Advanced GeSn/SiGeSn Group IV Heterostructure Lasers
Nils von den Driesch, Daniela Stange, Denis Rainko, Ivan Povstugar, Peter Zaumseil, Giovanni Capellini, Thomas Schröder, Peter Denneulin, Zoran Ikonic, Jean-Michel Hartmann, et al.

To cite this version:
Nils von den Driesch, Daniela Stange, Denis Rainko, Ivan Povstugar, Peter Zaumseil, et al.. Advanced GeSn/SiGeSn Group IV Heterostructure Lasers. Advanced Science, Wiley Open Access, 2018, 5 (6), pp.1700955. 10.1002/advs.201700955. cea-02186164

HAL Id: cea-02186164
https://hal-cea.archives-ouvertes.fr/cea-02186164
Submitted on 27 Aug 2019

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Advanced GeSn/SiGeSn Group IV Heterostructure Lasers

Nils von den Driesch,* Daniela Stange, Denis Rainko, Ivan Povstugar, Peter Zaumseil, Giovanni Capellini, Thomas Schröder, Thibaud Denneulin, Zoran Ikonic, Jean-Michel Hartmann, Hans Sigg, Siegfried Mantl, Detlev Grützmacher, and Dan Buca*

Growth and characterization of advanced group IV semiconductor materials with CMOS-compatible applications are demonstrated, both in photonics. The investigated GeSn/SiGeSn heterostructures combine direct bandgap GeSn active layers with indirect gap ternary SiGeSn claddings, a design proven its worth already decades ago in the III–V material system. Different types of double heterostructures and multi-quantum wells (MQWs) are epitaxially grown with varying well thicknesses and barriers. The retaining high material quality of those complex structures is probed by advanced characterization methods, such as atom probe tomography and dark-field electron holography to extract composition parameters and strain, used further for band structure calculations. Special emphasis is put on the impact of carrier confinement and quantization effects, evaluated by photoluminescence and validated by theoretical calculations. As shown, particularly MQW heterostructures promise the highest potential for efficient next generation complementary metal-oxide-semiconductor (CMOS)-compatible group IV lasers.

The ever-increasing demand for computational power, e.g., in data centers or consumer electronics, requires new approaches tackling energy efficiency of computer chips. Optical short-range and on-chip communication may here offer dramatically reduced power dissipation.[1] In this respect, Group IV photonics is the key technology,[2,3] allowing seamless integration of complementary metal-oxide-semiconductor (CMOS)-based electronics with optical components.[4,5] Since the indirect nature of their bandgap hinders Si and Ge from acting as efficient light emitters, heterogeneous integration[6] and epitaxy of III–V materials on Si are the solutions presently investigated, yielding optically[7] and electrically driven[8,9] lasers. Recently, solutions based on pure group IV alloys got renewed interest as incorporation of group IV element tin (Sn) in Ge and SiGe has led to (i) the formation of truly Si-compatible, direct gap GeSn[10] and SiGeSn[11] systems and, (ii) several demonstrations of GeSn optically pumped lasing.[10,12–14] Moreover, their bandgap in the mid-infrared range turns them into key materials for biological and chemical sensing applications.[3]

Electrically induced luminescence has been, however, so far demonstrated only for homojunction diodes and/or for indirect gap GeSn structures.[15–18]

Lessons learned from highly mature III–V laser materials can serve as a model for developing more efficient group IV
Epitaxial growth and characterization were, however, ternaries has been theoretically proposed for energy efficient heterostructures and quantum wells combining binaries and properties shall be strongly enhanced in the active region. \[28\] If the valley, later termed directness, emission \(L\)- and the direct properties increase the energy difference between the indirect \(\Gamma\)- and \(X\)-valley, respectively. Hence, the complete SiGeSn/GaAsSn DHS stack was grown at 350 °C. Thus, the main difference between the two structures is the stoichiometry of the bottom SiGeSn layers, which was extracted to a Si/Ge incorporation of 5.5/11.5 and 5.5/11.5 at% for DHS I and DHS2, respectively. Details can be found in the Experimental Section and Table 1. For DHS I, this difference leads to better carrier confinement both for \(L\)- and especially for \(\Gamma\)-electrons (110 meV for DHS1 vs 47 meV DHS2), which is shown later in Figure 3a.

X-ray diffraction reciprocal space maps (XRD-RSMs) indicate that the Ge\(_{0.85}\)Sn\(_{0.15}\) buffers are partly relaxed with a residual compressive strain of only \(-0.4\%\) (shown for DHS1 in Figure 1a). Both cladding layers are, if at all, hardly distinguishable in the RSM. The bottom SiGeSn cladding layer's signal largely superimposes the one from the GeSn buffer. The top SiGeSn layer is grown coherently on top of the active GeSn, and can be linked to a signal with the same in-plane reciprocal lattice constant \(Q_x\) (see Figure 1a). The active Ge\(_{0.855}\)Sn\(_{0.145}\) layer in DHS1 and Ge\(_{0.80}\)Sn\(_{0.14}\) layer in DHS2 are partly relaxed with respect to the Ge\(_{0.80}\)Sn\(_{0.20}\) buffers, being under compressive strain of \(-0.56\%\) and \(-0.70\%\), respectively. Transmission electron microscopy (TEM) analysis (Figure 1b) indicates the formation of additional unwanted misfit dislocations at the bottom SiGeSn/active GeSn interface, some of which are marked by white arrows. Apart from that, very homogeneous incorporation of Sn atoms throughout the structure is evidenced by the overlaid secondary ion mass spectrometry (SIMS) depth profile, showing that Si atoms are present only in the cladding regions.

Microdisk lasers were fabricated according to the procedure reported in ref. \[12\] to study the influence of confinement and misfit dislocations on lasing characteristics, such as threshold and maximum operating temperature. Both the heterostructures show lasing under 1550 nm wavelength optical pumping at 20 K, as depicted in Figure 1c,d in comparison to PL. The difference in PL emission wavelength of both the structures can be attributed to small Sn variations, while the shift of lasing emission is a consequence of both band filling (blueshift) and layer relaxation in underetched microdisks (redshift).

Table 1. Overview on the main structural properties of the grown heterostructures. Except for the bulk GeSn layer, all heterostructure samples feature a 200 nm thick partially relaxed Ge\(_{0.9}\)Sn\(_{0.1}\) buffer.

| Name  | Sn content in active region \(^a\) | GeSn active region \(^b\) thickness [nm] | Si/GeSn content in top cladding [at%] | Si/GeSn content in bottom cladding [at%] | Si/GeSn content in barrier [at%] | Barrier thickness [nm] |
|-------|----------------------------------|------------------------------------------|---------------------------------------|----------------------------------------|-------------------------------|----------------------|
| DHS1  | 14.5                             | 377                                      | 4.5/14.0                              | 5.5/11.5                               | –                             | –                    |
| DHS2  | 14.0                             | 342                                      | 5.0/13.5                              | 5.5/13                                 | –                             | –                    |
| MQW1  | 13.3                             | 10 × 22                                  | –                                     | –                                      | 4.8/13.0                     | 22                   |
| MQW2  | 13.5                             | 10 × 12                                  | –                                     | –                                      | 5.2/13.4                     | 16                   |
| Bulk GeSn | 12.5                           | 414                                      | –                                     | –                                      | –                             | –                    |

\(^a\)Well regions in case of the MQW structures.
This first demonstration of optically pumped lasing in group IV heterostructures discloses two important findings. First and foremost, lasing is achieved with SiGeSn ternaries as barriers to form heterostructures. As shown in Figure 1e, however, lasing ceases at temperatures above 120 K, where the linearity of the light-in light-out (LL) curve throughout all pumping levels can be taken as a sign of spontaneous emission dominated luminescence. This is linked to increased losses from nonradiative recombination through defects or Γ–L intervalley scattering, which cannot be overcome by the material gain anymore.

In comparison to laser structures reported earlier in literature,[12,14] introduction of different SiGeSn claddings for carrier confinement does not result in improved lasing thresholds. Their theoretical advantage seems to be shadowed by a common drawback of both structures: a newly formed misfit dislocation network at the GeSn/SiGeSn interface, making the structures behave similar to bulk GeSn layers. This major drawback can be overcome by using strain-adjusted heterostructures, such as GeSn/SiGeSn multi-quantum wells (MQWs), as we show in the following text.

Two such heterostructures, MQW1 and MQW2 (see Table 1), each containing ten periods of GeSn wells and SiGeSn barriers, were grown on top of a partially relaxed Ge_{0.9}Sn_{0.1} buffer, similar to the DHS samples. XRD and depth-calibrated SIMS measurements (not shown here) yield well thicknesses of 22 and 12 nm, respectively. Due to the complexity of the MQW stack, XRD measurements can deliver only average compositional information of the superlattices. Instead, high resolution elemental analysis was performed by means of atom probe tomography (APT). The reconstructed Si atom positions for MQW1 are shown in Figure 2a. Si atoms are strictly confined within the SiGeSn barriers, with only 0.03 at% found in the well region. Elemental profiles (Figure 2b) passing across six central wells, indicate a Sn content of 13.3 at% in the wells, while Si and Sn incorporation of 4.8 and 13.0 at%, respectively, are extracted for the SiGeSn barriers.
The distinct advantage of the MQW structure is revealed by dark-field electron holography measurements (MQW2 shown here). Due to its high spatial resolution of about 6 nm, in- and out-of-plane deformations can be locally resolved. Colored lattice deformation maps reflect lattice strain differences for out-of-plane (Figure 2c) and in-plane (Figure 2d) directions. The constant in-plane deformation, obvious in the line scans through buffer and MQW in Figure 2e, proves coherent growth of the MQW on top of the Ge\textsubscript{0.9}Sn\textsubscript{0.1} buffer. This should greatly enhance light emission, as the nonradiative recombination of carriers at the bottom interface with the buffer can be eliminated.\textsuperscript{[32]}

In Figure 3a–c, the calculated conduction band energies in DHS1, DHS2, MQW1, and MQW2 heterostructures are shown. Solid lines represent the lowest bulk conduction band energy, and dotted lines denote the first quantized electron state at Γ. For easier comparison, the displayed conduction band and state energies are measured from the top valence band energy, which in case of the MQW structures is the first quantized heavy hole state. Normalized low temperature PL is juxtaposed in Figure 3d–f to illustrate the effect of quantum confinement in the Ge\textsubscript{0.867}Sn\textsubscript{0.133} wells. For DHS2, the predicted bandgap of 0.474 eV closely fits PL emission at about 0.485 eV. A large blueshift of PL emission for the MQW heterostructure with 22 nm well thickness can be observed Figure 3e, which is attributed to a slightly smaller Sn incorporation (13.3 at% instead of 14 at% Sn) and to level splitting because of quantization. The additional blueshift of 20 meV for the MQW2 sample with 12 nm thin wells (Figure 3f) is solely a quantum confinement effect, as indicated by band structure calculations. Thus, clear emission from quantized states is observed in the MQW heterostructures.

Temperature-dependent PL emission between 4 K and room temperature offers additional information on carrier confinement. As shown in Figure 4a,b, DHS1 shows only a marginally increased PL signal at cryogenic temperatures compared to a 400 nm bulk GeSn layer with 12.5 at% Sn. A different picture emerges for the 22 nm MQW heterostructure (MQW1) in Figure 4c. Coherent growth leads to a strong increase of PL at low temperatures (an order of magnitude at 4 K). This distinct advantage is expected to result from both the confinement of carriers in the GeSn wells and their spatial separation from the GeSn/Ge–VS defective interface. Such a heterostructure is expected to improve lasing performance, such as the lasing threshold, which will be discussed in more details elsewhere. However, too strong quantization, as present in MQW2, diminishes the energy difference between Γ- and L-valleys due to the smaller effective mass of Γ-electrons in comparison to L.\textsuperscript{[16]}

In this case, the decreased directness (and even transition to indirect semiconductor for thinnest wells) leads to poor light emission (98% less PL emission at 4 K compared to MQW1).

The above described PL effects are quantified in a logarithmic plot of the integrated PL intensity in Figure 4d. The first observation is that DHS1 yields no light emission improvement over a bulk GeSn sample (see Table 1), as both are limited by the above mentioned misfit dislocation network at the GeSn/SiGeSn interface. In contrast, MQW1 shows the strongest PL emission of the investigated samples, at least at cryogenic temperatures, hinting to a considerably increased nonradiative lifetime. The particular advantage of MQW1 in terms of light emission, however, disappears for temperatures above \(\approx 150 \text{ K}\). This can be related to the relatively low conduction band offset between wells and barriers (13 meV determined by band structure calculations). Above this temperature, carriers are able to overcome the energetic barriers and nonradiative recombination at the GeSn/Ge interface sets in. In fact, at room temperature, the integrated MQW1 PL emission intensity is below the one of DHS1 and the bulk GeSn sample, which we attribute to the thinner optically active layers. Therefore, we can deduce that coherent growth, i.e., epitaxy schemes avoiding the formation of additional misfit dislocations, is the most vital pathway toward further improvement of the optical properties.
**Figure 3.** Conduction band alignment (and the lowest quantized states positions) for a) DHS1 and DHS2 and MQW samples with b) 22 nm well thickness (MQW1) and c) 12 nm well thickness (MQW2) at 4 K. d–f) The corresponding PL emission illustrates a peak shift, in line with bandgap changes.

**Figure 4.** Temperature-dependent PL emission for a) bulk GeSn, b) a double heterostructure, and c) a multi-quantum well with 22 nm well thickness. d) Integrated PL intensity shows the strongest emission from MQW1 at cryogenic temperatures.
For example, growth of thinner active regions and/or optimized GeSn buffers will be developed. Moreover, to effectively screen carriers from nonradiative recombination sinks at the bottom GeSn/Ge interface, we consider growing SiGeSn barriers with higher Si content. In that case, we expect a distinct advantage of MQW heterostructures, compared to double heterostructures and even more bulk GeSn layers. These features may allow room temperature, low threshold lasers made entirely from group IV materials.

In summary, we demonstrated the potential of group IV epitaxy for the fabrication of direct bandgap heterostructures. Different designs, from bulk to double heterostructures and multi-quantum wells, were epitaxially grown and characterized. A high sample quality is proven by the first demonstration of optically pumped lasing from group IV GeSn/SiGeSn heterostructures. For the MQW heterostructures, a shift in light emission is achieved by controlling the quantum confinement of carriers through varying well thicknesses. But most importantly, this type of structure outperforms double heterostructures, because of the spatial separation of misfit dislocations from the active region. We indicated that by increasing the heterostructure barriers, the favorable MQW emission properties can be extended to higher temperatures, possibly up to room temperature. In conclusion, we believe that, as demonstrated decades ago already for the III–V material system, group IV heterostructures and multi-quantum wells are potential enablers for high efficient integrated light emitters.

**Experimental Section**

**Epitaxy:** All investigated layers were produced by means of chemical vapor deposition (CVD) in an industry-compatible reactor design on 200 mm wafers. The employed precursors disilane (Si₂H₆), digermane (Ge₂H₄), and tin tetrachloride (SnCl₄) ensure high growth rates (typically thick Ge–VS.

**Material Characterization:** The local material parameters inside the complex heterostructures were determined by a large number of techniques. For the MQWs, exact compositions were obtained using APT, while energy-dispersive X-ray spectroscopy (EDX) mapping ensured precise values for the DHS. Layer thicknesses were determined likewise from EDX and validated from XRD (MQW) and SIMS (DHS). Strain values were obtained from XRD reciprocal space maps and validated by dark-field electron holography maps.

**Band structure values at critical energies were obtained at 0 K by the empirical pseudopotential method and appropriate strain correction within an 8-band k·p framework. The effect of temperature was included via Varshni’s empirical relations. All theoretical parameters, required for modeling the band structure, can be found in ref. [27] and the Supporting Information of ref. [10].

**Acknowledgements**

This research received funding for CVD growth investigations from the Federal Ministry of Education and Research (BMBF) under project UltraLowPow (16ES0060 K). The authors further acknowledge financial support by the BMBF in frame of the M-ERA.NET project CESNAPHOTO, support by the German Research Foundation (DFG) through the project SiGeSn Laser for Silicon Photonics, and from The Royal Society International Exchanges grant IE131593.

**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**

GeSn, heterostructures, lasers, multi-quantum wells, SiGeSn

Received: December 1, 2017
Revised: February 25, 2018
Published online: March 27, 2018

[1] D. A. B. Miller, Proc. IEEE 2000, 88, 728.
[2] R. Soref, D. Buca, S.-Q. Yu, Opt. Photonics News 2016, 27, 32.
[3] R. Soref, Nat. Photonics 2010, 4, 495.
[4] J. J. Ackert, D. J. Thomson, L. Shen, A. C. Peacock, P. E. Jessop, G. T. Reed, G. Z. Mashanovich, A. P. Knights, Nat. Photonics 2015, 9, 393.
[5] C. Sun, M. T. Wade, Y. Lee, J. S. Orcutt, L. Alloatti, M. S. Georgas, A. S. Waterman, J. M. Shainline, R. R. Avizienis, S. Lin, B. R. Moss, R. Kumar, F. Pavanello, A. H. Atabaki, H. M. Cook, A. J. Ou, J. C. Leu, Y.-H. Chen, K. Asanović, J. R. Ram, M. A. Popović, V. M. Stojanović, Nature 2015, 528, 534.
[6] G. Morthier, G. Roelkens, D. Van Thourhout, in Compact Semiconductor Lasers (Eds. R. DeLa Rue, J.-M. Lourtioz, S. Yu), Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany 2014, pp. 195–230.
[7] B. Tian, Z. Wang, M. Pantouvaki, P. Absil, J. Van Campenhout, C. Merckling, D. Van Thourhout, Nano Lett. 2017, 17, 559.
[8] Y. Sun, K. Zhou, Q. Sun, J. Liu, M. Feng, Z. Li, Y. Zhou, L. Zhang, D. Li, S. Zhang, M. Ikeda, S. Liu, H. Yang, Nat. Photonics 2016, 10, 595.
[9] S. Chen, W. Li, J. Wu, Q. Jiang, M. Tang, S. Shutts, N. E. Sutin, A. Sobiesiarski, A. J. Seeds, I. Ross, P. M. Smowton, H. Liu, Nat. Photonics 2016, 10, 307.
[10] S. Wirths, R. Geiger, N. von den Driesch, G. Mussler, T. Stoica, S. Mantl, Z. Ikonić, M. Luysberg, S. Chiussi, J.-M. Hartmann, H. Sigg, J. Faist, D. Buca, D. Grützmacher, Nat. Photonics 2015, 9, 88.
[11] N. von den Driesch, D. Stange, S. Wirths, D. Rainko, I. Povstugar, A. Savenko, U. Breuer, R. Geiger, H. Sigg, Z. Ikonić, J.-M. Hartmann, D. Grützmacher, S. Mantl, D. Buca, Small 2017, 13, 160321.
[12] D. Stange, S. Wirths, R. Geiger, C. Schulte-Braucks, B. Marzban, N. von den Driesch, G. Mussler, T. Zabel, T. Stoica, J.-M. Hartmann, S. Mantl, Z. Ikonić, D. Grützmacher, H. Sigg, J. Witzen, D. Buca, ACS Photonics 2016, 3, 1279.
[13] V. Reubod, A. Gassenq, N. Pauc, J. Aubin, L. Milord, Q. M. Thai, M. Bertrand, K. Guillou, D. Rouchon, J. Rothman, T. Zabel, F. Armand Pilon, H. Sigg, A. Chelnokov, J. M. Hartmann, V. Calvo, Appl. Phys. Lett. 2017, 111, 92101.
[14] J. Margetis, S. Al-Kabi, W. Du, W. Dou, Y. Zhou, T. Pham, P. Grant, S. Ghetmiri, A. Mosleh, B. L. Liu, G. Sun, R. Soref, J. Tolle, M. Mortazavi, S.-Q. Yu, ACS Photonics 2017, https://doi.org/10.1021/acsphotonics.7b00938.
[15] J. D. Gallagher, C. L. Senaratne, C. Xu, P. Sims, T. Aoki, J. D. Smith, J. Menéndez, J. Kouvetakis, J. Appl. Phys. 2015, 117, 245704.
[16] D. Stange, N. von den Driesch, D. Rainko, C. Schulte-Braucks, S. Wirths, G. Mussler, A. T. Tiedemann, T. Stoica, J.-M. Hartmann, Z. Ikonic, S. Mantl, D. Grützmacher, D. Buca, Opt. Express 2016, 24, 1358.

[17] B. Schwartz, M. Oehme, K. Kostecki, D. Widmann, M. Gollhofer, R. Koerner, S. Bechler, I. A. Fischer, T. Wendav, E. Kasper, J. Schulze, M. Kittler, Opt. Lett. 2015, 40, 3209.

[18] Y. Zhou, W. Dou, W. Du, T. Pham, S. A. Ghettmri, S. Al-Kabi, A. Mosleh, M. Alher, J. Margetis, J. Tolle, G. Sun, R. Soref, B. Li, M. Mortazavi, H. Naseem, S.-Q. Yu, J. Appl. Phys. 2016, 120, 23102.

[19] Z. Alferov, IEEE J. Sel. Top. Quantum Electron. 2000, 6, 832.

[20] G. Sun, R. A. Soref, H. H. Cheng, Opt. Express 2010, 18, 19957.

[21] P. Moontragoon, R. A. Soref, Z. Ikonic, J. Appl. Phys. 2012, 112, 73106.

[22] S. Wirths, A. T. Tiedemann, Z. Ikonic, P. Harrison, B. Holländer, T. Stoica, G. Mussler, M. Myronov, J.-M. Hartmann, D. Grützmacher, D. Buca, S. Mantl, Appl. Phys. Lett. 2013, 102, 192103.

[23] A. Attiaoui, S. Wirths, A.-P. Blanchard-Dionne, M. Meunier, J. M. Hartmann, D. Buca, O. Moutanabbir, arXiv ID: 1702.00682 2017.

[24] C. Xu, R. T. Beeler, L. Jiang, J. D. Gallagher, R. Favaro, J. Menéndez, J. Kouvetakis, Thin Solid Films 2014, 557, 177.

[25] S. Wirths, D. Buca, Z. Ikonic, P. Harrison, A. T. Tiedemann, B. Holländer, T. Stoica, G. Mussler, U. Breuer, J.-M. Hartmann, D. Grützmacher, S. Mantl, Thin Solid Films 2014, 557, 183.

[26] S. A. Ghettmri, Y. Zhou, J. Margetis, S. Al-Kabi, W. Dou, A. Mosleh, W. Du, A. Kuchuk, J. Liu, G. Sun, R. A. Soref, J. Tolle, H. A. Naseem, B. Li, M. Mortazavi, S.-Q. Yu, Opt. Lett. 2017, 42, 387.

[27] D. Stange, N. von den Driesch, D. Rainko, S. Roegsgaard, I. Povstugar, J.-M. Hartmann, T. Stoica, Z. Ikonic, S. Mantl, D. Grützmacher, D. Buca, Optica 2017, 4, 185.

[28] D. Stange, S. Wirths, N. von den Driesch, G. Mussler, T. Stoica, Z. Ikonic, J. M. Hartmann, S. Mantl, D. Grützmacher, D. Buca, ACS Photonics 2015, 2, 1539.

[29] N. von den Driesch, D. Stange, S. Wirths, G. Mussler, B. Holländer, Z. Ikonic, J. M. Hartmann, T. Stoica, S. Mantl, D. Grützmacher, D. Buca, Chem. Mater. 2015, 27, 4693.

[30] R. Chen, Y.-C. Huang, S. Gupta, A. C. Lin, E. Sanchez, Y. Kim, K. C. Saraswat, T. I. Kamins, J. S. Harris, J. Cryst. Growth 2013, 365, 29.

[31] S. Takeuchi, A. Sakai, O. Nakatsuka, M. Ogawa, S. Zaima, Thin Solid Films 2008, 517, 159.

[32] F. Pezzoli, A. Giorgioni, D. Patchett, M. Myronov, ACS Photonics 2016, 3, 2004.

[33] J. M. Hartmann, A. Abbade, N. Cherkashin, H. Grampeix, L. Clavelier, Semicond. Sci. Technol. 2009, 24, 55002.