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RESEARCH

Excitation of Laguerre–Gaussian and geometric modes in a dye laser

Olga Burdukova[1,2] · Vladimir Petukhov[1] · Mikhail Semenov[1] · Yuri Senatsky[1]

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Abstract
Investigations of structured laser beams formation are one of the attractive areas of laser research in recent years. Compared to solid-state lasers, dye lasers were rarely in demand in these studies. Here we present several transverse mode selection experiments in a pulsed Rhodamine 6G dye laser ($\lambda = 580$ nm) with a plano-spherical resonator under hollow beam pumping by 20 ns pulses of 532 nm radiation from the Nd:YAG laser 2-nd harmonic. Along with high-order vortex and petal Laguerre–Gaussian modes, a whole family of non-planar geometric modes of different structures was obtained at the frequency degenerate states of this dye laser resonator. The ensemble of geometric modes in the related degenerate resonator configurations is illustrated by fractal frequency spectrum calculations. The experiments performed show that a dye laser represents a fairly simple and convenient object for testing various mode selection techniques. Dye lasers operating in the visible and near-infrared spectral regions can complement the range of solid-state laser sources currently used for obtaining and studying structured laser beams, which have many applications in scientific research and applied problems.

1 Introduction

The study of structured laser beams, which have numerous applications in scientific research and applied problems, is one of the attractive fields of laser physics for several last decades. Structured beams at the laser output arise due to variety of resonator transverse modes. Several comprehensive reviews and original studies on methods of transverse modes selection in lasers, both "classical" like Hermite–Gaussian, Laguerre–Gaussian, Ince–Gaussian and so-called geometric modes were published recently, see for example [1–5]. The "toolkit" for controlling the mode composition inside a resonator includes various techniques that affect the amplitude, phase, polarization of laser radiation, its gain and losses [1–6]. The recent impressive achievement here was the "digital laser" based on an intra-cavity spatial light modulator (SLM), with which one can set almost any required beam profile on the SLM matrix and obtain it at the laser output [7]. However, as the use of SLMs is limited by their low radiation resistance, more simple and cheap intracavity devices and different methods of mode selection are still in demand. One of such techniques that continue to be actively studied and applied is the mode selection by controlling the inversion spatial distribution in the laser medium.

Annular pumping of an active medium turned out to be an effective method of selecting Laguerre–Gaussian modes (LG_{pl}, p,l—radial and azimuthal indices) of the LG_{0l} type (annular beams) [8, 9]. This approach was successfully used in a number of subsequent experiments. The overwhelming majority of work on mode selection has been carried out with solid-state lasers. In the present work, we performed mode selection experiments in a dye laser. Dye lasers were rarely in demand in such type of experiments and they are significantly inferior in energy and radiation power to solid-state lasers. However, as will be shown, mode selection experiments with a dye laser are quite illustrative as well. We applied schemes of hollow beam pumping similar to [8, 9] to a Rhodamine 6G (Rh6G) dye laser with a plano-spherical resonator, operating near the wavelength $\lambda = 580$ nm. Along with annular vortex and petal modes of the LG_{0l} type, non-planar geometric modes of different structures were obtained in the frequency degenerate states of the resonator. Several non-planar modes, as far as we know, were obtained experimentally for the first time. The ensemble of geometric modes in the related degenerate configurations of a resonator is illustrated by fractal frequency spectrum calculations.

✉ Olga Burdukova
burdukovaoa@lebedev.ru

1 Division of Quantum Radiophysics, P.N. Lebedev Physical Institute of the Russian Academy of Sciences (LPI RAS), Moscow, Russia 119991

2 I.M. Sechenov First Moscow State Medical University, Moscow, Russia 119991

The results obtained confirm the validity of the presented methods of mode selection and the usage of dye lasers for structured laser beams formation.

2 Excitation of Laguerre–Gaussian and geometric modes in a Rh6G dye laser under hollow beam pumping

Elements of the Rh6G dye laser and the pumping system are shown in the scheme at Fig. 1. A glass laser cuvette with a wall thickness of 1.3 mm and a gap of 1 mm, filled with a solution of the Rh6G dye in ethyl alcohol (optical density D ≈ 2.5 at $\lambda = 532$ nm), was installed in a plano-spherical resonator with an effective length L (including the cuvette). The reflection coefficients of the spherical mirror M1 with a radius $R = 100$ mm and the flat output mirror M2 at the dye laser wavelength $\lambda = 580$ nm were 99% and 88%, respectively. The dye laser was pumped by 20 ns pulses of the 2nd harmonic of a multimode Nd:YAG laser. The spherical mirror transmitted 80% of the pump radiation at $\lambda = 532$ nm. The energy of linearly polarized pump pulses in the range of 0.1–3 mJ at the output of the Nd:YAG laser was adjusted by the attenuator 1 based on an $\lambda/2$ plate and a polarizer. The oval-shaped Nd:YAG laser beam had a non-uniform intensity distribution across its cross section. The beam passed through a lens f_1, contracted to a size of ≈1 mm and directed into the glass cuvette 2 (1 mm gap) with a dense mixture of LiF crystal particles (70–140 μm) and isobutyl alcohol (LiF/iso) with similar refractive indices, n ≈ 1.39. The LiF/iso mixture is transparent in the visible and near-IR regions of the spectrum and manifests itself in this region as a small-angle radiation scatterer (analogue of a Christiansen filter) [10]. A steel ball with ⌀380 μm was placed in the mixture in order to form a hollow pump beam at the output of the cuvette 2.

An oval hollow pump beam was projected by an f_2 lens into the dye solution at the front wall of the laser cuvette. A photo of the pump beam and its profile are shown in Fig. 2. The laser cuvette was located at a distance of ≈35 mm from the spherical mirror. The pump radiation was almost completely absorbed already in the first 400 μm of the dye solution. In a number of experiments, a ⌀2 mm diaphragm was installed in the resonator in front of the cuvette, which cut the periphery of the pump beam and contributed to the formation of annular laser beams. Low-intensity 532 nm radiation at the center and the periphery of the pump spot, which arose presumably due to pump beam diffraction and scattering, had no noticeable effect on the dye laser beam formation.

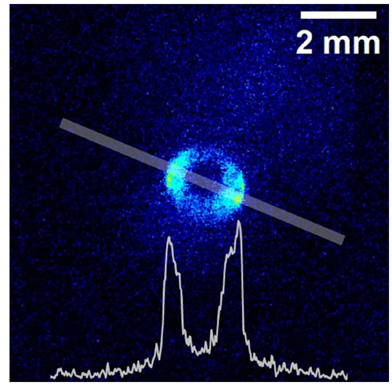

Fig. 2 An image and a profile of the pump spot inside the Rh6G dye laser cuvette when pumped according to the Fig. 1 scheme

The study of the dye laser was carried out at various cavity lengths by moving the flat output mirror M2 from a semi-confocal to a near semi-concentric configuration (L = 50–99 mm). In this case, the radius of the fundamental ω_{00} mode in the cuvette varied from ≈100 μm to ≈270 μm (calculation). When pumped near threshold, 15–20 ns pulses of linearly polarized dye laser radiation with an energy of up to 0.4 mJ at $\lambda = 580$ nm were obtained. Laser beams in the near or far field were projected by an f_3 lens (500 mm focal length) onto a CCD camera. Figure 3 shows the far-field profiles of laser beams when a ⌀2 mm diaphragm was used in the resonator. When mirror M2 was moved, annular beams with a smooth intensity distribution like Fig. 3a alternated with beams consisting of individual spots distributed around a ring, Fig. 3b–d. The full angle divergence of the recorded laser beams was 20–25 mrad. Both smooth and

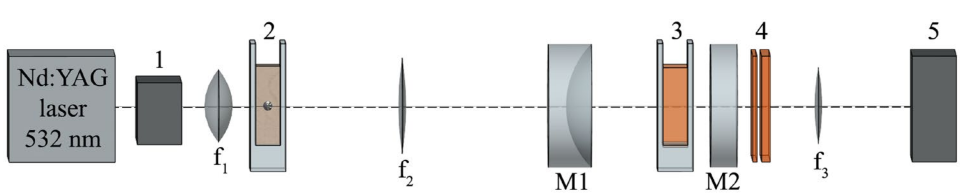

Fig. 1 Scheme of the experiment with Rh6G dye laser pumped by the 2nd harmonic of the Nd:YAG laser via a cuvette—immersion diffuser with a steel ball on the pump axis. 1—attenuator; 2—cuvette-diffuser (LiF particles of 70–140 μm in isobutanol) with a ball; 3—laser cuvette (1 mm gap filled with an Rh6G dye solution in ethanol); 4—long-pass filters; 5—CCD camera; f_1, f_2, f_3—lenses, M1—concave mirror with radius of curvature R = 100 mm, M2—output coupler, $R = \infty$

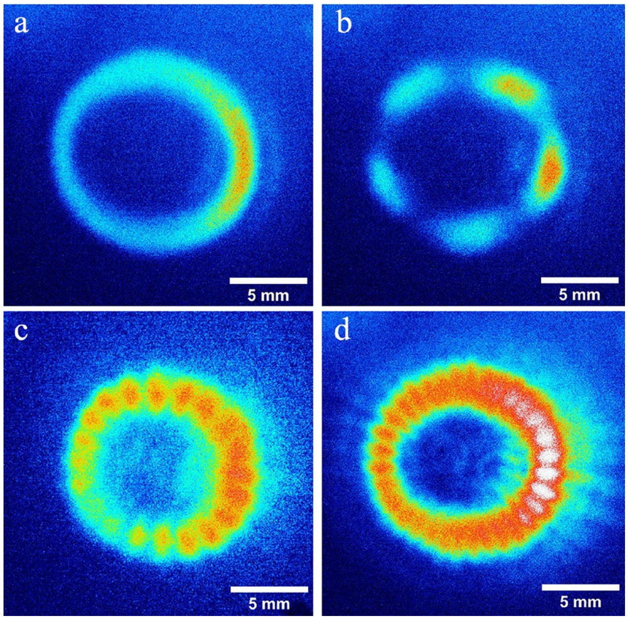

Fig. 3 Far field profiles of Rh6G dye laser beams when using an intra-cavity ⌀2 mm diaphragm

structured annular beams at the exit from the laser resonator propagated without changing the profile, that is, they manifested themselves as laser modes. Taking into account the cylindrical symmetry of the resonator, structureless annular beams should be classified as Laguerre–Gaussian $LG_{0,1}$ vortex modes. The structured beams were similar to $LG_{0,1}$ petal modes, which are known to be a result of coherent addition of 2 vortex modes with opposite helicities [1–6]. Registration of the profiles in Figs. 3a–d was carried out at small adjustments of the resonator length L around the values 9 cm and 10 cm, respectively. The radii of the fundamental mode at the active element were $\omega_{00} = 270\ \mu$m at $L = 99$ mm (Fig. 3c), $\omega_{00} = 220\ \mu$m at $L = 97$ mm (Fig. 3d) and $\omega_{00} \approx 150\ \mu$m at $L \approx 90$ mm (Fig. 3a, b). Thus, the ratios of the radii of the pump beam in the cuvette $\omega_{pump} = 0.9$ mm and the ω_{00} values were 3–6 and corresponded to the conditions for excitation of high-order modes, like petal modes $LG_{0,10}$ and $LG_{0,16}$ shown in Fig. 3c, d.

Unlike $LG_{0,1}$ petal modes, which must contain an even number of petals, the beam presented in Fig. 3b contained an odd number of spots and caused confusion. The origin of structures with odd numbers of spots was clarified in experiments without an intra-cavity diaphragm. Under these conditions, when the resonator mirror M2 moved, oval-shaped laser beams with a smooth intensity profile appeared (like Fig. 3a), alternating with structured beams, consisting in most cases of an odd number of spots along the ellipse (Fig. 4). Figure 4a–c, e, f show photos of beam profiles with odd (3, 5, 7, 11, 13) numbers of spots obtained at certain values of the ratio of the cavity length L to the mirror M1 radius R in the interval L/R = 0.5–0.95. Beams with an even number of spots on the ellipse (4 and 8) were observed at $L = 0.5R$ and $0.85R$, respectively.

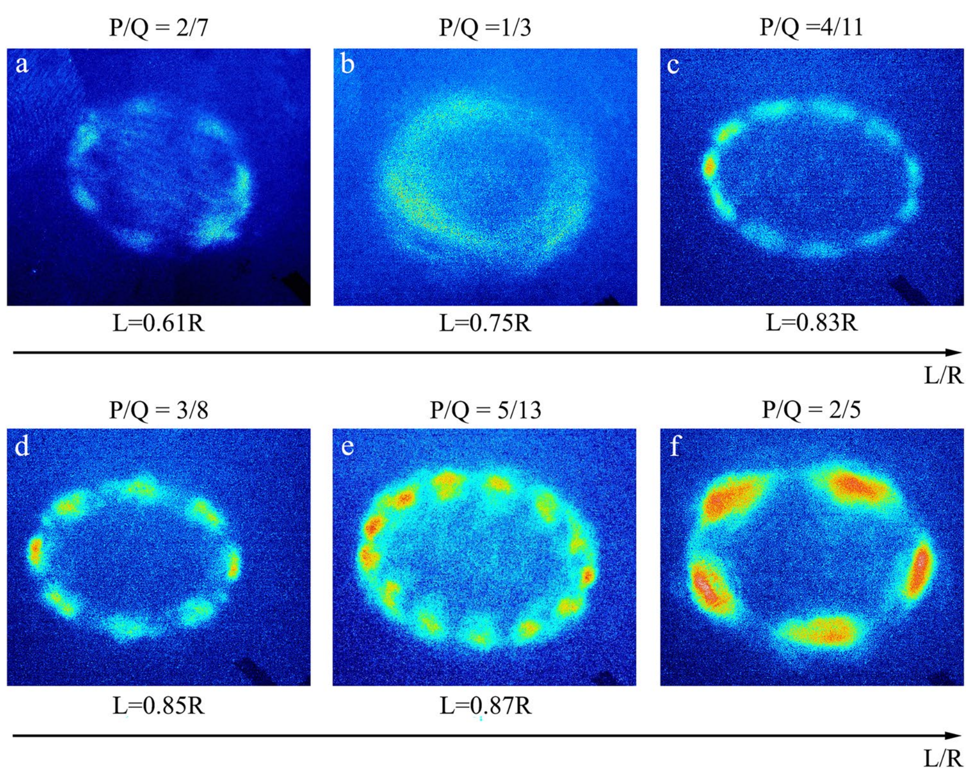

Fig. 4 Far field profiles of non-planar geometric modes for several frequency-degenerate resonator configurations marked by parameters P/Q and L/R, obtained in a Rh6G dye laser under non-uniform hollow beam pumping

Generation of beams with 3 (Fig. 4b) and 4 (photo not shown) low-intensity spots was difficult, probably due to the weak overlap of these beam structures with the pump profile. A comparison of all these dye laser beam profiles with the known beam profiles of geometric modes of solid-state lasers [2, 3] indicates the nature of the modes in Fig. 4 as a family of non-planar geometric modes. The distribution of geometric mode structures not on circles, but on ellipses (Fig. 4) is apparently associated with the inhomogeneous oval profile of the pump beam. Profiles on Fig. 4 can be compared in qualitative form to the data [11], where the formation of several types of non-planar elliptical geometric modes was observed during asymmetric selective end-pumping of a Nd:YVO_4 laser in a plano-spherical resonator. In works [2, 3, 11–13] and in a number of other publications, non-planar modes with 3, 4, 5 and 7 spots were observed. In our experiments, we observed these modes as well, and we also obtained non-planar modes with 8, 11 and 13 spots of azimuthal structures (Fig. 4c, d, e).

The cavity is called the frequency degenerate one, if the ratio $\Delta f_T/\Delta f_L = P/Q$, where Δf_L is the longitudinal mode spacing, Δf_T is the transverse mode spacing in the given resonator and P and Q are co-prime positive integers [2, 3]. For a given normalized length L/R of the degenerate resonator, the type of the trajectory and the number of spots in the structure of the geometric mode are determined from the relation $L/R = \sin^2(\pi P/Q)$ [2, 3]. To check the consistency of our experimental results with the theoretical data, we calculated the frequency-degenerate spectrum of the ideal plano-spherical resonator as a function of the normalized length L/R, similar to [13]. The eigenmode frequencies of the resonator are determined by the relation

$$
\begin{aligned}
f_{n,m,s} &= \left[s + (n+m+1)\frac{\Delta f_T}{\Delta f_L}\right]\Delta f_L \\
\frac{\Delta f_T}{\Delta f_L} &= \frac{1}{\pi}\arctan\left[\frac{L}{\sqrt{L(R-L)}}\right],
\end{aligned}
\tag{1}
$$

where n, m and s are the transverse and longitudinal mode indices respectively [2, 3].

The results of calculating the ratio $(f_{n,m,s} - f_{n_0,m_0,s_0})$ to Δf_L are presented in Fig. 5 for resonator lengths L from $0.5R$ to R, with indices $|n - n_0| < 15$; $|m - m_0| < 15$; $|s - s_0| < 15$, where n, n_0, m, m_0 and s, s_0 are the orders of transverse and longitudinal modes. Red arrows mark the positions of the degenerate states realized in our experiments.

Using pumping with improved symmetry, we were unable to excite separately geometric modes in a Rh6G dye laser assembled according to the same scheme as in Fig. 1. To obtain more uniform pumping, the beam of the multimode Nd:YAG laser was passed through a fiber with a core diameter of 550 μm and a length of 10 m (a cuvette-diffuser with a ball was not used). After collimating the beam at the output of the fiber, an axicon/lens pair was used to obtain hollow pump beams [5]. Pump radiation at the fiber output was non-polarized. In this case dye laser radiation was non-polarized as well. A photo and a profile of the pump spot formed with an axicon/lens pair are shown in Fig. 6. Figure 7a shows one of the typical photo of a structureless annular dye laser beam ($LG_{0,1}$ vortex mode), obtained outside degenerate resonator configurations when pumped with an axicon/lens pair. In degenerate configurations, lasing on $LG_{0,1}$ modes was also observed, but some marks of non-planar geometric modes structures appeared often over these modes profiles. Lasing on modes with 3, 5, 8 spots and some others was observed.

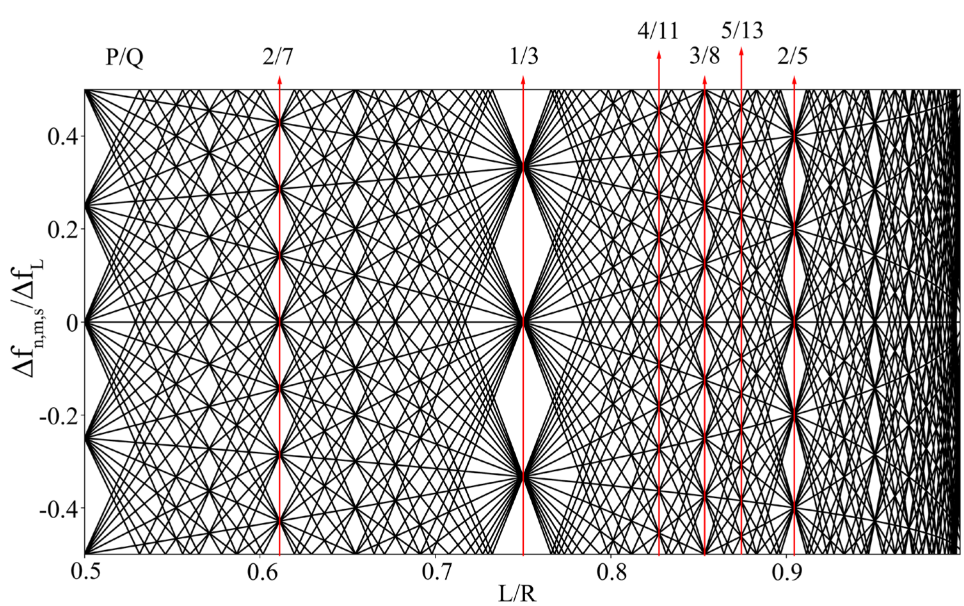

Fig. 5 Fractal frequency spectrum as a function of a normalized cavity length ($L/R = 0.5 - 1.0$) for the ideal plano-spherical cavity. Red arrows indicate some of frequency-degenerate P/Q states. The calculation was carried out for $|n - n_0| < 15$; $|m - m_0| < 15$; $|s - s_0| < 15$

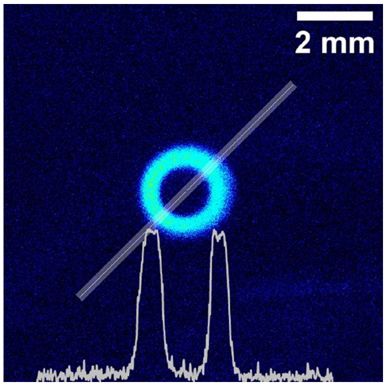

Fig. 6 An image and a profile of the pump spot inside the Rh6G dye laser cuvette when pumped using a fiber and an axicon/lens pair

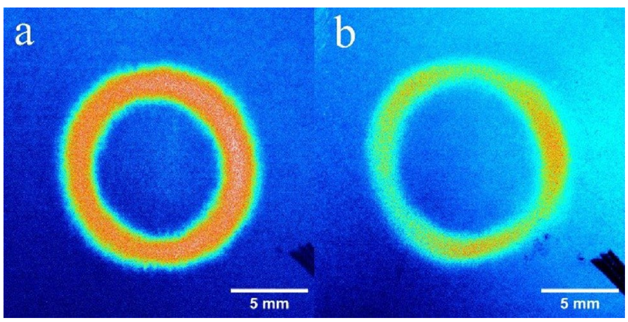

Fig. 7 Far field profiles of Rh6G dye laser beams outside (a) and inside (b) the resonator degeneracy region ($L = 0.75R$, $P/Q = 1/3$) when pumped with a fiber and an axicon/lens pair

An example of such a superposition of an $LG_{0,1}$ annular beam and a geometric mode structure with 3 spots is presented in Fig. 7b.

3 Discussion

Since the first publications on dye lasers, research on mode selection in these lasers has focused mainly on methods for obtaining lasing with a narrow linewidth and a single longitudinal mode [14, 15]. The number of works on transverse mode selection in dye lasers is extremely limited compared to the huge number of publications on this topic for solid-state lasers. Methods for obtaining the fundamental TEM_{00} and the TEM_{01} modes in dye lasers were reported in [16, 17]. In the work [18] on a CW Rh6G dye jet laser study, the authors presented photos of higher-order Hermite–Gaussian modes as illustrations of the achieved optical homogeneity of the active medium. Due to the recent increased interest in the development of methods for mode selection in lasers and obtaining structured laser beams of various types, in the present work we tried to demonstrate the possibility of including into this problem a family of laser sources in the visible and near-IR ranges based on dye lasers. Although dye lasers cannot compete with solid-state lasers in terms of energy and output power, our experiments show that a dye laser represents a fairly simple and convenient test object for developing mode selection methods and obtaining structured laser beams. The large absorption cross section of pump radiation and the high gain in the dye laser make it possible to use small volumes and thicknesses of the active medium in mode selection experiments and to work at low radiation loads with pump and lasing pulses energies ≈1 mJ. Note also that when one used a dye solution (an isotropic active medium), there are no problems with laser alignment and mode selection associated with the active medium anisotropy which, for example, were reported by the authors of the publication [11] who worked with the $Nd{:}YVO_4$ crystal.

To excite annular transverse modes in a dye laser with a plano-spherical resonator, we used a known approach: profiling the pump beam and obtaining a hollow profile of inversion in the active medium. In the first series of experiments, a cuvette was introduced into the pump optical channel: an immersion diffuser with a dense mixture of LiF crystal particles and isobutanol, containing an obstacle—a small steel ball. In papers [10, 19, 20] we reported the use of a cuvette-diffuser in the resonators of dye lasers and neodymium lasers to produce radiation with low spatial coherence. The diffuser in the resonator divided the laser aperture into fragments with a chaotic spread of the optical path length, promoting the development of low-coherence lasing. The possibility of using an immersion diffuser in pumping systems for lasers is also of some interest, since the diffuser allows, by changing the level of immersion and causing nonlinear effects in the mixture, to influence the spatial-angular characteristics of the pump radiation passing through the cuvette. When anisotropic particles are used in the diffuser, the polarization of the pump radiation can also be influenced. The practical implementation of all these possibilities requires, however, separate studies, which were not carried out within the framework of this work.

In the present work, we used a cuvette-diffuser with a LiF/iso mixture primarily as a convenient hosting for an opaque obstacle—a steel ball. When the image of the ball was transferred to the active medium, a hollow pump beam profile was formed in the laser cuvette. The optical scheme made it possible to adjust the size of the pump spot in accordance with the cavity configuration and mode selection tasks. A low-intensity Poisson diffraction spot that appeared when the image of the ball was transferred to the resonator was usually located outside the active medium and did not affect the laser operation. Since the radius of the hollow inversion profile in the dye active medium was several times greater than the radius of the ω_{00} mode, it was possible to excite vortex and petal LG modes of high orders, and in the points of the resonator frequency degeneracy, excitation of various non-planar geometric modes was possible. The preferential excitation of geometric modes rather than LG modes here was facilitated, apparently, due to the strongly inhomogeneous profile of the pump beam, individual maxima of which coincided with the discrete structures of geometric modes and supported their amplification rather than the generation of annular LG modes. Various non-planar geometric modes with the number of spots from 3 to 13 were observed in our experiments. The numerical simulation of the fractal frequency spectrum of the laser resonator as a function of a normalized cavity length (Fig. 5) confirms the experimental results shown in Fig. 4.

To improve the homogeneity of the pump spot, in the second series of experiments the Nd:YAG laser beam was passed through a fiber, at the output of which an axicon/lens pair was used [5] to obtain a hollow pump beam profile. In this case, the simultaneous lasing of high order LG modes and non-planar geometric modes was recorded at the resonator frequency degenerate configurations. We associate the joint lasing of geometric and LG_{0l} modes here due to an almost uniform ring inversion profile that was formed in the dye layer and can support both LG_{0l} and geometric modes, Fig. 7b. Outside the regions of resonator degeneracy, we observed separate generation of a dye laser in LG modes, Fig. 7a. Research on optimizing regimes of pumping and the formation of Laguerre–Gaussian and geometric modes in dye lasers is planned to be continued. The formation of beams both on traditional modes like Hermite–Gaussian, Laguerre–Gaussian et al., and on geometric modes in dye lasers may help complement areas of application of structured laser beams in scientific research and applied problems, discussed in [1–4].

4 Conclusion

In this work, demonstration experiments on transverse mode selection in a liquid dye laser were performed. Schemes of hollow beam pumping were applied to an Rh6G dye laser with a plano-spherical resonator pumped by 20 ns pulses of the 2nd harmonic of Nd:YAG laser. Along with Laguerre–Gaussian vortex and petal modes of the LG_{0l} type, a family of non-planar geometric modes of different structures was obtained in the frequency degenerate states of the resonator. Several non-planar modes were obtained, as far as we know, for the first time. The experiments performed showed that a dye laser represents a fairly simple and convenient test object for developing mode selection methods. Dye lasers operating in various parts of the visible and near-infrared spectrum can complement the range of solid-state laser sources currently used for obtaining structured laser beams, which have many applications in scientific research and applied problems.

Author contributions Yu.V. Senatsky and V.A. Petukhov wrote the main manuscript text . O.A. Burdukova and M.A. Semenov prepared the calculations and the figures. All authors participated in the experiments and reviewed the manuscript.

Data availability No datasets were generated or analysed during the current study.

Declarations

Conflict of interest The authors declare no competing interests.

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