## 4.12 Report of Project Part 12: IR detection and emission by group IV nanostructures

## Detection and emission of infrared radiation based on group IV nanostructures

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### 4.12.1 Summary

Room temperature photoluminescence (PL) of randomly nucleated SiGe islands in a photonic crystal (PhC) and in microdisc resonators have been observed. Islands ordered with respect to the PhC have been grown, for details see P02. To avoid the necessity of active alignment, a set of two stamps for nanoimprint lithography has been designed with that perfect alignment in a large enough area (~200 × 200 µm$^2$) achievable via the Moiré effect. The necessary, homogeneous island growth of ordered islands over ~10 mm$^2$ large areas required for such an approach has been demonstrated. The careful analysis of island growth and a correlation with low-T PL emission resulted in a detailed understanding of the growth conditions required for perfect ordering and to avoid PL quenching. A comparison between excitation intensity dependent PL measurements on ordered and randomly nucleated SiGe islands revealed a distinctly different electronic structure of the ground states in these two island ensemble types. For randomly nucleated islands, it is dominated by size quantization within small areas of high Ge concentration at the island apexes. In ordered island ensembles, a homogeneous Ge concentration within the islands and within island ensembles results in rather narrow PL emission bands, the shift of which at high excitation intensity we ascribe to multi-excition interactions.

From small ensembles of ~10 SiGe islands, clear PL was observed. However, the PL spectra showed no indication of single islands contributing to the spectral shape but showed the same width as spectra measured on larger ensembles of 30 000 islands. We attribute this observation to the large required excitation intensity for observing the PL of 10 islands (~7 kW/cm$^2$).

With a newly implemented process, p-i-n diodes with a single SiGe island layer in the intrinsic region showing small saturation current and series resistance could be fabricated. With these diodes a strong photo-signal in the energy region below the Si bulk absorption could be observed. However, structures that unambiguously can be attributed to island inter band absorption are absent in these spectra.

On SiGe pseudo-substrate, a favorable new QWIP concept based on light-hole to light-hole (LH-LH) transitions has been demonstrated. Our new QWIPs show a richly structured response PC spectrum extending from the THz into the MIR region. From our experiments, we estimate a detectivity of at least $D^* = 5.7\times10^7$ cm Hz $^{0.5}$ / W at 7 THz for these new LH-LH QWIPs.

New blocked impurity detectors with a design optimized according to the results of the first IR-ON funding period were designed. Device processing is still in progress. For high accuracy characterization free of thermal background radiation, an experimental setup in that the sources of thermal radiation are shielded by cooled pinholes has been set up.

### 4.12.2 Scientific Background - State of the Art

Si based optoelectronics is a very active area of research as there is common agreement in the scientific community as well as in the electronics industry that optical interconnects will fulfill the main requirements of future Si based information technology (high data rate, large bandwidth, low loss) more likely than electrical ones (Miller 2009, Ohashi 2009, ITRS 2009). In particular, SiGe nanostructures are regarded as promising active building blocks for a Si based integrated optics platform (Tsybeskov 2009). The global research is mainly focused on the telecom wavelength range around 1.5 µm. However, as pointed out in (Soref 2010), Si based devices for larger wavelengths in the mid-infrared (MIR) to THz spectral range are required for example for integrated systems for medical and environmental sensing. Consequently, increasing efforts are targeted towards the development of a Si based integrated optics also for the MIR and THz wavelength range. Recently, large non-linear optical constants have been demonstrated in Si waveguides at wavelengths around 2 µm (Zlatanovic 2010, Liu 2010), leading the authors to the conclusion that Si is highly attractive for parametric light generation for MIR applications.

In this project we addressed the NIR, MIR and THz spectral ranges with our work on SiGe islands, SiGe quantum well photo detectors (QWIPs) and blocked impurity band (BIB) detectors. Via three dimensional SiGe island growth on Si substrate locally, the fundamental bandgap of the Si-SiGe island heterostructured can be reduced so that detection and emission of photons in telecom wavelength region becomes possible. Due to the three dimensional shape of the islands, the misfit strain between Ge and Si can be relaxed elastically, and coherent integration of large local Ge concentrations into a Si matrix becomes possible. For randomly nucleated SiGe islands, room temperature electro-luminescence (Chang 2003) and photocurrent (Tong 2002) in the telecom spectral range have been reported. By integration of the randomly nucleated island in resonator structures (photonic crystal microcavities, microdiscs), the room temperature PL and electroluminescence efficiency (Xia 2010, Xia 2009) was shown to be drastically enhanced. However, up to now, no results with respect to islands aligned with the maxima of the electric field of a resonator or photonic crystal mode exist in literature. We expect an increased rate of optical transitions as a consequence of the large modal field at the SiGe island position. The most common method to determine the nucleation sites of islands uses pits etched into the substrate, in that the islands nucleate preferentially (Zhong 2003, Hu 2008, Katsaros 2008, Kiravittaya 2009). In order to be able to grow SiGe islands with uniform optical properties at positions dictated by electric field pattern of an optical eigenmode over large areas, the interplay between SiGe island growth conditions, pattern period, optical properties, and homogeneity of the SiGe islands has to be precisely controlled. Thus within this project, the dependence of the islands' luminescence properties and morphology on the pit pattern period was established for a given set of growth parameters (Hackl 2011). Based on these parameters, ~10 mm$^2$ large areas with addressable SiGe islands showing homogeneous and energetically narrow PL emission spectra ideally suited for aligned integration in photonic crystals could be demonstrated (Lausecker 2011).

A quantitative correlation of structural SiGe island properties with the observed PL or absorption spectra and the calculated electronic level structure has so far only be performed on island ensemble level (Brehm 2009, Brehm 2010, Grydlik 2010), since experimental data on the optoelectronic properties of single islands in the Si/Ge material system are scarcely available. To my knowledge, absorption data for quantum dots consisting of group IV semiconductors are only published for $Ge_{1-x}Sn_x$ islands (Naruse 2009) and no PL emission of single islands composed of group IV elements has been reported so far. I am also not aware of any published single island photocurrent results similar to those published for III-V material islands by for example (Fasching 2005, Zrenner 2006). Transport through single SiGe islands has been observed recently (Katsaros 2010), but from these results no conclusions with respect to the energy level scheme in the isands can be drawn.

With respect to intersubband devices in the SiGe systems, most of the recent work on QWs, was focused on conduction band quantum wells, both with tensilely strained Si as Δ-valley QW (Ciasca 2009) and for compressively strained L-valley Ge QWs (De Seta 2009). For both QW systems, growth on SiGe pseudo-substrates is necessary for adjusting the required conduction band offsets between the layers. Such QWs are considered as promising for the realization quantum cascade emitters for the IR and THz spectral region. For SiGe islands, quantum dot infrared photo detectors (QDIPs) operating at elevated temperatures up to room temperature have been reported recently (Singha 2010, Yakimov 2011). The work within this project on the further development of SiGe based quantum well infrared photodetectors (QWIPs) is complementary to these approaches utilizing the advantageous properties of light-hole states for the realization of QWIPs sensitive both in the THz and MIR spectral range (Rauter 2011).

A new approach to develop Si based detectors for single photon detection was suggested in (Stepina 2011). In their work, the authors observed giant fluctuations of the electron hopping conductivity along a lateral channel containing a strongly localized SiGe QD system upon irradiation with a weak photon flux. They attribute these fluctuations to a modification of the hopping paths through the channel due to charging/discharging individual QDs in the channel by the incident photon flux and claim that this effect is suitable for single photodetection in the wavelength range around 1.55 µm. The approach followed in this project to establish single photon detection is based on the excitation of an electron from the valence band into the acceptor impurity band, and thus, aims at a region of much larger wavelengths in the THz and MIR range.

### 4.12.3 Results and Discussion

For future applications of SiGe islands as sources in a SiGe based optoelectronic platform, room temperature emission of the SiGe islands is required. By integration of SiGe islands into microdisc resonators and photonic crystals, room temperature PL and EL has been demonstrated (Xia 2009, Xia 2010). Fig. 1 and Fig. 2 show that room temperature PL could also be demonstrated within this work, both for islands integrated into PhC as well as into microdiscs.

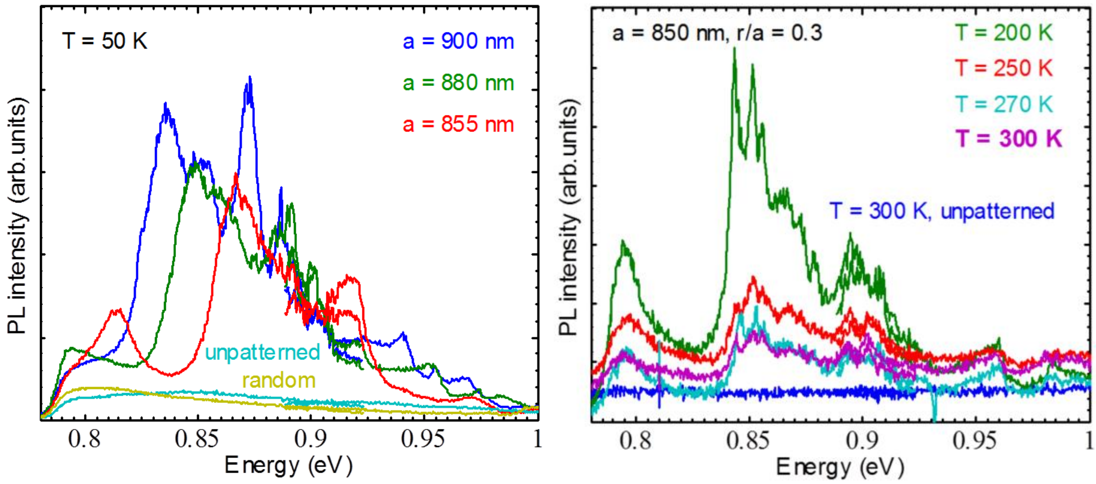

**Figure 1:** left: PL spectra of SiGe islands in PhCs. A clear dependence of the emission maximum on the period of the PhC is observed. Compared to unpatterned sample areas and areas with randomly positioned holes, the PhC emission is enhanced by an order of magnitude; right: Due to the enhancement, SiGe island PL is observable up to room temperature, whereas from regions outside the PhC, no PL is observed at 300K

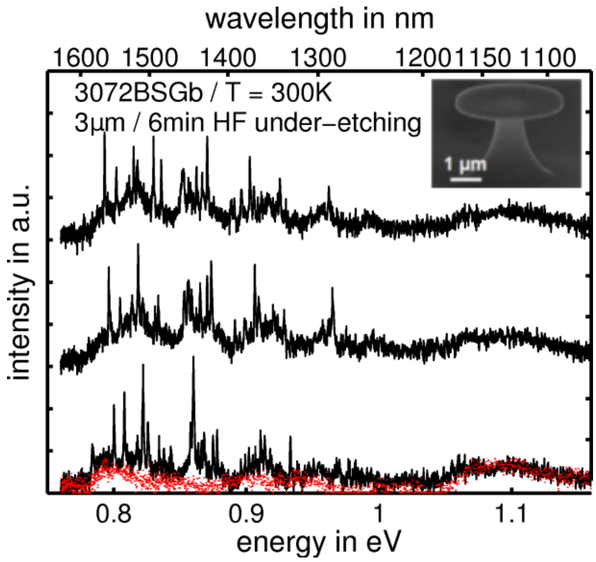

**Figure 2:** Room temperature PL emission spectra of several microdisc resonators (3 µm diameter) with integrated SiGe islands (black). In the spectral region of SiGe island emission (0.8-0.9 eV) sharp emission resonances (whispering gallery modes) prevail up to room temperature. On reference positions without resonators, these peaks are absent (red)

In Fig. 1 it is shown that the peak emission sensitively depends on the PhC period a. A thorough analysis of the electromagnetic eigenstates of the PhC obtained by a three dimensional simulation (including the finite thickness of the PhC membrane) allowed to tentatively identify the modes that dominate the PhC's emission spectra. These are hybrid modes between free-space propagating and PhC guided modes, for that the in plane electric component of the modal field is dominated by the PhC guided mode. In order to improve the PL efficiency, PhCs with islands positioned at the maxima of these guided modes were fabricated. A detailed description of these PhCs is given in the report of P02. Despite the positioning of the islands at the maxima of the electric field, the PL emission is not significantly improved. However, systematic experiments that monitor the PL efficiency as a function of the positioning accuracy of the islands with respect to the field maxima have not been done yet, since it is not straight forward to control the island positioning with the required resolution over different PhC fields by e-beam lithography. Therefore a pair of stamps for nanoimprint lithography was defined for large area (1 x 1 mm$^2$) patterning of island positions and PhC definition, the periods of which being slightly different, so that a Moiré beating between exactly aligned and completely misaligned islands occurs. The difference in the periods of the stamp pair was chosen such that approximately two beating periods appear across the diameter of the PhC (1 mm). As a consequence, within the detection area of our micro-PL setup (~30 µm), the alignment variation is negligible, and the influence of the alignment accuracy can be observed by spatially resolved PL experiments across the PhC field.

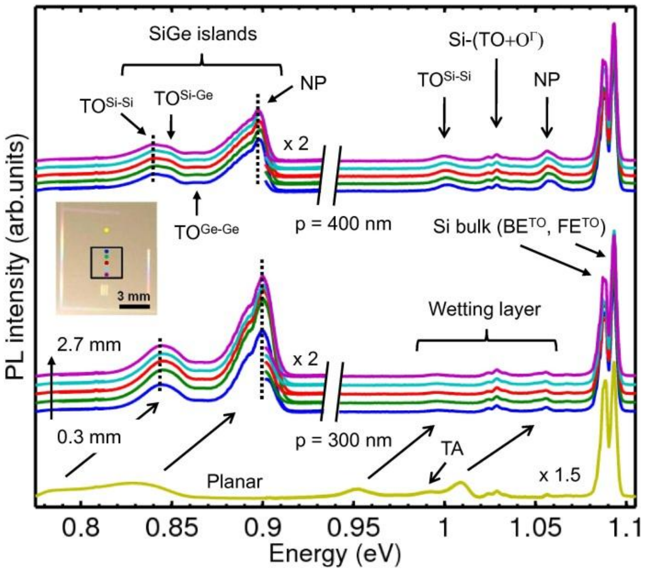

**Figure 3:** SiGe island PL spectra measured at several positions equally distributed over a 3 x 3 mm$^2$ area of ordered islands. Excellent homogeneity of the islands' optoelectronic properties is achieved

These experiments are still in progress, however we have demonstrated that the growth of a homogeneous and ordered island ensemble over mm-large areas defined by nanoimprint lithography is possible under the established growth conditions (Lausecker 2011). In Fig. 3 it is shown that for template periods of 300 nm and 400 nm no variation of the PL spectra across an island field of 3 x 3 mm$^2$ is observable. For the realization of ordered islands on templates with periods dictated by the periodicity of the photonic crystal, which in turn depends on the desired energy of the emission maximum (see Fig. 1), the optoelectronic island properties have to be optimized in dependence of the pattern period. In Fig. 4 it is shown that for a given set of growth parameters, the PL emission efficiency is drastically quenched if the pattern period is larger than a threshold value (400 nm in Fig. 4). In addition, for even larger pit periods (800 nm in Fig. 4), island nucleation in the flat regions between the pits occurs as evidenced by the appearance of island PL with the same spectral shape as observed on the un-patterned reference fields. In (Hackl 2011) it is shown that this interpretation is consistent with AFM images. A careful analysis that will be discussed in a forthcoming publication shows that both the rate and the amount of the deposited Ge has to be matched to the pattern period for a given growth temperature in order to obtain perfectly ordered islands (no island nucleation between the pits) and islands with finite PL efficiency suitable for optoelectronic applications, respectively.

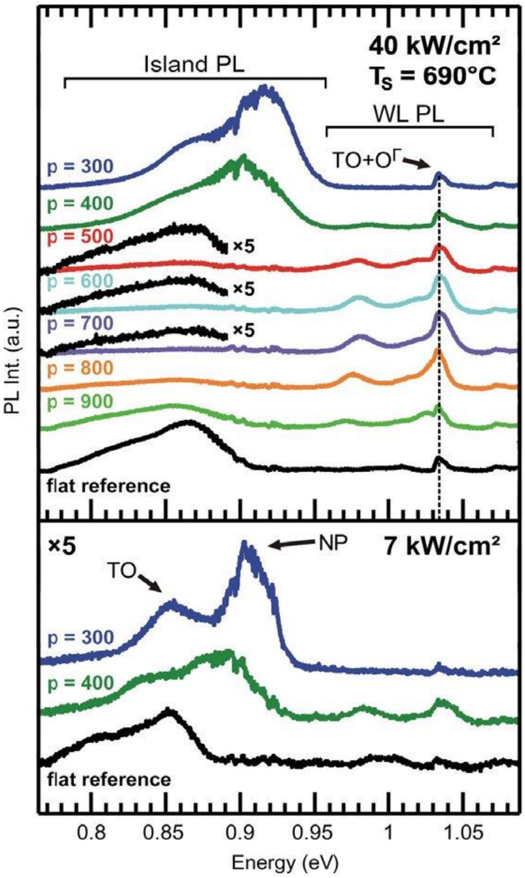

**Figure 4:** PL island emission of ordered SiGe islands for various template periods grown under identical conditions. Beyond a critical amount of Ge per pattern unit cell, PL quenching is observed. A deposition rate in excess of the global Ge incorporation rate results in island nucleation between the pits

Both, Fig. 3 and Fig. 4 show, that for the ordered islands a richly structured PL spectrum is observed, whereas for the randomly nucleated islands on the unpatterned areas these structures appear to be smeared out. In addition, the emission maximum of the PL of the randomly nucleated islands is observed at lower energetical position that depends much more on the excitation energy than for the ordered islands. AFM measurements on these samples have shown that both the mean volume and the width of the statistical size distribution of the ordered and randomly nucleated island ensembles are almost equal. However, in several publications evidence is given that the Ge distributions in randomly nucleated and ordered islands are different, the former showing a larger Ge gradient and a less homogeneity than the latter (Rastelli 2008, Schülli 2009, Pezzoli 2009). Thus, we ascribe the smearing and the lower energetical position of the PL maximum observed for the randomly nucleated islands to the more pronounced fluctuation of the maximum Ge content at a larger average value Ge present in these islands (Brehm 2010). Assuming realistic Ge distributions for energy level calculations based on the nextnano3 code, the observed difference in the PL emission maxima for ordered and randomly nucleated islands can be explained. In addition, the calculations show that due to the larger Ge gradient assumed for the randomly nucleated islands, size quantization and the energetic splitting of the lowest few heavy hole eigenstates results in a small density of states at the fundamental bandgap of the SiGe island. As a consequence, at elevated pump intensities the filling of the hole groundstates results in a stronger PL emission broadening towards larger energies as compared to ordered island ensembles, where the shallow Ge gradient results in a much larger density of states at the PL transition energy. In fact, for the ordered islands, a detailed analysis of changes in the PL emission spectrum induced by increasing the pump intensity reveals the appearance of an almost quadratically growing PL line shifted by ~15-20 meV to larger energies with respect to the no-phonon (NP) island emission. We tentatively ascribe this line to a biexciton emission. The magnitude of this anti-binding energy is in agreement with values calculated by the configuration interaction method (Rodt 2005) using single-particle wavefunctions obtained by nextnano3.

Alternative island shapes obtained by the growth on substrates patterned by inverse pyramidal pits have been demonstrated (Grydlik 2010). However, even at low growth temperatures around 550°C a pronounced intermixing of Si and Ge is observed. As concluded from the PL emission energy and a comparison with energy level calculations using nextnano3, even at this low growth temperature, the Ge concentration is already reduced to 25%. At a growth temperature of 700°C, the inverted pyramidal pits become nearly completely smeared during the first few monolayers of deposited Ge and the same island types as observed on substrates patterned with standard pits are observed. Due to this observed unfavorable tendency of enhanced intermixing, the work on islands in pyramidal pits was abandoned.

PL spectra of only ~10 SiGe islands were measured by limiting the diameter of the detection area to ~1 µm by a pin-hole at the position of the intermediate image in the microscope optics of the µ-PL setup. Fig. 5 shows, that a clear PL signal is observed for the small number of islands (green line) down to a pump intensity of ~7 kW/cm$^2$, however, in the PL spectrum the contributions of the single islands cannot be identified, since the spectral shape is the same as observed for an ensemble of ~30 000 islands (red lines in Fig. 5). Most likely, the high pump intensity broadens the PL spectra. Such behavior at large pump intensity was also observed for InAs islands, for that narrow emission lines were only observed for pump intensities below ~100 W/cm$^2$ in the same µ-PL setup under the same experimental conditions.

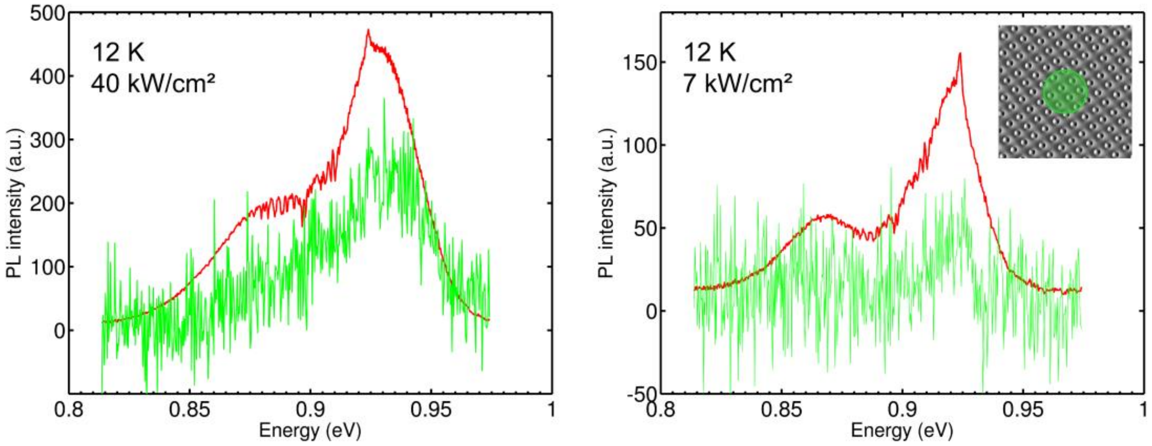

**Figure 5:** PL spectra of ~10 (green) and ~ 30 000 (red) ordered SiGe islands for two excitation intensities. No indications of single island PL are observed at these large excitation intensities

Also the attempts to obtain single island spectra via photocurrent measurements on islands embedded in a p-i-n junction were so far not successful. In Fig. 6, a typical photocurrent spectra of two samples with a single island layer in the intrinsic zone (red, green) are compared to the PC response of a reference diode without islands (blue) and to the PL spectra of the two samples with the islands (lower panel). For the PC spectra shown in Fig. 6, a Si filter was used in order to suppress the huge PC signal originating from the Si substrate absorption. The PC response of the samples due to the Si substrate was measured without Si filter. These spectra were used for normalization. As shown in Fig. 6, also the diode without SiGe island shows a small absorbance below the Si bandgap, the origin of which is not clear yet. After normalization, this background spectrum was subtracted from the measured PC spectra of the diodes with the islands to obtain the additional response due to the presence of the islands. The results after the background subtraction are shown in Fig. 6 by the red and green line for two samples. Evidently, a much stronger Si sub-bandgap absorption is observed for the samples with the islands that we ascribe both to islands and wetting layer, however, at the energy of the island PL emission shown in the lower panel of Fig. 6 no structure is observed in the PC spectrum. Interestingly, for energies smaller than 0.7 eV, the PC signal increases with decreasing energy and is cut off by the absorption of the $SiO_2$ beamsplitter (BMS) used in our experimental Fourier-transform setup. Whether or not this signal is due to VB intersubband transitions in the dot has still to be clarified. If so, the QDs must be occupied by a significant number of non-equilibrium holes, since the islands are located in the intrinsic region of the diode.

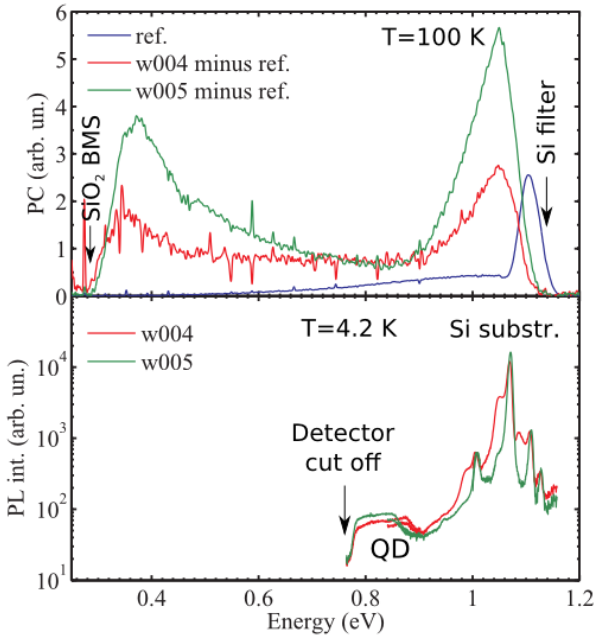

**Figure 6:** Comparison of PC and PL spectra of a single randomly nuclated SiGe island layer integrated into a p-i-n diode

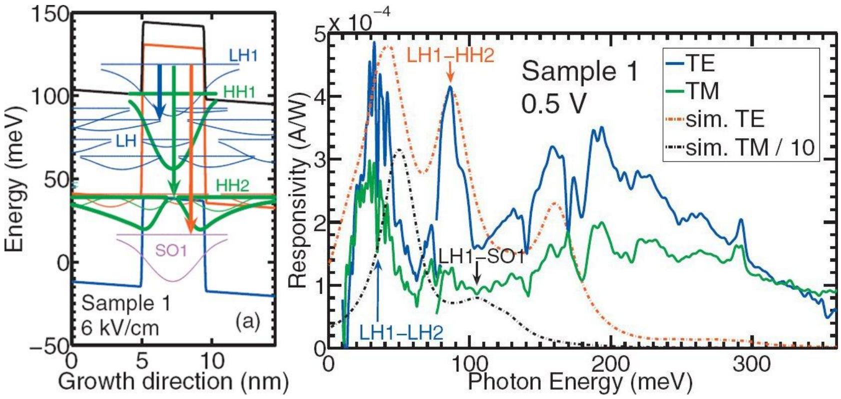

**Figure 7:** Energy level structure (left) and PC spectrum (right) for a new type of QWIP based LH ground states

A new type of QWIP was designed where the intersubband transitions are based on light-hole (LH) QW states (Rauter 2011). The advantages of LH transitions for QWIP performance are discussed in (Jiang 2008). Such a design is only possible using a pseudosubstrate with a finite Ge fraction, so that tensile strained valence band (VB) QWs can be grown for that the LH states become the ground states. Fig. 7, left panel shows the energy level structure of typical QWs investigated within this project together with the observed PC response spectrum (right panel in Fig. 7). The QW in Fig. 6 is grown on a $Si_{0.74}Ge_{0.26}$ pseudosubstrate with $Si_{0.78}Ge_{0.22}$ VB QWs and $Si_{0.9}Ge_{0.1}$ barriers. Fig. 7 left panel, shows a PC spectrum measured at 6 K in that the LH1-LH2, LH1-HH2 (heavy-hole 2) are very pronounced even at a moderate bias of 0.5 V. At this bias, a detectivity of $5.7x10^7$ $cm^2Hz^{0.5}/W$ in the THz spectral region (30 meV) is determined. Increasing the bias voltage leads to a drastic enhancement of the LH1-SO1 (split-off 1) transition that at 4 V dominate the photoresponsivity spectrum (Rauter 2011). The good agreement with the model calculations as well as the relatively large responsivities observed for the LH1-LH2 transition show that the LH band is attractive for designing QWIPs and, thus, also cascade emitter structures.

Detector structures based on the blocked-impurity-band (BIB) absorption were optimized following (Szmulowicz 1988) by decreasing in a series of samples the phosphor compensation doping to $2x10^{13}$ $cm^3$ and increasing the boron BIB doping to $10^{18}$ $cm^3$. The samples were processed and the characterization of the MIR response is in progress. For ultra-low background radiation experiments, an experimental set up in that a LED emitting at 7 µm is cooled to 30 K and in that the BIB detector is cooled to 4 K and shielded from the background radiation by a pinhole cooled to 40 K was developed. In that setup, only photons emitted by the LED or by the 40 K temperature both at the position on the LED or by the 40 K pinhole can generate a response of the BIB detector. By electrically modulating the LED, the BIB detector's response to the 7 µm radiation emitted by the LED and attenuated by the pinhole can be measured at an ultra-low background radiation.

### 4.12.4 Collaboration within and beyond the SFB

Within the SFB, a strong collaboration was established with P02, in that all SiGe islands investigated in this work were grown. In addition, the processed required for the realization of the photonic crystal membranes as well as the microdisc resonators positioned on $SiO_2$ posts were developed in P02 and the devices investigated in P12 prepared. PL experiments performed in P12 were the basis for the optimization of the growth condition with respect to the optical properties of the islands. Using know-how from P13, the energy level structures of the different island types were calculated for a comparison with the experimentally observed PL spectra and their dependence on the excitation energy. Micro-PL measurements aiming at the observation of PL from single QDs and nanocrystalls grown and developed in P03, P04, and P05 have been performed within P12. Additionally, MIR absorption measurements on nanocrystals synthesized in P05 have been performed. All QWIPs investigated in this work were grown at the Forschungszentrum Jülich by G. Mussler in the group of D.

Grützmacher. Together with S. Winnerl and M. Helm at the Helmholtzzentrum Dresden-Rossendorf, the doping profiles for the BIB structures investigated in this project were established by ion-implantation. In collaboration with P. Klenovsky from Masaryk University, Brno, Czech Republic, a large part of the work on the dependence of the PL emission on the excitation intensity was performed.

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