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Solid-state organic laser with a wavelength tuning range of 78 nm

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Abstract. **We report the results of studies of the lasing characteristics of an organic solid-state tunable laser with a polymethylmethacrylate matrix doped with Chromene 3, Pyrromethene 567, and Pyrromethene 597 dyes. The features of lasing when using selective and nonselective cavities are described. Tuning of the laser wavelength with a spectral linewidth of 0.018 nm in the range 550–628 nm is obtained on three polymer laser-active media emitting in the yellow and red spectral regions.**

Keywords: *dye lasers, organic solid-state laser, solid-state laser-active media, wavelength radiation tuning.*

1. Introduction

Tunable organic dye lasers have found wide application in science and technology due to the unique parameters of the output radiation: high pulsed power, wide wavelength tuning range, and narrow lasing line. The main advantages of tunable dye lasers include the relative simplicity of the design and a wide variety of organic laser-active media (LAMs), as well as the possibility of targeted changes in the structure and properties for specific pumping conditions. Furthermore, the technological conditions for the production of dye-based LAMs are much simpler and cheaper than for the production of nonlinear media used in optical parametric oscillators, which are direct competitors of dye lasers. However, the use of tunable dye lasers is limited by their main disadvantage: LAMs are liquid solutions. Recently produced solid-state dye-based organic media are capable of converting pump radiation into coherent radiation with a high energy and power, and have an efficiency and service life that are not inferior to their liquid counterparts, which stimulates the development of work in this field [1, 2].

To implement a specific version of a solid-state tunable laser, it is necessary to ensure the matching of the LAM parameters, which mainly determine the characteristics of the device, with the choice of a pumping scheme and the configuration of a selective cavity. When developing effective organic LAMs, it is necessary to solve a number of problems: to choose the dye and the polymer that combine well with each other and are commercially available; to ensure that the absorption bands of active media coincide with the pump radiation wavelength; and select compounds with a low level of induced absorption.

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Currently, LAMs using polymethylmethacrylate (PMMA) doped with various dyes have enjoyed the widest use among solid-state media. Despite the fact that there are quite many works devoted to solid-state LAMs, there are hardly any specifically implemented designs of solid-state tunable dye lasers. In our previous studies, we showed the promise of using solid-state LAMs based on known dyes: Pyrromethene 567 (RM567), Pyrromethene 597 (RM597), and Chromene 3 (Ch3) in PMMA [3, 4]. In the case of a nonselective cavity, a high conversion efficiency (over 80%) and a long service life (more than 10^5 lasing pulses when pumped into one region of a solid-state laser optical element) were achieved. In addition, when designing a tunable dye laser, the choice of the LAM pumping scheme is of no small importance. Three pumping schemes are widely used: longitudinal, quasi-longitudinal, and transverse [5, 6]. The latter allows pumping significant LAM volumes, which is important for achieving a high output energy and power.

The aim of this work was to implement a version of a tunable solid-state organic laser based on the produced high-efficiency solid-state LAMs and to study the features of the output characteristics of the laser in the variation of selective cavity parameters.

2. Experimental facility

An important factor determining the operating characteristics of a tunable laser is the choice of an efficient selective cavity configuration. Of the many versions of dispersive cavities with diffraction gratings (DGs), the most attractive is the version with grazing incidence of radiation on the DG (angle of incidence 86°–89°) [7, 8]. With this cavity configuration, the dispersion of the DG is higher in comparison with the dispersion in the autocollimation regime. With an increase in the angle of incidence over 89°, the adjacent grating grooves are shadowed by the previous ones, and the grating efficiency decreases sharply. For the grating operation in the grazing-incidence regime, the cavity bandwidth is defined by the expression

$$\delta\lambda_{\rm res} \approx \frac{\lambda^2 \cos\beta}{w_0 X(1 + \sin\beta)},$$

where β is the diffraction angle; w_0 is the Gaussian beam waist; X is the DG telescopicity; and λ is the radiation wavelength [9]. In this case, the DG combines the functions of a dispersive element and a one-dimensional telescope, which significantly reduces the cavity length and increases the con-

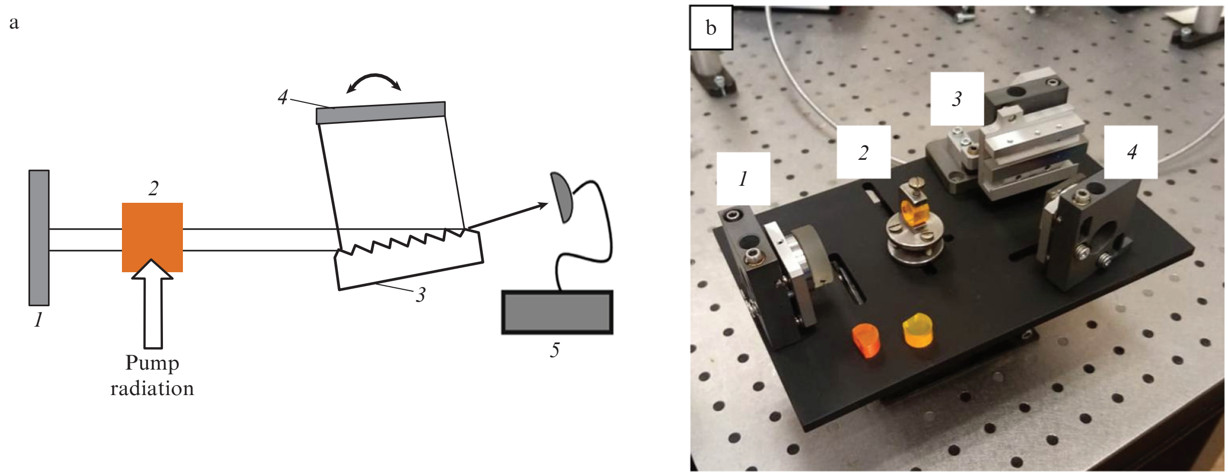

Figure 1. (Colour online) (a) Schematic of a tunable solid-state dye laser and (b) photograph of the setup: (*1*) nontransmitting mirror; (*2*) LAM; (*3*) DG; (*4*) folding tuning mirror (or DG in the autocollimation regime); (*5*) spectrometer; pumping was performed by a YAG: Nd^{3+} laser (LQ-529B).

version efficiency. An additional rotating mirror is used to control the tuning of the laser wavelength. The output radiation is the zero-order beam reflected from the DG. The cavity dispersion in this case is defined by the expression

$$\left(\frac{\partial\phi}{\partial\lambda}\right)_{\lambda_0} \approx \frac{2}{\lambda_0(\pi/2-\psi)},$$

where ψ is the angle of incidence of radiation on the DG, and in the spectral range 550 – 600 nm it can be ~0.2 nm^{-1}. In this case, the emission linewidth is as low as ~0.002 nm [10]. In the literature, this configuration is referred to as the Littman scheme.

The combined use of the above-described cavity configuration and a transverse pumping scheme greatly simplifies and reduces the cost of laser design, since there is no need for a dichroic mirror, which is used in the longitudinal pumping scheme. Its purpose, as is well known, is to freely transmit the pump radiation and be broadband with 100% reflection at the lasing wavelength over the entire tuning band. In the quasi-longitudinal scheme, there is no dichroic mirror, but the small length of the selective cavity makes it difficult to use this option. In a transverse arrangement, it is sufficient to use a cheap broadband nontransmitting aluminium mirror.

The selected version of the configuration of the tunable solid-state dye laser and a photograph of its layout are shown in Fig. 1. The cavity of the tunable laser consisted of a DG (1200 lines mm^{-1}), the normal to which was at an angle of 86°–89° with the axis of the incident beam, and two nontransmitting aluminium feedback mirrors, one of which was tunable, installed in the first diffraction order of the grazing-incidence grating. The radiation was extracted from the zero order of diffraction. In a number of experiments, the folding tuning mirror was replaced by a feedback DG (1200 lines mm^{-1}) operating in the autocollimation regime. The laser design made it possible to change the cavity length by about 50% by increasing the distance between the folding tuning mirror and the grazing-incidence DG.

To measure the energy characteristics of solid-state LAMs, we used Ophir Nova II and Gentec energy metres. The spectra were recorded with an AvaSpec-ULS2048 fibre-optic spectrometer. The intensity profile of the output laser radiation was measured using a Spiricon L11059M profilometer.

3. Results and discussion

Solid-state LAMs were synthesised according to the method described in Ref. [11]. Note that the Ch3 compound is known in the industry as Solvent red 197 and is widely used as a dye for various polymeric materials, and PM567 and PM597 are known as effective laser dyes. The structural formulas of the LAM compounds and a photograph of the solid-state active elements are shown in Fig. 2.

The laser elements were made in the form of a cylinder with a diameter of 10 mm and a length of 10 mm, truncated along the lateral surface, with a sample thickness of 7 mm in a plane perpendicular to the cylinder axis. Then, the ends of the element and the flat face were manually polished. After fabrication, all laser elements were tested for lasing efficiency with the use of a nonselective cavity. The facility is schematically shown in Fig. 3.

The LAM samples were pumped by the second harmonic of a neodymium laser with a wavelength of 532 nm by pulses with an energy of up to 30 mJ and a duration of 12 ns in a transverse excitation scheme. The cross section of the pump beam was a rectangle 10 × 0.5 mm in size formed by two crossed cylindrical lenses. The cavity was formed by the outer end face of the laser element and a plane aluminium mirror with 100% reflectivity. The main spectral-luminescent and lasing characteristics of the laser elements obtained using the non-selective cavity are shown in Table 1. A system of neutral light filters was used to vary the pump radiation intensity. In our earlier works, it was shown that in the case of a transverse excitation scheme, the pump intensity of 9 – 11 MW cm^{-2} was optimal in terms of service life and conversion efficiency, depending on the dye used.

Table 1 shows the lasing characteristics of the fabricated samples with two dye densities (10^{-3} and 5×10^{-4} M L^{-1}). Despite the fact that the efficiency of samples with a dye density of 10^{-3} M L^{-1} is higher than that of samples with a density of 5×10^{-4} M L^{-1}, in the proposed version of the laser, we used the samples with a lower dye density. It turned out that putting the samples with a dye density of 10^{-3} M L^{-1}

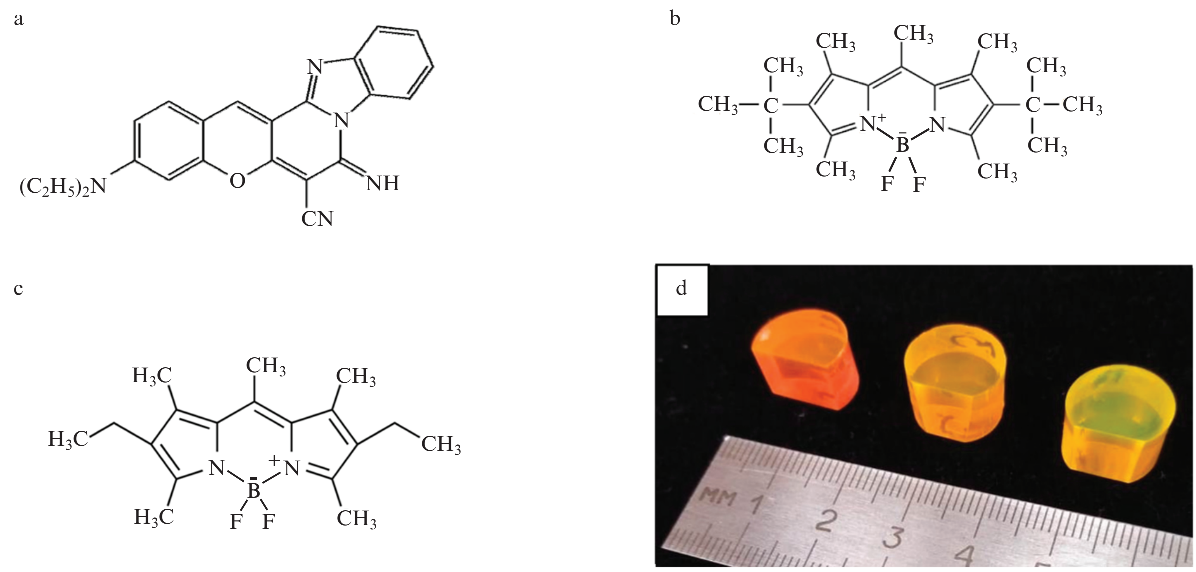

Figure 2. (Colour online) Structural formulas of LAM compounds: (a) 3-diethylamino-7-imino-7H-chromene [3′,2′-3,4] pyrido [1,2-a]-benzimidazole-6-carbonitrile (Ch3); (b) 1,3,5,7,8-pentamethyl-2,6-di-tert-butylpyrromethene-difluoroborate complex (PM597); and (c) 1,3,5,7,8-pentamethyl-2,6-diethylpyrromethene-difluoroborate complex (PM567), as well as (d) a photograph of the active elements.

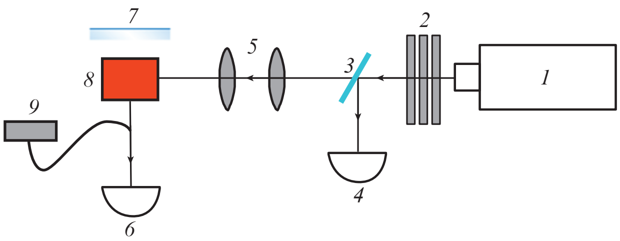

Figure 3. Schematic of the facility for measuring the spectral-luminescent and lasing characteristics of the LAMs:
(*1*) YAG: Nd3+-laser (LQ-529B); (*2*) system of nonselective light filters; (*3*) beam splitter; (*4*) Gentec energy metre; (*5*) a system of crossed cylindrical lenses; (*6*) Ophir Nova II energy metre; (*7*) nontransmitting mirror; (*8*) LAM; (*9*) Avantes spectrometer.

into the selective resonator entailed a significant narrowing of the tuning band in comparison with that for samples with a dye density of 5×10^{-4} M L^{-1}. This effect is due to the appearance of parasitic amplified spontaneous emission at the maximum of the LAM luminescence band when the

Table 1. Spectral-luminescent and lasing characteristics of laser elements for a pump radiation intensity of 9 MW cm^{-2} and the number of pulses greater than 10^5.

LAM	Dye density/M L^{-1}	Efficiency (%)	λ_{gen}/nm	$\Delta\lambda_{lum}$/nm
Ch3 in PMMA	5×10^{-4}	49	605	52
Ch3 in PMMA	10^{-3}	53	606	52
PM567 in PMMA	5×10^{-4}	60	563	60
PM567 in PMMA	10^{-3}	67	563	60
PM597 in PMMA	5×10^{-4}	23	566	26
PM597 in PMMA	10^{-3}	27	566	26

wavelength of the laser line is 5–8 nm away from the maximum wavelength. In this case, the regime of two-band lasing is realised, and on further tuning-away, lasing on the tunable line is disrupted.

For a solid-state sample based on PM567, we studied how varying within 86°–89° the angle of radiation incidence on the grazing-incidence DG affected the lasing linewidth. We also studied for the PM567-based sample the effect of cavity length on the conversion efficiency at a given laser line halfwidth. An aluminium mirror with 100% reflection or a DG (1200 lines mm^{-1}) operating in an autocollimation regime was used as a tuning mirror. Figure 4a shows the half-widths of the laser line and the conversion efficiency. When the highly reflecting aluminium mirror was replaced by the autocollimation grating, it was possible to obtain a two times narrower laser line (less than 0.02 nm) at an angle of incidence of 88°, the conversion efficiency in this case being 20%.

With an increase in the total cavity length by 46% (from 11 to 19 cm), the conversion efficiency decreases by 6% for a maximum value of 20% reached in the case of a short cavity. When the mirror was replaced with a grating, the overall efficiency of the oscillator lowered only slightly (by 2%).

As an example, Fig. 5 shows the typical distribution of the radiation intensity of the PM567-based LAM, which is fairly symmetric about the spot centre and is close to the Gaussian distribution.

Figure 6a shows the normalised gain profiles of the LAMs under investigation. The shaded part shows the total region of the oscillation wavelength tuning. Figure 6b shows the LAM tuning characteristics in the selective cavity. The maximum tuning range was obtained for Ch3 and was 52 nm. The difference between the shape of the tuning curve and the gain curve is explained by the fact that the spectrum of spontaneous emission of Ch3 consists of three bands, the middle band being highest in intensity. The PM567 and PM597 dyes permit expanding the tuning range towards shorter wavelengths. For both dyes, it is equal to 25 nm.

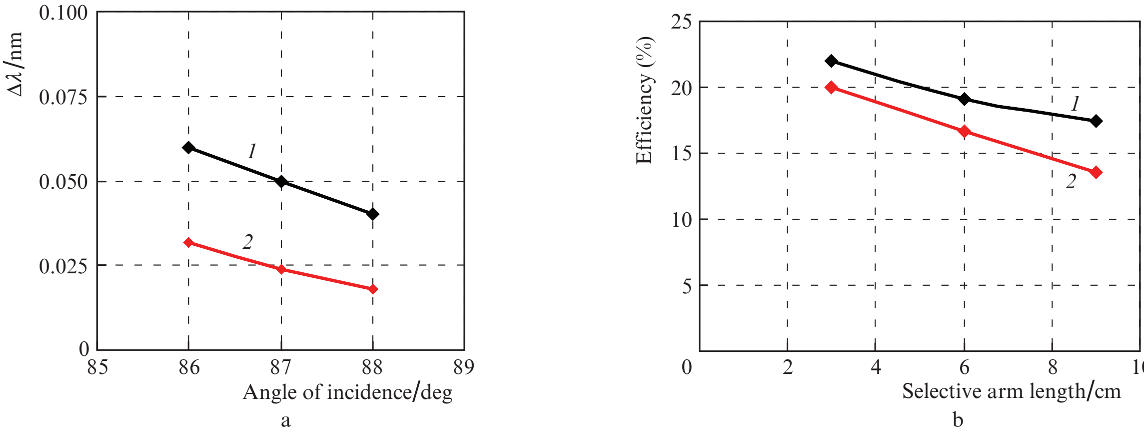

Figure 4. (Colour online) Dependences of (a) the laser line half-width on the angle of incidence on the DG as well as of (b) the selective-cavity laser efficiency on the distance between the tuning mirror and the grazing-incidence DG (on the selective arm length) when an aluminium mirror (*1*) and DG (*2*) are used as a tuning mirror.

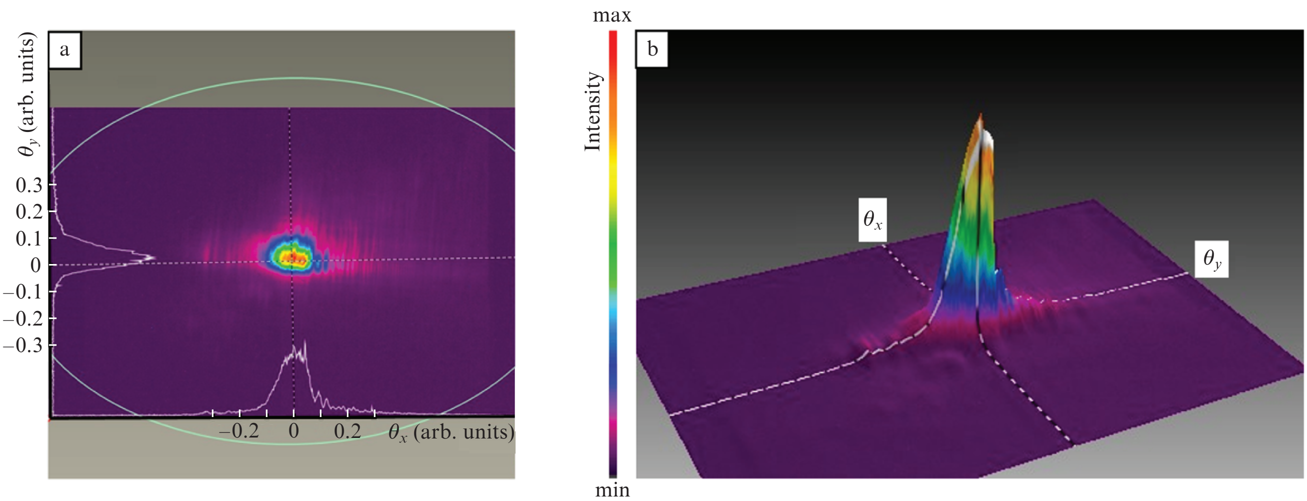

Figure 5. (Colour online) (a) Far-field radiation intensity distribution of a solid-state PM567-based LAM and (b) three-dimensional view of this distribution. The radiation pulse energy is 5 mJ, the wavelength is 560 nm, and the linewidth is 0.018 nm. The radiation of the laser pump propagates parallel to the abscissa. The distribution was obtained using a Spiricon L11059M profilometer. The green oval in Fig. 5a is the boundary of the recording area of the profilometer.

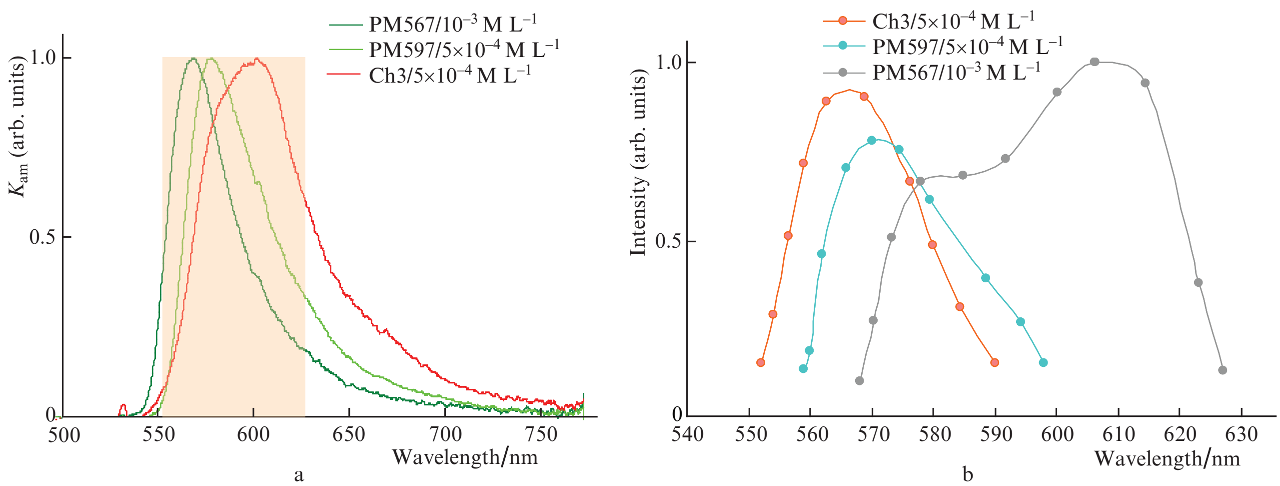

Figure 6. (Colour online) (a) Normalised gain contours of solid-state Ch3-, PM567-, and PM597-dye based LAMs, and (b) tuning characteristics of these LAMs.

4. Conclusions

In this paper, we present the results of investigations of the lasing characteristics of an organic solid-state tunable laser based on Ch3-, PM597-, and PM567-dye doped PMMA pumped by the second harmonic of a YAG:Nd^{3+} laser (532 nm). The features of lasing with the use of non-selective and selective cavities were noted. We investigated how the angle of radiation incidence on the grazing-incidence DG affected the width of the laser line and how the cavity length affected the conversion efficiency at a given half-width of the laser line. It was found that replacing a highly reflecting aluminium mirror by an autocollimating DG allowed us to obtain a laser line twice as narrow (less than 0.02 nm) at an angle of incidence of 88°. The conversion efficiency in this case was 20%. For the three organic LAMs in the yellow and red regions of the spectrum, the laser linewidth was 0.018 nm, and the total tuning range was 78 nm. The results obtained showed that the synthesised solid-state organic media hold promise for the development of solid-state tunable dye lasers using grazing-incidence configurations. Realisable in this case is a simple and technology-friendly design of a tunable laser with radiation characteristics that are not inferior to its liquid counterparts.

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