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Liquid laser cavities and waveguides. IV. Line lasing spectra

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Abstract. The unusual line structure observed earlier in the emission spectra of dye-laser liquid macrocavities is explained by the presence of transparent outer shells in which the generated and interfering laser radiation partly propagates, as in a complex cavity.

As already mentioned [1 – 3], a regular line (mode) structure has been observed in a number of experiments with liquid cavities in the usually broad-band and smooth emission spectra of dye lasers.

In conformity with the theoretical study [4], the presence of modes in a spherical drop is determined by the dimensionless Mie parameter

$$x = 2\pi r \lambda^{-1} , \tag{1}$$

(where r is the drop radius and λ is the radiation wavelength) and by the relative refractive index $m = n_1/n_2$ (where n_1 and n_2 are, respectively, the refractive indices of the drop liquid and of the surrounding medium). The spacing between neighbouring modes for spherical drops is given by the following expression for large values of x:

$$\Delta\lambda = \frac{\lambda^2}{2\pi r}\Delta x , \tag{2}$$

where

$$\Delta x = \frac{\tan^{-1}(m^2-1)^{0.5}}{(m^2-1)^{0.5}} . \tag{3}$$

The spacings between the modes observed in our experiments reached several nanometres, which corresponds to linear dimensions of the cavities of the order of fractions of a millimetre. Such dimensions do not match at all the $\sim$1 cm perimeters of the drops employed. The reason for the appearance of the line spectra in these experiments could not be discovered because this kind of mode structure was poorly reproducible, i.e. did not appear in all the forms of liquid cavities and in every experiment.

The physical reason of the appearance of the microlaser mode line structure in our macrocavities became better understood after its controlled and reproducible observation under the following conditions. A solution of rhodamine B in ethanol was poured into a cylindrical glass test tube with an external diameter of 8 mm and a wall thickness of about 0.9 mm. Lasing was excited by radiation with $\lambda = 532$ nm focused into a horizontal strip approximately 5 mm long and 0.5 mm high. The radiation outcoupling was enhanced sometimes by clamping a glass prism to the test tube. Since the refractive index of glass ($n = 1.5$) is greater than that of ethanol ($n = 1.35$), the laser radiation in the solution incident on the ethanol – glass interface does not undergo total internal reflection and this appreciably lowers the cavity Q-factor, as we have already demonstrated for a drop of ethanol in kerosene [3].

It was expected that there would be a high lasing threshold in such a cavity without total internal reflection and that the generated radiation would be unable to propagate along the boundary of the solution and would therefore emerge not from two but from only one front point of the ring, i.e. directly from the pump zone. However, it was found that, for a thin glass wall (Fig. 1), the laser radiation can undergo total internal reflection from the external glass – air interface, which ensures a low excitation threshold. In a side view, the laser radiation emerges from the front and from the rear point of the ring and, what is especially important, has a reproducible line spectrum with a distinct mode structure, illustrated in Fig. 2. The mode spacing is about 0.25 nm. This corresponds to a circular cavity [4] with a radius of 0.23 mm or a linear cavity 1.4 mm long, which is close to the optical thickness of the cell wall.

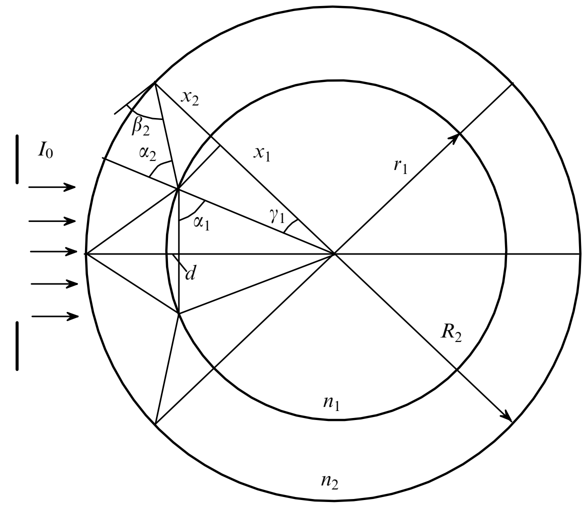

Figure 1. Schematic illustration of the beam paths in a laser cavity consisting of a liquid medium having a refractive index n_1 and an outer shell having a refractive index n_2 with total internal reflection from the outer shell – air interface.

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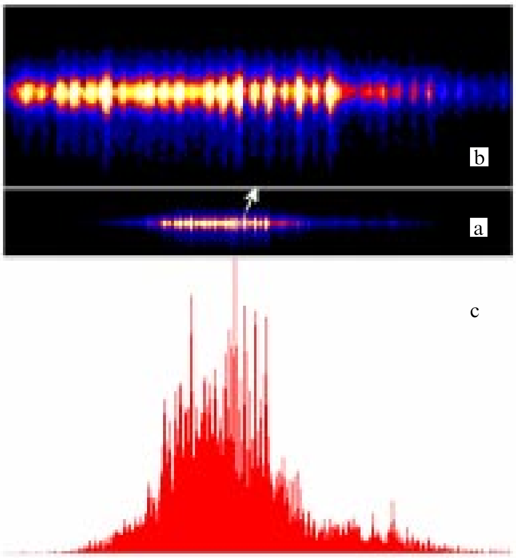

Figure 2. Emission spectrum of a liquid laser (a) in a glass shell with an external diameter of 8 mm and a wall thickness of 0.9 mm for the pump energy $E = 0.24$ mJ and a typical spectral mode spacing of 0.25 nm together, a section of this spectrum after threefold magnification (b), and the distribution of the radiation intensities along the spectrum (c) (the emission maximum is at a wavelength of 622.15 nm).

In similar experiments with a glass test tube (external diameter 15.8 mm, wall thickness 0.57 mm) with an ethanol solution of rhodamine, the lasing-mode spacing increased to 0.45 nm, as shown in Fig. 3. When a solution of rhodamine in dibutyl phthalate (DBP) ($n_1 = 1.491$) was poured into the same test tube (instead of the ethanol solution), the mode spacing in the lasing spectrum increased to 0.8 nm.

It became clear that the appearance of the modes in our experiments is caused by the influence of the complex cavity and is due to the presence of a thin shell in which the laser radiation partly propagates virtually without losses. It enters the laser solution, is amplified in it, and then propagates via other paths — both in the solution and in the shell (Fig. 1). The difference between these optical paths, the interference of the beams, and the complex intracavity angular and spectral selection of the radiation lead to the appearance of a mode line structure in it. Examples of calculations and modelling of the propagation of a beam through the shell within the framework of geometrical optics are presented below.

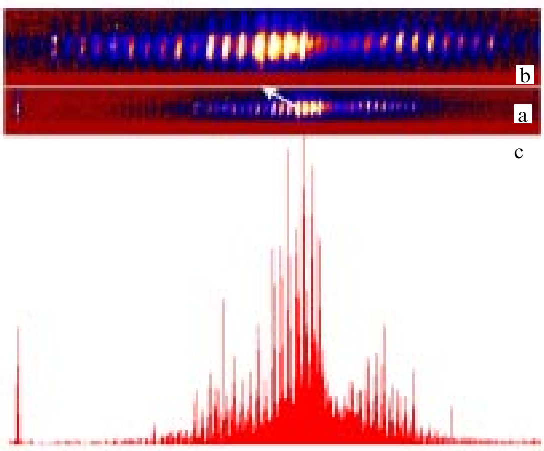

Figure 3. Emission spectrum of a liquid laser (a) in a glass shell with an external diameter of 15.8 mm and a wall thickness of 0.57 mm for the pump energy $E = 0.06$ mJ and a typical spectral mode spacing of 0.45 nm, a section of this spectrum after twofold magnification (b), and the distribution of the emission intensities along the spectrum (c) (the emission maximum is at a wavelength of 616 nm).

If the thickness of the surrounding glass wall is nonuniform, an additional modulation of the mode intensities with a period determined by the difference between the thicknesses of the wall during a round trip through it appears in the spectrum. It was demonstrated experimentally that, for a low pump energy and in the presence of optical inhomogeneities within the wall thickness, sometimes only one mode is the most intense in the lasing spectrum owing to additional selection (Fig. 4, curve 1). With increase in the excitation strip length D (and in the quantity d in Fig. 1), the number of modes also increases and they become less distinct (Fig. 4).

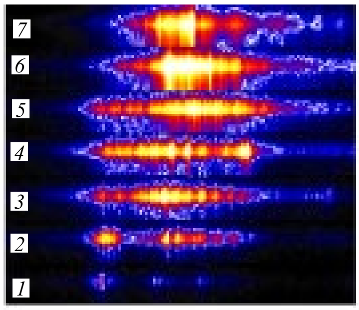

Figure 4. The influence of the pump energy E and the width of the pump zone D on the position and mode structure of the radiation of a liquid laser in a glass shell with an external diameter of 8 mm and a wall thickness of 0.9 mm for $D = 1.5$ mm and $E = 0.13$ mJ (1), $D = 2.0$ and $E = 0.23$ mJ (2), $D = 2.5$ mm and $E = 0.36$ mJ (3), $D = 3.0$ mm and $E = 0.34$ mJ (4), $D = 3.5$ mm and $E = 0.43$ mJ (5), $D = 4.0$ mm and $E = 0.55$ mJ (6), and $D = 9.0$ mm and $E = 0.722$ mJ (7) (the brightness has been reduced by a factor of 2). For $D = 0.8$ mm, there is no lasing.

The influence of the wall thickness on the mode structure was confirmed in experiments where the wall thickness was varied locally with the aid of a liquid DBP film deposited on the outside of the wall. This altered the positions of the intense modes in the lasing spectrum or led to their complete spreading to form a continuous spectrum. When the thickness of the DBP film was altered, the positions and intensities of the lasing modes were also changed. Consequently, the decisive parameter for the line structure of the radiation is indeed the thickness of the cell wall and the changes in it are clearly manifested by changes in the lasing spectrum.

Returning now to earlier studies [1 – 3], one may say that we already noted there that a thin shell appeared occasionally after a time on the DBP – water interface on the surfaces of the drops (when these were present in solutions under a thin layer of water for several days). This shell was particularly notable when the laser solution was cautiously removed from a suspended drop and the latter lost as a result its spherical shape. However, we usually carried out experiments on freshly prepared drops and such shells (and the mode structure in the spectra) were then absent.

Glass shells on liquid cavities with line spectra made it possible to achieve a reproducible demonstration of the high sensitivity of liquid cavities to minute changes in their

Figure 5. The change in the emission spectra of a liquid laser in a glass shell with an external diameter of 8 mm and a wall thickness of 0.9 mm after mechanical squeezing of the shell: the initial spectrum in the absence of mechanical influence on the shell (1, $E = 0.17$ mJ), the spectrum during the squeezing of the shell (2, $E = 0.15$ mJ), the spectrum immediately after the release of mechanical pressure (3, $E = 0.17$ mJ), and the spectra 1 min (4, $E = 0.16$ mJ), 10 min (5 $E = 0.16$ mJ), and 20 min after the removal of the pressure (6, $E = 0.14$ mJ).

shape, which was discussed earlier [1 – 3]. When the first glass shell was mechanically squeezed and the pressure was then released, the lasing spectra changed from line spectra to almost single-mode spectra (Fig. 5). After the complete removal of the mechanical pressure, the positions and the initial line structure of the lasing spectra were restored, but not immediately. This required $\sim$30 – 40 min, which is associated with the slow removal of the residual internal mechanical stresses in the glass. Measurement of the spectra of liquid cavities showed that the mechanical stresses in the glass shell persist for further tens of minutes after the removal of the external influence and that the spectra of liquid cavities reflect the small mechanical changes in the shape of the cavity.

In order to elucidate the influence of the shell on the mode structure of the radiation, we shall consider the problem of the beam travel in a liquid laser cavity consisting of a circular cylinder with an active medium having a refractive index n_1, surrounded by a cylindrical circular shell having a refractive index $n_2 > n_1$ (Fig. 1). We may note that this relationship between the refractive indices is diametrically opposite to the situation in waveguides for which usually $n_2 < n_1$.

Suppose that the radius of the active medium is r_1. It is necessary to determine the radius R_2 for which the laser beam travelling from the active medium and incident on the shell undergoes total internal reflection at the outer shell – air interface (needed for a laser cavity with a high Q-factor). For a light beam travelling along a chord, separated by a distance d (see the designations in Fig. 1) from the inner circle with a radius r_1, and entering the shell, we have

$$\alpha_1 = \arcsin\frac{r_1 - d}{r_1}\,, \tag{4}$$

$$\alpha_2 = \arcsin\left(\frac{n_1}{n_2}\sin\alpha_1\right)\,, \tag{5}$$

$$\beta_2 = \arccos\frac{1}{n_2}\,, \tag{6}$$

$$\gamma_1 = \pi - (\pi - \alpha_2) - \left(\frac{\pi}{2} - \beta_2\right)\,. \tag{7}$$

After introducing the notation $R_2 = x_1 + x_2$ (Fig. 1) and noting that

$$x_1 = r_1\cos\gamma_1\,, \tag{8}$$

$$x_2 = \frac{r_1\sin\gamma_1}{\tan(\pi/2 - \beta_2)}\,, \tag{9}$$

we obtain

$$\frac{R_2}{r_1} = \cos\gamma_1 + \frac{\sin\gamma_1}{\tan(\pi/2 - \beta_2)}\,. \tag{10}$$

Expression (10) can be simplified:

$$\frac{R_2}{r_1} = \frac{\cos\gamma_1\sin(\pi/2 - \beta_2) + \sin\gamma_1\cos(\pi/2 - \beta_2)}{\sin(\pi/2 - \beta_2)}$$

$$= \frac{\sin(\gamma_1 + \pi/2 - \beta_2)}{\cos\beta_2} = \frac{\sin\alpha_2}{\cos\beta_2} = \frac{(n_1/n_2)\sin\alpha_1}{1/n_2} = n_1\sin\alpha_1\,. \tag{11}$$

After substituting expression (4) into expression (11), we obtain

$$\frac{R_2}{r_1} = \frac{n_1(r_1 - d)}{r_1}\,. \tag{12}$$

For $d \ll r_1$, we have

$$\frac{R_2}{r_1} = n_1\,. \tag{13}$$

An extremely unexpected feature is that, for $n_2 > n$, the limiting value of R_2 (and the thickness of the shell) is altogether independent of n_2 according to formulas (12) and (13).

Thus the total internal reflection of light from the outer boundary of the shell is observed when the ratio R_2/r_1 [formula (12)] does not exceed n_1, which is 1.35 for ethanol. In other words, the thickness of the shell around the active laser medium should not exceed 35% of its internal radius or 26% of its external radius.

In our experiments with the first glass test tube, where a distinct line structure of the spectra was observed, the wall thickness was about 0.9 mm with the external radius $R_2 = 4$ mm, i.e. amounted to 29% of the internal radius or 22.2% of the external radius, which satisfies the condition for the total internal reflection of light at the outer boundary.

Despite the fact that in this kind of complex cavity with a shell light may propagate via many paths with incursions into the active medium and the shell, the path with the lowest losses (i.e. proceedings mainly via the glass and only partly via the active medium) and with the greatest gain (i.e. following the longest chord in the active medium for the specified length of the amplification section) has the highest Q-factor. Thus the beam, the point of the emergence of which from the active medium after a series of reflections coincides with the entry point, again travels along the longest chord and for this reason is amplified to the greatest extent and has the largest Q-factor.

Examples of the calculation of the paths of such beams are presented below, but yet another feature must be emphasised. The path followed by such a beam along a complex cavity consists of an integral number of identical 'cells', in each of which the beam splits into two beams after incidence on the glass – solution interface: one beam travels along glass after reflection, is reflected from the outer glass – air interface, and again travels to the glass – solution interface, where it encounters and interferes with the other beam which had penetrated into the solution and travelled in it along a chord. Thus the path with the highest Q-factor represents a set of an integral number of cavities with identical lengths and properties and the same mode structure. Among these 'cells', one is active (with amplification) while the others are passive, but the mode spacing in them is the same and is determined by the difference between the optical light paths in the glass and in the solution. Below are presented estimates which demonstrate that this difference between the paths is comparable with the shell wall thickness (i.e. is of the order of magnitude of millimetres) and actually determines the mode spacing [despite the long (centimetre) light path in the cavity throughout the length of the glass shell].

The fraction of the light penetrating into the active medium after reflection from the glass – solution interface depends on the polarisation. Most of the beam, the polarisation of which is perpendicular to the incident plane, penetrates into the amplifying medium and returns to the shell, but in subsequent 'shells' it again travels through the passive solution and is further absorbed. The beam, the polarisation of which is in the incidence plane, penetrates into the active medium to a lesser extent and after amplification a smaller beam fraction returns to the shell. However, in subsequent 'shells' it is absorbed to a lesser extent. We did not carry out a more detailed estimation of the losses for beams with different polarisations in a complex cavity of this kind, but measurements have shown that radiation with both types of polarisation and with comparable intensities is present in liquid ring cavities when lasing line spectra are generated.

We shall now quote examples of calculations of beam paths with an integral number of 'cells' for a liquid laser with a shell.

Example 1 (see the designations in Fig. 1). For a liquid cavity with $n_1 = 1.35$, $n_2 = 1.5$, $r_1 = 1$, $d/r_1 = 0.034077$, $\beta_2 = \arccos(1.0677/n_2)$, we obtain $R_2/r_1 = 1.22131$, $(\pi/2-\alpha_1)(180/\pi) = 15°$, $\gamma_1 = 15°$ by using formulas (4) – (10). For the given parameters, the beam enters precisely the starting point in the amplifying medium after twelve reflections from the outer glass – air interface. Thus, during the round trip through the shell the beam traverses 12 identical 'cells'. The optical difference between the paths followed by the two beams in each 'cell'. The optical difference between the paths followed by the two beams in each 'cell' is $\Delta l = 0.392$, which is comparable with the optical thickness of the shell, given by $0.22131 n_2 = 0.332$.

Example 2. In the case where $n_1 = 1.35$, $n_2 = 1.55$, $r_1 = 1$, $d/r_1 = 0.034112$, $\beta_2 = \arccos(1.0428/n_2)$, we obtain from formulas (4) – (10) $R_2/r_1 = 1.25043$, $(\pi/2 - \alpha_1)(180/\pi) = 15°$, $\gamma_1 = 15°$. For the given parameters, the beam traverses 12 identical 'cells' during the round trip through the shell. The optical difference between the paths followed by the two beams is $\Delta l = 0.493$, which is comparable with the optical thickness of the shell given by $0.25043 n_2 = 0.388$.

For the second test tube with ethanol, the light undergoes 24 reflections from the outer shell during the round trip through the cavity according to estimates, whereas the difference between the beam paths diminishes by a factor of 1.85, which actually leads to an increase in the mode spacing. Since the refractive index of DBP is close to that of glass, the difference between the beam paths diminishes, whereas the mode spacing increases to 0.8 nm.

The line structure of the spectra becomes less distinct (it spreads) when the wall thickness of the shell proves to be non-uniform along the circumference and when the gain band contains no wavelengths for which the length of the shell is not a multiple of an integral number of 'cells'. In this case, the lasing spectra are of the broad-band type usual for dye lasers without a visible mode structure.

Thus, it has been established that the mode structure in the lasing spectra of liquid laser cavities is associated with the presence of transparent outer shells in which the laser radiation generated partly propagates. The uniqueness of experiments described, involving the measurement of the line spectra, is that such experiments are extremely difficult to carry out in other condensed laser media.

The case where $n_2 > n_1$ while the central zone is amplifying may be implemented in waveguide fibres. However, as a consequence of the small gain in fibres, such modes are not excited (there are no selected 'amplifying chords'). On the other hand, in diode lasers there is a high gain but, owing to the characteristic high value of n_1, it is difficult to construct a shell surrounding the active medium and having $n_2 > n_1$.

Further study and detailed calculation of the mode structure of macrodimensional liquid laser cavities with transparent rigid and flexible shells is of interest. This is also applicable to the construction of optical sensors, making it possible to detect external influences comparatively easily and with a high precision from changes in the positions and intensities of lasing modes by means of spectroscopic instruments.

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