Electrical tuning of the oscillator strength in type II InAs/GaInSb quantum wells for active region of passively mode-locked interband cascade lasers

Mateusz Dyksik, Marcin Motyka, Marcin Kurka, Krzysztof Ryczko, Jan Misiewicz, Anne Schade, Martin Kamp, Sven Höfling, and Grzegorz Sek

1Laboratory for Optical Spectroscopy of Nanostructures, Department of Experimental Physics, Faculty of Fundamental Problems of Technology, Wrocław University of Science and Technology, Wrocław 50-370, Poland
2Technische Physik, University of Würzburg and Wilhelm-Conrad-Röntgen-Research Center for Complex Material Systems, Würzburg D-97074, Germany
3SUPA, School of Physics and Astronomy, University of St. Andrews, North Haugh, St. Andrews KY16 9SS, U.K.

Received June 14, 2017; accepted August 14, 2017; published online September 29, 2017

Two designs of active region for an interband cascade laser, based on double or triple GaInSb/InAs type II quantum wells (QWs), were compared with respect to passive mode-locked operation in the mid-infrared range around 4 μm. The layer structure and electron and hole wavefunctions under external electric field were engineered to allow controlling the optical transition oscillator strength and the resulting lifetimes. As a result, the investigated structures can mimic absorber-like and gain-like sections of a mode-locked device when properly polarized with opposite bias. A significantly larger oscillator strength tuning range for triple QWs was experimentally verified by Fourier-transform photoreflectance.

Interband cascade lasers (ICLs) have been proven to be reliable laser sources in the mid-infrared spectral range, providing room temperature, continuous wave (cw), single mode operation up to 7 μm and low threshold current densities below 100 A cm−2. Their low power consumption and cw operation allowed for application in compact gas sensors. Tunable diode laser absorption spectroscopy of ethanol, formaldehyde, or nitric oxide have been already demonstrated. Such sensors require an ICL with a peak emission matched to the absorption line of a given harmful or toxic substance. In principle, the respective monomode device might be electrically or thermally tuned over several nanometers, nevertheless, usually only one substance can be detected with a single device. However, there is a growing demand for a robust and compact mid-infrared spectrometer capable of detecting multiple species at once.

Recently, a multi-mode ICL has been employed in a multi-mode absorption spectroscopy (MUMAS) technique, resulting in a possibility of detecting several species simultaneously. Although this approach offers fast response times while detecting known species, an extensive data processing of the detected signal is required for sensing of unknown gases. Also, the minimum detection limit (e.g., 70 ppm for methane) might be considered insufficient for specific applications. Multi-mode Fabry–Perot ICLs also show the feasibility of employment in dual-comb spectroscopic technique utilizing two optical frequency combs with slightly different repetition rates. Such multi-heterodyne approach allows for multi-species detection with very short acquisition times, and orders of magnitude higher spectral resolution comparing to MUMAS. Recently, a dual-comb spectrometer has been implemented in the mid-infrared spectral region by means of an actively mode-locked quantum cascade laser (QCL), allowing for broadband measurements with a resolution of 0.0027 cm−1 around 7 μm, whilst with passively mode-locked lasers resolution below 0.001 cm−1 can be achieved.

Although QCLs have become standard semiconductor laser sources in the mid-infrared above 4 μm, the possibility of passive mode-locked operation in the saturable absorber scheme is questionable due to very short upper state lifetimes. In contrast, ICLs might be considered as an alternative since they exhibit relatively long (nanoseconds) upper state lifetimes, operate in cw mode up to 7 μm at temperatures above ambient, have much lower threshold current densities than QCLs, and still produce cw output power exceeding 500 mW. In the only communication reporting on the mode-locked operation of an ICL there has been used a common active region without any optimization and employed a post-growth ion bombardment to tailor the losses and reduce the recovery time in the absorber section.

In this work we present experimental verification of an ICL’s active region which is better suited for passive mode-locked operation than the commonly used InAs/GaInSb W-shaped type II quantum wells (QW). Our approach assumes the application of a fast saturable absorber (τa) scheme, with recovery time significantly faster than the pulse duration (τp), τa ≪ τp. To realize that the semiconductor laser is divided into two parts with a common bottom contact. As a result, two sections are present, the gain and absorber, and both might be biased individually. Throughout this report the impact of external electric field on the oscillator strength (OS) of optical transitions will be studied. Since the assumption is to have significantly larger optical transition rates in the absorber section than in the gain section, the aim is to probe the active region design exhibiting large difference in the OS when the bias is reversed (the respective carrier lifetimes are inversely proportional to the OS). Two designs, based on asymmetric type II QW, will be compared. The first studied design utilizes a standard W-shaped band alignment, whereas the second one benefits from a triple QW design. Preliminary design considerations of similar structures with respect to the passive mode-locking have been recently published. Based on these results two well-optimized designs with respect to the oscillator strength tuning by external electric field were selected for fabrication, processing and optical study. Our approach to optimize...
the oscillator strength in order to tailor the losses is supported by the recent presentation of GaSb-based laser diode emitting at 2.1 µm, where the type I QW active region, non-optimized with respect to the oscillator strength at opposite bias, was employed in two-section device and passive mode-locking was achieved. Hereby, we make an important step forward which is the first practical realization and experimental verification of the modified ICLS’s active regions with respect to the use in a mode-lock operation scheme.

The studied samples were grown on an n-type GaSb substrate in a solid source molecular beam epitaxy system equipped with valved cracker cells for both antimony and arsenic. Directly on the substrate a 100-nm-thick 3 × 10¹⁸ cm⁻³ n-doped buffer layer was grown and followed by a 100-nm thick undoped GaSb buffer preventing diffusion of doping atoms. The core part of the first sample was designed as the “W-like” shape quantum well (therefore called further W-QW), consisting of two InAs layers with intentionally slightly different thicknesses [see Fig. 1(a)] and one GaInSb layer in-between. The second sample had three InAs wells for confinement of electrons and two GaInSb layers confining holes (hence named T-QW). Each QW in both samples is surrounded by 2.5-nm-thick AlSb barriers. In order to enhance the overall optical response, the wells have been repeated five times and separated from each other by 20 nm of GaSb. Both structures are terminated with a 100-nm-thick, 3 × 10¹⁸ cm⁻³ doped, GaSb cap layer. The band alignment of the core part of the W-QW and T-QW samples is presented in Figs. 1(a) and 1(b), respectively. The buffer and cap layers were Te-doped in order to allow for application of external electric field. The n-type backside contact formed of AuGe, Ni and Au was vapor-deposited and alloyed at 300°C for 30 s. The p-type top surface contact was formed as a 980 × 4400 µm² rectangular shape mesa of vapor-deposited Ti, Pt, and Au.

Photoreflectance (PR), as an absorption-like technique, was employed in order to determine the OS of optical transitions. For each value of the bias voltage a PR spectrum was measured. For that purpose a Fourier-transform spectrometer Bruker Vertex 80v, operated in the step-scan mode, was utilized with a liquid-nitrogen cooled InSb photodetector and a KBr beamsplitter. In order to reduce the absorption by ambient gasses the spectrometer was evacuated to the pressure of 1.2 hPa. The pump beam was provided by the 640 nm line of a 60 mW semiconductor laser diode, which was mechanically chopped at the frequency of 1200 Hz. At this point it is worth mentioning that the used illumination intensities are such that the photovoltage generated due to the PR pump laser beam, which could affect the electric field, is negligible (1% of the externally generated bias in the active region), whereas the photovoltage due to the broadband glowbar lamp was not observed at all. The phase sensitive detection of the optical response was performed using a lock-in amplifier. The experimental and theoretical considerations presented below were performed at room temperature as the best representative for the conditions of operational device.

The photoreflectance studies were supported by electronic structure modeling by means of k·p theory using the 8 × 8 k·p Hamiltonian defined for the [001] growth direction and including strain effects. The carriers’ wavefunctions and subband energies were determined by numerically solving the Schrödinger equation and employing the finite difference method. Supplementary simulations were performed in order to test the impact of the sample temperature on the band structure and thus on the relative oscillator strength tuning dependence on the electric field, which however, appeared negligible. As a result, the analysis presented below was performed for data acquired at room temperature as corresponding to the conditions of the operational device. All the material parameters were taken from Ref. 23.

Figure 1 presents the band alignment for the W-QW (a) and T-QW (b) samples, together with the calculated lowest energy states and their respective probability densities. For both samples the electron probability density (red) is localized not only in the InAs wells but also significantly penetrates into the GaInSb barriers. The first sample is an asymmetric W-shaped quantum well, thus the electron probability density at zero bias is mostly localized in the wider well. Since the T-QW exhibits an asymmetry in the GaInSb wells for confinement of holes as well (2.4- and 3.3-nm-thick layers, respectively), at zero bias the two lowest energy heavy hole-like states h1 and h2 are spatially separated in the adjacent wells—the probability density of both states is localized in each of the wells. These two states are also separated in energy by 35 meV. The presence of these two h1 and h2 levels in the valence band has been confirmed experimentally. Figures 2(a) and 2(b) present the results of PR measurements for both samples under investigation (open symbols). For W-QW [Fig. 2(a)] a transition between the heavy-hole and electron states is manifested by a singularity at around 0.3 eV, whereas the spectrum for T-QW [Fig. 2(b)] exhibits two spectral lines, separated by about 35 meV. According to our calculations [Fig. 1(b)] the low-energy transition at 0.27 eV corresponds to the absorption between h1 and electron states (e1h1), whereas the high-energy one is understood as the e1h2 transition. The integrated intensity of the PR signal corresponds directly to the OS of the observed optical transitions. In order to determine the OS, a modulus of the PR line shape has been derived according to the following expression:

\[
\frac{\Delta R}{R}(E) = \frac{C}{(E - E_0)^2 + \Gamma^2 m^{3/2}}
\]
where the transition energy $E_0$, intensity of the singularity $C$ and its broadening $\Gamma$ have been extracted from the fitting of the PR data with the common formula:\(^{24}\)

$$\frac{\Delta R}{R}(E) = \text{Re}[Ce^{-i\theta}(E - E_0 + i\Gamma)^{-m}]. \quad (2)$$

In Eq. (2) $\theta$ is a phase parameter, whereas $m$ refers to the type of the optical transition and the critical point in the Brillouin zone.\(^{25}\)

The respective PR moduli for each transition are presented in Figs. 2(c) and 2(d). The OS is obtained by calculating the area under the PR modulus spectrum. One can notice that for the T-QW sample the ground state transition has larger OS (by 40%) compared to the W-QW, which is due to increased overlap integral in the triple QW.\(^{16}\)

In order to determine if the designed samples are suitable for an active region of passively mode-locked ICLs we measured the PR spectra as a function of external electric field (employed in order to simulate the conditions occurring in an operational device). Figures 3(a) and 3(c) show the results of PR measurements for three values of electric field for samples W-QW and T-QW, respectively. The obtained data seem to meet the main design requirements.

A clear difference in the PR intensity is noticeable for opposite biases, thus reflecting also the changes in the OS. Figures 3(b) and 3(d) present the shift of confined states energies in the analyzed quantum systems due to the quantum confined Stark effect induced by the band realignment when the external electric field is reversed. The transition energies extracted from the fitting procedure and represented by symbols follow the theoretical predictions (solid lines). In contrast to a typical type I quantum well, where the introduction of external electric field leads to non-monotonic dependence of the transition energy, the studied systems with asymmetric band alignment exhibit an almost linear blue shift of the ground state transition for both samples W-QW (diamonds) and T-QW (triangles).

The OS for both samples was extracted from PR spectra in the range of electric fields from $-20$ to $20\,\text{kV/cm}$. Figure 4(a) summarizes the obtained values in terms of relative change of the OS, i.e.,

$$\frac{\Delta OS}{OS_{F=0}} = \frac{OS - OS_{F=0}}{OS_{F=0}} \quad (3)$$

as a function of the external electric field $F$. The T-QW exhibits larger difference in the relative OS for a given range of the external electric field. For instance, at forward bias of $10\,\text{kV/cm}$, when the investigated structures imitate the gain section of a mode-locked ICL, the T-QW sample exhibits 125% lower OS than in the zero bias conditions, while the sample W-QW shows only 45% reduction with respect to the OS at zero bias. Consequently, at reverse bias of $-10\,\text{kV/cm}$, where both samples are supposed to be the fast absorber
section, the T-QW has a relative OS of almost 70% whilst the W-QW exhibits only 15% of the values at zero field.

For both the structures under investigation the squared overlap integral, normalized to its value for the non-biased structure ($I/I_0$), was calculated as a function of external electric field. The simulation results are presented in Fig. 4(b) in the dash-dot region of moderate electric fields, from $-20$ to $20 \text{kV/cm}$. As in the experimental data, the simulated dependence for the T-QW exhibits a larger change of the squared overlap integral in the considered range of electric fields. The low electron effective mass and the narrow and shallow GaInSb barriers allow for the e1 electron state wavefunction to be spread across the three InAs wells and to penetrate significantly into the GaInSb barriers. When the structure is reverse biased, the maximal probability of finding an electron is transferred from the 2.4 nm GaInSb layer into the 3.3 nm one, greatly enhancing the overlap with the h1 hole wavefunction. Although the probability density of finding an electron in the 2.4 nm GaInSb layer decreases, at significant change of the squared overlap integral for electric field values below $-30 \text{kV/cm}$.

In conclusion, two different designs of the ICL active region structures emitting around 4 µm have been realized and investigated. We have shown that a triple quantum well structure offers an additional degree of freedom in engineering the electron and hole wavefunctions in order to obtain a large difference in the oscillator strength of the fundamental optical transition under external bias of opposite signs. This makes such a type II QW structure suitable as the absorber-like and gain-like sections of a passively modelocked interband cascade lasers and opens a pathway for practical realization of a dual comb spectrometer operating in the mid-infrared spectral range, based on ICL devices.

Acknowledgements This project has received funding from the European Commission’s Horizon 2020 Research and Innovation Programme (iCspec under grant agreement No. 636930) and has also been supported by the National Science Centre of Poland within Grant No. 2014/15/B/ST7/04663.

1) M. Dallner, F. Hau, S. Höfling, and M. Kamp, Appl. Phys. Lett. 106, 041108 (2015).
2) R. Weih, M. Kamp, and S. Höfling, Appl. Phys. Lett. 102, 231123 (2013).
3) L. Dong, C. Li, N. P. Sanchez, A. K. Gluszek, R. J. Gilllin, and F. K. Tittel, Appl. Phys. Lett. 108, 011106 (2016).
4) C. Li, L. Dong, C. Zheng, and F. K. Tittel, Sens. Actuators B 232, 188 (2016).
5) S. Lundqvist, P. Kluczyński, R. Weih, M. von Edlinger, L. Näble, M. Fischer, A. Bauer, S. Höfling, and J. Koeth, Appl. Opt. 51, 6009 (2012).
6) M. von Edlinger, J. Scheueremann, R. Weih, C. Zimmermann, L. Näble, M. Fischer, J. Koeth, S. Höfling, and M. Kamp, IEEE Photonics Technol. Lett. 26, 480 (2014).
7) M. von Edlinger, R. Weih, J. Scheueremann, L. Näble, M. Fischer, J. Koeth, M. Kamp, and S. Höfling, Appl. Phys. Lett. 109, 201109 (2016).
8) J. H. Northern, S. O’Hagan, B. Fletcher, B. Gras, P. Ewart, C. S. Kim, M. Kim, C. D. Merritt, W. W. Bewley, C. L. Canedy, J. Abell, I. Vurgaftman, and J. R. Meyer, Opt. Lett. 40, 4186 (2015).
9) C. L. Patrick, L. A. Sterczewski, J. Westberg, W. W. Bewley, C. D. Merritt, C. L. Canedy, S. Kim, M. Kim, I. Vurgaftman, J. R. Meyer, and G. Wysocki, Proc. SPIE 10123, 101231L (2017).
10) G. Villares, A. Hugi, S. Blaser, and J. Faist, Nat. Commun. 5, 5192 (2014).
11) J. Faist, G. Villares, G. Scalari, M. Rösch, C. Bonzoni, A. Hugi, and M. Beck, Nanophotonics 5, 272 (2016).
12) W. W. Bewley, J. R. Lindle, C. S. Kim, M. Kim, C. L. Canedy, I. Vurgaftman, and J. R. Meyer, Appl. Phys. Lett. 93, 041118 (2008).
13) C. L. Canedy, J. Abell, C. D. Merritt, W. W. Bewley, C. S. Kim, M. Kim, I. Vurgaftman, and J. R. Meyer, Opt. Express 22, 7702 (2014).
14) M. Bagheri, C. Frez, I. Vurgaftman, M. Fradet, C. L. Canedy, W. W. Bewley, C. D. Merritt, C. S. Kim, S. Forouhar, and J. R. Meyer, Proc. IEEE Photonics Conf., 2016, p. 82.
15) H. A. Haus, J. Appl. Phys. 46, 3049 (1975).
16) M. Motyka, K. Ryczko, M. Dyksik, G. Sęka, J. Misiewicz, R. Weih, M. Dallner, S. Höfling, and M. Kamp, J. Appl. Phys. 117, 084312 (2015).
17) Y. Jiang, L. Li, Z. Tian, H. Ye, L. Zhao, R. Q. Yang, T. D. Mishima, M. B. Santos, M. B. Johnson, and K. Mansour, J. Appl. Phys. 115, 113101 (2014).
18) K. Ryczko, J. Misiewicz, S. Höfling, M. Kamp, and G. Sęka, APJ Adv. 7, 015015 (2017).
19) K. Mergheim, R. Teissier, G. Aubin, A. M. Monakhov, A. Ramsdane, and A. N. Baranov, Appl. Phys. Lett. 107, 111109 (2015).
20) K. Ryczko, G. Sęka, and J. Misiewicz, J. Appl. Phys. 114, 223519 (2013).
21) G. L. Bir, E. G. Pikus, P. Shelnitz, and D. Louvish, Symmetry and Strain-Induced Effects in Semiconductors (Wiley, New York, 1976).
22) J. W. Thomas, Partial Differential Equations, Finite Difference Methods (Springer, New York, 1995).
23) I. Vurgaftman, J. R. Meyer, and L. R. Ram-Mohan, J. Appl. Phys. 89, 5815 (2001).
24) D. E. Aspnes, Surf. Sci. 37, 418 (1973).
25) F. H. Pollak, in Handbook of Semiconductors, ed. T. S. Moss (Elsevier, Amsterdam, 1994) Vol. 2, p. 527.