

DEVELOPMENT OF A TUNABLE ULTRASHORT DYE LASER SYSTEM BY USING MOLECULAR PHOTONIC PROCESSES AND A NANOSECOND PUMPING LASER

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ABSTRACT

The paper presents the physical processes and operational characteristics of an original sub-picosecond dye laser system which is designed and successfully developed at our laboratory basing on the combination use of three photonic processes of dye molecules: 1) Fast spectro-temporal evolutions in the broadband dye laser emissions; 2) Generation of a chain of ultrashort pulses (spikes) from low-Q micro-cavity dye lasers; and 3) Non-linear resonant interaction between ultrashort laser pulses and highly-saturated dye media. Such a laser system provides a stable generation of single 500 fs pulses with a peak power of 300 MW at 606 nm. Particularly, the whole sub-picosecond laser system is pumped by a single nanosecond laser (Q-switched Nd:YAG laser). The spectral and time processes involved in these pulse-shortening methods are analyzed with a rate-equation model extended to wavelengths. A white light continuum generation was obtained by focusing the 500 fs pulses into 2 cm water cell. As a result, it enables us to produce widely tunable ultrashort laser pulses with a spectral selection and amplification of the supercontinuum spectrum.

1. INTRODUCTION

Over the years, ultrashort pulse lasers have become important tools in many fields of scientific research and applications. They have greatly improved time-resolved spectroscopic measurements and, therefore, led to many important progresses in the study of ultrafast phenomena and new ultrafast laser-based applications [1 - 3]. Production of ultrashort laser pulses is generally based on mode-locking techniques and often requires a combination of two expensive pump lasers, one CW and one pulsed as well as synchronization electronics. This is unavailable to many laboratories, and particular, in the developing countries. However, conventional nanosecond lasers such as Nd: YAG, ruby, nitrogen, metal vapor or excimer laser have been widely used there as optical pump sources. Therefore, much attention devoted to the developments of various methods to produce ultrashort laser pulses with a (non-mode locked) nanosecond pump laser such as cavity transient [4, 5], cavity quenching [6], distributed feedback [7, 8] and spectro-temporal selection methods [9 - 11].

Transient behavior in laser resonators, especially spiking phenomenon, is well observed for the laser media whose the fluorescence lifetimes are longer than the resonator round-trip time. For

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homogeneously broadened active media of organic dyes, the fluorescence lifetimes are short (of the order of nanosecond), relaxation oscillations in the laser emissions well observed with short laser resonators [4, 5]. In the cases of short and low-Q dye laser resonators, the above-threshold laser emission is typically a chain of short pulses (spikes) and strongly dependent on wavelength [9 - 11]. As a result, the first spike can be isolated from the pulse chain by a spectral selection or non-linear resonant interactions of the spikes with highly-saturated dye media. This suggest one to develop simple ways for generating single ultrashort dye laser pulses.

In this paper, we presents the physical processes and operational characteristics of a original dye laser system which is designed and successfully developed at our laboratory using the combination of three photonic processes of dye molecules in order to generate high-power tunable sub-picosecond laser pulses. The spectral and time processes involved in these pulse-shortening methods are analyzed with a rate-equation model extended to wavelengths. Such a laser system uses two low-Q and short dye laser cavities followed by an extra-cavity pulse shaper in a highly saturated absorber and amplifier, provides a stable generation of single 500 fs pulses with a peak power of 300 MW at 606 nm. The whole laser system is pumped by a single standard Q-switched Nd:YAG laser. A white light continuum generation was obtained by focusing the 500 fs pulses into 2 cm water cell. As a result, it enables us to generate high-power tunable sub-picosecond pulses from 450 nm to 720 nm by a simple spectral selection and amplification of the continuum spectrum.

2. SUBPICOSECOND DYE LASER SYSTEM: DESCRIPTION AND PHYSICAL PROCESSES

The dye laser system for converting nanosecond pulses to subpicosecond pulses, as presented in Fig. 1, consists of 3 parts arranged in cascade. A single standard nanosecond Q-switched Nd:YAG laser (6 ns, 20 Hz, Brilliant - Quantel) is used as the pumping laser at 532 nm for the whole laser system. Part 1 is used to convert 6 ns pumping pulses into < 100 ps pulses; Part 2 functions as a sub-picosecond pulse laser converter; Part 3 is a power amplification chain of single sub-picosecond dye laser pulses.

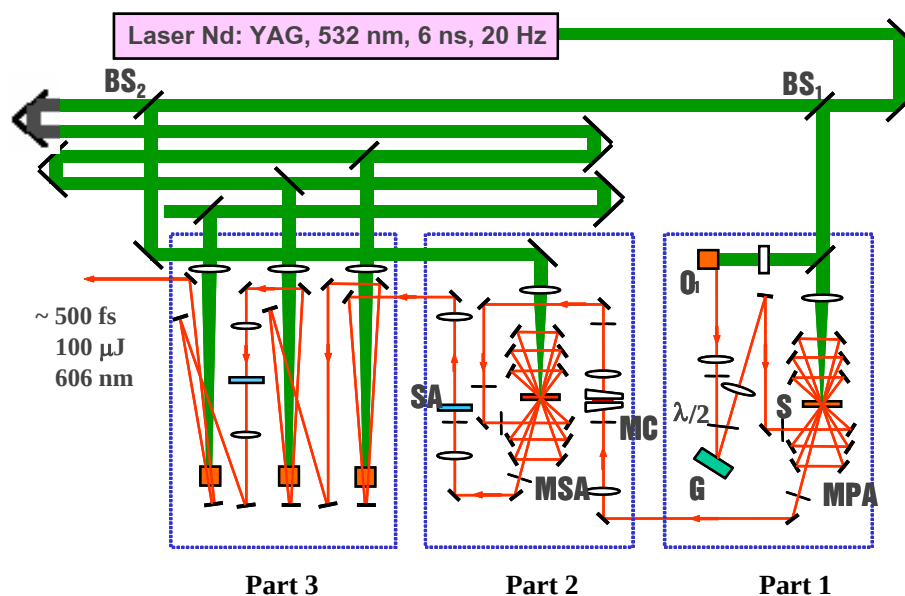


Fig. 1: 500 fs dye laser system

2.1 Part 1: Generation of picosecond dye laser pulses with spectro-temporal selection (STS) method

Part 1 is to produce picosecond dye laser pulses using the spectro-temporal selection (STS) method. Details about picosecond STS dye lasers were reported [9, 10]. The principle of STS method is based on a fast spectral evolution in the broadband nanosecond dye laser emissions from short and low-Q resonators. This important spectral feature is recalled here.

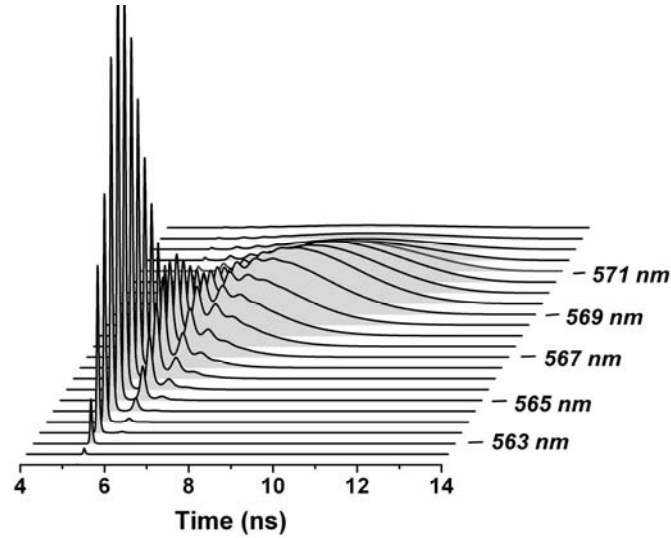


Fig. 2: Typical laser emission from 1 cm dye cell filled with Rh6G/ethanol, $C = 4.10^{-4}$ M/l, and pumped at 10 times above threshold

We consider the broadband laser emission from a standard 1 cm $\times$ 1 cm spectroscopic cell (DL-type Hellma), filled with solution of Rhodamine 6G in ethanol (4.10^{-4} M/l) and is transversally pumped at 10 times above threshold with 6 ns pump pulses. The dye cell constitutes a short and low-Q cavity with two un-coated side faces of 0.04 reflectivity. Figure 2 shows the spectral evolution in the broadband laser emission from such a dye laser by solving the rate equations extended to wavelengths [9 - 11] and using the dye molecular parameters [12, 13]:

$$\frac{\partial N_1}{\partial t} = \left(\sigma_{ap} I_p + \sum_{i=1}^n \sigma_{ai} I_i \right) N_0 - \left(\frac{1}{\tau} + \sum_{i=1}^n \sigma_{ei} I_i \right) N_1$$

$$\frac{\partial I_i}{\partial t} = [2(\sigma_{ei} N_1 - \sigma_{ai} N_0) l - \alpha_i] \frac{I_i}{T} + A_i N_1$$

It should be noted that on the short-wavelength wing of the broadband laser spectrum, the laser oscillation lasts inherently for a short duration. This single short pulse can be obtained by a simple spectral selection. In practice, therefore, the STS method is to select, with an extra cavity filter, a spectral narrow band on the short-wavelength wing of the broadband laser emission from a short and low-Q dye laser oscillator. Its STS laser output is single short pulses, for current dyes, of less than 100 ps [9 - 11].

In our laser, the STS dye laser (Part 1- Fig. 1) is pumped by 6 ns pulses at 532 nm from the Nd:YAG laser. The broadband laser emission from O₁ dye cell (Rh6G /ethanol solution, 4.10^{-4} M/l) is collimated and directed to a small grating G (600 grooves/mm, 1 cm $\times$ 2 cm), the second order diffracted beam is imaged and a spectral narrow band is selected with slit S of 0.3 mm wide. By that way, single short laser pulses of 75 ps and an energy of 15 nJ are produced.

Sequently, the pulse is amplified in a 6-pass amplifier to 30 μJ . The amplifier used a bow-tie configuration with 1-mm thick dye cell (Rh6G/ethanol solution, $3 \cdot 10^{-4}$ M/l, circulated). Such a STS dye laser generated single laser pulses of 75 ps (Fig. 3, assuming sech^2 pulse shape), an pulse energy of 30 μJ at 562 nm and a spectral linewidth of 0.5 nm (FWHM).

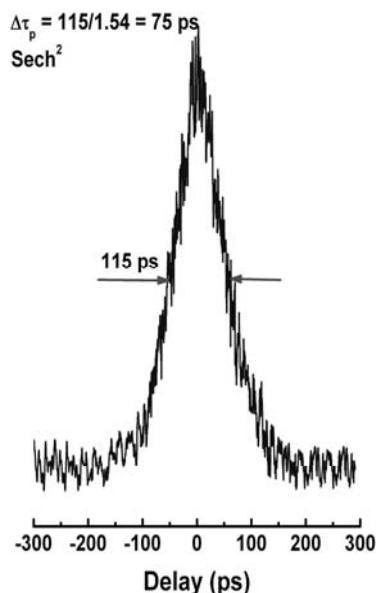


Fig. 3: Autocorrelation trace of output pulses from the Part 1

We have successfully operated the picosecond STS laser with different dyes (in liquid and solid state matrices) pumped at 355 nm or 532 nm by the third and second harmonic generation of the Nd:YAG laser in order to produce picosecond dye laser pulses (< 100 ps) spectrally adjustable from 386 nm to 860 nm [9 - 11]. In comparison with the mode-locked lasers that are used as optical pumping sources, these STS picosecond dye lasers have advantages such as compactness (25 cm $\times$ 35 cm), ease of operation, economy, possible use of many pump wavelengths and tunability of the output wavelength.

2.2 Part 2: Generation of subpicosecond pulses

Part 2 is to convert the 75 ps pulses generated from the STS dye laser into sub-picosecond laser pulses (Fig. 1). For this purpose, we used an ultralow-Q micro-cavity dye laser (MC) to produce a chain of ultrashort pulses (spikes) and the highly- saturated dye amplifier and absorber for extra-microcavity pulse treatment. The ultralow-Q micro-cavity dye laser (MC) is made of two wedged silica plates (preferably wedged to avoid parasitic reflection) pressed together with a very thin layer of solution of Rhodamine 640/methanol ($3 \cdot 10^{-2}$ M/l). Similar small-path cells are commercially available (e.g., Hellma). MC is pumped longitudinally by the at 562 nm, 75 ps pulse that emitted from Part 1 through a 10 cm focal length lens.

Applying the same type of calculations of the rate equation system extended to wavelengths, we have computed the spectral evolution of the laser emissions from the 50 μm low-Q dye laser cavity, as shown in Fig. 4, which is a chain of sub-picosecond pulses (spikes). Such a pulse structure well presents the spiking phenomenon in the laser relaxation oscillations of an ultralow-Q ultrashort laser cavity and, in particular, that well agrees with experimental one measured by a streak camera (Fig. 5). The MC laser emission spectrum could be changed by translation or rotation the MC in respect to the pumping beam axis (Fig. 4b).

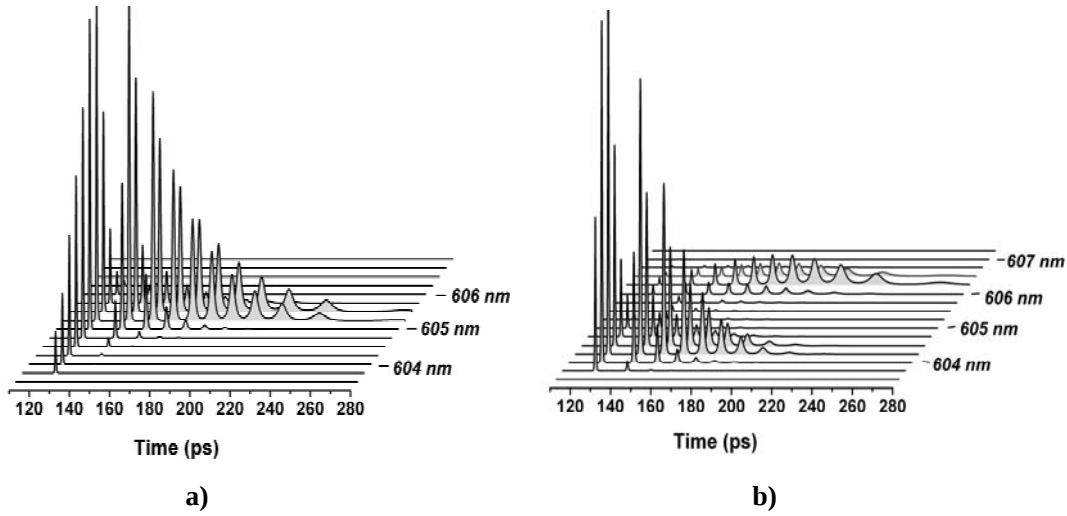


Fig. 4: Laser emission from a 50 μm microcavity filled with Rh640/methanol ($C = 3.10^{-2} \text{ M/l}$) pumped by Gaussian shape pulses of 100 ps at 6 times above laser threshold. a) MC is perpendicular to the pumping beam; b) MC is tilted 4°

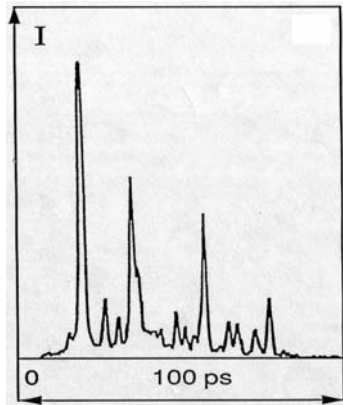


Fig. 5: Typical emission from a 25 μm cavity

In order to obtain single ultrashort laser pulses by extra-micro cavity pulse treatment, the micro cavity dye laser (MC) is followed by a multi-pass saturable dye amplifier (MSA), its configuration is similar to MPA in Part 1 and the Rh640/ethanol solution (3.10^{-4} M/l) is used as a gain medium. Because of highly-saturated gain, this amplifier functions mainly as a pulse shaper, the chain of spikes is strongly distorted pass by pass and the first spike becomes dominant in the amplified pulse chain. This pulse-shaping process is demonstrated by solving the following rate equation for gain G at each pass [14 - 16] and the results obtained are presented in Fig. 6. This pulse-shaping process is still effective until the sixth pass, that is why a six - pass saturable amplifier (MSA) configuration was used in our laser.

$$\frac{dG}{dt} = v(\sigma_{es} + \sigma_{as})G[I_{p0}(t + \Delta t)(1 - e^{-\kappa NL} G^\gamma) - I_{s0}(t)(G - 1)] - \frac{1}{\tau}G[\ln G + \sigma_{as}NL]$$

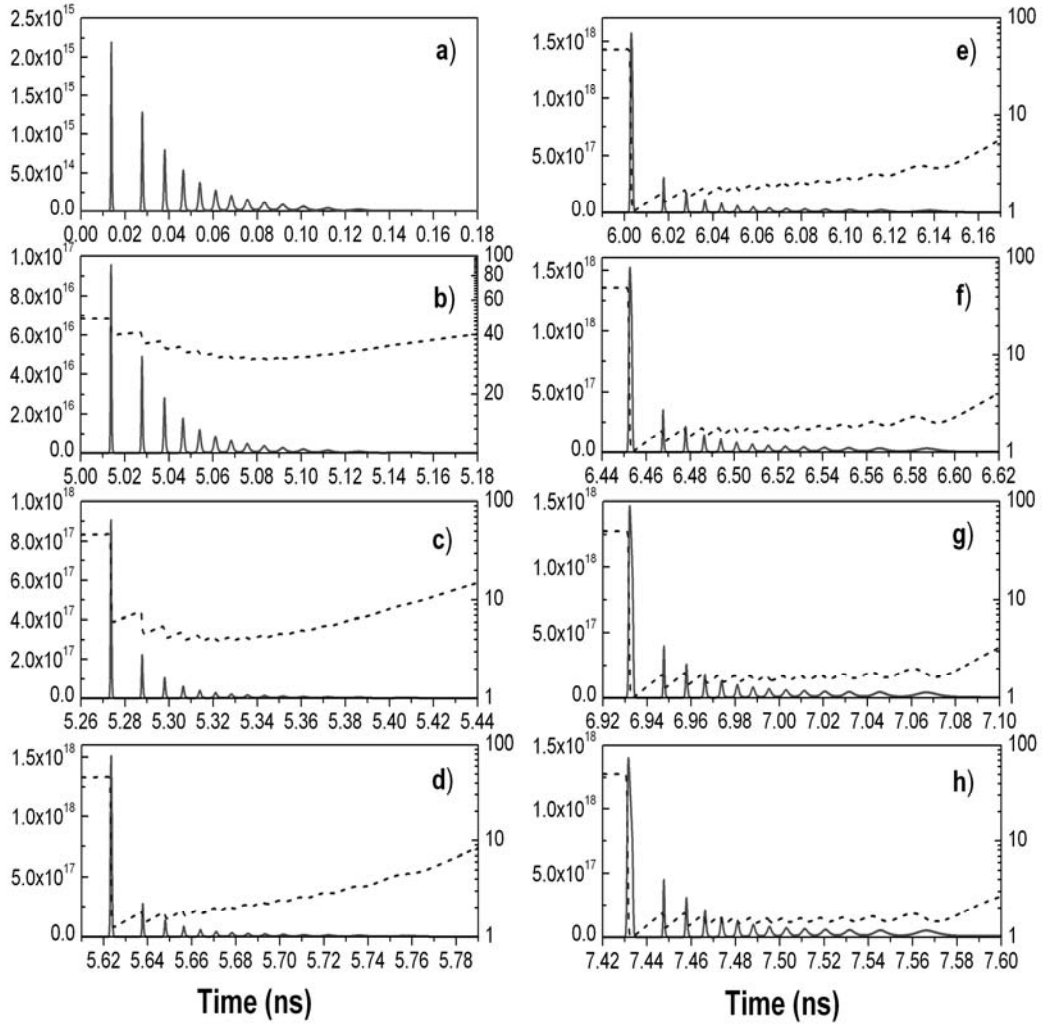


Fig. 6: a) The input signal; b) – h) Evolution of gain (dotted curves) and amplified signal intensity (solid curves) after each pass. (the left vertical axis: Signal intensity [$\text{photons.cm}^{-2}.\text{s}^{-1}$]; The right vertical axis: Gain)

The distorted output from MSA is focused into a saturable absorber (SA). Due to absorption saturation, the first spike is isolated from the chain of spikes and well passed SA. This provides us single sub-picosecond pulses. Fig. 7 presents the computation results for this pulse isolation process using a 1 mm dye cell filled with Malachite Green (MG) in methanol as a saturable absorber [17]. As analyzed in [11], suitable selection of concentration, pumping energy and input signal intensity for MSA and SA is a critical task due to their significant influences on pulse distortion.

In our laser system, the 25 μm MC is used and filled with Rh640/methanol solution (2.10^{-2} M/l) and pumped by 75 ps pulses from Part 1. The MC laser output is collimated and sent to MSA (Rh640/ethanol solution, 3.10^{-4} M/l). MSA is pumped semi-longitudinally by pulses of 6 ns (FWHM) and 3 mJ at 532 nm from the Nd:YAG laser. The synchronization between the signal and the pump pulses is performed by an optical delay line. The MSA output is of $\sim 30 \mu\text{J}$ with an amplified spontaneous emission energy of $\sim 50\%$. Adequate focusing of the MSA output pulse

into the Malachite Green saturable absorber cell (5.10^{-4} M/l) wipes out the spike tail and a single ultrashort spike - the first strongest one of ~ 1 μ J is obtained.

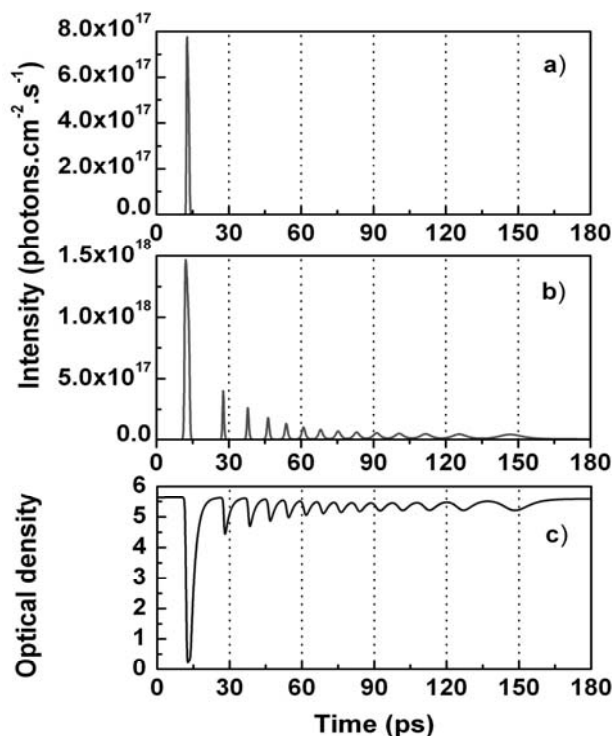


Fig. 7: Action of saturable absorber in extracting single sub-ps pulses. a) output; b) input; c) optical density of absorber

2.3 Part 3: Power amplifier

The single ultrashort laser pulse at 606 nm from Part 2 is amplified with three double-pass 1 cm amplifiers containing a circulated Rh640/ethanol solution (5.10^{-5} M/l). The second and the third amplifiers are separated by a 1 mm Malachite Green saturable absorber cell in order to eliminate ASE. The pumping pulse energies for the amplifiers are of 8, 10 and 10 mJ, respectively. Such a power amplification chain provides an amplifying factor of more than 100 that led to the output pulse energy of ~ 125 μ J.

The pulse duration is measured with the autocorrelator which used a non-collinear second harmonic generation with a BBO nonlinear crystal. The autocorrelation traces (Fig. 8) are corresponded to two positions of the Malachite Green saturable absorber (SA). In both cases, output pulse duration is about 500 fs assuming the sech^2 pulse shape. However, the obtained autocorrelation traces present the different actions of extra-cavity pulse treatment. For example, as the saturable absorber is at the tightly focused position of the input beam, the output could be multiple pulses corresponding to the enlargement of the autocorrelation trace on its foot (Fig. 8a). The trace in Fig. 8b corresponds to the shift of the saturable absorber 0.3 mm away from the previous position. At this position, the MG absorber is only saturated with the first and strongest spike, therefore, the output is the single pulses.

In brief, at the output of Part 3 we have obtained the single dye laser pulses of ~ 500 fs in duration, 125 μ J in energy at 606 nm.

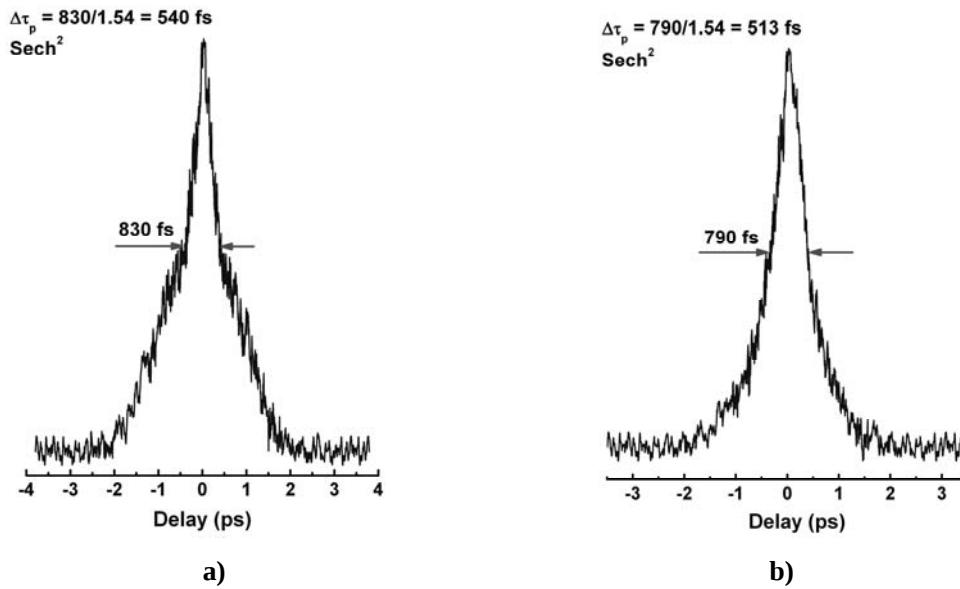


Fig. 8: Autocorrelation traces of output laser pulses from Part 3

3. SUPERCONTINUUM GENERATION AND WIDELY TUNABLE 500 FS PULSE

A white light continuum is generated by focusing the $\sim 100 \mu\text{J}$ - 500 fs pulse into 2 cm water cell with a 10 cm focal length lens. As a result of self-phase modulation, the supercontinuum spectrum is obtained, as presented in Fig. 9, and strongly broadened to cover over the UV, visible and near infrared spectral regions. The energy of the white light supercontinuum varies by 4 orders of magnitude from the central wavelength to both continuum wings (345 and 860 nm).

Significantly, the white light continuum generation enables one to produce widely tunable ultrashort pulses. By selecting a narrow band in the supercontinuum with an interferential filters of 1 nm FWHM bandwidth, we have produced nearly transform-limited subpicosecond pulses at a desired wavelength. Furthermore, widely and continuous tunability of ultrashort laser pulse from 540 nm to 720 nm is obtained by using suitable spectral filters and several dyes in dye amplifiers pumped at 532 nm. With the 355 nm pumping by the third harmonic of the same Nd:YAG laser, the system generates tunable sub-picosecond pulses between 540 nm to 450 nm with Coumarin dyes. Detail presentations of spectral selection and amplification of a desired spectral band from the supercontinuum spectrum were showed [18, 19].

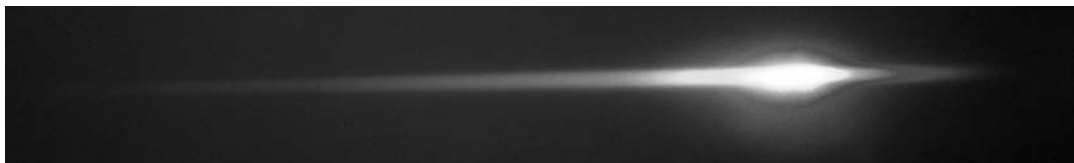


Fig. 9: Supercontinuum Spectrum of 500 fs laser pulse after passing through a water cell of 2 cm long

4. CONCLUSION

In this paper we have presented a simple, low-cost and relatively compact (80 cm wide $\times$ 100 cm long)

cm long) sub-picosecond dye laser system using a standard nanosecond pumping laser and photonic processes of organic dye molecules. The ultrashort dye laser system generates simultaneous 100 ps pulses (Part 1), single powerful 500 fs pulses at 606 nm (Part 3) and tunable laser 500 fs pulse resulted from the white light supercontinuum generation. Such an ultrafast laser does not need any expensive and specific optical components and therefore, it is reproducible at the laboratories of the developing countries and directly applied to time – resolved laser spectroscopic measurements and applications.

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