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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.

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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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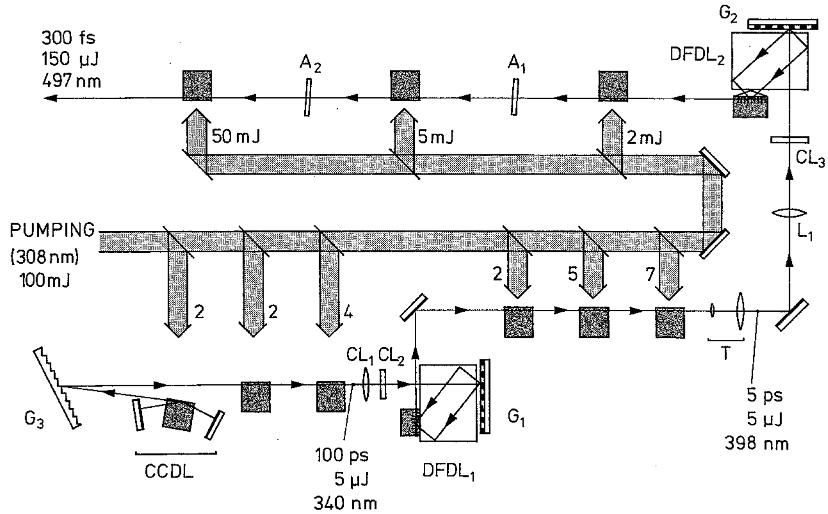

Fig. 1. Experimental arrangement. CCDL: competing cavity short pulse dye laser; $DFDL_1$ and $DFDL_2$: cascade distributed feedback dye lasers. The numbers indicate the approximated pump energies in mJ

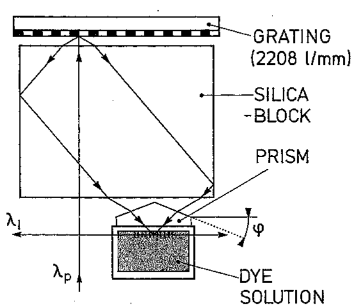

Fig. 2. Optical scheme of $DFDL_2$

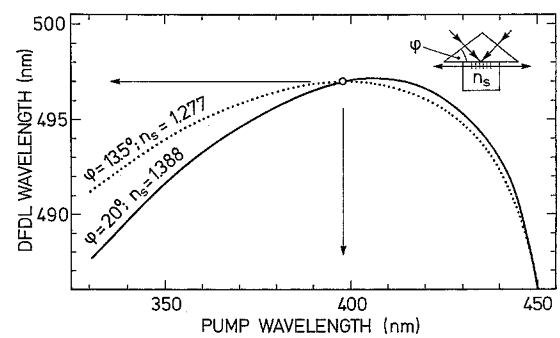

Fig. 3. Wavelength of $DFDL_2$ as a function of the pumping wavelength. At 398 nm the solid curve has a slope of 0.0334. This means that the arrangement is nearly achromatic

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

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

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

$$S = \frac{\lambda_1}{\lambda_p} \frac{\Delta\lambda_p}{\Delta\lambda_1}, \quad (2)$$

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.

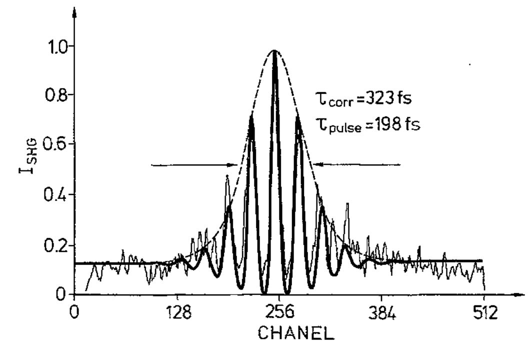

Fig. 4. Phase sensitive intensity autocorrelation of a pulse. (Heavy line: calculated, broken line: envelope, solid line: measurement). The 323 fs autocorrelation corresponds to 198 fs pulse duration assuming a $sech^2$ pulse shape

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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