Lasing of Rhodamine 6G in the Thin Hybrid Organic-Nonorganic Mesostructural Film

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Introduction

Major advances of integrated optoelectronics now are linked at making the source light of the submicron dimensions. One of most appropriative for this purpose are waveguide (WG) lasers which confine lasing emission in active host medium resulting in increasing optical gain and decreasing of the lasing threshold [1]. These lasers may be easily aggregated on the same chip with other optoelectronic devices (transmission lines, modulators, filters, etc.) [2].

The organic/inorganic hybrid films prepared by the sol-gel approach have become a new field of research in materials for WG laser [2]. This method allows forms the film of some hundreds nanometers thickness as a result of self-organization of the hybrid micelles and polycondensation of the inorganic precursor and allows managing independently two most important parameters of planar WG laser: gain value and refractive index of the matrix material. In the formed hybrid- film, the inorganic component determines of the refractive index and dye incorporated in micelles specifies value and contour of the gain. In this way film will acts as waveguide laser if its refractive index is higher than of substrate.

Fabrication of ultra-thin WG laser based on hybrid films requires solving two principal problems. The first one is ensuring of high concentration of dye molecules without concentration quenching of luminescence to provide sufficient absorbance of the pumping radiation in thin film and gain. The second to provide a higher refractive index of the film relative to the substrate to provide waveguide conditions at film/substrate interface.

Experimental

Two sets of sol-gel films based on SiO_2 and TiO_2 matrices were prepared and tested. The fabricated films represent mesoscopic ordered inorganic matrix of dioxide silica or titanium with oriented channels of a diameter 12 nm filled with micelles surfactant (P123) contained dye R6G molecules (fig.1). Thickness of the films measured with AFM method was about 200 – 300 nm.

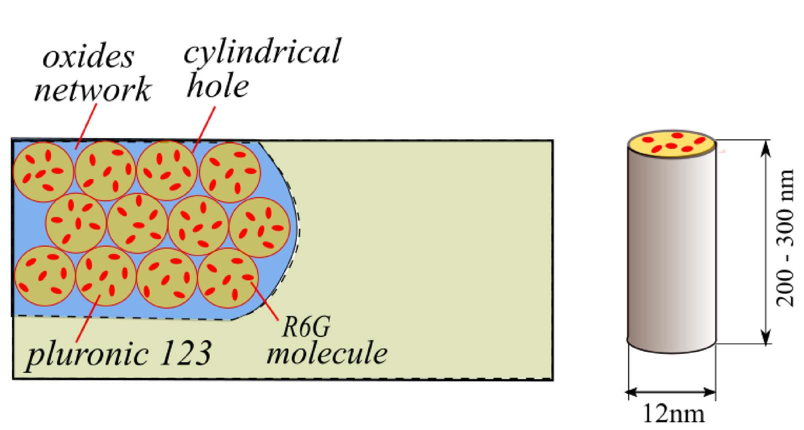

Fig1. Design of hybrid organic-noborganic film dyed with rhodamine 6g.

Refractive index films were measured with a modified Brewster method [10]. Its value varied in the range 1.499 – 1.68 in dependence on the type of inorganic component and technological conditions.

The fabricated films on the glass substrate together with air as cover material form asymmetric wave guides having the order of interlacing of layers with refractive indices of 1 / n_f / n_s, where n_f =1.499 (1.565) – refractive index of the SiO_2 (TiO_2) films and n_s= 1.515 – refractive index of the substrate. The film is doped with R6G of concentration 0.1 – 0.5 mmol/l . is. Quenching of the luminescence was overcame due to confining of Rh6G in P123 micelles that prevent planar packing dye molecules in non-luminescent H-type dimers [3].

Lasing of the films was excited by the second-harmonic radiation of a Q-switched YAG-Nd^{3+} (532 nm, 12 ns) focused on the film in the strip of 3x0.5mm^2 size. The spectrum of radiation emerging from the end of the excited luminous track was registered in directions along the track and under set of the angles relative to the track. The output end of the luminous track (or substrate) was projected by a fiber-lens system onto the entrance slit of the spectrograph. It allows possible to flexibly change the display of the emission area, orientation of the image relative to the slit and the direction of radiation propagation. Radiation spectra of the films were single-shot registered by diffractive spectrograph and CCD camera.

We have detected that radiation emerged from sample end contains two parts which substantially differ by directivity and spectrum. The first one spreads over a wide range of angles about 50° - 60 ° (fig. 2). Its spectrum sharply narrows by an order of magnitude up to 4- 5 nm (fig. 3b) with increasing pump intensity, which is inherent in lasing of conventional dye lasers. The threshold intensity of the appearance of this narrowing in the TiO2 film decreases by about an order of magnitude in comparison with the SiO2 film. Its radiation emerges from the film end and could be associated with WG lasing. However, the fabricated films do not provide the conditions for the appearance of the true type of this radiation, since the refractive index of SiO_2 is less than that required for total internal refraction (TIR), and the TiO_2 film thickness (250 nm) is less than the critical value.

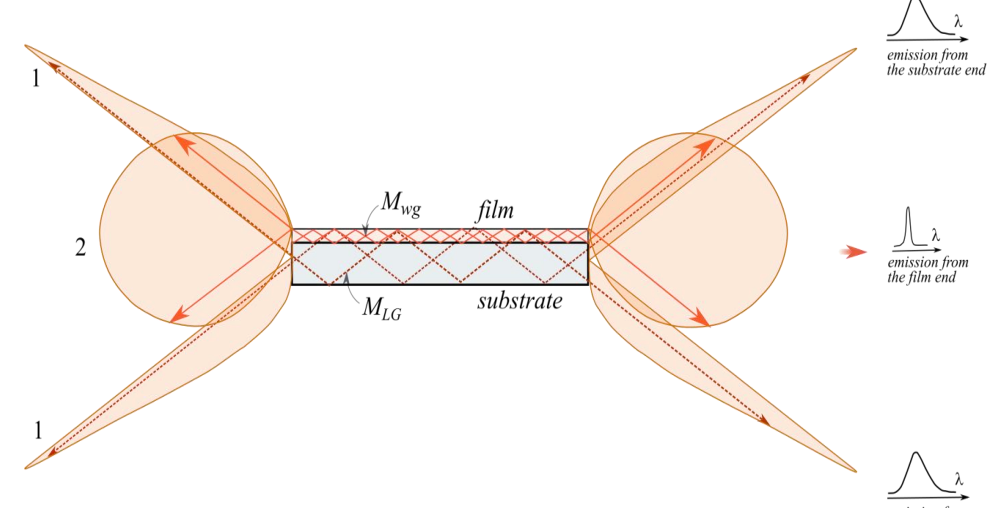

Fig2. Diagram of lasing formation in the film.

The second part of the emerged radiation spreads over a much narrower range of angles and propagates under 25° - 30° approximately relatively to the track (fig.3) and depends on refractive index of the film. Its spectrum much wider (fig. 3a) and narrows under substitution SiO_2 film with TiO_2 (25 nm to 7 nm). In SiO_2 film this lateral radiation forms well defined hyperbola-like arcs on the screen perpendicular to the track.

In both samples the narrowing of the spectrum arises sharply for radiation emerging from the film end and rather smoothly for radiation from substrate. The spectrum of the radiation from the substrate end contains narrow lasing component (2) and wide luminescence one (3), ratio of their intensity increase with growth pump.

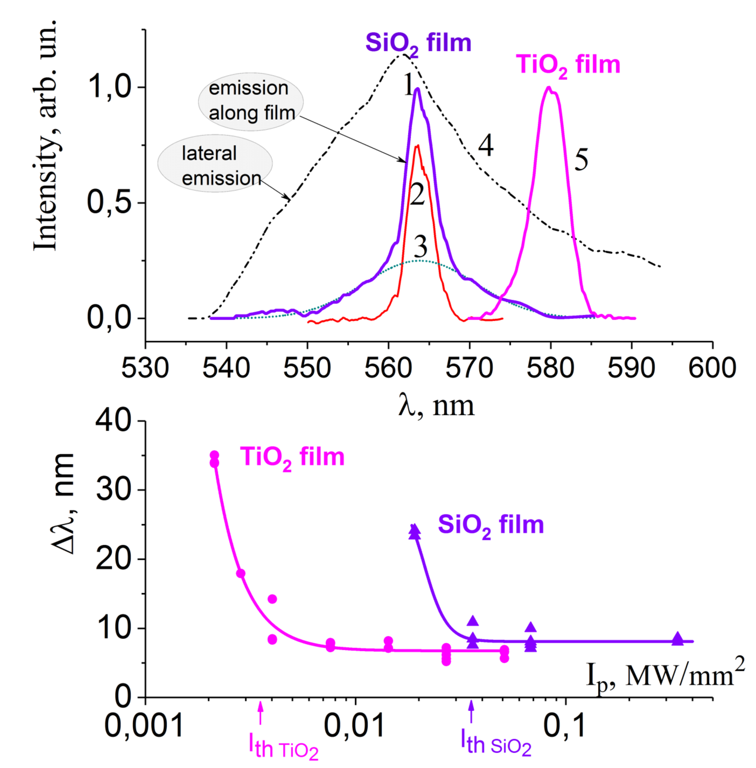

Fig3. a) Spectra of radiation emerging from end of the film along track (1, 5) and in direction of lateral beam (4); 2 and 3 – lasing and luminescent component of lateral beam.
b)Dependence of spectrum width of radiation from film end on pump intensity.

Relative intensity of these two emission depends on reflectivity of free surface of the substrate: intensity of the radiation from substrate reduces when immersion applying (fig. 4). It indicates that this radiation forms with assistance of reflection from this surface. This ratio depends on pump intensity differently: in the samples without immersion intensity in the substrate increase more rapidly. And contrary In the sample with immersion the radiation from the film growth more rapidly but tends to saturation..

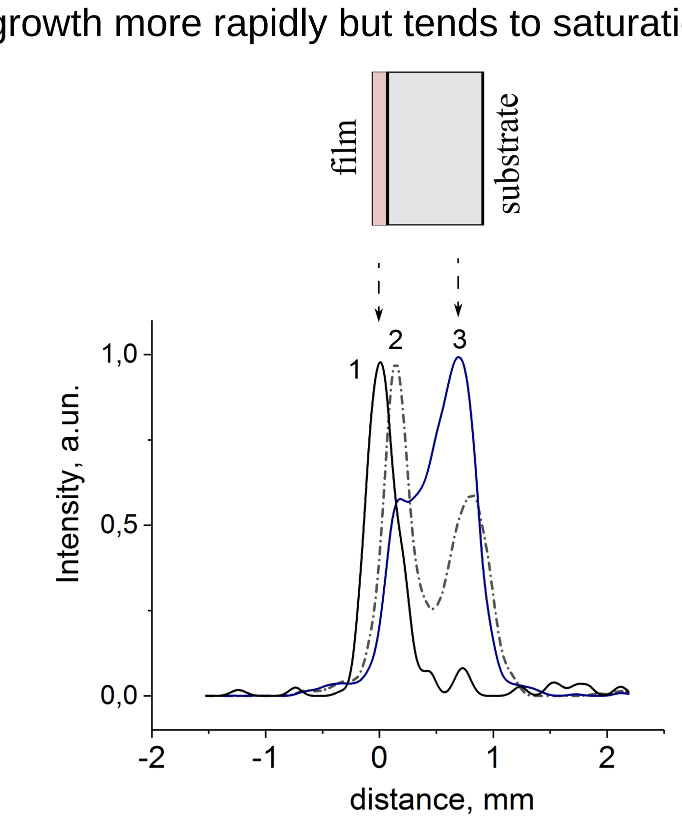

Fig4. a) Distribution of radiation over the ends of the TiO_2 film and substrate with immersion of free substrate surface (1, 2) and without it (3) under pump intensity 0.1 (1) and 2.7 (2, 3) MW/mm^2.

It is interesting to note that under pump intensity round the threshold the radiation from the substrate practically disappears and exists only in the film. This results may be important for application in optoelectronics.

Presented results confirms assumption made in the work [4] about possibility of the lasing on Lummer-Gehrke modes. They forms by many parallel plane waves satisfying to condition of self-consistency but incidence on the interface film/substrate at the angle which does not satisfy TIR condition. Part of these waves which stays in the film forms the quasi WG modes M_{wg}- waveguide mode which energy loses owing to coming out to the radiative mode of the substrate (fig. 2). This quasi mode is able to survival only due to light amplifying in the WG core (film) that compensates its radiative loss and therefore may arise only under high intensity of pumping.

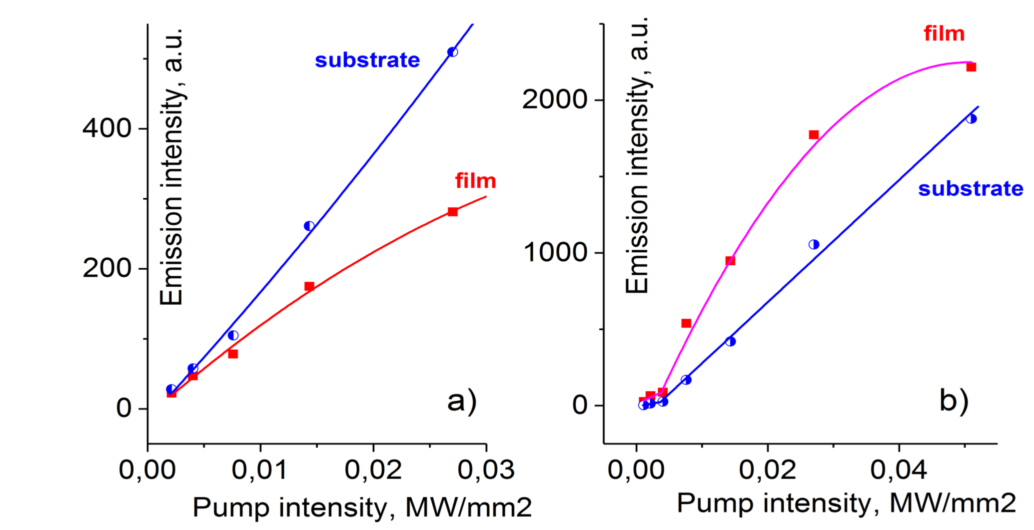

Fig5. Dependence of radiation intensity emerged from the film and substrate in the samples without immersion (a) and under immersion (b).

Other part of these wave coming out from the film to the substrate and undergoes TIR on the second surface (adjacent with air). In such way in common space of the film and substrate form the special modes which are similar to radiation in Lummer-Gehrke plate and which may be named as Lummer-Gehrke modes M_{LG}.

Radiation of both modes linked each other but developed differently.

Conclusions

In the hybrid organic-nonorganic films doped with rhodamine 6G with violate condition for true waveguide lasing emission of two types may appear:

- - quasi wavequide mode which energy loses into radiative mode of substrate
- mode Lummer-Gehrke – radiative mode of substrate formed as interference of plain wave coming out the film.
- These modes differs by directivity, direction of propagation an spectrum.

Bibliography

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