Appl. Phys. B 29, 191–193 (1982)

Applied Physics B Photophysics and Laser Chemistry



# Tunable Picosecond Pulse Generation from the Passively Mode-Locked Coumarin 6 Dye Laser

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Received 2 August 1982/Accepted 17 August 1982

**Abstract.** Using the saturable absorber 2-(p-Dimethylaminostyryl)-benzothiazolylethyl iodide, coumarin 6 has been passively mode-locked for the first time to give fully modulated trains of pulses of ~4 ps duration and with peak powers of ~3 MW tunable over the spectral range 526–547 nm.

**PACS:** 42.55 Mv, 42.60 Fe

The simplest source of frequency-tunable picosecond pulses is provided by the passively mode-locked flashlamp pumped dye laser [1, 2]. Although many suitable saturable absorbers have been identified [3], few reports have been given of passive mode-locking of dye lasers in the green [4–6] and green-yellow [7] spectral regions. In this communication, we report for the first time, on the passive mode-locking of coumarin 6 for the generation of tunable picosecond pulses over the spectral range 526–547 nm.

A $2 \times 10^{-4}$ M ethanolic solution of coumarin 6 was continuously flowed via an in line 2 μm pore sized filter, through a 127 mm long (3.5 mm i.d., 9 mm o.d.) dye cell, which was placed along the common focal axis of a standard double elliptical cylindrical cavity head, with semi major and semi minor axes of 49 mm and 45 mm, respectively. Excitation of the dye was achieved using 100 Torr Xe filled flashlamps of 4 mm i.d. bore (9 mm o.d.) and an overall bore length of 127 mm. Brewster-angled quartz block windows were "O" ring sealed to the ends of the dye cell and wedged optical components were used throughout the cavity. A maximum electrical energy of 100 J could be deposited into each lamp from a 0.5 μTF capacitor via a common ceramic-metal triggered spark gap, and the resulting excitation light pulse had a full width at half maximum of 2 μs. The dye laser cavity was formed by plane dielectric coated mirrors of 100% and 85% reflectance over the spectral range 470–550 nm, placed 35 cm apart. An intra-cavity 3.5 μm air spaced Fabry Perot etalon permitted wavelength selectivity. The saturable absorber was placed in an optically contacted dye cell of 500 μm thickness in contact with the 100% reflector and static solutions of the saturable absorber were used, only being renewed after very many laser shots. No attempt was made to separately temperature stabilize the active or passive dyes.

The saturable absorber exclusively used in the work reported here was 2-(*p*-Dimethylaminostyryl)-benzothiazolylethyl iodide (DASBTI), the molecular structure of which is shown in Fig. 1. This dye in ethanolic solution exhibits an absorption maximum at 530 nm with a cross section $\sigma_{max}$ of $1.1 \times 10^{-16}$ cm$^2$ ($\varepsilon = 2.75 \times 10^4$ l mol$^{-1}$ cm$^{-1}$) at this wavelength. Measurements of the fluorescence recovery time of the dye have shown a strong viscosity dependence with measured lifetimes of 55 ps in pure ethanol and 212 ps in a 1:1 solvent mixture of ethanol and glycerol [8]. The intracavity power and parameters of the dye was such that saturation occurred for ethanolic solutions of the saturable absorber and these were used throughout. Typically, the concentration of the saturable absorber was varied in the range

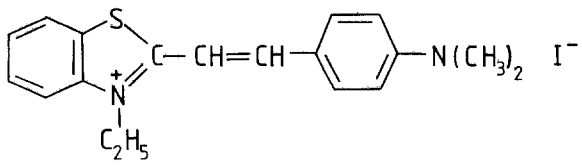


Fig. 1. Molecular structure of the saturable absorber DASBTI (see text)

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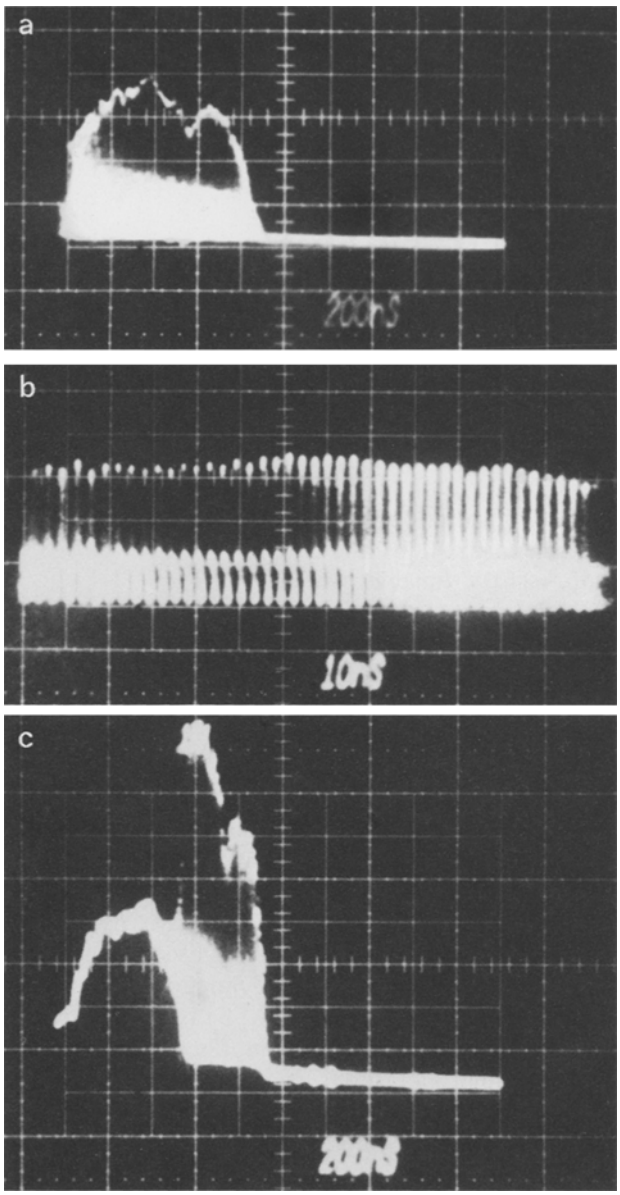


Fig. 2a–c. Temporal profiles of the mode-locked pulse trains from the coumarin 6 dye laser (a) tuned to operate at 526 nm on a timescale of 200 ns/small division, (b) as in (a) only 10 ns/small division, and (c) with no intracavity tuning element on 200 ns/small division

$5 \times 10^{-5}$ M – $2 \times 10^{-4}$ M for optimum output, depending on the wavelength of operation. Figure 2a shows a representative oscilloscope trace of the mode-locked output from the coumarin 6 laser tuned to 526 nm. The output trains were stable and reproducible. Depending on the operational wavelengths, a build up time to complete modulation was present in the trains. For example, at 540 nm, this required 150–200 ns. Generally the period of full modulation lasted 800 – 900 ns, as can be see in Fig. 2a. From Fig. 2b, it

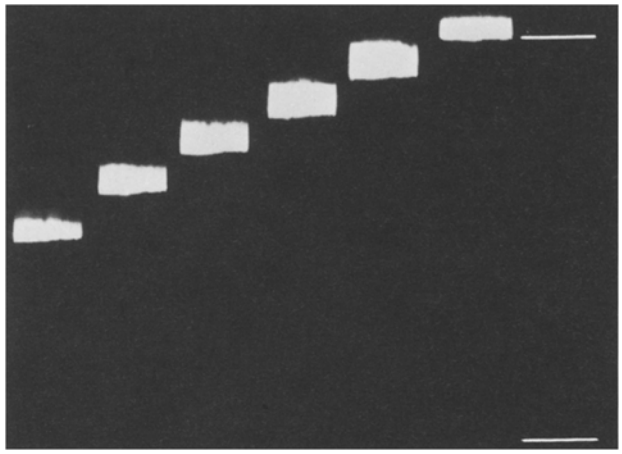

Fig. 3. Typical tuning range of coumarin 6 mode-locked with DASBTI, calibrations lines are at 507.3 and 546.1 nm, wavelength on vertical axis

can also be seen that to within the resolution obtainable with the scope-diode combination, a modulation depth of 100% and single pulse production was achieved. At maximum, typically the energy in a complete mode-locked train was 1–2 mJ which corresponded to an average energy per pulse of 3–6 μJ. This depended on operational wavelength and decreased towards the wings of the tuning range. Figure 3 shows the range of the tuning obtained with the etalon used, and can be seen to extend from 526–547 nm, calibration lines at 507.3 and 546.1 nm are also included in Fig. 3.

Passive mode-locking could also be achieved without the presence of the tuning element and Fig. 2c shows a typical output pulse profile under these condition. A much longer time to complete modulation was evident (~600 ns) with a fully modulated period of ~400 ns and the laser tended to operate at longer wavelengths and with a wide lasing spectrum. In Fig. 2c the central lasing wavelength was at 535 nm. A coarse tuning of the lasing frequency was possible by variation of the saturable absorber concentration and operation was similar to that of Rhodamine 6 G mode-locked with DODCI [9].

To determine the duration of the individual mode-locked pulses a Photochron II electron-optical streak camera with an S-20 photocathode was used. The standard experimental arrangement was used both to provide calibration and detection of the pulses [10]. Synchronization of the Krytron deflection circuitry was such that pulses 100–150 ns from the commencement of modulation were examined, by which time single-pulse evolution should have taken place in a passively mode-locked dye laser [11]. Figure 4 shows a microdensitometer trace of a recorded pulsewidth of 5.5 ps for the laser tuned to operate at 530 nm. The

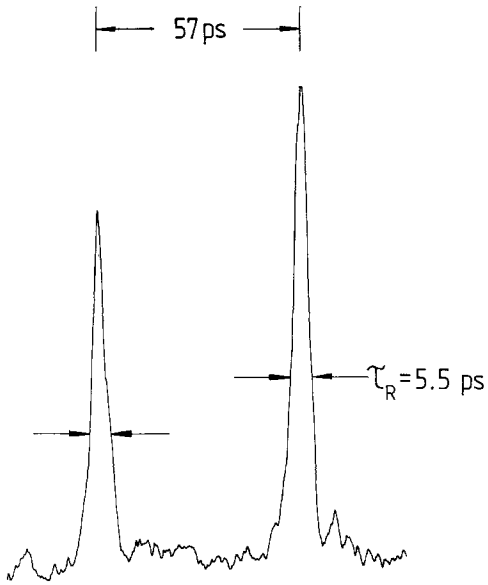


Fig. 4. Microdensitometer trace of streak camera recorded pulsewidths of 5.5 ps from the passively mode-locked coumarin 6 laser operating at 530 nm

second pulse delayed by 57 ps is for calibration purposes. Interpulse noise is slightly high due to the fact that the complete mode-locked train was incident on the streak camera photocathode and as a consequence, scattering of photoelectrons internally in the image tube gave rise to an increased background noise. The overall time resolution of the camera at a sweep speed of $6 \times 10^9$ cm s$^{-1}$ and at 530 nm is 3 ps. Deconvolution of the recorded pulsewidth $\tau_R$ of 5.5 ps in Fig. 4 would indicate an actual pulsewidth of ~4.5 ps, which is a fair representation of the pulses generated throughout the complete tuning range, although some as short as the camera resolution were recorded. Measurements of the generated pulsewidths in the absence of the Fabry-Perot were also in the range 4–6 ps.

In conclusion, we have shown for the first time passive mode-locking of the coumarin 6 dye laser, tunable from 526–547 nm with the production of pulses ~4 ps in duration with powers ~3 MW, in highly reproducible and stable pulse trains. Direct application to cw operation should be possible to produce subpicosecond pulses in this wavelength range, and this is at present under investigation.

*Acknowledgements.* The overall financial support for this work by the SERC is gratefully acknowledged.

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