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14. ABSTRACT Significant progress was achieved in the epitaxy of deep UV AlN/ AlGaIn Bragg mirrors and microcavity structures paving the way to the successful fabrication of vertical cavity emitting laser structures and polariton lasers. For the first time DBRs providing sufficient high reflectivity for polariton emission were demonstrated. Thanks to a developed strain balanced Al _{0.85} Ga _{0.15} N template, the critical thickness before cracking could be enhanced to 1.95 µm. The fabricated 25.5 pair AlN/ Al _{0.65} Ga _{0.35} N DBR exhibit a maximum reflectivity above 98% at a Bragg wavelength of 270 nm.					
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Report Title

Final Report: Ultraviolet Polariton Laser

ABSTRACT

Significant progress was achieved in the epitaxy of deep UV AlN/ AlGa_N Bragg mirrors and microcavity structures paving the way to the successful fabrication of vertical cavity emitting laser structures and polariton lasers. For the first time DBRs providing sufficient high reflectivity for polariton emission were demonstrated. Thanks to a developed strain balanced Al_{0.85}Ga_{0.15}N template, the critical thickness before cracking could be enhanced to 1.95 μm . The fabricated 25.5 pair AlN/ Al_{0.65}Ga_{0.35}N DBR exhibit a maximum reflectivity above 98% at a Bragg wavelength of 270 nm.

In addition a relaxation mechanism by introducing periodic low temperature relaxation layers during DBR growth was proposed and demonstrated. No effect of the LT-AlN layers on the optical properties of the DBR was found. A partial relaxation of the DBR structure by the incorporated interlayers allows to increase the critical layer thickness in order to grow crack-free high reflective DBRs above 99%.

The practical implementation of our DBRs and multiple quantum well active regions was demonstrated by the fabrication of a full resonant half microcavity structure. Spectral resonance between the center wavelength of the stop band of the DBR and the MQW emission wavelength was found across the full radius region of the wafer.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

International Workshop on Nitride Semiconductors (IWN), Wroclaw, Poland, Aug. 24 – 29 2014
“Observation of High Carrier Concentrations in Ge-doped GaN: Growth, Characterization & Application”
M.P. Hoffmann, R. Kirste, C. Nenstiel, E. Sachet, M. Bobea, D. Alden, Z. Bryan, I. Bryan, J.P. Maria, A. Hoffmann, R. Colalazo, M. Gerhold, Z. Sitar

Materials Research Society Fall Meeting 2014, Nov. 30 – Dec. 5 2014, Boston
Talk1 – Session AA
“Strain Management for Crack Free High Al Content AlGaIn/AlN-based Bragg Reflectors with Many Layer Pairs”
Marc Hoffmann, Alexander Franke, Milena Bobea, Felix Kaess, James Tweedie, Isaac Bryan, Zachary Bryan, Ronny Kirste, Ramon Collazo, Zlatko Sitar, Michael Gerhold
Talk 2- Session AA
“Strain Relaxation Mechanisms and Point Defect Control of AlGaIn Films Grown on AlN Single Crystal Substrates”
Marc Hoffmann, Alexander Franke, Milena Bobea, Felix Kaess, James Tweedie, Isaac Bryan, Zachary Bryan, Ronny Kirste, Ramon Collazo, Zlatko Sitar, Michael Gerhold

Talk 3 – Session T
“High reflectivity AlN/AlGaIn DBR suitable for vertical UV emitting laser structures”
A. Franke, M. P. Hoffmann, I. Bryan, Z. Bryan, M. Bobea, J. Tweedie, F. Kaess, R. Kirste, C. Shelton, J.P. Maria, M. Gerhold, R. Collazo, and Z. Sitar

Lester Eastman Conference on High Performance Devices, Aug. 5-7 2014
“AlN/AlGaIn Distributed Bragg Reflectors for Deep UV Microcavity Lasers”
M. P. Hoffmann, A. Franke, M. Bobea, J. Tweedie, I. Bryan, Z. Bryan, F. Kaess, R. Kirste, R. Collazo, Z. Sitar, M. Gerhold

SPIE Photonics West, Feb. 12-18 2015, San Francisco
“All nitride confinement structures for deep UV microcavity laser”
A. Franke, M. P. Hoffmann, I. Bryan, Z. Bryan, M. Bobea, J. Tweedie, F. Kaess, R. Kirste, C. Shelton, J.P. Maria, M. Gerhold, R. Collazo, Z. Sitar

57th Electronic Materials Conference, Jun. 24-26, Columbus, Ohio
“Submicron Periodically Poled AlN Waveguides Grown by Metal Organic Chemical Vapor Deposition”
Dorian Alden, Ronny Kirste, Wei Guo, Felix Kaess, Isaac Bryan, Alexander Franke, Michael Gerhold, Ramón Collazo, and Zlatko Sitar

Number of Presentations: 7.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

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(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Book

TOTAL:

TOTAL:

Patents Submitted

Patents Awarded

Awards

none

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Felix Kaess	0.50
FTE Equivalent:	0.50
Total Number:	1

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Zlatko Sitar	0.02	
FTE Equivalent:	0.02	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

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Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME

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Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See Attachment

Technology Transfer

Final Report: UV Polariton Laser

Introduction

The conducted research within the project “UV Polariton Laser” significantly enhanced the fundamental understanding of AlGa_N based materials suitable for deep UV lasers. Major material limitation for the epitaxy of thick nitride based vertical emitting laser structures as well as a profound knowledge of microcavity lasers and the optical properties of nitride based multi quantum well emitters were gained.

III-nitride materials are the most promising candidates for development of vertical cavity surface emitting laser structures operating in the deep UV spectral range. The ternary AlGa_N alloy enables the fabrication of versatile lasers emitting in a wavelength region between 6.2 eV (AlN) and 3.39 eV (Ga_N) with low absorption losses and high efficiency. In addition polariton lasing at ultra-low thresholds and with no need of population inversion is feasible. Polariton formation is promoted by the high excitation binding energy and strong light-matter coupling strength in nitride materials.

However, deep UV or polariton lasing from vertical emitting III-nitride laser structures has not been reported so far. A major challenge remains the fabrication of highly reflective low loss Bragg mirrors (DBR), the growth of suitable, highly efficient multiple quantum well (MQW) active regions, the integration of both in a microcavity structure and the doping of the lasers for electrical carrier injection. All of these issues were addressed during the period of the project. Significant breakthroughs were achieved in the growth and design of high reflectivity, low absorption deep UV DBRs operating at 270 nm. In addition, AlGa_N multiple quantum well emitters having high output efficiency matched to the DBR resonance wavelength (spectral resonance) were demonstrated for the first time.

1. Epitaxy of high reflective deep UV distributed Bragg mirrors

The fabrication of high reflectivity DBRs require the fabrication of an alternating $\lambda/4$ layer stack of a high and low refractive index material. The refractive index contrast and number of layer pairs directly defines the maximum reflectivity and bandwidth of the mirror. For the applied low and high index material AlN and Al_{0.65}Ga_{0.35}N respectively, the achieved refractive index contrast at the intended resonance wavelength of 270 nm is 6%. The dispersion of the nitride materials compared to literature values is depicted in Fig. 1. In order to achieve a sufficient high reflectivity above 90% required for lasing, more than 15 layer pairs needed to be grown (Fig. 2 and simulated graph in Fig. 7).

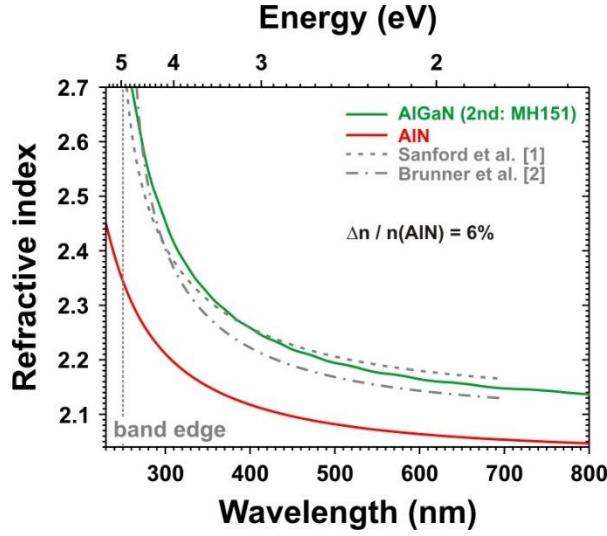


Fig. 1: Dispersion of the AlN (red) and $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ (green) materials used within the deep UV DBRs. The refractive index contrast at the resonance wavelength of 270 nm is 6%.

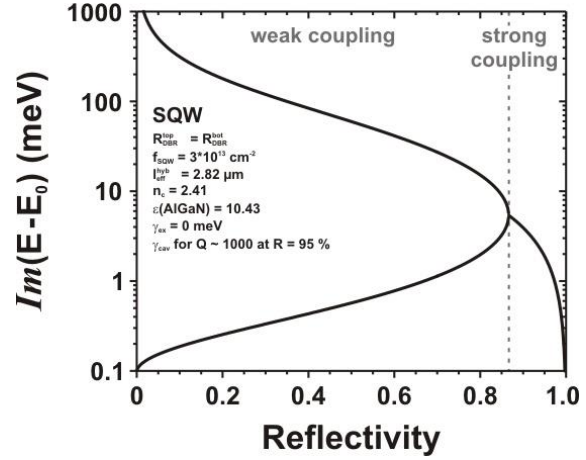


Fig. 2: Reflectivity required to achieve photon lasing (VCSEL) in the weak coupling regime and polariton lasing in strong coupling for a symmetric MQW sample having 10 multiple quantum wells.

However, thick DBR structures are prone to cracking. A large in-plane lattice mismatch and differences in the thermal expansion coefficient between the nitride materials AlN and $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$, and the sapphire substrate during the heteroepitaxy limits the maximum thickness. The critical thickness before cracking for a single $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ layer grown on an AlN template on top of a sapphire substrate was found to be about 1.0 μm corresponding to 15 DBR pairs. In order to increase the critical thickness for AlN/AlGaIn DBRs a profound strain relaxation study was conducted. During the study three major questions were pursued:

- role and interplay of dislocations and mismatch strain during growth
- design of proper template structure for the heteroepitaxy of thick DBRs on sapphire substrates
- strain relaxation concepts for the growth to enhance the critical thickness

The structural design of the used AlN/ $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ DBR is shown in Fig. 3. On top of a sapphire substrate a 300nm thick AlN template was grown. It provides a low dislocation density in the order of 10^{10} cm^{-2} and an atomic smooth surface as indicated by Fig. 4 (left image). To verify the suitability of the template for the growth of AlN/ $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ DBRs a structure having 20.5 layer pairs was fabricated. The DBR exhibit a high structural and crystalline quality as shown by the XRD $\theta/2\theta$ scan in Fig. 5 (top image). Prominent superlattice fringes up to the 4th order indicate smooth interfaces between the AlN and $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ layers as well as a high periodicity. Both anticipate enhanced optical properties of the Bragg mirror. The reflectivity spectra under normal incidence are shown in Fig. 6. A pronounced spectral region of high reflectivity, the stop band, of 8 nm is achieved. The maximum reflectivity at the center wavelength of the stop band is 93%.



Fig. 3: Sketch of the AlN/Al_{0.65}Ga_{0.35}N DBR on an AlN template on (0001)-sapphire.

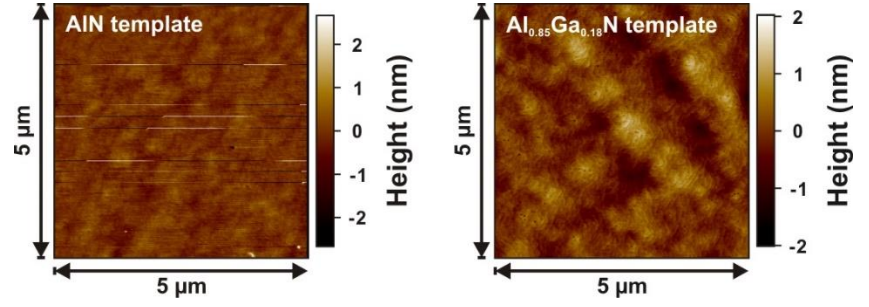


Fig. 4: Surface morphology of the AlN (left) and Al_{0.85}Ga_{0.15}N (right) template layer.

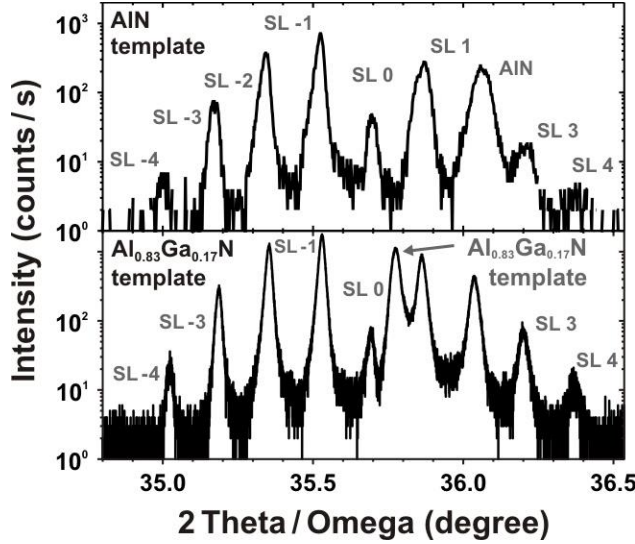


Fig. 5: XRD $\theta/2\omega$ scans of a 20.5 layer pair DBR grown on top of an AlN template (top) and Al_{0.85}Ga_{0.15}N template (bottom).

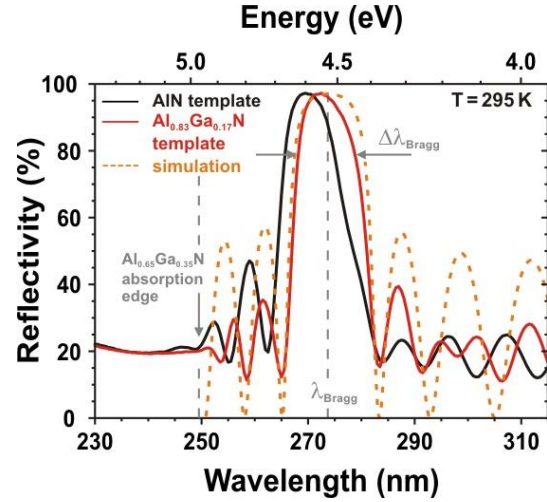


Fig. 6: Reflectivity of a 20.5x DBR grown on an AlN (black) and Al_{0.85}Ga_{0.15}N template (red). The dashed curves shows a calculated spectrum for the sample on the Al_{0.85}Ga_{0.15}N template.

However, during the growth of the DBR on the AlN template Al_{0.65}Ga_{0.35}N layer are exposed to compressive strain. The total strain energy increases gradually with increasing the number of layer pairs. An additional thermal strain component is introduced by cooling the structure from growth temperature (1100 °C) to room temperature. Both resulting in a relaxation of the complete structure by cracking. The critical thickness was found to be 1.57 μm equal to 23 layer pairs. A strong increase of the cracking limit was achieved by replacing the AlN by a 500 nm thick Al_{0.85}Ga_{0.15}N template. The Al composition was chosen at the average value between the AlN and Al_{0.65}Ga_{0.35}N used within the DBR. By relaxing the Al_{0.85}Ga_{0.15}N template layer the total

strain energy could be reduced. Now both layers, the tensile strained AlN and the compressive strained $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$, contribute with a decreased fraction to the overall strain energy. To guarantee good optical performance of the DBR grown on the $\text{Al}_{0.85}\text{Ga}_{0.15}\text{N}$ template first the template surface and crystalline quality needed to be verified. The sample morphology of the template is depicted on the right hand side of Fig. 4. The average RMS roughness is negligible increased to 0.4 nm compared to the AlN template (0.2 nm). The structural quality in terms of dislocation density and crystalline quality was found to be similar to the AlN.

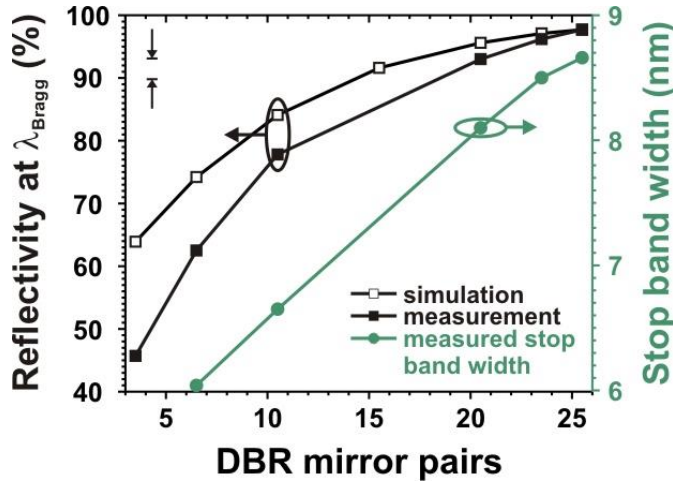


Fig. 7: Maximum reflectivity of AlN/ $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ DBRs of a various number of layer pairs grown on a strain engineered $\text{Al}_{0.85}\text{Ga}_{0.15}\text{N}$ template. Cracking occurs for DBRs of more than 26 pairs.

A proof of the strain engineering approach is given by the growth of AlN/ $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ DBRs of various number of mirror pairs. Figure 7 shows the achieved reflectivity for DBRs having 3.5 to 25.5 layer pairs deposited on top of the $\text{Al}_{0.85}\text{Ga}_{0.15}\text{N}$ template. The maximum reflectivity increases nonlinear following the trend of the calculated graph. The maximum reflectivity achieved for 25.5 layer pairs is 98%. Thicker structures above 26 pairs suffer from cracking. Therefore, the critical thickness could be enhanced by 3 mirror pairs to $1.98\text{ }\mu\text{m}$ compared to the DBRs on an AlN template while the optical performance is retained. The reflectivity spectra of a 20.5 pair DBR grown on both template structures is compared in Fig. 6. Both DBRs exhibit a similar stop band width and maximum reflectivity of 93%. A slight shift of the center wavelength of the stop band (Bragg wavelength) of the DBRs could be attributed to minor thickness deviations within the DBR.

The demonstrated maximum reflectivity of above 98% for an AlN/ $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ DBRs marks the highest reported value for nitride deep UV DBRs reported so far.

2. Strain relaxation concepts to enhance the maximum reflectivity

To increase the critical thickness above the demonstrated limit further strain relaxation concepts needed to be applied. Even if the total strain energy is reduced by using the $\text{Al}_{0.85}\text{Ga}_{0.15}\text{N}$ template, strain is still accumulated during DBR growth. In order to grow thick crack-free structures a partially relaxation and relief of the total strain energy is required. However, relaxation in the wurtzite crystal system is a topic of ongoing research and strongly discussed.

Besides cracking, favored relaxation mechanisms include: nucleation of new dislocations, “bending” or transformation of already existing dislocations, surface roughening, buckling, or cracking [3].

In order to partially relax the structures we pursued the concepts of the periodic incorporation of thin relaxation layers during the DBR growth. A strain relief is provided by dislocation bending and surface roughening by a 3 dimensional grown low temperature renucleation layer. The concept and a schematic cross section of the fabricated samples are shown in Fig. 8.

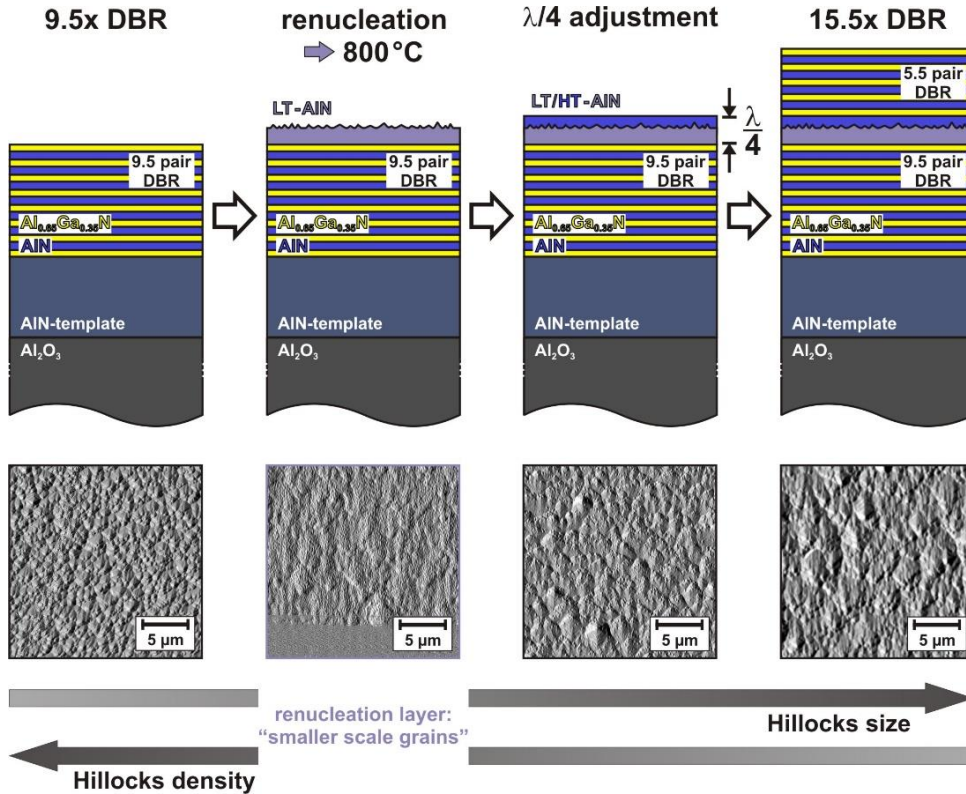


Fig.8: Strain relaxation by introducing a low temperature AlN interlayer during growth (top). The bottom AFM amplitude images show the sample surface of the structures before and after the renucleation step.

On top of a 9.5 layer pair AlN/ $\text{Al}_{0.65}\text{Ga}_{0.35}\text{N}$ DBR, grown at 1100°C , a low temperature AlN (LT-AlN) layer at 800°C was deposited (Fig. 8, middle left). The total thickness of the layer is about 17 nm. In order to keep the right $\lambda/4$ thickness periodicity within the DBR the layer was covered by an 15 nm thick high temperature AlN layer (1100°C , Fig.8 middle, right) and a 5.5 pair DBR (grown at 1100°C , Fig. 8 right). The sample morphology before and after the deposition of the low temperature renucleation layer is depicted in the bottom AFM images of Fig. 8. A significantly surface roughening and increase of the structural grain size is observed for the low temperature renucleation layer (second column in Fig. 8). The achieved three dimensional grains are expected to be suitable for a partially relief of the accumulated strain of the DBR. With continuing the growth at higher temperatures the surface features increases in size while their density is decreased. The final morphology of the 15.5 pair DBR is decorated by Hillocks composed of single atomic steps as known for the growth of AlGaIn materials.

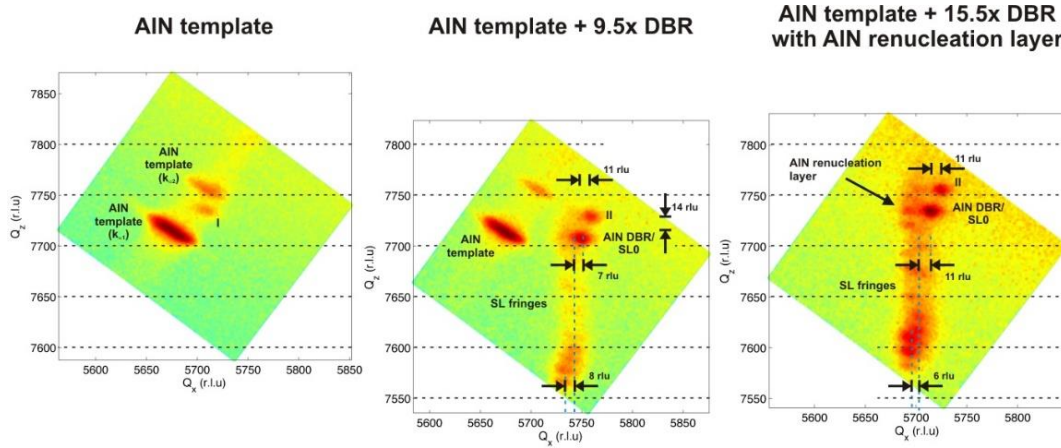


Fig. 9: XRD reciprocal space maps on the structure before (two left images) and after the renucleation (right) indicating a pseudomorphic growth and a high periodicity as well as sharp interfaces within the DBR.

In order to investigate the relaxation behavior of the renucleation layer XRD reciprocal space maps (RSM) were conducted. Fig. 9 summarized the RSMs before (left, middle) and after the renucleation (right). All layers are grown pseudomorphic with respect to the AlN template. Furthermore, an equidistant layer distribution and sharp interfaces are indicated by pronounced superlattice fringes at low Q_z values in both DBR structures. No influence of the low temperature interlayer on the periodicity could be observed. However, a verification of the relaxation behavior of the LT- AlN and the subsequently grown DBR is difficult. A clear distinction between the AlN used within the DBR, the template, and the low temperature AlN renucleation layer is not possible. An evidence for a changed strain state in the structure including the renucleation layer is given by additional diffraction features at low Q_x and high Q_z indicated by the arrow in right image of Fig. 9.

Optical properties of the DBR including a LT-AlN strain relaxation layer

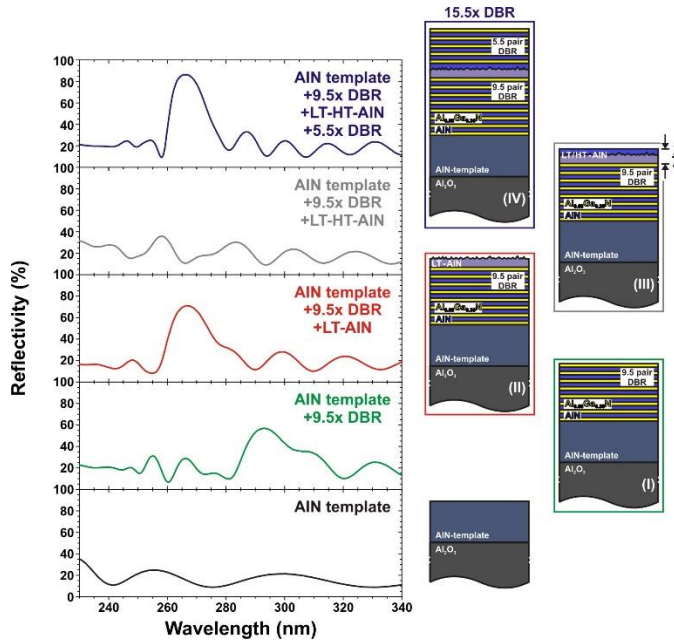


Fig. 10: Comparison of the reflectivity spectra for different growth steps before (bottom) and after the deposition of the low temperature AlN renucleation layer (top).

The effect of the renucleation layer on the evolution of the stop band of the DBR is shown in Fig. 10. Starting from the AlN-template (bottom) an almost constant reflectivity of 20% superimposed by thickness fringes is achieved. By introducing the DBR the reflectivity increases within the stopband region. A clear stop band, centered at around 265 nm emerges for the structure including the LT-AlN renucleation layer. No optical degeneration is observed. The final 15.5 pair DBR structure (Fig. 10, top) exhibit a well-defined stopband consistent with the previously observed center wavelength of the structure including the LT renucleation layer. The achieved maximum reflectivity matches with previous observations on DBRs without renucleation layer as shown in Fig. 7. Consequently, no deviation of the optical performance of the DBR by the renucleation layer is expected.

3. Fabrication of a resonant half microcavity structure

The fabrication of a vertical emitting laser structure requires the integration of an active medium, emitting at the intended resonance wavelength, between a top and bottom Bragg mirror. In order to verify spectral resonance between the Bragg wavelength of the bottom AlN/ Al_{0.65}Ga_{0.35}N DBR and the active medium a half microcavity was grown. As depicted in the cross-section scanning electron microscopy image Fig. 11 it consist of a 3.5x layer pair bottom DBR an optical λ thick cavity including a 5 fold multiple quantum well (MQW) active region. In order to allow a maximum light-matter coupling, the MQW was positioned at the maximum of the standing wave pattern in the center of the λ cavity.

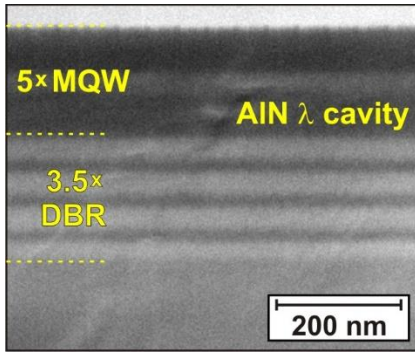


Fig. 11: Cross section scanning electron microscopy image of a half microcavity structure consisting of a 3.5 pair bottom DBR and an optical λ thick AlN cavity. Multiple quantum wells as active medium were centered in the λ cavity.

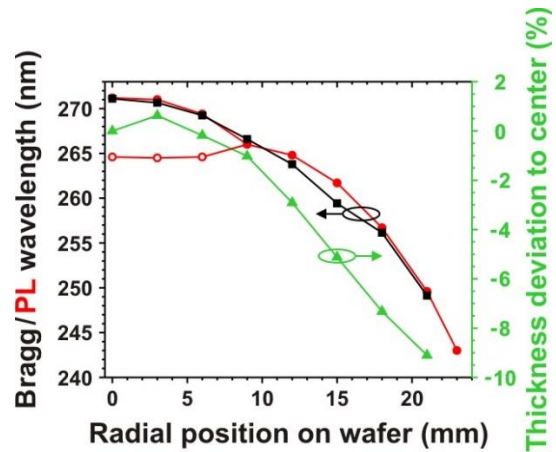


Fig. 12: Spectral resonance between the Bragg wavelength (black) of the DBR and the MQW emission (red) is found across the full radius range of the wafer.

Spectral resonance between the Bragg mirror and the active medium

The optical properties of the bottom DBR and the emission behavior of active MQW region were investigated by spatially resolved reflectivity and photoluminescence (PL) measurements. Reflectivity and PL spectra were recorded at the same spot across the diameter of the wafer. The spatial behavior of the center wavelength of the stop band of the DBR is shown by the black line plotted in Fig. 12. The Bragg wavelength in the center of the wafer is located at 271 nm and decreases nonlinearly toward the edge by 25 nm. The change could be attributed to a monotonously changed DBR layer thickness from the center to the edge of the wafer, as indicated by the green line in Fig. 12.

The exact same spatial behavior across the wafer as found for the stopband is observed for the MQW emission (red graph in Fig. 12). Consequently, spectral resonance between the Bragg wavelength and the MQW emission wavelength is achieved on the full wafer. It indicates the suitability of the structure for the fabrication of microcavity lasers operating in the weak (photon laser) or strong coupling regime (polariton laser).

4. Conclusion

Significant progress was achieved in the epitaxy of deep UV AlN/ AlGa_N Bragg mirrors and microcavity structures paving the way to the successful fabrication of vertical cavity emitting laser structures and polariton lasers. For the first time DBRs providing sufficient high reflectivity for polariton emission were demonstrated. Thanks to a developed strain balanced Al_{0.85}Ga_{0.15}N template, the critical thickness before cracking could be enhanced to 1.95 μm . The fabricated 25.5 pair AlN/ Al_{0.65}Ga_{0.35}N DBR exhibit a maximum reflectivity above 98% at a Bragg wavelength of 270 nm.

In addition a relaxation mechanism by introducing periodic low temperature relaxation layers during DBR growth was proposed and demonstrated. No effect of the LT-AlN layers on the optical properties of the DBR was found. A partial relaxation of the DBR structure by the incorporated interlayers allows to increase the critical layer thickness in order to grow crack-free high reflective DBRs above 99%.

The practical implementation of our DBRs and multiple quantum well active regions was demonstrated by the fabrication of a full resonant half microcavity structure. Spectral resonance between the center wavelength of the stop band of the DBR and the MQW emission wavelength was found across the full radius region of the wafer.

5. Literature

- [1] Sanford et al., "Refractive index study on Al_xGa_{1-x}N films grown on sapphire substrate" Jour. Appl. Phys, **94**, 2980, (2003)
- [2] Brunner et al., "Optical constants of epitaxial AlGa_N films and their temperature dependence", Jour. Appl. Phys, **82**, 5090, (1997)
- [3] Claeys et al., "Fundamental and Technological Aspects of Extended Defects in Germanium", Springer Verlag, Berlin, Heidelberg, pages 153-240, (2009)