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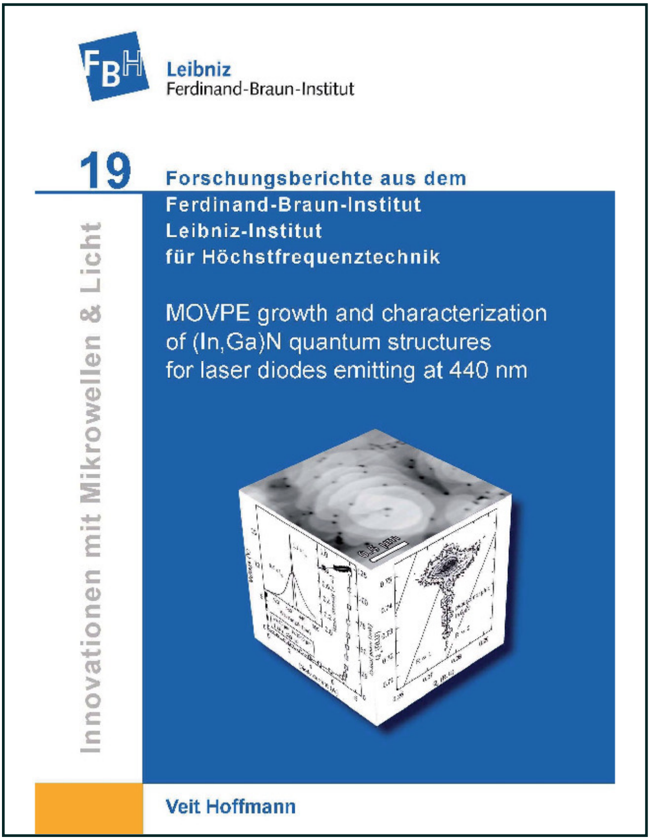
# MOVPE growth and characterization of (In,Ga)N quantum structures for laser diodes emitting at 440 nm

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# 1 Introduction

## 1.1 Motivation

Compound semiconductor opto-electronic devices are an inherent part of many everyday objects, e.g. light emitting diodes (LED)s, and modern technologies such as laser diodes (LD)s based fiber-optic communication. Due to the narrow band gap of the commonly used arsenide and phosphide compound semiconductor materials the emission and thus the applications are limited to the infra-red to the yellow/green range of the spectrum. Wurzite gallium nitride (GaN) and its alloys exhibit a direct band gap that theoretically covers the emission spectrum from the deep ultra violet (UV) (aluminum gallium nitride - $Al_{1.00}Ga_{0.00}N \simeq 6.2\,eV = 200\,nm$ [1]) to the red (indium gallium nitride - $In_{1.00}Ga_{0.00}N \simeq 0.7\,eV = 1750\,nm$ [2, 3]) wavelength region. Similar to conventional phosphide or arsenide based lights emitters, (In,Ga)N based opto-electronic devices can be cost-efficiently produced in a high number on one wafer enabling new applications in the whole mentioned range of the spectrum. Nevertheless, (In,Ga)N LDs are commercially available only for a limited number of distinct wavelengths and properties that mainly fit the requirements of strong-selling products such as blue-ray disk, laser projectors [4], laser printers and reprographics [5]. Beside the usage in new consumer electronic products nitride based LDs enable more compact and more efficient systems for a wide range of existing applications since LDs outperform conventional solid-state laser systems in terms of lifetime, robustness, size and power consumption.

Fig. 1.1 shows the absorption spectra of pure water with a distinct minimum in the blue wavelength region. Since water covers more than 70% of the earth's surface and is component of every life form the analysis of its resolved ingredients is of great interest in many fields of research and application. For instance, solid state light sources emitting at the absorption minima of water are potentially used in medical applications like single cells cytometry [7] or undersea optical communications [8]. Since the emission wavelength of nitride based LDs is adjustable over a wide range, tailor-made light sources can be realized for solutions with specific absorption minima or for spectroscopic applications in general, e.g. Raman spectroscopy [9], laser- induced fluorescence emission for in-vivo chlorophyll fluorescence [10] or DNA sequencing [11].

The goal of the presented work is the realization of a current injection semiconductor laser diode (LD) emitting at 435.9 nm. The wavelength corresponds to the $7^3S_1 - 6^3P_1$

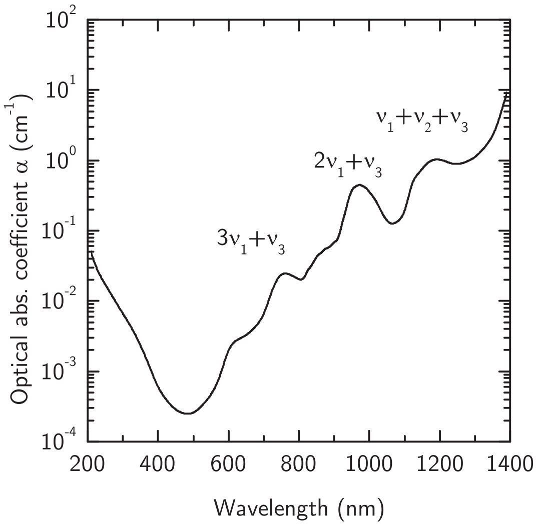


Figure 1.1: Absorption spectra of pure water showing lowest absorption in the blue wavelength region [6]. Wavelengths of increased absorption due to the distinct vibrational modes (symmetric stretching $\nu_1$, symmetric bending $\nu_2$ and asymmetric stretching $\nu_3$) are marked.

line of atomic mercury [12] in gas-discharge lamps, which is used for many biomedical and technical applications such as malaria [13] or tuberculosis [14] diagnosis, cell [15] and neural [16] research, fluorescence microscopy for chemical analysis [17] food safety and environmental testing [18].

## 1.2 (Al,In,Ga)N growth challenges

(Al,In,Ga)N device development starts with the specification of the heterostructure design. In general, LD heterostructures involve a high number of layers with different alloy compositions. The layer heterostructure is deposited on a GaN or a hetero-substrate, such as sapphire, silicon or silicon carbide by metal organic vapor phase epitaxy (MOVPE). Detailed information on the growth method can be found elsewhere [19, 20]. In order to reveal the optimum growth conditions for every single layer, the preparation of the different alloys is investigated before assembling the heterostructure. The realization of (Al,In,Ga)N with a high material quality represents a huge challenge due to the big differences in the material properties (especially the lattice constant) and the optimum growth conditions of the different binary compounds. Historically, the mastering of the MOVPE alloy formation limitations was the key to the realization of GaN based opto-electronic devices. Still, the MOVPE growth process significantly determines the material properties and quality and its mastery is of great importance for the realization of efficient GaN-based devices. Hereafter, crucial aspect of the (Al,In,Ga)N laser diode heterostructures growth are discussed.

The breakthroughs of the GaN technology was the achievement of p-doping using magnesium (Mg) [21, 22]. Due to the high activation energy of the acceptor of around 170 meV in GaN [23] and its passivation through the formation of Mg-H complexes [24] high Mg concentrations are required in order to realize sufficient p-type conductivity as well as low resistance p-type contacts. However, a high Mg concentration can cause compensation [25] of the holes and deteriorate the crystal perfection of the p-doped material by formation of clusters and other defects [26]. In order to obtain p-type doped material with both a high conductivity and a high material quality the optimum doping level and growth conditions resulting in a low compensation and a high Mg incorporation need to be determined.

The light emitting region of the LDs is embedded in a GaN waveguide core between AlGaN layers with a lower refractive index with respect to the effective index of the mode in the active region. This way the mode is vertically confined in theGaN waveguide core, guaranteeing a high optical intensity in the gain region of the device. The refractive index difference as well as the lattice mismatch strain in the heterostructure increase as the Al mole fraction of the cladding layer increases. Exceeding a critical AlGaN layer thickness and / or Al mole fraction layer cracking occurs [27] that massively disturbs the laser operation. The challenge regarding device realization is the optimization of the waveguiding structure, e.g. finding a good compromise between(Al,Ga)N layer thicknesses and Al mole fraction in the layers.

The active region consists of an InGaN/(In)GaN multiple quantum well (MQW) structure in order to confine the carriers in the InGaN QWs by /(Al,In)GaN barriers with a higher band gap energy. This way efficient radiative recombination is achieved. As a consequence of the different spontaneous and piezoelectric polarization of GaN and indium nitride (InN) [28] a non-vanishing dipole moment occurs resulting in high sheet carrier densities at the interfaces. The consequential electrical field strength is proportional to the gradient of the dipole moment at the interface. The electric field is perpendicular to the {0001} direction and spatially separates the holes and electrons in the quantum well when growing on the c-plane parallel to (0001). The separation of the carriers reduces the oscillator strength [29] and red-shifts the emission energy due to the quantum confined Stark effect [30].

Beside the oscillator strength, the crystal perfection of the active region, e.g. number of defects, interface roughness and material property uniformity, determines the material gain of the laser structure. The material quality in turn strongly depends on the In mole fraction in the solid of the active region [31] since the high lattice mismatch between InN and GaN [32] is a dominant driving force for material deterioration. The compressive strain of the InGaN forces defect formation or transition from layer by layer to 3D growth mode. Secondly, the low In incorporation efficiency at optimum GaN growth conditions of around 1000 °C requires low InGaN deposition temperatures of 700 °C. Because of the high In vapor pressure [33] and the high covalent radius of the In atom (1.44 Å) in comparison to the Ga atom (1.26 Å) the In is likely to desorb from the surface instead of being incorporated into the GaN. At low InGaN deposition temperatures the decomposition of the nitrogen precursor ammonia ($NH_3$) is reduced which causes the formation of point defects due to N deficiency in the layer [34]. Furthermore, the reduced mobility of the adducts on the growth surface at low growth temperatures prevents layer by layer growth. The challenge regarding InGaN growth is the determination of the best compromise between well thickness and In mole fraction in the solid and the optimization of the growth conditions for the best possible material quality.

## 1.3 Approach and organization of the work

The work focuses on the MOVPE growth and analysis of (In,Ga)N quantum well structures as well as the optimization and simulation of (Al,Ga)N waveguiding structures in order to improve the material properties as well as the modal gain of laser diodes emitting around 440 nm. Therefore, various sample structures and analysis methods are used. The growth and characterization parameters are described in chapter 2.

In chapter 3, the preparation and analysis of 15 nm and 120 nm thick InGaN single layers on GaN are discussed. Due to the high layer thicknesses the lateral and vertical distribution of the In can by determined by several methods. Additionally, the lower number of interfaces as well as the lower intrinsic fields strengths in the single layer structure, in comparison to MQW structures, simplifies the interpretation of the measurement results. The analysis and interpretation of the results reveal aspects of the In incorporation processes into GaN as well as fundamental InGaN material deterioration processes.

Next, described in chapter 4, InGaN layers are embedded into GaN barrier layers in order to form MQW structures. By varying the well width the indium gallium nitride (InGaN) /GaN heterostructure properties, e.g. In mole fraction in the solid, and their effect on the intrinsic field strength, the crystal perfection and luminescence efficiency are revealed. As a consequence of the increased structural complexity the exact determination of material properties is challenging and will be extensively described. The analysis of the influence of the QW width on the material quality and luminescence in the described experiments allows the specification of heterostructure parameters for the laser diode structures. Furthermore, by comparing the experimental results with theory the material parameters, used in the laser simulation later on, are adjusted.

In chapter 5, the influence of the MOVPE growth conditions, e.g. the growth temperature, on the material as well as luminescence properties of MQW structures is revealed. Furthermore, an experimental procedure is introduced that allows to correlate material properties, derived on MQW structures, with actual device properties. Using a multi-step epitaxy approach different heterostructures, e.g. MQWs, optical pumpable and current injection laser structures, are produced allowing for a valid comparison. The different structures contain identical active regions, which enables the determination of the material properties, e.g. the active region interface roughness, the In mole fraction in the solid and its spatial uniformity, and the device properties, e.g. threshold current density, output power and modal gain, at the same time. Unfortunately, the analysis of the results reveals a huge impact of the varied growth parameters on both the material quality and the optical confinement resulting in a simultaneous variation of the material gain and the modal gain.

In order to distinguish between the different effects on the device properties and enable growth optimization for laser heterostructures samples with identical modal gain but different material quality in the active region are prepared and analyzed (see chapter 6). Similar to chapter 5 the active region growth temperature is varied in order to affect the material quality. But this time the In mole fraction in the vapor is adjusted in order to realize identical In mole fractions in the quantum wells and barriers and thus identical modal gain in all samples. Analyzing the MQWs as well as the optical pumpable structures, the correlation of low threshold power densities with different growth conditions or respectively material properties is revealed. Using the characterization method that is most sensitive to the desired material properties a growth optimization scheme for laser heterostructures is established.

The basic developments have been made using laser diodes emitting around 400 nm since this wavelength is used in Blue-Ray disc storage systems and thus allows for a comparison with the well documented state of the art. Secondly, the 400 nm LDs feature moderate In mole fraction in the QWs facilitating the preparation of active regions with a good material quality. In order to realize emission at longer wavelengths both the material but also waveguiding losses at longer wavelengths need to be reduced. The optimization of the heterostructure layout as well as the growth conditions for 440 nm LD structures is addressed in chapter 7.

Due to economic reasons the heterostructures were deposited on sapphire based GaN templates with a high defect density. In the first part in section 7.1, the growth on low defect GaN substrates is discussed in order to decrease the number of threading dislocations in the active region and thus increases the material gain. Transferring the growth to GaN substrates one has to deal with different wafer bow resulting in different surface temperatures across the wafer. This in turn results in a lateral non-uniformity of the material properties and thus device characteristics. Growth experiments as well as simulations of the wafer bow during MOVPE growth will be described in order to specify GaN substrate properties that result in an identical uniformity of the wafer surface temperature as on sapphire substrate.

Next, in section 7.2, the influence of the waveguiding design on the optical confinement of the optical mode is discussed. Using device simulation as well as optical pumpable laser heterostructure variations the optical confinement at longer wavelengths is improved. After increasing the modal gain, the influence of modifications of the active region heterostructure layout on the material quality or respectively material gain is revealed in section 7.3. Using the optimization scheme established before the growth conditions of the active region are improved. Applying the optimized active region growth conditions together with the optimized waveguiding structure a 440 nm laser heterostructure is grown on low defect density GaN substrate. After processing of the wafer to an broad area laser diode (BA-LD) lasing at 436 nm in pulsed mode will be shown.