A Subpicosecond Tunable Ring Dye Laser and Its Applications to Time-Resolved Fluorescence Spectroscopy

D. ROSEN, A. G. DOUKAS, Y. BUDANSKY, A. KATZ, AND R. R. ALFANO

Abstract—**The design and operation of a subpicosecond CW passively mode-locked dye laser is described. The laser is tunable, stable, and operates at a low power threshold (below 1 W). The use of this laser in a new time-resolved fluorescence technique is also described.**

I. Introduction

THE generation of ultrashort light pulses through the use of mode-locking techniques [1] has opened up a new era for time-resolved spectroscopy [2]. The dye laser was first mode locked by Dienes *et al.* [3]. A variety of cavities have been developed by different research groups to achieve subpicosecond pulse generation [4]-[6]. Recently, Shank and his collaborators [7] have reported on a ring cavity which has shown greater stability, easier alignment, and fewer satellite pulses than previous cavity designs. This paper describes a stable, mode-locked ring cavity which operates at a very low threshold, less than 1 W, and which is wavelength tunable over a limited range. This laser has been utilized for time-resolved fluorescence measurements using a population mixing technique.

II. Description of the Laser

The schematic of the ring laser is shown in Fig. 1. The rhodamine 6G (Rh-6G) jet stream is located between the mirrors 7 and 8, with radii 10 cm. The 3,3′-diethyloxadicarbocyanine iodide (DODCI) jet stream is located between the mirrors 3 and 4, with radii 5 cm. Three other flat mirrors (1, 2, 5) complete the cavity. All mirrors are wedged 1° and are of laser quality (flatness $\lambda/10$, scratch-dig 10-5). Mirrors 2-5 and 8 have dielectric coatings of reflectivity R greater than 99.9 percent with low scatter loss in the range 590-630 nm. Mirror 7 is a broad-band reflector with R greater than 99.9 percent in the region 500-630 nm, with low scatter loss. The output coupler (mirror 1) has a typical transmission T of three percent. A second coupler has also been used with a T of six percent for higher power output. The mirror substrates were purchased from Special Optics and coated by either Trans World Optics or Airtron. All mirrors are mounted on Klinger mounts. Both jet stream nozzles are stainless steel, (Coherent Radiation) point downwards, and are set at Brewster's angle. The thickness of the jets at the lasing spot was measured to be 150 μm. The Rh-6G concentration is set so that transmission of the 514.5 nm argon beam is fifteen percent. Ethylene glycol solutions of each dye were used, where the dyes were first dissolved in small amounts of methanol. The concentration of the DODCI at 585 nm is set at 3 OD/cm. The flow rate in the two jets is adjusted to 1.5 l/min, which corresponds to a bulk velocity at the jet of 25 m/s. Both the dyes are kept at room temperature using a cooling chamber. Modified Spectra-Physics pumps are used to flow the dye solutions (model 376). All components are stainless steel in order to avoid dye deterioration [8]. The Brewster prism (element #6) provides the capability of tuning the wavelength of the laser. The Rh-6G jet is pumped by an argon ion laser at 514.5 nm. A $\lambda/2$ retardation plate in the laser path (element #9) changes the polarization of the argon beam to "*P*," which coincides with the polarization of the dye laser. When the polarizations of the two lasers match, the threshold of the dye laser is 0.9 W. Normally, the polarization of the argon laser is "*S*." In this case the threshold is over 3 W. The dramatic decrease in the threshold cannot be explained only by the decrease in the reflection off the Rh-6G jet stream, which is on the order of fifteen percent. This decrease in threshold power may arise from the anisotropic absorption and gain cross sections of the Rh-6G molecules, which result in a more efficient use of the pumping energy.

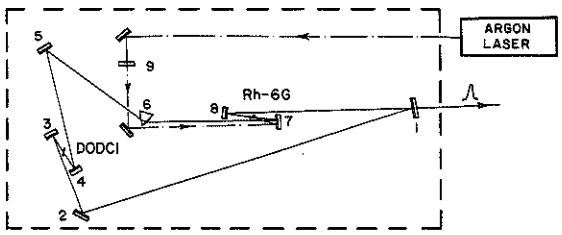

Fig. 1. Schematic of the subpicosecond ring dye laser. The components are explained in the text. The cavity dimensions and angles are L_{12} (distance between elements 1 and 2) = 78 cm, L_{23} = 16 cm, L_{34} = 5 cm, L_{45} = 28 cm, L_{56} = 30 cm, L_{67} = 25 cm, L_{78} = 10 cm, L_{81} = 50 cm, $\measuredangle 456 = 30°$, $\measuredangle 812 = 14°$, $\measuredangle 123 = 85°$.

The laser produces pulses of 0.3-0.5 ps at the rate of 125 MHz. To measure the duration of the pulses the autocorrelation SHG technique was used [9]. A typical autocorrelation function from a 1 cm thick KDP crystal is shown in Fig. 2. Assuming a sech^2 pulse the pulse duration was estimated to be 0.36 ps. The bandwidth has been (FWHM) measured to be 1.5 nm. The pulses are very close to being bandwidth limited. However, the prism is not the limiting factor of the pulse duration. A calculation of the prism-limited bandwidth

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The authors are with the Picosecond Laser and Spectroscopy Laboratory, Department of Physics, City College of New York, New York, NY 10031.

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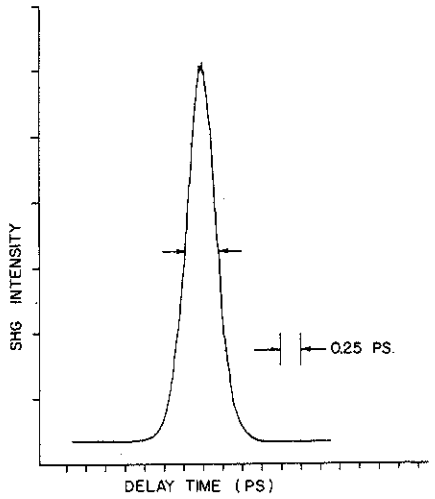

Fig. 2. A typical autocorrelation of the pulse taken by means of a shaker. Assuming a sech^2 pulse, the pulse duration (FWHM) is 0.36 ps.

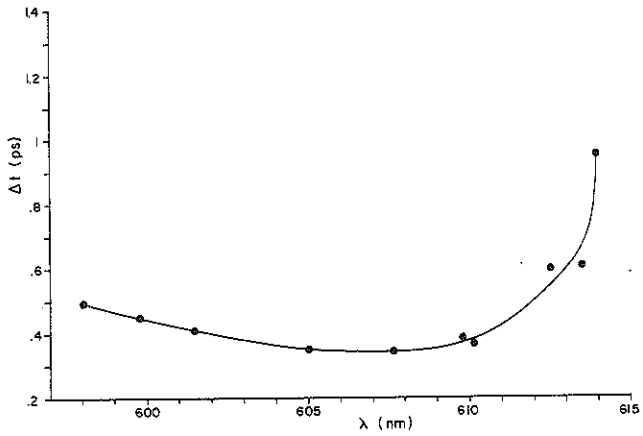

Fig. 3. Pulse duration as a function of the operating wavelength at 3 OD/cm of DODCI.

FWHM gives a value of ~6 nm, which is much greater than the actual bandwidth of the pulses (1.5 nm). The limiting factor is probably in the dispersion within the dye jets. Fork *et al.* [5] have recently produced pulses as short as 0.09 ps using a 10 μm jet for the DODCI dye. Therefore, it is possible to shorten the duration of the pulses and still retain the tunability by replacing the present jet by a thinner one.

The addition of a thin etalon in the laser cavity can decrease the bandwidth with a corresponding increase in the pulse duration. When a 100 μm etalon was added the pulse duration increased to 6 ps with a resolution-limited bandwidth of less than 0.1 nm. Therefore, one can make various combinations of pulse duration and bandwidth to suit the requirements of a particular application by using an intracavity etalon of appropriate thickness. The laser is isolated from air currents.

The laser is tunable at 3 OD/cm over a range of 595–615 nm. The pulse duration as a function of the wavelength of the laser pulses is shown in Fig. 3. The pulse duration remains below 1 ps except in the long wavelength region where it increases rapidly, perhaps due to the low absorption of the DODCI in this region. By increasing the DODCI concentration, the tunability range has been shifted to longer wavelengths, from 600 to 620 nm. Overall the laser is quite stable and it can operate for weeks with only minor adjustments.

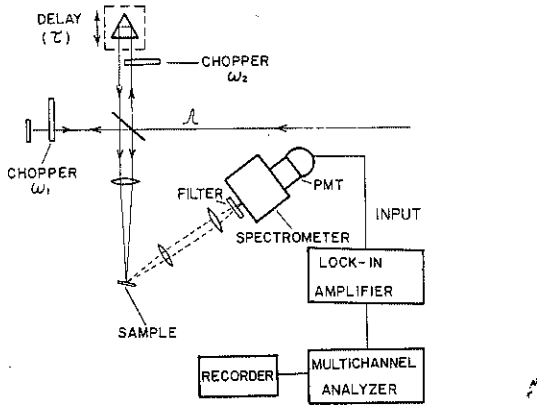

Fig. 4. Subpicosecond fluorescence time-resolved apparatus using the population mixing technique.

III. Picosecond Population Mixing Technique

We have recently developed a new technique for time-resolved fluorescence spectroscopy without the necessity of a multiple-stage amplifier and a streak camera. The method, called population mixing technique allows one to build a subpico-generated carrier densities. Fig. 4 shows a schematic of the apparatus. The pulse train is divided by a beam splitter into two pulse trains which are modulated at frequencies ω_1 and ω_2, respectively. A variable delay τ is added to one of the beams. Both beams are focused by a lens onto the sample. The fluorescence is collected and focused on the slit of a monochromator and detected by a photomultiplier and a lock-in amplifier, set at the difference frequency $|\omega_1 - \omega_2|$. In this way only the fluorescence signal produced by the two pulse trains is detected. The two pulses create free carrier populations $n(t)$ and $n(t+\tau)$, respectively. The detected signal is

$$I_s(\tau) \propto \int_{-\infty}^{\infty} n(t)n(t+\tau)\,dt. \tag{1}$$

If a single exponential decay time T of the carrier is assumed, we obtain

$$I_s(\tau) \propto \exp(-|\tau|/T). \tag{2}$$

The correlation function provides information about the relaxation of the photogenerated carriers. The method has subpicosecond time resolution limited only by the duration of the pulse. Using this method we have measured the relaxation time of 40 ps photogenerated carriers in highly doped p-GaAs [10].

IV. Conclusion

The ring laser we have described in conjunction with the population mixing technique allows one to build a subpico-second luminescence apparatus without the need for a high

power argon laser, dye laser amplifier, and streak camera. The technique can measure weak transient fluorescence signals with a subpicosecond time resolution.

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