



AFRL-AFOSR-UK-TR-2019-0048

**Development and Experimental Realization of Semiconductor Lasers
under Non-equilibrium Operating Conditions**

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**09/11/2019
Final Report**

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REPORT DOCUMENTATION PAGE				<i>Form Approved</i> OMB No. 0704-0188	
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1. REPORT DATE (DD-MM-YYYY) 11-09-2019		2. REPORT TYPE Final		3. DATES COVERED (From - To) 01 Jun 2014 to 31 May 2017	
4. TITLE AND SUBTITLE Development and Experimental Realization of Semiconductor Lasers under Non-equilibrium Operating Conditions				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER FA9550-14-1-0159	
				5c. PROGRAM ELEMENT NUMBER 61102F	
6. AUTHOR(S) Wolfgang Stoltz				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Philipps-Universität Biegenstr. 10-12 Marburg, Hessen, 35032 DE				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) EOARD Unit 4515 APO AE 09421-4515				10. SPONSOR/MONITOR'S ACRONYM(S) AFRL/AFOSR IOE	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-AFOSR-UK-TR-2019-0048	
12. DISTRIBUTION/AVAILABILITY STATEMENT A DISTRIBUTION UNLIMITED: PB Public Release					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT Finished research.					
15. SUBJECT TERMS maxwell bloch equations, diode pumped solid state laser physics, MBE for laser development, EOARD					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT SAR	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON LOCKWOOD, NATHANIEL
a. REPORT Unclassified	b. ABSTRACT Unclassified	c. THIS PAGE Unclassified			19b. TELEPHONE NUMBER (Include area code) 011-44-1895-616005

Technical Project Report

“Development and Experimental Realization of Semiconductor Lasers under Non-equilibrium Operating Conditions”

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Project period: June 1, 2014 – May 31, 2017**

DESIGN AND EPITAXIAL GROWTH OF LASER AND ABSORBER STRUCTURES

The very intense and closely interlinked collaboration between the group of Prof. J. Moloney at the Optical Sciences Center (OSC) and the group of Prof. W. Stolz at the Material Sciences Center of Philipps-University Marburg (UMR) was key for the design, fabrication and characterization of high quality semiconductor laser devices capable of generating ultrashort pulses duration in the sub-100fs regime. The microscopic many-body theory developed during this project at OSC was used extensively to understand and simulate the pulse formation dynamic and properties with specific theoretical epitaxial structures. These simulations showed that non-periodic ultra-broadband gain structures with adequate dispersion compensation is key for ultrashort sub-100fs pulse formation. These novel and unique epitaxial layer stacks design were subsequently grown at UMR by metal organic vapor phase epitaxy (MOVPE). These multiple quantum wells (MQW) structures are inherently non-periodic and highly strained, which presented a great challenge for low-defects high quality epitaxial growth. The structure design and targeted wavelength were subsequently adapted to remediate these difficulties. Accordingly, we designed and realized several designs of these sophisticated layer stacks for emission wavelengths around 1040nm, 865 nm, 980 nm and 1030 nm. In addition, we designed and grew ultrafast quantum-well absorbers suitable for the generation of ultrashort pulses at these wavelengths. The final designs have shown a great potential for the generation of high power ultrashort pulses. These structures, combined with a precise dispersion management and a novel cavity design were used to produce pulse durations as short as 95fs. Here we present some of the structures designed and fabricated and discuss the technical challenges that they represented.

MULTIPLE QUANTUM WELL GROWTH CHALLENGES

In order to realize these novel more complex and sophisticated semiconductor gain structures in particular for the intended extreme non-equilibrium conditions, novel designs and unique epitaxial layer stacks applying metal organic vapour phase epitaxy (MOVPE) had to be realized and elaborated in detail. Specific non-periodic gain layer stacks for VECSEL structures have to be developed also in conjunction with an appropriate non-destructive analysis technique. The quantitative structural analysis of the intended non-periodic multiple quantum well heterostructure (MQWH) gain layer stacks was not established at project start. This analysis, however, is a prerequisite for the experimental verification of the intended layer design and, thus, for the quantitative theory-experiment comparison of the VECSEL laser properties. In

addition, shorter carrier lifetimes in specifically designed SESAM-layer stacks also applying novel material systems have to be achieved in order to potentially verify the sub 100 fsec laser pulse regime.

The project partner at UMR have applied their broad knowledge and in-depth experience in the epitaxial growth of resonant periodic gain structures (RPG) in order to optimize designed non-periodic gain structures for VECSEL-based applications. For these non-periodic layer stacks the experimental verification by detailed high-resolution X-ray diffraction (HR-XRD) and subsequent full dynamical diffraction modeling has been a challenging novel research task. In addition, UMR have explored surface-coupled SESAM-QWH absorbers also applying specific surface passivations techniques to improve the SESAM reliability and realized novel SESAM-structures with designed short carrier lifetimes.

These research studies for the novel epitaxial layer stacks have been performed both on small 2" wafer size R&D-MOVPE-systems (*Figure 1*) as well as on production type multi 4"-wafer MOVPE-reactors (*Figure 1*). Production-type multi-wafer MOVPE-systems offer the advantage of reduced particulate and defect formation, drastically reducing scattering related losses in VECSEL. On the long-term, the studies on MOVPE production reactors form the basis for a potential subsequent commercial transfer of the obtained results. In addition, the commercial usage of 4"-GaAs-wafers for a variety of optoelectronic as well as electronic device applications has led to significant improvements in crystalline quality of the 4"-GaAs-wafers in particular with respect to residual threading dislocations density.



*Figure 1. Available MOVPE reactors: R&D MOVPE-reactor tool for 2" substrate wafers (Aixtron Aix 200-GFR-reactor) (left) and MOVPE-production reactor tool (Aixtron Aix2600-G3-reactor) in a 12 * 4" wafer configuration (right).*

For each and every active layer design the strain management had to be optimized iteratively with the cooperation of UMR and OSC. Due to the large number of compressively strained (GaIn)As-QWH layers and their specific positioning as active layers in the VECSEL structure, carefully designed tensile-strained Ga(AsP)-barrier layers had to be optimized with respect to their precise composition and thickness values in order to optimize the precise positioning of the active (GaIn)As-QWH layers but never exceed the integrated strain-energy density borders for the onset of strain relaxation processes (formation of dislocations) during the MOVPE growth process. Detailed high-resolution X-ray diffraction (HR-XRD) and atomic force microscopy

studies had to be performed in order to experimentally clarify these strain limits for both the compressively as well as the tensile strained constituent layers.

The structural analysis of crystalline but non-periodic layer stacks of high-strained individual layer stacks has been performed by experimental HR-XRD-analysis in comparison to full dynamical XRD-theory modelling developed for periodic layer stacks. While the analysis of periodic layer stacks is nowadays straightforward, the analysis of the designed non-periodic crystalline layer stacks proved to be very complex, as exemplified in *Figure 1*.

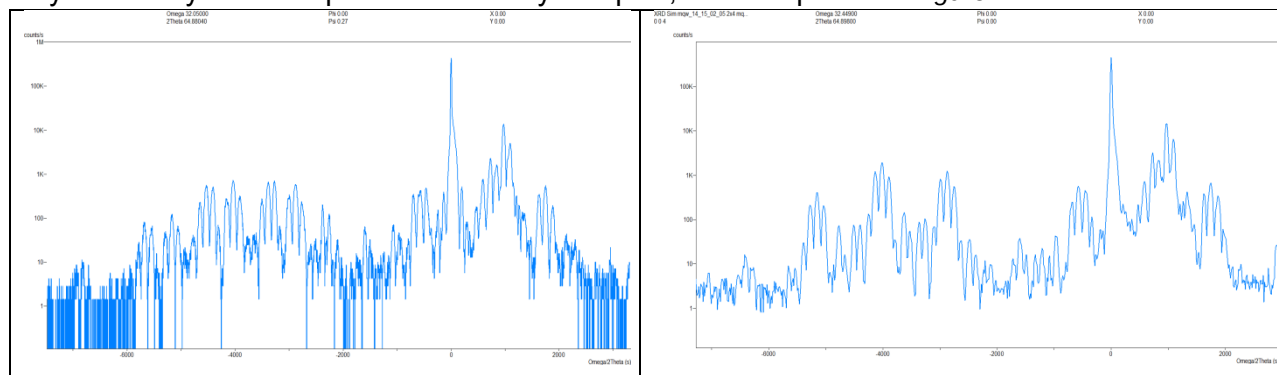


Figure 1 Comparison of experimental (left) and theoretical high-resolution X-ray diffraction (HR-XRD) pattern (right) of non-periodical (GaIn)As/Ga(PAs)-MQWH around the (004) reflection of GaAs indicating the high crystalline perfection of the realized non-periodic MQWH.

On the left side, an experimental HR-XRD-pattern of one of the realized non-periodic (GaIn)As/Ga(AsP)-MQWH layer stacks is shown. On the right side, the present best description of this pattern is summarized. While the overall XRD-pattern with respect to the positioning and the individual satellite linewidths as well as the envelope structure show a good agreement in between the experimental pattern and the theoretical modelling, it becomes clear that the intensity substructure of various reflections differ significantly. At present, detailed efforts are underway to clarify these differences also applying higher order corrections to the full dynamical HR-XRD modelling.

MULTIPLE QUANTUM WELLS STRUCTURES AT 1040NM

Due to previous experience, diagnostic equipment available, and record CW performance demonstrated by our groups with the (GaIn)As/GaAs material system for VECSEL structure emitting around 1040nm, we started to design VECSEL structures around this wavelength. One of the design consisted of a short active region containing a stack of 4QWs per antinode, with a total of 2 antinodes (4+4QWs). Another design consisted of only 1 stack of 4 QWs. To compensate the reduced pump absorption in these short active regions, we designed a transparent signal (AlGa)As/AlAs DBR followed by a pump (AlGa)As/AlAs DBR to recycle the pump not absorbed on the first pass. The structure layout and the temperature dependent reflectivity are presented on Figure 3.

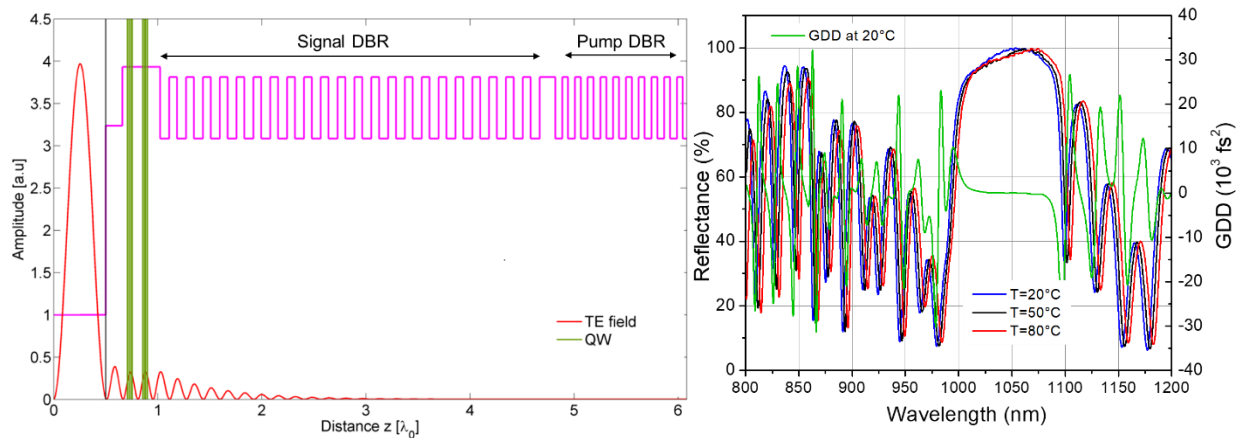


Figure 3. Structure layout of a 4+4QWs VECSEL at 1040nm with a double Signal+Pump DBR (left), and corresponding reflectivity and GDD spectrum (right).

After thorough characterizations, it appeared that these structures have a very limited gain available due to a high density of dislocations, inhibiting any modelocking operation. Images of surface photoluminescence of a sample are presented on Figure 2, showing a high density of threading dislocations (cross-hatch darklines).

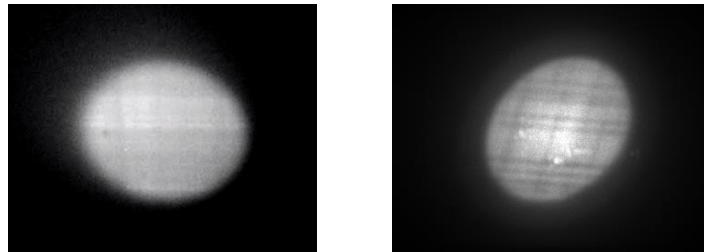


Figure 2. Surface photoluminescence images of a highly strained MQW structure at 1040nm.

To alleviate the inherent strain accumulated by the QWs in the active region and reduce the defect density, we designed and fabricated structures at shorter wavelength for a strain-free and low strain profile at 865nm and 980nm respectively.

STRAIN-FREE MULTIPLE QUANTUM WELLS STRUCTURES AT 865NM

The structure layout of the MQW structure at 865nm is presented on Figure 3. It consists of 2 stacks of 6 unstrained GaAs/ $\text{Al}_{0.064}\text{Ga}_{0.936}\text{As}$ QWs (6+6QWs) followed by 23 pairs of (AlGa)As/AlAs signal DBR and 14 pairs of (AlGa)As/AlAs pump DBR. It is designed to be pumped in the QWs by a 808nm pump. The photoluminescence spectra show a good calibration of the QW emission wavelength. However, when the structures were tested in a VECSEL cavity, these structures emitted at much longer wavelengths with very poor power performance and efficiency. These results suggested a saturation of the pump absorption when the structure is inverted, due to the proximity of the pump wavelength to the lasing wavelength. This effect was confirmed by comparing the output power using a different pump having slightly different wavelength (Figure 4). The shorter pump wavelength gives a lower threshold and higher power, suggesting a reduced pump absorption saturation.

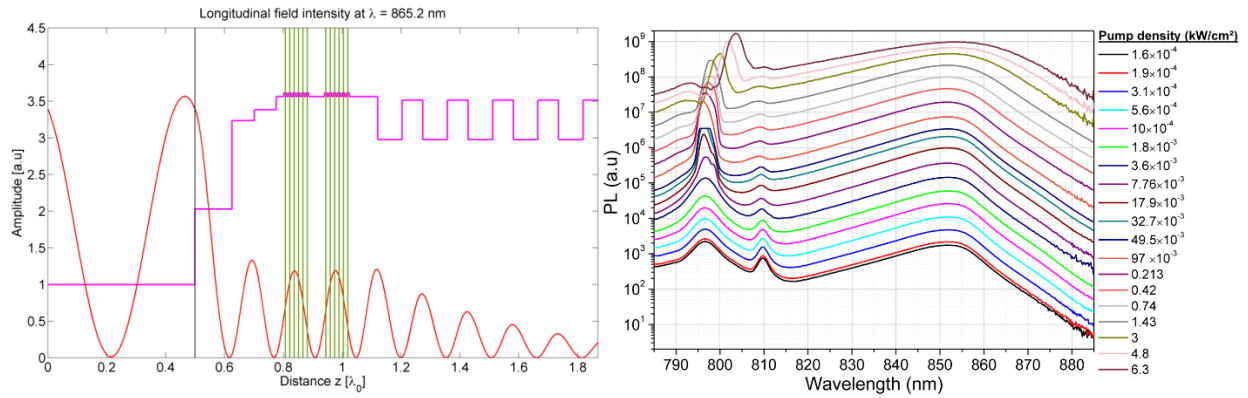


Figure 3. Structure layout of a MQW structure at 865nm and corresponding PL spectra at different excitation.

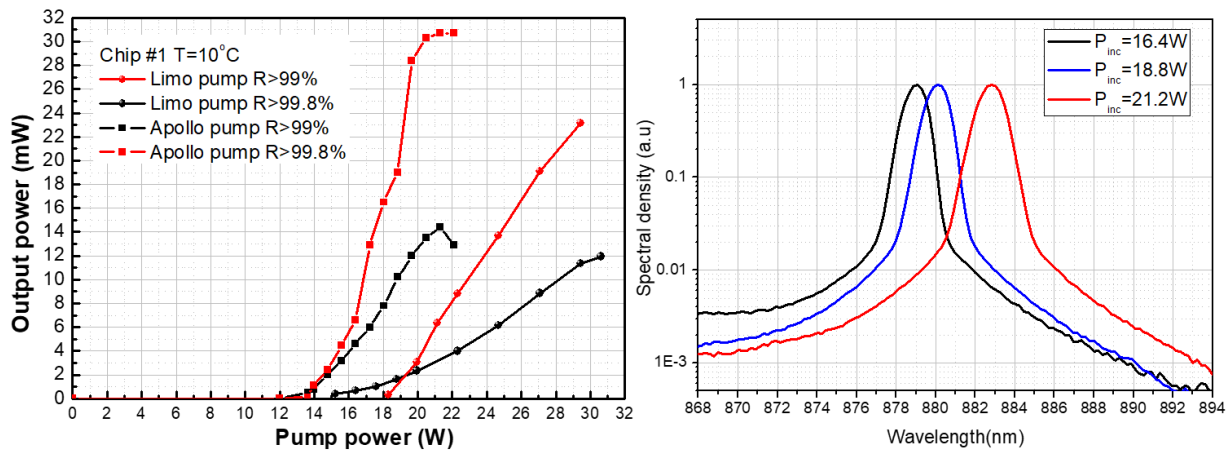


Figure 4. Output power and spectral characteristic of a MQW VECSEL at 865nm. The Apollo pump has a slightly shorter wavelength than the Limo pump (802nm vs. 808nm).

To resolve this problem, we designed some barrier-pumped structures at 980nm, a wavelength far enough from the pump wavelength while keeping a moderate strain.

LOW STRAIN MULTIPLE QUANTUM WELLS STRUCTURES AT 980NM

Over the course of this project, we designed and realized numerous structures at 980nm. We changed iteratively key parameters such as the number of QWs stacked per antinodes of the field, the total number of antinodes, the spacing between QWs, the strain compensating layers, the type of DBR (double DBR or hybrid metal-semiconductor DBR), the dispersion management layers, and the type/quality of GaAs substrate used. The goal was to realize and optimize a MQW structure with a very broad gain spectrum and low dispersion, while keeping the integrated strain below the critical limit that favor dislocations formation or spread (~15% nm). The following list is a short description of the different structures realized.

Active Region	GaAs(P) Barriers	DBR type	Dispersion management	GaAs Substrate	Performance
4+4 QWs	Low P content	Double*	Dielectric coating	Low defect	Poor (defects)
2+2+2+2 QWs	Low P content	Double*	Dielectric coating	Low defect	Good [11]
6 QWs	Low P content	Double*	Dielectric coating	Low defect	Poor (defects)
4+4+4 QWs	High P content	Double*	Dielectric coating	Low defect	Poor (defects)
4+4 QWs	High P content	Hybrid†	Dielectric coating	Low defect	Poor (defects)
4+4 QWs	High P content	Hybrid†	Multilayer SC + dielectric	Low defect	Poor (defects)
4+4 QWs	High P content	Double*	Multilayer SC + dielectric	Low defect	Poor (defects)
2+2+4 QWs	High P content	Double*	Multilayer SC + dielectric	Low defect	Poor (defects)
121+121+1 QWs	Low P content	Double*	Bi-layer dielectric	Very low defect	Poor (DBRs)
121+121+1 QWs	Low P content	Double*	Multilayer SC + dielectric	Very low defect	Poor (DBRs)
121+121+1 QWs	Low P content	Hybrid†	Bi-layer dielectric	Very low defect	Limited (gain)
3x121+1 QWs	Low P content	Hybrid†	Bi-layer dielectric	Very low defect	Very good [12]
3x121+1 QWs	Low P content	Double*	Bi-layer dielectric	Very low defect	Poor (DBRs)
3x121+1 QWs	Low P content	Double*	Multilayer SC + dielectric	Very low defect	Poor (DBRs)

(*)Double Semiconductor AlGaAs/GaAs DBR: 23pairs (signal) + 13 pairs (pump).

(†)Hybrid Semiconductor-metal DBR: short AlGaAs/GaAs DBR + gold reflector on patterned matrix [3].

During the project period, all relevant design structures could be realized successfully at UMR by MOVPE growth and have been transferred to OSC for subsequent processing and analysis of the respective pulse laser performance.

An example of structure layout at 980nm and its integrated strain profile is presented on Figure 5. We can see that despite having multiple strained QW stacked together, it is possible to keep the integrated strain below the theoretical/empirical “critical” limit of 15%nm. This strain limit is, however, dependent on many factors, such as the localized strain, the substrate quality, the growth parameters, etc.

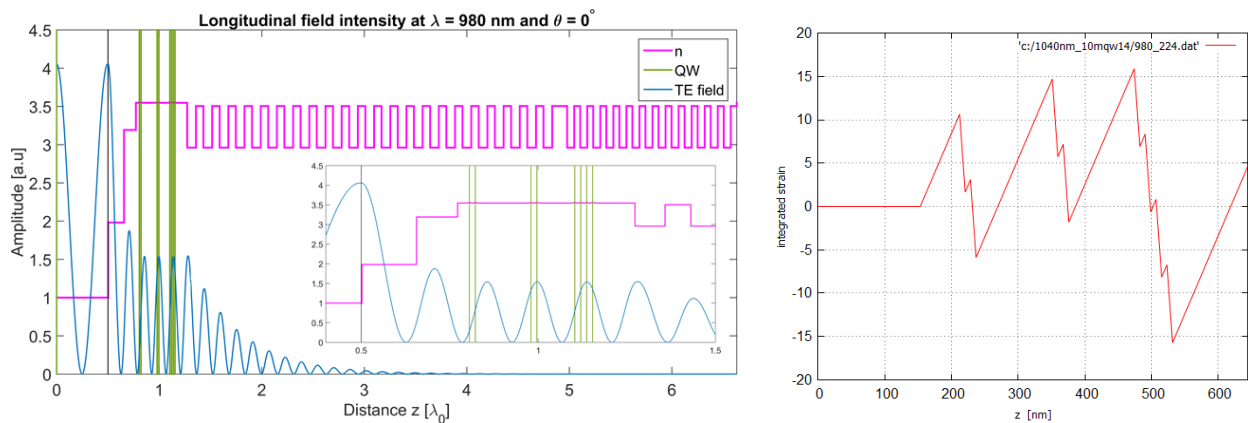


Figure 5. Structure layout of a 980nm MQW (2+2+4 QWs) VECSEL structure with a double DBR 23 (AlGa)As/GaAs pairs for signal + 13 pairs for pump (left) and its integrated strain profile (right).

An evaluation of the crystal quality and dislocation density can be done by looking at the surface photoluminescence of the structure, as dislocation are non-radiative recombination center and will appear as a dark spot or lines. The Figure 6 shows the PL images on three different areas of a VECSEL chip, clearly showing a relatively high density of dislocation lines.

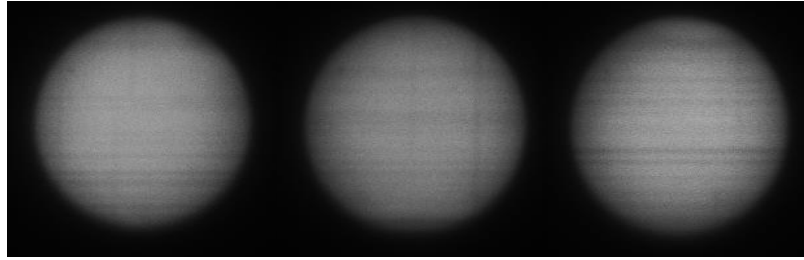


Figure 6. Surface photoluminescence images of a (2+2+4 QWs) VECSEL structure at 980nm.

During the course of the project, after the growth of multiple structures having different strain profiles, it became clear that the remaining threading dislocation density in the available GaAs-substrate wafers influences the performance of the designed active VECSEL layer stacks significantly. This is due to the bending of threading dislocation segments at the various interfaces of the highly strained (GaIn)As/Ga(AsP)-MQWH layer stack. This mechanism leads to the formation of non-radiative recombination centers near the active (GaIn)As-QWH layers as verified by spatially resolved imaging techniques and, thus, to a significant reduction in optical gain provided for the optical pulse formation process. This challenge has been solved by specifically characterizing and selecting the highest quality 4" GaAs-substrate wafers.

With these higher quality wafers we were able to realize MQW with very low density of dislocations and demonstrate state of the art performance in term of pulse duration [2], validating the concept of MQW structures design for ultrashort pulse generation.

OPTIMIZED MULTIPLE QUANTUM WELLS STRUCTURES AT 1030NM

After the proof of concept validation at 980nm, we designed and realized similar structures at 1030nm, accounting for the additional strain at this wavelength. This wavelength range is more suitable for a possible Yb-fiber amplification (frequency comb application), and for the ultrafast diagnostic equipment available. The short description of the optimized structures is shown below:

Active Region	GaAs(P) Barriers	DBR type	Dispersion management	GaAs Substrate	Performance
2x121+21+1 QWs	Low P content	Hybrid: 16 DBR pairs	Bi-layer dielectric	Very low defect	Record 95fs
2x121+21+1 QWs	Low P content	Hybrid: 22 DBR pairs	Bi-layer dielectric	Very low defect	Very good

The Figure 9 shows the structure layout and the integrated strain of the 2x121+21+1QWs structure with 16 DBR pairs. The number and spacing of the QWs were optimized to keep the strain below a critical limit while minimizing the gain available (larger spacing, one less QW on the third antinode compared to the 980nm structure, boost QW on the last antinode). The Table 1 shows the detailed structure design of the record-performance VECSEL structure.

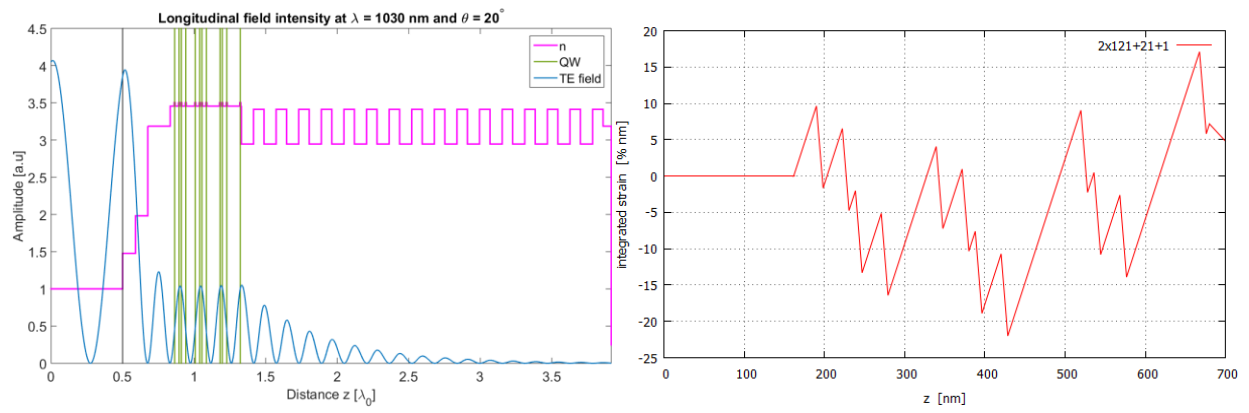


Figure 9. VECSEL structure layout (left) and its integrated strain profile (right), The position $z=0$ is the start of the barrier layers. The graph ends at the start of the DBR.

Table 1. Detailed structure design of a 1030nm MQW VECSEL yielding 95fs pulses.

Object	Repeats	Thickness [nm]	Material	n _r (1030nm)	Strain [%]
substrate			GaAs	3.4949	
	1	200.0	In _{0.484} Ga _{0.516} P	3.1912	
	1	30.0	GaAs	3.4949	
etch stop	1	163	In _{0.484} Ga _{0.516} P	3.1912	
barrier	1	28.3	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	24.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	8.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	24.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	60.1	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	24.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	8.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	24.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	91.1	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	8.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	24.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	90.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
well	1	8.35	In _{0.19} Ga _{0.81} As	3.5108	-1.352
barrier	1	4.0	GaAs _{0.905} P _{0.095}	3.4587	0.341
		88.19	AlAs	2.9484	-0.119
DBR	16	76.40	Al _{0.175} Ga _{0.825} As	3.4031	
phase-match	1	64.00	In _{0.484} Ga _{0.516} P	3.1912	0.000
metallization			Au/Pt/Au/In		

SATURABLE ABSORBER STRUCTURES

The pulse duration achievable with a modelocked VECSEL is not only dependent on the VECSEL structure design but is also greatly influenced by the properties of the saturable absorber. The most important parameters to consider for the generation of sub-100 fs laser pulses are: the carrier relaxation rate, the saturation fluence, the saturable losses and the dispersion of the structure. Simulations of pulse formation dynamics at OSC showed to which extent each parameter could affect the outcome of the pulse formation [4,5,6]. Commercially available SESAM (Batop) are generally not matching our stringent requirements and result in very long pulse durations. Therefore, OSC designed SESAMs that meet the needs, in particular in term of ultrafast recovery dynamic. The strategy employed for the fast recombination of excited carriers is to place the absorbing QW very close to the surface of the structure (3 to 7nm), allowing the carrier to tunnel through the surface and rapidly recombine on the numerous surface states (near-surface (GaIn)As-QWH configuration). The saturation fluence is then tailored by positioning the QW on a specific location of the standing wave of the field. This is done by adjusting the thickness of the GaAs spacer layer separating the QW and the DBR. Finally, the dispersion of the structure is carefully optimized, taking into account the angle of incidence of the field on the SESAM, by precisely centering the stop-band of the DBR on the lasing wavelength and by applying a very accurate dielectric coating on the surface (generally Ta₂O₅). This way, a large optical spectrum (>10nm) benefit from the highest reflectivity of the DBR and the lowest third order dispersion. Additionally, the coating thickness was slightly adjusted to account for the dispersion and the number of pass per round trip on each element of the cavity, in particular the VECSEL structure. The goal is to have a total GDD with a very broadband flat spectrum and a near zero value.

Multiple design-fabrication iterations were necessary to obtain ultrafast SESAM structure supporting sub-100fs pulses. In this joint research project, OSC has designed, realized and tested SESAM structures for emission wavelengths of 865nm, 980nm, 1030nm and 1040nm. Most of these structures were fabricated by molecular beam epitaxy (MBE) in the group of Dr. Ganesh Balakrishnan at the University of New Mexico (UNM). A few SESAM structures were also fabricated by MOVPE at UMR for wavelengths of 980nm and 1030nm in order to compare the influence of the growth method on the SESAM performance, in particular the long-term degradation observed in the sub-300fs range. For this purpose, we at UMR also realized a specific surface passivation of the thin GaAs-cap layer that was applied after growth, transferring part of this GaAs-layer into a stable Ga-oxide layer. Work is still ongoing to verify if this leads to an improvement of the SESAM lifetime. However, with an ever reducing pulsewidth down to the 100fs range, with ever increasing pulse powers, a significant degradation in SESAM-performance is still detected.

This characteristic has led to the development of a so-called bottom-absorber SESAM-design, where the actual epitaxial layer stack is grown in reverse order on a specific (GaInP-based etch-stop layer at UMR. In this way the SESAM-layer stack is directly mounted on a diamond heat spreader with subsequent etching-off of the entire substrate. This leads to a significant improvement in heat extraction from the laser spot on the SESAM layer stack. In addition, different (AlGa)As/GaAs-mirror designs have been realized in order to specifically influence the non-saturable losses of the SESAM part of the short pulse laser set-up.

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