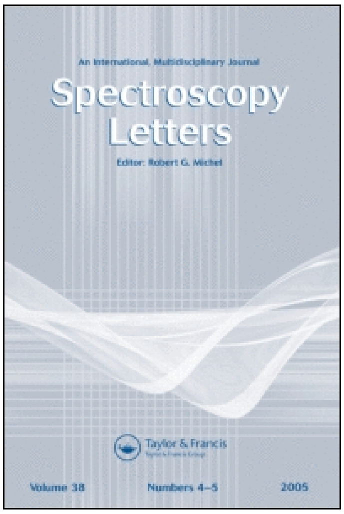

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WAVEGUIDE DYE SOLUTION LASERS

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1. Introduction

Since the development of first lasers the possibility of light amplification and oscillation in waveguide structures has been investigated. Waveguide lasers have been constructed using a variety of lasing materials, either solid, gaseous, or liquid.

One of the principal advantages of using a waveguide structure at the optical frequencies is the possibility of miniaturizing the laser components and to set up very compact lasers. The field of optical communications has often requested the development of fiber optic amplifiers and oscillators to be inserted into the fiber transmission systems(1-2). More recently, with the fast development of the integrated optics technology, the importance of waveguide lasers is further increased(3).

Although the feasibility of laser action in guiding structures has been widely demonstrated for solid, gaseous, and liquid materials, only solid-state lasers have been extensively investigated for the above mentioned applications. More precisely, dye lasers have been considered impractical for integrated optics because of limited lifetime due to photobleaching, and because of emission in the visible region, where the transmission losses of optical fibers are higher than for near IR sources. The availability of compact, rugged inexpensive narrow-band dye laser oscillators with integrated resonator structure will certainly be of great interest for a variety of applications beyond the field of integrated optics when improvements of dye characteristics will be achieved(4). Therefore, in this paper we will only briefly review the results obtained with liquid fiber and liquid thin-film dye lasers. We will discuss in more details the guiding effects which occur in liquid solution dye lasers as a consequence of the inhomogeneous distribution of the (complex) refractive index, produ ced by the pumping pulse, in view of their importance for a full understanding of dye laser performance, and especially for a more efficient design of dye laser geometries for high energy and high average power operation. These thermal-lens type waveguide dye lasers have been investigated by the authors, and in several aspects turned out to have superior operating characteristics than conventional dye lasers.

2. Active Waveguide Structures

Waveguide structures encountered in laser applications can be roughly divided into three groups (Fig.1):

a) fiber dielectric waveguides (with homogeneous core)

b) hollow dielectric waveguides (with homogeneous inner medium)

c) graded-index waveguides.

The study of light propagation in passive waveguides has recently received an increasing attention, and has been extended to active guiding structures as a consequence of the important developments in the field of fiber optics and integrated optics[5-8]. We now briefly illustrate the basic concepts and the principal results reported for cases a) and b); the practical applications of the principles presented in c) will be discussed in the next section.

a) Fiber dielectric waveguide

Circular cylindrical waveguide

An optical fibre is constituted by a glass-core cladded with another glass of lower refractive index. This fiber acts as a dielectric waveguide. The guiding mechanism is total internal reflection. Liquid-core optical fibres can be obtained by drawing a hollow glass fibre and by filling it with a suitable liquid of slightly higher refractive index. If the cross section of the core is sufficiently small (a few microns) only a small number of possible field distributions, or modes, can be allowed to propagate down the fibre. For example, the energy of the lowest order mode is confined into the waveguide structure if:

$$n_1 = n_2 \left[1 - \frac{3}{4} \left(\frac{\lambda}{a} \right)^2 \right] \qquad (1)$$

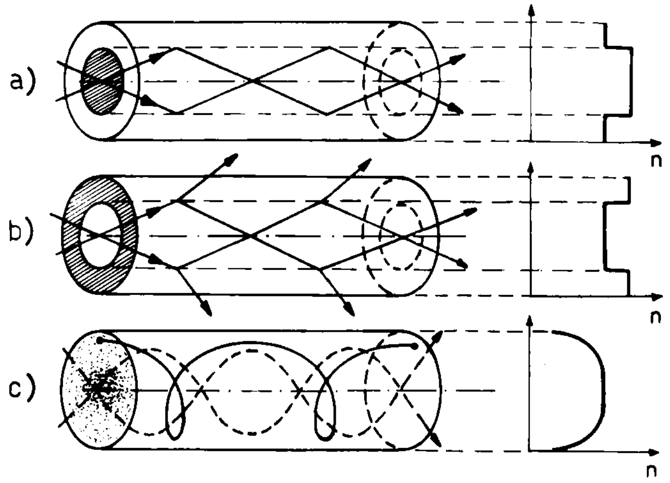

Fig.1 : Schematic representation of the three prin^c^ipal types of optical waveguides. a) fiber waveguide with homogeneous core and cladding : $n_{core} > n_{cladding}$. b) hollow dielectric waveguide with homogeneous inner medium and cladding : $n_{core} < n_{cladding}$. c) graded index fiber. Typical ray trajectories are shown. In case c) meridional rays and helical ray of constant radius are indicated.

where n_1 and n_2 denote the index of refraction of the glass capillary and of the solution, respectively; λ the free space wavelength; and a the radius of the liquid core. For a BK7 glass capillary (n_1 = 1.516) and benzyl alcohol (n_2 = 1.538) single mode operation at λ = 6.300 Å requires : a = 4 μm. Mode patterns corresponding to the well known fiber laser[5] have been observed by pumping with a frequency doubled Nd:YAG giant pulse laser a 10 μm bore capillary filled with a solution of Rhodamine B in benzyl alcohol[9].

Amplification of an input signal in a liquid-core fibre has been obtained by winding the fiber around a linear flashtube. With a solution of Rhodamine B in benzyl alcohol an amplification factor of 10 dB/m has been measured at the He-Ne wavelength[10].

<u>Slab-type (Thin-film) waveguide</u>

Light waves can be guided in thin dielectric films, which are the two-dimensional analog of optical fibers.

Thin film organic dye lasers have been built by filling tiny capillary gaps between polished glass surfaces with a dye solution. The distance between the glass surface can be determined by spacers made by evaporated dielectric or plastic film material. Laser action has been reported with Rhodamine B in benzyl alcohol with both traveling wave and evanescent wave pumping by a frequency doubled YAG:Nd laser[11]. The evanescent field pumping of a dye film has also been demonstrated [12].

In the above experiments the feedback necessary for the build-up of laser oscillation is provided by reflection at the exit windows or at the dye solution/air interface at the end of the waveguide. Waveguide structures with a distributed feedback have been obtained by means of a spatially periodic modulation of the liquid film's thickness. The forward mode is then coupled to its corresponding backward mode by intramode diffraction. Narrow band emission can be obtained by this kind of feedback[3]. Zory[13] has operated successfully a distributed feedback laser (DFL) consisting of a film of liquid organic dye solution in glycerol ($n = 1.4654$) held between a blazed reflection grating and an optical flat (quartz, $n = 1.4566$), and side pumped by a N_2 laser. Feedback is provided by the fourth spatial harmonic of the grating, and the output is coupled out from the grating under the first-order angle in the form of a leaky-wave. Distributed feedback can also be produced by periodic variations of refractive index in an adjacent dielectric medium of lower refractive index. The feedback mechanism then involves the evanescent waves of the waveguide. Hill and Watanabe[14] successfully operated a DFL by sandwiching a liquid dye solution film (15 ÷ 100 μm thick) between a holographic phase grating and an optical flat. Rhodamine 6G in a mixture of benzyl alcohol and ethanol has been used and side pumped by a N_2 laser.

b) Hollow dielectric waveguide

Conventional gas lasers with resonant cavities, formed by end mirrors, operate with free-space modes that are determined by the shape and spacing of the mirrors, but are not influenced by the presence of the laser tube. As the tube diameter is decreased the tube begins to interfere with the e.m. fields formed by the laser mirrors and operation of the laser is altered in a fundamental way. We are then in the presence of an interesting type of optical waveguide, in which the refractive index of the inside tube is lower than that of the surrounding material. Marcatili and Schmeltzer[15] demonstrated that, if the free space wavelength is much smaller

than the internal radius of the tube, the energy propagates essentially within the tube, bouncing at grazing angles against the wall. Consequently, there is little energy loss due to refraction. The hollow waveguide can be expected to propagate the low order mode with very small attenuation due to the grazing incidence.
The attenuation constant of the EH_{nm} modes for straight dielectric guides is given by

$$\alpha_{nm} = \left(\frac{u_{nm}}{2\pi}\right)^2 \cdot \frac{\lambda^2}{a^3} \cdot \frac{1}{2} \frac{(\nu^2+1)}{\sqrt{\nu^2-1}} \tag{2}$$

where u_{nm} denotes the m-th zero of the J_{n-1} Bessel function, and ν the relative refractive index of the external medium. For $\lambda = 1\ \mu m$, $\nu = 1.5$, $a = 1$ mm, the EH_{11} mode exhibits the lowest power attenuation of only 1.85 dB/Km.

The hollow waveguide laser still depends on external mirrors to provide the required feedback, but the transverse shape of the field distribution is no longer determined by the laser mirrors, but by the modes of the hollow tube. Distributed feedback has also been proposed for these lasers[16].

Hollow (cylindrical and planar) waveguides have been successfully used mainly with gas laser, where they permit to exploit the inverse dependence of the gain on tube diameter. Experiments with dye solution have proved that thermal effects due to laser or flashlamp pumping produce a strong inhomogeneous distribution of the complex refractive index which greatly affects the propagation characteristics of the waveguide. They will be then discussed in next section.

Zeidler[11] investigated the mode behavior for a slab waveguide as the refractive index of the dye solution was changed by temperature variation from the condition $n_{glass} < n_{dye}$ to $n_{dye} < n_{glass}$. In the case $n_{dye} < n_{glass}$ the waves are guided by the high reflection at the interfaces caused by the grazing incidence near the critical angle and give rise to modes which radiate both at the exit window and through the walls. The light penetrating the wall is radiated in cones and the brilliant hyperbolae of the Lummer-Gehrcke modes[19] have been observed. Traveling wave and evanescent pumping has been used.

c) Graded-Index Waveguide

A very interesting type of optical waveguide is represented by a dieletric medium with an inhomogeneous distribution of the index of refraction in the plane perpendicular to the guide axis. In this case the medium surrounding the inhomogeneous fiber plays a secondary role in determining the allowed modes of propagation and the light can be guided along the fiber by the distributed inhomogeneity. Light propagation in passive fibers with graded-refractive index has been extensively studied in view of their great interest for optical communication systems[1]. Fibers with parabolic profile of the index of refraction have received the greatest attentions. They present quite good characteristics for image display and pulse transmission[18]. One of the main features is that such a fiber can transmit 'stationary' Gaussian-Hermite beams[7], i.e. without changing the beam width with distance. The focusing effect of the fiber compensates the divergence of the free-space Gaussian beam. Wave propagation in a medium with radially symmetric refractive index profile can be discussed on the basis of the scalar wave equation:

$$\nabla_\perp^2 \psi(r) + \left[k^2 n^2(r) - \beta^2\right] \psi(r) = 0 \tag{3}$$

where $\psi(r)$ denotes the mode radial function, $n(r)$ the refractive index, β the propagation constant in the fiber axial direction. If n(r) is real (lossless passive fiber) the mode has a caustic at the 'turning point', r_t, defined by

$$k^2 n^2(r_t) = \beta^2 \tag{4}$$

It is known from the theory of WKB approximation that the field has an oscillatory character in the range $r < r_t$, and an exponentially decaying behaviour in the range $r > r_t$. The condition:

$$r_t = a \qquad \text{(a = fiber radius)} \tag{5}$$

has been chosen as a 'cut-off' condition to define the modes which begin to strongly interact with the fiber boundary, and become very lossy[19].

If the fiber has gain, n(r) is now complex and the field has an oscillatory and real exponentially behaviour in both regions $r \gtrless r_t$.

Since the variations of the index of refraction and of the gain over a wavelength are typically negligible, the more intuitive ray picture can be used to describe the light amplification. The ray equation is given by[20]:

$$\frac{d}{ds}\left(n\,\frac{d\underline{R}}{ds}\right) = \mathrm{grad}\; n \tag{6}$$

where s denotes the distance along the ray measured from some point on the ray, $\underline{R}$ a position vector of a typical point on the ray. The intensity $\mathcal{I}$ of the light ray as it travels in the amplifying medium is given by:

$$\mathcal{I}(s) = \frac{n(s)}{n(s_0)}\,\mathcal{I}(s_0)\,\exp\left[-\int_{s_0}^{s}\frac{1}{n}\,\nabla^2 \mathcal{S}\,ds + \int_{s_0}^{s} g(s)\,ds\right] \tag{7}$$

where s_0 denotes a reference point on the trajectory, S represents the eikonal: $\mathcal{S} = \int_{s_0}^{s} n(s)\,ds$, and g(s) the small-signal gain. The first term in the exponential in eq. (7) takes into account the spreading of the ray tube in the inhomogeneous media. For radially symmetric medium the solution of eq. (6) can be easily reduced to quadratures, and the ray path and the gain factor $G = \mathcal{I}/\mathcal{I}_0$ can be evaluated for any given index and gain profile [21].

The study of light transmission and amplification in media with complex inhomogeneous refractive index is receiving an increasing number of contributions[22÷26]. Solid-state self-focusing active fibers are now available[27]. The importance of these studies relies on the fact that lasing materials are only approximately homogeneous, and that in many cases the effects of refractive index and/or gain inhomogeneities must be taken into account to understand the laser performance. More precisely, in flash lamp pumped media the distribution of the absorbed energy cannot be uniform if a good utilization of the pumping radiation is required. The lateral focusing effect of cylindrical rods and the spectral composition of the pumping radiation and of the lasing material give rise to complicated distributions of the absorbed energy, and hence of the gain. The heat released in non-radiative transition, proportional to the absorbed energy, produces a non-uniform distribution of temperature, and hence of refractive index. Lens effects have been observed since long time in solid-state lasers. Even gas lasers with longitudinal electrical discharge pumping have nearly parabolic gain and refractive index profiles, with the maximum on the tube axis, which produces gain and dispersion focusing effects[23]. Thermally induced refractive index gradients have been considered as a serious obstacle to the development of liquid lasers and thermal lens waveguides have been proposed for these lasers since 1964[26]. Prism-like and lens-like effects were observed in dye lasers and considered the cause of deleterious effects, as early termination, beam distortion, etc. Only recently the authors have pointed out the importance of the waveguide structure produced by the pumping radiation for the construction of more efficient laser geometries.

3. Self-Guiding Dye Laser Amplifiers and Oscillators.

The total intensity I(r) absorbed at a distance r from the axis in the dye cell is given by:

$$I(r) = \int I_0(\lambda)\, \alpha(\lambda)\, f(r,\lambda,a)\, d\lambda \tag{8}$$

where:

- $I_0(\lambda)$ denotes the light intensity at the wall of the cell, whose spectral distribution depends on the particular pumping source used;

- $\alpha(\lambda)$ denotes the absorption coefficient of the dye solution;

- $f(r,\lambda,a)$ describes the profile of the illumination at the wavelength λ, at a distance r from the cell axis, for a cell with total thickness (or diameter) 2a;

- the integration is performed over the entire absorption spectrum of the dye.

The function I(r) has been determined by several authors in the case of solid-state and liquid laser materials[28,29]

A fraction of the absorbed intensity is converted into heat in the host medium. The heat transfer is associated with non-radiative transitions among rotational-vibrational levels of the singlet states, intersystems crossing, and triplet state relaxations of the dyes[30]. Part of this heat is evolved before initiation of the laser pulse itself, and the remaining heat during the pulse. Therefore a laser pulse is forced to propagate from the very beginning in a non-uniform, medium whose refractive index varies with time. Owing to the different decay constants of the re laxation processes, the relative importance of one process with respect to the others depends on the rise time and time duration of the pulse.

The complex refractive-index $\tilde{n}(r)$ of the active solution can then be written as:

$$\tilde{n}(r) = \left[n_0 - \Delta n(r)\right] - i\, g(r)/k \qquad (9)$$

where n_0 denotes the refractive index of the solution in the absence of pumping, $\Delta n(r)$ the refractive index variation produced by the exciting pulse, and $g(r)$ the gain-coefficient per unit length. Both $\Delta n(r)$ and $g(r)$ are proportional to $I(r)$. The waveguide characteristics of the pumped solution will substantially depend on the refractive index and gain distributions which in turn depend on the cell and pumping geometry and on the spectral distribution of the pumping radiation.

Their knowledge is important for the construction of efficient dye laser amplifiers and oscillators.

Let us now briefly discuss the lasing characteristics of these selfguiding structures.

a) Planar Cell

In planar cell illuminated from both sides by a parallel, uniform beam the absorbed intensity is expressed by

$$I(r) \propto \int I_0(\lambda)\, e^{-\alpha(\lambda)a} \cosh\left[\alpha(\lambda) r\right] d\lambda \qquad (10)$$

$I(r)$ is maximum at walls of the cell and minimum at the center. Correspondingly, the index of refraction is maximum on the cell axis, and decreases towards the walls, as shown in Fig. 1c. In such a medium light propagates in the same plane of the incident radiation along curved zig-zag paths (meridional rays in Fig. 1c). If the gain is sufficiently high, the threshold for superfluorescence (amplification of spontaneous emission) can be reached. Laser action occurs for ray paths of maximum gain, i.e. for those rays which impinge at nearly grazing incidence onto the cell surface, where the inversion is maximum. From each side of the cell the radiation is emitted into two beams, whose angular separation depends on the waveguide parameters. Since organic dye solutions have a high quantum yield still at high dye concentrations ($10^{-3} \div 10^{-2}$ M), high efficiency amplification can be achieved. For a given value of the cell thickness, an optimum concentration exists resulting from a compromize of uniform inversion over the cross section and maximum utilization of pump intensity. The maximum of the absorbed intensity at the axis, I(o), occurs for:

$$(\alpha_{peak})_{opt.} \cdot a = \begin{cases} 1 \text{ monochromatic pumping} \\ 2,\ 20 \text{ for} \end{cases}$$

a 30.000 °K black-body pumping with and without UV filtering, respectively (31). Since the absorption coefficients of the commonly used dye solutions are very large (α_p = 121 cm^{-1} for a 10^{-3}M solution of R6G in ethanol), very thin cells must be used at high concentration. The transverse dimension normal to the pumping beam and the length of the cell can then be suitably chosen to get the desired active volume. Two arrays of linear flashtubes closely coupled to the cell can be used for pumping. The planar cells have been successfully operated as laser amplifiers and oscillators (32,33). Maximum energy output occurs typically for a 0.8 10^{-3}M solution of R6G in ethanol and for a cell thickness of 0.4 mm. If one end of the waveguide is terminated by a flat mirror, and the other one by a Brewster angle window, an input signal can be injected in correspondence of one of the superfluorescence lobes and, after a double pass in the amplifier, extracted from the other lobe. By flashlamp pumping, gain factors up to 85.000 have been obtained at low signal input, and saturated gains of 5.000 at 100 mJ output (overall efficiency 0.1 %) has been achieved with the complete quenching of the broadband superfluorescence (33). By replacing the Brewster angle window by a flat window placed normal to the cell axis a compact and efficient oscillator can be constructed accordingly to Fig. 2 by using the narrow rectangular termination of the cell as the entrance slit of a spectrograph with the film plate replaced by the feedback mirror. Preliminary results obtained with the Gillieson mounting of Fig. 2 indicate that these lasers are well promising for the generation of narrow-band, energetic pulses. They can find practical applications in the fields of photochemistry, pollution detection, etc. Moreover, simultaneous operation at two or more wavelengths can be achieved by placing other mirrors at the focal curve of the spectrograph. With the Rowland mounting tuning should be achieved by simply sliding the feedback mirrors along the Rowland circle. To further reduce the output spectrum Fabry-Perot etalons can be inserted in the oscillator.

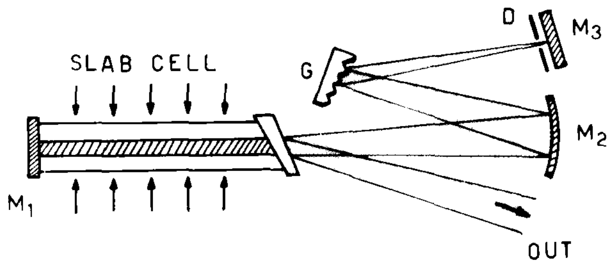

Fig.2 : Schematic drawing of a tunable slab oscillator.

b) Cylindrical Cell.

In an uniformly and isotropically illuminated cylindrical cell the profile of the absorbed intensity is complicated by the lateral lensing effect produced by the dye cell[28]. For $\alpha a \ll 1$ the profile is nearly flat and superfluorescence emission occurs uniformly over the entire cross section of the cell[34]. As the product αa is increased, the gain distribution favours the modes with off-axis maxima. The intensity gradually decreases at the center of the near field pattern, until it assumes a well defined ring distribution at large value of αa. For $\alpha a \gg 1$ the highest gain region is limited within a very thin sheet near the wall of the cell. Superfluorescence can occur only along helical rays (Fig. 1c) which travel near the cell wall ('whispering gallery' modes). Meridional rays (Fig. 1c) travel mostly in scarcely pumped regions and cannot break into oscillation without the feedback by an optical resonator.

Superfluorescent emission has been investigated in capillary cells with 0.1 mm bore filled with R6G in ethanol[34]. In spite of the poor coupling between flashlamp and capillary, relatively intense emission has been observed. This demonstrates the low propagation losses of these selfguiding structures, which permit the use of the more efficient low index solvents (as methanol and ethanol) for the dye solution.

However, the peculiar angular distribution of the radiation emerging from the cell operating on the whispering modes makes it difficult to be handled, and limits the utilization of cylindrical cells as high efficiency amplifiers and oscillators.

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