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## **GENERATION AND MEASUREMENT OF 200 FEMTOSECOND OPTICAL PULSES \***

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Pulses as short as 0.2 ps have been obtained from a passively mode locked dye laser. The technique is simpler than those previously used, but one must pay the price of a loss of tunability. The pulse durations were determined from their second order correlation functions measured with a highly stable interferometer which is described.

We report the generation of optical pulses at 6120 Å with durations as short as 0.2 ps in a passively mode locked dye laser cavity. Previously, subpicosecond pulses have been reported [1, 2] using dye laser cavities containing intracavity tuning elements. The replacement of all intracavity tuning and frequency limiting elements by a narrow band, highly reflective output mirror resulted in the production of nearly bandwidth limited pulses.

A standard three mirror folded dye laser cavity was used, including the jet stream and pump beam focusing optics of a Spectra Physics model 375 dye laser. Optimal mode locking was obtained with an extended cavity length of approximately 60 cm. With a tuning wedge in the cavity, conventional mode locking was achieved by mixing the saturable absorber 3, 3'-diethyloxadicarbocyanine iodide (DODCI) with the dye laser solution (Rh6G in ethylene glycol). While a bandwidth limiting element in the cavity is essential to limit the free spectral range of the dye laser, its dispersive characteristics limit the pulse bandwidth. Hence a tuning element will set a shorter pulse duration limit, as well as influence the waveform [2, 3, 4]. The ideal tuning filter would reject all frequency components outside the spectral range of optical mode locking, but leave those components contained within that region unperturbed. Instead of such an ideal filter we propose a similar solution which consists of a high order multi-

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layer dielectric reflector, which can be manufactured to have a reasonably "square" reflection band. The third order reflector that we used had a broad reflection band centered around 1.8  $\mu$ m and a narrow band with an average reflectivity of more than 99.6% at ± 8 nm from a central wavelength of 612 nm. With such a cavity, mode locked pulses of 0.2 ps duration were measured by a second order autocorrelation technique [5] using phase matched second harmonic generation in a KDP crystal. An "autocorrelator" with a resolution of 0.03 ps was developed for these measurements, and is briefly described below.

A prism assembly as shown in fig. 1 is used to split the incoming beam  $I_1$  into two optical paths  $I_2$  and  $I_3$ . An accurate setting of the intensity ratio  $I_2/I_3 = 1$  is made possible by the use of frustrated total internal reflection beam splitters. This is accomplished by applying pressure (P) on prism 1, thereby adjusting the gap between prisms 1 and 3 within a fraction of a wavelength. Beam I2 traverses a fixed optical path in the glass structure (1-2-7), while the optical pathlength of beam I<sub>3</sub> can be adjusted with the position of the mobile prism. Beams  $I_2$  and  $I_3$  recombine at the frustrated total internal reflection interface between prisms 6 and 7, into the complementary outputs of this interferometer I<sub>4</sub> and I<sub>5</sub>. All right angles of the prisms were cut within a few seconds of accuracy. Prisms 1-2-3-5-6-7 are sintered together and glued on a flat substrate of the same glass. The error of a few seconds of arc in the cut of each prism results in beams I2 and I3 not recom-

93

Volume 25, number 1



bining exactly at right angles in the plane of the figure. This small cumulative error is compensated by a small wedged glass plate W. The only remaining angular adjustment required is the tilt of the movable prism 4 around an axis X - X in the plane of the figure. Prism 4 is mounted on a translation stage bolted to a platform which can be tilted within one half second of arc accuracy around the axis X-X by a spring pivot assembly. The mechanical range of adjustment (10  $\mu$ m to 30 mm) of the optical path difference (between beams  $I_2$  and  $I_3$ ) is complemented by pressure tuning of SF<sub>6</sub> in the space between the fixed prisms assembly and prism 4 (range 0 to  $15 \,\mu m$ ).

Background free second order autocorrelation measurements by two photon fluorescence or SHG type II can be made by inserting a  $\lambda/2$  plate in beam 3 to rotate its plane of polarization by 90°. This technique could not be used in the present situation because:

1. The energy of the picosecond pulse is too weak to make the detection of two photon fluorescence practical;

2. No crystal can be found to phase match for SHG of the second kind (crossed polarization) at 612 nm.

Since the delay between beams  $I_2$  and  $I_3$  can be easily adjusted to within a fraction of a wavelength, by varying the pressure of SF<sub>6</sub> inside the chamber containing the interferometer, the device of fig. 1 can provide the expected contrast ratio of 8 to 1 in the second order autocorrelation measurement with type I (parallel polarization) SHG [1]. It is however also possible

#### OPTICS COMMUNICATIONS

April 1978

Fig. 1. Schematic top view of the autocorrelator. The prism assembly 1-2-3-4-5-6-7 is contained in a vacuum tight chamber.

to use the interferometer to make background free autocorrelation measurements as proposed by Ippen and Shank [6]. By laterally moving the mobile prism 4 (dashed line in fig. 1), the beams  $I_5$  and  $I'_5$  will become displaced parallel to each other. Without further alignment both beams are next focused into the KDP crystal. As shown in fig. 1, the KDP crystal is phase matched for SHG along the bissectrix of the angle made by each beam. The fundamental radiation is filtered out by blue color filters, F, and the SHG generated by each beam is partially blocked by the slit, S. The result of such an autocorrelation measurement is plotted in fig. 2. The small background in the wings of fig. 2 corresponds to



Fig. 2. Second order autocorrelation measurement for a DODCI concentration of  $4.5 \times 10^{-6}$  mole/2.

### Volume 25 number 1

an equal amount of SHG generated directly by each beam. Because of the presence of this background, interference fringes could be measured. The hatched region in fig. 2 indicates the amplitude of the interferences as measured by varying the pressure of SF<sub>6</sub> in the chamber containing the interferometer of fig. 1. The minima correspond to the fields of I2 and I3 combining out of phase. The contrast ratio of the fringes to the background is close to the theoretical maximum value of 8. The fact that fringes are observed for all pulse overlaps shows that the pulses are not chirped. The FWHM of the autocorrelation curve shown in fig. 2 corresponds to a pulse duration (FWHM) of 0.24 ps assuming a gaussian shape, 0.22 ps assuming a hyperbolic secant shape.

The mode locking characteristics and pulse duration depend critically on pump power intensity, dye concentration, output mirror reflectivity and laser alignment. Mode locking occurrred only in a very narrow range of pump power above threshold. The reflectivity of the output mirror (on the average 99.6% at the center of the reflection band) was not uniform over its entire surface. Mode locking could only be observed on the spots of highest reflectivity. This leads to the conclusion that cavity dumping would be the optimal way of extracting the mode locked pulse from the cavity. In the present situation, the peak power of the pulses was approximately 10 W. Because of the critical dependence of the parameters just mentioned, the autocorrelation measurements such as that shown in fig. 2, could not be fitted to a gaussian or hyperbolic secant shape. It appeared instead that the fluctuations in pump power intensity and dye concentration made the laser emit a train of pulses of durations distributed around the measured value, thus leading to the shape shown in fig. 2.

The dependence of the pulse duration on DODCI dye concentration keeping all other parameters constant, is shown in fig. 3. The full line indicates the pulse durations assuming hyperbolic secant shaped pulses. This curve should be translated upwards by the amount indicated by the arrow, G, for gaussian shaped pulses. Mode locking started beyond a concentration threshold. The pulse duration decreased to a sharp minimum. thereafter increased rapidly as the concentration increased.

It should be noted that the duration of the pulses measured by the autocorrelator is longer than that of

## OPTICS COMMUNICATIONS



Fig. 3. Concentration dependence of the pulse duration. The curves shown are fit to the data points. The full line is the pulse duration assuming a hyperbolic secant shape. The dashed line is a correction of the previous line to account for glass dispersion.

the pulses entering the interferometer. The duration of femtosecond pulses propagating through the glass prisms will increase because of their linear dispersion. The beams I<sub>2</sub> and I<sub>3</sub> (fig. 1) propagate respectively through .114 and 102 mm of glass. The dashed curve in fig. 3 is the pulse duration (assuming hyperbolic shape), as corrected for the linear dispersion of the BK7 glass [7].

In conclusion, it has been demonstrated that pulses of 200 femtosecond duration can be generated in a single jet cavity. The stability of our system, (pump power, dye flow and mechanical) has to be improved in order to determine accurately the shape of the individual pulses. Cavity pumping techniques should be used to extract maximum power from the cavity.

We are indebted to Professor F.P. Schäfer for making available the optical delay line, designed by one of us (J.-C. Diels) while at the Max Plank Institute (Göttingen).

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April 1978

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"Generation and Measurement of 200 Femtosecond Optical Pulses"\*

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## ABSTRACT

Pulses as short as 0.2 psec have been obtained from a passively mode locked dye laser by replacing the tuning element with a narrow band high reflectivity mirror. The pulse durations were determined from their second order correlation functions.

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The mode locking characteristics and pulse duration depend critically on pump power intensity, dye concentration, output mirror reflectivity and laser alignment. Mode locking occurred only in a very narrow range of pump powers above threshold.

The dependence of the pulse duration on DODCI dye concentration keeping all other parameters constant, is shown in figure 1. The full line indicates



Figure 1. Concentration dependence of the pulse duration. The curves shown are fit to the data points. The full line is the pulse duration assuming a hyperbolic secant shape. The dashed line is a correction of the previous line to account for glass dispersion.

the pulse durations assuming hyperbolic secant shaped pulses. This curve should be translated upwards by the amount indicated by the arrow, G, for Gaussian shaped pulses. Mode locking started beyond a concentration threshold after which the pulse duration decreased to a sharp minimum, thereafter increased rapidly as the concentration increased.

It should be noted that the duration of the pulses measured by the interferometric autocorrelator is longer than that of the pulses entering it. Because it is made of glass prisms, the duration of femtosecond pulses propagating through the autocorrelator will increase because of linear dispersion. The beams in each arm of the autocorrelator propagate respectively through 114 and 102 mm of glass. The dashed curve in figure 1 is the pulse duration (assuming a hyperbolic secant shape), as corrected for the linear dispersion of the BK7 glass.<sup>6</sup>

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