Planar waveguide nanolaser configured by dye-doped hybrid nanofilm on substrate

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Received 5 November 2017, revised 5 December 2017
Accepted for publication 18 December 2017
Published 23 February 2018

Abstract
Dye-doped hybrid silicate/titanium nanofilms on the glass substrate structures of asymmetrical waveguides were studied by way of laser systems. The threshold, spatial and spectral features of the laser oscillation of genuine and hollow waveguides were determined. The pattern of stimulated radiation included two concurrent processes: single-mode waveguide lasing and lateral small divergence emission. Comparison of the open angle of the lateral beams and grazing angles of the waveguide lasing mode provides an insight into the effect of leaky mode emission followed by Lummer–Gehrcke interference.

Keywords: waveguide lasing, hollow and genuine waveguide, dye-doped hybrid film, leaky and guided modes, Lummer–Gehrcke interference, wavelength cutoff

(Some figures may appear in colour only in the online journal)

1. Introduction

The inclusion of organic molecules in a solid inorganic matrix at a high concentration, but with minimal interaction to maintain luminescent efficiency, provides considerable reason for the promotion of research and development in this field. The high concentration of luminescent centers is a useful feature in laser materials, because actually, the dopant determines the specific amplification or nonlinear response (dye stream lasers, dyed liquid crystal laser, harmonic generation and others). Completing this task is possible with a structured inorganic matrix. The presence of matrix cells (pores) permits the organic molecules to be placed separately. When the matrix pore concentration is comparable to the guest molecules, the interaction between them is attenuated.

Regular mesopores (i.e. filled with intermediate layers) in silicate glasses were first obtained using sol–gel technology by the authors in [1–3]. The first porous matrices were created by the acid-alkaline etching of soda-borate-silicate glasses [4]. The dye deposition in the matrix pores was performed by sorption from the solution. In this approach the ‘individual cells’ of the separate molecules are absent. A similar dye-doped solid matrix with increased thermo-optical stability was gained in this way and used successfully. Frequency tunable lasers with similar dye-doped mesoporous matrices were realized in a few earlier works [5].

Another type of room temperature sol–gel chemical reaction technology for the production of hybrid laser materials with random pores was studied in [6]. The output product here is called ‘ormosil’ and is obtained in the usual 3D form. Significant research has been done with lasing on dye-doped ormosil matrixes at typically low concentrations, precluding the negative consequences of dye dimerization [7].

In our work, structured dye-doped material was produced with modified sol–gel technology. The technology relies on the ability of amphiphilic molecules to self-assemble in modular structures called micelles. During the dehydration stage, the self-organization of dyed micelles in spatially ordered ensembles with various symmetries takes place. Concurrently with the mentioned process, the relevant structuring of the silicon/titanium matrix occurs. With this technology, dye-doped hybrid material in the form of extended films with nanoscale thickness is produced. Herewith, the given films demonstrate higher luminescent quantum yield at dye concentrations that
are higher than the usual impurity solution and solids [8, 9]. Changing the concentration relation between the organic–inorganic parts of the material recovers control of the index of refraction (IR), the thickness of the films and some other properties. The properties of dye-doped hybrid nanofilms explain their application in nanoscale laser media.

The miniaturization of lasers to nanoscale dimensions has been the subject of heightened research in recent times. Small mode volumes and strongly enhanced fields are necessary as a prerequisite to nonlinear interaction followed by the appropriate nonlinear phenomena. One-dimensional semiconductor nanolasers have been investigated in [10–12]. Hybrid plasmonic zinc oxide nanowire lasers near the surface plasmon frequency were considered in [13]. A 3D fully dielectric laser was proposed and theoretically justified in [14]. For the cavity of this designed nanolaser, it is proposed to use a gain layer with a diameter of 100–200 nm covered with a dielectric silicon sphere.

In our work, for the dye-doped nanoscale films on the substrate, a 1D laser system is proposed as the active medium, based on a one-mode asymmetrical waveguide by way of the appropriate cavity. Consequently, the given work targets the creation and first examination of a 1D nanolaser with a dye-doped hybrid nanofilm.

2. Characterization of dye-doped hybrid nanofilms

The mesoporous hybrid SiO2/TiO2 films were fabricated by sol–gel technology using the surfactant nonionic three-bloc copolymer Pluronic P123 (from Sigma-Aldrich). A sol-solution was prepared using chemically pure tetra ethoxy silicate (Si(OC2H5)4) or tetra ethoxy titanium (Ti(OC 2H5)4), ethanol with distilled water and 35% HCl. After hydrolysis of the mixture and sol origination, the previously made R6G gel and the surfactant Pluronic P123 were mixed to the first critical concentration. This step resulted in the self-assembling of micelles in a random spatial distribution. The ratio of the gel and intermixture concentrations in gram-molecule units was varied from 1:32 to 32:1. The basic regular composition of the material was agreed to the following component ratio:

TEOSi(Ti):C2H5OH:HCl:H2O = 1:20:0.5:8. The final intermixture was suitable for film formation after aging at continuous stirring for (40 ÷ 60) min. Film deposition on the glass substrate was carried out at grivation and drive velocities of 2040 spins min−1 and (0.1–20) cm min−1, respectively. The deposited silicate (titanium) films were dried at the regular temperature/dampness levels until their thicknesses reached values of 100–300 nm. Due to drying, the concentration of dyed micelles in the film increased to the second critical value. As a result, the spatial ordering of dyed micelles takes place simultaneously with the SiO2 (TiO2) grid structure formation. Grid walls of (3, 5) nm are built around the spatially ordered micelle structure. Micelle ordering is manifested in the plane and across the nanofilm. With the conservation of organic micelles in the gained film, a similar structure is referred to as mesoporous. The heating of similar samples to 500 °C causes the evaporation of the organic micelles and, respectively, the formation of true porous films.

Formation of the ordered structure was confirmed by the small-angle x-ray diffraction technique. The results confirm the formation of the ordered structure and its invariance at the supplementation of non-ampiphilic laser dyes. Infrared spectroscopy specifies the almost full elimination of water from the films after drying. The film thickness was in the range of (100, 300) nm according to atomic force microscopy measurement. The next plain estimation provides the parameters of the shaped spatial structures. The diffraction maximum of the x-ray radiation (λCuKα = 0.15405 nm) satisfies the Bragg condition mλ = 2λ sin(θm) at a discrete diffraction angle. This permits the determination of the spatial periods of the grating Λ1 = 10204 nm and Λ2 = 10155 nm for m = 1 and 2 respectively.

Herewith the density of micelle packing is about ≈1018 cm−3 for the cubic symmetry if the spatial period and orderliness of the structured SiO2 films corresponds to the spherical micelles. The micelle packing density is about one order higher for hexagonal packing. At the measured optical density ε(TeO5Si(Ti)):C2H5OH:HCl:H2O = εT = 1.5 (dye R6G, molar extinction εmax = 1.2 × 105, film thickness T = 200 nm) in the maximum absorption band, the dye concentration ε = 3.6 × 1020 cm−3. Comparison of the micelle and dye concentrations shows about a ten-fold excess of dye above the pores. However, the shapes of the dye absorption bands do not show common signs of dimerization.

The IR measurements for dye-doped SiO2 and TiO2 were conducted using the modified Brewster method [15]. Both optical structures satisfy the following configuration of layer-interleaving ‘air-film–glass substrate’, which is relevant to the asymmetrical planar waveguides [16]. The IR Brewster measurements of the nanofilms were taken at two wavelengths: 532 nm and 660 nm. The smaller wavelength overlaps the absorption maximum of R6G in the SiO2 and the larger one fits the fluorescence band. The real n and imaginary x parts of the complex IR become commensurable when the concentration of R6G in the nanofilm reaches a relatively high level. Then, measurements of the complex IR by the Brewster method become ambiguous compared to the common case: n ≫ x = λ/2π. The ambiguity can be removed by independent measurement of the relevant absorption coefficient (α). In the case when strong absorption attenuates the second reflected beam at 532 nm from the substrate, a light reflection curve is observed without the accompaniment of interference (figure 1(a)). The measurement outside the absorption band at 660 nm shows the genuine Brewster angle for the nanofilm with Pluronics 123 + dye (figure 1(b)). Two-beam interference accompanies the relevant curve due to the presence of two beams in the reflection, but it does not affect the accuracy.

The Brewster angular minimum in the absorption band depends on the complex IR according to the relation n = 1/\cos(ϕmin) \sqrt{(1 + x^2)} and = λ/2π/15. The waveguiding of the planar structure is satisfied when the condition nB > n2,5 becomes valid. Here n2,5 means the IR for the substrates and cover, and nB is the IR of the relevant film. So, fabrication of the mesoporous film with the application of TiO2 (IR = 2.62) is imperative. The IR of the hybrid film reduces to a value of ≈(1.58–1.68), because the organic part of the film remains in kind but can partly fluctuate.
There is definite interest in the dependence of the absorption/luminescence spectrum at various dye concentrations in the films. Until the dye concentration stays below $10^{-4}$ M, the study shows no deviations from the typical monomolecular spectrum of R6G in water taking place. The evolution of the fluorescence spectrum with the concentration change of the hybrid films SiO$_2$ and TiO$_2$ is presented in figure 2 [8, 9]. The fluorescence efficiency R6G in the nanofilms is conserved until dimerization starts.

3. Experimental laser procedures, results and discussion

The considered dye-doped nanofilm on the glass substrate shapes the planar waveguide. This waveguide configuration has an asymmetrical structure due to the difference in IR between the substrate and the cover. The main features of the planar dielectric waveguides are presented in [16] and the other works cited there. To describe the waveguide behavior in our case, the ray-optical approach turns out to be enough. According to the ‘zigzag’ path model, the wave propagation has two groups of orthogonal modes: TE and TH types. If the light incident angle inside the core film satisfies $\theta_I < Q_{\text{TIR}}$ (TIR is the total internal reflection), light reflection with $R < 1$ takes place without a phase shift but with a jump $\pm \phi$. When the incident angle exceeds $Q_{\text{TIR}}$, reflection reaches 100% and includes the variable phase shift. The phase shift grows when $\theta_I$ exceeds $Q_{\text{TIR}}$ and reaches $\pi/2$ at $\theta_I = \pi/2$. These features are described by the classic Fresnel theory. The particular features of light propagation in an asymmetric waveguide depend on the relation between the grazing angle and the two different TIR angles. At $\theta_I < (Q_{\text{TIR1}}, Q_{\text{TIR2}})$ light leaks from the waveguide and creates leaky mode propagation. The same is also valid when $\theta_I \leq Q_{\text{TIR2}}$, but light only emanates from the substrate. So a necessary condition of the existence of guided
modes is the next inequality: $\theta_1 > Q_{\text{TIR1}}$. $Q_{\text{TIR2}}$: Sufficient conditions for the existence of guided (or leaky) modes appear when the sum of all phase shifts along the ‘zigzag’ trajectory is equal to the multiple number $2p$.

The self-consistency condition (or transverse resonance) takes the following form:

$$2k_n T \cos \Theta_1 - 2\vartheta_s - 2\vartheta_c = 2m\pi.$$  \hfill (1)

Only a discrete set of TIR angles satisfies the self-consistent survival of guided and leaky modes. Here $k$ is a wave number, and $2\vartheta_s$ and $2\vartheta_c$ are phase shifts for waves in the substrate and cover interfaces respectively. They are written with a minus sign because of the time delay at the reflection; $m = 0, 1, 2 \ldots$ are the numbers of the modes. The phase shift in the first member of (1) is inversely proportional to the grazing angle $\theta_1$.

The arbitrary small thickness $T$ is permitted by this equation (including $T = 0$) for the symmetrical waveguide on the main guided mode ($m = 0$). This implies that there is no cutoff frequency for the fundamental mode. However, multimode operation ($m > 0$) according to (1) is only allowed for waveguide thicknesses $T > 0$. For the asymmetrical waveguide, equation (1) shows that the waveguide thickness has to be $T >$, even for one-mode operation. The decrease of the core thickness is followed by a decrease of the grazing angle $\theta_1$. As $\theta_1$ approximates the larger angle $Q_{\text{TIR1}}$, the respected phase shift reaches zero and stops changing. From this region of grazing angles, guided mode operation will be changed by the leaky mode. This means that the cutoff thickness on the main mode of the asymmetrical waveguide can be found from relation (1) at condition $2\vartheta_c = 0$:

$$T_c = \vartheta_c / k_n \cos \theta_1.$$ \hfill (2)

The phase shift of the TE-polarized wave at an incident angle above the TIR2 angle is determined according to Fresnel theory:

$$\vartheta_c = \sqrt{(n_s^2 \sin^2 \vartheta_c - n_t^2)} / n_t \cos \theta_1.$$ \hfill (3)

Now the determination of the cutoff-thickness of the asymmetrical waveguide $T_c$ becomes plain because the grazing angle is equal to the bigger angle $Q_{\text{TIR2}}$. Taking into account that the phase shift $J_s \approx 0$, and substituting (2) and (3) in (1):

$$T_c = \lambda \arctan \sqrt{n_f^2 (\sin^2 \theta_{\text{TIR2}} - \sin^2 \theta_{\text{TIR1}})} / 2\pi n_t^2 \cos^2 \theta_{\text{TIR2}}.$$ \hfill (4)

When the difference between the angular values TIR2 and TIR1 approximates zero, the cutoff-thickness of the waveguide core $T_c$ also approaches zero. This means the asymmetrical waveguide allegedly transforms into a symmetrical one. The ratio of $T_c / \lambda$ is called the wavelength or frequency cut-off and it shows the smallest ‘part’ of the wavelength where emission can be transferred to the given asymmetrical waveguide.

Our genuine asymmetrical waveguide on the dye-doped hybrid TiO2 film with an IR $n_t \approx 1.68$ on the glass substrate ($n_s = 1.51$) has angles of $Q_{\text{TIR2}} = 64^\circ$ and $Q_{\text{TIR1}} = 36.53^\circ$ on the interfaces of the film-substrate and the film-air respectively. The evaluation of $T_c$ by the expression (3) at $n_t = 1.68$ provides the cut-off thickness $T_c = 131$ nm. The reduction of the film IR up to 1.58 gives an increase of TIR2 up to the value $Q_{\text{TIR2}} = 72.88^\circ$. The cutoff thickness drops to $T_c = 80.9$ nm in this connection.

A hollow asymmetrical waveguide is obtained with the dye-doped hybrid SiO2 film. This film has a wavelength-dependent IR = 1.49 (figure 1) that is less than the IR of the glass substrate $n_t = 1.51$. The given waveguide is characterized by higher radiation loss and all the guided modes are leaky. In spite of this, through an amplification lasing process, the leaky guided modes can be excited [17, 18].

Equation (1) can apply to the description of the discrete angular distribution of leaky modes of the hollow waveguide as well. In such a case, the light transfer process has two parts in leaky modes: the first part propagates inside the waveguide core and the second part of the power is emitted outside through the lateral wall. The phase jump of $p$ with the light reflection from the denser substrate media has to be taken into account. The grazing angles of the leaky modes can be found using the presented equation (1):

$$\Theta_m = \arccos((2m + 1) \lambda / 4n_t T)$$ \hfill (5)

where $m = 0, \pm 1, \pm 2 \ldots$, the grazing angles of the fundamental mode at $m = 0$ Q1 are 59.8° and 70.4° for the two core film thicknesses $T$ of 200 nm and 300 nm, respectively, $n_t = 1.491$ and $\lambda = 600$ nm. The highest modes $m > 0$ are prevented because the transverse resonance condition is not obtainable for the given film thickness.

The spatial and spectral features of the stimulated emission of the planar asymmetrical waveguide with the different IR of the core nanofilm are considered below. There are two types of asymmetric waveguide with the core dye-doped R6G at approximately equal concentrations. As noted above, the SiO2 hybrid core film forms a hollow waveguide and the TiO2 hybrid core film forms a genuine waveguide. The transverse laser pumping ($\lambda = 532$ nm, $\tau \approx 30$ ns) at the beam cross-section $18 \times 0.1$ mm² is injected into the waveguide through its lateral wall. The pump intensity can change stepwise by attenuators within the confines (0.05–0.35) MW mm⁻². The stimulated emission of high diffraction divergence is excited inside the dye-doped waveguide core film. Two symmetrically located lateral beams of much lesser divergence also emerged under pumping. The named beams were intercepted by the optical system in turn and directed alternately to the spectrograph for analysis. The divergence of the main waveguide radiation appeared naturally in the plane orthogonal to the waveguide plane. Herewith, the divergence in the plane of the waveguide remained lower and was determined by the shape/size of the excited film region. Next, measurements were taken for the comparison of the stimulated emission (threshold, angular and spectral characteristics) of the genuine and hollow waveguides under variable pumping.

It is well known that owing to the TIR, the luminescence of transparent films becomes much stronger around all the side ends compared to the wide plane surfaces. With the enhanced excitation of the similar planar ‘waveguide’, the luminescent power will increase much faster than for systems without TIR when the light can propagate freely in $4\pi$-steradian. It is more likely for the conditions and thresholds of laser
oscillation to be optimal in a genuine waveguide than in a hollow one. Herewith, laser oscillation requires the conditions for the survival of the minimal number of available modes of spontaneous emission. These conditions are known as phase and amplitude lasing. Equation (1) displays the phase conditions that highlight the lowest loss of the guided modes in the waveguide. The radiation losses of the waveguide modes must compensate for the amplification. Spectrally depend-ent feedback may also appear through counter-propagated ASE-beams (amplified spontaneous emission or superluminescence). No wavelength selective elements are necessary if coupling between the counter running waves owing to interference and gain grating recording takes place.

All observed types of stimulated emission in the asymmetrical hollow waveguides are presented schematically in figure 3. The excited region of the dye-doped waveguide core film emits the specular symmetrical spatial pattern of the stimulated radiation. The central lobe of the pattern is characterized by the relevant diffraction-limited divergence in the plane orthogonal to the waveguide plane $\lambda T \approx 2 \text{rad}$ at $T = 300 \text{nm}$ and $\lambda \approx 580 \text{nm}$. The angular divergence of the central lobe in the orthogonal waveguide plane is less than $1^\circ$. This divergence only depends on the pumping spot geometry. The total patterns of the spatial beam distributions for hollow and the genuine waveguides differ only slightly, despite the different properties of the leaky and guided modes.

The main lasing beam demonstrates sharp spectral narrowing from the initial broad luminescence band of $R6G \approx 550 \text{nm}$ to the lasing linewidth $\approx 4 \text{nm}$ when pumping reaches the threshold levels (figures 4(a) and (b)). It is well known that the lasing process differs from ASE by sharp spectral narrowing. The same dramatic narrowing with pumping is manifested in stimulated emission for the genuine and hollow waveguides. However, the lasing thresholds (at the points of spectral narrowing) for the genuine and hollow waveguides differ by orders of $\approx 1.5$. The reason for this is connected to the low radiation losses in the genuine waveguide compared to the same losses in the hollow one. Figure 4(a) also illustrates the behavior of the spectral lines when pumping is over the threshold value. One can see that the spectral bandwidth remains approximately the same.
The stimulated emission of lateral beams resolutely differs from waveguide lasing mode \((m = 0)\) with respect to the spectral widths and angular divergences (figure 3). These lateral beams in both types of waveguides have a smaller angular divergence \((\approx 1°)\) in the waveguide plane and a bigger divergence in the orthogonal plane as compared to the main mode emission. The low angular divergence of the lateral beams immediately suggests another output emission spot other than the face of the waveguide core. As an output spot of lateral beams, one should consider the side wall of the waveguide, although it is difficult to explain the origin of the lateral beams outside the genuine waveguides in this way.

The lateral beams for both waveguides appear with opening angles of about 52° in the same plane where all beams overlap each other. Conclusions about the beam overlapping were made after comparison of the spectral distributions of the single and overlapped beams. The emission spectra outside the beams are manifested in the known luminescence spectrum of R6G (see figure 2). The emission of lateral beams at an angle of about ±26° shows the small smooth narrowing of the initial broad R6G spectrum—like the evolution of the ASE spectra versus pumping. Spatial overlapping of the waveguide lasing emission and lateral beam emission (see figure 3) develops in their mutual spectrum: they have a different spectral bandwidth.

The small angular divergence of the lateral beams results from them emanating through the lateral waveguide walls without the diffraction angle broadening. They should be considered as leaky waveguide modes. The next summation of successive coherent beams along the excited waveguide region transforms into a Lummer–Gehrke multibeam interference pattern [17, 18]. The pattern is unwrapped in the plane parallel to the waveguide plane with a divergence of about 20° (figure 3). Fewer questions have arisen regarding comprehension of the lateral beams in the case of the hollow waveguide. However, for a genuine waveguide, it is necessary to suggest partial TIR violation at least! To be consistent with the phase equation (1), a small deflection from the angle of the TIR may be supposed for the grazing angle of the lasing mode. Some support in favor of this thought shows the distinction of the spectral widths of the main lasing line and the ASE in the lateral beams.

Furthermore, consider the correlation between the open angle for the lateral beams \(W \approx 52° (\pm 26°)\) to the axis of the waveguide core assigned to the pumping beam spot, figure 3) and the grazing angle for the waveguide lasing mode. The measured open angle \(W\) leads to the discovery of the grazing mode angle using Snell’s law. At the same time, the grazing mode angle can be calculated from equation (1), and the known values \(T\) and IR of the dye-doped film. The equality of these angular values should confirm the joint origin of the lasing mode and lateral beam emission.

The following results have been gained from the above-mentioned notes. Taking into consideration the initial incident angles ±26° and their succeeding refraction in transits across the substrate up to the film core, angular values of ±73° were found. The grazing angle for the main mode of the hollow waveguide at a film thickness of \(T = 300\) nm, \(n_t = 1.491\) and \(\lambda = 600\) nm reaches values of ±70.4°. It makes no sense to undertake a more accurate comparison, due to the deficiency of accurate thickness data using the TiO2 nanofilm.

The explanation of the lateral beam origin as a leaky mode at the same open angle ±26° in the case of the genuine waveguide was more complicated in comparison with the hollow one. A small violation of the TIR condition at accidental overlapping angles of \(Q\) and the grazing mode are proposed above as a solution. The determination of the incident angle inside the film core from the lateral beam directory again leads to the value ±73°. Now it is necessary to settle the TIR angle and the grazing mode angle of the genuine waveguide based on a TiO2 nanofilm. Angular consistency was achieved for the film of the same thickness \(T = 300\) nm and IR = 1.58 (instead of the expected IR = 1.68) that resulted in the grazing angle ±70.4° for the main mode \(m = 0\) and \(Q\) = 72.88°. Again, it makes no sense to perform a more accurate comparison of the experimental and calculated magnitudes now, due to the deficiency of the more accurate IR value using TiO2-nanofilm.

4. Summary

Lasing based on a dye-doped nanofilm of \((200 \div 300)\) nm thickness in a planar asymmetrical waveguide structure is realized and studied for the first time. The stimulated emission reveals a complicated structure, which depends on either genuine or leaky waveguide operation. The evaluation of the wavelength cut-off of the used asymmetric waveguides provided values of 130(80) nm, which are lower than the virtual thickness \(T = 300\) nm. The studied core nanofilms belong to a class of hybrid organic–inorganic material with ordering due to the organic micelles inside the inorganic matrix. Because of this, dye luminescence efficiency is preserved at higher concentrations than for the usual system. The hybrid SiO2 film forms a hollow waveguide on the glass substrate and operates in leaky mode. The hybrid TiO2 film works as a genuine waveguide on good quality guided modes. Both types of waveguide operate in the main single mode. The measurements confirm that the lasing threshold for the genuine waveguide is about two orders lower than for the hollow one. The spatial pattern of radiation also includes lateral beams with relatively low angular divergence. This radiation is referred to as leaky modes followed by Lummer–Gehrke interference. Its origin is evident for the hollow waveguide, but is not so obvious for the genuine one. The estimations show that a leaky mode can appear when genuine waveguide parameters have a mode grazing angle that is nearly equal to the bigger angle of the TIR. More study should be done for a better understanding of this observation.

Attending to the technological deficiencies as well as carrying out further development of the dyed structured ormosil films and their application to nano-optics are on the agenda. One of the promising conclusions of the given project is the possibility of applying planar waveguide structures to the laser testing of new nonlinear and active materials.
Acknowledgments

This research has been supported by the National Academy of Sciences of Ukraine under project #0106U002454 and fulfilled by the Institute of Physics NAS UA together with the Institute of Physical Chemistry NAS UA and the Department of Physics of T. Shevchenko National University.

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