Distributed Feedback Sol-gel Channel Waveguide Lasers

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Abstract

Distributed feedback (DFB) waveguide lasers allows the generation of picosecond short pulses with sub-nanometer narrow linewidths, and are thus compact tunable laser sources for applications in integrated optics. Zirconia is a very useful optical material because of its wide optical transparency, high mechanical strength and resistance to chemical reaction. The rectangular channel waveguide provides better lateral confinement and is very compatible with planar processing technologies. In this thesis, the sol-gel fabrication of dye-doped zirconia and zirconia-organically modified silicate (ORMOSIL) channel waveguides is presented and the demonstration of tunable DFB laser action in the channels is reported.

Rhodamine 6G (R6G) and LDS 925 were doped into zirconia and zirconia-ORMOSIL materials as prepared by the low temperature sol-gel technique. Embedded channel waveguides were fabricated using wet or dry etching of glass or fused silica substrates followed by deposition of the sol-gel solutions in the channels. DFB laser action tunable from the visible to the near infrared (NIR) was generated in the channel waveguides by crossing two nanosecond, frequency-doubled Nd:YAG laser beams of wavelength of 532 nm. Narrow linewidth lasing (<0.5 nm) was achieved for R6G-doped zirconia channel waveguides with widths of 5, 6.5 and 10 μm, and for LDS 925-doped zirconia-ORMOSIL channel waveguides with widths of 15.6, 25 and 30 μm. The wavelength tuning ranges were from 570 nm to 608 nm and from 787 nm...
to 933 nm, respectively. The output laser mode was identified as the fundamental $E_{11}^z$ mode. The cross-sectional profiles of the output beams resembled Gaussian shapes. The dispersion behavior of the laser output was checked by comparing experiments with the predictions of Marcatili’s theory. Additionally, NIR wide-band tuning and high-order DFB operations were realized in LDS dye-doped planar waveguides.
摘要

分佈反饋波導激光器是一種結構緊湊的可調諧激光光源。這種激光器採用動態增益調製技術產生皮米量級窄縫寬和皮秒量級短脈衝的激光輸出。二氧化錳是一種具有良好光學特性的無機玻璃，它具有寬的光學透明度，高的機械強度和抗化學反應的能力。二維通道光波導對光波的傳播比平面光波導有更好的約束能力，因而它被廣泛應用於集成光學的領域。在本論文當中，應用低溫溶膠凝膠技術，成功製備了摻雜激光染料的二氧化錳和摻有機改性硅的二氧化錳通道光波導，並研究了這些通道光導的分佈反饋激光輸出特性。

通過乾法刻蝕和濕法刻蝕兩種光刻方法，分別在玻璃和熔融石英基片上加工了各種尺寸的微通道。將摻雜了染料諾丹明 6G 和 LDS 925 的溶膠填充在微通道裏，分別獲得了能夠在可見和近紅外波段產生增益的主動通道光波導。通過將兩束橫向聚焦的倍頻 Nd:YAG 激光器輸出的 532 納米的納秒激光脈衝在通道波導中重複干涉形成周期性的增益調製光柵，成功地觀測到小于 0.5 納米的窄縫寬分佈反饋激光輸出，並通過改變調製光柵的周期，實現了激光波長的調諧。利用上述方法，成功地製作了寬度分別為 5, 6.5 和 10 微米的摻雜諾丹明 6G 的二氧化錳分佈反饋波導激光器和寬度分別為 15.6, 25 和 30 微米的摻雜 LDS 925 和有機改性硅的二氧化錳分佈反饋波導激光器，獲得了從可見到近紅外光譜範圍的可調諧激光輸出。激光的調
諧範圍分別是 570 納米到 608 納米和 787 納米到 933 納米。輸出激光的模式來源于通道波導的基模 $E_2$，輸出光斑的橫截面符合激光光束的高斯分佈。從實驗和理論上分析了通道光波導分佈反饋激光器的色散特性，這種波導激光器的色散能夠很好地符合 Marcatili 的二維光學波導理論。這說明了溶膠凝膠通道光波導有着良好的光學性能。另外，我們從摻雜 LDS 染料的平面光波導中實現了近紅外波段分佈反饋激光的寬帶調諧輸出和滿足高階 Bragg 反射條件的分佈反饋激光的可調諧輸出。
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Chapter I

Introduction

Dye lasers, with their broad tuning range and high efficiency, are useful tools in science and technology. After the first dye laser was invented by P.P. Sorokin et al. in 1966 [1], liquid solution of dyes in organic solvents became the standard media for dye lasers. Nevertheless, this approach was never fully satisfactory because of the serious inconveniences evidenced by the liquid dye lasers, mainly related to the need to handle large volumes of messy and sometimes toxic liquids. To overcome these inconvenience, an important effort has been dedicated from the mid-1980s to the development of compact and reliable solid-state dye lasers [2]. A solid state dye laser avoids the problems of toxicity and flammability, presents a low cost gain medium, and is easy to operate and maintain. The basic requirements imposed on a host matrix to be used as a host for the lasing dye molecules are transparency at both pump and lasing wavelengths, high optical quality with a low level of scattering, high damage threshold to laser radiation, and good thermal and photochemical stabilities. A simple technology for doping the matrix material with different classes of organic dyes is also desirable. Up to now, a large number of solid-state dye lasers based on different host matrices and laser dyes have been demonstrated, such as dye-doped polymeric materials lasers [3, 4], dye-doped sol-gel silica lasers [5-7] and dye-doped organically modified silicates (ORMOSIL) lasers [8, 9].
As laser materials in common use, inorganic glasses and transparent polymers have their respective advantages. Polymers have better chemical compatibility with organic dyes and superior optical homogeneity. In contrast, inorganic glasses exhibit in general better thermal properties (higher thermal conductivity and lower thermal expansion and thermal coefficient of refractive index, $\partial n/\partial T$) [10] and a higher damage threshold than polymers [11]. The low-temperature sol-gel process allows the incorporation of organic dyes into inorganic glasses (viz., silica, titania and zirconia, etc.), resulting in relative photostable materials with good laser efficiency [12]. Zirconia ($\text{ZrO}_2$) is a very useful inorganic glass because of the wide optical transparency and the high mechanical strength and resistance to corrosion. Due to its ability to form high refractive index stable glasses, zirconia films on glass or fused silica substrates have a wide range of applications in integrated optics. The sol-gel method has been successfully developed for the preparation of zirconia and zirconia-ORMOSIL thin films by Y. Sorek et al [13]. Zevin et al demonstrated zirconia waveguide amplifiers doped with a number of laser dyes [14]. The optical properties of infrared dye-doped zirconia thin films were reported by Casalboni et al [15].

In 1971, a novel tunable dye laser, distributed feedback (DFB) laser, was demonstrated by C. V. Shank et al [16]. In place of a conventional resonator cavity, optical feedback for a DFB laser was provided via backward Bragg scattering from periodic perturbations of the refractive index and/or the gain of the laser medium [17]. This oscillation mechanism results in a compact laser
structure and realizes narrow linewidth lasing emission easily. Since then, many research interests have been concentrated on DFB lasers. J. E. Bjorkholm et al observed high-order DFB oscillations [18] and obtained DFB laser actions in thin-film optical waveguides [19]; Z. Bor carried out many works on picosecond pulse generation in self Q-switched DFB dye lasers [20] and obtained the continuous press tuning [21]. At the same time, theories on DFB lasers were developed. The first DFB theory based on a coupled-wave model was presented by H. Kogelnik et al. in 1972 [17]. Then, a related coupled-mode theory was developed by A. Yariv in 1973 [22]. More detailed works were carried out by S. Wang [23] and H.A. Haus [24] etc.

DFB solid-state lasers developed rapidly during the last decade. Many new materials as host matrices and functional dopants were synthesized. Most of the work was done using organic polymers, such as dye-doped polymers and light emitting polymers. Y. Oki et al. published several papers on DFB dye-doped polymer lasers on topics as ultrashort pulse generation [25] and wide-wavelength-range tuning [26]; G. Kranzelbinder et al. reported optically written DFB MEH-PPV waveguide lasers [27]; G.A. Turnbull et al. observed DFB lasing in MEH-PPV films [28]; D. Wright et al. fabricated the holographically multiplexed organic DFB lasers [29]. J.M. Nunzi’s group and A. Penzkofer’s group studied DFB laser actions in dye-doped polymers and photopolymers [30, 31]. In addition, J. F. Pinto et al. realized tunable DFB Ce³⁺-doped LiSrAlF₆ lasers [32]; M. Ozaki’s group reported electro-tunable DFB laser action in dye-doped nematic liquid crystal [33].
In 2000, Prof. Dennis Lo started to devote himself to studying the applications of sol-gel glass materials in DFB lasers. D. Lo's group conducted many works on sol-gel silica dye lasers [7, 34-38]. They realized DFB dye-doped silica lasers tunable in the visible [39] and in the ultraviolet [40] and demonstrated temperature tuning of DFB solid-state dye lasers. Recently, they fabricated many kinds of DFB lasers based on sol-gel glass planar waveguides: wavelength-tunable DFB titania-silica waveguide lasers [41], single- and multi-wavelength DFB zirconia and zirconia-ORMOSIL waveguide lasers [42, 43], DFB sol-gel glass symmetric waveguide lasers [44] and two-photon-pumped DFB zirconia waveguide lasers [45], etc. However, of the several types of waveguides in use, the planar waveguide lacks lateral confinement and the circular waveguide is not compatible with planar processing technology. In practical applications in integrated optics, the rectangular dielectric waveguide is the most commonly used structure. The fabrication of active rectangular waveguides devices using standard planar processing technology for integrated circuit is generally laborious and expensive. Deposition of the active layer by sol-gel method affords the opportunity of an inexpensive processing approach. Casalboni et al. reported recently light amplification in dye-doped sol-gel channel waveguides [46].

In this thesis, following our previous works [39-46], we report our recent results on DFB dye-doped zirconia and zirconia-ORMOSIL channel waveguide lasers tunable from the visible to the near infrared (NIR). Firstly,
the fabrication of dye-doped zirconia and zirconia-ORMOSIL channel waveguides is described in chapter II.

Secondly, DFB R6G-doped zirconia channel waveguide lasers tunable in the visible are presented in chapter V. Several important properties, such as wavelength tuning range, threshold pump energy, slope efficiency, output polarization, operation lifetime and the dispersion behavior of the laser output are given. The experimental setup and optical arrangement are illustrated in chapter III. Some of the results have been published in *Applied Physics Letters* [47].

Thirdly, NIR DFB LDS 925-doped channel waveguide lasers are reported in chapter VI. Additionally, narrow linewidth DFB laser action achieved for the first, second and third Bragg orders in LDS 925-doped zirconia-ORMOSIL planar waveguides is described. NIR wide-band tuning based on four types of LDS dye-doped zirconia-ORMOSIL thin films (viz., LDS 759, LDS 798, LDS 867 and LDS 925) is demonstrated. Parts of the works have been published in *Optics Express* [48].

The coupled-wave theory of DFB lasers is introduced in chapter III. The one- and two-dimensional optical waveguide theory is presented in chapter IV. Conclusions are given in chapter VII. Future works are also discussed there.
Chapter II

Sol-gel channel waveguides

2.1 General sol-gel process

Sol-gel processing has been known for a long time. The first silica gels were made as early as 1845 by M. Ebelmen at the "Manufacture de Céramiques de Sévres" in France [49]. After stagnating for about 120 years, the sol-gel technique revived in the last three decades. Many kinds of glass and ceramic materials in various forms, such as powder, fibers, coating films, monoliths and ordered pores have been fabricated by this processing technique. In particular, sol-gel technology arises as a method of fabrication of high quality optical oxide materials and is well adapted for the preparation of thin films. It presents many well-known advantages for thin films elaboration over the other physical or chemical deposition techniques such as high purity of precursors, high homogeneity of material, low processing temperatures (which allows the incorporation of organic additives such as laser dyes [50] or indicators into the waveguide), no need of vacuum, low cost and high flexibility and feasibility [51]. Sol-gel material is a kind of material that can be easily improved and can be fabricated into the desired shapes and configurations [52]. The ability to prepare uniform films of large area makes this method highly practical. The surface of a film prepared by sol-gel technology is generally very smooth, which is important to achieve optical waveguiding properties. Until now, sol-gel thin films with various functions
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have been fabricated such as optical, optoelectronic and photonic films, films with electric and magnetic functions, films with chemical functions and films with mechanical functions, etc [53].

The word "sol" implies a stable dispersion of colloidal solid particles within a liquid [54]. Colloids are solid particles with dimensions in the range of 10 to 1000Å, each containing $10^3$ to $10^9$ atoms. The solvent used to stably disperse the colloidal particles of a sol is often either pure water or a solution composed mostly of water. When the viscosity of a sol increases sufficiently, usually through the partial loss of its liquid phase and/or polymerization of the solid particles, it becomes a porous solid body that is termed a "gel". A gel is a porous three-dimensional interconnected solid network that expands in a stable fashion throughout a liquid medium and is only limited by the size of the container. The network forming depends mainly on the structure of the sol and can be divided into two alternative mechanisms. The first is a mechanism which is based on the growth of molecules, leading to macromolecules which then connect together to form an infinite network. This mechanism leads to the so-called polymerized gels and is very common in the acid-catalyzed hydrolysis and condensation of tetraalkyl silicates. The other type is based on the aggregation of colloidal particles from a so-called colloidal sol and requires a fairly stable sol as the intermediate [53]. The nature of gels depends on the coexistence between the solid network and the liquid medium. The liquid is present between the mesh of solid network that composes the gel; it does not flow out spontaneously and is in thermodynamic equilibrium with the solid
network. If the liquid is mostly composed of water, and if that aqueous phase is the one present in greatest proportion, then the corresponding gel is an aquagel (or hydrogel). An aquagel is a soft material that can be easily cut with a knife. If the liquid phase is largely composed of an alcohol then the gel is an alcogel. Finally, if most of the liquid is removed, then the brittle solid obtained is called dry gel. The whole process from a sol to a gel is called gelation.

It is representative to use silica as an example. In the most simplistic terms, the sol-gel process consists of three reactions:

\[ \text{RO} \xrightarrow{\text{Si}} \text{OR} + \text{H}_2\text{O} \rightarrow \text{RO} \xrightarrow{\text{Si}} \text{OH} + \text{ROH} \]  
\[ \text{RO} \xrightarrow{\text{Si}} \text{OR} + \text{HO} \xrightarrow{\text{Si}} \text{OR} \rightarrow \text{RO} \xrightarrow{\text{Si}} \text{O} \xrightarrow{\text{Si}} \text{OR} + \text{ROH} \]  
\[ \text{RO} \xrightarrow{\text{Si}} \text{OR} + \text{CH} \xrightarrow{\text{Si}} \text{OR} \rightarrow \text{RO} \xrightarrow{\text{Si}} \text{O} \xrightarrow{\text{Si}} \text{OR} + \text{H}_2\text{O} \]

In (2.1), a silicon alkoxide such as tetraethoxy silane (TEOS), usually dissolved in an alcoholic solution, is hydrolyzed. The alkoxide group (OR) is replaced by a hydroxyl group (OH), then forms a silanol group. This reaction is called as hydrolysis reaction. Subsequently, the OR group of one TEOS molecule reacts with the OH group of an adjacent molecule (reaction (2.2)) or the OH group reacts with each other to form an Si-O-Si bond (reaction (2.3)). We refer to (2.2) as the alcohol condensation reaction and refer to (2.3) as the water condensation reaction. Usually, condensation commence before hydrolysis finishes. If these processes repeat themselves, then more and more Si-O-Si bonds are formed. As polymerization continues, viscosity of the
solution increases until a solid gel is formed. One major advantage of this process is the formation of the Si-O-Si bond at room temperature in a liquid solution.

The silicon can be replaced by another element M such as Ti or Zr. Thus TiO₂ and ZrO₂ gels can be prepared. If two alkoxides are used such that M is Ti and N is Ba, then a BaTiO₃ gel is formed. Ideally, any metal alkoxides can be employed and any number of alkoxides can be mixed to form a homogeneous solution. Thus, theoretically, any oxide glass composition or any crystalline oxide can be prepared in the gel form. Additionally, there are many interdependent factors which can influence the structure and microstructure of the gel and hence the structure and properties of the final oxide product. These include the particular alkoxide used (e.g. methoxide vs ethoxide), its concentration in the solution, the catalyst used, the pH value, the temperature, concentration of water, the particular solvent and other additives [55].

The interconnected three-dimensional pore network structure of the solid gel is often weak and brittle after aging and drying. In order to solve this problem, organic groups which do not contribute to cross-linking are introduced into the oxide network. The presence of the organic modifier can increase the flexibility and mechanical strength of sol-gel materials enormously. Such materials are known as “ORMOSILs” (organically modified silicates), “ORMOCERs” (organically modified ceramics) or “CERAMERs” (ceramic polymers) [56, 57].
2.2 Dye-doped sol-gel zirconia and zirconia-ORMOSIL materials

Zirconia ($\text{ZrO}_2$) is a very useful optical material because of the wide optical transparency and the high mechanical strength and resistance to chemical reaction. Combined with its high refractive index, zirconia films on glass or fused silica substrates hold good promise for a wide range of applications in integrated optics. Zirconia thin films appear to be superior to titania films in view of the optical transmission of zirconia deep into the UV and the absence of catalytic photodegradation of organic dopants [13]. Hence, zirconia seems ideal as a host matrix for functional organic dopants.

A major obstacle in preparing $\text{ZrO}_2$ thin films from zirconium alkoxides is rapid hydrolysis and subsequent precipitation of colloidal zirconia upon water addition to the $\text{Zr(OR)}_4$-containing precursor solutions [13]. Ganguli and Kundu were the first to overcome the fast precipitations problem by dissolving zirconium propoxide in dried solvents such as 2-propanol, cyclohexane and benzene. Transparent $\text{ZrO}_2$ films were prepared successfully [58]. Alternatively the zirconia precursor can be stabilized by complexing agents such as acetic acid or acetylacetonate [59].

We largely followed the preparation procedures of Sorek et al. to fabricate dye-doped zirconia and zirconia-ORMOSIL materials [13]. Fig. 2.1 is the typical sol-gel flow chart of zirconia (A) and zirconia-ORMOSIL (B) synthesis. The starting solution consisted of zirconium $\eta$-propoxide and acetic acid. 2.46 g of zirconium $\eta$-propoxide (70% solution in 1-propanol) was mixed in a hermetically closed glass vial with 0.3 mL of glacial acetic acid (analytical grade). After the solutions were magnetically stirred for an hour, 0.6mL of 2-
propanol was added to adjust the viscosity that, in combination of the speed of
spin coating, determines the thickness of the films on glass or fused silica
substrates or the depth of the dye-doped zirconia layer in the channels. The
water needed for hydrolysis was mixed with acetic acid (1:3 by volume) and
introduced drop by drop into the solutions. The molar ratio of zirconium n-
propoxide to acetic acid was about 1:4 in the final solutions. Finally, laser dyes,
such as R6G and LDS dyes, were added until the desired concentration was
reached. For zirconia-ORMOSIL films, the organic modifier γ-
glycidoxypropyltrimethoxysilane (GLYMO) (see Fig. 2.2) was introduced in
the initial solutions without any purification. Molar percentage of GLYMO in
the films was calculated by $X_{\text{GLYMO}} = \left[\frac{n_{\text{GLYMO}}}{n_{\text{GLYMO}} + n_z} \right] \times 100$.

![Diagram](image)

**Fig. 2.1** Schematic block diagram of preparation procedures of dye-doped zirconia and zirconia-ORMOSIL waveguides.
During preparation a white colloidal precipitate appeared either during water dropping to the solution or before the addition of the propanol and water. The precipitate was pepticized by adding more water to the solution. After the disappearance of the precipitate the solution is extremely stable, and no turbidity is developed even at long period (up to two months) of storage in a closed container. These observations can be explained by the following mechanism [13]:

1. Upon adding acetic acid a ligand exchange reaction occurs to form zirconium propoxide acetate according to (2.4):

$$Zr(OPr^\eta)_4 + x\text{Ac-OH} \rightarrow Zr(OPr^\eta)_{4-x}(OAc)_x + x\text{Pr-OH}, \quad (2.4)$$

where Pr$^\eta$ stands for $n$-C$_3$H$_7$, Ac stands for CH$_3$CO.

2. At this stage when both zirconium acetate and propoxide are present in the solution a nonhydrolytic oxide formation is possible according to (2.5):

$$2Zr(OPr)_4(OAc)_{x-4} \rightarrow Zr_2O(OAc)_{2x-1}(OPr)_{8-2x-1} + 4\text{AcOPr} \quad (2.5)$$

3. Adding water to the solution promotes hydrolysis of zirconium propoxyacetate oligomers. At this stage stable soluble species of oligomeric zirconium oxide-alkoxy acetate are formed.
The hydrolysis of zirconium propoxyacetate can be started before addition of water and can be promoted by "internal" water, produced in an esterification reaction between acetic acid and propanol [60]:

\[
Zr(\text{OPr})_4(\text{OAc})_4 + y\text{H}_2\text{O} \rightarrow Zr(\text{OAc})_2(\text{OPr})_{4-x-y}(\text{OH})_y + y\text{PrOH} \quad (2.6)
\]

There can occur also condensation reaction, leading to formation of products similar of those of reaction (2.5):

\[
2Zr(\text{OPr})_{4-x-y}(\text{OAc})_2(\text{OH})_y \leftrightarrow Zr_2(\text{OAc})_2(\text{OPr})_{8-2x-y}(\text{OH})_y + \text{H}_2\text{O} \quad (2.7)
\]

Introducing organically modified silicates into the films allows the control of porosity [61], refractive index, and the thickness of the coating and to improve the mechanical properties, the flexibility and transparency of the film without heat treatment. For examples, pure zirconia films can be removed from glass or fused silica substrates with a blade easily and cleanly. In contrast, it is hard to remove hybrid zirconia-ORMOSIL films from the substrates. And some of the pure zirconia channels showed cracks upon aging, but the zirconia-ORMOSIL channels can be kept crack-free over ten days at room temperature.

When organic groups are incorporated into the glass, the shrinkage is low, because the bulky organic components fill the pores between the inorganic oxide chains. The material reaches its final density at low temperature, especially if the organic groups can be cross-linked by epoxy or methacryl polymerization, as is the case for GLYMO in our preparations [13].

The waveguiding properties of doped and undoped zirconia and zirconia-ORMOSIL films were characterized using a commercial prism coupler (Metricon model 2010) at 633nm. Refractive index and the thickness of the
film were simultaneously determined from the propagation angles of the waveguide modes. Comparing the scanning prism coupler results for zirconia and zirconia-ORMOSIL films, it was revealed that zirconia-ORMOSIL films had better waveguiding properties as evidenced by the sharper and better resolved guiding modes. There were no measurable differences in refractive index for dye-doped and undoped waveguides. A Hitachi spectrophotometer was used to study the absorption/transmission properties of the waveguides. The transmission trace of a typical undoped zirconia waveguide of 0.6 μm thickness on fused silica showed excellent transmission from 210 nm to the near infrared (Fig. 2.3). For zirconia-ORMOSIL films, increased absorption in the deep UV (210-280 nm) was observed. Fig. 2.4 showed the absorption spectra of R6G-doped zirconia and LDS 925-doped zirconia-ORMOSIL thin
films. The R6G-doped thin films had an absorption peak near 530 nm, thus allowing efficient pumping by frequency-doubled neodymium-yttrium-aluminum-garnet (Nd:YAG) laser. The absorption peak of LDS 925-doped zirconia-ORMOSIL thin films was at 490 nm. Wide absorption band allowed the relative efficient pumping by frequency-doubled Nd:YAG laser.

![Absorption spectra of R6G-doped zirconia thin film (solid line) and LDS 925-doped zirconia-ORMOSIL thin film (dash-dotted line).](image)

2.3 Fabrication of sol-gel channel waveguides

2.3.1 General process of the photolithographic technique

In a clean-room environment, using the standard photolithographic technique to make a photo-mask and the subsequent etching of the fused silica or glass substrates, we obtained wedge-shaped shallow channels with wide tops and narrow bottoms in glass substrates and true rectangular channels in the fused silica substrates. The basic sequence for imaging a substrate with either
negative or positive photoresist is shown in Fig. 2.5. Typical imaging processes of the etching are as follows [62]:

1. Pretreatment: substrates are chemically and mechanically cleaned to remove surface contaminants. They are then forced-dried and given a short bake to remove residual surface moisture and thereby permit good resist adhesion. Careful control of wafer pretreatment is essential to maintaining high imaging quality. The step is critical for this reason: any defects or contamination allowed at this stage of wafer processing can only be magnified by subsequent processing. The final stage of pretreatment usually involves the

![Diagram of resist imaging process]

Fig. 2.5 Schematic of resist imaging process.
application of a resist adhesion promoter. This serves as an interfacial layer between the substrate and the photoresist. Substrates are coated immediately after priming.

2. Coating: Spin-coating is the most widely used technique for applying photoresist to substrates. The resist is dispensed onto the surfaces of the wafers, which are then accelerated on the coater to provide a thin uniform film across every substrate surface. Coating is performed in an environment with solvent exhaust. The objective of this step is to provide a uniform and defect-free layer of a photo-sensitive masking material.

3. Softbaking: Softbaking is used to remove the solvents present in the spin-coated film of resist and is usually performed immediately downstream from the coater. Infrared, conduction, and microwave heating are used to drive the solvents from the resist coating and render it sensitive to exposure energy. Solvents left in the resist film from improper softbaking will cause poor imaging quality, usually from attack by the developer.

4. Exposure: The goal of exposure is to create a latent image of a desired pattern in the resist film. The role of the photomask is paramount since the mask is a passive tool that gives the resist its latent image. It must therefore be clean and defect-free as the coated and softbaked wafer hopefully was. The result of exposure is a latent image closely matching the pattern of the photomask. Environment cleanliness is very important here as well, since contamination on either the wafer or the mask will be reproduced in the final developed image and cause a reject die or reject wafer, depending on the extent
of the contamination. The important parameters to control are exposure energy uniformity and exposure time control.

5. Development: The developer is sprayed or washed over the exposed resist coating, rinsing and drying follow the develop step, and the parts are agitated, either by spinning or movement, to provide uniform developing action. The concentration, temperature, and the time of application of the developer must be closely controlled and monitored. The development process is one in which the developer selectively attacks and removes either exposed regions (positive resist) or unexposed regions (negative resist), and leave behind the image to serve as the mask for etching. After developing, the wafers are rinsed and dried and sent for inspection prior to being postbaked.

6. Postbaking, or hardbaking: Postbaking, or hardbaking is included to provide “insurance” that the resist is well bonded to the underlying substrate. It is accomplished in the same manner as soft baking. The heat removes any residual moisture from the developing operation and further bonds the resist to the wafer. Intensive postbaking additionally “hardens” the resist by making it chemically inert to the etchant fluid (e.g. Hydrofluoric acid (HF)). Major factors to monitor and control in postbaking are bake time and temperature. Wafers are sent directly etching after postbaking, since delays may negate the effects of the bake.

7. Etching: The goal of etching is to precisely remove the substrate layer left exposed by the developing process. For this reason, complete removal of the developed resist is essential, and any residues left on the substrate layer may prevent or inhibit etching action, a phenomenon called “blocking”. The
etching control parameters are time, uniformity, temperature, and concentration of the etch species (e.g., HF solution). Numerous techniques, often called “black magic”, are associated with the etching process, such as special rotation or movement of wafers during etching that may enhance uniformity of etching.

After etching, wafers are transported to the removal station.

8. Removal: Resist removal is typically done by immersing wafers in a heated resist-stripping solution. The goal of removal is to leave behind a surface completely free of any resist material. After removal, wafers are inspected for defects; the undercutting which is a result of poor resist adhesion or overetching is very possible.

2.3.2 Channels in glass substrates by using photolithographic wet etching technique

Using the photolithographic technique to make a photo-mask combined with the subsequent wet etching of the glass substrates in HF: NH₄F: H₂O solution, we obtained wedge-shaped shallow channels. Since the wet-etching process is isotropic, this means that the maximum channel depth in the chip will be: (channel width - mask width)/2 (see Fig. 2.6). But considering the true situation, when the masking film has a good adhesion on the glass, a semi-circular wall is formed [63], delamination of the photoresist during etching at a fixed rate will result in a tapered wall structure at the edge of the window (Fig. 2.7) [64, 65].

In pure HF solutions delamination of the photoresist occurs due to attack at the glass-photoresist interface. This can be avoided by introducing the buffer
solution NH$_4$F to increase the pH value [66, 67]. However, we still met the channels with a tapered wall structure caused by the delamination after the buffer was used; we attributed that to the contamination at the interface.

![Fig. 2.6 Dimensions of a wet-etched channel. Channel width = mask width + 2 × depth](image)

The detailed block diagram of the preparation procedures of channel waveguides by adopting the wet etching approach to create channels in glass substrates (index of 1.51) is shown in Fig. 2.8.

The main reaction during etching is:

$$\text{SiO}_2 + 6\text{HF} \rightarrow \text{H}_2\text{SiF}_6 + 2\text{H}_2\text{O} \quad (2.8)$$
Wet etching in glass substrates

**Pretreatment:** clean glass substrates
1. Rinse glass substrates in ultrasonic cleaner with DECON (4% in volume), ethanol, acetone, and deionized (DI) water for five minutes respectively and in succession.
2. Boil them in a bath made up of H₂SO₄ and H₂O₂ for about half an hour.
3. Wash them carefully with DI water.
4. Dry them with dry nitrogen (N₂) flush.

**Coating:** apply photosress to glass substrates
1. Bake the substrates for one minute on hot plate.
2. Dip them in a resist adhesion promoter (HMDS).
3. Spin-coat them with the negative resist (AZ5214E) immediately after baking.

**Softbaking:**
Bake the substrates on hot plate about 70 seconds at 80°C to remove the solvents present in the spin-coated film of resist and render them sensitive to exposure energy.

**Exposure:**
1. Expose the coated and softbaked substrate with UV light for 70-80 seconds.
2. Bake them for 70 seconds at 100°C.
3. Expose them with UV light for 70-80 seconds again.

**Development:**
1. Immers the exposed film in a developer solution (AZ 300 MIF) at room temperature for about two minutes.
2. Wash them with DI water and dry them with N₂ flush carefully.

**Postbaking:**
Bake the substrates on hot plate for about 70 seconds at 100°C to remove any residual moisture from the developing operation and further bond resist to the underlying substrate.

**Etching:**
1. Immers the substrates in an etchant (HF: NH₄F: H₂O=1:6:7 in volume) at room temperature. Control the time of etching to get channels with different depth and width.
2. Rinse them with DI water carefully and dry them with N₂ flush.

**Removal:**
1. Rinse the substrates in a photosress stripping solution (acetone) for some minutes.
2. Clean them with DI water and dry them with N₂ flush.

**Characterizing:**
Measure the depth of the channels with a step profiler and the width with microscope.

Fig. 2.8 The block diagram of the preparation procedure of channel waveguides by adopting the wet etching approach.

This equation is a simplification of the reactions occurring during the heterogeneous SiO₂ dissolution [63]. The resist adhesion promoter HMDS (Hexamethyldisilazane) is used to promote the adhesion of resists to wafer.
surfaces. The mechanism whereby HMDS bonds resist to the oxide surface is shown in Fig. 2.9 [62]. HMDS ties up the molecular water on a hydroxylated silicon dioxide surface with a portion of the complex molecule and bonds to the ends of the resist molecule with the other. Thus a twin-ended molecular bonding mechanism makes HMDS a surface-linking adhesion promoter.

Fig. 2.9 HMDS bonding mechanism with silicon dioxide.

Fig. 2.10 shows the microscope images of the cross-sections of the wet-etching channels corresponding to the two situations in Fig. 2.7. Fig. 2.10(a) shows the semi-circular wall structure. In contrast, Fig. 2.10(b) shows the tapered wall structure.

The relationships between the geometric dimensions (viz., top width, base width and depth) of the wet-etching channels and the etching time are summarized in Fig. 2.11. We can see that the widths of the top and the base of channels increased as the etching time. Channels using a wider mask were wider than those using a narrower mask generally. But in Fig. 2.11 (a), channels using the mask of 20 μm were narrower than those using the mask of 10 μm possibly due to the delamination between the photoresist and the
substrates during the etching process. The depths of channels linearly increased as the etching time, which were independent on the mask widths.

![Microscope images of the cross-sections of the wet-etching channels. (a) semi-circular wall structure, (b) tapered wall structure.](image)

Fig. 2.10 Microscope images of the cross-sections of the wet-etching channels. (a) semi-circular wall structure, (b) tapered wall structure.
2.3.3 Channels in fused silica substrates by using photolithographic dry etching technique (Inductive-coupled plasma etching) [62]

Dry etching by inductive-coupled plasma etching allowed the creation of truly rectangular channels in the fused silica substrates. Accurate and reproducible pattern transfer to maintain dimensional control is a prominent criterion in dry-etching technology.

Plasma etching uses molecular gases that contain one or more halogen atoms. In a plasma, species of these become reactive with semiconductor layers and form volatile compounds. The inert molecular gas in a slow discharge generates ions, radicals, and reactive atomic species that combine with oxides, silicon, and metals to produce a volatile by-product. Under an operating pressure of $10^{-1}$ to $10^{-11}$ torr, these species are absorbed on the semiconductor surface, the reaction occurs, and the by-products that are subsequently diffuse in the etch chamber environment.

Here we will simply describe the fabrication process of the dry etching samples (Fig. 2.12).
Fig. 2.11 Variations of geometric dimensions (top width (a), base width (b) and depth (c)) of wet-etching channels as a function of etching time using masks with different widths.
Dry etching in fused silica substrates

**Preparation:** Clean quartz substrates

1. Boil quartz substrates in baths \(\text{H}_2\text{SO}_4\cdot\text{H}_2\text{O}_2\) (3:1) and \(\text{HNO}_3\cdot\text{H}_2\text{O}_2\cdot\text{H}_2\text{O}\) (1:2:7) at 80°C for 10 minutes respectively and subsequently.
2. Rinse them in ultrasonic cleaner with DI water for 10 minutes at 80°C.
3. Dry them with \(\text{N}_2\) flush.

**Evaporation of a Cr layer:**

Evaporate a protective layer Cr on the surface of the wafer with an accelerating voltage 10 KV and an electric current 0.36 A. The coating rate is 0.2 nm/second and the final thickness of this layer is 100 nm.

**Coating:** Apply photoresist to quartz substrates

Spin-coat the wafer with e-beam photoresist (UVIII). Control the coating speed (3000 nm/minute) to get a uniform photoresist layer with thickness of 300 nm.

**Softbaking:**

Bake the substrates on hot plane for 90 seconds at 130°C.

**Exposure:**

Expose the coated and softbaked substrates with e-beam at a current (100 pA).

**Postbaking:**

Bake the substrates on hot plane for 90 seconds at 130°C.

**Development:**

1. Immers the exposed substrates in a developer solution (CD 26) for 10 minutes, then rinse them with DI water and dry them with \(\text{N}_2\) flush.
2. Remove the exposed photoresist with acetone in an ultrasonic cleaner for about 2 minutes and dip the min ethanol solution subsequently.
3. Wash them with DI water and dry them with \(\text{N}_2\) flush carefully.

**Etching:**

Inductively coupled plasma etches the substrates with the plasma gas \(\text{SF}_6\) at a pressure 22 Pa. The etching speed is 40 nm/minute.

**Removal:**

1. Rinse the substrates with a Cr stripping solution CR7 (Ce \(^{+7}\)/HIO4) for 3 minutes
2. Clean them with DI water and dry them with \(\text{N}_2\) flush.

**Characterizing:**

Measure the depth of the channels with a top profiler and width with microscope.

Fig. 2.12 The block diagram of the preparation procedure of channel waveguides by adopting the dry etching approach.
Chapter III

Coupled-wave theory and experimental setup of distributed feedback channel waveguide lasers

3.1 Coupled-wave theory of distributed feedback lasers [17]

Distributed feedback (DFB) laser action is made by the presence of periodic perturbations in the gain medium that provide feedback by backward Bragg scattering [68]. Instead of the conventional cavity mirrors, the feedback for lasing oscillation was provided via backward Bragg scattering in periodic structures. So the DFB structures are compact and provide a spectral selection of high degree. The periodic perturbations can be realized from the spatial modulation of the refractive index [68] or gain, or a combination of both [16]. In a waveguide structure, a periodic change of the guiding film thickness was also proven effective in producing DFB laser action [69]. The periodic perturbations can be permanent or transient, with the transient effect often produced by crossing two beams from the output of the same laser to generate a concentration grating [16, 19]. The coupled-wave model based on the scalar wave equation developed by Kogelnik and Shank is described in this thesis.

Fig. 3.1 shows a simplified illustration which demonstrates the oscillation mechanism of a DFB structure. There are two waves in the diagram represented by arrows, one which travels to the left and the other to the right. As each wave travels in the periodic structure, it receives light at each point along its path by Bragg scattering from the oppositely traveling wave. This
creates a feedback mechanism which is distributed throughout the length of the periodic structure. So we name it as “distributed feedback”. Since the periodic structure has gain, one can see that, with sufficient feedback, there will be a condition for laser oscillation. Also, spectral selection occurs due to the wavelength sensitivity of the Bragg effect.

Coupled-wave theory of DFB lasers is a linear theory. A linear analysis is made to describe the modes of a DFB structure, and to predict the resonant frequencies, the corresponding threshold gain and the spectral selectivity. Nonlinear effect such as gain saturation is not considered. The model is based on the scalar wave equation for the electric field.
\[
\frac{\partial^2}{\partial z^2} E + k^2 E = 0,
\]  

(3.1)

where \( E \) is the complex amplitude of a field of angular frequency \( \omega \), which is assumed to be independent of the \( x \) and \( y \) coordinates. The constants of the laser medium are also independent of \( x \) and \( y \), but vary periodically as a function of the \( z \) coordinate, which points in the direction of propagation (Fig. 3.1).

We assume a spatial modulation of the refractive index \( n(z) \) and of the gain constant \( \alpha(z) \) of the form

\[
\begin{align*}
    n(z) &= n + n_1 \cos 2\beta_0 z, \\
    \alpha(z) &= \alpha + \alpha_1 \cos 2\beta_0 z,
\end{align*}
\]

(3.2)

where \( n \) and \( \alpha \) are the average values of the parameters of the medium and \( n_1 \) and \( \alpha_1 \) are the amplitudes of the spatial modulation. At the Bragg condition we have

\[
\beta_0 = n\omega_0 / c = n\omega / c,
\]

(3.3)

which implies a spatial periodicity \( \pi / \beta_0 \) equal to half the wavelength \( \lambda / n \) of the light in the medium. In addition, it is assumed that the DFB laser oscillates at or near the Bragg frequency \( \omega_0 \) (i.e., \( \omega \approx \omega_0 \)), that the gain is small over distance of the order of a wavelength \( \lambda_0 \), and that the perturbation of the refractive index and the gain are small, i.e.,

\[
\begin{align*}
    \alpha &\ll \beta_0 \equiv 2\pi n / \lambda_0, \\
    n_1 &\ll n, \\
    \alpha_1 &\ll \beta_0.
\end{align*}
\]

(3.4)

These assumptions, discussed in greater detail in [70], allow us to write the \( k \) constant of the wave equation (3.1) in the form
\[ k^2 = \beta^2 + 2j\alpha\beta + 4k\beta \cos 2\beta_0 z, \]

where \( \beta = \omega / c \) and the coupling constant \( \kappa \) is defined by

\[ \kappa = \pi n_i / \lambda_0 + \frac{1}{2} j\alpha. \]

The coupling constant is a central parameter of our model. It measures the strength of the backward Bragg scattering and thus the amount of feedback (per unit length) provided by the structure.

In principle, a periodic perturbation of the medium generates an infinite set of diffraction orders. But in the vicinity of the Bragg frequency only two orders are in phase synchronism and of significant amplitude. All other orders are neglected in the coupled-wave model [70]. As indicated in Fig. 3.1 (b), the two significant waves in the DFB structure are two counter-propagating waves \( R \) and \( S \). These waves grow because of the presence of gain and they feed energy into each other due to Bragg scattering. We describe these waves by complex amplitudes \( R(z) \) and \( S(z) \), and write the electric field as the sum

\[ E(z) = R(z) \exp(-j\beta_0 z) + S(z) \exp(j\beta_0 z). \]

In view of Eq. (3.4) these amplitudes are varying slowly so that their second derivatives \( \partial^2 R / \partial z^2 \) and \( \partial^2 S / \partial z^2 \) can be neglected [70].

With all these assumptions we insert Eq. (3.7) into the wave equation, compare terms with equal exponentials, and obtain a pair of coupled-wave equations of the form

\[ -R' + (\alpha - j\delta)R = j\kappa S, \]
\[ S' + (\alpha - j\delta)S = j\kappa R. \]

The parameter \( \delta \) is a normalized frequency parameter defined by

\[ \delta = (\beta^2 - \beta_0^2) / 2\beta \approx \beta - \beta_0 = n(\omega - \omega_0) / c. \]
It is a measure for the departure of the oscillation frequency $\omega$ from the Bragg frequency $\omega_0$. At the Bragg condition, we have $\delta = 0$.

The coupled-wave equations (3.8) describe wave propagation in the DFB structure in the presence of a gain and periodic perturbations of the medium. Since this model is that of a self-oscillating device, there are no incoming waves, and the internal waves start with zero amplitudes at the device boundaries, receiving their initial energy via scattering from the counter-running wave, we consider a structure of length $L$, extending from $z = -\frac{1}{2}L$ to $z = \frac{1}{2}L$. The boundary condition for the wave amplitudes are then

$$R(\frac{1}{2}L) = S(\frac{1}{2}L) = 0.$$  (3.10)

The coupled-wave equations (3.8) together with the boundary conditions of (3.10) specify the electromagnetic field in this model of the DFB laser. The corresponding solution yields self-consistent steady-state field configurations or oscillation states, i.e., the “modes” of the periodic structure.

The general solution to the coupled-wave equations (3.8) is of the form

$$R = r_1 e^{\gamma z} + r_2 e^{-\gamma z},$$  
$$S = s_1 e^{\gamma z} + s_2 e^{-\gamma z},$$  (3.11)

with the complex propagation constant $\gamma$ obeying the dispersion relation

$$\gamma^2 = \kappa^2 + (\alpha - j\delta)^2.$$  (3.12)

Because of the assumed symmetry of the device, symmetric $[E(-z) = E(z)]$ and anti-symmetric $[E(-z) = -E(z)]$ field solutions can be got, which implies the relationships

$$r_1 = \pm s_2,$$  
$$r_2 = \pm s_1.$$  (3.13)
The boundary condition (3.10) provides a further set of relations, namely,
\[ \frac{\eta_1}{r_2} = \frac{s_2}{s_1} = -e^{yL}. \]  
(3.14)

Using the above results, the longitudinal field distribution of the modes of a DFB structure can be described in the compact form

\[ R = \sinh \gamma(z + \frac{1}{2}L), \]
\[ S = \pm \sinh \gamma(z - \frac{1}{2}L). \]  
(3.15)

where \( L \) is the length of the structure. A discrete set of eigenvalues \( \gamma \) corresponds to a set of modes and this set corresponds to a structure with given length and given coupling \( \kappa \). To determine the eigenvalues, insert (3.15) into the coupled-wave equations (3.8), form the sum and the difference of the resulting equations, and obtain

\[ \gamma + (\alpha - j\delta) = \pm jke^{\gamma L}, \]
\[ \gamma - (\alpha - j\delta) = \mp jke^{-\gamma L}. \]  
(3.16)

Note that one obtains dispersion relation (3.12) by multiplying these two expressions. Addition of the two expressions yields a transcendental equation for the eigenvalue \( \gamma \)

\[ \kappa = \pm j\gamma / \sinh \gamma L. \]  
(3.17)

These eigenvalues are generally complex valued and each value of \( \gamma \) has a corresponding threshold gain constant \( \alpha \) and a resonant frequency \( \delta \). They obey the relation

\[ \alpha - j\delta = \pm j\kappa \cosh \gamma L = \gamma \coth \gamma L, \]  
(3.18)

which is derived by subtraction of equations (3.16).

Hereto, the (threshold) modes of the DFB structure have been obtained from the above solutions of the coupled-wave equations. For a given length \( L \)
and coupling $\kappa$, the eigenvalue $\gamma$ of each mode can be calculated by solving equation (3.17), to obtain the characteristic field pattern, the characteristic threshold gain and the resonant frequency of each mode.

3.2 Experimental setup

The transverse pumping arrangement was adopted in our DFB laser experiments. Distributed feedback and gain were obtained by optically pumping the waveguides with fringes formed by the interference of two coherent laser beams. A nanosecond frequency-doubled Nd:YAG laser (Continuum Surelite II) at 532 nm was chosen as the pump source considering the absorption of R6G and LDS 925 (Fig. 2.4). A holographic grating was employed as a beam splitter to get the two coherent pump beams from the same laser source, which considerably reduced the coherence requirement on the pump laser [20]. The Nd:YAG laser at 532 nm is an s-polarized (~95%) laser source. When the two coherent beams overlap on the zirconia films, pure intensity interference patterns are generated dynamically [71]. Thus we attribute the DFB modulation to the periodic gain modulation. DFB laser oscillation arises from the coherent Bragg scattering in the periodic gain modulation of the active medium. In such an arrangement, grating-like interference patterns with high visibility are required to reduce the pump threshold energy, to lead to narrow linewidth of DFB laser output and to improve the output energy and stability.

The optical arrangement of the DFB zirconia and zirconia-ORMOSIL channel waveguide lasers is illustrated in Fig. 3.2. A polarizer and a filter were
placed in the exit path of the Nd:YAG laser to set the polarization direction of
the pump beam to be s-polarized, and to filter out the unwanted 1.06 µm
component, respectively. Then the pump beam was perpendicularly reflected
on the holographic grating by a high-reflection (HR) plane mirror. The
holographic grating of 1800 lines/mm diffracted the pump beam into two parts
(±1 orders) of approximately equal intensities. These two diffracted beams
were redirected by two identical rotatable HR plane mirrors to combine in the
waveguide samples at an intersection angle of 2θ. Two cylindrical lenses with
short focal length, f = 100 mm, were used to focus each of the pump beams
independently. The lenses with horizontal optical axis were chosen to give a
long narrow transversely pumped region in the thin films. The size of the
pumped region, which was equal to the size of the image line of the pump
beams on the surface of the sol-gel samples, was approximately 6.8 × 0.1 mm².
The interference patterns were generated in the combined region. The period of
the gain modulation (the fringe pattern) is given as
\[ \Lambda = \frac{\lambda_p}{2 \sin \theta}, \]  
(3.19)
where \( \lambda_p \) is the wavelength of the pump laser source and \( \theta \) is half of the pump beam intersection angle at the gain medium. The samples, which are mounted
on a translation stage, can be finely translated along the Y direction. Hence the
intersection angle of the two crossing beams can be varied continuously by
finely rotating those two HR plane mirrors and by moving the samples,
resulting in the continuous variation of the period of the interference patterns.
Distributed feedback occurs near the Bragg condition [17], that is, when the
wavelength in the medium is equal to $2\Lambda$. Denoting $\lambda_e$ as the DFB oscillation wavelength in air, the Bragg condition can be written as

$$2\Lambda = M \frac{\lambda_e}{n_{\text{eff}}},$$  \hspace{1cm} (3.20)

where $n_{\text{eff}}$ is the effective index of the waveguides at $\lambda_e$ as determined by the guide parameters and the mode being considered. $M$ is the Bragg reflection order. Substituting (3.19) into (3.20), we can obtain the spectral selection equation of DFB waveguide lasers

$$\lambda_e = \frac{n_{\text{eff}} \lambda_p}{M \sin \theta}.$$ \hspace{1cm} (3.21)

This equation is the theoretical base of the wavelength tuning of DFB waveguide lasers.

The desired pump energy density can be adjusted by the translational movement of the two cylindrical lenses back and forth combined with attenuators. Increasing the pump energy above the threshold at an appropriate intersection angle (an appropriate period of modulation), the DFB lasing will emit along the length of the pumped zone. The output beams of DFB lasers have large divergence angles, so that a 100 mm convex lens was used to collect and collimate the output beams. The DFB laser signal was coupled into an optical fiber probe that is connected to a 0.3 m focal length spectrograph in conjunction with an intensified charge-coupled device (ICCD) for spectral measurement (zone A in Fig. 3.2). A fast sub-nanosecond phototube (Hamamatsu R1193U-52), in combination with a fast digital oscilloscope (1.5GHz, LeCroy 9362), was used to measure the waveforms and output.
energy of the DFB laser pulses (zone C in Fig. 3.2). The polarization property of the DFB laser output can be determined by a polarizer inserted in front of the fiber probe. The output beam profiles were captured using a digital camera (zone B in Fig. 3.2). The pump energy was measured by a joulemeter (Gentec DE-200) in conjunction with a digital oscilloscope (300MHz, LeCroy 9361).

Fig. 3.2 The experimental setup of DFB sol-gel waveguide lasers. The function of zone A is of spatial spectra measurement; the function of zone B is of beam profile measurement; and the function of zone C is of temporal waveform and output energy measurements.
Chapter IV

One-dimensional and two-dimensional optical waveguide analysis

In our experiments, DFB lasers were realized in both one-dimensional (1-D) and two-dimensional (2-D) optical waveguide structures. The DFB lasing action will also obey the mode and propagation properties of the waveguides. Hereafter, a brief introduction on the theories of 1-D and 2-D waveguides is presented.

4.1 1-D planar waveguide analysis [72, 73]

In this section, a brief introduction on the theory of a one-dimensional planar waveguide based on TE mode and TM mode propagations in a dielectric slab is presented.

Fig. 4.1 Cross-section structure of a planar optical waveguide and the configurations of transverse electric (TE) mode and transverse magnetic (TM) mode. A cross indicates the field entering the page, and “o” indicates the field pointing out of the page.
Fig. 4.1 illustrates the simplest structure of a planar optical waveguide. It consists of three layers: the guiding layer, the substrate layer and the cover layer. The guiding layer is sandwiched by the substrate and the cover. We will just consider the general situation of the asymmetric waveguide. Here, the refractive index of the guiding layer $n_f$ is larger than those of the substrate and the cover ($n_f > n_s > n_c$). Due to the total reflection at the two interfaces, the light ray can be confined and zigzags in the guiding layer. There exist five key parameters characterizing the waveguide. They are $n_f$, $n_s$, $n_c$, the guiding layer thickness $h$ and the propagation constant $k = 2\pi / \lambda = \omega / c$.

Assuming a light ray is traveling in the guiding layer, the total wavevector, $k = k_0 n_f$, can be decomposed into the longitudinal component $\beta$ and the transverse component $\kappa$, respectively. They obey the relation

$$k^2 = \kappa^2 + \beta^2. \quad (4.1)$$

The longitudinal wavevector $\beta$ is used to identify the individual modes, that is to say, $\beta$ is the eigenvalue of the guiding mode. For a guided mode wave, $\beta$ must satisfy the following condition

$$k_s n_s < \beta < k_s n_f. \quad (4.2)$$

For the TE modes in an asymmetric waveguide, through solving the wave equation

$$\frac{\partial^2 E_y}{\partial x^2} + (k_0^2 n_f^2 - \beta^2) E_y = 0. \quad (4.3)$$

and combining with the continuous boundary conditions, we can derive the transverse portions of the electric field amplitudes in the three regions:
\[ E_y = A e^{-\gamma_x}, x > 0 \]
\[ E_y = A[\cos(\kappa_f x - \frac{\gamma_s}{\kappa_f} \sin(\kappa_f x))], -h < x < 0 \]  
\[ E_y = A[\cos(\kappa_f h + \frac{\gamma_s}{\kappa_f} \sin(\kappa_f h))] e^{\gamma_s(x+h)}, x < -h \]  

(4.4)

where \( A \) is the amplitude at the \( x=0 \) interface, \( \gamma_c = \sqrt{\beta^2 - k_0^2 n_0^2} \) and \( \gamma_s = \sqrt{\beta^2 - k_0^2 n_s^2} \) are the attenuation coefficients, \( \kappa_f = \sqrt{k_0^2 n_s^2 - \beta^2} \) is the transverse wavevector. Using the continuity of \( \frac{\partial E_y}{\partial x} \) at \( x=-h \), the eigenvalue equation for \( \beta \) can be deduced from Eq. (4.4), with the form

\[ \tan(h \kappa_f) = \frac{\gamma_c + \gamma_s}{\kappa_f [1 - \frac{\gamma_c \gamma_s}{\kappa_f^2}]} \]  

(TE mode)

(4.5)

Similar results can also be obtained in the TM case.

According to Eq. (4.2), we define the effective refractive index of the guiding mode as

\[ n_{eff} = \frac{\beta}{k_0}. \]  

(4.6)

4.2 2-D channel waveguide analysis using the Marcatili method [74]

Fig. 4.2 shows two kinds of basic geometries for channel waveguides.

The guide is a dielectric rod of refractive index \( n \) immersed in another dielectric of slightly smaller refractive index \( n(1-\Delta) \); both are in contact with a third dielectric which may be air (Fig. 4.2 a) or a dielectric of refractive index \( n(1-\Delta) \) (Fig. 4.2 b). These geometries are attractive not only because of their simplicity, precision of construction, and mechanical stability, but also because by choosing \( \Delta \) small enough, single-mode operation can be achieved with the
transverse dimensions of the guide large compared with the free space wavelengths. This relaxes the tolerance requirements.

![Diagram of 2-D optical waveguides](image)

Fig. 4.2 Configurations of 2-D optical waveguides.

For a general analysis, we redraw in Fig. 4.3 the cross section of the immersed channel waveguide (Fig. 4.2 b) and subdivide it into several different areas. Five of the areas have their refractive indices specified by \( n_1 \) to \( n_5 \); we do not specify the four shaded areas. The reason for these choices will become obvious. We will introduce a drastic simplification which enables one to get a closed form solution. This simplification arises from observing that, for well-guided modes, the field decays exponentially in regions 2, 3, 4 and 5; therefore, most of the power travels in region 1, with a small part of it travels in regions 2, 3, 4, 5, and even less travels in the four shaded regions. Consequently, only a small error would be introduced into the calculation of fields in region 1 if one does not properly match the fields along the edges of the shaded areas.

The matching of modes only along the four sides of region 1 can be achieved by assuming simple field distributions. Thus the field components in region 1 vary sinusoidally in the \( x \), \( y \) direction; and those in 2 and 4 vary sinusoidally along \( x \) and exponentially along \( y \); and those in region 3 and 5
vary sinusoidally along $y$ and exponentially along $x$. The propagation constants $k_{x1}$, $k_{x2}$ and $k_{x4}$ along $x$ in media 1, 2 and 4 are identical and independent of $y$. Similarly, the propagation constants $k_{y1}$, $k_{y3}$ and $k_{y5}$ along $y$ in the regions 1, 3 and 5 are also identical and independent of $x$.

Fig. 4.3 Cross section of a channel waveguide immersed in different dielectrics.

All the modes in the channel waveguides are hybrid and wave guiding occurs because of total internal reflection. The largest field components are perpendicular to the axis of propagation; the modes are essentially of the TEM kind and can be grouped in two families, $E_{pr}^x$ and $E_{pq}^y$. The main field components of the members of the first family are $E_x$ and $H_y$, while those of the second are $E_y$ and $H_x$. The sub-index $p$ and $q$ indicate the number of extrema of the electric or magnetic field in the $x$, $y$ direction, respectively. Naturally, $E_{11}^x$ and $E_{11}^y$ are the fundamental modes; we will concentrate on them when we discuss the transmission properties in the next chapter.
4.2.1 The $E_{pq}^y$ modes: Polarization in the $y$ direction

As discussed above, the main transverse field components of $E_{pq}^y$ modes are $E_y$ and $H_z$. They are depicted in solid and broken lines, respectively, in Fig. 4.3 for the fundamental mode $E_{11}^y$. We solve the Maxwell's equations by neglecting the power propagating through the shaded areas, so the fields must be matched only along the sides of region 1.

The field components in the $\nu$th area in Fig. 4.3 are [75]:

$$
\begin{align*}
M_1 \cos(k_{1x}x + \alpha) \cos(k_{1y}y + \beta) \quad (\nu = 1) \\
M_2 \cos(k_{1x}x + \alpha) \exp(-ik_{1y}y) \quad (\nu = 2) \\
M_3 \cos(k_{2y}y + \beta) \exp(-ik_{2x}x) \quad (\nu = 3) \\
M_4 \cos(k_{2y}y + \alpha) \exp(ik_{2x}y) \quad (\nu = 4) \\
M_5 \cos(k_{2y}y + \beta) \exp(k_{2x}x) \quad (\nu = 5)
\end{align*}
$$

$$
H_{\nu y} = \exp(-ik_{2z}z + i\omega t)
$$

$$
H_{\nu y} = 0,
$$

\begin{align*}
H_{\nu y} &= -\frac{i}{k_z} \frac{\partial^2 H_{\nu y}}{} , \\
E_{\nu y} &= -\frac{1}{\omega \varepsilon n_{\nu}^2 k_z} \frac{\partial^2 H_{\nu y}}{} , \\
E_{\nu y} &= \frac{k^2 n_{\nu}^2 - k_{\nu y}^2}{\omega \varepsilon n_{\nu}^2 k_z} H_{\nu y} , \\
E_{\nu y} &= \frac{i}{\omega \varepsilon n_{\nu}^2} \frac{\partial H_{\nu y}}{}.
\end{align*}
$$

(4.7)

in which $M_\nu$ determines the amplitude of the field in the $\nu$th medium; $\alpha$ and $\beta$ locate the field maxima and minima in region 1; $\omega$ is the angular frequency; $\varepsilon$ and $\mu$ (appearing in $k^2 = \omega^2 \varepsilon \mu$) are the permittivity and permeability of free space.

The functional dependences of the $E_{pq}^y$ modes and $E_{pq}^z$ modes are depicted in Fig. 4.4. In the $\nu$th medium the refractive index is $n_\nu$, and the propagation constants $k_\nu$, $k_{\nu y}$, and $k_z$ are related by
\[ k_{z}^{2} + k_{x}^{2} + k_{y}^{2} = \omega^{2} \mu n_{z}^{2} = k_{0}^{2}. \]  

(4.8)

We call \( k_{z} \) the axial propagation constant and \( k_{x} \) and \( k_{y} \) the transverse propagation constants along the \( x \) and the \( y \) directions, respectively, in the \( \nu \)th medium \((\nu = 1, 2, \cdots 5)\). Furthermore, let us call

\[ k_{\nu} = k n_{\nu} = \frac{2\pi}{\lambda} n_{\nu}, \]  

(4.9)

which is the propagation constant of a plane wave in a medium of refractive index \( n_{\nu} \), and free-space wavelength \( \lambda \). To match the fields at the boundaries between region 1 and the regions 2 and 4, we have assumed that in Eq. (4.7)
and similar to match the fields between media 1, 3 and 5,
\[ k_{x1} = k_{x3} = k_{x5} = k_x, \]
This means that the fields in media 1, 2 and 4 have the same \( x \) dependence and similarly those in medium 1, 3 and 5 have identical \( y \) dependence.

Before finding the characteristic equations, let us assume the refractive index \( n_1 \) of the guide to be only slightly larger than the others. That is
\[ \frac{n_1}{n_{2, 3, 4, 5}} - 1 \ll 1. \]

As a consequence, only modes made of plane wavelets impinging at grazing angles on the surface of medium 1 are guided. Since this implies that
\[ k_x \ll k_z, \]
the field components \( E_z \) in Eq. (4.7) can be neglected.

Matching the remaining tangential components along the edges of region 1 and from Eq. (4.7) we can obtain the following transcendental equations
\[ k_xa = p\pi - \tan^{-1} \frac{\eta_2 k_x}{n_x^2} - \tan^{-1} \frac{\eta_4 k_x}{n_x^2}, \]
and
\[ k_yb = q\pi - \tan^{-1} \frac{n_x^2}{n_y^2} - \tan^{-1} \frac{n_x^2}{n_y^2} k_xk_y, \]
in which
\[ \xi_{3, 5} = \frac{1}{|k_{x3, 5}|} \left[ \left( \frac{\pi}{A_{3, 5}} \right)^2 - k_x^2 \right]^{1/2}. \]
\[ \eta_{2, 4} = \frac{1}{|k_{12, 4}|} = \frac{1}{\left[ \left( \frac{\pi}{A_{2, 4}} \right)^2 - k_y^2 \right]^{\frac{1}{2}}} , \quad (4.17) \]

and

\[ A_{2, 3, 4, 5} = \frac{\pi}{(k_1^2 - k_{2, 3, 4, 5})^{\frac{1}{2}}} = \frac{\lambda}{2\left( n_1^2 - n_{2, 3, 4, 5}^2 \right)^{\frac{1}{2}}} . \quad (4.18) \]

In the transcendental equations (4.14) and (4.15), \( a \) and \( b \) are the transverse dimensions of the guiding rod, \( p \) and \( q \) are the arbitrary integers characterizing the order of the propagating mode, and the \( \tan^{-1} \) functions are to be taken in the first quadrant. The amplitude of each field component in medium 3 (Fig. 4.3) decreases exponentially along the \( x \) direction. It decays by \( 1/e \) in a distance \( \xi_3 = 1/|k_{13}| \). Similarly, \( \xi_5, \eta_2 \) and \( \eta_4 \) measure the "penetration depths" of the field components in media 5, 2, and 4, respectively.

For well-guided modes, most of the power travels within medium 1, which implies that

\[ \left( \frac{k_x A_{3, 5}}{\pi} \right)^2 \ll 1 \quad \text{and} \quad \left( \frac{k_y A_{2, 4}}{\pi} \right)^2 \ll 1 . \quad (4.19) \]

This allows us to solve the transcendental equations in a closed form through approximates. Their solutions are

\[ k_x = \frac{p\pi}{a} \left( 1 + \frac{A_3 + A_5}{\pi a} \right)^{-1} , \quad (4.20) \]

\[ k_y = \frac{q\pi}{b} \left( 1 + \frac{n_5^2 A_2 + n_4^2 A_4}{\pi n_5^2 b} \right)^{-1} . \quad (4.21) \]
Substituting equations (4.20) and (4.21) in equations (4.8), (4.16) and (4.17), explicit expressions for \( k_x, k_y, \xi, \eta_2 \) and \( \eta_4 \) can be obtained (note that Eq. (4.8) should be deduced to \( k_x^2 + k_y^2 + k_z^2 = \omega^2 \varepsilon \mu n_z^2 = k_i^2 \) in medium 1). The propagation constant \( k_z \), which corresponds to \( \beta \) of the planar waveguide described in the 4.1 section, is:

\[
    k_z = \left[ k_i^2 - \left( \frac{\pi p}{a} \right)^2 \left( 1 + \frac{A_3}{\pi a} \right)^2 - \left( \frac{\pi q}{b} \right)^2 \left( 1 + \frac{n_i^2 A_3 + n_i^2 A_4}{\pi n_i^2 b} \right)^2 \right]^{\frac{1}{2}}. \tag{4.22}
\]

Also, the effective index of the guiding mode can be finally obtained by:

\[
    \eta_{eff} = \frac{k_z}{k_0}. \tag{4.23}
\]

### 4.2.2 The \( E_{pq}^x \) modes: Polarization in the x direction

The main transverse components of the \( E_{pq}^x \) modes are \( E_x \) and \( H_y \). The \( E_{pq}^x \) modes are qualitatively similar to the \( E_{pq}^y \) modes; they differ quantitatively. Using the same process of deduction, we can get the axial propagation constant and the "penetration depth" of the \( E_{pq}^x \) modes in media 2, 3, 4 and 5, which are:

\[
k_z = \left( k_i^2 - k_x^2 - k_y^2 \right)^{\frac{1}{2}},
\]

\[
    \xi_{2, 3, s} = \frac{1}{\left| k_{xs, s5} \right|} = \frac{1}{\left[ \left( \frac{\pi}{A_3} \right)^2 - k_x^2 \right]^{\frac{1}{2}}}, \tag{4.24}
\]

\[
    \eta_{2, 4} = \frac{1}{\left| k_{ys, y4} \right|} = \frac{1}{\left[ \left( \frac{\pi}{A_4} \right)^2 - k_y^2 \right]^{\frac{1}{2}}}
\]

in which \( k_x \) and \( k_y \) are the solutions of the transcendental equations

\[
    k_x a = p\pi - \tan^{-1} \frac{n_i^2}{n_i^2} k_x \xi, \quad k_y b = q\pi - \tan^{-1} \frac{n_i^2}{n_i^2} k_y \xi,
\]

\[
    k_x a = p\pi - \tan^{-1} k_x n_2, \quad k_y b = q\pi - \tan^{-1} k_y n_4. \tag{4.25}
\]
The approximate closed form solutions of these equations are

\[ k_x = \frac{p \pi}{a} \left( 1 + \frac{n_1^2 A_1 + n_2^2 A_2}{\pi n_1^2 a} \right)^{-1}, \]

\[ k_y = \frac{q \pi}{b} \left( 1 + \frac{A_2 + A_4}{\pi b} \right)^{-1}. \] (4.26)

Substituting Eq. (4.26) in Eq. (4.24), the explicit result of the propagation constant is:

\[ k_z = \left[ k_x^2 - \left( \frac{p \pi}{a} \right)^2 \left( 1 + \frac{n_1^2 A_1 + n_2^2 A_2}{\pi n_1^2 a} \right)^2 - \left( \frac{q \pi}{b} \right)^2 \left( 1 + \frac{A_2 + A_4}{\pi b} \right)^2 \right]^{1/2}. \] (4.27)

Finally, we will note that if

\[ \frac{1}{n_1} (n_1 - n_2, 3, 4, 5) \ll 1, \] (4.28)

this result coincides with that in Eq. (4.22), indicating that the \( E_{\nu} \) and \( E_{\mu} \) modes become degenerate. According to the exact numerical solutions of the transcendental equations for the propagation constant \( k_z \) and the result derived using the closed form approximations by Marcatili [74] for a rectangular waveguide shown in Fig. 4.2 (a) (\( n_1/n_2 \approx 1.5, n_3 = n_4 = n_5, n_1/n_4 \approx 1.01 \sim 1.05 \) and the ratio of the width and the height \( a \approx 2b \), which are just the characteristics of the channels in our work), the fundamental modes \( E_{11} \) and \( E_{14} \) are almost degenerate.

We note that the results obtained by Marcatili show that the closed form approximation is just within a few percent of the exact value. Therefore, for simplicity, we will use the closed form to analysis the channel waveguides in our experiments.
4.3 2-D channel waveguide analysis using the effective index method [72]

The effective index method converts a two-dimensional problem into one-dimensional problems. Consider a buried rectangular waveguide shown in Fig. 4.5. To use the effective index method, we first stretch the waveguide out along its thin axis, in this case along the $y$ axis, forming a planar slab waveguide.

![Diagram of a buried rectangular waveguide](image)

Fig. 4.5 General dielectric structure: the core ($n_1$) is surrounded on every side by a lower-index material. The index is neglected in the first-order approximation to solutions to the waveguide.

The thin one-dimensional slab waveguide can be analyzed in terms of TE or TM modes to find the allowed value of $\beta$ for the wavelength and mode of interest (see section 4.1). Once $\beta$ is found, the effective index of the slab is determined through the expression

$$n_{\text{eff}} = \frac{\beta}{k_o},$$

(4.29)
where $k_0$ is the vacuum wave vector of the light being guided. After this effective index is determined, we return to the original structure and stretch it along the thick axis (in this case vertical), forming a slab waveguide in the $x$ direction. The modes can now be found, only instead of using the original value of the index for the guiding film, the *effective index* found in the first step must be used. The value of $\beta$ found from this last step is the true value for the mode.

Here we will skip the particular deduction of the effective index of the waveguide since it is analogous to the slab waveguide described in section 4.1. By the way, when performing the wave analysis, we must be careful to use the proper characteristic equations for each waveguide. For example, if the electric field is polarized in the $y$ direction in the Fig. 4.5, then for the thin waveguide the field will appear to be a TM mode, and the appropriate characteristic equation must be used. When the thick slab is analyzed, the field will look like a TE mode, and so the TE characteristic equation should be used to find $\beta$. 
Chapter V

Distributed feedback channel waveguide lasers tunable in the visible

Passive and active planar optical waveguides based on sol-gel materials have generated much research interest because of the versatility of the sol-gel technique and the potential applications in integrated optics. Single-mode waveguides [76], multi-mode waveguides [43] and symmetric waveguides [44] can be fabricated by sol-gel method. Organic dye [77] and rare earth elements [78] can be introduced into these waveguides as functional dopants. Light amplification was observed in dye-doped sol-gel waveguides [77, 14]. DFB laser action in titania silica films [41], in zirconia films and in zirconia-ORMOSIL films [42] have been successfully demonstrated. The laser output was found to be in excellent agreement with the one-dimensional waveguide theory, demonstrating the high optical quality of the sol-gel planar waveguide lasers [42].

However, the planar waveguide lacks lateral confinement. In most practical applications in integrated optics, the rectangular dielectric waveguide is the most commonly used structure on which many of the active or passive devices (i.e., waveguide filters, optical switches, multiplexors, etc.) are in fact based. The rectangular waveguides are rectangular dielectric strips embedded in other dielectrics of lower refractive index. Active layers which can be easily deposited by sol-gel method are built in the dielectric strips to render the
waveguides optically active (e.g., waveguide lasers). In this chapter, we will report the fabrication and the characteristics of dye-doped sol-gel channel waveguide lasers operated in the visible region.

5.1 Rhodamine 6G-doped zirconia planar and channel waveguides

By low temperature sol-gel method, we prepared high quality Rhodamine 6G (R6G)-doped zirconia thin films. The preparation procedures have been described in chapter II. The initial dye concentration was typically $3 \times 10^{-3}$ M. R6G-doped zirconia films were obtained by spin-coating the final solutions upon glass or fused silica substrates. The refractive indices of the glass and fused silica substrates were 1.51 and 1.46, respectively. Fig. 5.1 shows an atomic-force microscopy (AFM) image of R6G-doped zirconia films. The smooth surface revealed that the sol-gel zirconia was an ideal material for the fabrication of optical waveguides.

![Fig. 5.1 AFM image of R6G-doped zirconia films with a roughness of 0.17 nm. Refractive index 1.56; film thickness 0.62 μm; concentration of R6G $2 \times 10^{-3}$ M.](image)
The waveguiding properties of R6G-doped zirconia planar waveguides were characterized with a prism coupler system at 633 nm. Depending on the spin speed and viscosity of the sol-gel solution, and also the storage conditions, the thickness and refractive index of zirconia films varied from 0.6 to 1.4 µm and from 1.53 to 1.64, respectively. Fig. 5.2 shows that refractive index variation of R6G-doped zirconia films that was aged at room temperature for about 48 hours. It remained essentially constant after the sharp increases within the initial 10 hours. Refractive index up to 1.64 was obtained when the films were kept in an oven at 60 °C for two days. Cladded on one side by the low-index substrate (glass or fused silica) and the other by air, the film-on-substrate structure behaved as an asymmetric waveguide.

![Graph showing refractive index variation of R6G-doped zirconia material.](image)

Fig. 5.2 Refractive index variation of R6G-doped zirconia material.
The fabrication process of channels was presented in detailed in chapter II. By adopting the wet-etching and dry-etching approaches, wedge shaped shallow channels with wide tops and narrow bottoms in glass substrates and truly rectangular channels in fused silica substrates were obtained, respectively. The top and bottom widths of the wet-etching channels, ranged from 11 \( \mu \text{m} \) to 70 \( \mu \text{m} \) and 11 \( \mu \text{m} \) to 58 \( \mu \text{m} \), were roughly determined by the mask width and the etching time, while the depth, ranged from 0.86 \( \mu \text{m} \) to 14.21 \( \mu \text{m} \), as shown in Fig. 2.11, was just dependent on the etching time. Since dry etching by the inductive-coupled plasma etching allowed the creation of truly rectangular channels, rectangular channels of widths varying from 10 \( \mu \text{m} \) to 5 \( \mu \text{m} \) and a depth fixed at 5 \( \mu \text{m} \) were produced.

The R6G-doped zirconia solution was deposited on top of the wet or dry-etching channels by spin coating at a spin speed of 1000 rpm for 70 seconds. The zirconia layer outside of the channel was removed, leaving only that inside to serve as the laser gain medium and to form a channel waveguide. The spin speed and viscosity of the sol-gel solution determined the depth of the zirconia layer in the channel. The typical value was 3 \( \mu \text{m} \). The refractive index of the zirconia layer was obtained by measuring that of zirconia films having the same aging time with the prism coupler. A microscope image is shown in Fig. 5.3 for a rectangular channel waveguide sample with a width of 5 \( \mu \text{m} \). Visually the R6G-doped zirconia layer stood out in red tone against a palely pink background. DFB laser action was realized in these active channel waveguide samples. The relevant DFB experimental setup was illustrated in Fig. 3.2.
Rhodamine 6G, also called rhodamine 590 chloride, is one of the best known of all laser dyes. It is a xanthene derivative, ionic and highly polar, with its chemical structure shown in Fig. 5.4. In ethanol solution, R6G exhibits an absorption peak at 530 nm and a fluorescence peak at 556 nm. It has a high fluorescence quantum yield of 95%, a low intersystem crossing rate, and low excited-state absorption. These properties make it a highly efficiency dye for both pulsed and continuous-wave laser action with fairly good chemical stability.

Figure 5.5 shows the spectra of absorption, fluorescence and amplified spontaneous emission (ASE) of R6G-doped zirconia waveguide. The fluorescence and ASE were measured along the optical axis of the waveguides. The absorption peak was at 536 nm which allowed efficient pumping at 532 nm by frequency-doubled Nd:YAG laser. Broad fluorescence emission
spectrum, which almost liked a mirror image of the absorption spectrum, is centered at 575.7 nm with a bandwidth of about 66 nm. At higher pump energy, narrowing of the spectral width (down to 12 nm) that is indicative of ASE was achieved. In addition, the fluorescence peak at 576 nm in zirconia films was red-shifted compared with that in ethanol at 530 nm. The ASE was further red-shifted to 598 nm. DFB lasing was tuned within the gain range of the ASE spectrum.

![Molecular structure of rhodamine 6G dye](image)

Fig. 5.4 Molecular structure of rhodamine 6G dye.

![Absorption, fluorescence and amplified spontaneous emission spectra for an R6G doped zirconia waveguide](image)

Fig. 5.5 Absorption, fluorescence and amplified spontaneous emission spectra for an R6G doped zirconia waveguide.
5.2 Results and discussion

DFB laser action was first attempted to be observed in R6G-doped zirconia planar waveguides. The dye-doped zirconia solution was spin-coated upon the glass substrate at a spin speed of 4000 rpm for 50 seconds. Then it was sealed and aged at room temperature for one week. The film had a thickness of 0.62 μm and a refractive index of 1.56. Tunable narrow linewidth DFB laser output was obtained when the pump energy was up to 50 μJ per pulse. Accounting for the reflection, scattering and transmission loss of the pump laser, the actual energy delivered to the film was about 8 μJ. Continuous tuning of the output wavelength was achieved by varying the intersection angle $\theta$ of the two crossing beams. Single mode lasing was observed throughout the tuning range from 586 nm to 615 nm. Figure 5.6 (a) shows the spectral variation on the intersection angle. The laser had a structure of a single prominent peak near the centre of the gain profile (598 nm). As the laser was tuned away from the gain centre, significant ASE appeared in the background. The linewidth of the DFB laser output was as narrow as 0.5 nm, which was in fact the resolution limit of our detection system. Figure 5.6 (b) shows the experimental data of the angle tuning versus the theoretical fit (solid line) of Bragg condition Eq. (3.21), in which the effective refractive index 1.53 determined independently by the prism coupler system was used. Very good agreement between the experimental data and the theory was seen.

The output energy of DFB lasing increased linearly as the pump energy increased. It decreased to 50% of the initial energy after 800 shots pumped by 220 μJ at a 0.5 Hz repetition rate, which was 4 to 5 times of the threshold
energy. The state of DFB output was s-polarized which indicated that the DFB lasing came from the TE₀ guiding mode.

Fig. 5.6 (a) Laser emission spectra for the R6G-doped zirconia waveguide laser on glass substrate; (b) Data of angle tuning vs theoretical fit.
Subsequently, we started our investigation of DFB laser action in R6G-doped zirconia channel waveguides. DFB laser action with continuous angle tuning was obtained in both the wet-etched and the dry-etched zirconia channel waveguides. The output wavelength followed the Bragg condition Eq. (3.21), where $\eta_{\text{eff}}$ took on the values of the effective indices for $E_{pq}^+$ modes and $E_{pq}^-$ modes, where $p$ and $q$ were the indices specifying the mode (see chapter IV). Tuning of $\lambda_t$ was achieved by varying the intersection angle and thus the period of gain modulation.

DFB lasing emission was observed in R6G-doped channel waveguides in fused silica substrates when the pump laser energy exceeded 20 µJ. Accounting for the scattering and transmission loss, the actual energy deposited in the film was about 1.6 µJ. Fig. 5.7 shows the DFB laser emission spectrum and the angle tuning results for a channel waveguide with a width of 5 µm and a depth of 3 µm embedded in a fused silica substrate. The linewidth of the DFB lasing was less than 0.5 nm. Typical output energy was 10 nJ. The tuning range was from 570 nm to 608 nm. The tuning data generally followed the solid line, which was the prediction by the Bragg condition for a zirconia layer with a refractive index of 1.55. We noted that the experimental data laid below the fit curve at long-wavelength-range and above the fit curve at short-wavelength-range. It is because we used a fixed value for the effective refractive index to fit the experimental data and ignored the dispersion effect of R6G-doped zirconia layer. In general, the refractive indices of zirconia and zirconia-ORMOSIL films decreased slightly as the propagation wavelength increased in the tuning region.
Fig. 5.7 DFB waveguide laser output spectrum and tuning data for a 5-μm-wide, 3-μm-deep rectangular waveguide. The dye concentration was 3×10⁻⁵ M.

Figure 5.8 displays the lasing output energy at 594 nm varied by the increase of the pump energy. As the pump energy was increased from 1.36 μJ to 26 μJ, the output energy increased essentially linearly and the threshold pump energy was about 1.6 μJ. DFB laser emission spectra near the threshold
pump energy are shown in Fig. 5.9. Fig. 5.10 shows the slope efficiencies for different laser output wavelengths. The straight lines represent the linear fit curves. Obviously, the DFB lasing at 599.4 nm, which was the center of the gain profile, had highest slope efficiency as expected.

The state of polarization of the DFB laser output was also determined. By rotating the polarizer in front of the fiber probe, the output intensity varied periodically as the azimuth angle of the polarizer was varied, as shown in Fig. 5.11. The output energy reached the maximum at 0° and 180°. By contrast, it decreased to almost zero at 90° and 270°. The ratio of the maximum and the minimum was several tens, which indicated that the main component of the laser output was polarized parallel to the substrate. According to the Marcatili theory, we identified the laser output as a $E_{11}^{\parallel}$ mode since the main field components of $E_{pq}^{\parallel}$ modes are $E_{\parallel}$ and $H_{\parallel}$.

The output beam profile was examined using a digital camera positioned close to the exit end of the channel (see Fig. 3.2). A three dimensional plot of light intensity on the focal plane of the camera is shown in Fig. 5.12. A roughly circular beam shape was observed. Figure 5.13 shows the cross sections of the intensity profile in directions parallel ($x$ direction) and perpendicular ($y$ direction) to the surface of the fused silica substrate. The intensity maximum was chosen as the origin of the plot. The cross-section profiles resembled Gaussian shapes as indicated by the solid lines.
Fig. 5.8 Variation of the DFB laser output energy at 594 nm as a function of pump energy.

Fig. 5.9 Lasing spectra at different pump energy near the threshold.
Fig. 5.10 Output energy characteristic for different output wavelength.

Fig. 5.11 Polarization of R6G-doped zirconia channel waveguide laser output.
As mentioned before (chapter IV), the guiding modes are determined by the waveguide parameters, one guiding mode has one effective refractive index and hence corresponds to one specific output wavelength of the DFB lasers. So, the output wavelength of the DFB waveguide laser is a function of the waveguide parameters. The functional relationship between the DFB output wavelengths and the waveguide parameters can be simulated numerically and compared with the experimental data. For better comparison with theory, we just concentrated on the results of the dry-etched channel waveguides.

The $E_{eq}$ modes eigenvalue equation (4.27) was the basic equation of our simulations. The effective indices of the guiding modes can be deduced from this equation. Combined with the Bragg condition, we can obtain the relationship between the DFB laser wavelengths and the waveguide parameters.
According to the nature of the R6G-doped zirconia channel waveguides, we set the refractive index of the zirconia layer to 1.55, and the refractive indices of the cover and the fused silica substrate were taken as 1.00 (air) and 1.46.
respectively. The DFB laser output wavelength for rectangular channels of various widths and a fixed depth of 3 μm at an intersection angle 44° is plotted in Fig. 5.14. The solid line is the prediction of $E_{||}$ mode based on the Marcatili theory. Dark dots represent the experimental data from the channels with widths of 5 μm, 6.5 μm, and 10 μm. All three output wavelengths tend to be the preset propagation wavelength as the channel width increased. The preset wavelength was 591 nm. Reasonable agreement between theory and experiments is observed, attesting to the high optical quality of the sol-gel zirconia rectangular waveguides.

![Figure 5.14 DFB laser output wavelength for rectangular channels of various widths and a depth of 3 μm. The solid line is prediction based on Marcatili's theory.](image)

Our R6G-doped zirconia channel waveguides were photobleached after several thousand shots. Also, some of the zirconia channels showed cracks
upon aging. Close examination of the channels by optical microscopy revealed some surface irregularity inside the etched channels. We believe that the surface irregularity leads to the crack formation. Problems related to crack development could be alleviated by applying an additional dielectric coating before the introduction of the dye-doped layer.

5.3 Summary

We fabricated dye-doped zirconia planar waveguides by spin-coating a R6G-doped zirconia solution on glass or fused silica substrates directly and fabricated channel waveguides using wet or dry etching of glass and fused silica substrates followed by deposition of the sol-gel solution in the channels.

Tunable narrow linewidth DFB laser action was demonstrated in the planar and channel waveguides by crossing two nanosecond laser beams at 532 nm. Wavelength tuning was achieved from 586 to 615 nm and 570 to 608 nm, respectively. The performances of the DFB planar waveguide lasers and channel waveguide lasers were summarized in Table 5.1.

The output mode of the channel waveguide DFB laser was identified as the fundamental $E_{11}$ mode. The beam shape was observed to be roughly circular and can be fitted with Gaussian lines. The dispersion behavior of the laser output was checked by comparing experiments with the predictions of Marcatili's theory.
<table>
<thead>
<tr>
<th>Samples</th>
<th>Planar waveguides</th>
<th>Channel waveguides</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dye</td>
<td>Rhodamine 6G</td>
<td>Rhodamine 6G</td>
</tr>
<tr>
<td>Initial dye concentration (M)</td>
<td>$4 \times 10^{-3}$</td>
<td>$3 \times 10^{-3}$</td>
</tr>
<tr>
<td>Refractive index of the guiding layer</td>
<td>1.56</td>
<td>1.55</td>
</tr>
<tr>
<td>Thickness of the guiding layer (µm)</td>
<td>0.62</td>
<td>3</td>
</tr>
<tr>
<td>Refractive index of the substrate</td>
<td>1.51 (glass)</td>
<td>1.46 (fused silica)</td>
</tr>
<tr>
<td>Fundamental guiding mode</td>
<td>$TE_0$</td>
<td>$E_{11}^\parallel$</td>
</tr>
<tr>
<td>Effective index of the fundamental mode</td>
<td>1.53</td>
<td>1.54</td>
</tr>
<tr>
<td>Pump wavelength (nm)</td>
<td>532</td>
<td>532</td>
</tr>
<tr>
<td>Bragg reflection order</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Center of gain (nm)</td>
<td>598</td>
<td>599.4</td>
</tr>
<tr>
<td>Tuning range (nm)</td>
<td>586 — 615</td>
<td>570 — 608</td>
</tr>
<tr>
<td>Threshold pump energy (µJ)</td>
<td>8</td>
<td>1.6</td>
</tr>
<tr>
<td>Linewidth (nm)</td>
<td>&lt; 0.5</td>
<td>&lt; 0.5</td>
</tr>
<tr>
<td>Lasing mode</td>
<td>$TE_0$ guiding mode, s-polarized</td>
<td>$E_{11}^\parallel$ guiding mode, s-polarized</td>
</tr>
</tbody>
</table>

Table 5.1 Performances of DFB planar waveguide lasers and channel waveguide lasers.
Chapter VI

Near infrared distributed feedback channel waveguide lasers

Lasers emitting in 700-900 nm are extensively used as light sources in scientific research. However, relatively little work has been reported on the properties of solid-state dye lasers emitting in this range. LDS (styryl) series dyes show positive gain from red to near infrared (NIR) spectral range and thus are useful for applications that require NIR laser sources. In the 1980s, NIR dye lasers based on LDS dye-doped solutions were intensively studied [79-84]. After that, a few works reported on the demonstrations of LDS dye-doped polymer lasers. M. Zevin and R. Reisfeld prepared LDS 730-doped zirconia thin films and observed the strong fluorescence emission [14]. Y. Oki et al. realized NIR DFB laser action in LDS dye-doped copolymer \((p-MMA_h : HEMA_i)\) thin films [26, 85]. T. Kobayashi et al. observed the lasing emission at 820 nm from LDS 821-doped plastic waveguides [86]. In this chapter, we will report the fabrication of LDS dye-doped zirconia-ORMOSIL channel waveguides and the demonstration of wavelength tunable NIR DFB lasers.

6.1 LDS dye-doped zirconia-ORMOSIL planar and channel waveguides

The preparation procedures of zirconia-ORMOSIL were described in chapter II. Four types of dyes, namely, LDS 759, LDS 798, LDS 867, and
LDS 925 were doped to make the sol-gel layer active. A few drops of propylene-carbonate (PC) were added in the final solution for fabricating films of LDS dyes in high concentration. Typical dye concentration was $5 \times 10^{-3}$ M. Prepared at room temperature, the refractive index of the zirconia-ORMOSIL layer was around 1.53 as determined by the prism coupler at 633 nm. The chemical formulas and molecular structures of LDS 798 and LDS 925 are illustrated in Fig. 6.1. Those of LDS 759 and LDS 867 are not available because they are proprietary information of Exciton Inc.

![Chemical formulas and molecular structures of LDS 798 and LDS 925.](image)

The dye-doped zirconia-ORMOSIL solution was then deposited on top of glass substrates with channels by spin coating. The depth of the zirconia-ORMOSIL layer in channel was typically 2-4 $\mu$m. At room temperature, zirconia-ORMOSIL channel waveguides can be kept crack-free over ten days.
since the GLYMO introduced enhanced the density and flexibility of the
zirconia matrix. In this work, the molar ratio of GLYMO to zirconia (viz.,
Si:Zr) was kept at 1:1.

A home-made scanning ellipsometer fitted with synchronously rotating
polarizer and analyzer [87, 88] was used to measure the refractive index (n)
and the extinction coefficient (k) of the dye-doped zirconia-ORMOSIL films
from 400 nm to 1200 nm. Three types of detectors, viz. photomultiplier tube
(Hamamatsu R1104), photomultiplier tube (Hamamatsu R316), and InGaAs
detector (Oriel 70348), were used in different spectral regions. The scanning
ellipsometer approach yields spectroscopic information for n and k, both are
critical parameters that define the propagation and loss of an optical wave in
the waveguide [89]. Silicon wafers were used as the substrates. The
measurements followed the standard procedures of ellipsometry. From the ratio
of the intensities of the reflected polarized beams, the values of elliptical
azimuth $\Psi$ and phase angle $\Delta$ were extracted. n and k were then determined at
10 nm interval by a numerical routine [87]. The ellipsometry results for a 0.9-
$\mu$m-thick zirconia-ORMOSIL film with an LDS 925 concentration of $5\times10^{-3}$ M
are illustrated in Fig. 6.2. n and k were 1.523 and $7\times10^{-4}$ around 900 nm,
respectively. Hence the LDS 925-doped zirconia-ORMOSIL layer surrounded
by the glass substrate of lower refractive index behaved as an embedded
channel waveguide with low propagation loss.
Fig. 6.2 Variation of $n$ (refractive index) (a) and $k$ (extinction coefficient) (b) from 400 nm to 1200 nm taken at 10-nm interval.
6.2 Results and discussion

Fig. 6.3 shows the traces of absorption, fluorescence and ASE of the LDS dyes-doped zirconia-ORMOSIL films. The dye concentration was $5 \times 10^{-3}$ M. Wide absorption band allowed the relatively efficient pumping at 532 nm by frequency-doubled Nd:YAG laser. Under the low energy density excitation at 532 nm, wide-band fluorescence was observed. By increasing the pump energy density, narrowing of the fluorescence spectral width indicative of ASE was achieved. Compared with the peak of the fluorescence, ASE showed a red shift for each dye, which can effectively extend the output wavelength of the DFB lasers. The detailed photophysical parameters are listed in Table 6.1.

The optical arrangement for the DFB channel waveguide lasers has been described in chapter III.

DFB laser action was observed in LDS 925-doped channel waveguides in glass substrates when the pump energy exceeded 300 µJ. Accounting for reflection and transmission loss, the actual energy deposited in the film was about 20 µJ. Fig. 6.4 shows the DFB laser emission spectrum and the angle tuning results for a zirconia-ORMOSIL channel waveguide with a width of 30 µm and a depth of 4 µm. The tuning range was from 787.2 nm to 932.7 nm. The line width of the DFB laser was less than 0.5 nm, which was the resolution limit of the spectrograph/ICCD system. The tuning data followed the solid line, which was the prediction by the second-order Bragg condition Eq. (3.21), where $n_{eff}$ took on the value of the effective index for $E_{11}$ mode. The
waveguide mode was deduced from the Marcatili theory by approximating the half-round channel waveguide to a rectangular channel waveguide.
# Table 6.1 Photophysical and DFB lasing properties of LDS dyes-doped waveguide lasers

<table>
<thead>
<tr>
<th>Samples</th>
<th>Zirconia-ORMOSIL thin films</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dyes</td>
<td>LDS 759</td>
</tr>
<tr>
<td>Initial dye concentration (M)</td>
<td>$5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Peak (nm)</td>
<td>492</td>
</tr>
<tr>
<td>Absorption FWHM (nm)</td>
<td>104</td>
</tr>
<tr>
<td>Band (nm)</td>
<td>436-540</td>
</tr>
<tr>
<td>Peak (nm)</td>
<td>713.3</td>
</tr>
<tr>
<td>Fluorescence FWHM (nm)</td>
<td>95.3</td>
</tr>
<tr>
<td>Band (nm)</td>
<td>669.6-764.9</td>
</tr>
<tr>
<td>Peak (nm)</td>
<td>735.7</td>
</tr>
<tr>
<td>ASE FWHM (nm)</td>
<td>23.4</td>
</tr>
<tr>
<td>Band (nm)</td>
<td>721.7-745.1</td>
</tr>
<tr>
<td>Refractive index of the guiding layer</td>
<td>1.538</td>
</tr>
<tr>
<td>Thickness of the guiding layer (µm)</td>
<td>0.9</td>
</tr>
<tr>
<td>Refractive index of the substrate</td>
<td>1.51</td>
</tr>
<tr>
<td>Effective index of TE&lt;sub&gt;0&lt;/sub&gt; mode</td>
<td>1.521</td>
</tr>
<tr>
<td>Pump wavelength (nm)</td>
<td>532</td>
</tr>
<tr>
<td>Bragg reflection order</td>
<td>2</td>
</tr>
<tr>
<td>Intersection angle (°)</td>
<td>32.4-35.3</td>
</tr>
<tr>
<td>Tuning range (nm)</td>
<td>696.4-747.4</td>
</tr>
<tr>
<td>Wavelength span (nm)</td>
<td>51.0</td>
</tr>
<tr>
<td>Linewidth (nm)</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>Lasing mode</td>
<td>TE&lt;sub&gt;0&lt;/sub&gt; mode, s-polarized</td>
</tr>
</tbody>
</table>

Table 6.1 Photophysical and DFB lasing properties of LDS dyes-doped waveguide lasers
Chapter VI Near infrared DFB channel waveguide lasers

Fig. 6.4 DFB waveguide laser tuning data (a) and output spectrum (b) for a 30-μm-wide, 4-μm-deep channel waveguide. The dye concentration was 0.005 M.
The DFB laser output wavelengths for channels of various widths and a fixed depth of 1.8 \( \mu \text{m} \) at an intersection angle of 28.7° are plotted in Fig. 6.5. The solid line is the prediction based on the Marcatili theory. The prediction was for \( E_{11} \) mode for a rectangular waveguide of a depth of 1.8 \( \mu \text{m} \). Refractive index for zirconia-ORMOSIL was 1.53 and that for glass was 1.51. The experimental data were corresponding to channel waveguides with widths of 15.6 \( \mu \text{m} \), 25 \( \mu \text{m} \), and 30 \( \mu \text{m} \). The experimental data fitted quite well with the theoretical curve.

DFB lasers operated at high Bragg orders has been reported in earlier works [88, 18]. High order DFB lasing is of interest because it reduces the resolution required for producing the periodic structure of a DFB laser. The
second order DFB operation, in particular, can be employed to realize the
surface-emitting lasers [90]. We studied DFB lasing of LDS 925-doped
zirconia-ORMOSIL thin films at first, second and third orders of the Bragg
condition. The films were obtained by spin-coating glass substrates with the
dye-doped zirconia-ORMOSIL solution. The film on substrate behaved as an
asymmetric waveguide. DFB lasing at first, second, and third orders was
achieved by crossing the pump beams at the intersection angles required by the
Bragg condition. The results of the tuning of DFB lasing at different Bragg
orders are summarized in Fig. 6.6. For first-order Bragg operation, tuning from
825 nm to 943 nm was realized. For the second Bragg order, the tuning range
was from 809 nm to 932 nm. The larger intersection angle (> 76°) limited the
short-wavelength tuning of DFB lasing at first Bragg order. For the third Bragg
order, tuning range was from 809 nm to 881 nm. The tuning narrowed
considerably as the Bragg order increased, corresponding to a decrease of the
intersection angle. Table 6.2 shows the performances of LDS 925-doped
zirconia-ORMOSIL planar waveguide lasers and channel waveguide lasers.

We also demonstrated the NIR wide-band tuning of DFB waveguide
lasers. By adopting four types of LDS dye-doped zirconia-ORMOSIL thin
films (viz. LDS 759, LDS 798, LDS 867 and LDS 925), continuous tuning
from 696 nm to 932 nm was obtained for the second Bragg order. Fig 6.7
shows the experimental data of the angle tuning versus the theoretical fit of
Bragg condition (solid line). The refractive indices were determined
independently by the prism coupler. Very good agreement between
experimental data and theory was seen. All the results are summarized in Table 6.1.

Fig. 6.6 Wavelength tuning of first-, second- and third-order DFB lasing of LDS 925-doped zirconia-ORMOSIL waveguide laser.

Fig. 6.7 NIR wide-band wavelength tuning vs theoretical fit for LDS dye-doped zirconia-ORMOSIL waveguide laser.
### Table 6.2 Performances of DFB planar waveguide lasers and channel waveguide lasers.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Zirconia-ORMOSIL thin films</th>
<th>Channels</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dye</td>
<td>LDS 925</td>
<td>LDS 925</td>
</tr>
<tr>
<td>Initial dye concentration (M)</td>
<td>$5 \times 10^{-3}$</td>
<td>$5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Refractive index of the guiding layer</td>
<td>1.546</td>
<td>1.538</td>
</tr>
<tr>
<td>Thickness of the guiding layer (μm)</td>
<td>0.84</td>
<td>1.0</td>
</tr>
<tr>
<td>Refractive index of the substrate</td>
<td>1.51</td>
<td>1.51</td>
</tr>
<tr>
<td>Fundamental guiding mode</td>
<td>$TE_0$</td>
<td>$TE_0$</td>
</tr>
<tr>
<td>Effective index of the fundamental mode</td>
<td>1.526</td>
<td>1.523</td>
</tr>
<tr>
<td>Pump wavelength (nm)</td>
<td>532</td>
<td>532</td>
</tr>
<tr>
<td>Bragg reflection order</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>Intersection angle (°)</td>
<td>57.0-76.0</td>
<td>25.5-29.8</td>
</tr>
<tr>
<td>Tuning range (nm)</td>
<td>824.9-943.1</td>
<td>809.1-932.4</td>
</tr>
<tr>
<td>Wavelength span (nm)</td>
<td>118.2</td>
<td>123.3</td>
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<tr>
<td>Linewidth (nm)</td>
<td>$&lt; 0.5$</td>
<td>$&lt; 0.5$</td>
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<tr>
<td>Lasing mode</td>
<td>$TE_0$ mode, s-polarized</td>
<td>$TE_0$ mode, s-polarized</td>
</tr>
</tbody>
</table>

Table 6.2 Performances of DFB planar waveguide lasers and channel waveguide lasers.
6.3 Summary

LDS dyes were doped into zirconia-ORMOSIL materials prepared by sol-gel technique. Embedded channel waveguides were fabricated using wet etching of glass substrates followed by deposition of the LDS 925-doped zirconia-ORMOSIL in the channels. NIR DFB laser action was generated in the channel waveguides. Narrow linewidth (<0.5 nm) tuning of the output wavelength was achieved by varying the period of the gain modulation generated by Nd:YAG laser at 532 nm. Tuning range was from 787 nm to 933 nm. The dispersion behavior of the laser output was checked by comparing experiments with the predictions of Marcatili’s theory. Additionally, NIR wide-band tuning and high-order DFB lasing operation were realized in LDS dye-doped planar waveguides.
Chapter VII

Summary

Sol-gel zirconia is a very useful optical material and an ideal host matrix for functional dopants. Organic dyes (viz. R6G, LDS series) doped zirconia and zirconia-ORMOSIL thin films were fabricated by a low-temperature sol-gel technique. Under different aging and drying conditions, the refractive index of zirconia and zirconia-ORMOSIL films varied from 1.53 to 1.64. The thickness was controlled from 0.6 to 1.4 μm by adjusting the viscosity of the initial solution and the spin speed. Good guiding performances of zirconia and zirconia-ORMOSIL films upon glass or fused silica substrates were revealed by a prism coupler system. Undoped zirconia films upon fused silica showed excellent transmission from 210 nm to the NIR. R6G- and LDS dye-doped zirconia and zirconia-ORMOSIL films have absorption peaks near 532 nm, allowing efficient pumping by frequency-doubled Nd:YAG laser.

The channel waveguides are rectangular dielectric strips embedded in other dielectrics of lower refractive index. Active layers can be easily built in the dielectric strips by sol-gel method to render the waveguides optically active. Compared with the planar waveguides, the channel waveguides have better lateral confinement. Using the standard photolithographic technique and the subsequent etching of glass substrates, we obtained wedge-shaped shallow channels with wide tops and narrow bottoms in glass substrates. Depending on the mask width and the etching time, the top and bottom widths varied from 11
μm to 70 μm and 11 μm to 58 μm, respectively; while the depth ranged from 0.86 μm to 14.21 μm, which was estimated by the etching time. Truly rectangular channels of widths varying from 10 μm to 5 μm and a fixed depth at 5 μm in fused silica substrates were produced by the inductive-coupled-plasma dry etching. The dye-doped sol-gel layer was then deposited on top of the substrates by spin coating. The sol-gel film outside of the channel was removed, leaving that inside to serve as the laser gain medium. Using this method, we successfully prepared R6G-doped zirconia channel waveguides and LDS 925-doped zirconia-ORMOSIL channel waveguides.

DFB lasers tunable in the visible were demonstrated in both R6G-doped zirconia planar waveguides and R6G-doped zirconia channel waveguides. DFB lasing emission was observed in R6G-doped zirconia waveguides when the pump energy was about 50 μJ per pulse. The net threshold pump energy was about 8 μJ per pulse, after accounting for reflection and transmission loss. Continuous tuning of the output wavelength was achieved by varying the period of gain modulation as formed by crossing two transversely focused pump beams. The tuning range was from 586 nm to 615 nm. Under the s-polarized pumping, the polarization of DFB output from zirconia films was s-polarized, which indicated that the DFB laser came from the TE guiding mode. DFB laser action was generated in R6G-doped channel waveguides in fused silica substrates when the pump laser pulse energy exceeded 20 μJ. Accounting for reflection and transmission loss, the actual energy deposited in the zirconia layer was about 1.6 μJ per pulse. Maintain the depth of the active layer at 3 μm, narrow linewidth lasing was achieved for rectangular channel
waveguides with widths of 5, 6.5 and 10 μm. Typical laser output energy was
10 nJ. Continuous tuning was achieved from 570 nm to 608 nm. The output
beam shape observed was roughly circular and can be fitted with Gaussian
profiles. We identified the output mode of the channel waveguide DFB laser as
the fundamental $E_{11}^0$ mode. The dispersion behavior of the laser output was
checked by comparing the experimental results with the predictions of
Marcatili's theory. The prediction was for a rectangular waveguide of a depth
of 3 μm. Good agreement was obtained between experiments and theory,
indicating the good performance of zirconia channel waveguides. The
linewidths of both the DFB zirconia planar waveguide laser and the DFB
zirconia channel waveguide laser were less than 0.5 nm, which was the
resolution limit of our detection system. Also, the laser output energy increased
linearly as the pump energy increased.

NIR DFB laser action was obtained in both LDS dye-doped
zirconia-ORMOSIL thin films and LDS 925-doped zirconia-ORMOSIL
channel waveguides. High-order DFB lasing operation was realized in LDS
925-doped planar waveguides by crossing the pump beams at the intersection
angles required by the Bragg condition. The tuning range was 825-943 nm,
809-932 nm and 809-881 nm for the first, second and third Bragg orders,
respectively. By adopting four types of LDS dye-doped zirconia-ORMOSIL
thin films (viz. LDS 759, LDS 798, LDS 867 and LDS 925), NIR wide-band
tuning of DFB waveguide lasers was demonstrated. Continuous tuning from
696 nm to 932 nm was achieved for the second Bragg order. Narrow linewidth
(<0.5 nm) DFB laser output tunable in the NIR was observed in the LDS...
925-doped zirconia-ORMOSIL channel waveguides when the pump energy exceeded 300 μJ. Accounting for the scattering and transmission loss, the net threshold was about 20 μJ. DFB lasing was achieved in channel waveguides with widths of 15.6 μm, 25 μm and 30 μm. The output wavelength for channels of various widths and a fixed depth of 1.8 μm at an intersection angle of 28.7° was predicted based on the Marcatili’s theory. The prediction was for $E_{11}^{v}$ lasing mode. Reasonable agreement between theory and experiments was observed.

In conclusion, dye-doped zirconia and zirconia-ORMOSIL channel waveguides were fabricated using wet or dry etching of glass and fused silica substrates followed by sol-gel deposition of the dye-doped zirconia and zirconia-ORMOSIL in the channels. DFB laser action tunable from visible to the NIR was realized in the channel waveguides. Corresponding laser properties were characterized.

Futures works to be carried out for the DFB sol-gel lasers are as follows:

1. Investigate the polarization properties of the dye-doped sol-gel channel waveguide lasers. This work is significant to the control of the output polarization of DFB waveguide lasers.

2. Realize DFB laser action in other spectral regions, such as IR and UV. Especially, IR DFB waveguide lasers at 1.3 μm are very useful in optical communications. This work can be carried out by doping IR dyes, such as IR 26, IR1051 and PY-1, into sol-gel waveguides.

3. In order to improve the lifetime of dye-doped sol-gel glass waveguide
lasers, we can introduce the two-photon-pumped (TPP) dyes into the host matrix as the functional dopants. Under low energy photon pumping, the lifetime of DFB waveguide lasers should be improved enormously.

4. Demonstrate surface-emitting DFB lasers based on 2-dimensional permanent surface relief gratings (SRG) in sol-gel waveguides. Due to the confinements in two orthogonal directions, the output beam of surface-emitting DFB lasers has much lower divergence angle, which makes the DFB lasers more practical [90].

5. Electro-tunable DFB solid-state lasers could be realized by introducing laser dyes and nematic liquid crystal (LC) into sol-gel thin films. With an applied electric field on the thin film, continuous tuning of the lasing wavelength could be achieved due to the variation of the effective refractive index of the film caused by the field-induced reorientation of LC molecules [33].
References


[49] Eliezer and M. Rabinovich, Sol Gel Processing – General Principles


References


