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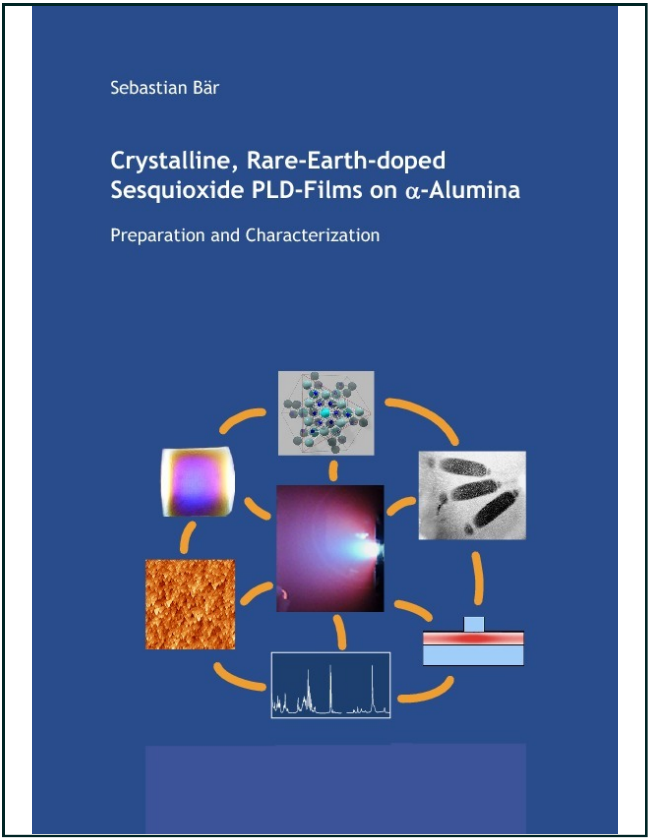
Crystalline Rare-Earth-doped Sesquioxide PLD-Films on alpha-Aluminia



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Preparation and Characterization

1 Introduction

1.1 Motivation

In recent years the data transfer created by computer-based business processes and internet applications has been growing exponentially. This development requires an increasing transmission capacity at lower cost, which can only be met by increased use of optical fibre and associated advanced photonic technologies. The operation of these new devices must be described in terms of optics as well as of electronics, giving birth to a mixed discipline called *(integrated) photonics*. The basic idea behind integrated photonics is the use of photons instead of electrons, creating integrated optical circuits similar to those in conventional electronics, i. e. the fabrication and integration of several photonic components on a common planar substrate.

The replacement of electronic by photonic devices is forced by fundamental physical reasons that limit the information transmission rate using purely electronic devices: as the frequency of an electrical signal propagating through a conductor increases, the impedance of the conductor also increases, thus the propagation characteristics of the electrical interconnection become less favorable. In contrast, optical signals propagate through non-conducting dielectric media, operating in the wavelength range where these materials are highly transparent. In general, this transparent window falls in the visible and near-infrared range of the electromagnetic spectrum, which corresponds to light frequencies around 300 THz, 10^6 times the frequency used in electrical transmission, which allows a very large bandwidth for transporting a huge amount of information.

The optical elements present in integrated photonic devices should include basic components for generating, focusing, splitting, junction, coupling, isolating, polarization controlling, switching, modulating, filtering and detecting of light. The key element of these devices are optical waveguides, that provide not only guiding, but also coupling, switching, splitting, multiplexing, and demultiplexing of optical signals.

Although nowadays a long list of integrated photonic devices has been proposed, modelled, and fabricated, and their number is quickly increasing, the basic components remain almost unchanged. All the optical components in integrated photonics are constructed from three building blocks. These are the straight waveguide, the bend waveguide and the power splitter (Y-branch). Using these building blocks, several basic components have been developed to perform basic optical functions. A good overview on the specific elements is given in [Lif03].

The basic technology for these waveguides is the fabrication of high quality thin films. The methods most widely used for the production of such thin films are diffusion techniques (e. g. titanium indiffusion in lithium niobate) and deposition techniques (such as chemical vapor deposition used for silica). The integration of multiple functions within a planar optical circuit requires structures with lateral dimensions of a few microns, which can be achieved by photolithographic processes [Hai01]. For the fabrication of functional devices different materials are used. Apart from semiconductors such as indium phosphide (InP), gallium arsenide (GaAs) – an example is depicted in figure 1.1 or even silicon (Si), also dielectrics such as polymers, glasses, silica on silicon (SiO_2/Si), or lithium niobate ($LiNbO_3$) are suitable for the use in integrated photonics.

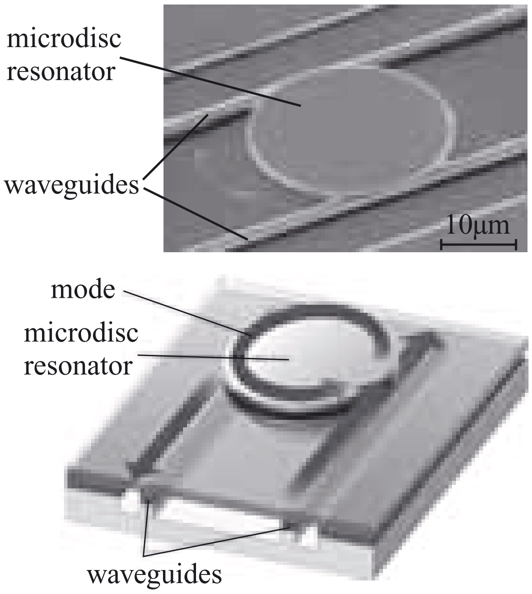

Figure 1.1: *InGasP ring resonator* [Tis99].

Photonic Crystals

Aside from this 'classical' way of realizing the necessary components for integrated optics, a new class of materials, termed *photonic crystals*, with optical properties that can be designed individually has been developed. In a photonic crystal, the periodic arrangement of refractive index variation controls how photons are able to move through the crystal. Similar to the periodic arrangement of ions in a lattice, which gives rise to the energy band structure e. g. in semiconductors, the refractive index contrast results in a photonic band structure characterized by a photonic band gap. Breaking the periodicity introduces new energy levels within the photonic band gap. This defect mode or microcavity formed by breaking the periodicity of the crystal amplifies only those wavelengths of light that are able to pass freely through the crystal. Photonic crystal microcavities are more efficient than conventional semiconductor diode lasers since there are few directions in the which the photons can escape. Two examples of photonic crystal structures are shown in below.

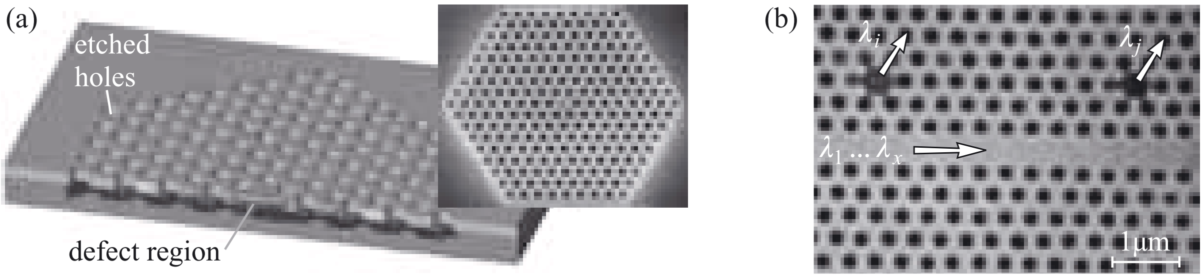

Figure 1.2: *Photonic crystal structures: (a) microcavity formed by a 'point defect'* [Vah03], *(b) Add-drop filter* [Nod00]

Dielectric Oxides

The materials used for integrated optics are mainly compounds of semiconductors as they are the most promising materials for monolithic integration of the components.This means that the highest level of integration (whether serial or parallel) can be achieved, where all the optical elements including light sources, light control, electronics, and detectors are incorporated in a single substrate. In certain cases, the hybrid integration technology might be favorable. Hybrid technology means that different elements are built on different substrates and then directly attached to each other or interconnected by optical fibres.

Compared with the highly developed standard fabricating and structuring techniques for semiconductor devices, the techniques for structuring crystalline oxide dielectrics are not well known, even if the processes are basically the same. Since this class of materials offers a range of advantageous properties, however, the aim of this thesis is the preparation and characterization of thin sesquioxide films, i. e. yttria Y_2O_3, lutetia Lu_2O_3, and scandia Sc_2O_3.

An advantage of the dielectric oxides is their wide transparency range from the ultraviolet to the mid-infrared part of the electromagnetic spectrum. Due to the possibility of doping the oxides with rare-earth-ions, the optical properties can be influenced selectively. Compared with semiconductors, the refractive index of these oxides is moderate resulting in considerably reduced fresnel reflections at the endfaces designed for coupling light in and out of the device. This is an important point since these reflections determine the efficiency of the system. For example, at normal incidence of near-infrared light, the index difference between semiconductors and air is of the order of $\Delta n \approx 2.5$ resulting in a reflectivity of $\approx 30\%$. Under the same conditions, the reflectivity of dielectric oxides is less than 10% because the index difference is smaller ($\Delta n < 1$).

The growing interest in the fabrication and characterization of Y_2O_3 films over the last few years is related to the diverse range of potential applications that can be envisioned. Y_2O_3 is an important material for semiconductor applications as well as for optical applications such as phosphors or waveguide lasers because of its ability to be a host material for rare-earth atoms like europium or thulium [Pol97, Jon97, Hui00]. Especially in the semiconductor industry, the use of Y_2O_3 in metal–insulator–semiconductor (MIS) diodes, transistor gates, metal–oxide–semiconductor (MOS) capacitors, and dynamic random access memory (DRAM) gate dielectrics is favorable due to its large bandgap (5.8 eV) and large dielectric constant (14-18) [Jon97, Ras92, Zha98].

Eu-doped Y_2O_3 is a well-known red phosphor [Jon97] and still used – as a powder – in electroluminescent displays (ELDs), fluorescent lamps, and cathodoluminescent (CL) displays. However, thin film phosphors have several advantages over powders, such as higher lateral resolution from smaller grains, better thermal stability, reduced outgassing, and better adhesion to the solid surface [Hir97]. Seeing these advantages, there is a significant interest in the development of $Eu{:}Y_2O_3$ thin film phosphors for the use in field emission display (FED) technology replacing the $Eu{:}Y_2O_2S$ system which is used in the traditional cathode ray tube (CRT).

Resulting from its high melting point of 2410°C, Y_2O_3 is also widely used for high temperature corrosion protection. In combination with the efficient transmission of infrared (IR) radiation, this robustness makes yttria films a very promising material for protective and antireflective coatings for IR detectors, e. g. HgTe and HgCdTe photovoltaic IR detectors.

Although Y_2O_3 has a lot of beneficial properties as shown above, lutetia and scandia offer additional positive properties for thin film production. The lattice constants of Lu_2O_3 and Sc_2O_3 are 10.39 Å and 9.86 Å, respectively, and thus, these values match better the lattice constant of sapphire, a common substrate for yttria, leading to the production of films with fewer dislocations. Additionally, both materials have a higher refractive index than Y_2O_3, which might be interesting for waveguide applications and for the fabrication of photonic crystals, where a high index contrast is advantageous.

Up to now, however, only little attention has been paid to Lu_2O_3 and Sc_2O_3. Scandia, for example, might be suitable for reducing surface states on AlGaN/GaN high electron mobility transistors (HEMT) and, in addition, exhibit promise as gate dielectrics in GaN [Luo02]. Since Lu_2O_3 has the largest bandgap among the rare-earth oxides, it is also considered for high-dielectric constant (high-k) gate insulator applications [Ohm02]. Additionally, Lu_2O_3 can be used as an efficient scintillator material because of its high absorption coefficient for ionizing radiation. Because of the high Z-number an incoming gamma or X-ray particle can be preferentially stopped at a single position. Therefore, higher resolution images in medical diagnosis can be envisioned [Zyc02].

Another application for rare-earth-doped sesquioxide films is the fabrication of planar waveguide structures. These include passive elements as well as active waveguides (amplifiers and lasers), because these materials are well known bulk hosts for laser ions (e. g. Nd:Y_2O_3, Yb:Y_2O_3) [For99, For00]. One of the first steps towards these devices is to be able to produce planar optical waveguides in very good quality. There is no question about the predominant role of optical fibres in long distance data transmission, but for active processes crystalline optical waveguides offer substantial advantages: The larger emission and absorption cross sections available in crystalline matrices become accessible, and the confinement of light inside the waveguide generates a larger intensity-length product. Since nonlinear processes, like upconversion or frequency conversion, depend strongly on the intensity, the realization of these processes is much easier to achieve in waveguides. Additionally, the guiding of the pump mode as well as the signal mode leads to an excellent overlap of the modes resulting in lower laser thresholds.

Thus, lasing can be achieved on transitions that possess an impractically high lasing threshold in bulk materials [Han93]. Apart from waveguides of Nd:YAG [Bon00], Nd:GGG [And98, Bar01], and Ti:Al_2O_3 [And98], also first yttria waveguides have been fabricated, for example Er:Y_2O_3 [Hoe92, Kor01, Lec02].

Why Pulsed Laser Deposition?

There are many technologies existing for the fabrication of optical thin films and apart from the pulsed laser deposition technique, a variety of other physical and chemical methods have been used in the past to synthesize dielectric Y_2O_3 films, e. g. chemical vapor deposition (CVD) [Sha93, McK00], RF magnetron sputtering [Gur87], ion-assisted evaporation [Cho99], anodization [Gol68], electron-beam evaporation (EBV) [Fuk89], sol-gel techniques [Rao96, Lou01], and reactive synthesis [And94].

In recent years, however, pulsed laser deposition (PLD) has proven to be a very attractive and practical method not only for the growth of high-quality doped [Cho98, Gao99] and undoped [Zha98] Y_2O_3 films (pure or doped Y_2O_3 sintered targets were used) but also for a variety of other materials. There are many reasons why this method has become that attractive for the fabrication of thin films, namely the possibility

- to generate films that match the stoichiometry of the target due to an extremely high heating rate of the target (10^8 K/s) leading to congruent evaporation (whereas conventional thermal evaporation or sputtering can lead to non-stoichiometric deposits).
- to use small targets (in contrast to the large targets required for sputtering)
- to deposit in reactive gas environments (in contrast to conventional evaporation, where hot filaments and/or crucibles could be oxidized).
- to benefit from high energies of the plasma species (often 5 to $\geq$100 eV) that can have beneficial effects on the film properties, e.g. good film adhesion at low substrate temperatures
- to benefit from a high ionization degree of the plasma flux (up to 80%), positively influencing the film properties
- to accurately control of the film thickness by controlling the number of laser pulses

A more detailed description of the PLD process will be given in chapter 3. Apart from sintered targets of Y_2O_3, there have also been attempts to use a pure metal ablation target [Kor00]. In combination with alternate-target PLD, it is possible to have a precise control of dopant concentration levels and in-depth separation [Ser99].

For crystalline film growth, an appropriate substrate material is very important, because epitaxial film growth depends strongly on the lattice matching between film and substrate. Besides crystallinity, the desired application determines the substrate. In fact, thin films of yttria have been grown on various substrates, e. g. Si, SiO_2, MgO, Al_2O_3, $LaAlO_3$ and yttrium-stabilized zirconia (YSZ) [Cho98, Cho00, Dik03, Fuk89, Gao99, Gab02]. Generally, these films were highly textured along the $\langle 111 \rangle$ or $\langle 100 \rangle$ crystallographic axis.

Nanostructures

The capability of PLD to produce 'films' with thicknesses smaller than a monolayer makes this technique interesting for structures in the sub-nanometer range, which is like "playing LEGO at the atomic scale" [Bla04].