Active-Passive Mode Locking of a Flashlamp-Pumped Dye Laser

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Abstract—**A simple dye laser system for obtaining relatively high energy, synchronized, picosecond pulses is described. The novel combination of active and passive mode locking within a single flashlamp-pumped dye laser cavity produced 2.5 μJ, sub-10 ps pulses capable of being synchronized to an external event with better than 100 ps accuracy. The single pulse energy was subsequently increased to 40 μJ by using a larger twin flashlamp-pumped oscillator.**

Introduction

To an ever increasing degree, picosecond laser pulses are being utilized to generate or diagnose transient phenomena in many diverse fields. A common requirement of such studies is the generation of a single pulse, or a train of single or multifrequency pulses, in precise synchronism with an external event. This demand has been apparent for some time in the laser-fusion field where an auxiliary, intense, picosecond pulse, generally of short wavelength, is required for optical [1] or X-ray [2] probing of the plasma that is produced. For plasmas created by nanosecond CO_2 lasers, this problem has been addressed by the development of synchronizable actively mode-locked solid-state lasers, either Nd:glass [3] or ruby [4], and more recently, Nd:phosphate [5]. These systems, while adequately fulfilling many demands, suffer in one principal respect in that they typically produce pulses of $\gtrsim$50 ps duration, limited by either the spectral bandwidth or the build-up time in which effective pulse shortening can occur. Some applications, including laser-plasma diagnosis, require pulses of considerably shorter duration. To this end, we have developed a combined active-passive, mode-locked, flashlamp-pumped dye laser system capable of providing pulses of 250 kW power and <10 ps duration with external synchronism to better than 100 ps [6]. Although dual-modulation techniques have been used previously with flashlamp-pumped neodymium [7]-[10] and ruby [11] lasers, either for external synchronism or improving the pulse stability, this paper reports the first time that these techniques have been incorporated into a flashlamp-pumped dye laser system. Similar methods have been applied to CW dye lasers resulting in sub-picosecond pulses [12], but their peak powers are typically <1 kW, insufficient for many optical diagnostic applications.

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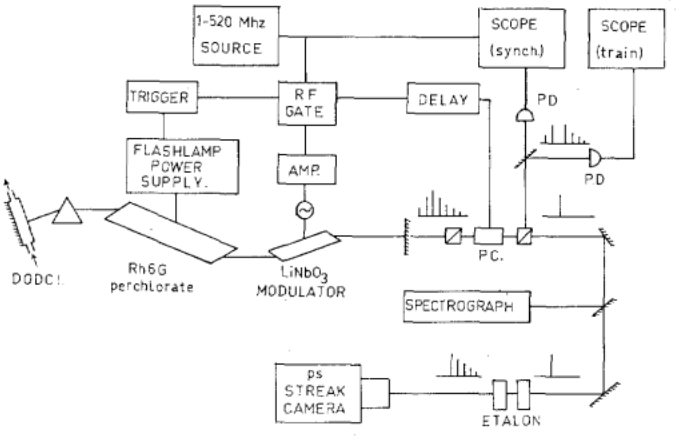

Fig. 1. Schematic diagram of the experimental layout.

Experimental Details of the Laser System

The design of the laser system and the principal diagnostics used to monitor its output are shown schematically in Fig. 1. The oscillator was comprised of a flowing-dye cuvette, a prism wavelength tuning element, an active modulator, and a passive mode-locking dye cell in a linear resonator of optical path length ~100 cm, formed by a 99.5 percent reflecting plane mirror and a partially transmitting (typically 85 percent) concave mirror of radius of curvature 2 m. The 76 mm long, 4 mm internal diameter flowing-dye cuvette contained a 1×10^{-4} M $\cdot l^{-1}$ concentration of Rh6G perchlorate in ethanol, and was optically pumped in a polished, focused elliptical cavity by a single linear flashlamp having a discharge time of ~4 μs, dissipating an energy of ~80 J at 12 kV. The 1 mm thick saturable absorber dye cell, in optical contact with the plane resonator mirror, contained a solution of DODCI in ethanol with a concentration that was varied between 1×10^{-6} and 1×10^{-4} M $\cdot l^{-1}$.

The active mode-locking element consisted of a 3 mm thick rectangular-sectional Brewster-ended $LiNbO_3$ electrooptic crystal (40 mm long, 9 mm wide) with crystal axes oriented to utilize the transverse electrooptic effect. In this mode, the crystal had a half-wave voltage $V_{\lambda/2}$ at 606 nm of 460 V. To prevent any thermally induced damage to the crystal, the 1 V CW RF drive from the signal generator was gated before amplification, producing a 100 μs burst of modulating voltage.

Great care was taken initially to maximize the RF voltage

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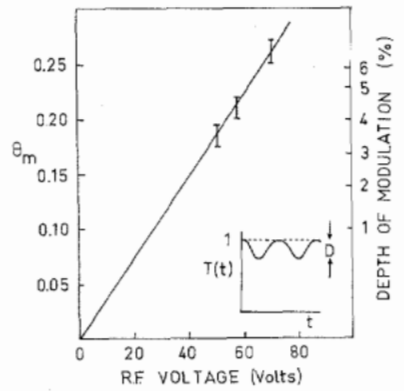

Fig. 2. Relationship between the depth of modulation D and θ_m and the deduced RF voltages that were applied. The inset shows how D was defined.

generated across the $LiNbO_3$ crystal, applied transversely to the optic axis, by making the crystal part of an L-C-R resonance circuit which had a maximum impedance at the required driver frequency (76 MHz). However, it was subsequently found that a depth of modulation of only a few percent was sufficient to stabilize the output. In fact, large RF voltages led to a deterioration in the output affecting both the mode-locked pulse train amplitude and duration, as if acoustic scattering of the beam were taking place. A separate experiment was carried out, with the $LiNbO_3$ crystal external to the cavity and no DODCI in the saturable absorber cell, to determine how the depth of modulation varied with applied RF voltage. This method provides exact knowledge of the depth of modulation. Any simultaneous electrical measurement of RF voltage is less exact, as the resonance circuit will be perturbed by the probe itself. The results, which can be seen in Fig. 2, were limited by measurement resolution of D at lower voltages. The inset in Fig. 2 shows how the depth of modulation was defined to be

$$D = 1 - \cos^2 \theta_m \tag{1}$$

with the transmission function being

$$T(t) = \cos^2 (\theta_m \sin 2\nu\pi t) \tag{2}$$

where $\theta_m = \theta/2 \cdot V/V_{\lambda/2}$ and ν is the frequency of the applied RF signal. The good linear fit to values derived using (1) and (2) at the higher voltages leads to confidence in extrapolating the values of D (or θm) from the lower end of the graph. The output performance of the laser system was characterized with the diagnostics shown schematically in Fig. 1. Single pulse selection from the output train was achieved with a KD*p electrooptic Pockels cell gate, activated after a variable delay. The contrast ratio was 1500:1 per pulse (5:1 in energy). The time of switch-out during the pulse train was monitored for each shot with an S-20 photodiode. The spectral content of the single pulse was analyzed by a 1 m Czerny-Turner spectrograph with a spectral resolution of 10 Å/mm. The spectra were recorded on Kodak Tri-X film.

The pulse duration of the isolated single pulse was measured using a variety of picosecond streak camera arrangements. Figs. 3 and 5 were recorded with an S-20 sensitive Picotron 100 streak camera [13] without and with the aid of a channel

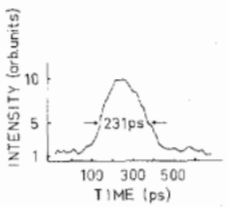

Fig. 3. Pulsewidth when only active mode locking was applied.

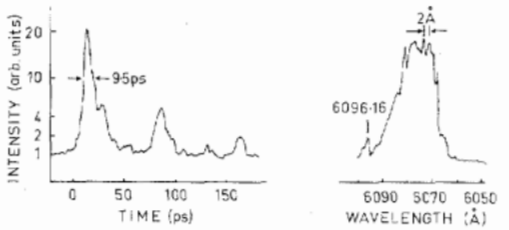

Fig. 4. (a) Pulsewidth when active and passive mode locking were applied. (b) Simultaneous single pulse spectrum.

plate intensifier, respectively. Fig. 4(a) was recorded utilizing an S-1 sensitive Photochron II streak camera with an intensifier. The instrumental time resolutions at which the systems operated for the above traces were 120, 4.5, and 3.5 ps, respectively. The switched-out laser pulse was passed through an etalon to generate the series of pulses seen in the figures. All of the streak records were fiber optically coupled onto Kodak 2475 film developed to an ASA exposure rating of 1500 and densitometered using an aperture of 100 μm × 1 mm.

The average single pulse energy of the above system was estimated to be about 2.5 μJ. By increasing the oscillator dye cuvette length to 250 mm, retaining a 4 mm diameter, and utilizing a double-focused elliptical cavity with two linear flashlamps [6], the single pulse energy was increased to ~40 μJ when pumped with an electrical discharge energy of ~130 J at 16 kV for a period (FWHM) of 4 μs.

Mode-Locking Characteristics

A. Active Mode Locking

With an applied RF voltage of 200 V ($\theta m = 0.44$), long, stable trains consisting of scope-limited pulses were readily produced. However, subsequent streak camera measurements showed these to be only 200–250 ps in duration. An example can be seen in Fig. 3. For AM modulation, one would expect the pulsewidth to be, after M roundtrips [14],

$$\tau_p = \underset{\sim 250 \text{ ps}}{\frac{\ln 2}{2}} \cdot \frac{1}{\pi} \cdot \frac{1}{\theta_m \sqrt{M f_m}} . \tag{3}$$

Agreement with (3) is sufficiently close to show the futility of pursuing very short pulse generation by active mode locking alone in a flashlamp-pumped system. Such a conclusion was also reached when CW lasers were used [15], [16].

B. Active-Passive Mode Locking

A gradual decrease in the duration of the individual pulses from more than 250 ps through less than 10 ps was accomplished by including progressively more DODCI in the satu-

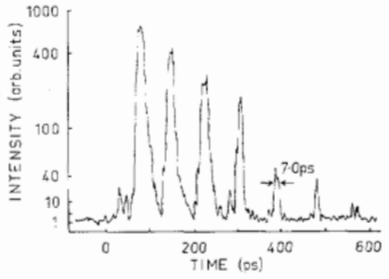

Fig. 5. Pulsewidth when only passive mode locking was applied.

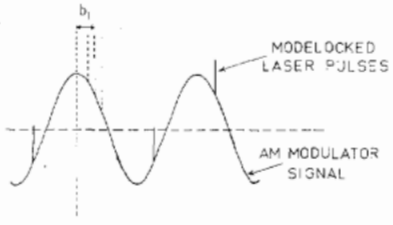

Fig. 6. Schematic diagram illustrating how the jitter was determined.

rable absorber cell, up to a maximum concentration of approximately 1×10^{-4} M $\cdot$ l^{-1}. Changing the passive mode-locking dye concentration also changes the laser spectrum at which maximum output occurs, with the wavelength increasing for increasing absorber dye concentrations. With no saturable absorber, the wavelength was centered around 582 nm and at the highest concentrations it extended beyond 610 nm. The optimum concentration (5×10^{-5} M/l) was found to be similar to that required for peak passive mode-locking performance. Greater concentrations than these led to substantial reductions in the overall pulse train amplitude and duration as the linear losses are increased. Single pulses of less than 10 ps duration were readily produced [see Fig. 4(a)]. The spectrum associated with this particular pulse, which was recorded simultaneously, is shown in Fig. 4(b). Subsequent analysis of the spectrum revealed a 2 Å modulation, which was attributed to the uncoated, planar, parallel surface of the front window of the saturable absorber cell.

C. Passive Mode Locking

Later work with passive mode locking alone incorporated a wedged saturable absorber cell to avoid multiple reflections. Utilizing the thinner side of this wedged cell gave rise to pulses substantially less than 10 ps (see Fig. 5), with camera deconvoluted half-widths of ~5 ps; the pulse durations were still apparently being governed by the width of the saturable absorber cell.

Measurement of Synchronism

The jitter in the synchronism of the mode-locked pulse train, as well as the actual level of the RF drive, was measured by splitting the RF signal, feeding part to the crystal and the rest to an oscilloscope together with the photodiode signal monitoring the rejected pulse train. A schematic diagram of the latter can be seen in Fig. 6. From an arbitrary reference point in the RF signal (here, for example, the voltage peak), n measurements of the relative delay b_i can be made. The jitter is then

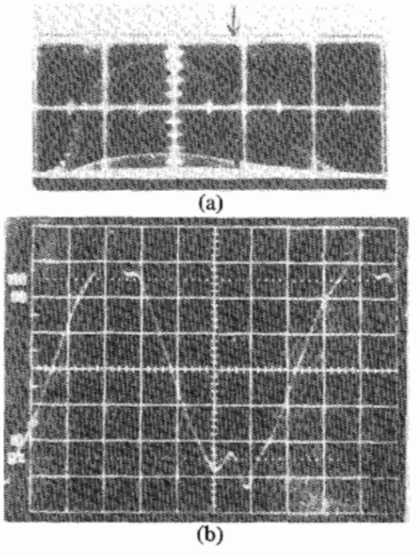

Fig. 7. (a) Typical mode-locked pulse train. The arrow indicates position of switched-out pulse. Timescale: 200 ns/div. (b) Superposition of a fraction of the above photodiode response with a part of the modulator RF drive voltage. Timescale: 2 ns/div.

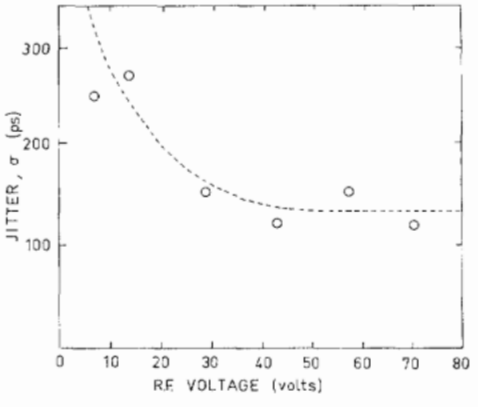

Fig. 8. Jitter as a function of applied RF voltage.

$$\sigma = \frac{\sum_{i=1}^{n} |b_i - b_0|^2}{n - 1} \tag{4}$$

where

$$b_0 = \frac{\sum_{i=1}^{n} b_i}{n} .$$

An example of the actual signals can be seen in Fig. 7. No attempt was made to ensure that the delay between the peak of the RF signal and the arrival time of the mode-locked laser pulses was calibrated in absolute terms. However, the relative timing of the fraction of the mode-locked train displayed on the oscilloscope remained constant.

For twenty consecutive shots at each of six RF voltages, the root mean square jitter was determined according to Fig. 6 and (4). The results are plotted in Fig. 8. The resolution limit was simply determined by the width of the trace recorded on polaroid film at a sweep speed sufficiently low to still provide clearly focused images (2 ns/div 7904 Tektronix). That the

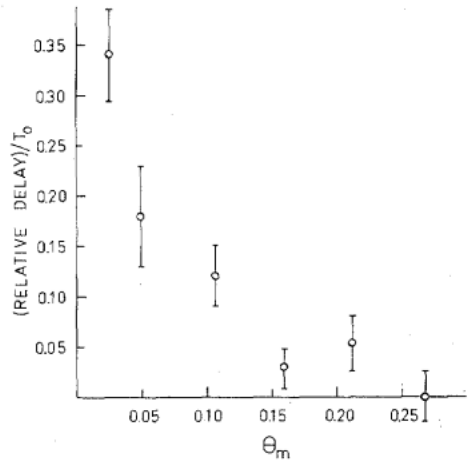

Fig. 9. Relative delay of the pulse, normalized to the interpulse separation, as a function of the modulation index θ_m.

statistics seem to saturate just above the estimate in measurement error of ~100 ps leads to confidence in suggesting that the jitter itself is considerably less than the quoted value of 100 ps.

Phase Shift Dependence on Depth of Modulation

Analysis of the data for the previous section revealed a relative shift in the time of arrival of the mode-locked laser pulses, with respect to the phase of the RF voltage, as the amplitude of the RF voltage was varied. This effect has not been observed previously. The relative phase shift, normalized as a fraction of the interpulse separation, is plotted as a function of applied depth of modulation in Fig. 9. Although such a delay is expected implicitly according to the model of Haus and Dyckman [17] which considers the case of active-passive mode locking, it has recently been explicitly predicted in a theory by Piché and Bélanger [18] that considers the case of active modulation alone. However, perhaps due to the effects of the saturable absorber, the experimentally observed delay would seem to vary more rapidly with θm then expected according to [18]. Also, the degree of absolute agreement cannot be verified as no temporal fiducials were employed.

Conclusions

Having proved the principle of combining active and passive mode locking in a flashlamp-pumped dye laser, and showing it to be a simple means of obtaining synchronizable picosecond pulses of relatively high energy and low jitter, a larger system was constructed capable of providing more than 40 μJ in a single pulse, more than sufficient for optical probing of plasmas [19]. Analysis of the results revealed the novel observation of pulse drift with variation in applied depth of modulation. Such a drift can be significant if varying values of the RF voltage are used. In fact, varying the applied RF voltage can lead to an in-line method of delaying the pulse relative to any external event without the need for separate electrical or optical delays. With a quasi-CW laser system, time histories of repetitive events could be followed by applying a ramp RF voltage.

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Substituent and Environmental Effects on the Picosecond Lifetimes of the Polymethine Cyanine Dyes

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Abstract—**Using the synchronously operated streak camera, the fluorescence lifetimes of polymethines of the cyanine family have been examined under low power CW picosecond dye laser excitation. The effects of steric hindrance on the lifetimes and of atomic substitution within the dyes have been measured, with lifetimes ranging from 14 ps to ~2 ns. The strong viscosity-dependent fluorescence lifetime exhibited by these dyes has also been investigated. In addition, deviation from a single exponential decay was recorded for several of the dyes.**

INTRODUCTION

SINCE the initial discovery of the cyanine dyes [1], considerable research has been carried out to characterize their properties. Primarily, interest was concentrated on their application to spectral sensitization in photography [2]. Various measurements were taken to correlate trends in the absorption and fluorescence profiles associated with dye structure [3], [4], in particular, to relate the shift in the absorption spectrum to the lengthening of the polymethine chain and to atomic substitution within the dye [5]. Fluorescence quantum yield measurements demonstrated a strong structural dependence, and steric hindrance within the molecules also contributed a major effect [6]. The general chemical effects of the cyanines are well documented in the review by Sturmer [7].

Interest was further directed towards the polymethine dyes as potential sources of highly tunable radiation after laser action was reported in several species [8]. Although usually laser pumped, flashlamp pumping is possible with laser action over the range of 690–1100 nm [9]–[11] with the polymethine dyes. However, a further important application followed the observation that 1,1′-diethyl-4,4′-carbocyanine iodide [12] and related dyes [13] acted as saturable absorbers with which the ruby laser could be passively mode locked to produce trains of picosecond pulses. Similarly, 3,3′-diethyl

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