4.B Generation of Pulses Shorter than 70 fsec with a Synchronously Pumped CW Dye Laser

Recently, Fork *et al.*[1] have reported on the generation of stable 90 fsec (10^{-15} seconds) pulses, by colliding pulse mode-locking with a CW pumped passively mode-locked dye laser. At LLE we have succeeded in generating laser pulses less than 70 fsec in duration with a synchronously pumped dye laser using a solution of rhodamine 6G and DQOCI in ethylene glycol. The spectral width of these laser pulses, larger than 100 Å, suggests a frequency chirp in the pulses or a limitation in our abilty to measure the real pulse width due to a restricted phase matching bandwidth and the converging beam geometry of our background-free autocorrelator. Minimum pulse widths are obtained at 615 nm when the laser reaches its optimum output power with an overall efficiency of 10%. The laser is somewhat tunable over a 590-615 nm range with an increase in pulse width. Unlike regular synchronously pumped dye lasers, no satellite pulses or coherence spikes are observed. Furthermore, because of the excellent synchronization of the short pulse with the pump pulses, this pulse can readily be amplified by a synchronously pumped dye amplifier system.[2]

A frequency doubled CW modelocked Nd:YAG laser is used to synchronously pump a four mirror-dye laser as shown in Fig. 21. A Z-cavity configuration was used to make provision for two outputs and was not essential for the generation of short pulses. A 200 μm thick jet is used with 5 cm focal length folding mirrors. It is noteworthy that the jet does not occupy any strategic position in the cavity. The dye laser is a mixture of 5x10^{4}M rhodamine 6G and 3x10^{5}M DQOCI. An output power of 30mW is obtained for 300mW pump power. A 2 μm thick uncoated pellicle tunes the laser wavelength without restricting the laser bandwidth. The pulse width measurements are performed using a background-free autocorrelator and the integrated laser spectrum is monitored with a 1/4

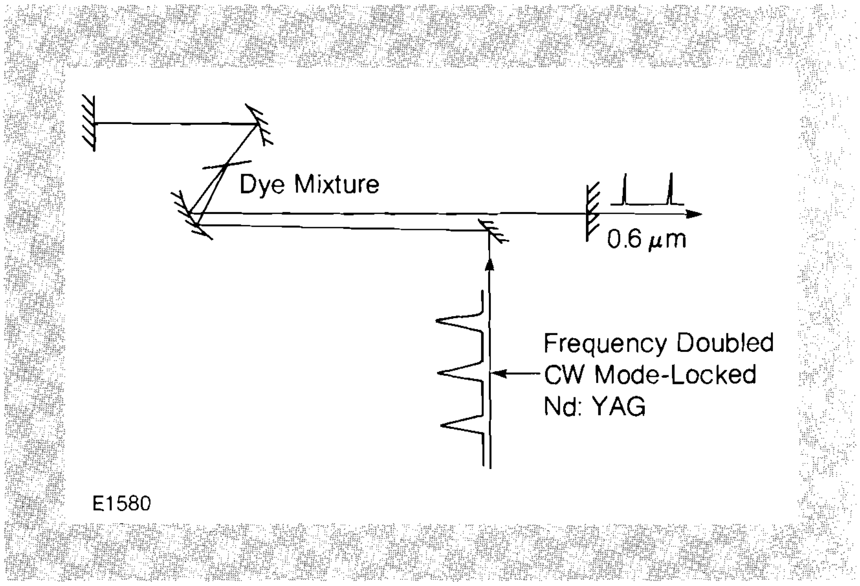

Fig. 21
Experimental set-up: a frequency-doubled CW mode-locked Nd:YAG laser pumps a dye mixture of rhodamine 6G and DQOCI in ethylene glycol in a 4-mirror cavity.

meter monochromator on an optical multichannel analyzer. Minimum pulse widths are achieved at 615 nm when the dye laser cavity length is carefully tuned to the length of the Nd:YAG laser cavity. To minimize the cavity length fluctuation, the two lasers are mounted on super invar slabs. The DQOCI[3] dye in ethylene glycol is remarkably well suited as a saturable absorber because its absorption band matches the rhodamine 6G emission band, leading to a large intensity discrimination and maximum wavelength tuning range. The dye lifetime has also been measured to be very short. Time-delay fluorimetry using a jitter-free streak[4] camera has shown that the DQOCI 1/e fluorescence time is equal to or less than 3 psec. When DQOCI is replaced by DODCI, longer pulses, around 200 fsec, are observed.

Figure 22 shows a typical autocorrelation trace suggesting a real pulse width of slightly less than 70 fsec for a hyperbolic secant pulse, which corresponds to a laser spectrum of 60 Å FWHM. The integrated spectral width is over 120 Å and suggests a restricted correlator bandwidth or a frequency chirp. This issue should be resolved with the use of KDP crystals less than 100 μm in thickness, working at a low convergence angle. The nonlinear crystal used presently in this work is 1 mm thick and exhibits a crossover length of 100 μm for converging beams. In addition to the crystal thickness which introduces a temporal broadening due to the phase matching bandwidth of the nonlinear crystal, a large waist size could lead to a significant temporal spread. For instance, a beam size of 50 μm at a 10 degree angle of convergence on the crystal, leads to a temporal spread of 30 fsec.

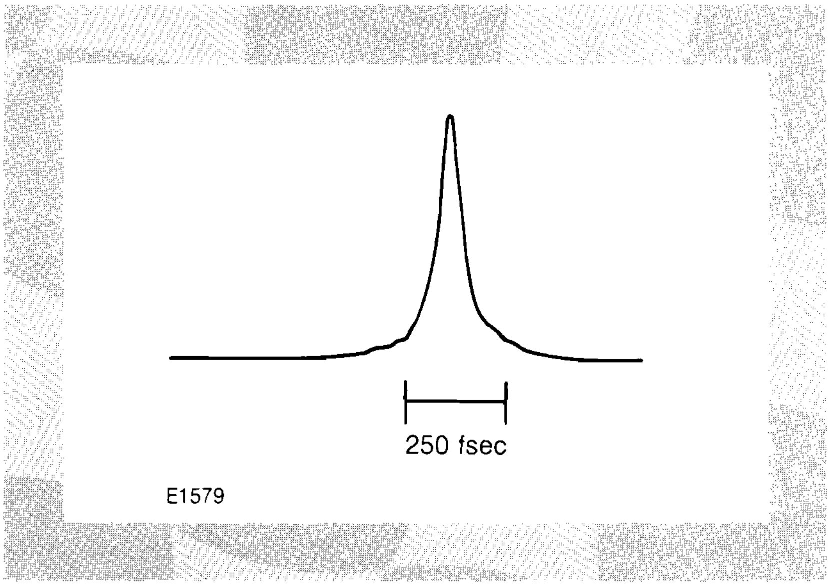

Fig. 22
Autocorrelation trace of the optical pulse. The scale has been established assuming a hyperbolic secant pulse.

In a test through dispersive media such as BK7 glass, we found that pulses starting from a different initial pulse width can be stretched or compressed according to the central frequency of the laser, suggesting a negative or positive frequency chirp across the frequency tuning range. The frequency chirp variation could stem from the interplay between the normal and anomalous dispersions in the solvent, rhodamine 6G, and DQOCI dye mixture. At frequencies corresponding to the minimum pulse width the use of dispersive glass stretches the optical pulse and seems to indicate a positive frequency chirp. The use of a grating pair outside the cavity, in an arrangement demonstrated by Treacy,[5] should help to compress the laser pulse width towards its Fourier transform limit of 30 fsec. Also, dispersion-free cavities could be built with a judicious choice of auxiliary dye molecules or metal vapors[6] working near their absorption line.

In conclusion, less than 70 fsec pulses have been generated with a synchronously pumped dye laser using a mixture of rhodamine 6G and DQOCI. The laser exhibits maximum stability and a maximum output power at minimum pulse width, reflecting the dramatic effect of the saturable absorber. The large laser spectrum and evidence of frequency chirp indicate that shorter pulses could be obtained in a dispersion-free cavity. The availability of this system at LLE will enable researchers to investigate physical processes occurring in a variety of material and biological systems in the 0.1 psec to 1.0 psec time domain.

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