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Wave-guided sol-gel glass lasers

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Abstract: Planar waveguides are obtained by deposition of sol-gel precursors with a variety of laser dyes on glass or quartz plates at low temperature. The laser radiation is emitted in the film or the support, depending on the difference between the refractive indices of the two media. Tunable laser emission was observed in the visible spectral range in films doped by perylimide, oxazine, xantene, cyanine and merocyanine dyes. Efficient laser amplifiers can be constructed.

Numerous modern applications in recording, communication printing, display etc. demand compact waveguiding lasers that can be tuned in the visible spectral range. Such a system was proposed by us about a decade ago suggesting theoretically an introduction of laser dyes into glass films [1,2,3]. Since then tunable lasers based on bulk glasses prepared by the sol-gel method have been reported by our, and by other groups [4-9].

The problem of introducing laser dyes which will be dispersed into monomers in films of high optical quality at room temperature is not trivial. The necessary conditions for efficient wave guided lasers is that the laser molecules do not form dimers or higher aggregates which dissipate the excitation energy by non-radiative energy transfer. This phenomenon is also responsible for photodecomposition of the dyes. In the present paper we present the experimental methods that allowed us to incorporate a large number of laser dyes into thin waveguiding films. We have observed laser action when pumped by the second harmonic of Nd:YAG laser and a low threshold of less than 50 μ J.

Waveguiding thin films made of commercial coating solutions containing silicon and esters of titanate acid were prepared by Herrmann [10] and by Hewak and Lit [11]. Their procedure consists of dip coating of the substrate, drying at 100-150°C and baking at 500-1000°C. While this procedure allows preparation of waveguides with a high optical damage threshold and a low attenuation, the high temperature involved in the processes (close to the T_g of the glass) prevents doping with organic dyes which are responsible for the laser properties. The high power density, caused by confinement of the radiation within a very thin film, can also be applied for pumping of laser materials or nonlinear dyes incorporated into the film. While this concept was already proposed by us [1,2], it is only in this work that we have found practical ways for preparing photostable wave guiding films that allow incorporation of organic dyes.

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High refractive index glass films were prepared as follows at room temperature. The glass synthesis was performed by hydrolysis and subsequent copolymerization of titanium tetraethoxide $\text{Ti}(\text{OEt})_4$ or titanium tetraisopropoxide $\text{Ti}(\text{OPr}^i)_4$ with the ORMOSIL namely glycidylpropyltrimethoxysilan (GLYMO).

Glass films with refractive index up to 1.66 were thus obtained.

In order to prepare films of lower refractive index of 1.48 and density of 1.68 gr/cm^3 we have used a procedure based on azeotropic distillation with benzene or toluene solution including the laser dye, tetra-ethoxysilane $\text{Si}(\text{OC}_2\text{H}_5)_4$ (TEOS), and triethoxyvinylsilane $\text{CH}_2=\text{CHSi}(\text{OC}_2\text{H}_5)_3$ (TEVS), and low molecular weight polymethylmethacrylate (PMMA). The block diagram of this procedure is given in Fig. 1.

Fig.1.
**Procedure for Preparation
of The Composite Glass Films**

Table I. presents examples of laser dyes introduced into the films (prepared in our laboratory) the lasing range, and the threshold of laser operation.

Fig. 2 presents laser emission as observed by narrowing of the fluorescence at threshold energy, of the Lumogene LFR 300 provided by BASF. The spectroscopy of this dye in various solvents has been discussed in detail in reference 12.

Table 1. Spectral characteristics of the dyes used for glass films doping.

Dye	Abs. max, nm	Tuning range nm	Spontaneous emission spec. width nm	Laser Spec. width nm	Threshold $\mu\text{J/pulse}$
Lumogen LFR 300	578	605-630	70	9	30
Lumogen LFO 240	525	568-583	30	3	50
R 6 G	546	560-610	44	11	50
DCM	472, 496	595-650	79	19	100
Rhodamine 610	560	585-635	45	10	40

Figure 2. Fluorescence and laser emission of LFR 300 in a composite glass.

Glass films from alumina, prepared by peptization of aluminum hydroxide doped by Rhodamine 6G, Rhodamine B and oxazine 4 showed laser action with calculated conversion efficiency of 21%. The intensity decreased linearly with the shot number of exciting N_2 laser [13].

Fluorescence intensity, photo degradation and kinetics of degradation of excited states of R6G in sol gel films was recently reported in ref. 14. Photo degradation of the dye increased with the concentration as a result of interaction of the monomers with higher aggregates.

The coupling of leaky modes from lower refractive index of the doped film than the support has been calculated theoretically in ref. 15. A laser configuration based on active dielectric thin film, acting as an optical amplifier, has been analyzed in detail theoretically [15].

It has been predicted that the resonant beam width is dominated by the active medium width of the film, not by the resonator-mirror curvature. The calculations were based on the leaky-mode concept [16].

An investigation of oblique plane wave scattering in active dielectric films reveals the existence of anomalously large resonance that occur at discrete plane wave angles of incidence. This fact allows application of the active films with lower refractive index than the support as efficient amplifier.

A large progress has been made in dyed doped glass lasers since the last two years [17], and many of the systems described here will be soon ready for application in novel optics.

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