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# Simple Generation of 400–700 nm Picosecond Dye Laser Pulses with Nanosecond Laser Pumping

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**Abstract.** New results in the experimental study of the spectro-temporal selection (STS) method to produce picosecond dye laser pulses are presented. Adjustability of the picosecond pulse wavelength, possibility of extension of the STS method to different dyes and to UV pump wavelength, stability of the output pulse duration and intensity, and the concentration effect on pulse duration, are reported for the first time. From these results, production of high power picosecond (50–100 ps) dye laser pulses spectrally adjustable between 400 and 700 nm is obtained with a standard nanosecond pump laser, in a compact and simple device.

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Dye lasers have become multipurpose light sources over the last 25 years in a wide range of applications; in particular ultrashort dye lasers are now widely employed [1]. Production of ultrashort pulses at desired wavelengths is generally based on mode locking techniques and often requires the combined use of two expensive pump lasers, one cw, one pulsed. However, different ways to produce short pulses with a (non-mode-locked) nanosecond pump have been proposed using the processes of cavity transients [2], cavity quenching [3], distributed feedback [4], and spectro-temporal selection (STS) [5, 6]. Thanks to the enlarged choice of pump lasers and noticeable simplifications offered by these methods, much attention was paid to their development.

The STS method of producing picosecond dye laser pulses was first reported in 1985 and successfully operated with rhodamine 6G dye pumped at 532 nm [5]. The principle of the method is to select with an extra-cavity filter a narrow spectral band in the short-wavelength side of the broadband laser output of an untuned short and low-$Q$ dye laser oscillator.

However in this early report some interesting questions related to the STS method were not fully answered. These concerned (1) possibility of adjustability of the output ps pulse wavelength over a fairly large range; (2) stability of the output pulse duration and intensity; (3) possibility of extension of the STS method to dyes other than the rhodamine 6G initially used; and (4) possibility of using pump wavelengths in the UV to generate ps dye laser pulses from 400 to 700 nm. The answers to the above points are not straightforward and require experimental demonstration since excited state molecular parameters of most dyes are not well known in general and even less for picosecond response times.

In this report, we wish to present new results in the experimental study of the STS method. From these results we have designed a compact and simple picosecond generator using dyes for optically converting a fixed-wavelength nanosecond pulse emitted from a standard nanosecond laser (such as Nd:YAG, ruby, $N_2$, excimer lasers) into high power ps pulses spectrally adjustable from 400 to 700 nm. This device is able to produce a pulse-shortening factor greater than 100 (from 10 ns down to 50–100 ps) with a single stage, and for output picosecond wavelengths between 400 and 700 nm.

## 1. Experimental

The present spectro-temporal selection (STS) dye laser shown in Fig. 1 is compact: 20 × 30 cm. The STS oscillator O is simply a standard 1 × 1 cm spectroscopy cell which constitutes a low-$Q$ cavity of 120 ps round-trip time between the uncoated external faces (0.04 reflectivity). The cavity is transversally excited through a cylindric lens CL of 5 cm focal length, by a small part (0.5 mJ) of the output (10 ns, 532 ns or 355 nm, 10 Hz) from a $Q$-switch Quantel Nd:YAG laser. The output from the STS oscillator is collimated by a 10 cm lens and diffracted in the first order of a 1800 g/mm grating G. The spectrum is

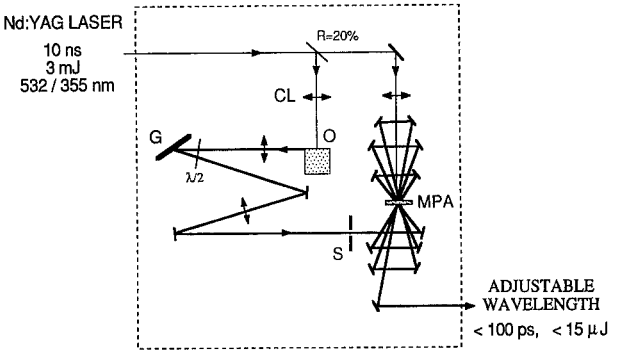


**Fig. 1.** The STS dye laser for generation of single (< 100 ps) picosecond pulses from nanosecond pump laser pulses. O: sweeping oscillator; CL: cylindrical lens; G: grating; S: slit; MPA: 6-pass amplifier

formed with a 30 cm lens on a screen with a 0.5 mm slit S. The transmitted beam is amplified by a dye multipass amplifier (MPA) pumped by the same Nd : YAG laser. A polaroid half-wave plate P is used in order to optimize the output intensities from the grating and the amplification in the MPA. All mirrors have 8 mm diameter and aluminum coatings.

With appropriate focusing and pump energy, the effect of tuning the spectral filter G in the STS laser is the following: when the grating G is tuned at the maximum of the oscillator broadband emission, the pulse transmitted through the slit S is several nanoseconds long. When the grating is tuned to the blue wing of the oscillator broadband, the transmitted pulse become shorter and at a given wavelength a single (< 100 ps) picosecond peak is obtained.

The amplifier MPA is a 1 mm path cell, longitudinally pumped with 2 mJ through a 10 cm $f-1$ lens. The path from the oscillator O to the MPA is long enough for the leading edge of the oscillator emission (emitted during the risetime of the pump pulse) to be sufficiently delayed and so amplified by the maximum of the pump pulse at the multiplass amplifier cell. The value of the saturated total gain of the 6-pass amplifier (MPA) is found to be about $10^3$ with rhodamine dyes. Other reasons for choosing this amplifier configuration are that it provides good beam quality and is easy to align. The shot-to-shot intensity stability of the picosecond pulses is much better after amplification if highly saturated amplification is established in the 6-pass amplifier [7]. Note that it is straightforward to further amplify the 10 μJ picosecond output pulses from the STS laser with two additional double-pass dye amplifiers to obtain high-energy picosecond pulses.

We have tested the production of picosecond pulses with this STS laser with 14 different dyes from Lambda Physik (listed in Table 1) which can absorb the different pumping wavelengths 532 nm or 355 nm.

The output pulse shape of the STS laser is monitored by an streak camera (ARP) with sweep speeds from 1 ns to 50 ps per millimeter. The time resolution is 4 ps. The average pulse durations produced at the 10 Hz repetition rate are measured by the autocorrelation method of non-collinear second harmonic generation. The pulse energies are measured with a RK-3332 Joulemeter (Laser Precision) and spectral widths with a monochromator SPEX equipped with an OMA SSR.

## 2. Results and Discussion

### *2.1 Adjustability of the Picosecond Pulse Wavelength in the STS Method*

In the first work on the STS laser, which used only rhodamin 6G [5], some variation of the ps output pulse wavelength was noted as a function of concentration but was not studied.

It is shown here for the first time that the STS method allows one to produce ps pulses continuously adjustable over a significant part of the gain range of a dye, for example 25 nm for rhodamine dyes pumped at 355 nm. The width of the tuning range depends on the width of the usable concentration range. The ps pulse tunability is obtained first by rotating the grating to the desired ps wavelenght and then by varying the dye concentration to obtain the short pulse at this wavelength.

The tuning range of the ps pulse wavelength, at a given pumping energy, is limited by the dye concentration range in which (broad band) laser action occurs in the short and low-$Q$ cavity. The lowest concentration limit is fixed by the laser threshold value and the highest one is fixed by the thickness of the excited layer of the transversally pumped active medium for which diffraction losses increase so much that they prevent laser action. Thus, it is preferable to use moderate absorption coefficient in order to widen the usable dye concentration range and, therefore, the tuning range. We found, with all rhodamine dyes used, that the highest lasing dye concentration was one order of magnitude larger for 355 nm ($\varepsilon \sim 5 \times 10^3\,\mathrm{l\,M^{-1}\,cm^{-1}}$) than for 532 nm ($\varepsilon \sim 10^5\,\mathrm{l\,M^{-1}\,cm^{-1}}$) pumping. Correspondingly, the widest tunability range, namely 25 nm, was obtained with rhodamine dyes pumping at 355 nm.

In the STS method, the shortest pulse is obtained at the shortest wavelength in the broadband laser output of a short and low-$Q$ dye laser oscillator. It is well known that dye concentration variation produces a general shift of the time-integrated broadband emission in untuned dye lasers. This is used to adjust the STS laser wavelength, but it must be noted that the concentration dependence of the maximum of the time integrated broadband laser emission is different from that of the ps pulse at the selected wavelength which is fixed by the grating. Figure 2 shows the wavelength shifts for both measured in the case of rhodamine 6G.

### *2.2 Picosecond Pulse Generation Between 400 and 700 nm with Other Dyes*

The STS method of producing ps pulses is in principle applicable to different dyes provided they lase in a short and low-$Q$ cavity, but this has not been experimentally demonstrated hitherto.

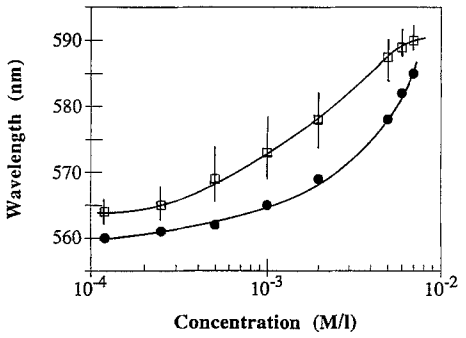


**Fig. 2.** Tunability of the picosecond dye laser pulses measured with rhodamine 6G in ethanol (black circles) pumped at 355 nm as compared to broadband emission at different concentrations. The tunable range is 25 nm. The squares and the bars give the intensity maxima wavelengths and bandwidths, respectively, of the broadband laser emission

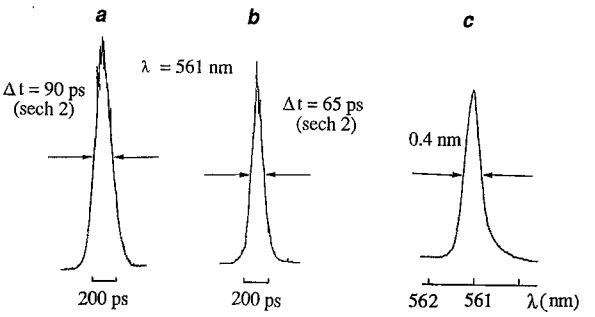


**Fig. 3a–c.** Autocorrelation traces of the output STS laser pulses, for R6G at 561 nm pumped at: **a** 532 nm with $c = 10^{-4}$ M/l, **b** 355 nm with $c = 1.5 \times 10^{-3}$ M/l; the picosecond pulses produced with 355 nm pumping are shorter than with 532 nm pumping. **c** Spectra of the STS laser pulses

We have successfully operated this STS laser with 14 different dyes and with different pumping wavelengths (532 nm and 355 nm) to generate picosecond pulses (50–100 ps) adjustable in the wavelength range from 400 to 700 nm. The operating characteristics are given in Table 1.

The spectral widths of the picosecond STS laser emission depends on the dispersion of the grating, the divergence of the laser beam and the slit width; it is here, with a 0.5 mm slit, less than 0.5 nm (FWHM), as shown in Fig. 3c.

### *2.3 Concentration Effect on Pulse Duration*

With rhodamine dyes pumped at 355 nm, the shortest STS pulses are observed when the concentration is the maximum possible. For example, the pulse duration is about 65 ps with the lowest dye concentration and 50 ps with the highest one. Pulse shortening by the concentration effect is even more noticeable for the rhodamine dyes, where pumping is possible at both 532 nm and 355 nm, as can be seen in Table 1. The ps pulses produced, at the same wavelength, with 355 nm pumping (with concentrations 10 times greater than with 532 nm pumping) are shorter by about 30% than with 532 nm pumping as shown in Fig. 3.

In the case of coumarine dyes, pulse shortening as a function of concentration is not observed. This can be due to the narrow concentration range usable, as presented in Table 1.

The concentration effect on picosecond pulse duration can be understood by the relation [Ref. 5, Eq. (2)] giving the spectro-temporal evolution time, $\tau_{ij}$, which is a parameter describing the spectral sweeping within the broadband laser emission

$$\tau_{ij} = \frac{T}{2LN(\delta_{ij}\sigma_{aj} - \sigma_{ai}) + \alpha(\delta_{ij} - 1)},$$

**Table 1.** Characteristics of the STS picosecond laser

| Dye/solvent | $\lambda_{pump}$ [nm] | Tuning range [nm] | Conc. [$10^{-3}$ M/l] | Pulse duration [ps] | Output energy [μJ] |
|---|---|---|---|---|---|
| Pyridin 1/MeOH (LDS698) | 532 | 695–700 | 0.6–7 | 54 | – |
| Cresyl violet/EtOH | 532 | 645–653 | 6–25 | 86 | 3 |
| | 355 | No lasing | | | |
| Rhodamine 640/EtOH | 532 | 605–607 | 0.1–1 | 90 | 12 |
| (Rhod. 101) | 355 | 605–632 | 1–15 | 65–50 | 8 |
| Rhodamine B/EtOH | 532 | 580–584 | 0.05–0. 4 | 90 | 12 |
| | 355 | 580–598 | 0.5–7 | 75–50 | 9 |
| Rhodamine 6G/EtOH | 532 | 561–564 | 0.1–0.7 | 90 | 14 |
| | 355 | 561–586 | 0.15–7 | 65–50 | 10 |
| Rhodamine 110/EtOH | 532 | 545–555 | 0.8–5 | 65 | 7 |
| | 355 | 545–565 | 1.5–13 | 60–50 | 6 |
| Coumarin 307/EtOH | 355 | 498–504 | 4–10 | 85 | 6 |
| Coumarin 102/EtOH | 355 | 481–484 | 1–35 | 85 | 5 |
| Coumarin 47/EtOH | 355 | 453–456 | 0.5–1.8 | 90 | 5 |
| Coumarin 2/MeOH | 355 | 441–445 | 0.5–1.6 | 95 | 4 |
| Coumarin 120/MeOH | 355 | 435–441 | 0.5–1.5 | 95 | 5 |
| Furan 1/MeOH | 355 | 422–425 | 0.3–0.5 | 90 | 7 |
| Stilbene 3/MeOH | 355 | 429–435 | 0.1–0.5 | 75 | 6 |
| Stilbene 1/MeOH | 355 | 415–417 | 0.1–0.5 | 80 | 3 |

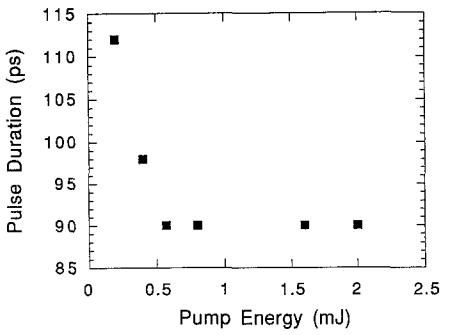


**Fig. 4.** Output pulse duration of the STS dye laser as a function of the energy of the 532 nm, 10 ns, multimode pump pulse in the case of rhodamine 6G. The output pulse duration is not sensitive to pump energy between 4 and 16 times above the lasing threshold

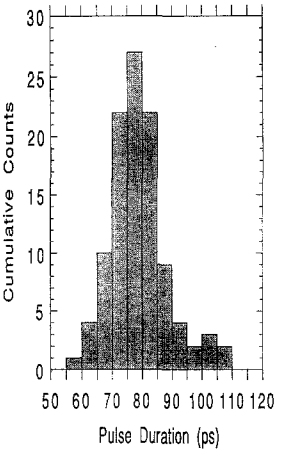


**Fig. 5.** Statistics of the output pulse duration of the rhodamine 6G STS dye laser pumped by a multimode 532 nm Nd:YAG pulse. Stability is found to be ±15% rms

where $T$ is the roundtrip time, $L$ the amplifying path length, $N$ the concentration, $\alpha$ the roundtrip loss given by $\alpha = -\ln R_1 R_2 + \alpha_D$, with $\alpha_D$ is the concentration dependent diffraction loss coefficient; $\sigma_a$ and $\sigma_e$ the absorption and stimulated emission cross sections at wavelengths $i$ and $j$, and

$$\delta_{ij} = \frac{\sigma_{ei} + \sigma_{ai}}{\sigma_{ej} + \sigma_{aj}}.$$

In our case, the increase of the dye concentration $N$, which appears in the $2LN$ factor and in $\alpha_D$, can increase the spectral sweep rate in the broadband laser output. This leads to a pulse shortening as observed.

### *2.4 Pulse Intensity and Duration Fluctuations*

In all types of picosecond dye lasers, fluctuations of the pump intensity lead to fluctuations in the output pulse intensity and duration which must be known before utilisation.

We have investigated the stability degree of the output pulse duration of the STS dye laser, in the case of rhodamine 6G pumped at 532 nm. In the region of single pulse operation, within the range of 4 to 16 times the lasing threshold, the average duration of the output pulse is measured to be little influenced by the pump laser power as shown in Fig. 4. The output of this laser was recorded by a streak camera with a 4 ps resolution. One hundred pulse traces were sampled and each FWHM duration measured. Figure 5 displays the statistics for the rhodamine 6G output pulse duration. The average pulse duration is found to be 78 ps and the stability is ±15% rms. The pulse energy fluctuation of the Nd:YAG (532 nm) pump laser is ±5% rms; this value cannot be the main cause for the 15% fluctuation in the output STS laser duration because the pump energy is well above threshold. The main reason for this fluctuation is the fact that the STS dye laser is pumped by highly structured pulses (with 40 ps spikes due to mode beating) from the multimode Nd:YAG laser.

The pumping intensity fluctuations due to random spikes during one shot disturb the lasing regime and restart an excited population overshot which leads to large fluctuations in both energy and duration of the output pulse. It is believed that the output pulse duration stability of the STS laser will be much improved if smooth pulses, such as those emitted from single-mode Nd:YAG or superfluorescent nitrogen or excimer lasers, are used for pumping.

First tests with pumping by the smooth pulse emitted from a 6 ns, 532 nm, single-mode injected Nd:YAG laser from Quantel show a large increase of stability in both the output ps pulse duration and intensity. With the same pulse energy fluctuations (±5%), the single-mode laser pumping reduces the ps pulse fluctuations by a factor of three.

The fluctuation value of ±15% rms in the output pulse duration of the STS laser is comparable to that of the other picosecond laser systems. The rms fluctuation in the output pulse of the passively mode-locked Nd:YAG laser was reported to be ±20% [9] and ±15% in the cascade transient oscillation laser [8]. Study of the theoretical pump intensity dependence of the output pulse duration showed the pulse duration stability of the distributed feedback laser to be not better than that of the cascade transient oscillation laser [4, 10].

## 3. Summary and Conclusion

The experimental results of the STS method used with UV pumping are significant in practice. This is not only because most laser dyes in the visible spectral range can be efficiently pumped at UV wavelengths but also because high-energy UV laser systems, for example Nd:YAG (355 nm) lasers, excimer or nitrogen lasers, are widely available in laboratories with output durations in the 1–25 ns region.

The STS method for ps pulse generation using a standard ns pump laser has been improved:

1) Adjustability of the output picosecond pulse wavelength over a fairly large range with a given dye in the STS method is shown for the first time. The adjustability range of the ps pulse within the spectral range of each

dye depends on the dye concentration range usable with the pump wavelength. The tuning range is about 25 nm for rhodamine dyes pumped at 355 nm.

2) The spectro-temporal selection method for ps-pulse generation using a standard nanosecond pump laser is successfully used with 14 different dyes and pumping at 532 nm and 355 nm. This allows one to produce directly picosecond dye laser pulses (50–100 ps), with narrow linewidth (0.5 nm), continuously adjustable from 400 to 700 nm by changing the dyes and/or concentration.

3) Fluctuations in both the output ps pulse duration and peak intensity from the STS method are measured. These fluctuations result from the strongly structured waveform of the pump pulse. The ps rhodamine STS laser, when pumped by the 10 ns, 532 nm, Nd : YAG multimode laser, has a pulse duration stability of $\pm 15\%$ rms. The stability value is comparable to that of other ps laser systems.

4) The use of moderate absorption coefficient at the pump wavelength, which permits the use of large concentration range, proved to be of advantage for pulse shortening, tunability and stabilization in the STS method. This suggests the particular interest of pumping STS dye lasers with smooth pulse UV lasers, such as $N_2$, excimer and single-mode UV lasers.

5) From the above new results in the study of the STS method, a device using dyes is designed for optically converting a fixed wavelength nanosecond pump pulse emitted from a standard nanosecond laser into a picosecond pulse spectrally adjustable from 400 to 700 nm. This device is simple to operate and very compact ($20 \times 30$ cm, or, with two power amplifiers, $40 \times 20$ cm). It can be directly applied to time-resolved measurements with a subnanosecond resolution and, in particular for pumping at a desired wavelength in the green and the blue regions, where a standard mode-locking technique is not efficient.

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