SIMPLE METHOD FOR TEMPORAL STUDY OF SUBPICOSECOND DISTRIBUTED FEEDBACK DYE LASERS

P. SIMON [1], H. GERHARDT
Laser-Laboratorium Göttingen e.V., Robert-Bosch-Breite 10, D-3400 Göttingen, Fed. Rep. Germany

and

S. SZATMÀRI [2]
Max-Planck-Institut für Biophysikalische Chemie, Am Fassberg, D-3400 Göttingen, Fed, Rep. Germany

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The spectral and temporal behaviour of subpicosecond DFDLs are studied. A simple and sensitive spectral diagnostic method is proposed to exhibit the presence and determine the temporal separation and relative amplitude of any following pulse. The measurements were performed in a hybrid excimer-dye laser system generating less than 100 fs pulses at 248 nm.

1. Introduction

Distributed feedback dye lasers (DFDLs) [1] are able to generate transform limited ultrashort pulses in a wide spectral range [2]. Although the shortest pulse duration that can be generated by DFDLs (several hundred femtoseconds) [3,4] is still longer by an order of magnitude than that of cw dye lasers [5], there are many applications where DFDLs are preferable due to their easy tunability and automatic synchronization to the following amplifiers. Perhaps the best example is the technique of femtosecond excimer pulse generation, where a hybrid excimer-dye laser system based on DFDLs has proved to be the simplest way for the generation of femtosecond pulses at most of the excimer wavelengths [6–9].

As it is shown in ref. [2] the DFDL generates a single transform limited pulse when pumped just above its threshold. With stronger pumping multiple pulse generation is expected. In this case the requirements for the pump stability are decreased, however pulse selection is required to obtain – the generally needed – single pulses [10,11]. The single pulse generation regime in picosecond DFDLs could easily be controlled by spectral measurements, since the spectra of the successive pulses were slightly shifted [12,13].

For subpicosecond DFDLs different spectral behaviour is found in ref. [8], but no explanation is given there.

Since the exclusion of any trailing pulse is crucial in many spectroscopic applications, we have studied the correlation between the spectral and temporal behaviour of subpicosecond DFDLs. In this paper we show a method to control the temporal behaviour through spectral measurements.

2. Experimental

The scheme of our experimental setup (fig. 1) is just the same as described in ref. [8]. One tube of a Lambda Physik EMG 150 MSC double discharge excimer laser, filled with XeCl pumps an intermediate dye laser as described in ref. [14]. The 2 μJ, ~ 8 ps output of this laser is used to pump the DFDL

[1] Permanent address: Jate University, Dept. of Exp. Physics, Szeged, Dòm tèr 9, H-6720 Szeged, Hungary.
[2] Permanent address: Jate University, Research Group on Laser Physics of the Hungarian Academy of Sciences, Dòm Tèr 9, H-6720 Szeged, Hungary.

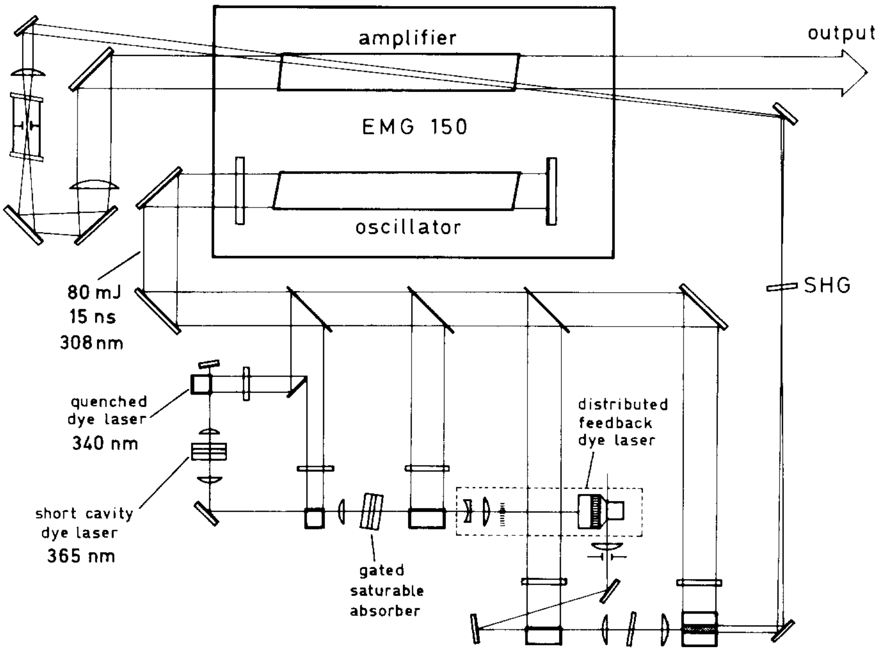

Fig. 1. Experimental setup.

master oscillator [15]. The subpicosecond 496 nm pulses generated are amplified in a two stage dye amplifier pumped by the same XeCl laser. After frequency doubling in a 0.2 mm thick BBO crystal the 248 nm, fs pulses are amplified in the other discharge channel of the excimer laser, filled with KrF. In a double pass arrangement 9 mJ output energy is obtained.

Although the principle of the arrangement is the same as in ref. [8], we introduced several changes resulting in more stable and reliable operation up to 20 Hz.

3. Results

Simultaneous spectral and temporal study of the 496 nm pulses, just before frequency doubling was carried out. For the spectral measurements a homemade spectrograph was used with a diode array attached, with a resolution of ~0.2 Å. The temporal measurements were done by a Hamamatsu C1587 streak-camera, giving ~2 ps resolution.

With pump energies up to two times the DFDL threshold single pulse operation was observed. A typical pair of a streak-camera trace and spectrum is shown in fig. 2a. The pulsewidth is limited by the resolution of the streak-camera as was checked by autocorrelation measurements. The typical pulse duration was measured to be 650 fs, close to the transform limit of the 4 Å wide spectrum, supposing sech^2 shape for the pulses.

When the DFDL is pumped 2–5 times above its threshold, two or sometimes three pulses are generated, as shown on the streak-camera traces in figs. 2b–d. It is seen on the corresponding spectra, that multiple pulsing is always accompanied by a structured spectrum. Note, that larger temporal separation of the subsequent pulses results in smaller modulation period in the spectrum, and a trailing pulse of smaller intensity causes smaller modulation depth.

Figs. 3a,b shows the spectra of the final UV output, when the DFDL generates a single pulse (fig. 3a) and two pulses (fig. 3b). In the latter case the modulation in the spectrum indicates the presence of a trailing component. Note, that the modulation appears only in a relatively narrow band of the full spectrum, and the modulation depth is much smaller than for the green pulses.

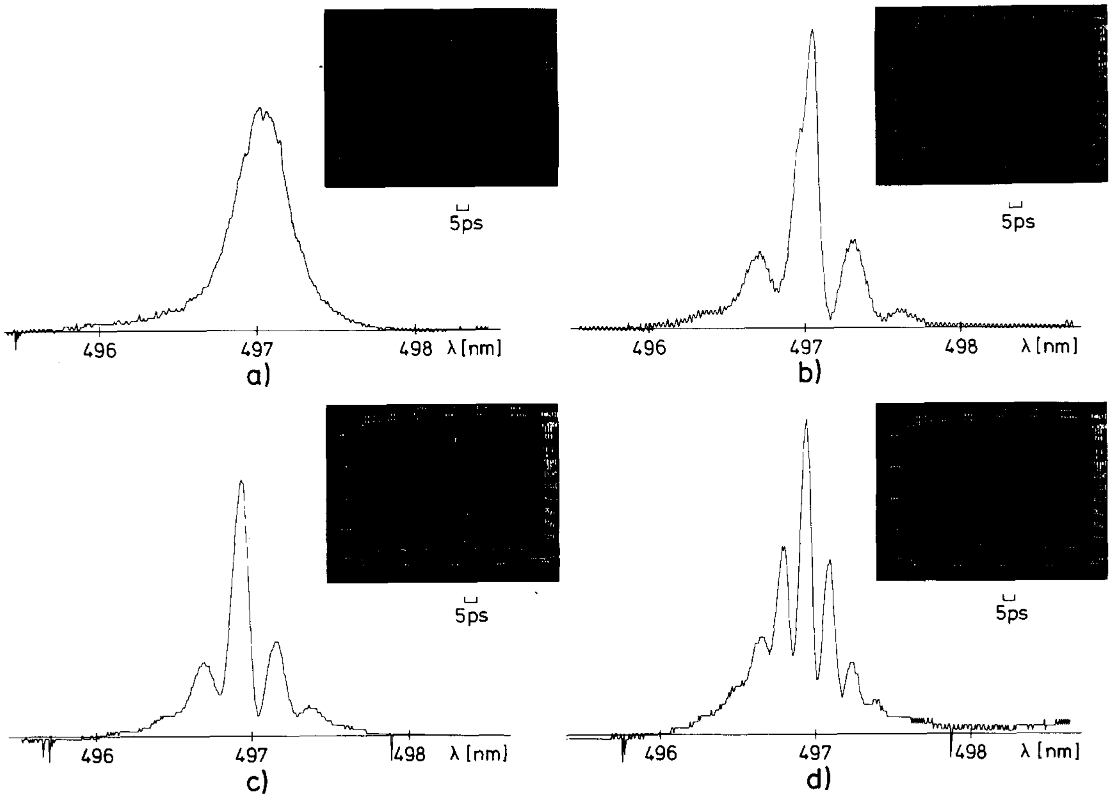

Fig. 2. Simultaneously measured spectra and streak camera traces of the amplified DFDL pulses. The DFDL operates (a) ~1.5, (b) ~4, (c) ~3, (d) ~2 times above threshold.

Using the result of the simultaneous spectral and temporal measurements of 50 shots when the DFDL pumped well above threshold, the modulation period observed in the spectra ($\Delta\lambda$) versus the temporal separation of the main and trailing pulses (ΔT) is plotted in fig. 4.

4. Discussion

One can fit the experimental results (fig. 4) by the function

$$\lambda = (\lambda^2/c)(1/\Delta T), \qquad (1)$$

where λ is the wavelength of radiation and c is the speed of light. Eq. (1) is set up with the assumption that the DFDL, when pumped well above threshold emits two coherent pulses.

Then it can be modelled by a single pulse generator whose output is split temporally by e.g. a Michelson interferometer. Eq. (1) expresses the free spectral range (FSR) of an interferometer as a function of the time delay between the two arms. Now, if two subsequent coherent pulses enter a spectrograph of sufficient dispersion, a temporal overlap is created between the pulses at the exit plane because of the temporal front delay introduced by the dispersive element [16,17]. Therefore the spectrograph shows the interference of the two pulses, i.e., according to the model, wavelength-dependent transmission of the Michelson interferometer modulated by its FSR.

A very important feature of this spectral method is its extremely high sensitivity. For comparison, let us regard the most often used second order autocorrelation technique. If a trailing pulse is present, the autocorrelation function has side maxima on both sides of the main peak. The contrast ratio between

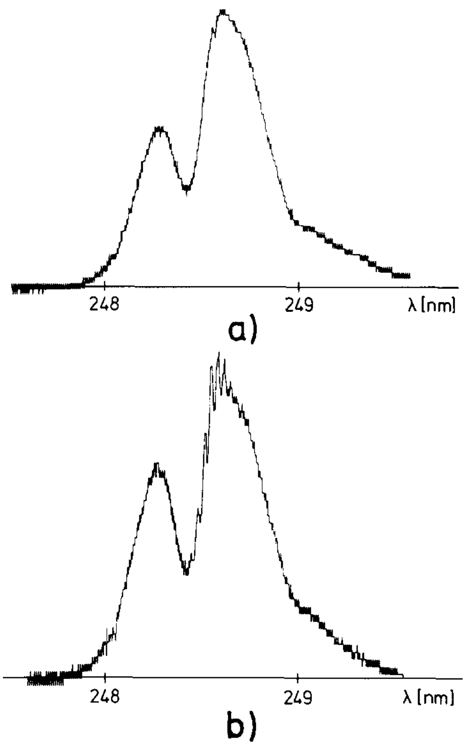

Fig. 3. Spectra of the fs excimer laser output when the DFDL generates (a) single, (b) multiple pulses.

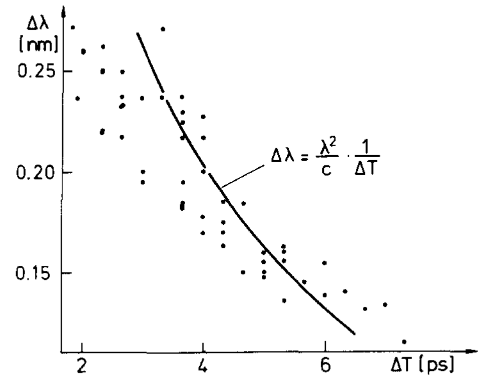

Fig. 4. Modulation period of the spectra versus temporal separation of the DFDL pulses.

the side maxima and background is

$$\mathrm{Max/Min} = 1 + 4/(I_2/I_1 + I_1/I_2), \quad (2)$$

where I_1 and I_2 are the intensities of the two pulses. With the use of our spectral method the contrast ratio between the maxima and the minima of the modulation of the spectrum is given by

$$\mathrm{Max/Min} = (1+\sqrt{I_2/I_1})^2/(1-\sqrt{I_2/I_1})^2. \quad (3)$$

Eqs. (2) and (3) are plotted in fig. 5 for different ratios of I_1 and I_2. It is seen that the spectrograph gives much higher contrast. Moreover, the comparison is only valid for single shot autocorrelations. When multiple-shot autocorrelation technique is used for DFDL pulse measurement, the contrast is further reduced due to the jitter between the first and second pulses.

It is worth noting that in contrast to picosecond DFDLs where the trailing pulses have slightly shifted spectra [12,13], the pulse train generated by a femtosecond DFDL is coherent. This behaviour has already been observed with a previously used pump arrangement [18], and with different dyes (Coumarin 47, Coumarin 102, Coumarin 307, Rhodamine B, DCM). In ref. [13] the spectral shift was attributed to gain induced refractive index changes related either to some unknown loss developing during the pulse train or to triplet state accumulation both leading to higher necessary gain for the following pulses. If the first explanation was right we should also observe a similar spectral shift of the trailing pulses. Since the main difference between the two

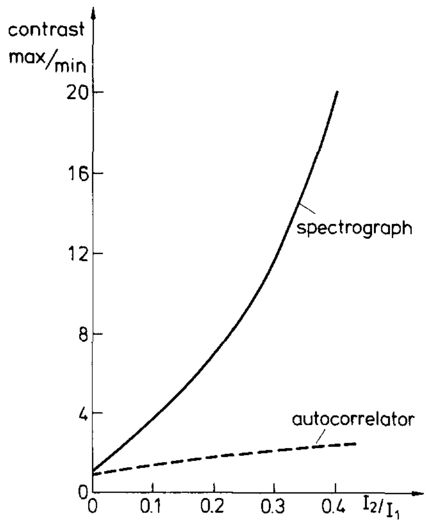

Fig. 5. Calculated sensitivity of the spectral and autocorrelation methods for the detection of trailing pulses.

experiments is the much different timescale, triplet accumulation seems a plausible reason for the earlier observed spectral shift.

As a combined effect of self-phase modulation (SPM) of the DFDL pulses in the OG 530 saturable absorber placed between the amplifier stages and saturation of amplification in the KrF gain medium, the UV output has a similar shape observed in ref. [8], shown in fig. 3a. Broadening of the spectrum is mainly attributed to the chirp introduced by the saturable absorber, the dip, located at twice the maximum frequency of the DFDL is presumably the result of some strong nonlinearity in the amplification process.

When the DFDL is pumped well above threshold some structure appears even in the UV spectrum (fig. 3b) indicating the presence of trailing pulses. The fact that this modulation seems to appear only in a limited range of the full spectrum is easily understood by considering that it is only the first pulse which undergoes SPM in the saturable absorber and the spectrum of the trailing component remains unchanged.

Another important feature of the UV spectrum is its weak modulation which can be explained by the pulse selection effect of amplifiers [10]. In saturated amplifiers, because of strong gain depletion by the first pulse being amplified, the trailing pulses see much less gain. From fig. 3b, 0.1% relative energy content of the trailing pulse with respect to the main pulse is evaluated. In applications where even this trailing pulse is disturbing, stable single pulse generation can be achieved by keeping the energy of the XeCl pump laser within ±7%.

An inherent property of the UV pulses generated by our system is their linear frequency chirp [7,8]. Thus, directing the output beam along a dispersive line with sufficient negative group velocity dispersion, the pulses can be compressed. To demonstrate this feature we used a pulse compressor consisting of a pair of prisms as in ref. [8].

Typical autocorrelation traces of the UV pulses before and after compression are shown in figs. 6a and b, respectively. When the DFDL generated either single or multiple pulses – that could easily be controlled by the spectral measurement – no difference was observed in the shape of the autocorrelation traces. This is understandable, since a trailing pulse

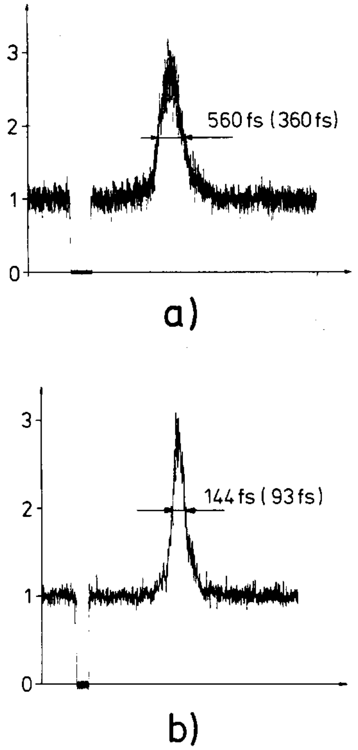

Fig. 6. Autocorrelation traces of the (a) direct and (b) compressed UV output.

having 0.1% energy content and some jitter with respect to the main pulse is expected to give a minor contribution to the autocorrelation trace which is far below the noise level. The broader autocorrelation width of the compressed pulses compared to that reported in ref. [8] is attributed to the narrower spectral width due to less saturation in the excimer amplifier. The autocorrelation curves were recorded without selection of pulses, without averaging and without using any reference. From these traces one obtains less than ±4% fluctuation both for the energy and for the pulse width.

5. Conclusion

In conclusion we have studied the spectral and temporal behaviour of subpicosecond DFDLs. A simple and very sensitive spectral method is proposed for the detection of trailing pulses, for determination of their relative amplitude and temporal separation. These measurements were performed in a DFDL-based hybrid excimer-dye laser system whose stability was significantly improved. As a result <100 fs pulses at 248 nm were routinely generated (at 20 Hz) without any need of maintenance for 10^6 shots.

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