FRONTIER DEVELOPMENTS IN OPTICS AND SPECTROSCOPY

Majorana Center: View from the San Domenico Lecture Hall

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Baldassare Di Bartolo
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FRONTIER DEVELOPMENTS IN OPTICS AND SPECTROSCOPY

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Felix qui potuit rerum cognoscere causas

Virgil,Georgics

PREFACE

This book is based on the Proceedings of the Institute “Frontier Developments in Optics and Spectroscopy,” held in Erice, Sicily, Italy, from the 17th of June to the 2nd of July 2007. The meeting was organized by the International School of Atomic and Molecular Spectroscopy of the “Ettore Majorana” Center for Scientific Culture.

Other Institutes organized by this School are listed on pp. vi-vii

There has been a consensus to put this book online to give it the maximum possible diffusion and to have it reach the people who could not attend the meeting.

The purpose of the Institute was to present the new developments in the field of spectroscopy including the realm of new phenomena and the new techniques that allow their exploration. New techniques open the possibility of probing into new and unexplored areas. The discovery of new effects spurs the growth of new and more refined techniques which may in turn be used to discover new phenomena, and so on, in a sort of inexorable and never ending spiral “technique-phenomenon-new technique-new phenomenon-new technique, etc.” This process has accelerated in recent times, leaving people entering the field of spectroscopy - as well as people in the field - with a serious difficulty of grasping the magnitude and variety of the new developments.

The Institute considered seriously this problem and presented in a pedagogical fashion the new aspects, both theoretical and experimental, of the spectroscopic optical research. Recent developments in laser technology and optical technology in general, coupled with rapid advancements in materials research, synthesis, and fabrication (e.g. nanomaterials and photonic metamaterials) have opened the door to completely new regimes of optical spectroscopy that the course explored. New developments in the fields of laser technology, semiconductors, luminescence and quantum entanglement were also reported.

The challenge in organizing such a meeting was to create a coherent framework in which the various techniques could be viewed. For this reason we dealt mainly with the physical principles on which the techniques are based, avoiding purely descriptive presentations, in accordance with the nature of a "school", whose impact has to be not only timely, but also lasting. The meeting was meant to be in the best tradition of our past Institutes because in it we tried to start from the consideration of fundamental principles, to reach the frontier of research in a systematic and didactic fashion.

The Institute provided the participants with an opportunity to present their research work in the form of short seminars or posters. The participants came from 10 different countries: France, Estonia, Germany, Ireland, Italy, Japan, The Netherlands, Spain, Turkey, and United States.

I wish to acknowledge the sponsorship of the meeting by NASA, the ENEA Organization, Boston College, the Italian Ministry of University and Scientific Research and Technology, and the Sicilian Regional Government.

I would like to thank the members of the organizing committee (Prof. Steve Arnold, Dr. Giuseppe Baldacchini, Dr. Norman Barnes, Prof. John Collins, Prof. Claus Klingshirn, Prof. Eric Mazur, Prof. Ralph von Baltz, and Prof. Martin Wegener) for their advise and support and the secretary of the course, Mr. Ottavio Forte, for his great help in organizing and running the course.

I am looking forward to our activities at the Majorana Centre in years to come, including the next 2009 meeting of the International School of Atomic and Molecular Spectroscopy.

Baldassare (Rino) Di Bartolo
Director of the International School of
Atomic and Molecular Spectroscopy of
the “Ettore Majorana” Center

LIST OF PAST INSTITUTES
of the
International School of Atomic and Molecular Spectroscopy

All the Institutes were held at the “Ettore Majorana” Centre in Erice, Sicily, Italy:

1974 – Optical Properties of Ions in Solids

1975 – The Spectroscopy of the Excited State

1977 – Luminescence of Inorganic Solids

1979 – Radiationless Processes

1981 – Collective Excitations in Solids

1983 –Energy Transfer Processes in Condensed Matter

1985 – Spectroscopy of Solid-State Laser Type Materials

1987 – Disordered Solids: Structures and Processes

1989 – Advances in Nonradiative Processes

1991 – Optical Properties of Excited State in Solids

1993 – Nonlinear Spectroscopy of Solids: Advances and Applications

1995 – Spectroscopy and Dynamics of Collective Excitations in Solids

1996 – Workshop on Luminescence Spectroscopy

1997 – Ultrafast Dynamics of Quantum Systems:

Physical Processes and Spectroscopic Techniques

1998 – Workshop on Advances in Solid State in Luminescence Spectroscopy

1999 – Advances in Energy Transfer Processes

2000 – Workshop on Advanced Topics in Luminescence Spectroscopy

2001 - Spectroscopy of Systems with Spatially Confined Structures

2002- Workshop on The Status and Prospects of Luminescence Research

2003- Frontiers of Optical Spectroscopy:
Investigating Extreme Physical Conditions with Advanced Optical Techniques

2004- Workshop on Advances in Luminescence Research

2005- New Developments in Optics and Related Fields:Modern Techniques,
Materials and Applications

2006-Workshop on Advances in the Study of Luminescent Materials

2007- Frontier Developments in Optics and Spectroscopy

THE PARTICIPANTS

THE DIRECTOR WITH HIS ASSISTANTS

DIELECTRIC WAVEGUIDE LASERS

Optical Gain in $KY(WO_4)_2:Yb^{3+}$ and α-$Al_2O_3:Ti^{3+}$ Waveguides

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1. Introduction

In recent years, the field of integrated optics has made remarkable progress. Optical gain and high output power can be obtained in rare-earth or transition-metal ion doped dielectric materials. These robust materials also offer high optical damage thresholds. As miniaturized waveguide lasers and amplifiers, these systems will find applications whenever there is a need for on-chip optical gain in combination with high optical power, wavelength diversity, ultra-short pulses, or second- and third-order optical nonlinearities which can be exploited in order to integrate various optical functions on a chip.

This chapter reviews our and our colleagues' recent results concerning planar waveguide lasers in $KY(WO_4)_2:Yb^{3+}$ (hereafter abbreviated as KYW:Yb) near 1020 nm [1] and channel waveguide lasers in α-$Al_2O_3:Ti^{3+}$ (hereafter abbreviated as Ti:sapphire) near 800 nm [2].

2. KYW planar waveguide lasers

Potassium yttrium double tungstate, KYW, is a strongly anisotropic biaxial crystal, which crystallizes in the monoclinic structure with space group C2/c [3, 4, 5]. It exhibits one of the largest absorption and emission cross-sections and broader linewidths when doped with rare-earth ions, which may be due partly to the high refractive indices and partly to the strong anisotropy [6, 7]. Another important advantage of rare-earth-ion-doped KYW is the relatively large ion separation allowing highest doping levels with minimum quenching effects.

Among the rare-earth-ion-doped solid-state materials, Yb^{3+}-doped crystals are particularly well suited as gain media in high-power lasers, femtosecond lasers, and amplifiers [8]. Yb^{3+} is a promising activating ion which possesses a number of advantages over Nd^{3+} for laser operation in the 1-µm spectral region [9]. These advantages are related to the very simple energy-level scheme constituted of only two levels: the $^2F_{7/2}$ ground state and the $^2F_{5/2}$ excited state. Effects such as excited-state absorption, cross relaxation, and energy-transfer upconversion, which can lead to reduced laser efficiency through alternative paths for depopulation of the upper laser level, are absent. The Yb^{3+} ion also has a small quantum defect as a result of the close pump and laser wavelengths, leading to low thermal load. The most intense Yb^{3+} absorption line near 980 nm is suitable for pumping with InGaAs laser diodes. The thermal conductivity of the double tungstates of approximately 3.3 $Wm^{-1}K^{-1}$ (averaged over the polarizations) is about three times lower than in YAG but four times that of a typical phosphate laser glass [10]. KYW:Yb has been recognized as a laser material with a high potential for efficient tunable continuous-wave (CW) and mode-locked operation in diode-pumped arrangements [6, 7, 10, 11, 12].

In general, waveguide structures exhibit high confinement and excellent mode overlap of pump and laser beams. This feature is of particular advantage for three-level transitions such as the 1-μm transition in Yb^{3+}. The monoclinic double tungstates possess high refractive indices, resulting in high refractive-index contrasts in structured devices, which makes rare-earth-ion-activated monoclinic double tungstates potentially useful for applications in integrated optics.

A method of liquid-phase epitaxy (LPE) with vertical substrate dipping has been developed to produce rare-earth-ion-doped KYW thin planar layers. LPE is a well-known technique for producing oxide films for laser applications, in which a crystalline layer can be grown from a molten solution onto an oriented single-crystal substrate [13]. The major advantage of LPE compared to epitaxial techniques from the vapor phase is that LPE is a near-thermodynamic equilibrium process, and therefore, high-quality single-crystalline KYW layers are feasible [14].

Figure 1 shows schematically an experimental LPE set-up, based on a non-vacuum resistance-heated furnace with vertical loading. Inside the furnace, there is a crucible filled with a molten solution of KYW in an appropriate solvent. Initially, we tested the low-temperature chloride mixture NaCl-KCl-CsCl as the solvent [15]. However, 3D island nucleation generated insertion defects, which limited the maximum layer thickness to approximately 10 μm and led to non-optimum interface quality. The tungstate solvent $K_2W_2O_7$, which we employed successfully in the present work, can offer larger thickness and good layer quality. The $K_2W_2O_7$ solvent contains no impurity ions and exhibits a high solubility for double tungstates [16]. Planar KYW layers with thickness d = 5 to 100 μm and a surface area of ~0.5 cm^2 can be grown at an average growth rate of 18 μm/h [17]. When adding a small amount of Yb_2O_3 powder to the initial solution, Yb^{3+}-doped KYW layers can be grown, where the Yb^{3+} ion substitutes for Y^{3+}. The incorporation coefficient of Yb^{3+} ions into the KYW matrix during growth from the $K_2W_2O_7$ solvent is close to unity [18]. Thus, the growth of KYW:Yb layers with tailored concentration of Yb^{3+} ions is possible, knowing the initial ratio of Yb^{3+}/Y^{3+} in the molten solution. Typical Yb^{3+} concentrations used in the present studies were well below the critical ones and varied from 1.2 to 2.4at.%. Higher dopant concentration can be achieved by using co-doping with two different rare-earth ions, e.g. Lu^{3+} and Gd^{3+}, which have opposite ionic-radius misfit with Y^{3+}. Thus, crack-free layers containing 25.3at.% Lu^{3+}, 13at.% Gd^{3+}, and 1.7at.% Yb^{3+} could be grown [19]. Since Lu^{3+} and Yb^{3+} possess similar ionic radii, rather high doping concentrations of the optically active Yb^{3+} ion can be obtained by replacing Lu^{3+} with Yb^{3+}.

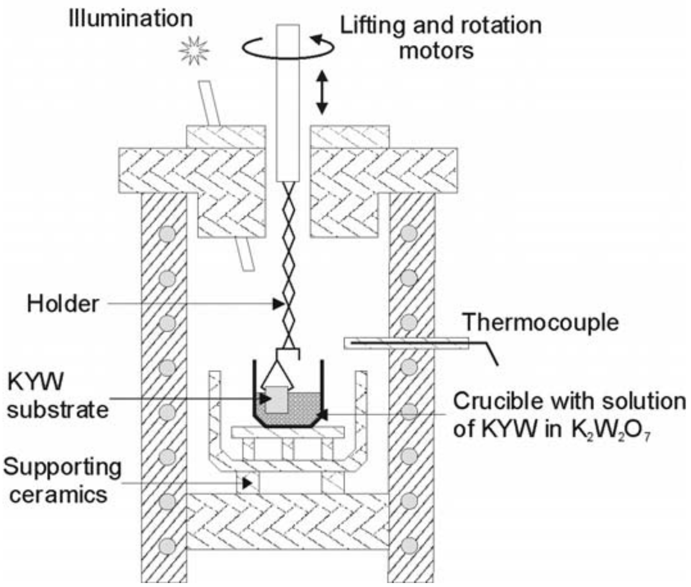

Figure 1. LPE set-up for the growth of KYW:RE^{3+} waveguides. (Figure taken from [1]).

Since KYW is isostructural to KREW for RE = Sm…Lu (100% of Y^{3+} is substituted by RE^{3+}) [4, 20], one can assume that the refractive indices of KYW:RE^{3+} layers increase linearly with increasing RE^{3+} concentration. Thus, the refractive-index change of a 1.8at.% Yb^{3+}-doped layer with respect to the undoped substrate is expected to be 6×10^{-4}, which was confirmed experimentally by dark m-line spectroscopy. The refractive-index change can be significantly larger in Lu^{3+} and Gd^{3+} co-doped KYW:Yb layers, where high concentrations of optically inert Lu^{3+} and Gd^{3+} dopants are incorporated to increase the refractive index change up to 7.5×10^{-3} [19].

Both endfaces of the waveguides were polished to laser-grade quality. Figure 2a shows the optical image of the polished endface for a KYW:Yb surface layer. The interface is sharp and straight without any detectable defects. The layers were tested as active and passive planar waveguides under laser excitation at 981 nm (InGaAs diode laser), 632.8 nm (He-Ne laser), or 488 nm (Ar-ion laser). The pump light was coupled into the active layer by focusing with a microscope objective. The propagated light was imaged onto the sensor of a CCD camera with another microscope objective. For the 11-µm-thick waveguide shown in Fig. 2b, the emitted Yb^{3+} fluorescence was guided together with the 981-nm pump light in the surface KYW:Yb layer. The vertical intensity profile of the outcoupled light is close to a Gaussian distribution, since only one TE mode at λ = 980 nm can be supported by the 11-µm-thick planar waveguide in vertical direction. Propagation losses of the optical waveguides were found to be only 0.1-0.2 dB/cm at 981 nm [17].

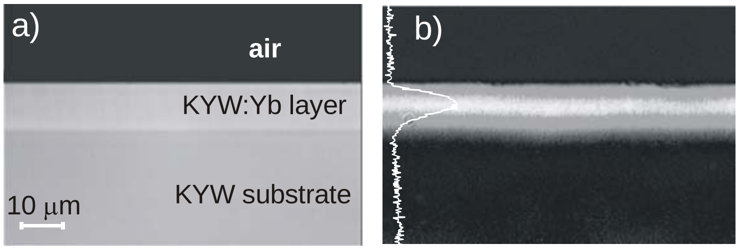

Figure 2. (a) Optical photograph of the endface of a 1.2at.% Yb^{3+}-doped KYW surface layer; (b) near-field image of the guided pump light and Yb^{3+} fluorescence outcoupled from the waveguide. (Figure taken from [1]).

For the waveguide laser investigations, 6-mm-long KYW:Yb surface and buried planar waveguides were prepared. The waveguide was positioned between the two folding mirrors of an open Z-type resonator (inset of Fig. 3). The waveguide was positioned at Brewster angle to minimize the loss in the laser cavity. The waveguide orientation corresponded to propagation approximately along the N_g principal optical axis and polarization parallel to the N_m axis. The pump polarization was parallel to the N_m axis. Due to the chosen thickness between 17 and 35 μm, each waveguide supported propagation of a relatively high number of transverse modes in its guiding direction. Nevertheless, the observed far-field laser intensity distributions indicate that the laser output was close to the diffraction limit in almost all samples and at all output powers [17].

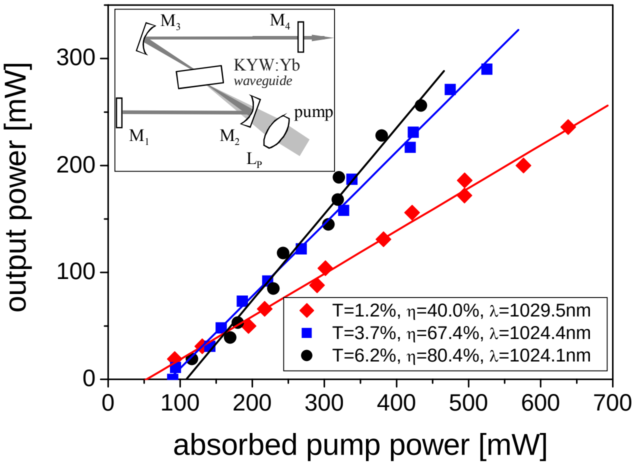

Figure 3. Laser output power versus absorbed pump power of 17-μm thick surface KYW:Yb planar waveguides for different transmissions of the output coupler. Inset: setup of the Z-shaped laser cavity: L_p – focusing pump lens, M_1, M_2, M_3 – high reflecting mirrors (M_2, M_3; ROC = -10 cm), M_4 – plane output coupler. (Figure taken from [17]).

The KYW:Yb layers were pumped by a CW Ti:sapphire laser operating at 980.5 nm. Stable CW oscillation near 1025 nm was achieved for all waveguides investigated [17]. The best laser performance was achieved with the 17-μm-thick surface waveguide doped with 1.2at.% Yb^{3+}. With an output coupling of 3.7% the laser threshold was only 80 mW of absorbed pump power and the maximum output power amounted to 290 mW, resulting in a slope efficiency versus absorbed pump power of 67.4%. A maximum slope efficiency of 80.4% was obtained for an output coupling of 6.2%, corresponding to a pump efficiency of 58.9% (Fig. 3).

Also KYW:Tm planar waveguide lasers near 2 μm have been demonstrated in a similar way as described above [21]. Other methods that have been used to fabricate KYW:Yb waveguides include ion-beam implantation [22] and femtosecond laser writing [23].

3. Ti:sapphire channel waveguide lasers

Transition-metal-ion-doped materials exhibiting $3d \leftrightarrow 3d$ electronic transitions are useful as broadly tunable laser sources and for the generation of ultrashort pulses, due to strong homogeneous broadening of the gain bandwidth by interaction with lattice vibrations. Ti:sapphire [24] allows continuous-wave tunability from ~670-1100 nm and the generation of pulses as short as ~5 fs [25]. Although laser thresholds as low as ~100 mW have been demonstrated [26], in general thresholds are much higher. This has prompted work on creating Ti:sapphire waveguides in order to lower the laser threshold. In addition, the waveguide geometry offers the opportunity of integrating devices with the laser for tuning or mode-locking for a high-repetition rate, compact femtosecond-laser source [27], and of integrating with other waveguide circuits, e.g. for the purposes of a sensing device that utilizes the large Ti:sapphire bandwidth.

Ion beam implantation has been used for the fabrication of waveguide laser structures in insulators. The first reported ion-implanted waveguide laser was a planar device produced in Nd:YAG [28]. For light ions, electronic damage is comparatively low; at lower energies near the end of the ions' track nuclear collisions predominate, leading to the displacement of lattice ions and resulting typically in a large cascade of damages. As a consequence, a (partial) amorphization of the material appears at the end of the ions' track, with a positive or negative refractive-index change depending on the ion fluence, the implanted material, and the nature of the interactions.

Buried channel waveguides have been produced by proton implantation in sapphire [29] and Ti:sapphire bulk crystals [30]. Proton irradiation in sapphire or Ti:sapphire results in a negative refractive-index change of the implanted zone. Therefore, buried channel waveguides have been fabricated by implanting regions of low refractive index surrounding the intended channel waveguide region. Implantations were performed with high energy protons of 0.5-1 MeV and doses between 2×10^{16} and 4×10^{16} H^+/cm^2. Vertical confinement was provided with a lower and upper barrier resulting from uniform irradiations of the whole surface of the bulk sample at different energies in order to produce a buried planar waveguide. Within the planar waveguide, horizontal confinement was obtained by successive irradiations through a slit on both sides of the intended channel. A better lateral confinement can be achieved by a vertically more uniform distribution of the refractive-index change within the side barriers. Therefore, irradiations with several angles of incidence were performed, resulting in vertically stacked low-refractive-index barriers. Typical examples of waveguides produced in undoped sapphire are shown in Fig. 4. In Ti:sapphire, several 5-µm-deep buried channel waveguides were fabricated with widths of 10, 15, and 25 µm [30].

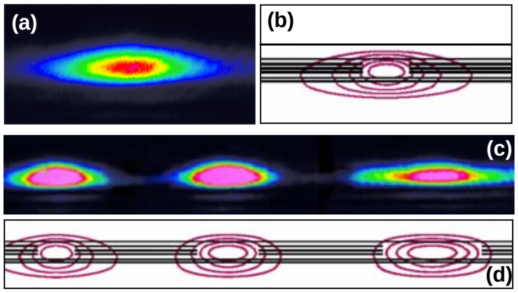

Figure 4. Mode patterns of 780-nm fundamental-mode laser light end-coupled to sapphire buried channel waveguides. (a) Experimental output profile recorded from a buried channel waveguide and (b) corresponding simulated contour plot. (c) Experimental output profile recorded from three parallel buried channel waveguides with different widths of 6, 10, and 15 µm and (d) corresponding simulated contour plot. (Figure taken from [29]).

Refractive-index changes of the planar waveguide parts were characterized by m-lines spectroscopy [31]. This technique is based on the measurement of the effective refractive index of propagating modes excited by prism coupling of light. Only negative refractive-index changes were observed in proton-irradiated sapphire, with gradient shape and maximum refractive-index decrease of typically 0.5-1%, for irradiation with 1-MeV protons at a fluence of 10^{16} H^+/cm^2. The damaged barriers were localized ~8-µm deep for 1-MeV protons, which is in good accordance with Monte-Carlo calculations of the probability of a target atom being displaced from its lattice site (TRIM calculations). Proton implantation induces well-controlled refractive-index decreases, hence the guided regions are formed by writing low-refractive-index barriers around the intended guided regions and their mode profiles can be easily adjusted by changing the implantation parameters and geometries. By choosing different channel dimensions and optical confinement in the horizontal and vertical directions, we produced elliptical fundamental-mode profiles, with larger modal fields in the horizontal direction (Fig. 4). Also cylindrical mode shapes can be achieved by adjusting the implantation energies, doses, and geometries. When measured with the self-pumped phase-conjugation [32] method, buried channel waveguides with cross-sections of 15 µm × 5 µm and 10 µm × 5 µm showed losses of 1.0 and 1.3 dB/cm, respectively, at 720 nm. The higher loss level of the narrower guides was due to the stronger interaction of the modal field with the optical barriers.

Lasing experiments [30] with the Ti:sapphire buried channel waveguides were performed at room temperature with an Ar^+ pump laser operating on all lines. The laser resonator was formed by attaching at the end-faces two thin dielectric mirrors with high reflectivity and transmission at the lasing and pump wavelength, respectively. CW laser action was obtained in channel waveguides with a height of 5 μm and widths of 10 and 15 μm at absorbed pump power thresholds of 230 and 260 mW, respectively. Lasing was observed near 780 nm and the output was π-polarized regardless of the polarization state of the pump beam. Measurements of the M^2 factors confirmed the near-diffraction-limited nature of the laser emission from the 10-μm-wide channels with values of $M_x^2 = 1.5$ and $M_y^2 = 1.2$ for the horizontal and perpendicular directions, respectively. By replacing the HR output mirror with one having a transmission of 4.6% at the lasing wavelength the threshold values increased to 260 and 290 mW for the 10- and 15-μm-wide channels, respectively. The laser input-output characteristics obtained with the 4.6% output coupler are shown in Fig. 5. Slope efficiency values of 3% and 2.2% were obtained from the 15- and 10-μm-wide channels, respectively. The corresponding output powers are 17.5 and 12.4 mW for 1 W of absorbed power.

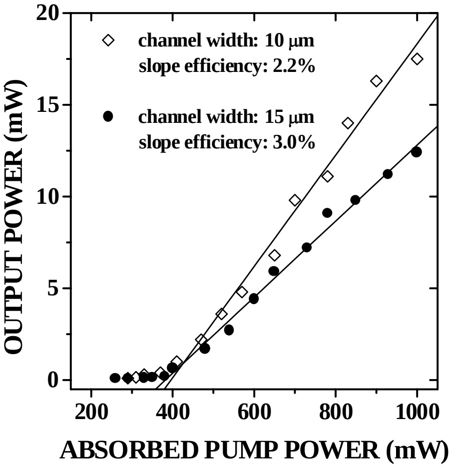

Figure 5. Dependence of the output power from two Ti:sapphire waveguide lasers with cross-sections of 5 μm x 15 μm and 5 μm x 10 μm, respectively, as a function of the absorbed power for an output coupler with a transmission of 4.6%. (Figure taken from [30]).

Other methods that have been used to fabricate Ti:sapphire waveguides include ion beam implantation and subsequent wet chemical etching [33], direct laser ablation [34], femtosecond laser writing [35], ion in-diffusion [36], and pulsed laser deposition [37] in combination with chlorine reactive ion etching [38] or Ar ion milling [39]. The latter methods have lead to the demonstration of broadband luminescent emitters [40], which show potential as light sources in optical coherence tomography [41], and planar [42], rib channel [43], and in-diffused channel [44] waveguide lasers.

4. Conclusions

Only recently, the significant potential of optical waveguide lasers in doped dielectric crystalline materials has surfaced. Planar fabrication methods such as liquid phase epitaxy or pulsed laser deposition followed reactive ion etching or Ar-ion milling, or direct methods to change the refractive index in bulk samples such as light-ion implantation, ion in-diffusion, or femtosecond laser writing have been explored successfully and surface and buried, planar and channel waveguide lasers have been demonstrated in KYW and Ti:sapphire for the first time. Since the propagation losses of these first-generation waveguides are still rather high, substantial improvement is required in order to obtain ultra-low-threshold waveguide lasers and to explore their full potential as light sources in various applications in integrated optics.

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