it was found that already the first pulse, which is the only one remaining after amplification [16] in a strongly saturated amplifier, shows the modulated spectrum. This could possibly mean that the various peaks belong to different modes of the DFDL which start to oscillate simultaneously or with slight delay when pumping high above threshold.

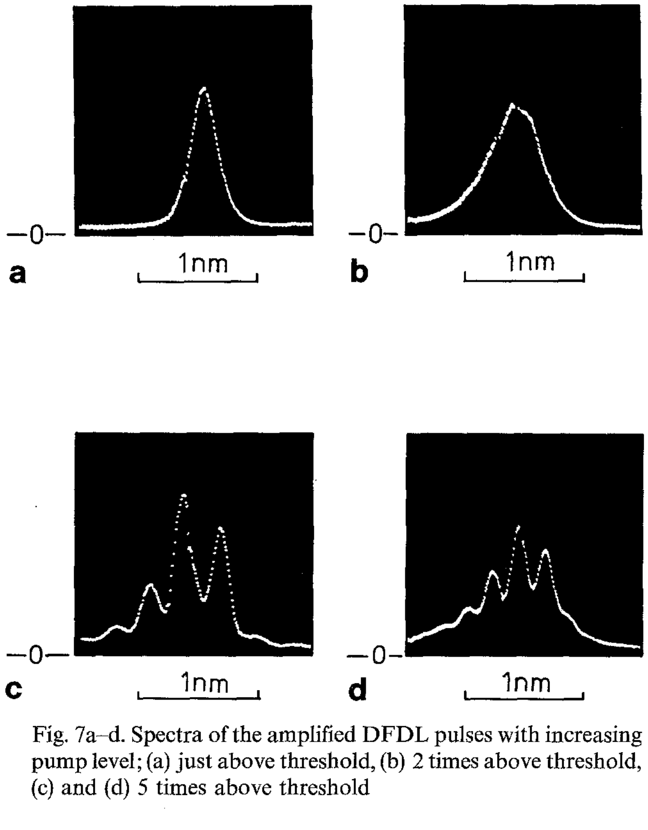

Fig. 7a–d. Spectra of the amplified DFDL pulses with increasing pump level; (a) just above threshold, (b) 2 times above threshold, (c) and (d) 5 times above threshold

Figure 8 demonstrates the tuning of the DFDL, while it was pumped two times above threshold, corresponding to the shortest pulses with smooth and well defined spectrum. The spectra were measured with a Spex 0.75 m spectrograph (SPEX Industries, Inc. 1704) with an optical multichannel analyzer attached, having 0.03 nm resolution. Four 10 nm wide samples were taken from the 70 nm wide tuning range for Coumarin 307 in HFP and several spectra recorded for different positions of the transmission grating, as shown in Fig. 8.

It is worth noting that tuning of the DFDL by shifting the transmission grating does neither change the position of the active volume nor the direction of the output beam. Even when the grating is shifted by more than a centimeter, corresponding to several 10 nm tuning, only a very minor deterioration of the visibility of the grating and hence the operation of the DFDL can be observed. For an even wider tuning this deterioration can be avoided by a very minuscule translation of the objective, until the grating image is sharp again. This operation could easily be automated and leaves the position of the DFDL and the direction of its output beam unchanged, thus eliminating the need for a realignment of the eventually following amplifier stages.

Figure 9 demonstrates the wide overall tuning range obtained with different dyes and solvents by translation of the transmission grating. The left column was measured with the Zeiss objective, the right one with the Olympus objective. The short wavelength limit of Quinolon 390/HFP (left column), Coumarin 307/HFP, Coumarin 153/dioxane, and Coumarin 153/DMF (right column) was determined by the numerical aperture of the objectives. With solvents of low refractive index in the long-wavelength region the tuning range was narrowed by the fact that the objectives had to be used relatively far from the optimum position of correction.

We would like to mention that the tuning method presented here is only one of several possibilities. For instance, the grating can be imaged by a zoom objective as an intermediate image, which is then imaged by the microscope objective into the dye solution. In this way the intermediate image grating period can be changed in a very simple way, while the position is stationary. The blocking of the zeroeth order can already be done in the zoom objective. Using only cylindrical lenses in the zoom objective, the cylindrical telescope mentioned above is superfluous. If the temporal coherence of the pump pulse is high enough, a Fizeau wedge or a simple beam splitter can be used instead of the transmission grating.

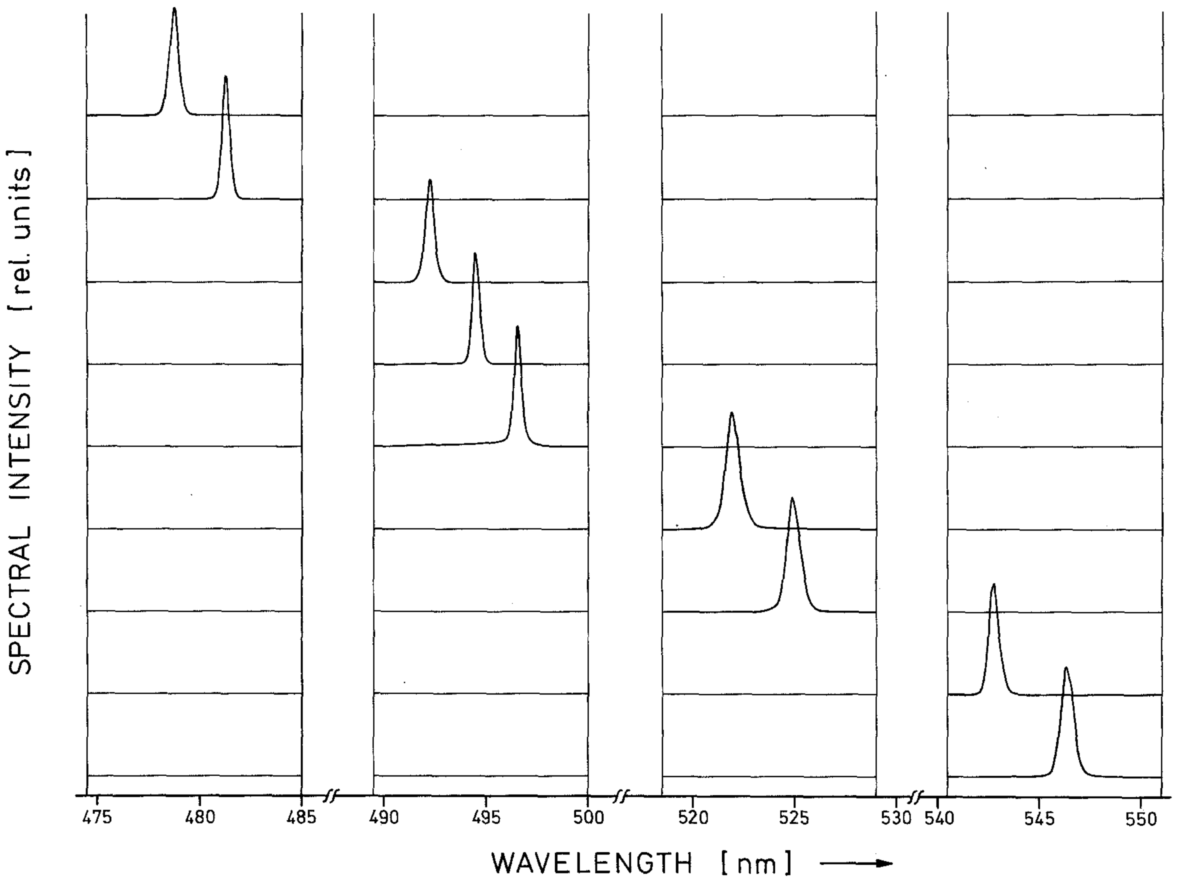

Fig. 8. Continuous tuning of the wavelength of the DFDL

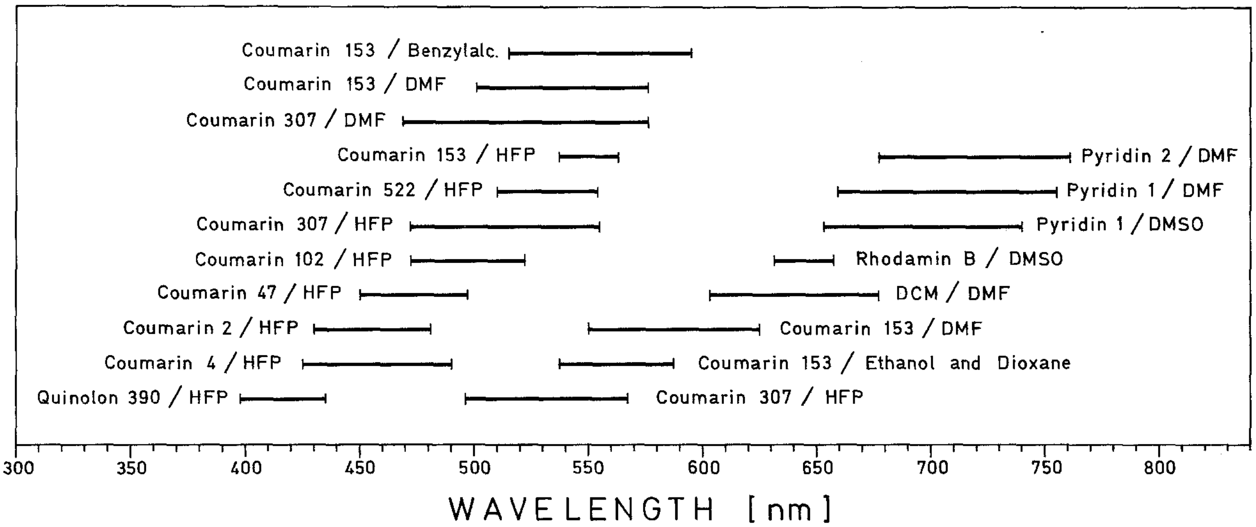

Fig. 9. The tuning range of the DFDL with the use of a 63/1.25 objective (left column), and a 40/1.00 objective (right column)

Conclusion

We have described an achromatic and widely tunable DFDL. It is based on the use of a microscope objective, which images a transmission grating into the active medium providing the periodic structure for the distributed feedback. The structure has high visibility even using a pump beam of several nm bandwidth and low spatial coherence. The period of the structure can easily be changed by a translation of the transmission grating, making continuous tuning of this DFDL very convenient. In this way 70 nm continuous tuning was achieved. The tuning procedure leaves both, the position and the direction of the DFDL output unchanged, which is very convenient when further amplification of the DFDL pulses is necessary.

We have reported the temporal and spectral behavior, in particular the wide tuning range from 400 to 760 nm. Typically 500 fs pulses could be achieved with this system.

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