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Generation of single picosecond pulses in a distributed-feedback binary dye mixture laser pumped by nanosecond pulses

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***Abstract*. A new technique for generation of stable single tunable picosecond pulses was proposed and realised in a distributed-feedback binary dye mixture laser pumped by nanosecond pulses with energies significantly exceeding the excitation threshold.**

Keywords: *distributed feedback, organic dyes, single picosecond pulses, spectral switching*.

1. Introduction

One of the most simple ways to obtain ultrashort pulses in dye lasers is passive Q-switching, which does not involve additional external control devices [1]. Of special interest from the practical standpoint is the operation of a dye laser when the laser generates a single tunable picosecond pulse upon pumping by nanosecond pulses with energies significantly exceeding the threshold excitation energy, which should appreciably reduce the influence of pump fluctuations on the lasing characteristics of the dye laser.

Our analysis shows that, because of several complicating circumstances, the conventional Q-switching is not the radical solution to the problem of obtaining stable single picosecond pulses in such dye lasers [2], while the employment of mode locking [3, 4] or the use of the regime of relaxation oscillations and the quenching of undesirable subsequent pulses [5, 6] are complex in realisation. For this reason, it is important to develop new lasing regimes in dye lasers for the generation of tunable and stable single picosecond pulses in a wide intensity range of nanosecond pumping pulses provided by inexpensive sources of a simple design, which should provide a broad field of their practical applications.

Consider a dye laser with a dynamic distributed feedback whose active medium consists of a solution of two dyes; one of them fulfils the function of a laser medium and the second one serves as a saturable absorber. It is known that in distributed-feedback dye lasers, unlike lasers with mirror resonators, self-Q-switching occurs (the photon lifetime in a resonator and, hence, its Q factor decrease sharply at the instant of laser pulse emission) [7, 9].

Because the amplification spectrum of organic dyes is rather broad (20 – 50 nm) and the emission wavelength of a distributed-feedback dye laser is strictly defined by the angles of incidence of the interfering pump beams on the active medium [10], it is possible to provide the conditions when lasing occurs in two spectral regions. One of these regions corresponds to the wavelength determined by the distributed feedback and the second one is determined by the spectral tuning of a selective external resonator. When an unsaturated absorber is present in the two-component active medium, the external resonator Q factor can be so selected that no lasing occurs for a given pump pulse energy.

The exciting radiation produces a population grating in the laser medium, when the lasing threshold is achieved, a picosecond pulse is emitted at the wavelength determined by the distributed feedback; simultaneously, the saturable absorber is saturated and the population of its ground state decreases almost to zero. The inverse population of the laser medium also decreases, resulting in a sharp shortening of the photon lifetime in the distributed-feedback resonator. When a saturable absorber with a long excited-state lifetime (of the order of several nanoseconds) is used, after the first laser pulse by the distributed-feedback structure it remains bleached for a time sufficiently long to reach the lasing threshold in the external resonator because of the continuing pumping.

Because the threshold with a bleached saturable absorber is reached for a relatively low inverse population of the laser medium, which is insufficient to produce an efficient distributed feedback, subsequent pulses will be generated at the wavelength determined by the external feedback. As a result, the distributed-feedback dye laser will operate in the regime of switching from the wavelength determined by the distributed feedback (the first pulse) to the wavelength determined by the detuned external resonator (subsequent pulses). The saturable absorber provides in this regime lasing in the distributed-feedback system at high threshold inverse populations. Therefore, one might expect that a distributed-feedback binary organic dye mixture laser operating in the regime of spectral switching can generate a single picosecond pulse at the wavelength determined by the distributed feedback upon pumping by nanosecond pulses with energies significantly exceeding the lasing threshold.

We will calculate the lasing characteristics for the proposed regime of spectral switching of the distributed-feedback dye laser with a two-component active medium

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using the rate equations for space-averaged parameters [1, 2, 11]:

$$\frac{\mathrm{d}n}{\mathrm{d}t} = I_\mathrm{p}\sigma_\mathrm{p}(N-n) - \frac{\sigma_{\mathrm{e}1}c}{\eta}nq_1 - \frac{\sigma_{\mathrm{e}2}c}{\eta}nq_2 + \frac{\sigma_{\mathrm{a}1}c}{\eta}(N-n)q_1 + \frac{\sigma_{\mathrm{a}2}c}{\eta}(N-n)q_2 - \frac{n}{\tau}, \quad (1)$$

$$\frac{\mathrm{d}n_\mathrm{a}}{\mathrm{d}t} = \frac{N_\mathrm{a}-n_\mathrm{a}}{\tau_\mathrm{a}} - I_\mathrm{p}\sigma_{\mathrm{pa}}n_\mathrm{a} - \frac{\sigma_{1\mathrm{aa}}c}{\eta}n_\mathrm{a}q_1 - \frac{\sigma_{2\mathrm{aa}}c}{\eta}n_\mathrm{a}q_2 - k\sigma_{\mathrm{aa}}\frac{n_\mathrm{a}n}{\tau}, \quad (2)$$

$$\frac{\mathrm{d}q_1}{\mathrm{d}t} = \frac{\sigma_{\mathrm{e}1}c}{\eta}nq_1 - \frac{q_1}{\tau_{\mathrm{c}1}} + \Omega_1\frac{n}{\tau} - \frac{\sigma_{1\mathrm{aa}}c}{\eta}n_\mathrm{a}q_1 - \frac{\sigma_{\mathrm{a}1}c}{\eta}(N-n)q_1, \quad (3)$$

$$\frac{\mathrm{d}q_2}{\mathrm{d}t} = \frac{L}{L_2}\frac{\sigma_{\mathrm{e}2}c}{\eta}nq_2 - \frac{q_2}{\tau_{\mathrm{c}2}} + \Omega_2\frac{n}{\tau} - \frac{L}{L_2}\frac{\sigma_{2\mathrm{aa}}c}{\eta}nq_2 - \frac{L}{L_2}\frac{\sigma_{\mathrm{a}2}c}{\eta}(N-n)q_2, \quad (4)$$

where n and n_a are the densities of laser-medium molecules in the excited state and saturable-absorber molecules in the ground state, respectively; N and N_a are the densities of laser-medium and saturable-absorber molecules; q_1 and q_2 are the photon densities in the laser medium at wavelengths determined by the distributed feedback (λ_1) and the spectrally selective external resonator (λ_2), respectively; I_p is the photon density of the excitation power; L is the length of the active medium; η is the refractive index of the solution; τ and τ_a are the excited-state lifetimes of the laser-medium and saturable-absorber molecules; c is the velocity of light; σ_p and σ_{pa} are the absorption cross sections of lasing molecules and saturable-absorber molecules at the pumping wavelength, respectively; $\sigma_{\mathrm{e}1}$ and $\sigma_{\mathrm{e}2}$ are the lasing cross sections at the wavelengths λ_1 and λ_2, respectively; $\sigma_{\mathrm{a}1}$ and $\sigma_{\mathrm{a}2}$ are the absorption cross sections of lasing molecules at wavelengths λ_1 and λ_2, respectively; $\sigma_{1\mathrm{aa}}$ and $\sigma_{2\mathrm{aa}}$ are the absorption cross sections of the saturable absorber at wavelengths λ_1 and λ_2, respectively; σ_{aa} is the average absorption cross section of the saturable absorber; k is the parameter that takes into account the saturable absorber excited-state population due to the nonradiative energy transfer from the laser medium and the reabsorption of the luminescence of this medium; Ω_1 and Ω_2 are the spectral spatial factors which determine the fraction of luminescence of the laser medium that falls within the corresponding spectral and spatial lasing regions;

$$\tau_{\mathrm{c}1} = \max\left[\frac{\eta L^3}{8\pi^2 c}(\sigma_{\mathrm{e}1}nV)^2, \frac{\eta L}{10c}\right], \quad (5)$$

$$\tau_{\mathrm{c}2} = \frac{2L_2\eta}{c\ln(1/R_1R_2)} \quad (6)$$

are the photon lifetimes in the distributed-feedback and external resonators [12], respectively; V is the interference pattern visibility; R_1 and R_2 are the reflectivities of the mirrors of the external resonator; and L_2 is the length of the external resonator.

The last term in Eqn (2), which is not used in the conventional rate-equation model, describes the population of the excited state of saturable absorber molecules due to the nonradiative inductive-resonance energy transfer (the transfer through the exchange-resonance mechanism can be neglected at the molecular concentrations employed) from excited molecules of the laser medium and due to the reabsorption of luminescence of these molecules, which is emitted in the direction different from the direction of the resonator axis.

Indeed, a subsequent analysis showed that the processes under discussion play a significant part in the dynamics of lasing development in the laser system involved. Detailed calculations of the parameter k require the correct inclusion of nonradiative energy transfer and also the description of reabsorption of luminescence of the laser medium by the saturable absorber for a specific geometry of the pumped region. These calculations are rather cumbersome (see, for instance, Refs [13, 14] where the reabsorption of laser and luminescence emission was calculated in a distributed-feedback binary dye mixture laser).

We made approximate estimates of the parameter k. In the case of nonradiative energy transfer, k can be taken equal to half the diameter of the pumped region, because the main contribution to the lasing characteristics of the laser system under study will be made only by the saturable-absorber molecules located in the pumped region. A typical diameter of the pump radiation focused in the active medium is $\sim 50-100$ μm; therefore, the parameter k is estimated as $(2.5-5)\times10^{-3}$ cm. For concentrations of the laser-medium and saturable-absorber molecules used in practice and the critical radius of nonradiative inductive-resonance energy transfer (~ 40 Å), the parameter k can be estimated as 10^{-3} cm [15]. Therefore, the nonradiative energy transfer makes a contribution to the excited-state population of the saturable absorber that is comparable to that made by the luminescence reabsorption, and the parameter k can vary between 10^{-3} and 10^{-2} cm.

Let the time profile of the exciting pulse be Gaussian,

$$I_\mathrm{p} = I_\mathrm{p}^0\exp\left[-4\ln 2\left(\frac{t}{T}\right)^2\right], \quad (7)$$

where I_p^0 and T are the peak power density and the FWHM duration of the excitating pulse, respectively. Then, the output powers P_1 and P_2 at wavelengths λ_1 and λ_2 are described by the expressions [16]:

$$P_1 = \frac{1}{2}\frac{hc}{\lambda_1}\frac{abL}{\tau_{\mathrm{c}1}}q_1, \quad (8)$$

$$P_2 = \frac{hc}{\lambda_2}\frac{abL_2}{\tau_{\mathrm{c}2}}q_2, \quad (9)$$

where $a = (N\sigma_\mathrm{p} + N_\mathrm{a}\sigma_{\mathrm{pa}})^{-1}$ is the exciting-radiation penetration depth in the dye solution (defined as the depth at which the pump is attenuated by a factor e); h is the Planck constant; and b is the height of the excited volume.

The system of rate equations (1)–(4) was solved by the fourth-order Runge–Kutta technique using initial conditions specified for $t \ll -T$. We used the following molecular parameters corresponding to the pair of dyes 6-aminophenalemine (AP) as a laser medium and oxazine-1 (OX) as a saturable absorber (the normalised absorption and

luminescence spectra of AP and OX are given in Fig. 1): $\sigma_p = 7 \times 10^{-17}$ cm^2, $\sigma_{pa} = 2 \times 10^{-17}$ cm^2, $\sigma_{e1} = 6.8 \times 10^{-17}$ cm^2, $\sigma_{e2} = 6.5 \times 10^{-17}$ cm^2, $\sigma_{a1} = 4.2 \times 10^{-18}$ cm^2, $\sigma_{a2} = 2.1 \times 10^{-18}$ cm^2, $\sigma_{1aa} = 1.7 \times 10^{-16}$ cm^2, $\sigma_{2aa} = 1.9 \times 10^{-16}$ cm^2, $\sigma_{aa} = 1.8 \times 10^{-16}$ cm^2, $\eta = 1.36$, $\Omega_1 = 5.2 \times 10^{-8}$, $\Omega_2 = 3 \times 10^{-6}$, $\tau = 4$ ns, $\tau_a = 3$ ns, $k = 3.5 \times 10^{-3}$ cm, $\lambda_1 = 595$ nm, $\lambda_2 = 610$ nm, $L = 0.9$ cm, $L_2 = 5$ cm, $V = 1$, $b = 200$ μm, $T = 10$ ns, 2×10^{-3} mol L^{-1}, $C_a = N_a/A = 5 \times 10^{-5}$ mol L^{-1} (A is the Avogadro number).

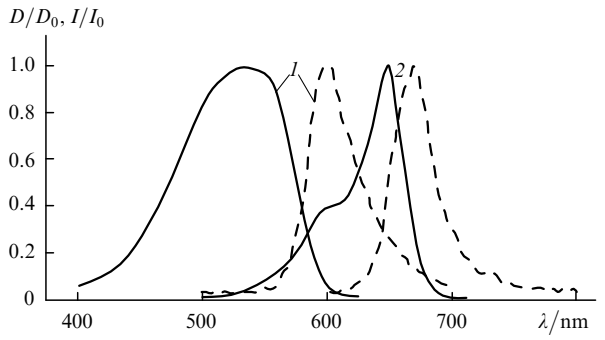

Figure 1. Absorption D/D_0 (solid curves) and luminescence I/I_0 (dashed curves) spectra of 6-aminophenalemine (*1*) and oxazine-1 (*2*).

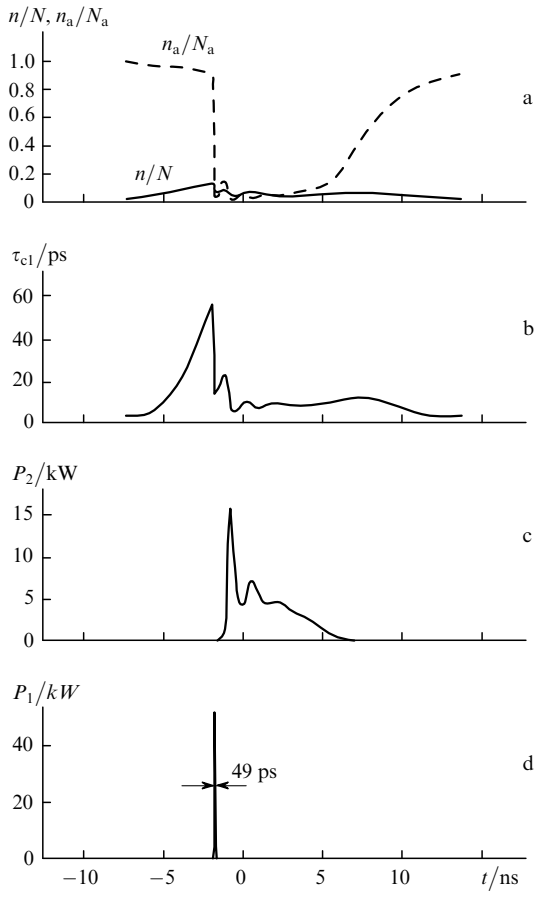

Figure 2. Time dependences of the laser-medium excited-state (n/N) and the saturable-absorber ground-state (n_a/N_a) populations (a), of the photon lifetime in the distributed-feedback resonator (b), and of the output laser power formed in the external resonator (c) and the distributed-feedback resonator (d) for the pump power two times the threshold; $R_1 = 1$, $R_2 = 0.04$.

Fig. 2 shows the dynamic output characteristics of the distributed-feedback dye laser with spectral switching calculated for the pumping power two times the lasing threshold. The duration of the laser pulse at the wavelength determined by the distributed feedback is 49 ps and the laser efficiency is 1.5 %. According to our analysis, even when the pumping power exceeds the threshold by a factor of four, the second laser pulse is emitted at the wavelength λ_1, its intensity is nearly an order of magnitude lower than the intensity of the first pulse. Note that this lasing is qualitatively different from the lasing which occurs in systems with relaxation oscillations upon 'conventional' passive Q-switching, where a group of pulses of comparable intensity is generated upon pumping high above the threshold. In the version under study, the weak subsequent pulses which follow the first 'quasi-single' pulse can be efficiently suppressed using the 'amplifier – nonlinear filter' combination, if necessary.

The lasing at the wavelength λ_2, which is determined by the tuning of the spectrally selective external resonator, develops after the saturable-absorber bleaching (see Figs 2a, c) and is delayed compared to lasing due to the distributed-feedback structure. In this case, the photon lifetime in the distributed-feedback dye laser decreases sharply (this corresponds to a low inverse population in the laser medium because of lasing in the external resonator when the saturable absorber is bleached); as a result the lasing threshold at the wavelength λ_1 is not reached (see Fig. 2b).

Therefore, our calculations showed that a distributed-feedback dye laser with a spectral switching of lasing is capable of generating a stable single tunable picosecond pulse when the pump intensity is high above the threshold.

Fig. 3a shows the scheme of the experimental setup. The distributed-feedback dye laser (*6*) and amplifier (*8*) were pumped by a vertically polarised second-harmonic 532-nm radiation from a solid-state Nd^{3+}:YAG laser (*1*) with the FWHM duration $T = 10$ ns and a pulse repetition rate of 5 Hz. Neutral filters (*2*) were used to vary the pump energy which was measured with an IMO-3 power meter. A Gnom-2 laser was used as a distributed-feedback dye laser (its schematic is shown in Fig. 3b). The laser wavelength was tuned by rotating adjustment mirrors (*13*). The external resonator was formed by two Littrow prisms (*15*) and (*16*). The rear reflecting face of one of them was coated with aluminium. The reflecting surface of other prism was not coated to afford the required low Q factor of the spectrally selective external resonator.

We used ethanol solutions of AP (the laser medium) at the concentration $C = 2 \times 10^{-3}$ mol L^{-1} and OX (the saturable absorber) at the concentration $C_a = 5 \times 10^{-5}$ mol L^{-1} (the molecular parameters of these compounds were used in the above calculations). The laser was tuned to the emission wavelength $\lambda_1 = 595$ nm and the external resonator to $\lambda_2 = 610$ nm (the observed spectral width of laser line in the external resonator was of about 2 nm). The length of the external resonator was $L_2 = 5$ cm and the length of the distributed-feedback structure produced in the active medium was 0.9 cm, which corresponded to the active medium length L.

A 1200 lines mm^{-1} diffraction grating (*7*) was used for the spatial separation of laser lines at the wavelengths λ_1 and λ_2. The amplified laser radiation at the wavelength determined by the distributed feedback (as the amplifier, a transversely pumped one centimetre-long cell with an ethanol solution of AP at the concentration 10^{-3} mol L^{-1} was used)

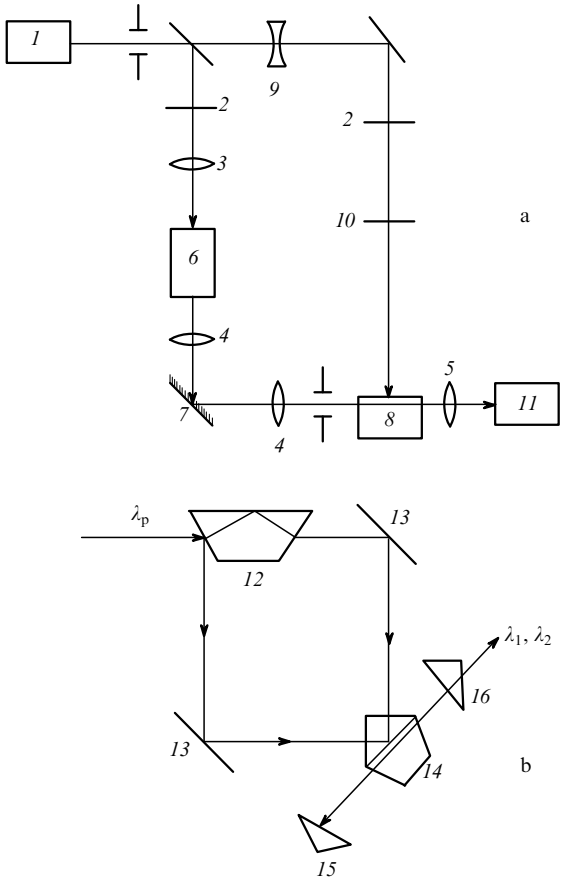

Figure 3. Schemes of the experimental setup (a) and the distributed-feedback dye laser with a spectrally selective external resonator (b): (*1*) Nd^{3+}:YAG laser; (*2*) neutral density filters; (*3*) telescope; (*4*, *5*) spherical lenses; (*6*) distributed-feedback dye laser; (*7*) diffraction grating; (*8*) dye cell (amplifier); (*9*) negative lens; (*10*) cylindrical lens; (*11*) recording system; (*12*) beamsplitter; (*13*) tuning mirrors; (*14*) prism cell; (*15*, *16*) Littrow prisms with and without an aluminium coating on the reflecting surface, respectively.

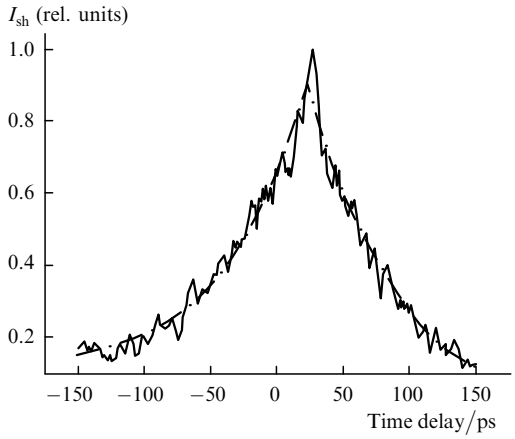

Figure 4. Experimental (the solid curve) and approximating (the dash-dotted curve) autocorrelation function of a single pulse from a distributed-feedback dye laser with a spectral switching.

was directed to the recording system (*11*) after recollimation with a lens (*5*). An MDR-6U monochromator was used to observe the laser emission spectrum.

Using an oscilloscope, we observed distinctly a single laser pulse at the wavelength determined by the distributed feedback (its FWHM duration determined by the instrumental function of the recording system was 1 ns), whereas the duration of the laser pulse at the wavelength λ_2 was about 7 ns; in this case, the pumping power exceeded the threshold by a factor of two. Cutting off the external resonator resulted in a sharp increase in the duration of the laser pulse (up to 6–8 ns) for the saturable absorberme pump power.

Fig. 4 shows the shape of the autocorrelation function of a single laser pulse measured by the technique of noncollinear second-harmonic generation. The duration of a single laser pulse estimated from the width of the autocorrelation function was 46 ps, which is in good agreement with the results obtained in the calculations.

Therefore, we have shown, both theoretically and experimentally, the possibility of stable generation of a single tunable picosecond pulse in a distributed-feedback binary organic dye mixture laser in the regime of spectral switching when the pump intensity is significantly higher than the threshold. The results of this work can provide the basis for the development of simple and reliable sources of single frequency-tunable ultrashort pulses upon pumping a dye laser by nanosecond pulses. In addition, the proposed technique of spectral switching of lasing can be used in lasers based on other broadband active media.

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