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Generation of 320 fs Pulses with a Distributed Feedback Dye Laser

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Abstract. A new achromatic distributed feedback dye laser (DFDL) arrangement is described. The experimental conditions for subpicosecond pulse generation with the new device were investigated. For the first time, stable generation of subpicosecond pulses ($\tau \leqq 350$ fs) at 616 nm was achieved with a DFDL. The simultaneous spectral and autocorrelation measurements showed that the amplified DFDL pulses are nearly transform limited, having a pulse form close to the sech^2 shape.

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Cw dye lasers are capable of directly generating pulses as short as 27 fs pulses [1]. For many applications no such a short pulse duration is required. Instead, broader tunability and single pulse generation capability may be desired. This can easily be fulfilled by a DFDL [2].

Up to now the shortest pulse which has been generated by a DFDL is in the 2–3 ps region [3, 4]. With a travelling wave excitation scheme [5], the shortest pulse duration achieved was estimated to be around 1 ps. These values are still far from the theoretical limit [6]. Our aim is to study the effect of different experimental conditions on the operation of a subpicosecond DFDL, and to approach the theoretical limit by the new DFDL pumping scheme which we reported in [6].

Experimental Arrangement

In our experiments we use the newly introduced arrangement [6], as is shown in Fig. 1. This setup ensures the creation of perfect, high visibility interference fringes, and of a well defined small-size active volume even in case of broadband pumping. The pump beam ($\lambda = 365$ nm) passes through the quartz (Q) block and hits grating 1 (2208 l/mm Zeiss holographic grating blazed for 365 nm). One of the two diffracted orders is reflected on the side wall of the Q block by total internal reflection; then the two beams are propagating parallel inside the Q block.

In the position of grating 2 an identical grating is used, but in second order autocollimation. This grating is glued to the Q block with EPO-TEC 305 – serving as immersion between the optical components – while the first is independently mounted on a special holder allowing the two most important movements for alignment: translation along and rotation around the axis perpendicular to the active surface of the gratings. The selective use of immersion resulted in a relatively high efficiency (30% for each of the diffracted first orders in air, and $\approx 80\%$ for second-order autocollimation in immersion) using the same type of grating. After diffraction on the surface of grating 2, the two

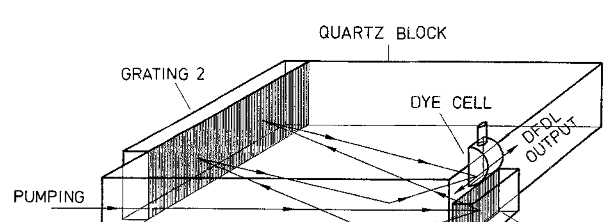

Fig. 1. Schematic of the new DFDL pump arrangement

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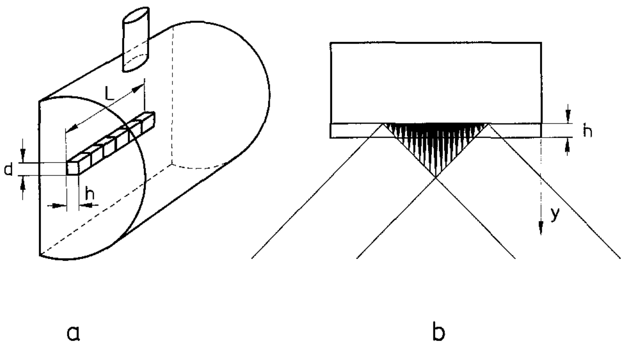

Fig. 2. (a) Schematic of the pumped volume in the DFDL (b) Top view of the front plane of the dye cell

pump beams propagate in a reversed way, and – due to a small tilting angle of the optical elements – are combined just above grating 1, at the inner surface of the dye cell. The dye cell – containing 7×10^{-2} M Rh B in methanol – is optically contacted to the quartz block.

The length of our subpicosecond DFDL (L) must be less than 100 μm [6]. Our experiments showed that if the dimensions of the active volume perpendicular to the DFDL radiation are comparable to the length of the DFDL structure, some directional unstability of the DFDL output appears. Then the far field of the DFDL output consists of circular illuminated points whose distribution is statistically fluctuating from shot to shot.

This situation is caused by the interference of numerous DFDL laser channels operating simultaneously. Since all of them fulfill the Bragg condition they are spatially coherent. However, because of the statistical phase condition of these individual channels, the resultant intensity distribution in the far field shows the above mentioned statistical shot-to-shot fluctuation. Decreasing the cross-section of the DFDL to the cross-section of an individual laser channel, this fluctuation disappeared. If only the vertical dimension of the DFDL (denoted by d in Fig. 2a) was comparable to L, the far field consisted of numerous horizontal lines – corresponding to the interference of parallel laser channels, lying in the plane of the inner front surface of the dye cell. In order to keep the pencil-like shape of the excited volume, the vertical size of pumping at the dye cell (d) had to be less than 10 μm.

The other dimension of the DFDL (h), in the direction perpendicular to the surface of the dye cell, was automatically small. In spite of the fact that the penetration depth of pumping – because of the limited solubility of the Rh B – was larger than 10 μm, the interference pattern, necessary for DFDL operation, was created only in the overlap of the two beams, and has a sharply decreasing length in the y direction (Fig. 2b). If the overlap of the two pump beams is complete just behind the surface of the dye cell, the length of the interference pattern has a maximum there, and the visibility of interference fringes is high. Along the y direction this visibility decreases, because of the limited spatial coherence of the pump beam.

If we assume that the gain (G) of the DFDL at complete overlap of the pump beams is two times above threshold ($G = 150$), the depth of the active volume (h) capable for DFDL operation can be calculated.

Even if we neglect the decrease of the visibility of the interference pattern and the pump intensity along the axis y the calculated depth

$$h \approx 0.2\,L.$$

In practice, the above-mentioned dependence of the visibility and of the pump intensity versus y must be taken into account, which leads to a further decrease of h. This means that h is smaller than L by at least one order of magnitude, which is in agreement with our experimental observations.

For pumping this DFDL the cascade dye laser setup which we reported in [7] was used. Since the pulse duration of the DFDL was not sensitive to the change of the pump pulse duration in the 3–8 ps region, the simplest cascade arrangement was used incorporating a quenched dye laser, a short cavity dye laser and one gated absorber, providing 8.2 ps $\pm$ 0.4 ps pump pulses for the DFDL.

The output of the cascade laser was nearly diffraction limited. In order to create the necessary 100 μm $\times$ 10 μm beam profile at the surface of the dye cell of the DFDL, a special telescope was used ($f_1 = -16$ mm planconcave spherical lens, $f_2 = 60$ mm planconvex cylindrical, $f_3 = 80$ mm planconvex cylindrical) having different magnification (and focal plane) for the horizontal and vertical direction.

Using the above apparatus the horizontal and vertical size of the excited volume could be changed independently. Figure 3 shows the horizontal (a), and vertical (b) intensity distribution of the pump beam at the surface of the dye cell.

Under optimal operational condition the divergence of the DFDL output showed only a slight shot-to-shot fluctuation, which could easily be eliminated by spatial filtering of the output, allowing only the central circular part of the beam to enter to the amplifier.

The DFDL wavelength is determined not only by the static refractive index of the solvent, but dynamic refractive index changes also influence the wavelength of the DFDL [8]. This problem is subject of further investigation. Since the latter refractive index changes are introduced by pumping – which has some shot-to-

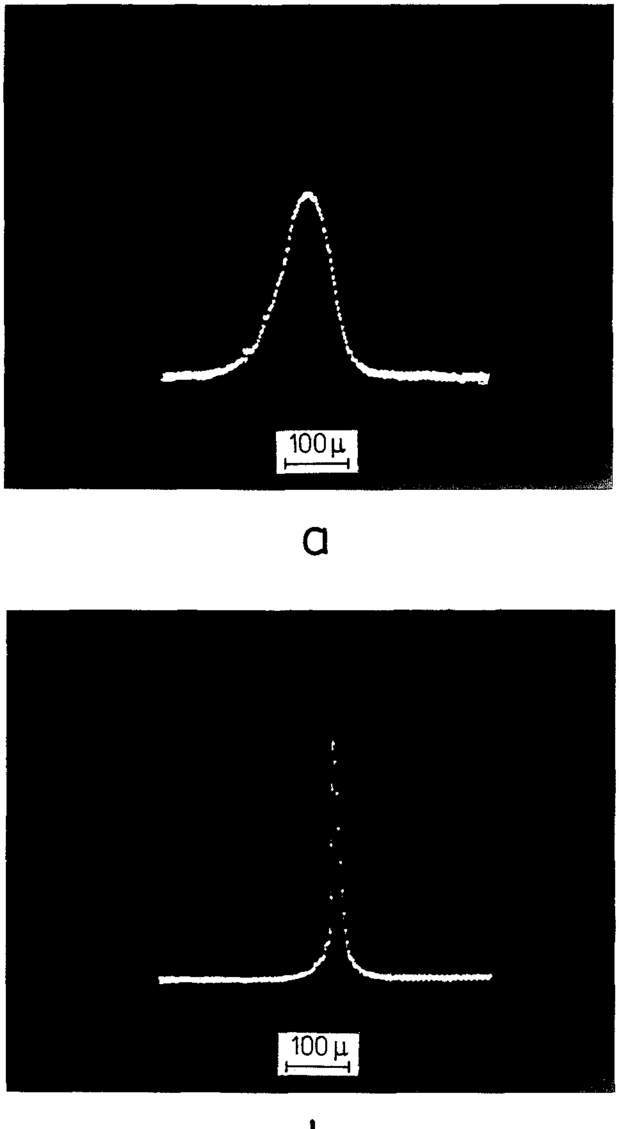

Fig. 3a, b. Pump intensity distribution at the front surface of the dye cell for horizontal (a) and vertical (b) direction

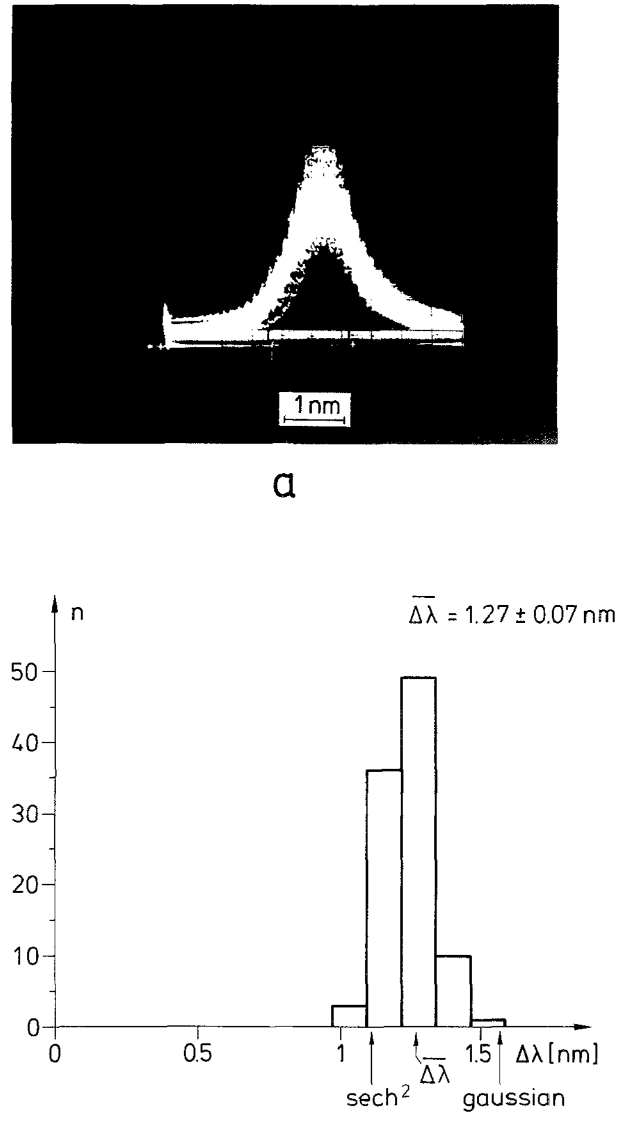

Fig. 4. (a) Superimposed spectra of 100 subsequent amplified DFDL pulses (b) Histogram of the bandwidth of 110 pulses

shot fluctuation – this kind of refractive index change can be the origin of a fluctuation in the central wavelength of the DFDL ($\Delta\lambda_c$). Figure 4a shows that under our experimental condition this fluctuation remained small compared to the bandwidth ($\Delta\lambda$). According to our observation

$$\Delta\lambda_c < \frac{\Delta\lambda}{3}.$$

Tunability of the DFDL can be accomplished by the change of the refractive index of the solution. By proper choice of the solvent, several 10 nm tuning can be realized. Convenient temperature tuning is also possible through the temperature dependence of the refractive index of the active medium.

The DFDL excited volume was imaged to the entrance of a three-stage amplifier of standard design. The amplifier stages – containing $c_1 = 1.2 \times 10^{-3}$ M, $c_2 = 7 \times 10^{-4}$ M, $c_3 = 5.5 \times 10^{-4}$ M Sulforhodamin B dye ethanolic solution – were separated by saturable absorbers (2 mm thick RG 645 Schott glass filter and 1 mm 5×10^{-4} M Malachite green). After the second amplifier, a polarizer cut the residual part of the unwanted polarization. As a last stage a Bethune type dye cell [9] was used (∅ = 1.5 mm) to get nearly diffraction limited output. The amplifier was not optimized for the highest output energy, which was actually 80 μJ in our experiment, because these pulses (after frequency doubling) served as input signals for amplification in XeCl in a later experiment [10]. The

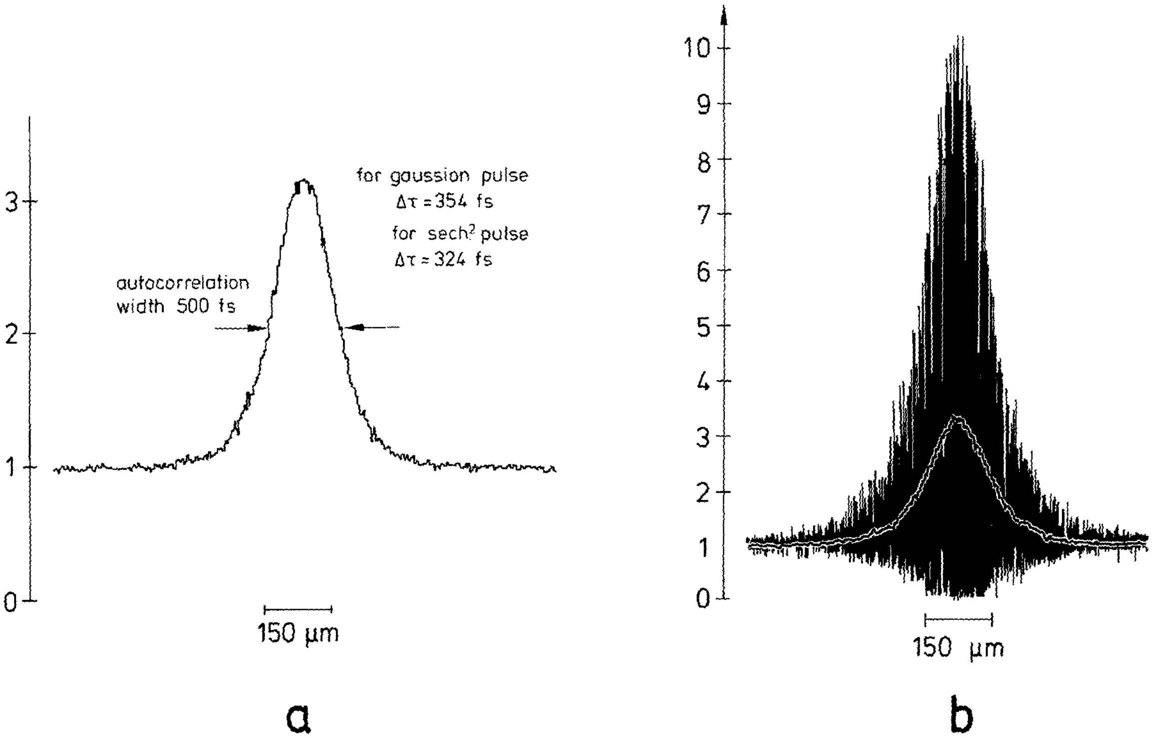

Fig. 5. (a) Autocorrelation curve of the amplified DFDL output, (b) the corresponding fringe-resolved autocorrelation curve

fluctuation of the output energy was measured to be $\pm 15\%$.

Results

The spectral and temporal properties of the output were studied after the last amplifier stage. The results are shown in Figs. 4 and 5.

Figure 4a shows the superimposed spectra of 100 subsequent DFDL pulses exhibiting relatively small fluctuation in bandwidth and in the central wavelength. Figure 4b is the histogram of the measured value of the bandwidth of 110 pulses. It can be read from the figure that the average is 1.27 nm with a standard deviation of 0.07 nm. The small value of the fluctuation is worth noting. The spectra were recorded with a home-made spectrograph, attached to a diode-array/monitor combination. For the calibration of this setup a Fabry-Perot etalon was used in reflection, having a free spectral range of 1.29 Å (which is responsible for the modulation of the spectrum seen in Fig. 4a).

Figure 5a shows the autocorrelation curve of the amplified DFDL output using the standard collinear SHG technique. Before the Michelson interferometer $\approx 20\%$ of the beam was sent to another SHG crystal to give a reference signal, which was used to eliminate the error caused by the intensity fluctuation of the pulses. Both, in the signal channel and in the reference channel, 1 mm thick KDP crystals were used. The beams were sent through the crystals without focusing, and the second harmonic signal was monitored by two uv sensitive photodiodes HAMAMATSU R1193U. The ratio of the two signals was formed and then averaged by a LAMBDA PHYSIK laser-photometer (LF 300).

The width of the autocorrelation curve was measured to be 500 fs, which corresponds to 320 fs (350 fs) pulse duration for $sech^2$ (Gaussian) pulse shape respectively. Assuming transform limited pulses, this corresponds to a bandwidth of 1.12 nm (1.57 nm). If we compare these two values to the average of the measured bandwidth in Fig. 4b, one can see that the average is somewhere in between, but closer to the value obtained by the assumption of $sech^2$ pulse shape. Since the shot-to-shot fluctuation cannot have significant influence on the measured pulse duration, one can conclude from the above results that the amplified DFDL pulse has practically $sech^2$ shape and nearly transform limited behaviour. The fringe-resolved autocorrelation curve in Fig. 5b also supports this assumption.

The accuracy of measurement is of course not high enough to decide whether this slight difference between the measured average bandwidth and the bandwidth obtained from the temporal measurement derives from the assumption of the transform limited behaviour of DFDL pulses or from the assumption of the $sech^2$ pulse shape. It is known from the DFDL theory [2] that the output of the DFDL is transform limited and has Gaussian shape. Possibly the combined effect of absorber and amplifier introduces some exponential behaviour, resulting in a final pulse shape of approximately $sech^2$ pulse shape.

Conclusion

The new DFDL arrangement is capable of generating subpicosecond pulses, close to the theoretical limit of the shortest pulse which can be generated by a DFDL. Using this arrangement, stable generation of transform limited, $\leqq 350$ fs pulse at 616 nm was achieved.

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