Deliverable 2.3: Spontaneous emission dynamics

1. Hex-Ge PL of single nanowires of different morphologies (TUM)

After having studied PL emission from nanowires of different material compositions, we continued by analyzing optical properties of single Hex-Ge nanowires at high excitation levels such at those for which gain might be expected.

As presented in Figure 1, we observed three different classes of nanowires that are identified by different luminescence features. Corresponding to different shapes we identified different excitation power dependences, as shown in Figure 2. The results are summarized as follows:

- The majority of nanowires, as labelled NW0 in green, show weak photoluminescence with a main emission feature at \( \sim 0.39 \text{eV} \) and a sub-linear power-dependence with an exponent of \( 0.83 \pm 0.02 \).
- Few nanowires, as labelled NW1 in red, exhibit a much larger signal and a second PL peak at \( \sim 0.49 \text{eV} \). The power dependence of those wires shows a strong super-linearity with an exponent of \( 8.17 \pm 0.74 \).
- A single nanowire, as labelled NW2 in blue, exhibiting a very strong PL signal was observed. This wire shows PL emission at \( \sim 0.42 \text{eV} \) and a slightly super-linear power dependence of \( 2.39 \pm 0.38 \) that decreases to \( 1.35 \pm 0.04 \) at \( \sim 0.1 \text{kW/cm}^2 \).

The recorded data suggests a classification of the nanowires in three morphology classes:

- **NW0**: Those nanowires contain lots of crystal defects and, therefore, have only weak optical emission form the fundamental band-transition at \( \sim 0.39 \text{eV} \). Due to non-radiative charge carrier recombination via defect channels, the excitation power dependence of the detected PL signal is sub-linear.
- **NW1**: Wires of this kind have fewer crystal defects and better optical quality than **NW0**. Our working hypothesis for those wires is as follows: Due to the lower defect density, charge carrier recombination channels related to crystal defects can be saturated with optically excited carriers. Therefore, in addition to the comparably weak luminescence from the fundamental
band-transition at $-0.39\text{eV}$, a strong PL signal at $-0.49\text{eV}$ is observed. This higher energetic luminescence is attributed to a transition from the second conduction band to the valence band. As described in the band structure calculations by Jena, the second conduction band transition has a much larger transition matrix moment. Therefore, this higher conduction band transition is optically much more active than the fundamental one. Since optically excited charge carriers recombine much slower via the fundamental transition, the wire is in population inversion. Light amplification via stimulated emission is possible and we observe a super linear slope in the excitation power dependent measurements. However, as the end- and side-facets of the NW1-nanowires are not smooth enough to enable efficient waveguiding, the wire cannot be pushed into the lasing regime when exciting it below its destruction threshold of $-10\text{KW/cm}^2$.

• NW2: As a working hypothesis we expect this nanowire to exhibit very good crystal quality with almost no defects. Therefore, as shown in Figure 2 in blue, we observe PL signal for very low excitation power densities of $-40\text{W/cm}^2$. Furthermore, a change in the slope of the excitation power dependence from $2.39\pm0.38$ to $1.35\pm0.04$ corresponds to a transition from a super linear regime to an almost linear one. In addition to the light amplification via stimulated in nanowires of type NW1, this observation indicates the presence of an optical feedback in NW2. This optical feedback in nanowires is typically provided via waveguiding and reflection from the end-facets. For this hypothesis to be correct we expect to see a rise in coherence time of the light detected from NW2 as compared to that detected from a wire of type NW1 or NW0. Therefore, we performed Michelson interference measurements as presented in the discussion below.

2. Probing charge carrier dynamics in Hex-Ge nanowires with non-resonant pump-probe spectroscopy (TUM)

![Figure 3](image_url)

*Figure 3: PL emission intensity as a function of excitation power density for a “NW 1” - type nanowire on the left. The data is shown for excitation with probe only (red), with pump only (green) and both pump and probe (black) for zero time delay $\Delta T$. The right side shows the normalized PL intensity as a function of pump-probe delay $\Delta T$ for a “NW 1” - nanowire in red and a “NW 2” - nanowire in blue.*
We continue by investigating carrier relaxation dynamics and, later in Sec. 3, the coherence properties of those wires. The emission of coherent laser light (already from a 1st generation) Hex-Ge nanowire constitutes a significant milestone on the road towards a Hex-Si$_x$Ge$_{1-x}$ nanowire laser.

The pump-probe PL data is summarized in Figure 3. The data was obtained at 7K for a “NW 1”-type wire and a “NW 2”-type wire. The experiment was performed with both pump and probe photon energy being above the bandgap with a probe energy of 1.3eV and a pump energy of 1.6eV, respectively. The pulse width was 60ps and the time delay $\Delta T$ was tuned between 0.00±0.06ns and 18±0.06ns using varying coaxial cable lengths at the trigger-inputs of pulsed laser diodes. The timing resolution is limited to 0.25±0.06ns, as given by the smallest coaxial connector. In the data shown on the left side of Figure 3 the time delay $\Delta T$ was fixed to 0.00±0.06ns. When focusing the excitation beams onto a nanowire of type “NW 1”, we can distinguish three different excitation scenarios:

1. Excitation with the probe-beam only, as shown in red: Here we see only lock-in noise and no PL signal, since the optically generated charge carriers are not sufficient to fill all the defects and generate luminescence.
2. Excitation with pump only, as shown in green: Here we see a super-linear rise of PL emission.
3. Excitation with pump and probe, as depicted in black: In this case we can see a clear enhancement of PL intensity by a factor of 1.7, despite the fact that no PL is detected from probe alone.

The super-linear enhancement by the weak probe beam, as observed above, is now studied for different pump-probe time delays $\Delta T$ and for a “NW 1”- nanowire as well as for “NW 2”. The corresponding data is shown on the right side of Figure 3 and was obtained for a fixed pump/probe excitation power density of $P_{\text{pump,NW1}}=5.6$ kW/cm$^2$, $P_{\text{probe,NW1}}=2.8$ kW/cm$^2$, $P_{\text{pump,NW2}}=0.74$ kW/cm$^2$ and $P_{\text{probe,NW2}}=0.64$ kW/cm$^2$, respectively. While increasing $\Delta T$ from 0.00±0.06ns to 18±0.06ns, no significant change in the normalized PL intensity could be observed in case of “NW 1”. However, for “NW 2”, a clear decrease from $\sim 1.0$ to $\sim 0.925$ was found with a decay constant of 0.23±0.06ns, thereby indicating a much faster charge carrier relaxation in case of “NW 2” compared to “NW 1”. Therefore, we came to the working hypothesis that temporal dynamics of charge carriers in case of “NW 2” are dominated by stimulated emission rather than spontaneous emission or non-radiative recombination. We will continue by investigating the coherence properties of the light emitted by “NW 2” in a Michelson experiment in order to further verify this assumption.
3. Coherence enhancement for “NW 2” compared to “NW 0” (TUM)

As described in the pump-probe measurements, we tentatively attribute the emission from Hex-Ge nanowires to originate from stimulated emission for wires of type “NW 2” compared to wires of type “NW 1” and “NW 0” where light stems from spontaneous emission. In order to verify this working hypothesis, we performed Michelson interference measurements on both “NW 2” and “NW 0” types.

![Figure 4](image.png)

**Figure 4**: Michelson interference measurement of a “NW 2”-type nanowire on the left in blue and of a “NW 0”-type wire on the right in green, respectively.

The measurements presented in Figure 4 were performed at 7K on a single, standing Hex-Ge nanowire of type “NW 2”, as shown on the left in blue and one of type “NW 1”, as shown on the right in green, respectively. After focusing the excitation laser on that specific nanowire using a spatially resolved PL map, the light emitted by the nanowire is guided through a Michelson interferometer and detected by an InSb detector. The corresponding interferograms in Figure 4 show clear interference fringes for both nanowires studied here. In case of “NW 2”, as depicted on the left in blue, a coherence time of $124\pm18$ fs can be measured. In contrast, the coherence time of “NW 0”, as shown on the right in green, is only $27\pm8$ fs, which is $\sim 4.6x$ smaller.

The clear coherence-enhancement of “NW 2” compared to “NW 1” constitutes a clear indication, that the light emitted from “NW 2” stems from amplified spontaneous emission rather than pure PL emission as in case of “NW 1”.

4. Moss-Burstein Effect in hex-SiGe

A strong bandfilling effect in hex-Ge can be observed. This phenomenon was found to hold true for hex-SiGe with a varying Ge content. **Figure 5 (a)** shows the emission spectra recorded from various hex-SiGe nanowires for excitation power densities of 2 kW/cm$^2$ (dashed curves) and 100 kW/cm$^2$ (solid curves). The Ge content of these hex-SiGe nanowires was measured by TU/e to be in the range x=86%-100%. **Figure 5 (b)** shows the integrated intensity and the peak width extracted from **Figure 5 (a)**.
As visible in Figure 5, we can observe a Moss-Burstein Effect for all samples. This observation supports the identification and a direct bandgap of hex-SiGe for Ge content ranging from 100% down to 86%. The data presented in Figure 5 (a) clearly illustrate a pronounced difference between the low and high-power density spectra recorded from the pure Ge sample and those with x<100%. For the pure-Ge sample, excitation at higher power results in a significant blueshift of the emission (27 ± 2 meV) and the clear emergence of an addition spectral feature 50 ± 6 meV above the lowest energy peak. Reducing the Ge-content successively to 97%, 89% and 86% results in power dependent blueshifts of 27 ± 1, 38 ± 9 and 20 ± 1 meV, respectively. A similar excitation induced shift of the lowest PL feature is observed for all the samples. We also compared the low and high-power emission intensities from the samples and found similar behavior for all NWs with x<100%. Clearly, the addition of Si to the shell (e.g. pure-Ge c.f. Si$_{1-x}$Ge$_x$) results in a profound change of the form of the high-power luminescence, notably the appearance of a high-energy emission band. However, the relative intensity of the lower energy feature is similar for all the samples investigated.

5. Photonic Crystal for gain enhancement

A new idea that arose during M12-M24 was to capitalize on our ability to grow ordered arrays of NWs and exploit photonic crystal phenomena to achieve stimulated emission and control the directionality of the emitted light into the far field. Hereby, the periodic array of NWs has associated with it a photonic bandstructure for in-plane optical propagation (perpendicular to the NW axis). Inevitable disorder in the NW photonic crystal array gives rise to Anderson localization of the optical field and, thereby, feedback required to produce lasing, as shown in (Javadi et al. 2014), (Reichert 2018), (John 1987) and (John 1991). Similar phenomena have been widely reported in the literature for III-V planar photonic crystals (see e.g. (Dowling et al. 1994), (Nomura et al. 2009), (Ryu et al. 2002)). In such a system, the group velocity of propagating photonic modes can also reduce dramatically for photon
energies near the photonic band edges. Therefore, photons near the photonic band edges propagate slowly, enhancing the strength of the light-matter interaction.

Consequently, we performed finite difference time-domain (FDTD) simulations of a 2D photonic crystal consisting of a hexagonal lattice of hex-Ge nanowires. **Fig. 6 (a)** shows a typical photonic band structure for Hex-Ge nanowires arranged in a hexagonal lattice with a lattice constant of \( a = 1.06 \) µm and a diameter of \( 2r = 0.3 \) µm. The upper photonic band (air band) and the lower band (dielectric band) are clearly visible, as is the photonic bandgap that extends over the spectral range from 0.40 - 0.61 eV. The energetic position of the photonic band edges can be controlled by the photonic crystal lattice constant \( a \) or the diameter of the Hex-Ge NWs \( 2r \). **Fig. 6 (b)** shows the results of varying \( a \) in the range 0.6 - 2 µm.

For gain enhancement, we plan to tune either air or dielectric band edge into resonance with the nanowire emission. For this, TU/e fabricated a series of initial samples with a lattice constant ranging from 0.78 µm up to 1.82 µm (see Section 1.1). The samples have fairly well-ordered nanowire arrays arranged in a hexagonal lattice, with the lattice constants 1.19 µm in (a) and 1.82 µm in (b).

**Figure 6**: Typical results of FDTD simulations for Hex-Ge nanowires placed on a hexagonal lattice on the growth surface. (a) shows a typical photonic band structure with a lattice constant \( a = 1.06 \) µm and \( 2r = 0.3 \) µm. (b) shows the tunability of the photonic band edges for \( 2r = 0.3 \) µm and \( a \) in the range 0.6 - 2 µm. The solid grey line shows selected PL spectra from the pure-Ge sample recorded at 20 kW/cm².

**Figure 7**: Optical microscope image of two nanowire arrays arrange in a hexagonal lattice, with the lattice constants 1.19 µm in (a) and 1.82 µm in (b).
as shown in Fig. 7, with defects that occur during the growth process. Hereby, Fig. 7 (a) shows a typical Hex-Ge nanowire array with $a_1 = 1.19 \, \mu m$ while Fig. 7 (b) shows a lattice constant of $a_2 = 1.82 \, \mu m$. Besides a number of point defects (vacant lattice sites), only slight irregularities in the nanowire radius and position are visible in Fig. 7. Fig. 8 (a) and (b) show the Fourier-transform of the Figures 7 (a) and (b), respectively. Hereby, the six-fold symmetrically ordered points show clearly the hexagonal lattice of the photonic crystal. In addition, the width of these points indicate the disorder of the lattice and the central Gaussian peak corresponds to the number of nanowire vacancies. In general, we observe more irregularities for lower lattice constants $a$, as visible in Fig. 8.

![Figure 8: (a) and (b) show the Fourier-transform of the optical microscope images displayed in Figure 6(a) and (b), respectively.](image)

Such local photonic disorder is essential to give rise to local optical trapping as a consequence of shifting photonic modes below the average band edge of the defect free photonic crystal. Hereby, areas are formed locally where the photonic bands are shifted in energy. As a result, this gives rise to optical modes that are localized over a specific length scale leading to the key-element of optical feedback needed for lasing operation. At the same time, such quasi-localized modes can extend over a large region of the photonic crystal, maximizing the available material gain able to contribute towards possible lasing action.

![Figure 9: PL results of the 1.19 $\mu m$ photonic crystal from Figure 6 (a). (a) Shows the emission spectra for increasing excitation powers. A clear non-linearity is observed after 4 kW/cm$^2$. In (b) these spectra are integrated and fitted to show the superlinear slope of 5.5.](image)
To investigate the photonic crystals optically, their photoluminescence response is measured for increasing excitation density. At excitation densities of upwards of 4 kW/cm$^2$ a bright spot is found on the lattice constant $a = 1.19 \mu m$ photonic crystal. At this position, the emission intensity increases rapidly with increasing excitation density. This behaviour is shown by measuring the spectrum for increasingly higher optical pump powers in Fig. 9 (a). To further analyze this result, the spectra are integrated and plotted logarithmically in Fig. 9 (b). The superlinear slope is determined to be $S = 5.5 \pm 0.6$ by performing a linear fit to the double-logarithmic dataset. A superlinear slope $>1$ is an indication that material gain is present.

This gain can be explained by Anderson localization. As stated above, the light is localized to certain areas of the photonic crystal since the disorder causes fluctuations in the energetic positions of the photonic band-edges. In this particular sample, we observe disorder of at least 50% in radius and approximately 30% in lattice constant. Therefore, the light is localized to small area. By chance, the crystal is locally more ordered, and we can observe amplification of light, as seen in Figure 9.

### 6. Strong Emission from Ge-rich spokes in SiGe Nanowires

Following iterative improvements of the growth parameters, new SiGe Nanowires were grown by TUE and analyzed by TUM / TUE. The observed sample (H06128) contains nanowires with a length of approximately 6 microns at a total radius of 590nm. The nominal Ge content was 80%, while the real Ge content measured using EDX is approximately 86%.

![Figure 10: In (a) a spectrally resolved power series at 7K on 3-4 nanowires on the sample ranging from an excitation power density 8 kW/cm$^2$ up to 500 kW/cm$^2$. The inset shows a magnified view of the spectra at lower excitation powers. (b) shows the emission intensities for the high-energy (black) and low-energy (red) from (a) as a function of the excitation power density in a double-log plot. The noted slopes correspond to the slope of a linear fit.](image)

In Figure 10 (a) a spectrally resolved power series on a few standing nanowires at 7K is shown. The excitation power density is varied from 8 kW/cm$^2$ (in green) up to 500 kW/cm$^2$ (in red) and focused on 3-4 nanowires. As visible in the inset, for low excitation powers only the photoluminescence peak at 0.51 eV is observed, as expected for this SiGe composition. Above a threshold excitation power, a second peak at 0.34 eV emerges rapidly. The peak behavior is analyzed in Figure 10 (b). Here, the emission intensity of the two peaks in Figure 10 (a) are plotted against the excitation power density in
a double-log plot. The peak at 0.51 eV (in black) exhibits a slow increase and can be fitted with a linear fit with slope 0.6. Furthermore, it saturates at higher excitation powers. However, for the peak at 0.34 eV (in red) we observe no intensity up to a certain threshold power and then a superlinear increase corresponding to slope of 12.4 with a linear fit.

While the high-energy peak at 0.51 eV coincides very well with the emission from SiGe and exhibits the already described Burstein-Moss effect, the peak at 0.34 eV is out of the ordinary. The emission energy corresponds to the emission energy of hexagonal Ge NWs hinting regions with high Ge content within the NWs. We observed earlier a Ge rich shell around the SiGe shell as well as Ge rich spokes leading from the corners of the core to the corners of the whole NW, as visible in the lamella TEM in Figure 11. The Ge rich spokes surrounded by SiGe form quantum wells across the cross section of the NW. Since these spokes are extremely narrow (approximately 25nm), the emission at low excitation powers is too weak to observe. However, stimulated emission from these quantum wells is possible and exceeds above a certain excitation power threshold the noise limit. Only at this point, we can detect the light. This is also the reason we cannot observe the transition from spontaneous to stimulated emission for this peak.

Comparing this data with the results from the photonic crystal sample in Figure 9, we can confirm that the signal originates form pure hexagonal Ge. The emission peak has the same emission energy and behaves similarly as well. The photonic crystal sample consists only hexagonal Ge and thus we have enough material to see the emission even at low excitation powers. While here the amplification originates from the Anderson localization of the photonic crystal, we observe stimulated emission stemming from a quantum well for the SiGe NW sample (H06128) in Figure 10. Although these are two different methods of amplification, the resulting emission is very similar.

**Figure 11:** TEM image of sample H04651 with nominal 81% Ge content. The Ge rich spokes from the center are visible as well as the Ge shell around the SiGe shell.

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**Publication bibliography**


Javadi, Alisa; Maibom, Sebastian; Sapienza, Luca; Thyrrestrup, Henri; García, Pedro D.; Lodahl, Peter (2014): Statistical measurements of quantum emitters coupled to Anderson-localized modes in
disordered photonic-crystal waveguides. In Optics express 22 (25), pp. 30992–31001. DOI: 10.1364/OE.22.030992.


