Emission properties of high-\(\beta\) nanolasers with continuous gain media

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“All around the wind blows
We would only hold on to let go”

MAJOR LAZER
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Chapter 1

Introduction

Information technology is a cornerstone of our modern society. In huge data centers enormous amounts of data are being processed and redistributed. Recent reports estimate that by the year 2020 the annual internet traffic will have increased up to 2.3 Zetabytes (1 ZB = 10^{21} B) globally [1]. The peer-to-peer traffic is, however, only a fraction of the traffic that stays inside data centers for queries to databases, procedure calls, etc, which amount on average to three times the volume that enters or leaves the data center [2]. For certain companies, handling huge amounts of HTTP requests, this factor is even up to 1000 as the volume of data sets that must be accessed makes it impossible to run everything from a single server [3]. The energy consumption that comes with this ever increasing volume of data processing is substantial. Estimates established that around the globe, operation of data centers accounted for 1.5% of the total worldwide electricity consumption in 2010 [4] with large impact for the environment. Around 2% of the global green house gas emission is produced by the information and communication technologies sector [5] with ever growing tendencies in the overall rate of energy consumption due to increasing demand for new and faster technologies as well as ever larger data sets that are processed in the age of big data.

In light of this ongoing technological transformation, the search for more energy efficient solutions for communication technologies is of high interest, both, from an economical and ecological standpoint. In data centers a large percentage of the power consumption falls to optical interconnects. In those, vertical cavity surface emitting lasers (VCSELs) are used to transmit data between chips or different sections of data centers with energy consumption of several tens of pJ/bit [6]. Bringing this rate down to 1 pJ/bit is an established goal as massive energy savings will be necessary to keep up with ever larger amounts of data that have to be processed [2]. Also, if optical data transmission will be possible with reduced energy per bit, electrical signals could be replaced by optical ones on increasingly shorter length scales. In this regime, optical interconnects could outperform electrical ones in terms of energy consumption if a rate of 10–20 fJ/bit per optical pulse generation can be achieved [7]. Taking this idea even further are integrated photonic circuits in which all logic operations are performed by optical components. They promise greatly increased computation speeds, low power dissipation and large bandwidths due to multiplexing. At the same time, miniaturization on the scale that is achievable with electronics is challenging due to the diffraction limited confinement of light in photonic components. That said, while the development of all-optical processors may still be future technology, hybrid system have emerged in which the combination of electronic and optical components leads to an increase in efficiency [8].

In the last decades nanolasers have sparked great interest as they present the next step in the quest for miniaturization from lasers with microcavities to lasers with cavities of sub-
CHAPTER 1. INTRODUCTION

Figure 1.1: Owing to the small size of the laser cavities, quantum optical effects take place in nanolasers, changing the fraction of spontaneous emission that is directed into the cavity mode and thus the balance of spontaneous and stimulated emission is changed which impacts the energy efficiency of the laser as well as the quality of the light emission.

micrometer dimensions. The miniaturization is not only crucial for densely integrated device applications, but is also associated in the literature with an increase in energy efficiency, sought after in optical interconnects, by lowering the laser threshold. This is facilitated by the small cavity volumes of nanolasers that introduce quantum-optical effects and allow to engineer the spontaneous emission rate of the gain material. In this thesis we revisit the notion of the laser threshold for nanolasers, defined for conventional lasers in terms of the output intensity, and investigate its relation to alternative approaches that rely on the laser’s photon statistics. We especially focus on the rate of spontaneous emission that is directed into the laser mode (see Fig. 1.1) as it plays different roles in nanolaser with different device geometries and gain materials employed.

Many reports on nanolasers in the literature focus on quantum dot (QD) based gain medium [9, 10, 11]. In QDs charge carriers are locally confined which allows for a strong light-matter interaction strength and thus potentially high gain. At the same time QDs suffer from manufacturing inhomogeneities [12] making them not universally applicable as gain media for nanolasers. More recently also continuous, two-dimensional quantum well (QW) gain media where brought from micrometer scale (e.g. VCSEL structures) into the nanolaser regime, made possible by improved manufacturing techniques and high-finesse (high-Q) cavities [13, 14, 15]. There is an ongoing investigation of new device geometries and material compositions for nanolasers and different types are under investigation including photonic crystal cavities, microdiscs, and metallic cavities [16].

Also recently, a new class of 2D materials attracted attention as potential gain medium for nanolasers. Transition metal dichalcogenides (TMD) form atomically thin two-dimensional layers, similar to graphene. Some of them turn out to be direct band gap semiconductors when single layers are exfoliated from the bulk material, which makes them interesting for optoelectronic applications [17, 18]. Two-dimensional TMDs, due to their atomic thickness, exhibit large excitonic binding energies and fast luminescence. Furthermore, they are susceptible to their external environment, which allows to tailor their electronic and optical properties. In this light, monolayers of semiconducting TMDs are also investigated as gain medium in nanolasers. Several groups reported lasing emission from single layers of TMD materials [19, 20, 21, 22, 23]. However, there is much debate, whether the observed signatures actually prove the onset of lasing operation, or not [24, 25].

The use of extended, two-dimensional gain media, such as QWs as well as TMD monolayers, holds opportunities to advance the fields of nanophotonics and provides new ways for developing sources of coherent light on the nanoscale. Here we contribute to this field by pro-
viding quantum-optical as well as semiclassical laser models that allow to assess the threshold behavior of a range of devices and to give guidelines for the design of future experiments. This thesis is organized as follows. Chapter 2 will introduce concepts that are fundamental for the discussion of (nano-) lasers and their threshold properties. Chapter 3 revisits the notion of the laser threshold and sheds light on how different notions of the laser threshold are applicable, especially in the context of two-dimensional gain media. The special class of atomically thin TMD materials is discussed in Chapter 4 and their applicability as gain medium for nanolasers is assessed. In Chapter 5 we introduce a quantum optical model for semiconductor nanolasers that we apply in Chapter 6 in close cooperation with experimental groups to investigate real devices and to unambiguously proof lasing in these structures.
Chapter 2

Foundations

Nanolasers, like conventional lasers, consist of three main components. An optical resonator or cavity confines the light field and provides optical feedback for light amplification. The gain medium, interacting with the light field of the cavity, emits light, either by spontaneous or stimulated emission, where the latter process is responsible for the emergence of coherence in the lasing process. Lastly, an external pump source, electrically or optically, provides energy to the process by exciting the gain medium and facilitating stimulated emission.

In the following we introduce essential laser components in the context of nanolasers with extended, two-dimensional gain media and discuss basic concepts, necessary for the description of their emission properties.

2.1 Micro- and nanocavities

Optical resonators or cavities are an integral component of any laser as they provide optical feedback. A perfect cavity would contain light for an indefinite time at a single well defined frequency. However, any realistic cavity leaks some light at a certain rate which broadens the cavity’s resonance spectrum and is quantified by its quality factor $Q$

$$Q = \frac{\Delta \omega}{\omega},$$

where $\Delta \omega$ is the width of the cavity mode’s spectral line and $\hbar \omega$ is its mode energy. In a more intuitive picture, the $Q$ factor is a measure of how often, on average, a photon is reflected by the cavity’s mirrors before it escapes the cavity. Another important figure of merit is the cavity’s mode volume as it indicates the degree of spatial confinement of the electric field. Neglecting the influence of cavity losses [26] it is commonly defined as

$$V_m = \frac{\int \epsilon(r)|E(r)|^2 d^3r}{\max[\epsilon(r)|E(r)|^2]},$$

where $\epsilon(r)$ is the relative permittivity. Lower mode volumes lead to higher field strengths in the cavity, thus increasing the light matter interaction with the gain medium.

The intuitive picture of a cavity is a pair of two flat mirrors facing each other, as it is e.g. the central component of a Fabry-Pérot-interferometer. However with the ongoing miniaturization of laser devices, other cavity designs were developed that allow for a maximal light confinement and higher $Q$ factors. A number of different cavity geometries on the micro- and nanoscale are pictured in Fig. 2.1. Photonic crystals refer to materials with a periodically changing refractive
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Figure 2.1: Examples of different micro- and nanocavities: a) Photonic crystal cavities exhibit a localized defect mode [27], b) microdisc cavities with whispering-gallery modes [28] and c) nanocavity with a metallic cladding allow to confine light below the diffraction limit [13].

index in which light propagation is suppressed for certain frequencies due to a photonic band gap. This effect is analogue to the emergence of an electronic band gap in solids with a periodic atom potential\(^1\). Similar to atomic crystals, a defect in the photonic crystal creates localized photon modes that confine light by back reflection into the defect region. Fig. 2.1 a) shows an example of a photonic crystal cavity (PCC). A two-dimensional array of air holes in a dielectric material (e.g. SiO\(_2\) or GaAs) creates the photonic band gap. A number of adjacent holes are missing creating the photonic defect cavity. Confinement in the vertical direction is achieved by total internal reflection due to a high refractive index contrast with the environment. Other types of PCCs are one dimensional photonic crystals with a hole structure (nanobeam) [15, 29] or vertical Bragg-reflector cavities (also 1D), found e.g. micropillars [30]. PCCs can achieve the smallest mode volumes close to the diffraction limit, i.e. with dimensions of half the light’s wavelength \(V_m \approx (\lambda/2n)^3\). On the other hand the total dimensions of the photonic crystal array is rather large with dimensions of several \(\mu m\). In theory, very high \(Q\) factors can be achieved in PCCs, especially when optimizing hole positions close to the cavity (\(Q > 10^6\)) [31]. However, the demands in the manufacturing precision are high and \(Q\) factors in the range of \(10^3 - 10^4\) are more typical [32, 33].

Microdisc cavities are disc shaped structures, fabricated from dielectric materials with diameters in the \(\mu m\) range. An example is shown in Fig. 2.1 b). They support whispering-gallery modes on the edge of the disc by total internal reflection. Microdisc cavities generally have larger mode volumes and lower quality factors in comparison to PCCs. However their whispering-gallery modes provide good overlap with gain media integrated in the microdisc which increases their light-matter interaction and thus their efficiency [34]. Furthermore, microdiscs (symmetrical or deformed) allow to study interesting optical phenomenon like wave chaos and non-Hermitian dynamics [35].

The minimal mode volume that can be achieved with dielectric media is set by the diffraction limit. This limit, however, can be overcome with metal coated nanolaser cavities, an example of which is shown in Fig. 2.1 c). Although these types of cavities introduce additional losses due to the ohmic resistance of the metal coating, it is indeed possible to achieve sufficient \(Q\) factors of the cavities to facilitate lasing [36, 37]. The significant reduction of device size, similar to electrical transistors, makes nanolasers with metal coated cavities good candidates

\(^1\)This can easily be seen if one considers the wave equation for a spatially stationary electric field \(E(r, t) = E_0 u(r) e^{-i\omega t}\), with \(u(r)\) being the mode function. From Maxwell’s equations one obtains \(\nabla^2 E(r) - (\alpha(r)\omega/c)^2 E(r) = 0\), which is structurally equivalent to the time independent Schrödinger equation \(-\frac{\hbar^2}{2m}\nabla^2 \psi(r) + (V(r) - E)\psi(r) = 0\). Thus, for a periodically changing refractive index \(n(r)\), an analogue to the Bloch theorem can be applied to the electric field which gives rise to a photonic band gap.
2.2 QUANTUM WELLS — TWO-DIMENSIONAL GAIN MEDIA

For application as light sources in on-chip integrated systems. We restrict our introduction of micro- and nanocavities to the discussion of cavity types shown in Fig. 2.1, as nanolasers with those types are discussed as examples in later chapters. However a comprehensive list of cavity types that were investigated as resonators for micro- and nanolasers is given in [16].

2.2 Quantum wells — two-dimensional gain media

Quantum wells (QW) are thin layers of a semiconductor surrounded by a barrier material that creates a confining potential for charge carriers along the vertical direction and restrict them to a two-dimensional motion along the plane. A sketch of a QW structure is shown in Fig. 2.2 a). The confining potential is created by the band gap difference to the surrounding barrier material. As indicated in Fig. 2.2 b), energy levels along the confinement direction are discretized and form subbands \((n = 1, 2, \ldots)\) for the otherwise unrestricted motion of the electrons along the QW plane. The benefit of using QWs instead of bulk media as gain media for semiconductor lasers is the spatial confinement of carriers. The electronic wave functions are localized more strongly and can be placed in the anti-node positions of the cavity mode for maximal field strength in the QW. This leads to stronger interaction of the carriers with the cavity light field and faster emission rates. Furthermore, with charge carriers concentrated in the QW plane, population inversion is created more easily, which is a prerequisite for efficient lasing operation.

The free motion of carriers in the 2D plane of the QW gives rise to a band structure as in the 3D case. An example band structure (GaAs) is shown in Fig. 2.3 a). As typical for other III/V semiconductors as well, the direct band gap at the \(\Gamma\) point is responsible for efficient interaction with the cavity light. The valence band maximum is split into light- and heavy-hole bands as well as the split-off band due to spin-orbit interaction. In order to obtain a simpler model picture we can focus our attention to the region of the band structure around the direct energy level.

Figure 2.2: a) Sketch of a QW structure. The QW layer (yellow) confines electron movement to a two-dimensional plane. A confinement potential \(V_0\) is created by the band gap difference to the surrounding barrier material (blue). b) Energy level structure of the confining QW potential with width \(d\). The Energy levels are discretized along the confinement direction and form subbands of the otherwise continuous energy levels of the 2D electrons. Example wave functions are shown for the vertical component of the QW wave functions in the lowest two subbands. c) The ultimate limit of QW confinement is achieved by atomically thin layers of TMD material as discussed in detail in Section 4.
Figure 2.3: a) The band structure of a typical semiconductor QW (here GaAs). The direct band gap at the Γ point contains the light- and heavy-hole bands as well as the split-off band due to spin-orbit interaction. b) Focus on the Brillouin zone just around the direct band gap. In the effective mass approximation, the various bands around the band gap can be approximated with parabolic dispersions.

Figure 2.4: The electronic DOS for a two-dimensional QW (solid line) is constant in energy in comparison to the $\sqrt{E}$ dependence of the bulk material DOS (dashed line). The discrete jumps in the DOS arise from the subsequent contributions of different subbands that are defined by the one dimensional confinement potential of the QW.
2.3. THE LASER THRESHOLD AND LASER RATE EQUATIONS

band gap (here the Γ point). In the effective mass approximation we describe conduction
and valence bands as parabolas with quadratic dispersion as

$$\epsilon_k = \Delta \epsilon + \frac{\hbar^2 k^2}{2m^*},$$

(2.3)

where \(m^*\) is the effective mass of the band in question (being positive or negative) and \(\Delta \epsilon\) is
the offset of the band from zero.

Another type of two-dimensional gain medium are monolayers of transition metal dichalcogenides (TMD).
They represent the ultimate limit with respect to carrier confinement as they consist of an atomically
thin layer of semiconductor material. A sketch of a typical TMD monolayer (MoS\(_2\)) is given in Fig. 2.2 c)
showing that electrons are confined (perpendicular to the layer) on atomic length scales in these materials. Although
they behave similar to typical QWs with respect to carrier confinement, they exhibit very distinct electronic properties
due to their reduced thickness. These effects as well as their applicability as nanolaser gain media
will be discussed in detail in Chapter 4

Additionally, due to spatial confinement the QW’s electronic density of states (DOS) changes,
in a free carrier picture, from a \(\sqrt{E}\) dependence \[38\] to a constant DOS as indicated in Fig. 2.4.
The discrete steps correspond to occupations in the individual subbands. At sufficiently low
excitation, carriers are confined to the lowest subband and thus, in addition to the spatial confinement,
also spectral confinement takes place. The increased light-matter interaction strength
between the active material and a light field due to spatial and spectral localization of carriers
is often desired in optoelectronic applications. Especially in semiconductor lasers, faster carrier
recombination and thus higher quantum efficiency can be obtained.

2.3 The laser threshold and laser rate equations

The laser threshold defines one the central concepts for characterizing the emission from a
laser. For increasing pump power it is determined by the point where the laser ”switches on”
and starts emitting coherent light. We will discuss and revisit this notion throughout this
thesis. We start here with introducing standard laser rate equations, as they capture the most
basic processes in laser physics and help to illustrate the issues that arise in the comparison
of conventional semiconductor lasers and nanolasers. They describe the dynamics of the excited
carrier density \(N\) in the gain medium and the photon density \(N_p\) in the cavity mode. In a
simplified version the laser rate equations are given by \[39\]

$$\dot{N} = P - G(N)N_p - (\gamma_l + \gamma_{nl})N^2 - AN - CN^3,$$  \hspace{1cm} (2.4a)

$$\dot{N}_p = G(N)N_p + \gamma_l N^2 - \frac{N_p}{\tau_c},$$ \hspace{1cm} (2.4b)

where \(P\) is the pump rate that excites carriers in the gain medium, \(\tau_c\) is the cavity decay time
and \(\gamma_l\) and \(\gamma_{nl}\) are the rates of spontaneous emission into the lasing and non-lasing modes.
The gain \(G(N)\), i.e. the rate of light amplification of the medium, is left unspecified here. For
semiconductor lasers, often linear or logarithmic gain models (as function of carrier density) are
used in the literature \[40, 41\]. The spontaneous emission factor \(\beta\) is the fraction of spontaneous
emission that is directed into the lasing mode defined by

$$\beta = \frac{\gamma_l}{\gamma_l + \gamma_{nl}}.$$  \hspace{1cm} (2.5)
CHAPTER 2. FOUNDATIONS

Figure 2.5: a) For conventional lasers the influence of spontaneous emission on the output intensity is negligible. Reaching the lasing regime with the threshold, all excess energy is converted into photons leading to a linear increase of the output intensity. A sharp transition is separating the two regimes. b) The same data on a logarithmic scale shows a distinct threshold jump in the intensity.

For conventional semiconductor lasers a quite simple picture can be found that clearly illustrates the concept of the laser threshold. This simplified picture holds true as the contribution of spontaneous emission processes are only small, i.e. the SE term in (2.4b) can be neglected with $\beta$ factor of the order $10^{-3}$ and below. Then, for stronger pump power the optical gain of the material increases until it compensates the optical losses from the cavity. In the language of the rate equations in (2.4) this means $G(N) = 1/\tau_c$. As a result, the output intensity suddenly changes from around zero to a macroscopic value as photons start to accumulate in the cavity mode and all excess energy provided by the pump is converted into photons by stimulated emission as depicted in Fig. 2.5 a). Nevertheless, randomly occurring fluctuations of the light intensity that are caused by SE provide the seed for lasing operation, i.e. the initial light field that is amplified by stimulated emission once the threshold is crossed. The emission from SE below threshold is visible in Fig. 2.5 b) showing the same data as in panel a) but on a logarithmic scale.

Fig. 2.6 shows the stationary solutions to the rate equations for different $\beta$ factors. Laser diodes or VCSEL devices generally have lower $\beta$ factors (red curves) [39] and show a clear threshold jump. At the same time the carrier density saturates abruptly (clamping) and excess energy is converted into photons by stimulated emission. Nanolasers, in comparison, operate in a different regime. Due to strong light-matter interaction and small cavity dimensions, quantum optical effects emerge [42, 43]. In particular $\beta$, the fraction of spontaneous emission that is directed into the lasing mode, can increase which reduces the threshold jump in the intensity while clamping of the carriers becomes more gradual. In the extreme case of $\beta = 1$ the threshold jump in the I/O characteristics vanishes [13, 44].

In lasers with high $\beta$ factors, contributions to the emission due to SE start to play a major role. As the threshold jump in the intensity vanishes, identifying the onset of coherent emission from the output power alone becomes ambiguous [45]. The resulting question, how to define the lasing threshold in this regime is an ongoing discussion and one of the central issues of this thesis. In the literature, different concepts for a definition of the threshold in nanolasers were put forward. With $\beta$ factors of $10^{-5}$ for conventional semiconductor lasers all
2.4. PURCELL EFFECT — TAILORING SPONTANEOUS EMISSION

definitions for the laser threshold coincide. The system undergoes a sharp transition at the
threshold pump power, that was discussed in the context of a second-order phase transition in
the thermodynamic limit ($\beta \to 0$) [44, 46]. With $\beta$ approaching 1, the influence of spontaneous
emission increases. Although the threshold non-linearity in the I/O curve shifts to lower pump
powers, it was realized early that this does not imply a zero-threshold lasing operation where
coherent emission is achieved at arbitrarily small pump powers. From rate equation analysis a
quantum threshold criterion was defined as the pump power, where spontaneous and stimulated
emission in the cavity mode are equal or, equivalently, where the average photon number in the
cavity is one [47].

It is clear that high-$\beta$ nanolasers operate far from a thermodynamic limit and thus a clear
transition does not occur. They are truly thresholdless in the sense that they show a gradual
transition between two regimes instead of a (near) discontinuous jump [44]. As discussed
below, unambiguous proof of coherent emission can be given by photon-correlation functions
which indicate the transition from a thermal to Poissonian photon statistics [15, 48, 49, 50]. In
nanolasers with QD gain media the quantum threshold criterion is indeed confirmed by photon-
correlation functions [42]. However, as we will discuss in the following chapters, nanolasers with
extended (2D) gain media operate in a different regime and the issue around the transition to
coherent emission has to be reevaluated (see Chapter 3).

![Figure 2.6](image_url)

Figure 2.6: Stationary results of the rate equations (2.4) for different $\beta$ factors. For low $\beta$ a
distinct threshold transition is visible in the I/O characteristics and the carrier density clamps
abruptly. With increasing $\beta$ the threshold jump shifts to smaller pump values. In the extreme
case of $\beta = 1$ the threshold jump in the I/O characteristics vanishes and also the carrier density
shows only a gradual transition.

2.4 Purcell effect — tailoring spontaneous emission

The emission of coherent light in lasers relies on the process of stimulated emission in the gain
medium which amplifies the light field in the optical resonator. However, spontaneous emission
CHAPTER 2. FOUNDATIONS

Figure 2.7: Different relation of emitter and cavity linewidth: a) Emitter and cavity linewidths according to \( \Delta \omega_e \ll \Delta \omega_c \) as it is commonly the case in atomic or QD based gain media. In this case the expression for the Purcell factor proportional to \( Q/V_m \) is valid. b) The opposite situation \( \Delta \omega_c \ll \Delta \omega_e \) is typically encountered in homogeneously and thermally broadened, extended gain media and nanocavities with sufficiently high \( Q \) factors.

plays an essential role as well, as it is not only a source of noise in the emission, but also provides the initial light field in the cavity that is to be amplified. The lasing threshold and thus the efficiency of the lasing process relies on the fraction of spontaneous emission that is directed into the lasing mode, i.e. on the \( \beta \) factor. One distinguishing feature of nanolasers is the strong confinement of the cavity mode that gives rise to quantum optical effects. Most notably, the photonic environment of the cavity changes the local density of states (DOS), which influences the spontaneous emission of the material itself. The Purcell effect [51] describes an increase of the spontaneous emission into a single mode of the lasers resonator. It can lead to an increase of the \( \beta \) factor and is often cited in the context of high-\( \beta \) lasers [11, 20, 21, 22, 23, 37, 52]. The Purcell factor is commonly given by the increase of the spontaneous emission rate into the lasing mode. It is related to the ratio of the density of states in the cavity \( \rho_c \) and the one of free space \( \rho_f \) assuming perfect resonance of the emitter with the cavity

\[
\frac{\rho_c}{\rho_f} = \frac{1}{8\pi} \frac{Q}{V_m} \left( \frac{\lambda}{n} \right)^3,
\]

where \( Q \) and \( V_m \) are the cavities quality factor and mode volume and \( \lambda/n \) is the wavelength inside the cavity with refractive index \( n \) [53]. This expression proportional to \( Q/V_m \) is derived under the assumption \( \Delta \omega_e \ll \Delta \omega_c \) where \( \Delta \omega_e \) and \( \Delta \omega_c \) are the linewidths of emitter and cavity, respectively. The assumption is indeed satisfied in systems with narrow emitter lines like atomic or QD gain media. However in nanolasers with extended gain media that offer broad
room-temperature gain profiles the situation is typically reversed and the cavity linewidth is narrow in comparison to the emission. Although Purcell enhancement can still take place it is not given by the proportionality \( Q/V_m \) and other expressions must be applied [53, 54]. The two situations are schematically depicted in Fig. 2.7.

Instead of increasing the spontaneous emission rate into the lasing mode, in nanolasers with extended gain media, high \( \beta \) factors can be obtained by suppression of emission into other, non-lasing modes, i.e. either other cavity modes or leaky modes that couple to free space [55, 56]. In photonic crystal cavities the photonic band gap efficiently decouples the gain medium from all other modes except one cavity mode. In general, the small dimensions of micro- and nanocavities lead to a large free spectral range of the cavity and thus facilitates single mode lasing and generally high \( \beta \) factors.

2.5 Photon correlations and states of light

Correlation functions (spatial or temporal) are important tools in statistical physics and quantum mechanics as they carry information about order and microscopic processes in a system. Here we are interested in correlations of in the light field that manifest themselves in amplitude and intensity fluctuations observed in the emission from a light source. Experiments by Hanbury Brown and Twiss on the correlation of intensity fluctuations from stellar radio sources [57] prompted the study of how the dynamics of emitters and their emission processes influence the observed correlations. In this context it was realized that photon correlations can serve as a witness of non-classical states of light by means anti-bunching or sub-Poissonian statistics [58].

In quantum optics, correlation functions of photons are used today as a tool to distinguish different states of light, be it classical or non-classical, according to their emission process [59]. Furthermore, correlation functions are intimately related to the light’s power spectrum [60] that contains information on the excitation levels in the system and its dephasing processes. In this thesis correlation functions play a central role for the characterization of a laser’s output characteristics in order to distinguish spontaneous- and stimulated-emission dominated operation.

First-order coherence

We first consider a classical picture as it provides an intuitive interpretation of coherence. The degree of first-order coherence is tied to fluctuation in the electric field’s amplitude and phase and the occurrence of interference pattern in interferometric experiments. As an example consider a Mach-Zehnder interferometer as depicted in Fig. 2.8. When superimposing two components of a light beam \( E(t) \) at the second beam splitter, the resulting output intensity for a stationary light field is given by

\[
I_3(t) = \frac{1}{2} \varepsilon_0 c |E_3(t)|^2
= \frac{1}{2} \varepsilon_0 c |R|^2 |T|^2 |E(t) + E(t + \tau)|^2
= \frac{1}{2} \varepsilon_0 c |R|^2 |T|^2 \left\{ |E(t)|^2 + |E(t + \tau)|^2 + 2 \text{Re} \left[ E^*(t)E(t + \tau) \right] \right\}, \tag{2.7}
\]

where \( R \) and \( T \) are the reflection and transmission coefficients of the beam splitters and \( \tau \) is the run time difference between the two arms of the interferometer. If we take the average value of the output intensity, (2.7) can be written as

\[
\langle I_3(t) \rangle = 2 |R|^2 |T|^2 \langle I(t) \rangle \left\{ 1 + \text{Re} g^{(1)}(\tau) \right\}, \tag{2.8}
\]
Figure 2.8: In a Mach-Zehnder interferometer light from a source is separated into two components by a beam splitter. Subsequently, the two resulting beams are recombined. The degree of first-order correlation of the beams governs the appearance of interference effects at the output of the setup. Depending on the run time difference $\tau$ the two components will have different phase relations and show constructive or destructive interference if the delay is smaller than the coherence time $\tau < \tau_c$.

where we wrote $\langle I(t) \rangle = \frac{1}{2} \epsilon_0 c (|E(t)|^2)$ and introduced the normalized first-order correlation function

$$g^{(1)}(\tau) = \frac{\langle E^*(t)E(t+\tau) \rangle}{\langle E^*(t)E(t) \rangle}.$$  \hspace{1cm} (2.9)

Thus, the visibility of interference patterns is directly linked to $g^{(1)}(\tau)$. The degree of first-order coherence is given by $|g^{(1)}(\tau)|$. It decays on a time scale given by the coherence time $\tau_c$, on which the light field maintains its coherence and, therefore, the ability to produce interference patterns. In theory, a plain wave of a single light mode shows perfect coherence for arbitrarily long times $\tau$, i.e.

$$E(t) = E_0 e^{-(\omega t + kz)} \rightarrow g^{(1)}(\tau) = e^{-i\omega \tau}, \quad |g^{(1)}(\tau)| = 1.$$  \hspace{1cm} (2.10)

However, any realistic light source will be subject to noise so that $g^{(1)}(\tau)$ will decay to zero corresponding to a finite coherence time $\tau_c$ as shown in Fig. 2.9. The coherence time can be interpreted as the time scale, on which the propagating light will maintain its coherence properties. Commonly also the coherence length is considered given by $l_c = c \tau_c$ with $c$ being the speed of light.

The first-order correlation also encodes information about the energy spectrum of the system. According to the Wiener-Khintchine theorem [61] $g^{(1)}(\tau)$ is related to the normalized energy spectral density $S(\omega)$ of the system via the Fourier transform

$$S(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau \, g^{(1)}(\tau) e^{i\omega \tau}.$$  \hspace{1cm} (2.11)

In the context of laser physics this indicates that the emission spectrum and thus the linewidth of a laser can be determined from the first-order correlation function and vice versa. In the
2.5. PHOTON CORRELATIONS AND STATES OF LIGHT

Figure 2.9: The degree of first-order correlation is given by $|g^{(1)}(\tau)|$. For perfectly coherent light as given (ideally) by a stable plane wave, coherence is maintained indefinitely (dashed line). For an incoherent light sources the degree of coherence will decay on time scales of the coherence time $\tau_c$.

particular case of a Lorentzian emission spectrum with FWHM of $\gamma$, the correlation function decays with coherence time $\tau_c = 2/\gamma$.

In this thesis we are considering laser models with a quantized description of the light that allow us to describe quantum optical effects in the emission. Therefore, we are interested in a quantized description of the correlation functions as well. Glauber introduced a quantum theory of coherence in 1963 [62] in which he introduced the quantum versions of photon-correlation functions as we use them today. The electric field operator $E(r, t)$ is split into a positive and negative frequency part

$$E(r, t) = E^+(r, t) + E^-(r, t) = E_0 \left( u(r) a(t) e^{-i\omega t} + u^*(r) a^\dagger(t) e^{i\omega t} \right), \quad (2.12)$$

where the positive part ($+$) is associated with the destruction of photons ($a(t)$) and the negative part ($-$) with their creation ($a^\dagger(t)$). In the classical picture this distinction is only a mathematical tool as the quantity that is measured in classical experiments is the full electric field $E(r, t)$. However this is different in experiments where quantum effects come into play. For most types of detectors the detection of individual photons involves their absorption and is thus related to $E^+(r, t)$. Assuming an idealized detector of negligible size and perfect time resolution, the state of the field changes in the absorption process by the detector from an initial state $|i\rangle$ to a final state $|f\rangle$. The matrix element of that process will be $\langle f | E^+(r, t) | i \rangle$. The rate at which photons are recorded is determined by Fermi’s Golden Rule. As the detection does not depend on the field’s final state, the rate is proportional to the sum over all final states

$$\sum_f |\langle f | E^+(r, t) | i \rangle|^2 = \sum_f \langle i | E^- (r, t) | f \rangle \langle f | E^+(r, t) | i \rangle$$

$$= \langle i | E^- (r, t) E^+(r, t) | i \rangle. \quad (2.13)$$
More generally, if we are uncertain about the exact initial state and thus have to describe the light field by a (mixed) state \( \rho \), the detection rate is given by

\[
\text{Tr} \left[ \rho E^- (r, t) E^+ (r, t) \right] = \langle E^- (r, t) E^+ (r, t) \rangle .
\] (2.14)

To connect this result with the interference experiment already discussed for the classical case, we neglect the spatial dependence from now on as we are only interested in the temporal dependence of the correlations. We notice that the measured intensity at the output of the Mach-Zehnder interferometer is proportional to the photon count rate at that point and

\[
E^\pm (t) = R T E^\pm_1 (t - \tau_1) + T R E^\pm_2 (t - \tau_2)
\] (2.15)

are the sums of the contributions from the individual arms with run times \( \tau_1, \tau_2 \). Inserting these contributions into (2.14) we find (again for stationary fields, i.e. no \( t \) dependence)

\[
\langle E^- (t) E^+ (t) \rangle = \langle E^- (t - \tau_1) E^+ (t - \tau_1) \rangle + \langle E^- (t - \tau_2) E^+ (t - \tau_2) \rangle + 2 \text{Re} \left[ \langle E^- (t - \tau_1) E^+ (t - \tau_2) \rangle \right]
\]

\[
= 2 |R|^2 |T|^2 \langle E^- (t) E^+ (t) \rangle \left\{ 1 + \text{Re} g^{(1)} (\tau) \right\},
\] (2.16)

with \( \tau = \tau_2 - \tau_1 \) and where we have introduced the normalized first-order quantum correlation function

\[
g^{(1)} (\tau) = \frac{\langle E^- (t) E^+ (t + \tau) \rangle}{\langle E^- (t) E^+ (t) \rangle} = \frac{\langle a^\dagger (t) a (t + \tau) \rangle}{\langle a^\dagger (t) a (t) \rangle} .
\] (2.17)

In the last equality we have inserted the electric field’s operator representation (2.12). The similarity to the classical case (2.8) and (2.9) is striking. Many results for correlation functions obtained with classical fields are reproduced in the quantum treatment, e.g. for thermal or coherent light fields. However, this is not the case when considering non-classical light sources, which can not be described in terms of classical fields.

**Second-order coherence**

While, in a classical picture, the first-order coherence function is linked to amplitude and phase fluctuations of the light field, second-order coherence relates to fluctuations of the intensity. It also contains information on the photon statistics of the light field and can thus be used to distinguish different states of light. In a classical Hanbury Brown and Twiss (HBT) setup the intensity of a light source at different times is correlated as schematically shown in Fig. 2.10. The second-order correlation function is defined accordingly for a stationary light field, i.e. \( \langle I(t) \rangle = \langle I(t + \tau) \rangle \), as

\[
g^{(2)} (\tau) = \frac{\langle I(t) I(t + \tau) \rangle}{\langle I(t) \rangle^2} .
\] (2.18)

A quantum description of the second-order correlation is derived in analogy to the first-order case. Now we are interested in the rate of coincidence detection events of a photon at detector 1 at time \( t \) and a second one at detector 2 at time \( t + \tau \). Experimentally this is realized in a HBT setup where, instead of intensity currents, the arrival times of single photons are detected [63]. Similar to Eq. (2.13) we write for the matrix element for the process of two-photon detection (neglecting the spatial dependence for clarity)

\[
\sum_f |\langle f | E^+ (t + \tau) E^+ (t) | i \rangle|^2 = \sum_f |\langle i | E^- (t) E^- (t + \tau) | f \rangle| \langle f | E^+ (t) E^+ (t + \tau) | i \rangle
\]

\[
= \langle i | E^- (t) E^- (t + \tau) E^+ (t + \tau) E^+ (t) | i \rangle .
\] (2.19)
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Figure 2.10: In a classical Hanbury Brown and Twiss (HBT) setup the intensity from a light source is split into two arms. The detected photo currents are multiplied (with a certain time delay $\tau$) and the average $\langle I(t) I(t + \tau) \rangle$ is taken. Normalized by the average intensity current $\langle I(t) \rangle$ the correlated intensities determine the two-photon correlation function $g^{(2)}(\tau)$. In the quantum version of the setup single photon detectors are used and the coincidence rate of two photons arriving with a delay $\tau$ at the two detectors is recorded.

The transition rate that we obtain from this matrix element will be in units of $(\text{time})^2$ as it describes the coincidence rate of photon detection (per unit time) at both detectors. We are interested in quantifying the degree of second-order coherence independent of the photon count rate at the detectors. Therefore, we define the normalized second-order quantum correlation function

$$g^{(2)}(\tau) = \frac{\langle E^-(t) E^-(t + \tau) E^+(t + \tau) E^+(t) \rangle^2}{\langle E^-(t) E^+(t) \rangle^2} = \frac{\langle a\dagger(t) a\dagger(t + \tau) a(t + \tau) a(t) \rangle}{\langle a\dagger(t) a(t) \rangle^2}. \quad (2.20)$$

In the last equality we have again inserted the expression (2.12) and the expectation values are evaluated according to (2.14). Note, that the normal ordering of the creation and annihilation operators as well as the particular ordering of time arguments naturally follows from the quantum theory of photon detection introduced in [62].

The second-order correlation function $g^{(2)}(\tau)$ is used to distinguish between different states of light according to their photon statistics. In this context, the value of $g^{(2)}$ at zero time delay

$$g^{(2)}(\tau = 0) = \frac{\langle a\dagger a\dagger a a \rangle}{\langle a\dagger a \rangle^2} = \frac{\langle n^2 \rangle}{\langle n \rangle^2} - \frac{1}{\langle n \rangle} \quad (2.21)$$

is of special interest. It is related to the probability of detecting two photons at the same time at the two arms of the HBT setup. For uncorrelated light sources, the rate of two photon detection events will be identical to the product of detection rates at the individual detectors. In that case the numerator of the correlation function (2.20) factorizes and we find $g^{(2)}(\tau) = 1$ for all time delays $\tau$. If, on the other hand, the rate of two-photon detections is increased relative to two uncorrelated light sources this is referred to as photon bunching ($g^{(2)}(0) > 1$). If photons are emitted individually, i.e. one after the other, this is called antibunching ($g^{(2)}(0) < 1$). Using these distinctions we discuss three basic states of the light field.

Coherent light is generated in the stimulated emission process of a laser. In the classical picture, it corresponds to the state of light that is represented by a stable wave with no fluctua-
Figure 2.11: Photon number distributions for coherent light with Poissonian photon statistics (upper), thermal light (middle) and Fock state distributions (lower) for increasing average photon number $\langle n \rangle = 1, 5, 10$. 

In the quantum description, however, also coherent light shows quantum fluctuations of the photon number and thus of the intensity. However, these fluctuations are minimal among all possible states, satisfying the Heisenberg uncertainty principle. Photons from a coherent light source are emitted completely uncorrelated. Its second-order correlation function is thus given by $g^{(2)}(\tau) = 1$. It is defined by the photon state (in the Fock basis) \[ |\alpha\rangle = e^{-|\alpha|^2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle, \tag{2.22} \]

where $\alpha = |\alpha|e^{i\theta}$ give the fixed amplitude and phase of the coherent state. Its photon statistics follows a Poissonian distribution given by

\[ p(n) = |\langle n|\alpha\rangle|^2 = e^{-\langle n \rangle} \frac{\langle n \rangle^n}{n!}, \tag{2.23} \]

with an average photon number given by $\langle n \rangle = |\alpha|^2$. The upper panel in Fig. 2.11 shows Poisson distributions for different mean photon numbers. The distribution is generally peaked around the mean and the variance increases proportional to the mean photon number as well. In statistics, Poisson distributions are related to the occurrence of independent events. It thus reflects the emission property of coherent light sources to emit photons completely uncorrelated from each other.

**Thermal light** is generated by a medium that emits photons in a random fashion by spontaneous emission, creating strong fluctuations of the light’s intensity. On the level of individual photons this results in photon bunching, i.e. a higher probability that two photons are emitted at the same time. An intuitive picture for this effect can be given in terms of the intensity...
fluctuations. If one photon is detected at time \( t \) the intensity is more likely to be higher than
the average at that moment which in turn also increases the probability to detect a second
photon at the same time or shortly after. A quantum mechanical argument for the bunching
of thermal photons was given by Fano [64] in terms of interference of which-path information
for indistinguishable particles, which directly shows the bosonic nature of photons (fermions
would show antibunching in the same type of experiment [65]). Thermal light is a state that
can be described as being in thermal equilibrium with a black body at inverse temperature
\( \beta = 1/(k_B T) \). It has a photon statistics that follows a Bose-Einstein-distribution

\[
p(n) = \frac{\langle n \rangle^n}{(\langle n \rangle + 1)^{n+1}}, \tag{2.24}
\]

from which follows for the two-photon correlation function \( g^{(2)}(0) = 2 \). Fig. 2.11 (middle panel)
shows thermal distributions for different average photon numbers. Notably, \( |0\rangle \langle 0| \) is the most
probable state irrespective of the average photon number. This reflects the higher fluctuations
of thermal states with respect to coherent ones.

\[
g^{(2)}(\tau)
\]

Figure 2.12: two-photon correlation function \( g^{(2)}(\tau) \) as a function of delay time \( \tau \) for states of
light with different photon statistics. For coherent light photons are emitted from the source
completely uncorrelated which is reflected in \( g^{(2)}(\tau) \) being 1 for all time delays. Thermal light
shows photon bunching with \( g^{(2)}(0) > 1 \) and thus an increased probability of two photons being
emitted at the same time. While \( g^{(2)}(0) \) can in principle take any value larger than one also for
classical fields, only non-classical light states allow for \( g^{(2)}(0) < 1 \), referred to as antibunching.
All types of light loose their correlations for sufficiently large \( \tau \) and \( g^{(2)}(\tau) \) approaches 1 in all
cases.

**Non-classical** states of light are those which can not be described by classical electromagnetic
fields as it is the case for coherent or thermal states. They require a quantum description
of light. One type of non-classical states is characterized by a two-photon correlation function
showing antibunching \( g^{(2)}(0) < 1 \), i.e. the probability of detecting two photons at the same
time is reduced. The best example are Fock states $|n\rangle$ with a fixed photon number $n$ satisfying

$$g^{(2)}(0) = 1 - \frac{1}{n}. \quad (2.25)$$

In the limit of single photon emitters ($n = 1$) only individual photons are emitted, the probability of detecting two photons at the same time is zero and $g^{(2)}(0) = 0$. This situation can be realized by single emitter systems like single quantum dots, inserted into photonic structures [66], or single nitrogen vacancy centers in diamond [67]. The lower panel of Fig. 2.11 shows Fock state photon number distributions which are basically Kronecker-$\delta$ functions.

Fig. 2.12 shows the delay-time dependent behavior of $g^{(2)}(\tau)$ for thermal, coherent and non-classical states of light. Starting from the initial value, that is the fingerprint of each state respectively, $g^{(2)}(\tau)$ approaches 1 in all cases as the photon emission events become completely uncorrelated for sufficiently long time delays. In this thesis we are primarily concerned with light in the thermal and coherent regimes as these characterize the light emission process of a laser below and above threshold. Non-classical states of light are found primarily in the regime of single photon emitters or in the context of cavity QED for the study of entanglement and applications in quantum information [68].

### 2.6 Quantum regression theorem

An important tool in quantum optics is the quantum regression theorem (QRT) [69]. Given certain assumptions, it allows to calculate two-time correlation functions of operators based on the dynamics of individual (single time) operators. In the previous section we discussed the photon-correlation functions $g^{(1)}(\tau)$ and $g^{(2)}(\tau)$ with dynamics given as a function of the time difference $\tau$ from the underlying two-time correlations that are accessible via the QRT. Classically, the regression theorem (or hypothesis) [70] relates microscopic fluctuations in the system to macroscopic dynamics, e.g. spatial correlations of temperature fluctuations in a material obey the same dynamics as the heat diffusion itself. In this thesis we deal with the quantum case, i.e. with photon-correlation functions that quantify fluctuations in the laser emission. For Markovian, open quantum systems, i.e. systems that are weekly coupled to a bath and stay approximately uncorrelated with it, the QRT can be applied [61]. Assume a complete set of operators $\{A_\mu\}$ acting on a system’s state $\rho$ in the sense that their expectation value for a system is given by

$$\frac{d}{dt}\langle A_\mu(t) \rangle = \sum_\nu M_{\mu\nu} \langle A_\nu(t) \rangle,$$  \quad (2.26)

with constant coefficients $M_{\mu\nu}$. In this case the QRT states that the two-time correlation function $\langle A_i(t) A_\mu(t + \tau) A_j(t) \rangle$ follows a dynamics with respect to $\tau$ according to

$$\frac{d}{d\tau}\langle A_i(t) A_\mu(t + \tau) A_j(t) \rangle = \sum_\nu M_{\mu\nu} \langle A_i(t) A_\nu(t + \tau) A_j(t) \rangle.$$  \quad (2.27)

We note that for Markovian systems the quantum regression theorem is exact and is found to mirror the classical case [71]. For systems that follow the Lindblad-von Neumann equation, the Born-Markov approximation is applied and thus the QRT is a valid as long as the assumptions of the Born-Markov approximation hold [72].
Chapter 3

Rethinking the lasing threshold

The laser threshold is a central concept in laser theory. It is well defined for conventional lasers as the pump power where the laser starts to emit coherent light and lowering the threshold pump power is an established avenue to achieve higher energy efficiency in this regime. In Section 2.3 we introduced laser rate equations and showed how they give rise to the laser threshold. In particular, with spontaneous emission coupling factors $\beta$ of the order $10^{-3}$ and smaller, conventional lasers show a distinct phase transition like threshold that can be unambiguously identified. Here we are revisiting the notion of the laser threshold in the context of the unique properties of nanolasers. In nanolasers quantum optical effects are mediated by high field intensities in the optical cavity. They can change the spontaneous emission (SE) of the gain medium and lead to $\beta$ factors close to the ideal limit of 1. This is often associated with an increase in energy efficiency as the threshold jump in the output intensity shifts to lower pump powers as $\beta \rightarrow 1$. Identifying the laser threshold from the I/O characteristics becomes ambiguous in this regime as the threshold jump also becomes more gradual and the threshold appears to “smear out” over a range of pump values.

To amend this, alternative definitions for the laser threshold were put forward like the quantum threshold at an intracavity photon number of 1 [44]. Nevertheless, an inherent issue remains, i.e. that from the average output intensity, by itself, it is difficult to infer the state of the emitted light or gain information about its photon statistics. At the same time, unambiguous proof of lasing can be given by measuring a change of the photon statistics of the output directly. For lasers, this is mostly not feasible in practice as photon number resolving detectors only work for smallest output intensities and have long dead times [73]. Therefore it has become an established method to measure changes in the two-photon correlation function $g^{(2)}(0)$, introduced in Section 2.5, as it serves as a fingerprint of the light’s photon statistics. This allows to detect indirectly the transition from thermal to Poissonian photon statistics as stimulated emission starts to dominate the emission properties of the laser. In the literature there are a number of reports on high-$\beta$ nanolasers with gain media consisting of discrete emitters (mostly QDs) where measurements of second-order photon correlations show the onset of coherent emission [43, 49, 52]. In this regime, the assessment of the lasing threshold in terms of the photon correlations seems to agree with the quantum threshold criterion [42]. More recently, 2D gain media like QWs, which are ubiquitous in conventional lasers, and more novel TMD monolayers were combined with ultrasmall micro- and nanocavities, pushing these QW based devices into the cavity-QED regime as well [13, 14, 15, 20, 21, 22, 23, 50].

In this chapter we examine the threshold behavior of high-$\beta$ nanolasers in terms of their coherence properties in a systematic way by using an idealized laser model that nevertheless captures the relevant effects. We distinguish between device regimes of nanolasers with QD
and extended 2D gain media. On the one hand we confirm in this chapter what can already be found in the literature [48], namely that the absence of a visible threshold in the output intensity does not imply zero-threshold lasing, i.e. coherent emission at arbitrary small pump powers. Instead, the transition to coherent emission always occurs at finite pump powers. On the other hand we show that nanolasers with QD and extended 2D gain media have different behavior regarding their lasing transition in relation to the quantum threshold criterion, that is rendered invalid under certain conditions for high-β nanolasers with extended gain media.

The discussion and results presented in this chapter have led to our publication [74]. Other publications discuss similar aspects of nanolasers and, albeit applying different methods, come to conclusions in agreement with our work for the discussed device regimes [14, 75, 76].

3.1 An idealized laser model

The laser model that we introduce here serves the purpose of investigating the influence of important device parameters while retaining a level of simplicity that allow to derive analytic expressions. In particular, we are interested in the interplay of radiative losses, cavity decay rate, and the amount of gain medium that interacts with a single cavity mode. The laser model that provides this and which we consider in the following is an implementation of the Tavis-Cummings Hamiltonian [77], describing \( N \) identical two-level emitters interacting with a single mode of an optical cavity. Additionally, dissipating terms (pump, radiative and cavity losses) are described in the Lindblad-formalism [72]. Fig. 3.1 shows a schematic picture of the model system.

We are using our laser model to compare high-β nanolasers that are QD based with those that use extended (2D) gain media and to investigate the role of spontaneous emission in these different device regimes. As QDs as gain medium are discrete, the analogy to the model is quickly established as \( N \) is the number of quantum dots that are in resonance with the cavity mode (typically 50-100 [49, 78]). Extended (2D) gain media exhibit a continuous density of states with large numbers of electron-hole pairs at different energy levels in the band structure. Nevertheless we can estimate the number of electron-hole pairs in an excited gain medium that resonantly interact with the cavity mode, given typical carrier densities and the extent of the gain medium’s active region (on the order of \( 10^4 \)). Although this simplified picture neglects effects like many-body interactions or carrier mobility in the gain medium, it is still suited to describe the effects discussed here. As we will see, the influence of spontaneous emission into the lasing mode is one of the key factors in understanding the coherence properties of the laser emission. As this influence is primarily determined by system parameters like the amount of gain medium \( (N) \), cavity decay rate, and the β factor, our simplified laser model captures the relevant mechanisms. The parameters that we are going to use in the next sections correspond to typical parameters found in the literature for QD based nanolasers [49] and nanolasers with 2D gain media [23].

Hamiltonian

The Hamiltonian of the \( N \)-emitter Tavis-Cummings model is given by (with \( \hbar = 1 \))

\[
H = \omega_c \sum_i c_i^\dagger c_i + \omega_b b^\dagger b + \sum_i \left[ g b^\dagger v_i^\dagger c_i + g^* b c_i^\dagger v_i \right],
\]

where \( b^\dagger, b \) are creation and annihilation operators for the photon mode, \( c_i^\dagger, c_i \) for the upper (excited) emitter states and \( v_i^\dagger, v_i \) for the lower (valence band) ones. We assume a single carrier
Figure 3.1: Sketch of the $N$ two-level systems interacting with a single cavity mode (Tavis-Cummings-Model). The cavity mode’s coherent interaction (with rate $g$) with the emitters is resonant with the transition of the two levels ($|1\rangle \leftrightarrow |2\rangle$). Excitation of the emitters by an external pump (rate $P$) as well as dissipation due to radiative losses ($\gamma$) and cavity decay ($\kappa$) is described in the Lindblad formalism.

Nanolasers are examples of open, dissipative quantum systems as they rely on an external energy supply and are subject to losses. Cavity decay, radiative losses, and incoherent pumping of the
emitters are described as dissipative processes by the Lindblad-master equation \([72]\). However, instead of describing the dynamics of the system’s full density matrix we calculate expectation values for relevant operators. In the Heisenberg picture, the equation of motion (EoM) for the expectation value of an operator \(A\) is given by

\[
\frac{d}{dt} \langle A \rangle = -i \langle [A, H] \rangle + \sum_{\alpha} \frac{\lambda_{\alpha}}{2} \left( \langle L^\dagger_{\alpha} A L_{\alpha} + L^\dagger_{\alpha} [A, L_{\alpha}] \rangle \right), \tag{3.4}
\]

where \(\lambda_{\alpha}\) is the rate associated to the dissipative process defined by the operator \(L_{\alpha}\). Radiative losses by spontaneous emission in each emitter takes place with rate \(\lambda_{\alpha} = \gamma\) and the corresponding operators \(L_{\alpha} = v_i^\dagger c_i\) describes the transition from the excited to the lower emitter level. The Lindblad term for the cavity losses have rate \(\kappa\) and operator \(b\) and an incoherent pump excites emitters at rate \(P\) where the corresponding operators are \(L_{\alpha} = c_i^\dagger v_i\). With that the whole master equation becomes

\[
\frac{d}{dt} \langle A \rangle = -i \langle [A, H] \rangle + \gamma \sum_i (2c_i^\dagger v_i A v_i^\dagger c_i - c_i^\dagger v_i v_i^\dagger c_i A - A c_i^\dagger v_i v_i^\dagger c_i) + P \sum_i (2v_i^\dagger A c_i^\dagger v_i - v_i^\dagger c_i c_i^\dagger v_i A - A v_i^\dagger c_i c_i^\dagger v_i) + \kappa \left( 2b^\dagger A b - b^\dagger b A - A b^\dagger b \right). \tag{3.5}
\]

### 3.2 Equations of motion

In this section we derive equations of motion for the relevant system characteristics, including correlation functions, relying on the cluster expansion approach. The \(g^{(2)}(0)\) function would, in principle, most directly be determined from the photon statistics, i.e. the diagonal elements of the reduced density operator of the cavity system \(\rho_c\), by tracing out the emitters’ degrees of freedom \(\rho_c = \text{Tr}_e [\rho]\) of the cavity. However, we rely on the equation of motion approach for operator expectation values, as the calculation of the full density matrix is not feasible for systems of the size of a laser. For technical details of the following we refer to Appendix A on the cluster expansion approach. The infinite hierarchy of equations that arise from this approach is truncated at the quadruplet level. This is necessary in order to access lasing characteristics and especially photon correlations as given by \(g^{(2)}(0)\). Higher levels of the truncation would allow for calculation of higher order correlation functions as well, but the resulting equations become increasingly complex. Other approaches, used in the literature, include the use of Langevin forces \([75, 79]\) or master equation approaches together with stochastic evaluation methods \([14, 80]\).

### Doublet level

Before considering \(g^{(2)}(0)\) we calculate, in a first step, quantities on the doublet level of the cluster expansion. From Eq. (3.5) we start by calculating EoM for the photon number \(n = \langle b^\dagger b \rangle\) and the upper level population \(f = \langle c_i^\dagger c_i \rangle\) (note that \(f\) does not carry the subscript \(i\) as all
emitters are identical)

\[ \dot{n} = -\kappa n + 2N \Re \psi_1, \quad (3.6a) \]

\[ \dot{f} = -\gamma f + P(1 - f) - 2 \Re \psi_1. \quad (3.6b) \]

Both quantities couple to the real part of the photon-assisted polarization \( \psi_1 = -ig (b^\dagger v^\dagger_i c_i) \). The EoM for \( \psi_1 \) brings in higher order expectation values, as discussed in Appendix A, giving rise to the hierarchy of equations. It is given by

\[ \dot{\psi}_1 = g \left( \langle b^\dagger b c_i^\dagger c_i \rangle - \langle b^\dagger b v_i^\dagger v_i \rangle + \sum_{i \neq j} \langle c_j^\dagger v_j^\dagger v_i c_i \rangle \right) - \frac{1}{2} \Gamma \psi_1. \quad (3.7) \]

The first two terms in (3.7) represent correlations between photon number and emitter populations. They are factorized according to the cluster expansion scheme as

\[ \langle b^\dagger b c_i^\dagger c_i \rangle = \langle b^\dagger b \rangle \langle c_i^\dagger c_i \rangle + \delta \langle b^\dagger b c_i^\dagger c_i \rangle = nf + \delta c, \]

\[ \langle b^\dagger b v_i^\dagger v_i \rangle = \langle b^\dagger b \rangle \langle v_i^\dagger v_i \rangle + \delta \langle b^\dagger b v_i^\dagger v_i \rangle = n(1 - f) + \delta v, \quad (3.8) \]

reducing them to products of lower order expectation values and pure correlation functions \( \delta(\ldots) \) defined in Appendix A. The third term in (3.7) reduces to \( f \) due to the one-electron assumption, while the sum represents correlation among different emitters. These interemitter correlations come into play when one is interested in superradiant effects [52, 81, 82] but we neglect them in the following (see also Appendix B.2). The last term in (3.7) corresponds to a dephasing with rate \( \Gamma/2 = (P + \gamma + \kappa)/2 \) due to the dissipative terms.

**Rate equation approximation**

At this level it is possible to apply the rate equation approximation [83] by adiabatically eliminating the polarization (3.7) and inserting it into Eqs. (3.6). In combination with the factorized result (3.8) and neglecting the correlation terms \( \delta c, \delta v \) this yields

\[ \dot{f} = -R \left[ n(2f - 1) + f \right] - \gamma f + P(1 - f), \quad (3.9a) \]

\[ \dot{n} = RN \left[ n(2f - 1) + f \right] - \kappa n, \quad (3.9b) \]

where we defined the rate of spontaneous emission into the cavity mode

\[ R = \frac{4|g|^2}{P + \gamma + \kappa}. \quad (3.10) \]

The \( \beta \)-factor is defined as the ratio of the spontaneous emission into the laser mode \( R \) to the total spontaneous emission including losses, i.e.

\[ \beta = R/(R + \gamma). \quad (3.11) \]

These rate equations are similar to those introduced in (2.4). However they are modified by the fact that the number of emitters is finite and thus the available gain is limited by \( RN \) and we find Pauli-blocking (\( \propto (1 - f) \)) in the pump term. Furthermore, the single-electron assumption changes the dependence of the spontaneous emission rate on the carrier excitation from quadratic to linear (\( \propto f \)).
3.3 Intensity threshold and coherence threshold

We can easily evaluate the rate equations in their steady state as a function of pump power and for different $\beta$ factors. The results are shown in Fig. 3.2 a) and Fig. 3.3 a). Both cases show the same behavior: With increasing $\beta$ the threshold non-linearity shifts to lower pump powers and gradually vanishes.

### Intensity threshold

Working with the stationary solutions of (3.9) we can derive an analytic expression for the pump power of the intensity jump as outlined in Appendix B.1. We refer to it as the intensity threshold. It obeys the equation

$$P_{th} - \gamma - (P_{th} + \gamma) \frac{\kappa}{RN} = 0$$

and is indicated with dashed vertical lines in Figs. 3.2 and 3.3. It is located exactly in the middle of the corresponding jump intervals of the output intensities and represents the change in scaling behavior of the photon number with the system size $N$, i.e. the photon number is proportional to $N$ above threshold while independent of $N$ below. As $\beta$ approaches unity, $P_{th}$ goes to zero, implying thresholdless lasing often referred to in the literature [9, 11, 13]. However, it is well established that coherent emission will take place at finite excitation even in the limiting case $\beta = 1$ [78]. In the next section we investigate this alternative notion for the different device regimes discussed here.

Note that due to excitation-induced dephasing, the $\beta$ factor here is pump power dependent as can be seen from Eqs. (3.10) and (3.11). In Figs. 3.2 and 3.3 we changed the rate of spontaneous emission into non-lasing modes $\gamma$ for different curves. Nevertheless, since a constant $\beta$ factor is well established throughout the literature to characterize laser efficiency, we choose to use $\beta$ rather than $\gamma$ to label the results. To that end, all given values of $\beta$ are evaluated at $P = P_{th}$.

While non-constant $\beta$ factors have been investigated in different contexts [84, 85, 86], our approach is well suited in the context of this work and makes the results more comparable.

### Coherence threshold

With no distinct threshold non-linearity in the output intensity of high-$\beta$ lasers, one has to rely on statistical properties of the emission in order to identify lasing as indicated before. But even when a threshold jump is visible we are interested in the coherence properties and to check, whether or not it reflects the behavior of the intensity threshold.

Accessing the second-order autocorrelation function requires to extend the EoM beyond the rate equation level, using the cluster expansion at least up to the quadruplet level. We write $g^{(2)}(0)$ in terms of the pure correlation as

$$g^{(2)}(0) = \frac{\langle b^\dagger b^\dagger bb \rangle}{n^2} = 2 + \frac{\delta \langle b^\dagger b^\dagger bb \rangle}{n^2}.$$  

(3.13)

We can access $\delta \langle b^\dagger b^\dagger bb \rangle$ by keeping the correlation terms $\delta c_1$, $\delta v_1$ in (3.8) and writing EoM for these and other higher order terms including $\delta \langle b^\dagger b^\dagger bb \rangle$. In Appendix B.2 we give an outline of the full derivation where we make use of the fact that all emitters are independent, which greatly simplifies the equations.

Considering only stationary solutions of the system \( \frac{d}{dt} \ldots = 0 \) the system of equations becomes algebraic and, interestingly, we can derive an analytic expression for the two-photon
3.3. INTENSITY THRESHOLD AND COHERENCE THRESHOLD

Figure 3.2: a) Output intensity and b) $g^{(2)}(0)$ values according to Eqs. (3.9) and (3.14) are shown for different values of $\beta$. The parameters $N = 100$, $\kappa = 0.04 \text{ ps}^{-1}$, and $g = 0.03 \text{ ps}^{-1}$ correspond to typical high-Q QD nanolasers [49]. The intensity thresholds are marked as dashed lines. In this parameter regime, they nearly coincide with the threshold to coherent emission, which are indicated by the shaded regions for each value of $\beta$.

The correlation function

$$g^{(2)}(0) = 2 - \frac{R}{\kappa} \left( 2n + 1 \right) \left( n + f \right) + \frac{2\kappa}{N} \right)$$

Fig. 3.2 b) shows the evaluation of Eq. (3.14) for the same set of parameters as in panel a). At low excitation levels, $g^{(2)}(0)$ has a value of 2 for all values of $\beta$. This reflects the regime of thermal emission that is dominated by random, spontaneous emission processes and that exhibits no phase coherence. The transition of $g^{(2)}(0)$ towards 1 (marked by the shaded regions) indicates the onset of coherent emission. We define the coherence threshold $P_{\text{coh}}$ as the point, where $g^{(2)}(0)$ crosses a fixed value close to 1. Here we choose $g^{(2)}(0) = 1.1$ as this threshold value, which is to a certain extent arbitrary, however, a lower value would be impractical given the gradual nature of $g^{(2)}(0)$. Depending on the application in mind and the required quality of the coherence, also other threshold values could be defined, while the conclusions drawn from our discussion here will remain the same. It is important to point out that an unique definition of $P_{\text{coh}}$ is only possible for $\beta \to 0$, corresponding to the thermodynamic limit, where the threshold behaves as a second-order phase transition [44]. Nanolasers with large values of $\beta$ operate far from the thermodynamic limit and $g^{(2)}(0)$ approaches 1 gradually over wide range of excitation powers.

In Fig. 3.2 we have chosen parameters typical for QD laser devices. The coherence threshold $P_{\text{coh}}$ (shaded areas) closely follows the intensity threshold (marked by the vertical lines). This
CHAPTER 3. RETHINKING THE LASING THRESHOLD

Figure 3.3: For a nanolaser with an extended gain material, a) output intensity and b) $g^{(2)}(0)$ values are shown for different $\beta$. The large carrier number in the gain media is reflected in $N = 3 \times 10^4$, whereas the other parameters are $\kappa = 0.7$ ps$^{-1}$, $g = 0.02$ ps$^{-1}$. For all values of $\beta$, the intensity threshold (dashed vertical lines) is clearly separated from the transition to coherent emission (shaded area). Furthermore, the coherence threshold indicates that increasing $\beta$ has no benefit in terms of reducing the pump power to reach coherent emission.

behavior corresponds to the common conception of the intensity threshold being interlinked with the transition to coherent emission. Only for $\beta = 1$ the intensity threshold vanishes and solely $g^{(2)}(0)$ indicates the existence of a transition. Also, with an increase of $\beta$, the pump power interval of the $g^{(2)}(0)$ transition shifts to lower pump powers and thus offers a threshold power reduction, which is one of the most prominent effects sought after in high-$\beta$ devices.

3.4 A new lasing regime

For parameters corresponding to nanolasers with extended gain media, the $g^{(2)}(0)$ transition shows a different picture, as shown in Fig. 3.3. In comparison to the previous case of QD gain media, the coherence threshold $P_{\text{coh}}$ (shaded area) is strongly offset to higher pump rates from the intensity jump (vertical lines). Here the coherence and intensity thresholds are no longer aligned. Coherent emission is reached at the same pump rate irrespective of the value of $\beta$, which implies that for devices with extended gain media a decrease of the threshold current is no longer possible by reducing radiative losses (i.e. increasing $\beta$). These results lead to a reevaluation of the role of $\beta$ in terms of energy efficiency in this new device regime.

Nanolasers that are QD based require large $Q$ factors ($\lesssim 10^4$) in order to facilitate lasing. With a lower number of emitters the available gain is limited and thus defines an upper bound on the cavity losses $\kappa$. In those high-$Q$ systems, radiative losses typically dominate cavity losses...
and, as a result, the threshold pump power strongly depends on $\beta$. In contrast, the peculiar behavior of nanolasers with extended gain media that we observed in Fig. 3.3 can be attributed to the distinct parameter regime they operate in. Room-temperature lasing from the extended gain media, as in Refs. [15] and [20], is reported with relatively low $Q$ factors ($\approx 2500$). This is possible due to the large number of carriers contributing to the gain. Therefore, in the high-$\beta$ regime, radiative losses become negligible in comparison to cavity losses. In effect, the roles of radiative and cavity losses are interchanged with respect to their influence on the coherence threshold.

This relation is depicted in Fig. 3.4 where the pump power of the coherence threshold $P_{coh}$ is shown as a function of cavity loss rate $\kappa$ for different values of $\beta$. The case of nanolasers with extended gain media (solid lines) is compared with the case of QD based gain (dashed lines). Parameters are the same as in Figs. 3.2 and 3.3, respectively. The corresponding cavity loss rates are indicated by vertical lines (black) in the plot. For QD gain media a substantial dependence of the coherence threshold on $\beta$ can be seen. The lines terminate at the maximal value of $\kappa$. Beyond that, the small number of emitters does not facilitate lasing any more as the maximal gain can not compensate the losses in that case. For extended gain media the transition between two regimes is visible. For small cavity loss rates the same bahavior as in the QD case is observed, i.e. a dependence on $\beta$ and vanishing dependence in $\kappa$ (lines converge to a constant). Only in the case of $\beta = 1$ the threshold is not limited from below and can in principle brought to zero, however this corresponds to an idealized situation with zero losses in the system that is not physical.

For increasing cavity losses the dependence on $\beta$ vanishes as radiative losses become less dominant in comparison. For the loss rate we considered in Fig. 3.3 all values with $\beta > 0.1$ become indistinguishable. We note at this point that all of our consideration relate to $\beta$ factors of 0.1 and larger, what is already high in the context of conventional lasers where $\beta$ is orders of magnitudes smaller. Conventional lasers, e.g. VCSELs or gas lasers, feature high number of emitters as well, however, they operate in a regime where radiative losses by far dominate those due to the cavity and their threshold behavior is clearly dominated by $\beta$.

More insight in the difference between the discussed lasing regimes is revealed when we combine the information in Figs. 3.2 and 3.3 into a depiction of $g^{(2)}(0)$ as a function of the mean photon number $n$. For the parameters of the extended gain media we show the results in Fig. 3.5 (solid lines). We see that the transition to coherent emission is solely determined by the mean intracavity photon number, irrespective of the value of $\beta$, which reflects the previous findings in this particular device regime. Furthermore, coherence is reached around $n \approx 1000$, which greatly exceeds the quantum threshold criterion given by $n = 1$ [42, 47, 52] as previously mentioned. In comparison, for few-emitter nanolaser the results are shown as dashed lines in Fig. 3.5. Here, the coherence threshold is reached at significantly lower mean photon numbers ($n \approx 10$ for $\beta \to 1$) and one observes a clear reduction in the photon number necessary to reach coherence with increasing $\beta$.

The origin of the delayed onset of coherence and the large required photon number can be understood by considering the different contributions of spontaneous and stimulated emission in the system. To observe coherence, stimulated emission has to dominate in order to quieten intensity noise introduced by spontaneous emission events. Both contributions, stimulated ($\propto n(2f - 1)$) and spontaneous emission ($\propto f$) scale with the amount of gain material $N$. In the lasing regime the population inversion $2f - 1$ is approximately given by $2f_L - 1 = \kappa/(2RN)$, i.e. it decreases with the emitter number $N$. Corrections to this approximation are provided by the right-hand side of Eq. (B.3) in the Appendix, which is again of the order $1/N$. This is the reason that for large numbers of emitters also large photon numbers are required in order for stimulated emission to dominate the spontaneous contributions. Technically, this behavior
CHAPTER 3. RETHINKING THE LASING THRESHOLD

Figure 3.4: Pump rate $P_{\text{coh}}$ at the coherence threshold as a function of cavity loss rate $\kappa$. Parameters $(N, g)$ correspond to those in Figs. 3.2 (dashed) and 3.3 (solid). Vertical lines indicate respective values for $\kappa$. We observe a different dependence on $\beta$ for the two cases as well as the transition between the regimes for varying $\kappa$. For a small number of emitters there is an upper bound for $\kappa$, beyond which lasing can not be achieved and thus the dashed lines terminate above a certain value.

Figure 3.5: Two-photon correlation function $g^{(2)}(0)$ as function of the intracavity photon number $n$. For high-$Q$ quantum-dot nanolasers (dashed lines) an increasing $\beta$ factor leads to fewer photons required for coherent emission. For a high number of emitters and lower cavity $Q$ (solid lines) coherent emission happens at much larger photon numbers and independent of $\beta$. 
3.4. A NEW LASING REGIME

is also present in conventional lasers (such as gas and edge-emitting lasers) but is rendered irrelevant and goes unnoticed as the transition to coherent emission occurs sharply on a narrow interval in such devices due to \( \beta \) factors that are orders of magnitudes smaller than those considered here.

In a last step, we turn back to the analytic expression (3.14). By using the stationary values of \( f \) and \( n \), obtained from the rate equations (3.9), we can eliminate the carrier population \( f \) and find an expression for \( g^{(2)}(0) \) that only depends on the intracavity photon number

\[
g^{(2)}(0) = 2 - \frac{R(2n + 2) + \frac{3\kappa}{\gamma} + \frac{1}{\kappa} NR + \kappa}{R(2n + 1) + \kappa + \frac{1}{\kappa} NR + \frac{3\kappa}{\gamma} }. 
\]  

(3.15)

Interestingly, this equation provides access to the statistical properties of the emitted light even if photon numbers obtained from rate equations are used. It might be helpful for analyzing future device realizations in term of its coherence properties. Inserting the rate equation results into Eq. (3.15) is in principle an approximation, as it corresponds to the doublet level of the cluster expansion. However, the correlation terms beyond the doublet level represent only small corrections to \( n \) and \( f \) in the present case. Therefore, this approximate treatment agrees well with the solution of the quadruplet equations to obtain \( g^{(2)}(0) \), which is shown in Fig. 3.6.

![Figure 3.6: Photon autocorrelation function \( g^{(2)}(0) \) obtained by using full quadruplet (solid, Eq. (3.14)) and rate-equation (circles, Eq. (3.15)) populations agree well showing the suitability of the rate equation approximation made in the derivation of Eq. (3.15). The parameters used are the same as in the previous Figures for high-\( Q \) QD nanolasers (left) and lower-\( Q \) nanolasers with extended gain medium (right).](image)

In conclusion, we have seen how in high-\( \beta \) nanolasers with extended gain media the lasing threshold in terms of the coherence properties is no longer determined by \( \beta \). Instead, the cavity losses become more dominant in determining the threshold behavior. This gives important direction for the design choices of future nanolaser realizations, where cavity geometries should be optimized towards higher quality factors \( Q \) instead of higher values of \( \beta \). On the other hand, the benefit of high-\( \beta \) nanolasers in terms of \( n = 1 \) at threshold and low threshold pump powers due to increasing \( \beta \) rely on being close to the few-emitter limit of lasing. Up to date only a small
number of publications report on measurements of $g^{(2)}(0)$ on high-$\beta$ nanolasers with extended gain media [14, 15, 50], some of which we will discuss in the following chapters. Especially in the field of TMD-based nanolasers employing atomically thin gain materials, direct experimental verification of coherent emission is still pending. Future reports will hopefully shed more light on this novel device regime.
Chapter 4

Monolayers of transition metal dichalcogenides as laser gain media

Single layers of transition metal dichalcogenides (TMD) exhibit unique properties. Similar to graphene, they form two-dimensional layers of atomic thickness in which strong 2D carrier confinement takes place. Their reduced dimensionality and strong electric dipole moments lead to strong light-matter interaction in the material, which moved them into focus for possible optoelectronic device applications. In this context, single layers of semiconducting TMDs are also investigated as a novel type of nanolaser gain material [25, 87] promising large quantum efficiency and reduced device dimensions.

With extended, two-dimensional gain media, TMD-based nanolasers fall into the class of devices that may exhibit a delayed onset of coherent emission in relation to the intensity threshold as we discussed in the previous chapter. Up to now, several groups have reported on realizations of nanolasers with optical gain from monolayer TMDs [20, 21, 22, 23]. However, experimental verification of coherent emission was not presented and lasing was inferred from the I/O characteristics and linewidth narrowing, only.

In most of the mentioned reports, the theoretical analysis of the laser characteristics was based on standard rate equation models. As we will see in the following, strong Coulomb interactions and weak dielectric screening due to the reduced dimensionality of TMD monolayers lead to pronounced material-specific effects in the optical response of the monolayer, rendering standard rate equation approaches unsuited for describing their performance as laser gain material. To amend this, we will combine results from microscopic gain calculations for highly excited TMD materials with a modified rate equation model that takes into account the material-specific gain effects. Furthermore, we will emphasize the necessity of correlation measurements for verification of coherent emission in those devices. The discussion in this chapter is based on our previous publication [88].

4.1 Atomically thin semiconductors

Two-dimensional materials are large class of materials that form atomically thin layers with many interesting properties. Graphene is certainly the most prominent member of the group. However, there exists a great range of materials that fall in different material classes [89]. While some materials form electronic insulators (e.g. hBN [90]) or metals (TaS2 [91]), also semiconductors can be found. Among these certain transition metal dichalcogenides (TMDs) attracted a lot of attention recently due to their interesting optoelectronic properties [92, 93, 94, 95]. In the single layers (monolayers), transition metal atoms (Mo or W) are sandwiched
CHAPTER 4. MONOLAYERS OF TRANSITION METAL DICHALCOGENIDES AS LASER GAIN MEDIA

Figure 4.1: a) Schematic view of individual TMD layers [96]. Transition metal atoms (black) are sandwiched between two layers of chalcogen atoms (yellow) connected by strong covalent bonds. Individual layers have a typical separation in the sub-nanometer range and are connected by weaker van-der-Waals interaction. Due to the weak interlayer bonding, single layers can be mechanically exfoliated from the bulk material quite easily. b) Transition metals (M) and chalcogenides (X), with compounds of the form MX₂ that are the most studied in the context of semiconducting 2D materials due to their strong optical activity and unique electronic properties as a monolayer.

between two layers of chalcogen atoms (S, Se, or Te) to form the 2D sheets as depicted in Fig. 4.1 a). In the following, when discussing TMD monolayers, we refer to these semiconducting material combinations.

Atoms inside the layer are held together by strong covalent bonds while different layers are only weakly bound by van-der-Waals interactions. As a result, single layers of TMD can be exfoliated from the bulk material and deposited on a substrate, making them accessible for examination of their optoelectronic properties [97] and for device integration [96, 98]. Fig. 4.2 shows the band structure of a typical semiconducting TMD (MoS₂). For the bulk material the band structure shows an indirect band gap. However, when the material is reduced to a monolayer a transition to a direct band gap is observed. This greatly increases the optical activity, making monolayer TMDs interesting for optoelectronic applications. In comparison to many III-V semiconductors with the band gap in the center of the Brillouin zone, the direct band gap in TMDs is located at the $K$ point of the hexagonal Brillouin zone. Additional band valleys are located at the $\Gamma$ point in the valence band and the $\Sigma$ point in the conduction band half way between $K$ and $\Gamma$.

Semiconducting TMD monolayers or, depending on the context, 2D materials in general feature a range of unique properties that lend themselves to various studies and possible applications. Due to the reduced dimensionality of the monolayers, charge carriers that interact with each other “feel” the dielectric environment surrounding the monolayer, i.e. field lines of the Coulomb interaction pass through the surrounding medium and this leads to different screening. Thus, the band structure of TMDs is highly susceptible to changes of the dielectric environment as well as strain [99, 100], which allows to engineering their electronic properties. The reduced Coulomb screening of electrons gives rise to very large exciton binding energies.
Figure 4.2: Band structure of bulk and monolayer MoS$_2$. The material’s band structure is strongly dependent on the dimensionality of the material. While the bulk material has an indirect band gap, a direct band gap emerges at the $K$ point of the Brillouin zone [111] as the material is reduced to a monolayer.

and results in strong photoluminescence due to large oscillator strengths. Furthermore, the symmetry of the monolayer leads to two different types of $K$ valleys in the hexagonal Brillouin zone. They are distinguished by their different coupling to circular polarized light. This allows to specifically exploit this valley-degree of freedom giving rise to the field of “valleytronics” [101, 102]. Additionally, due to their atomic thickness, monolayers can be stacked on top of each other to form so called van-der-Waals heterostructures [89, 103, 104, 105]. As the layers influence each other, interesting electronic phenomena can be studied like interlayer excitons [106] or strongly improved photoluminescence linewidth [107]. More recently also the topic of twisted bilayers, related to heterostructures, emerged. In this context the magic angle in twisted bilayer graphene was observed, that showed signs of strongly correlated behavior and superconductivity [108, 109]. The same concept of twisted bilayers can also be applied to TMD materials, where the emerging Moiré patterns between the layers create modulations of the electronic landscape. These span over a large number of unit cells, leading to periodic confinement potentials for intra- and interlayer excitons [110].

In this chapter we focus on single layer TMDs with their strong optical activity and photoluminescence in the visible range. In the context of nanolaser application of TMDs, the influence of excited carriers on their electronic properties must be considered as lasers operate far out of equilibrium. The band structures shown on Fig. 4.2 includes many-particle effects, but represents the electronic structure of the system in the ground state that would be experienced by a single excited electron-hole pair. If large carrier densities are excited in the material, additional many-body effects emerge. In TMD monolayers, with their strong Coulomb interaction, these many-body effects lead to a screening of the interaction that cause band structure renormalizations and additional dephasing due to carrier scattering. They result in significant changes of the optoelectronic properties [112]. Fig. 4.3 illustrates the important renormalizations of the band structure due to excited carriers that will influence the TMDs performance as laser gain medium. In the ground state (dashed, gray lines) the materials that we consider have a direct
band gap at the \( K \) point of the Brillouin zone\(^1\). If the material is excited the band gap of the material shrinks (red lines in Fig. 4.3) leading to a red shift in the materials photoluminescence [112]. At the same time the conduction band \( \Sigma \) valley shifts to lower energies relative to the \( K \) valley. Carriers that are in quasi-equilibrium in the conduction band distribute themselves among the \( K \) and \( \Sigma \) valleys. However, with their increasing energy difference at higher densities, carriers are drained from \( K \) into \( \Sigma \), reducing population inversion at the \( K \) point which is detrimental to the optical gain of the excited material. Additionally, scattering among excited carriers introduces additional dephasing in the material that effectively reduces the light-matter interaction and hinders efficient lasing operation. An extensive analysis of these many-body effects can be found in [114].

Figure 4.3: Illustration of the effect of band-structure renormalizations induced by excited carriers. The direct band gap \( E_G^0 \) at the \( K \) point shrinks to the value \( E_G \) at a certain carrier density. At the same time, the conduction-band valley at the \( \Sigma \) point shifts even further, becoming energetically more favorable for electrons than the \( K \) point at a critical carrier density. As a consequence, electrons are drained from the \( K \) to the \( \Sigma \) valley, thereby being lost to the optically active region around \( K \).

Application in micro- and nanolasers

Initial reports [20, 21, 22, 23] make use of the strong optical activity of semiconducting TMD monolayers by combining them with dielectric micro- and nanocavities to assess their capabilities for high-efficiency lasing applications. To that end, monolayers were positioned on top of photonic crystal or microdisc cavities and into the evanescent field of the localized cavity modes as depicted in Fig. 4.4. This procedure has advantages over more established epitaxially grown semiconductor nanostructures, such as self-organized quantum dots or quantum wells; mono-

\(^1\)The intrinsic direct band gap of TMD monolayers (especially in selenides) is disputed in the literature [113], while the minimum of the \( \Sigma \) valley is separated from the \( K \) valley by only few tens of meV. However, the \( K \) and \( \Sigma \) valleys of the conduction band are energetically close in these materials and the transition from direct to indirect band gap is a gradual one. Thus, this distinction is less relevant in the context considered here.
4.1. ATOMICALLY THIN SEMICONDUCTORS

Layers can be positioned deterministically into the field of a photonic-crystal or microdisc cavity, which offers control over the light-matter interaction without the detrimental effect on the $Q$ factor when integrating the medium inside the cavity. Interestingly, an atomically thin layer of material that interacts with the relatively weak evanescent field of a cavity mode, provides sufficient optical gain to sustain lasing. In this context improvement of the optical confinement factor $\Gamma$ is critical to increase the quantum efficiency of the laser. It is defined by

$$\Gamma = \frac{\int_{\text{Layer}} \epsilon(r) |E_{||}(r)|^2 \, dr}{\int_{V} \epsilon(r) |E(r)|^2 \, dr}$$

and quantifies the spatial overlap of the mode function with the 2D gain medium. $E_{||}(r)$ is the in-plane electric field component in the monolayer [115, 116] while the denominator is proportional to the cavity mode volume. The confinement factor is a measure for how strongly the gain medium can couple photons into the cavity mode and thus it determines the modal gain. Optimizing $\Gamma$ is a promising avenue for increasing device efficiency. For example in [23], a WS$_2$ monolayer was placed on top of a microdisc cavity and was capped with a layer of silica material, as shown in Fig. 4.4 a). By its lower refractive index the capping increases the extend of the evanescent field outside the cavity and thus the field strength at the monolayer position, which effectively increases $\Gamma$. Another proposal to increase the modal gain is to exploit the potential of van-der-Waals heterostructures [103, 104]. Alternating layers of gain medium and isolating 2D materials (e.g. hBN) could potentially multiply the confinement factor facilitated by the sub-nanometer length scale of the layer thickness, on which the evanescence field strength only decays slowly.

Figure 4.4: Examples of TMD nanolasers with intensity patterns of the lasing mode. a) Nitride based microdisc cavity with monolayer WS$_2$ as gain medium positioned on top [23]. The whispering-galley mode of the cavity (top view, bottom panel) interacts with the monolayer via its evanescent field, where the mode overlap is increased by the silica (HSQ, green) capping layer. b) Monolayer WSe$_2$ is positioned on top of a photonic crystal defect cavity [22]. The calculated cavity mode intensity profile is shown. The side view (bottom panel) indicates the extend of the evanescent field relative to the monolayers position.
4.2 Microscopic gain calculation

To create population inversion and achieve optical gain, TMD monolayers must be strongly excited. As discussed above, the excited carriers lead to pronounced band structure renormalizations. In the following we examine how these influence the gain properties of the material. Optical gain of a material is reached when the material’s absorption becomes negative due to external excitation of carriers, i.e. more photons leave the material than enter it. Absorption/gain is defined via the response to a weak optical test field $E(\omega)$. In the regime of linear optics this response is given by the optical susceptibility

$$
\chi(\omega) = \frac{P(\omega)}{\epsilon_0 E(\omega)},
$$

where $P(\omega)$ is the macroscopic polarization inside the optically active layer. The absorption is then proportional to the imaginary part $\text{Im}[\chi(\omega)]$ of the susceptibility, while its real part is related to a change in the refractive index. The macroscopic polarization $P(\omega)$ is given by the sum over microscopic ones $\psi^{\text{eh}}_k(\omega)$ and dipole moments $d^{\text{eh}}_k$ according to

$$
P(\omega) = \frac{1}{S} \sum_{k'} \psi^{\text{eh}}_k(\omega) (d^{\text{eh}}_k)^* + \text{h.c.}
$$

We determine the microscopic polarizations by solving the semiconductor Bloch equations (SBE) on the full Brillouin zone. Taking into account many-body effects on $GW$ level the SBE are given by

$$
\left(\hbar \omega - \epsilon^{\text{e}}_k - \epsilon^{\text{h}}_k - \Delta^{\text{eh}}_k(\omega) + i \gamma^{\text{el-ph}} \right) \psi^{\text{eh}}_k(\omega)
+ (1 - f_{k'}^e - f_{k'}^h) \left( d^{\text{eh}}_k \cdot E(\omega) + \frac{1}{S} \sum_{k'} V^{\text{ehhe}}_{kk'kk'} \psi^{\text{eh}}_k(\omega) \right)
+ \frac{1}{S} \sum_{k'} V^{\text{effeh}}_{kk'}(\omega) \psi^{\text{eh}}_k(\omega) = 0.
$$

On $GW$ level, the SBE describe two-particle states taking into account screened carrier-carrier interactions. In (4.4), $\epsilon^{\text{e}}_k$ and $\epsilon^{\text{h}}_k$ are the single particle energies with renormalizations on Hartree-Fock level [117]. Electron and hole population functions $f_{k}^{\text{e}}$ and $f_{k}^{\text{h}}$ enter the Pauli-blocking term in the second line of (4.4). It also contains the light-matter interaction of the materials dipoles with the external field $d \cdot E$ as well as Coulomb interaction terms with Coulomb matrix elements $V^{\text{ehhe}}_{kk'kk'}$, where $S$ is the area of the TMD layer. Dephasing due to carrier-phonon interaction is included by a phenomenological rate $\gamma^{\text{el-ph}}$. Many-body effects beyond the Hartree-Fock level are included by correlation terms $V^{\text{effeh}}_{kk'}(\omega)$ and energy renormalization $\Delta^{\text{eh}}_k(\omega)$. The material-realistic gain calculations where performed primarily by Dr. Alexander Steinhoff in the context of our cooperation work [88].

The results for the material gain are shown in Fig. 4.5 for carrier densities above the Mott transition [118]. All investigated materials exhibit gain at a certain excitation level. At the same time, all materials show a reduction of gain at high excitations that limits the maximum achievable gain. This “rollover” can be traced back to the band structure renormalizations, caused by many-body effects, discussed in the previous section. The $\Sigma$ valley drains carriers from the $K$ valley, effectively reducing the inversion at the optical transition around $K$ and eventually leading to a gradual direct-to-indirect band gap transition of the material. Together with additional carrier-induced dephasing that broadens the optical resonances, these band
4.2. MICROSCOPIC GAIN CALCULATION

Figure 4.5: Imaginary part of dimensionless optical susceptibility as obtained from the SBE for (a) monolayer MoS$_2$, (b) MoSe$_2$, (c) WS$_2$, and (d) WSe$_2$ on a SiO$_2$ substrate at $T = 300$ K and increasing excitation density $N$ given in $10^{13}/\text{cm}^2$. While a positive value of $\text{Im} \chi$ implies absorption, optical gain is characterized by a negative imaginary part of the susceptibility (shaded region). The energy axis is chosen relative to the energy $E_0^X$ of the A exciton at zero excitation density and low temperatures [107]. Vertical lines (dotted) indicate the energies at which maximum gain is achieved for each material.

structure renormalizations limit the achievable gain in the material and lead to the observed rollover.

We note that there is ongoing debate on the origin of the gain mechanism for TMD-monolayer based nanolasers. Gain from exciton population is claimed in [22, 23] while in [20] a gain mechanism involving trion populations is proposed. Here we discuss gain properties of highly excited TMD monolayers. Considering excitation levels beyond the Mott transition, all excitons are dissociated into an electron-hole plasma due to screening of the Coulomb interaction [118]. Nevertheless, the plasma gain still experiences renormalizations by the strong many-body effects discussed above. Although our consideration exclude gain mechanism related to populations of excitons and trions, interesting effects might occur at lower carrier densities, which merits further investigation of that regime in the future.
4.3 A TMD-nanolaser model

Experimental measurements of gain spectra can be performed for more conventional 2D semiconductors [119, 120, 121]. In comparison, for monolayer TMDs population inversion at elevated excitation levels have been measured [122], however, the direct measurements of their gain properties is more challenging in general. Therefore, we are interested in experimentally accessible signatures of the material-specific gain behavior, in order to relate the gain calculations discussed in the previous section to realistic devices. To that end, we introduce a rate equation theory that is suited to describe semiconductor nanolasers in which the localized mode of a cavity interacts with the extended 2D gain material. Furthermore, we provide gain parametrizations that can be used as input to the introduced rate equations. Our discussion presents a semiclassical model capturing the material-specific effects in comparison to the previous chapter where the source of optical gain is left unspecified. The model serves the purpose of bridging the gap between microscopic, many-body gain calculations and a laser theory that is readily accessible for the analysis of experimental TMD nanolasers on a material-realistic basis.

Modal gain

The microscopic gain calculations presented in 4.2 only specify the material susceptibility that is related to the material gain. For a particular device we are interested in the modal gain that accounts for the overlap of the gain medium with the cavity mode or, more specifically, for the spatially varying interaction strength of the localized cavity mode with the gain medium. To derive an expression for modal gain, standard rate equations often consider the concept of a propagating light field in an extended medium. Assuming a slowly varying envelope of the light field, this treatment gives rise to a gain expression with units of 1/m. It reflects the concept of light amplification per unit length of light traveling in the gain medium. Here, instead, we consider strongly localized modes (spatially and spectrally) as indicated in Fig. 4.4. In Appendix C.1 we give a derivation of the modal gain, along the lines of [79, 115], for a spatially stationary mode interacting with the 2D layer. The result includes the optical confinement factor Γ defined in (4.1). The modal gain is written as

\[ \Gamma G(N), \quad \text{with} \quad G(N) = -\frac{\omega_0}{d} \Im [\chi(\hbar \omega_0, N)], \]

(4.5)

where \( G(N) \) is the material gain in units of the 1/s. The difference in units of the modal gain results from the a different treatment of Maxwell’s equations leading to the expression. To obtain a modal gain in units of 1/m, one typically assumes the slowly varying amplitude approximation (SVEA) to hold, where the amplitude variation is meant to occur spatially [83]. For our derivation the mode function has a fixed spatial dependence but its amplitude is assumed to vary slowly in time.

We want to use the calculated material gain from the previous section as input to a rate equation theory. We start by comparing the gain values for MoS\(_2\), WS\(_2\) and WSe\(_2\) as a function of carrier density. MoSe\(_2\) is neglected as it provides only minimal gain. We collect results for the susceptibility of Fig. 4.5 at fixed energies (vertical lines) that maximize the gain for each material. We plot the resulting modal gain in Fig. 4.6, where the confinement factor \( \Gamma = 9 \times 10^{-4} \) is chosen to lie in the range of values reported in [20, 23] for photonic crystal and microdisc cavities. The gain curves clearly show the distinct rollover behavior that imposes an intrinsic upper limit on the gain that is achievable from a single monolayer. Solid lines show a fit to the calculated data (dots) with a fit formula that is given in Appendix C.3. In rate equations theories, linear or logarithmic gain models are often employed and have also been applied to
4.3. A TMD-NANOLASER MODEL

Figure 4.6: Modal gain $\Gamma G(N)$ as a function of carrier density for MoS$_2$, WS$_2$ and WSe$_2$ at energies where maximum gain is achieved. All materials show a characteristic rollover that limits gain from a single layer. In WS$_2$, gain sets in at lowest carrier densities and takes on the largest value among all four TMD materials. While MoS$_2$ requires much larger excitation densities to achieve gain, it still exhibits a larger peak gain than WSe$_2$. Solid lines indicate the parametrization according to Eq. (C.32). Dashed lines show logarithmic fits to the gain curves at low densities. For the used parameters, the minimal $Q$-factor necessary to achieve lasing is given for each material.

TMD-based nanolasers. Dashed lines in Fig. 4.6 show a logarithmic fit to the material gain that works well at low densities but fails to reproduce the rollover behavior in the absence of any mechanism that would limit the gain.

Rate equations

Using the modal gain derived above we introduce a suitable set of rate equations in order to investigate experimentally observable characteristics. Similar to standard rate equations [39] they describe the interaction of the carrier density in the active medium $N$ and the photon density $N_p$ in the cavity. They are given by (see Appendix C.2 for more details)

\[
\dot{N} = \frac{P}{A_s} - \frac{V_m}{A_s} \Gamma G(N) N_p - \frac{V_m}{A_s} (AN + BN^2 + CN^3), \tag{4.6a}
\]

\[
\dot{N}_p = \Gamma G(N) N_p + \beta BN^2 - \frac{N_p}{\tau_c}, \tag{4.6b}
\]

where $P$ is the rate of carrier generation in the active area $A_s$. The spontaneous emission coupling factor is $\beta$, $\Gamma$ is the optical confinement factor from Eq. (4.1), $V_m$ is the cavity mode volume and $1/\tau_c = \omega_0/Q$ is the cavity decay rate with the cavity resonance frequency $\omega_0$ and quality factor $Q$. The coefficients $A$, $B$ and $C$ are the rates of radiative and non-radiative losses known from standard rate equations. These rate equations are different from standard rate equations in the way they deal with the dimensions of the systems. They describe the density of excited carriers in the 2D active medium per area, while $N_p$ relates to the cavity
volume. This mismatch in dimensions is reconciled by the fraction \( V_m/A_s \). We note that in the standard treatment of rate equations a similar dimensionality factor \( V_m/V \) is encountered, relating the mode volume and the volume of the gain medium \( V \). Often it is also referred to as confinement factor. In conventional lasers, the gain material is typically completely enclosed inside the cavity and the \( V_m/V \) fraction approximately coincides with the optical confinement factor. As our case is different with respect to the device geometry, we leave this dimensionality factor explicit in the equations. In principle it has physical meaning, but its effect is merely a rescaling of the power axis (\( P \propto V_m \) for stationary solutions), while the relative contributions of gain and loss in the equations remain unchanged.

Looking at the rate equations (4.6), the significance of \( \Gamma \) becomes more clear as we can construct device specific criteria for the possibility to sustain lasing. Considering stationary solutions to (3.9b) and neglecting contributions by spontaneous emission, the threshold condition of gain to compensate losses is given by

\[
\Gamma G(N) = \tau_c^{-1}. \tag{4.7}
\]

Looking at the modal gain (4.5) we easily see that \( \Gamma Q \) needs to be exceed a certain material-specific value in order to satisfy (4.7). As all investigated TMD materials show the characteristic gain rollover, this establishes a device specific lower bound for the product \( \Gamma Q \) and we find 2.18 for WS\(_2\), 4.59 for MoS\(_2\), and 5.88 for WSe\(_2\). For the confinement factor chosen here, Fig. 4.6 shows minimal values of \( Q \) for each material to sustain lasing.

| \( \Gamma \)   | \( 9 \times 10^{-4} \) |
| \( \beta \)   | 0.1 |
| \( A \)       | 0 |
| \( B \)       | \( 6 \times 10^{-6} \) nm ps\(^{-1} \) |
| \( C \)       | 0 |
| \( V_m \)     | \( 1.4 \times 10^8 \) nm\(^3 \) |
| \( A_s \)     | \( 5.6 \times 10^6 \) nm\(^2 \) |

Table 4.1: Parameters used for the results in Fig. 4.7. Note that while the quantities \( V_m \) and \( A_s \) possess physical meaning [23], they merely define a scale for the excitation-rate axis while leaving the input-output characteristics unchanged.

We evaluate stationary solutions for the rate equations (4.6) as a function of the input pump power \( P \). Parameters are chosen close to those in [23] which we summarize in Table 4.1 for better overview. Here, we refrain from adding non-radiative losses in the calculation \( (A = C = 0) \) as they act on top of the material-intrinsic gain rollover. In realistic scenarios however, they might contribute significantly to the dynamics, e.g. by recombination at defects. Therefore, when applying the rate equation theory to realistic devices, (non-) radiative recombination coefficients \( (A, B, C) \) as well as the confinement factor \( \Gamma \) are ideally determined from individual measurements or calculations. The results of the evaluation are shown in Fig. 4.7 for MoS\(_2\), WS\(_2\) and WSe\(_2\) with increasing quality factors of the optical cavity.

Upper panels in Fig. 4.7 show the output intensity of the laser. In all cases an (apparent) threshold non-linearities in the I/O characteristics, similar to those in [20, 22, 23], can be observed. Here, however, only clamping of the carrier density (middle panels) indicate above-threshold operation. At insufficient values of \( \Gamma Q \), the apparent threshold non-linearities can instead be attributed to the onset of amplified spontaneous emission [52] in the high-\( \beta \) regime. This is the case for all three materials at the lowest \( Q \) factor (left panels). This is also reflected in the ratio of modal gain and cavity losses (lower panels), showing the TMD-specific gain
Figure 4.7: Input-output curves (top), carrier densities (middle) and the ratio of modal gain and cavity losses (bottom) for increasing $Q$-factors of 1500, 3000 and 6000. Results are obtained from Eqs. (4.6) using input from the material-realistic gain calculation as described in Section 4.3 for MoS$_2$, WS$_2$ and WSe$_2$. Dashed lines use the logarithmic gain fit indicated in Fig. 4.6. In WS$_2$ the lasing threshold is reached at lowest excitation rate and the lowest $Q$-factor is required as the maximum gain is larger in comparison to MoS$_2$ and WSe$_2$. If the product $\Gamma Q$ falls below a material-specific value, all materials experience a gain rollover (left panels) and lasing cannot be achieved by stronger pumping. With a logarithmic gain model the threshold can be reached at sufficient pumping and the material-specific lasing signatures are no longer reflected in the calculations. Parameters are chosen close to those in [23].
densities (together with WSe$_2$) due to lower effective hole masses in those materials, causing a more localized carrier population that helps the formation of inversion [88].

4.4 Outlook: Correlation measurements on TMD nanolasers

When relying on rate equation theories that are based on conventional linear or logarithmic gain models, we saw that the results can be misleading when used to analyze the I/O characteristics of high-$\beta$ TMD nanolasers. Also the relatively small linewidth narrowing observed in first reports on TMD nanolasers can be attributed to the onset of amplified spontaneous emission and is not necessarily a clear indicator that the threshold has been crossed. From the rate equations, clamping of the gain or the carrier density in a nanolaser would prove lasing operation in a device, but it is challenging if not impossible to measure experimentally. Furthermore, TMD monolayers represent a class of extended 2D gain media as discussed in the previous chapter and are thus expected to show a delayed onset of coherent emission with respect to the intensity jump even if the intensity threshold has been crossed. These findings emphasize the need to investigate photon-correlation functions in order to unambiguously prove lasing from a given device. Only then, the change in photon statistics from a thermal to a Poisson distribution is made visible, as it is imprinted in the change of the second-order correlation function $g^{(2)}(0)$. In the following chapters we will expand our discussion on coherence properties of semiconductor nanolasers. To that end we develop a quantum-optical lasers model that gives access to photon correlations as well as a broad range of device characteristics. While the model in the previous chapter was more idealized, in the following we will also include a microscopic modeling of the semiconductor gain medium, allowing to compare the theoretical analysis to experimental measurements of QW-based semiconductor nanolasers. This lays the ground also for cooperation of theory and experiment in future realizations of TMD nanolasers where, by measurements of photon correlations, the capabilities of TMD as gain media will be further investigated.
Chapter 5

Quantum-optical models for quantum well nanolasers

The previous chapters highlighted challenges that arise when assessing the emission characteristics of semiconductor nanolasers in general and nanolasers with extended 2D gain media in particular. Especially in the regime of high $\beta$ factors, identifying lasing becomes difficult when one relies solely on I/O characteristics and linewidth narrowing. As losses in the laser become small, the disappearance of a clear-cut laser threshold and the transition to a gradual onset of coherent emission necessitate a confirmation of a Poissonian emission statistics. Furthermore, we saw in the context of TMD-monolayer gain materials that the material specific gain behavior of the monolayers provides additional difficulties due to the predicted rollover (see Section 4.2).

Experimentally, it has become an established method to distinguish between coherent and thermal emission characteristics by measurement of the two-photon correlation function $g^{(2)}$ [14, 15, 48]. However, often the direct observation of photon bunching at the two arms of a HBT setup is limited by the finite time resolution of the single photon detectors and the short coherence time that is inherent to incoherent light. Therefore, the theoretical understanding of the photon statistical properties becomes important in order to understand experimental results and to correctly identify lasing operation. In Chapter 3 we shed light on the emission properties of high-$\beta$ nanolasers in terms of their photon correlations and found a delay for the onset of coherent emission that is specific to nanolasers with extended (2D) gain media. In particular, the quantum threshold, i.e. an average photon number of 1 in the cavity, is not a viable criterion for coherent emission in those systems.

In order to connect our theoretical discussion on nanolasers to experimental studies, in the following we will present quantum optical studies of (near) thresholdless nanolasers with semiconductor quantum wells as gain medium. As we will see, signatures of the delayed onset of coherent emission can be observed in

Figure 5.1: The examples of high-$\beta$ nanolaser with QW gain medium, to which we apply the model derived in this chapter. a) Coaxial nanolaser with metallic cladding as cavity with integrated multi QWs. b) Photonic crystal nanobeam laser with an embedded single nitrite based QW.
the reported systems. Schematic views of the examined devices are shown in Fig. 5.1. In the following sections we will develop a semiconductor laser theory that combines a microscopic description of the laser gain medium with the quantum-optical treatment of the photon correlations introduced previously. Starting in Section 5.1 we introduce the Hamiltonian and basic concepts of the laser model and derive equations of motion on the doublet level. Section 5.2 will show how our model relates to standard rate equations (RE) that are often employed to model the emission characteristics of conventional as well as micro- and nanolasers. Following that, beginning from Section 5.3, we will discuss how first- and second-order correlation functions can be included in the model as well as the calculation of time-resolved photoluminescence. Our theoretical model gives access to a number of emission characteristics, while taking as input device specific parameters that are determined from experiment. This will be applied in Chapter 6 where we combine our theoretical analysis with experimental results. The discussion of our model is based on our previous publications [15] and [50] (under review at the time of writing).

5.1 A quantum optical model

In Chapter 3 we started with the system Hamiltonian to derive equations of motion for the system dynamics. We derive our semiconductor laser model here along the same lines, using the cluster expansion approach to calculate photon correlations functions. However, we expand the description of the system to include aspects of the extended nature of the semiconductor gain medium. Most importantly, we assume a parabolic and quasi-continuous band structure for the semiconductor where electron-hole pairs of different energies can interact resonantly or non-resonantly with the modes of the cavity as indicated in Fig. 5.2. On a phenomenological level, we also include effects of carrier-carrier and carrier-phonon interaction in the material, giving rise to a more comprehensive picture of the processes in the system. The treatment of these effects on a phenomenological level makes it possible that we can separate the time scales of the system dynamics. Microscopic carrier-carrier interactions generally happen on time scales orders of magnitude faster (e.g. 1 ps) than those of the laser dynamics, while being computationally much more demanding. Modeling these effects on the level presented here allows us to calculate the dynamics on much longer time scales, which is necessary to obtain a model that make it feasible to model the dynamics of devices.

Hamiltonian

We start with the system Hamiltonian and derive equations for the time evolution of operator expectation values according to the Heisenberg equation of motion

$$\frac{d}{dt} \langle A(t) \rangle = -i \langle [A(t), H] \rangle,$$  \hspace{1cm} (5.1)

for any operator $A(t)$ where we use $\hbar = 1$ everywhere in the following. The Hamiltonian is given by

$$H = H_{\text{carr}} + H_{\text{ph}} + H_I.$$  \hspace{1cm} (5.2)

Explicitly,

$$H_{\text{carr}} = \sum_k \epsilon_k c_k^\dagger c_k + \epsilon_k^h v_k^\dagger v_k$$  \hspace{1cm} (5.3)

is the Hamiltonian of the excited carriers in the material, $c_k, c_k^\dagger$ are creation and annihilation operators for electrons in the conduction band, and $v_k, v_k^\dagger$ for the valence band, respectively.
5.1. A QUANTUM OPTICAL MODEL

Figure 5.2: Band structure model with two parabolic bands used for modeling the semiconductor gain medium. The red arrows indicate the elementary processes representing the interaction with the light field. These are the absorption of a cavity photon under the creation of an electron-hole pair in the bands or, in reverse, the recombination of electrons and holes under emission of a photon. When interacting with a single cavity mode, only electron-hole pairs with energy \( \epsilon_k^e - \epsilon_k^h = \hbar \omega \) are resonant with the cavity.

for different in-plane carrier momenta \( k \). Also given in Fig. 5.2, according to the effective mass approximation, the energies for electrons and holes \( \epsilon_k^e, \epsilon_k^h \) in the band structure are given by

\[
\epsilon_k^e = \epsilon_g + \frac{\hbar^2 k^2}{2m_e}, \quad \text{and} \quad \epsilon_k^h = -\frac{\hbar^2 k^2}{2m_h},
\]

with \( \epsilon_g \) being the electronic band gap. Similarly, the photon Hamiltonian is

\[
H_{\text{ph}} = \sum_{\xi} \omega_{\xi} \left( b_{\xi} b_{\xi}^\dagger + \frac{1}{2}\right),
\]

with creation and annihilation operators \( b_{\xi}^\dagger, b_{\xi} \) for photons in different modes \( \xi \) and mode energy \( \hbar \omega_{\xi} \). The light-matter interaction Hamiltonian is given by

\[
H_1 = i \sum_{k, \xi} \left( g_{\xi,k} b_{\xi} b_{\xi}^\dagger c_k - g_{\xi,k}^* b_{\xi}^\dagger c_k^\dagger \right).
\]

It represents a Jaynes-Cummings-type interaction between electron-hole pairs and the photon modes where the localized lasing mode of the cavity \( \xi_l \) plays the dominant role. The form of the light-matter interaction strength \( g_{\xi,k} \) is discussed in detail in Appendix D.

Equations of motions

The dynamics of individual operators is governed by Eq. (5.1). We find for the creation operators (and analogously the hermitian conjugate for the annihilation operators)

\[
\frac{d}{dt} v_k^\dagger = i \epsilon_k^h v_k^\dagger + \sum_{\xi} g_{\xi,k} \xi^h b_{\xi},
\]
\[ \frac{d}{dt} c_k^\dagger = i\epsilon_k c_k^\dagger - \frac{1}{\hbar} \sum_\xi g_\xi v_\xi^k b_\xi^k, \] (5.8)

\[ \frac{d}{dt} b_\xi^k = i(\omega_\xi - i\kappa_\xi) b_\xi^k + g_\xi^2 \sum_k c_k^\dagger v_k, \] (5.9)

where for \( b_\xi^k \) we introduced a complex cavity resonance frequency with imaginary part \(-i\kappa_\xi\) to account for the finite lifetime of photon in each particular mode.

Using Eq. (5.1) we derive the dynamics of the carrier populations for conduction and valence band electrons. For the conduction band we write \( f_k^c = \langle c_k^\dagger c_k \rangle \) and for the valence band \( f_k^v = 1 - \langle v_k^\dagger v_k \rangle \), which indicates that holes represent the absence of an electron in a particular state and that all populations are bound by 1 due to the fermionic character of the electrons. The corresponding EoMs are given by

\[ \frac{d}{dt} f_k^c = -2 \sum_\xi \text{Re} \left[ g_{\xi,k}^* \langle b_\xi^\dagger v_\xi^k c_k \rangle \right], \] (5.10)

\[ \frac{d}{dt} f_k^v = -2 \sum_\xi \text{Re} \left[ g_{\xi,k}^* \langle b_\xi^\dagger v_\xi^k c_k \rangle \right]. \] (5.11)

The dynamics are coupled to the photon-assisted polarization \( \langle b_\xi^\dagger v_\xi^k c_k \rangle \) that represents the process of an electron-hole recombination under emission of a photon into mode \( \xi \). Similarly, for the average photon number in mode \( \xi \) we find

\[ \left( \frac{d}{dt} + 2\kappa_\xi \right) \langle b_\xi^\dagger b_\xi \rangle = 2 \sum_{k'} \text{Re} \left[ g_{\xi,k}^* \langle b_\xi^\dagger v_\xi^k c_{k'} \rangle \right]. \] (5.12)

The photon-assisted polarization has its own dynamics given through the Hamiltonian in (5.2). However it is coupled to three-particle quantities \( \langle b_\xi^\dagger b_\xi^\dagger c_k^c \rangle, \langle b_\xi^\dagger b_\xi^\dagger v_\xi^k v_k \rangle \) and electron-electron correlations \( \langle c_k^c v_k v_k^c \rangle \). Coupling of the equations to ever higher-order terms is a manifestation of the hierarchy problem discussed in Appendix A. We factorize the three-particle expectation values according to the cluster expansion. This results in the EoM

\[ \left( \frac{d}{dt} + \kappa_\xi + \Gamma \right) \langle b_\xi^\dagger v_\xi^k c_k \rangle = -i(\epsilon_\xi - \epsilon_k^c - \hbar\omega_\xi)\langle b_\xi^\dagger v_\xi^k c_k \rangle \]
\[ + f_k^c f_k^v + \langle b_\xi^\dagger b_\xi \rangle (f_k^c + f_k^v - 1) \]
\[ + \delta(\langle b_\xi^\dagger b_\xi^\dagger c_k^c \rangle - \langle b_\xi^\dagger b_\xi^\dagger v_\xi^k v_k \rangle). \] (5.13)

The resulting terms can readily be interpreted. The first line in Eq. (5.13) is the evolution due to the non-interacting Hamiltonian while the second line corresponds to contributions of spontaneous and stimulated emission. It is important to note that the spontaneous-emission term results from the quantum description of the light field. In a semiclassical theory that considers a classical light field this term does not arise from the Hamiltonian and spontaneous emission must be included on a phenomenological basis. This will become even more clear when connecting Eqs. (5.10)–(5.13) to REs in Section 5.2. The last two terms in (5.13) are pure electron-photon correlations beyond the doublet level. On this level we neglect these terms, which corresponds to a truncation of the EoM hierarchy on the doublet level (c.f. Appendix A). They are, however, vital for accessing \( g^{(2)}(0) \) in Section 5.5.
5.1. A QUANTUM OPTICAL MODEL

Figure 5.3: Optical or electrical pumping of carriers in the barrier material is modeled as incoherent excitation of carriers with a Gaussian energy distributions centered high above the band gap (red). Subsequently, carriers are scattered to lower energy states with rate $\gamma_{\text{rel}}$ due to carrier-carrier and carrier-phonon interaction. They relax towards Fermi-Dirac quasi-equilibrium distributions $f_{k}^{\text{FD}}$ (indicated in blue), separately for conduction and valence band.

**Carrier relaxation and pumping**

Carrier relaxation due to electron-phonon as well as Coulomb scattering is treated on a phenomenological level and in relaxation-time approximation. Collision effects drive the carrier populations towards a quasi-equilibrium for electrons and holes on a material specific time scale [83, 124, 125]. We add the relaxation terms to the dynamics of the population functions given and by

$$\frac{d}{dt} f_{k}^{\nu} \bigg|_{\text{rel}} = -\gamma_{\text{rel}} (f_{k}^{\nu} - f_{k}^{\text{FD}}) ,$$

(5.14)

where $\nu = \{e, h\}$ with the relaxation rate $\gamma_{\text{rel}}$ and Fermi-Dirac quasi-equilibrium functions $f_{k}^{\text{FD}}$. Furthermore, we account for scattering induced dephasing of the polarization by adding the phenomenological dephasing rate $\Gamma$ in the polarization equation (5.13).

Pumping of the gain medium is modeled as incoherent carrier generation high above the band gap with subsequent carrier equilibration and relaxation to lower energy states. We assume a carrier generation with a Gaussian distribution $f_{k}^{P}$ at a rate $P$. The generation is limited by Pauli blocking, which results in the pumping term

$$\frac{d}{dt} f_{k}^{\nu} \bigg|_{P} = P f_{k}^{P} (1 - f_{k}^{\nu}) .$$

(5.15)

**Adiabatic elimination of non-lasing modes**

A main feature of our theory is that it describes the dynamics of a single lasing mode that interacts with the gain medium. However, in general there will be radiative losses by interaction with other modes in which photons are emitted as well. The sums in (5.10) and (5.11) account for these unwanted emission channels by including other cavity modes as well as free space modes into which electron hole pairs can emit photons when recombining. We describe the
combined effect of these non-lasing (nl) modes by adiabatically eliminating their photon-assisted polarization equations (5.13). We assume that photon numbers are negligible for all non-lasing modes, i.e. $\langle b^\dagger_k b_k \rangle = 0$ for any $\xi$ that is not the laser mode, and we also neglect the corresponding correlation functions. After inserting (5.10) and (5.11) we find with $\nu = \{e, h\}$

$$\frac{d}{dt} f^\nu_k |_{nl} = -\sum_{\xi \neq \xi_l} \frac{2|g_{k,\xi}|^2(\Gamma + \kappa)}{(\Gamma + \kappa)^2 + (\epsilon^c_k - \epsilon^h_k - \hbar \omega^\xi)^2} f^e_k f^h_k = -\gamma_{nl} f^e_k f^h_k,$$

(5.16)

which defines the total loss rate $\gamma_{nl}$ into all other modes. It is clear from (5.16) that the rate $\gamma_{nl}$ will in principle be $k$-dependent. However, as we deal with systems where fast relaxation according to (5.14) redistributes the carriers quickly, we use an effective, $k$-independent rate of radiative losses. As we combined the influence of all non-lasing modes into a collective emission rate, from now on we drop the label $\xi$ as we are left with a single lasing mode only in our description.

We summarize the resulting EoM adding the terms that we introduced to the dynamics of the carrier populations, Eqs. (5.10) and (5.11):

$$\frac{d}{dt} f^e_k = -2|g_k|^2 \text{Re} \left[ \langle b^\dagger_k v^e_k c_k \rangle \right] - \gamma_{nl} f^e_k f^h_k - \gamma_{rel} (f^e_k - f^{FD}_k) + Pf^e_k (1 - f^e_k),$$

(5.17)

$$\frac{d}{dt} f^h_k = -2|g_k|^2 \text{Re} \left[ \langle b^\dagger_k v^e_k c_k \rangle \right] - \gamma_{nl} f^e_k f^h_k - \gamma_{rel} (f^h_k - f^{FD}_k) + Pf^h_k (1 - f^h_k),$$

(5.18)

$$\left( \frac{d}{dt} + 2\kappa \right) \langle b^\dagger b \rangle = 2 \sum_k |g_k|^2 \text{Re} \left[ \langle b^\dagger_k v^e_k c_k \rangle \right],$$

(5.19)

$$\left( \frac{d}{dt} + \kappa + \Gamma \right) \langle b^\dagger_k v^e_k c_k \rangle = -i(\epsilon^c_k - \epsilon^h_k - \omega)\langle b^\dagger_k v^e_k c_k \rangle$$

$$+ f^e_k f^h_k + \langle b^\dagger b \rangle (f^e_k + f^h_k - 1),$$

(5.20)

where for now we neglected the pure correlation functions in (5.13). Furthermore, we scaled the photon assisted polarization by $g_k$ according to $\langle b^\dagger v^e_k c_k \rangle \rightarrow g_k \langle b^\dagger v^e_k c_k \rangle$ so that we only have to deal with the absolute square $|g_k|^2$ of the light-matter interaction. This closed set of equations allows us to access the intracavity photon number and thus the output intensity which is the most basic laser characteristic. In Fig. (5.4) we show stationary solutions for the intracavity photon number as a function of pump power $P$ as well as the carrier density according to

$$N_e = \frac{1}{S} \sum_k f^e_k, \quad N_h = \frac{1}{S} \sum_k f^h_k,$$

(5.21)

where $S$ is the area of the active medium. We assume equal carrier densities for electrons $N_e$ and holes $N_h$ as we describe an optically excited, undoped gain medium. The general structure of Eqs. (5.17)–(5.20) gives rise to a similar behavior to what is found with standard REs (see Fig. 2.6) and with the quantum-optical model discussed in Chapter 3. For larger rates of radiative losses (darker lines) we see the phase-transition-like laser threshold, i.e. a distinct jump in the intracavity photon number over several orders of magnitude, as well as a sudden clamping of the carrier density at the threshold pump power. With decreasing radiative losses (lighter blue lines), the threshold transition becomes more gradual while shifting to lower pump powers as it was the case for standard REs as well. In the extreme case of $\gamma_{nl} = 0$ the I/O
5.2 Rate Equation Limit, Optical Gain, and the $\beta$-Factor

It is instructive at this point to highlight the connection of the equations we developed in the previous section to those from standard RE theory. In doing so we gain a better understanding for how the different contributions in the equations must be interpreted and what the scope of our model is. Eqs. (5.17)–(5.20) can be developed into a RE-like form. To do so, we assume that changes in the carrier populations and photon number happen on a much longer time scale than the polarization lifetime, which is given by $\kappa + \Gamma$. Then, the photon assisted polarization follows the other quantities adiabatically and can be eliminated. This is referred to as the rate equation limit, optical gain, and the $\beta$-factor

### Table 5.1: Parameters used in Fig. 5.4.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\kappa$</td>
<td>0.6 ps$^{-1}$</td>
</tr>
<tr>
<td>$\Gamma$</td>
<td>5 ps$^{-1}$</td>
</tr>
<tr>
<td>$\gamma_{nl}$</td>
<td>30 ps$^{-1}$</td>
</tr>
<tr>
<td>$S$</td>
<td>1.58 $\mu$m$^2$</td>
</tr>
<tr>
<td>$g_0$</td>
<td>0.05 ps$^{-1}$</td>
</tr>
</tbody>
</table>
equation approximation [83] giving
\[
\frac{d}{dt} f_k^e = -2\frac{|g_k|^2}{\kappa + \Gamma} L(\delta_k) \left(f_k^e f_k^h + \langle b^\dagger b \rangle (f_k^e + f_k^h - 1) \right) \\
- \gamma_m f_k^e f_k^h - \gamma_{rel} (f_k^e - f_k^{FD}) + P f_k^p (1 - f_k^e),
\]
(5.22)
\[
\frac{d}{dt} f_k^h = -2\frac{|g_k|^2}{\kappa + \Gamma} L(\delta_k) \left(f_k^e f_k^h + \langle b^\dagger b \rangle (f_k^e + f_k^h - 1) \right) \\
- \gamma_m f_k^e f_k^h - \gamma_{rel} (f_k^h - f_k^{FD}) + P f_k^p (1 - f_k^h),
\]
(5.23)
with the \( k \)-dependent detuning \( \delta_k = (\epsilon_k^e - \epsilon_k^h - \hbar \omega) \) and the Lorentzian linewidth function
\[
L(\delta_k) = \frac{(\kappa + \Gamma)^2}{(\kappa + \Gamma)^2 + \delta_k^2}.
\]
The photon number EoM becomes
\[
\left( \frac{d}{dt} + 2\kappa \right) \langle b^\dagger b \rangle = \sum_k 2\frac{|g_k|^2}{\kappa + \Gamma} L(\delta_k) \left(f_k^e f_k^h + \langle b^\dagger b \rangle (f_k^e + f_k^h - 1) \right).
\]
(5.24)
We are mainly interested in steady state solutions of the system, in which the adiabatic elimination of the polarization becomes analytically correct. Furthermore, for \( k \)-states with large detuning from the cavity mode the first term in the right hand side of (5.20) leads to small but fast oscillations of the polarization, which are numerically unfavorable and are conveniently removed in the set of equations resulting from the adiabatic elimination. We therefore used these equations to evaluate the I/O characteristics and the population functions as shown in Fig. 5.4.

In order to derive REs we now sum (5.22) and (5.23) over all \( k \) states according to (5.21) and take the continuous limit
\[
\frac{1}{S} \sum_k \ldots \rightarrow \int dk^2 \ldots
\]
(5.25)
where \( S \) is the area of the 2D gain material. This removes the \( k \) dependence from the equations. Carrier-population functions are shown in Fig. 5.5. They are given by Fermi-Dirac, quasi-equilibrium distributions at low densities and show only small deviations from thermal equilibrium at higher densities as seen in the inset of Fig. 5.5. Therefore, we approximate the non-equilibrium distribution functions \( f_k^\nu \), \( \nu = \{ e, h \} \) with thermalized distributions
\[
n_k^\nu = \frac{1}{e^{(\epsilon_k^\nu - \mu_\nu)/k_B T} + 1}.
\]
(5.26)
which is referred to as quasi-equilibrium approximation [83]. In general, to find the right chemical potential \( \mu_\nu \) for the Fermi-Dirac distributions for a fixed carrier density, the expression
\[
N = \frac{1}{S} \sum_k n_k(\mu) = \int dk^n n_k(\mu)
\]
(5.27)
has to be solved numerically for \( \mu \). However, in the special case of a 2D density of states and carrier energies as given in (5.4) the chemical potential can be calculated analytically as a function of carrier density, i.e.
\[
\mu_e = k_B T \ln \left( e^{\frac{N_e k_B^2}{\hbar^2}} - 1 \right) + \epsilon_g, \quad \mu_h = k_B T \ln \left( e^{\frac{N_h k_B^2}{\hbar^2}} - 1 \right).
\]
(5.28)
We use these expressions in the following to calculate the optical gain as well as the spontaneous-emission contributions in the quasi-equilibrium approximation.
5.2. RATE EQUATION LIMIT, OPTICAL GAIN, AND THE $\beta$-FACTOR

Figure 5.5: Population functions for a) electrons and b) holes with increasing pump power. The population functions closely follow Fermi-Dirac distributions, due to the thermalization of the carriers. For high pump powers (red curves) the population do not grow any further due to fast stimulated emission leading to clamping of the carrier density. Inset: Small deviations from the quasi-equilibrium functions (spectral hole burning) are observed due to fast stimulated emission.

Semiconductor gain

The gain mechanism in our treatment is described by a free carrier model. In our equations it is calculated dynamically from the non-equilibrium distribution functions $f_{\nu k}$. For derivation of the REs we calculate the gain in the quasi-equilibrium approximation as

$$G(\hbar\omega, N) = \sum_k \frac{2|g_k|^2}{\kappa + \Gamma} L(\delta_k)(n^e_k + n^h_k - 1) = S \int \frac{d^2k}{4\pi} \frac{2|g_k|^2}{\kappa + \Gamma} L(\delta_k)(n^e_k + n^h_k - 1),$$

(5.29)

which is found by integrating the stimulated emission part of Eqs. (5.22) and (5.23) over all carrier momenta $k$. In Fig. 5.6 we show gain spectra for different carrier densities as a function of the detuning of the incident light from the optical band gap. At carrier densities of around $N = 10^{12} \text{ cm}^{-2}$, gain sets in and facilitates stimulated emission. Fig. 5.7 shows the gain as a function of carrier density with detuning $\delta = \hbar\omega - \epsilon_g = 10 \text{ meV}$ above the band gap as given by the vertical line in Fig. 5.6. For most of the density range the gain (at fixed energy as well as the peak gain) approximately has a logarithmic dependence on the density.

The free-carrier gain model that we discuss here does not include the effects of many-body interactions to the gain such as Coulomb interactions among the carriers. For semiconductor quantum wells the influence of Coulomb effects has been investigated thoroughly in the literature [83, 128, 129] and they result in some non-negligible changes in the gain spectra of semiconductor QWs. However, in conventional semiconductors these changes amount to a shift in the gain spectrum and a difference in the peak gain height. Here we are considering a single mode laser theory where the cavity mode with a narrow spectral linewidth picks up material gain at a single frequency (c.f. vertical line in Fig. 5.6). Shifts in the gain spectrum can be accounted for by an adjusted resonance frequency and changes in peak height by an adjusted effective confinement factor. Our main focus lies on calculating the emission properties of nanolasers in terms of the photon correlations in the emitted light. To that end, using our
semiconductor laser model for III/V gain media, we obtain a consistent description of several key device characteristics, namely the I/O-curve, first- and second-order correlation functions, coherence time and linewidth. The simplicity of the gain material’s description allows us to calculate this range of emission characteristics on a microscopic level, while providing avenues for future extensions of the material description.

Spontaneous emission and the $\beta$ factor

After analyzing the gain mechanism described by our model we now turn to the spontaneous-emission contribution, where our description of the emission dynamics differs in two important ways from standard REs. The quantum-optical treatment of the light field naturally gives rise to the spontaneous emission proportional to $n_e^k n_h^k$. This is not the case in the semiclassical description where SE must be introduced on a phenomenological basis. Secondly, the rates of spontaneous emission into lasing and non-lasing modes depend on the excited carrier density in different ways. As a result, the $\beta$ factor does not appear in the equations as a constant device parameter. Instead, we calculate $\beta$ as a function of carrier density $N$ from the quasi-equilibrium distributions and it represents an excitation-dependent quantity.

The radiative losses by spontaneous emission into non-lasing are evaluated for the RE description from Eqs. (5.22) and (5.23) as

$$B_{SE}^{nl}(N) = \frac{1}{\mathcal{S}} \sum_k \gamma_{nl} n_e^k n_h^k,$$

with $n_e^k$, $n_h^k$ the Fermi-Dirac distribution functions in the quasi-equilibrium approximation. We evaluate (5.30) as a function of carrier density, shown in Fig. 5.8 (solid line), and find that for small densities the SE follows a $N^2$ dependence (dashed line for comparison) while for high densities the dependence is $\sim N^{3/2}$. Thus, in the relevant density range we find an effective
spontaneous-emission rate \( \tilde{\gamma}_N N^2 \) in agreement with standard RE theories. The same holds true for the spontaneous-emission rate into the lasing mode. We evaluate the related term from (5.22)

\[
B_{SE}(N) = \frac{1}{B} \sum_k \frac{2|g_k|^2}{\kappa + 1} L(\delta_k) n_k^e n_k^h, 
\]

(5.31)

where again for small densities we find an effective SE rate \( \tilde{\gamma}_N N^2 \), as shown in Fig.5.9. For higher densities the emission saturates due to Pauli blocking of states in resonance with the cavity mode, i.e. the population functions \( n_k^{e/h} \) approach 1 in the vicinity of the cavity mode energy.

We pointed out that \( \beta \) is not a free parameter in our theory. Instead, we calculate \( \beta \) as the fraction of total spontaneous emission that is emitted into the lasing mode. It becomes an excitation dependent quantity and is evaluated as

\[
\beta(N) = \frac{\sum_k \frac{2|g_k|^2}{\kappa + 1} L(\delta_k) n_k^e n_k^h}{\sum_k \left( \frac{2|g_k|^2}{\kappa + 1} L(\delta_k) n_k^e n_k^h + \gamma_{nl} n_k^e n_k^h \right)}. 
\]

(5.32)

Non-constant \( \beta \) factors were also discussed in [85, 86, 130, 131, 132] in the context of QD based nanolasers, where superradiant effects come into play. Similarly, when the full equations (5.17)–(5.20) are solved, \( \beta \) can be evaluated from the non-equilibrium populations \( f_k^{e/h}, f_k^{h} \) according to (5.32). In Fig.5.10 \( \beta \) is shown as function of the carrier density. We see that a constant \( \beta \), as normally assumed in REs, is a good approximation for low densities, which are typically sufficient to reach the lasing threshold. The constant approximation relates to the effective rate of spontaneous emission as \( \beta = \tilde{\gamma}_N / (\tilde{\gamma}_N + \gamma_{nl}) \).
Figure 5.8: Density dependence of the spontaneous-emission contribution into non-lasing modes in the rate equation approximation according to Eq. (5.30). For small carrier densities we find a $\tilde{\gamma}_n N^2$ law (dashed line) in agreement with standard REs. Typical carrier densities (clamping levels) stay in the regime where the square dependence is valid. Here we have $\tilde{\gamma}_n = 1.88 \times 10^{-5} \text{cm}^2 \text{s}^{-1}$.

Figure 5.9: Density dependence of the spontaneous-emission contribution into the lasing mode. For low densities we again find a $N^2$ dependence, which is indicated by a dashed line. For higher densities, Pauli blocking of $k$-states in resonance with the cavity mode lead to saturation of the emission rate. Here we have $\gamma_l = 3.96 \times 10^{-5} \text{cm}^2 \text{s}^{-1}$. 
5.3. FIRST-ORDER COHERENCE AND LINEWIDTH

Combining all results from the previous discussions results in the REs

\[
\dot{N} = P - \tilde{\gamma}_l N^2 - \frac{V_p}{S} G(N) N_p - \tilde{\gamma}_{nl} N^2, \quad (5.33)
\]

\[
\dot{N}_p = \frac{S}{V_p} \tilde{\gamma}_l N^2 + G(N) N_p - N_p / \tau_p, \quad (5.34)
\]

with emission rates \(\tilde{\gamma}_l\) and \(\tilde{\gamma}_{nl}\) that are approximately constant if clamping of the carrier density occurs at sufficiently low densities. Here, \(N\) is the carrier density (identical for conduction or valence band) and \(N_p\) is the photon density \(n/V_p\) in the cavity volume \(V_p\) and the cavity decay time is given by \(\tau_p = 1/(2\kappa)\). The active area \(S\) and cavity mode volume \(V_p\) appear in the equations in order to reconcile the relative dimensions of the carrier density in the 2D gain medium and the photon density, similar to the REs in Section 4.3. In the literature many different conventions for formulating REs can be found. Although different versions exhibit the same (or similar) mathematical structure in general, they will reflect the specific device geometry of cavity and gain medium, respectively. This explains the differences in the REs given here and those in the previous chapters.

5.3 First-order coherence and linewidth

Up to now we have studied I/O characteristics and carrier densities that are also accessible with standard REs. We now go towards a more comprehensive picture of the emission properties of semiconductor nanolasers, by calculating additional quantities that go beyond the RE description of nanolasers. For now we still treat the dynamics on the doublet level of the cluster expansion and calculate the first-order coherence properties of the light emission from time dependent operators of the quantized light field. The degree of first-order coherence given by
the normalized first-order correlation function

$$g^{(1)}(\tau) = \frac{\langle \hat{b}^\dagger(t)\hat{b}(t+\tau) \rangle}{\langle \hat{b}^\dagger(t)\hat{b}(t) \rangle},$$

(5.35)

where $\langle \hat{b}^\dagger(t)\hat{b}(t) \rangle$ is the steady state photon number in the cavity. We apply the quantum regression theorem (QRT) discussed in Section 2.6. It states that the dynamics two-time correlation functions is related to single-time expectation values which helps us to evaluate the expression $\langle \hat{b}^\dagger(t)\hat{b}(t+\tau) \rangle$. From the Hamiltonian (5.2) we find for the average mode amplitude

$$\frac{d}{dt} \langle \hat{b}(t) \rangle = -i(\omega + i\kappa)\langle \hat{b}(t) \rangle + g \sum_k \langle p_k(t) \rangle,$$

(5.36)

and similarly for the microscopic polarizations $\langle p_k(t) \rangle = \langle v_k^\dagger c_k(t) \rangle$

$$\frac{d}{dt} \langle p_k(t) \rangle = -i (\epsilon_k^e - \epsilon_k^h) \langle p_k(t) \rangle + g^* \left( \langle bc_k^\dagger c_k(t) \rangle - \langle bv_k^\dagger v_k(t) \rangle \right) .$$

(5.37)

The time derivatives are linear equations of other operators’ expectation values and the QRT can be applied according to [72]. In order to derive an equation of motion for $g^{(1)}(\tau)$ we define

$$G(\tau) = \langle \hat{b}^\dagger(t)\hat{b}(t+\tau) \rangle e^{i\omega \tau},$$

(5.38)

$$P_k(\tau) = \langle \hat{b}^\dagger(t)v_k^\dagger(t+\tau)c_k(t+\tau) \rangle e^{i\omega \tau} .$$

(5.39)

Figure 5.11: Coherence times (left) calculated according to Eq. (5.42) and emission linewidth (right) determined from the emission’s power spectrum (5.43). Both quantities show a gradual transition that indicates the lasing threshold. The coherence time saturates around $\tau_p = 10$ ps, which becomes important in the quantum optical modeling of experimental correlation measurements where the influence of limited detector resolution becomes visible if the coherence time is too small.
Then, using the QRT, we find from the time derivatives of $\langle p_k(t) \rangle$ and $\langle b(t) \rangle$ the dynamics of $G(\tau)$

$$
\hbar \frac{d}{dt} G(\tau) = \sum_k g^* (b^\dagger(t) v^\dagger_k(t + \tau) c_k(t + \tau)) e^{i\omega \tau} - \kappa (b^\dagger(t) b(t + \tau)) e^{i\omega \tau} = \sum_k g^* P_k(\tau) - \kappa G(\tau),
$$

(5.40)

and of $P_k(\tau)$

$$
\hbar \frac{d}{dt} P_k(\tau) = g (b^\dagger(t) b(t + \tau)) \left( \langle c^\dagger_k(t + \tau) c_k(t + \tau) \rangle - \langle v^\dagger_k(t + \tau) v_k(t + \tau) \rangle \right) e^{i\omega \tau} - (\Gamma + i\delta_k) (b^\dagger(t) v^\dagger_k(t + \tau) c_k(t + \tau)) e^{i\omega \tau} = g (f^c_k - f^v_k) G(\tau) - (\Gamma + i\delta_k) P_k(\tau),
$$

(5.41)

where we again used the cluster expansion approach to truncate the arising hierarchy of equations on the doublet level. Initial conditions $G(\tau = 0)$ and $P_k(\tau = 0)$ are given by the steady state values of the photon number and the polarization amplitudes.

The first-order coherence function gives direct access to the coherence time as well as the linewidth of the emission [60]. The coherence time $\tau_c$ is related to the degree of first-order coherence according to

$$
\tau_c = \int_{-\infty}^{\infty} d\tau \left| g^{(1)}(\tau) \right|^2 = \int_{-\infty}^{\infty} d\tau \frac{|G(\tau)|^2}{|G(0)|^2}.
$$

(5.42)

It defines the time scale on which the electric field maintains its coherence and can show interference phenomena. The normalized emission spectrum is given by the Fourier transform of the correlation function from which the emission linewidth can be determined

$$
F(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau \left| g^{(1)}(\tau) \right| e^{i\omega \tau}.
$$

(5.43)

Fig. 5.11 shows the coherence time and linewidth for nanolaser parameters as given in Table 5.1 with decreasing rates of radiative losses. The increase in coherence time (left panel) associated with the laser threshold occurs in the same pump power range as the intensity threshold jump shown in Fig. 5.4. However, the coherence time stays in a range of the order of 10 ps which corresponds to coherence lengths of around 3 mm while conventional laser have coherence lengths of 20 cm and more. The reduced coherence time even above threshold can be traced back to the strong spontaneous emission into the cavity mode. Although $\tau_p$ can be determined experimentally [133, 134], it is often much easier to determine the spectral linewidth of the emission. For perfectly Lorentzian line shapes, the linewidth $\gamma$ at full width at half maximum (FWHM) relates to the coherence time as $1/\tau_p = \gamma/2$. In Fig. 5.11 (right panel) the emission linewidth is shown to decrease with increasing carrier density.

### 5.4 Time-resolved Photoluminescence

The doublet equations of motion also allow to model the temporal emission dynamics from nanolaser devices as measured in time-resolved photoluminescence (TRPL) experiments. Investigating TRPL time traces is important as they are, via the rate of spontaneous emission, connected to the light-matter interaction strength between the active material and the cavity mode. The interaction strength is otherwise difficult to access in extended gain media.
nanolasers. In QD nanolasers, for comparison, the value for the light-matter interaction has been determined from TRPL measurements [78] by examining the lifetime of the photoluminescence assuming that all QDs, which are in resonance with the cavity, have a similar interaction strength. The procedure is generally more difficult in the case of nanolasers with extended gain media. Here, charge carriers in a quasi-continuum of states interact with the cavity mode at different detunings and thus have effectively uneven interaction strengths. However, in reproducing TRPL traces that are recorded in experimental measurements, we can fix the value for the interaction constant $g_0$ (c.f. Eq. (D.22)). The ability to assess the light matter interaction in QW based nanolasers is a feature of our theory that significantly adds to the consistency of the device modeling.

Assuming fast carrier generation in the gain material by an ultrashort laser pulse, we solve the EoM (5.22)–(5.24) starting with a fixed carrier density in the conduction and valence band. Results for typical device parameters are shown in Fig. 5.12. For low excitation, i.e. low initial carrier densities we find a non-exponential decay that approximately follows a $1/t$ dependence. For higher carrier densities the initial onset of stimulated emission quickly depletes the carrier densities to below the threshold value as can be seen in the inset. Below threshold the decay time is mainly determined by non-radiative losses, governed by $\gamma_{nl}$. The fast recombination, observed for the highest density value in Fig. 5.12, is governed by $g_0$, i.e. the light matter interaction. In Section 6.1 we will apply the model to experimentally recorded time traces in order to determine the values of $\gamma_{nl}$ and $g_0$ in a consistent manner.

### 5.5 Second-order correlation function

So far, all calculated laser characteristics were accessible from the equations of motion on the doublet level of the cluster expansion. However, as discussed in the introduction, we are

![Figure 5.12: Time-resolved photoluminescence time traces, calculated from the doublet equations (5.22)–(5.24) without optical pump. A fast optical pulse is assumed to create an initial carrier density $N_0$. For the largest $N_0$ the system is above threshold, resulting in a fast depletion of carriers by stimulated emission, also reflected in a fast decay of the intensity. The inset shows a magnification of the initial intensity peak for better visibility.](image-url)
interested in a definitive criterion to distinguish thermal and coherent emission. This can be given by the photon statistics of the emitted light that imprints itself onto the second-order correlation function

\[ g^{(2)}(0) = 2 + \frac{\delta(b^{\dagger}b^{\dagger}bb)}{\langle b^{\dagger}b \rangle^2}, \tag{5.44} \]

which we introduced previously in (3.13). We can access it by deriving EoM for the pure two-photon correlation \( \delta(b^{\dagger}b^{\dagger}bb) \). In order to do so, we extend the doublet equations (5.20)–(5.20) by the triplet correlation terms in (5.13). The addition of higher correlation functions represents only a minor correction to the population functions and the photon number (see also discussion around Fig. 3.6). It nevertheless gives direct access to \( g^{(2)}(0) \) by expanding the system of equation up to the quadruplet level of the cluster expansion. The additional EoM are given by

\begin{align*}
\left( \frac{\hbar}{d} + 2\kappa \right) \delta\langle b^{\dagger}b c_k^e c_k^c \rangle &= -2|g_k|^2 \text{Re} \left[ \delta\langle b^{\dagger}b^{\dagger}b v_k^e c_k \rangle + (\langle b^{\dagger}b \rangle + f_k^b) \langle b^{\dagger}v_k^e c_k \rangle \right], \tag{5.45} \\
\left( \frac{\hbar}{d} + 2\kappa \right) \delta\langle b^{\dagger}b v_k^e v_k^e \rangle &= 2|g_k|^2 \text{Re} \left[ \delta\langle b^{\dagger}b^{\dagger}b v_k^e c_k \rangle + (\langle b^{\dagger}b \rangle + f_k^b) \langle b^{\dagger}v_k^e c_k \rangle \right], \tag{5.46} \\
\left( \frac{\hbar}{d} + 3\kappa + \Gamma \right) \delta\langle b^{\dagger}b^{\dagger}b v_k^e c_k \rangle &= -i(\epsilon_k^e - \epsilon_k^b - \hbar\omega) \delta\langle b^{\dagger}b^{\dagger}b v_k^e c_k \rangle \\
&\quad - 2|g_k|^2 \langle b^{\dagger}v_k^e c_k \rangle^2 - (1 - f_k^c - f_k^b) \delta\langle b^{\dagger}b^{\dagger}bb \rangle \\
&\quad + 2f_k^b\delta\langle b^{\dagger}b c_k^e c_k \rangle - 2f_k^c\delta\langle b^{\dagger}b v_k^e v_k \rangle \\
&\quad + 2\langle b^{\dagger}b \rangle (\delta\langle b^{\dagger}c_k^e c_k \rangle - \delta\langle b^{\dagger}v_k^e v_k \rangle), \tag{5.47} \\
\left( \frac{\hbar}{d} + 4\kappa \right) \delta\langle b^{\dagger}b^{\dagger}bb \rangle &= 4 \sum_k |g_k|^2 \text{Re} \left[ \langle b^{\dagger}b^{\dagger}b v_k^e c_k \rangle \right], \tag{5.48} 
\end{align*}

which form a closed set of equations. Again, we neglected correlations between emitters in different states as well as correlations of more than four particles and we scaled the extended polarization term \( \delta(b^{\dagger}b^{\dagger}b v_k^e c_k) \) by the light matter coupling \( g_k \) analogue to \( \langle b^{\dagger}v_k^e c_k \rangle \). The triplet correlations in Eqs. (5.45) and (5.46) couple to doublet quantities as well as the extended polarization. Its dynamics, in turn, is similar to the dynamics of the photon assisted polarization (5.20). A generalized stimulated emission term is given by the second term in the second line of (5.47) being proportional to the inversion. The spontaneous-emission contribution in (5.20) is here reflected in the coupling to the triplet correlations. Finally, the extended polarization is the source term for the two-photon correlations \( \delta(b^{\dagger}b^{\dagger}bb) \) in (5.48), in the same way as the photon assisted polarization is the source term for the photon number \( \langle b^{\dagger}b \rangle \).

We evaluate the quadruplet laser equations in the steady state for the same set of parameters as those used in the previous sections. The results are shown in Fig. 5.13. The transition of \( g^{(2)}(0) \) from a value of 2 to 1 clearly indicates the change in photon statistics from a thermal distribution below threshold to a Poissonian one above threshold. For comparison, the I/O characteristics and linewidth are plotted with pump power axes aligned. Several features are clearly visible. For large rates of radiative losses (darker curves) the threshold transition, as given by \( g^{(2)}(0) \), is very sudden. It becomes more gradual (lighter curves) for decreasing losses. 

5.5. SECOND-ORDER CORRELATION FUNCTION
Figure 5.13: Calculated photon-correlation function $g^{(2)}(0)$ (left panel) as function of pump rate. With decreasing radiative losses (increasing $\beta$) the transition to coherent emission ($g^{(2)}(0) = 1$) becomes more gradual and ceases to shift to lower pump rates. Comparing the results with the photon number and linewidth (right panel) e.g. for $\gamma_{nl} = 2 \times 10^{-3}$ ps$^{-1}$ at pump rates indicated by the red vertical line shows the delayed onset of coherent emission w.r.t the intensity threshold as discussed in Chapter 3.

$(\gamma_{nl} \rightarrow 0$, i.e. $\beta \rightarrow 1$). This is also reflected in the output intensity and linewidth (right panel). The threshold jump in the intensity becomes smaller and approaches zero, while $g^{(2)}(0)$ and linewidth still show a transition at finite pump powers. The delayed onset of coherent emission as discussed in Chapter 3 is also observable here. Take as an example the results in Fig. 5.13 for $\gamma_{nl} = 2 \times 10^{-3}$ ps$^{-1}$ at the pump power indicated by red vertical lines. While intensity and linewidth are clearly beyond of what could be identified as the threshold transition, $g^{(2)}(0) \approx 1.8$ still indicates largely incoherent emission. Another important observation is that $g^{(2)}(0)$, with decreasing radiative losses, converges to its extremal value much faster than the other quantities do. This shows that our findings here reflect our results from Section 3.3, i.e. we again find that lowering the coherence threshold, in comparison to the intensity threshold jump, stops being dependent on an increase in $\beta$.

5.6 Device geometry and scaling behavior

The quantum-optical laser theory in this chapter aims at the description of high-$\beta$ nanolasers with two-dimensional materials as gain media. The basic interaction mechanisms of light and carriers in these systems are the same as in more conventional semiconductor lasers (e.g. VCSELs). However, they exhibit a different device geometry that needs to be accounted for in the modeling. It leads to the specific form of the laser equations we derived and subsequently of the rate equations discussed in Section 5.2. In particular, the extent of the gain material and its overlap with the cavity mode appear in a different relation in comparison to VCSEL devices, which justifies an independent discussion.

Appendix D discusses the derivation of the light-matter interaction of the cavity light field with the carriers in the 2D gain medium. Bloch waves are considered as electronic basis states
5.6. DEVICE GEOMETRY AND SCALING BEHAVIOR

![Diagram](image)

Figure 5.14: Examples of laser structures (schematic) with different device geometries. For both examples the form of the cavity mode function $u(r)$ is given. a) VCSEL structure with a QW active region (white) sandwiched between upper and lower distributed Bragg reflectors and a capping layer (red). The in-plane mode function is assumed to be a plane wave extending over the whole area in the 2D gain medium. b) Nanolaser with 2D gain medium (red) interacting with the spatially localized mode of a photonic crystal cavity. Localization takes place in all three dimensions and the mode function has no free-wave component.

In the 2D layer with in-plane momenta $k_\parallel$ [38]

$$\varphi_{k_\parallel}(r) = \frac{1}{\sqrt{S}} e^{i k_\parallel \cdot r} \xi(z) u(z),$$

(5.49)

where $S$ is the quantization area that arises in the standard treatment of QW electronic states in the Bloch formalism and $\xi(z)$ is the electron wave function confined along the $z$-direction perpendicular to the 2D plane. This is the same ansatz as it is used in the theoretical modeling of VCSEL devices. The difference, however, lies in the description of the light field. For VCSELs a cavity light field is assumed that is spatially homogeneous along the in-plane direction of the cavity/QW [117], given by

$$u(r) = \frac{1}{\sqrt{S}} e^{i q_\parallel \cdot r} u(z),$$

(5.50)

i.e. we have a plain-wave part in the in-plane direction that is separated from the confinement along the $z$-direction, as is illustrated in Fig. 5.14 a). This translational invariance in the QW plane has the consequence that the quantization area $S$ drops out of the equation when transitioning to a quasi-continuum of states as in Eq. (5.25). This allows us to formulate a scale invariant description of the theory.

When describing the localized mode of the nanolaser cavity, as depicted in Fig. 5.14 b), the encountered situation is different because the strong localization of the mode breaks the translation invariance of the system along the QW plane. The Bloch wave formalism in this case is treated as an approximation where finite size effects are neglected and the normalization area $S$ can be interpreted as the physical area of the 2D layer. As a result, the model is not scale invariant (relative to its in-plane extension) and an explicit value for $S$ must be chosen for the modeling. At the same time, the strong electric fields in the localized cavity mode lead to a range of local effects, like spatial hole burning and spatially varying interaction strength.
When we consider furthermore the finite carrier mobility, it becomes clear that never all carriers in the 2D gain layer will interact with the mode. As a consequence, and since including the mentioned effects is beyond the scope of our model, we treat the QW area $S$ as an effective parameter. It can be thought of as a measure for the amount of charge carriers that interact with the cavity mode, which is similar to the number of quantum dots, each containing one exciton, in a QD-based nanolaser. Thereby, a connection to the more idealized laser model in Chapter 3 is established.

We investigate the scaling behavior of the lasing characteristics with the system size. This is possible due to the explicit nature of how the gain medium area enters the laser equations, which becomes more clear when looking at the stationary solution of the laser equation (5.22)–(5.24). One observes that the equations for the photon number (5.24) stay invariant under the scaling $|g_k|^2 S = \text{const.}$, while the same is not true for the populations in (5.22) and (5.23). However, in all quantities that we calculate as a function of the pump power, only the sums over all carrier momenta $k$ enter (cf. Eq. (5.40) for the first-order coherence as an example). If we sum the EoM for the populations, we obtain a symmetric set of equations in the sense that we can absorb the equations’ dependence on the area into $|g_k|^2$. For that we also rescale the radiative losses $\gamma_{nl}$ as well as the pump power $P$ and relaxation rate $\gamma_{\text{rel}}$ in order to obtain a scale invariant model. Results, where this scaling behavior can be seen for different device characteristics are shown in Fig. 5.15. We observe that all results stay invariant for different amounts of gain medium $S$. For better visibility we did not scale the pump power in this plot.

These observations permit two independent interpretations of the area $S$. On the one hand we can associate it with an abstract system size as it is introduced in the formulation of the electronic Bloch functions. In this respect we can absorb it in the light-matter interaction strength, as well as in the radiative loss, relaxation, and pump rates $\gamma_{nl}$, $\gamma_{\text{rel}}$, and $P$. In this way we obtain a scale invariant model. On the other hand, we can interpret $S$ as the physical size of the system, i.e. the area of the active material, which makes intuitive sense as we deal with localized modes of micro- and nanocavities and their overlap with the gain medium is a function of the area and should have an influence on the laser dynamics. This second interpretation is further justified by results shown in Fig. 5.16. There, we only reduced $S$ without the scaling of other parameters as discussed above. If the area becomes too small, i.e. the amount of gain medium decreases, insufficient gain does not facilitate lasing. In this case the laser stays below the threshold while the intensity saturates, the carrier density shows no clamping and $g^{(2)}(0)$ stays in the thermal regime close to a value of 2.
5.6. DEVICE GEOMETRY AND SCALING BEHAVIOR

![Graph](image)

Figure 5.15: Different device characteristics are shown for decreasing quantization area $S$ while keeping $S \times |g_k|^2 = \text{const}$. We also scale radiative losses $\gamma_{\text{nl}}$ and the relaxation rate $\gamma_{\text{rel}}$ accordingly, so that the full set of laser equations stays invariant. For better visibility we did not scale the pump rate, effectively shifting the results along the x-axis.

![Graph](image)

Figure 5.16: When only scaling the area $S$, it becomes clear that it can be interpreted as the quantitative amount of gain medium that interacts with the cavity mode. For insufficient $S$ the laser stays below threshold as the medium provides too little gain to compensate losses. The I/O curve does not show a threshold jump and saturates, the carrier density does not show clamping, there is no linewidth narrowing and $g^{(2)}(\tau)$ stays close to 2 indicating thermal emission over all pump rates.
Chapter 6

Modeling experimental devices

In this chapter we present results of theoretical and experimental investigations of two types of nanolasers and apply our semiconductor laser theory developed in the previous chapter. The investigated device classes are metal clad coaxial nanolasers (CNL) as reported in [50] and nitride based nanobeam lasers [15], both featuring semiconductor QWs as gain medium. These studies were done in collaboration with the research team of S. Reitzenstein at the Technical University Berlin who performed quantum optical measurements of various device characteristics. Most notably, two-photon correlation were measured, in order to assess the transition to coherent emission in the investigated devices. Our theoretical modeling supports the experimental findings by calculating device characteristics from different point of view and combination of the results provide a comprehensive picture of the lasing process. In this way, experiment and theoretical analysis work side by side to advance the understanding of lasing in the nanoscale regime and provide avenues for the improvement of future experimental designs.

We assess the lasing behavior in the discussed structures that all operate in the high-$\beta$ regime and feature metallic or photonic crystal nanocavities. These devices are, with respects to their geometry and material compositions, quite different from TMD-based nanolasers reported in the literature. However, in principle they work in the same device (or parameter) regime and they are equally applicable to the discussion on the delayed onset of coherence from the previous chapters. The fingerprints of this behavior in the device characteristics are visible in the theoretical modeling as well as in the experimental measurements.

6.1 Metal-clad coaxial nanolasers

The devices investigated in [50] are metal-clad coaxial nanolasers (CNL) [13, 135]. They are comprised of a multi QW structure embedded in a metallic cavity that allow to confine light on smaller dimension than what would be possible with dielectric cavities [37]. Therefore, they present a further step towards miniaturization of nanolasers for optoelectronic integration. In Fig. 6.1 a) a schematic view of the CNL is depicted. Six ring-shaped InGaAsP QWs shown in red are separated by barrier material (orange) and surrounded by the metallic (silver) inner core and the outer cladding. A SiO$_2$ plug to the bottom separates the dielectrics from the metallic cavity. Fig. 6.1 b) shows a scanning electron microscope image of the dielectric components after dry etching of the structure, but prior to applying the metallic layer by vapor deposition. Details on the manufacturing process can be found in the Supplementary Information of [50]. Two different CNLs where investigated with similar geometry but varying inner and outer radii for the QWs. This has the effect of changing the $Q$ factor of the modes supported by the cavity and to alter their spectral positions. With that, also the effective $\beta$-factor is modified as the
Figure 6.1: Investigated laser structures. 

a) Schematic (cross sectional) view of the metal clad CNL. The gain medium consist of 6 InGaAsP QWs (red) separated by the barrier material (orange). The silver inner core and a metal cladding (silver applied by vapor deposition) constitute the cavity for optical feedback. 

b) Scanning electron microscope image of the dielectric components of the CNL prior to the metal deposition. The CNL structures were developed and fabricated in the group of M. Khajavikhan at CREOL, University of Central Florida.

spectral overlap of the cavity mode and the active medium’s excitation spectrum changes. We label the investigated structures CNL1 ($R_{out}$: 295 nm, $R_{in}$: 55 nm) and CNL2 ($R_{out}$: 315 nm, $R_{out}$: 75 nm).

Device characteristics

In Fig. 6.2 we show a range of device characteristics for CNL1(2). Blue dots indicate measurement results from experiment while black, solid lines are obtained from the theoretical investigation of the devices that is based on the model introduced in the previous chapter. The output intensity Fig. 6.2 a) of CNL1 shows an almost linear dependence on the pump power, which is associated with a (near) perfect spontaneous emission coupling into the lasing mode, also indicated by the $\beta$ factor close to unity. Thresholdless, i.e. linear I/O characteristics have been demonstrated before in QD-based nanolasers with dielectric cavities [9, 10, 11]. However, observing this behavior in a metallic-cavity nanolaser with with extended (QW) gain medium, is what distinguishes the presented investigation from previous publications. In comparison, CNL2 shows a distinct S-bend in the I/O characteristics indicating a clear (intensity-) threshold (c.f. Chapter 3), which is also reflected in the effective $\beta$ factor that stays below 0.15\(^1\). Dashed lines in Fig. 6.2 (a-b) indicate a mean cavity photon number of 1, which is the quantum threshold criterion. CNL1 and CNL2 meet this criterion at pump rates of approximately 65 kW cm\(^{-2}\) and 155 kW cm\(^{-2}\), respectively. In view of the results on photon correlation (Fig. 6.5) that are discussed below, we will see that at these excitation intensities the photon correlations will indicate largely incoherent emission, reflecting the discussion on the delayed onset of coherence in Chapter 3.

The pump power dependent $\beta$ factor is shown in Fig. 6.2 (c-d). The reduction of $\beta$ at higher pump powers can be explained by considering the quasi-equilibrium states of the carrier populations. In quasi-equilibrium, clamping of the total carrier density sets in at the laser

\(^1\)From the point of view of more conventional lasers (gas-, or dye lasers or conventional semiconductor lasers) a $\beta$ factor around 0.1 is still considered high as other lasers exhibit $\beta$ factors order of magnitude smaller ranging from $10^{-4}$ to $10^{-7}$. It is interesting that in the context of nanolaser a $\beta$ of 0.1 is not considered particularly high, showing the shifting baseline for the perception of these devices, that operate in a different regime.
6.1. METAL-CLAD COAXIAL NANOLASERS

Figure 6.2: Optical properties of both examined CNLs. While experimentally obtained results are indicated by blue dots, solid lines show results from the theoretical modeling. a), b) I/O characteristics. Dashed lines indicate a mean cavity photon number of 1. CNL1 and CNL2 meet this criterion at pump rates of approximately 65 kW cm\(^{-2}\) and 155 kW cm\(^{-2}\), respectively. c), d) Excitation intensity dependent $\beta$ factor calculated from theory. e), f) Emission wavelength of the CNL devices. g), h) Emission linewidth fitted from emission spectra (Fig. 6.4) and calculated from theory (Eq. (5.43). i), j) Change in emission lineshape (see [50] for details).

threshold, which has the effect that the total carrier density remains constant while excess pump power is converted into emitted photons. On the microscopic level described by our model, we see that the carrier populations may still increase in parts of the band structure that are not depleted by stimulated emission into the laser mode. This is illustrated in Fig. 6.3. As a consequence, the carrier distribution functions become non-thermal and exhibit hole-burning around the cavity resonance as discussed previously in Sec. 5.2. The SE rate in the laser mode no longer increases, as the population of $k$-states in the vicinity of the cavity mode (indicated in red) are fixed by the hole-burning effect. At the same time, carriers accumulate in higher-lying
CHAPTER 6. MODELING EXPERIMENTAL DEVICES

Figure 6.3: We illustrate the influence of the carrier population functions for electrons ($f^e$) and holes ($f^h$) on the SE rate in a schematic picture. At the threshold (left panel), the population functions still resemble the thermal case. Above the threshold (right panel), hole burning clamps electron and hole populations in the vicinity of the cavity-mode energy, shown in red. Increasing the excitation power results in the population of energetically higher states in the band structure, which increases the overall SE losses into non-lasing modes (indicated in blue) and thereby lowering $\beta$.

Figure 6.4: Excitation power dependent $\mu$-PL spectra for both CNL devices. At low excitations a broad SE peak is visible in the spectrum, while for higher excitations strong emission from a single cavity mode starts to dominate the spectrum.

$k$ states, which effectively increases radiative losses into non-lasing modes (indicated by the area shaded in blue) and, thereby, lowers the $\beta$ factor.

The emission spectra from the investigated CNLs is depicted in 6.4. For both devices a sharp cavity emission line appears for increasing pump power, from which the output intensity is determined. Comparison of the spectra reveals that the overlap of the cavity mode with the free emission spectrum is larger for CNL1, which again reflects the stronger interaction of mode and gain medium leading to a higher $\beta$ factor. From these spectra, the cavity emission wavelength and linewidth are determined as shown in Fig. 6.2(e-h). Using our theoretical model we calculate the linewidth via the first-order correlation (c.f. Sec. 5.3) and the results agree well with the measured linewidth in Fig. 6.2(g-h). The linewidth has units of angular frequency (rad./ps), as it is obtained from the normalized emission power spectrum (5.43). The
experimentally measured linewidth are given in units of wavelength (nm), instead. In order to convert the units, we use the relation $\Delta \omega / \omega \approx \Delta \lambda_c / \lambda_c$, which approximately holds for small linewidths.

Experimentally, a change in emission wavelength is detected, as well as a change in the emission lineshape from a Lorentzian to a partially Gaussian spectral line. These effects are not modeled by the theory presented here. The changing lineshape might indicate an additional mechanism that introduces an inhomogeneous broadening of the laser emission, e.g. heating effects. These are, however, beyond the scope of our model. Further details can be found in the method section of [50].

Photon correlation measurements

As discussed previously, a measurement of the two-photon correlation function $g^{(2)}(0)$ is necessary to verify the onset of coherent emission from the device. The time-dependent second-order correlation function $g^{(2)}(\tau)$ is measured using a Hanbury Brown and Twiss (HBT) setup with superconducting nanowire single-photon detectors (c.f. Section 2.5). Two of the recorded histograms are depicted in Fig. 6.5 (a-b) (black lines). The measured data is fitted with Gaussian temporal profiles (red) in order to extract the raw values of $g^{(2)}(0)$. The extracted peak heights are shown in Fig. 6.5 (c-d) as black squares. We see that the raw $g^{(2)}(0)$ approaches 1 in the limit of large excitation, but with a maximum value of 1.02 it diverges only weakly from the uncorrelated case. In comparison to the idealized HBT setup introduced in Section 2.5, the experimental data are, among other things, subject to timing jitter of the electronics as well as dark counts and dead times of the photon detectors. As a result, the temporal resolution of the setup is limited and does not allow to resolve photon bunching ($g^{(2)}(0) > 1$) if the decay of $g^{(2)}(\tau)$ happens on a significantly faster time scale than the resolution time. To account for this effect, we model the effect of the limited detector resolution and apply it to our theoretical results for $g^{(2)}(0)$. We assume that the Sigert relation, $g^{(2)}(\tau) = 1 + a |g^{(1)}(\tau)|^2$ (which technically only holds for thermal light with $a = 1$) [60], can be extended to partially coherent light where we use $a = g^{(2)}(0) - 1$. We convolute the estimated $g^{(2)}(\tau)$ with the detector response function, which we model as a Gaussian with $\sigma = \Delta t/(2 \sqrt{2 \ln(2)})$. The FWHM is $\Delta t = 80$ ps and represents the detector’s temporal resolution. We only plot the value of the convoluted correlation function (denoted by $\tilde{g}^{(2)}(\tau)$) at $\tau = 0$, so we specifically calculate

$$\tilde{g}^{(2)}(0) = 1 + (g^{(2)}(0) - 1) \int_{-\infty}^{\infty} d\tau |g^{(1)}(\tau)|^2 \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{\tau^2}{2\sigma^2}}.$$  \hspace{1cm} (6.1)

The results of these convolution procedure are shown in Fig. 6.5 (c-d) as solid lines, which agree well with the results obtained from measurement. The opposite direction was taken in the lower panels of the figure. Here the theoretical $g^{(2)}(0)$ values are shown (solid lines) together with correlation values (blue squares) that represent a deconvolution of the measured $g^{(2)}(\tau)$ and the detector response function. Details on the procedure can be found in [50]. This dual approach of assessing the photon correlations in the system theoretically and experimentally from two different directions, provides a consistent picture of the processes leading to the observed emission characteristics. It supports the interpretation that indeed a thresholdless transition to lasing was observed for one of the nanolaser devices (CNL1).

Discussion of model parameters

The choice of parameters used in the theoretical investigation of the CNL devices are either directly determined from experiment or they reflect the underlying physical properties of the

Photon correlation measurements

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Figure 6.5: Quantum-optical investigation of the CNLs. a), b) Examples of experimentally measured correlation histograms (black) for CNL1 yielding $g^{(2)}(\tau)$ at two different excitation intensities. Red lines show Gaussian fits from which raw values of $g^{(2)}(0)$ are obtained. c), d) Raw values of $g^{(2)}(0)$ as function of excitation intensity. Measured values (black squares) stay close to the Poisson limit for low pump rates due to short coherence times that prevent the resolution of photon bunching. Solid lines show the calculated values of $g^{(2)}(0)$ taking into account the coherence time and the finite detector resolution of 80 ps. e), f) $g^{(2)}(0)$ values as obtained from the quantum optical modeling (solid line) agree well with the measured values (blue dots) that were deconvoluted from the detector response function.

device geometry and the materials used. For better overview the explicit values used in the modeling of the CNLs are given in Table 6.1. The $Q$ factor as well as the wavelength of the most prominent cavity mode were determined by FDTD calculations (see Supplement of [50]). They relate to the cavity decay rate via $\kappa = \frac{\pi c}{Q \lambda}$. For both CNLs we assume relaxation rates of $\gamma_{\text{rel}} = 10 \text{ps}^{-1}$ and a constant dephasing rate $\Gamma = 5 \text{ps}^{-1}$, which corresponds to typical values found for III/V-based semiconductor gain media [83]. The role of the QWs’ area as an effective parameter was discussed in Section 5.6. Nevertheless, the values we used for the modeling of the CNLs reflect the relative size of the devices as well as their relative overlap (confinement factor) with the cavity mode. Finally, the light matter interaction strength is fixed by modeling experimentally recorded TRPL time traces as discussed in Section 5.4. In Fig. 6.6 (a-b) luminescence traces of the cavity emission are shown for the two CNLs for low excitation (blue) and high excitation (black) above the threshold, where stimulated emission quickly depletes the carrier density. For comparison, Fig. 6.6 c) shows the TRPL for the unstructured gain medium,
6.1. METAL-CLAD COAXIAL NANOLASERS

Figure 6.6: a), b) Time-resolved photoluminescence (TRPL) traces for the CNL devices after excitation with a short laser pulse. Below threshold excitation (blue) can be clearly distinguished from excitation above threshold (black) where stimulated emission quickly depletes carriers. Experimentally recorded traces are well reproduced by our calculations (orange/red dashed lines) taking into account the finite detector resolution (solid lines). Doing so allows to fix values for the light-matter interaction in the devices, which is a distinguishing feature of our theoretical investigation. c) Reference measurement of TRPL traces of the unstructured, planar QW gain medium without cavity with a slower decay on a timescale around 6 ps. As the CNLs have lifetimes \(< 0.4\) ps, this indicates a pronounced Purcell effect mediated by the cavity.

\[
\begin{array}{lcc}
\kappa & 0.634 \text{ps}^{-1} & 0.782 \text{ps}^{-1} \\
\Gamma & 5 \text{ps}^{-1} & 5 \text{ps}^{-1} \\
\gamma_{\text{rel}} & 10 \text{ps}^{-1} & 10 \text{ps}^{-1} \\
g_0 & 0.4 \text{ps}^{-1} & 0.38 \text{ps}^{-1} \\
S & 0.036 \mu\text{m}^2 & 0.04 \mu\text{m}^2 \\
\gamma_{\text{nl}} & 0.003 \text{ps}^{-1} & 0.2 \text{ps}^{-1} \\
\end{array}
\]

Table 6.1: Summary of explicit parameter values as used for modeling the CNL1 and CNL2.

i.e. the planar QW without the influence of the cavity. Although the decay due to spontaneous emission does not follow an exponential law, the planar material’s luminescence decays on a timescale of \(~ 6\) ps while the CNLs’ lifetimes are \(< 0.4\) ps, suggesting a clear Purcell effect mediated by the cavity of around 15. Calculating the TRPL traces allows to determine the appropriate rate of light-matter interaction for the modeling of the CNLs. Red/orange lines show the result of the calculation, where dashed lines are the raw results. The large stimulated emission peak above threshold can not be resolved by the limited temporal resolution of the photon detectors. Therefore, in order to obtain good agreement with the experimentally recorded traces, we convolute our calculated result with the detectors response function, assumed to be a Gaussian with 80 ps FWHM. Modeling the TRPL traces in combination with
all other laser characteristics discussed previously, confirms the consistency of our choice of parameters.

### 6.2 Room-temperature lasing from nitride based nanobeam nanolaser

The second type of nanolasers that we investigated in a cooperative, theoretical-experimental project were single-QW nanobeam lasers. The device is depicted in Fig. 6.7. Operated at room-temperature under cw excitation a clear transition to coherent emission, witnessed by the change in photon statistics, is observed. In this device a single InGaN QW is embedded in a GaN barrier material layer that is grown on a SiO$_2$ substrate. The barrier material also forms the nanobeam cavity, i.e. a type of 1D photonic crystal cavity as shown in Fig. 6.7 a). The devices where designed, fabricated, and initially characterized in the group of N. Grandjean at EPFL Lausanne. For more information on the fabrication process of the samples we refer to [15, 136, 137]. Nanobeam lasers offer a small footprint, large β factors, and have been demonstrated to operate under electrical pumping [138]. Furthermore, theoretical Q-factors in the range of $10^7$, which are obtained by cavity optimization, have been predicted for nanobeam cavities [139]. The cavity is formed by a taper region in the center of the nanobeam with decreasing hole size (numerically optimized), where a color plot of the mode’s intensity profile is given in Fig. 6.7 b). The nitride-based device material offers several beneficial properties such as high thermal robustness, chemical inertness, and biocompatibility, while the emission wavelength in the blue spectral range makes it interesting for certain optoelectronic photonic applications [136]. However, until recently, the fabrication of nitride-based nanostructures considered here was more challenging [137], underlining the novelty aspect of the discussion here.

![Figure 6.7: a) Schematic view of the free standing nanobeam laser. The InGaN QW gain medium (yellow) is embedded in GaN barrier material (blue). The cavity consists of two photonic crystal mirrors along the beam direction with a tapered region in the center forming the cavity. b) Intensity profile of the mode function’s electric field obtained from FDTD simulations. c), d) Scanning electron microscope images of typical nanobeam structures in top and side views. The scale bar in c) is equal to 100 nm in length.](image-url)
6.2. ROOM-TEMPERATURE LASING FROM NITRIDE BASED NANOBEAM
NANOLASER

Emission characteristics and correlation measurements

Emission characteristics were measured experimentally for two different sample structures that had nominally the same device geometry while only one showed lasing behavior, possibly due to differences in the manufacturing quality. Fig. 6.8 shows characteristic quantities, i.e. the output intensity, emission linewidth and wavelength. Also shown are measured values for the two-photon correlation function \( g^{(2)}(0) \) obtained from a HBT setup using single-photon detectors with temporal resolution \( \Delta t = 225 \text{ ps} \). The first nanobeam shows a close to linear I/O characteristics seen in Fig. 6.8 a). This indicates an efficient funneling of spontaneous emission into the lasing mode. A slight S-bend is visible when comparing with the linear reference line (dashed). In comparison to the linewidth reduction observed for the CNLs in the previous section, here the linewidth after an initial slight decrease, starts to increase rapidly with larger excitation power. Furthermore, a redshift of the lasing wavelength is observed. This can be attributed to thermal effects. With increasing power, heating of the nanobeam occurs, leading to the expansion of the nanobeam, which changes the emission wavelength. Additionally, jumping of the cavity resonance frequency due to thermal fluctuations on time scales smaller than the integration time in the experiment leads to the observed broadening of the linewidth. Photon-correlation measurements of the first nanobeam sample are shown in Fig. 6.8 c). A clear transition to the Poisson limit \( g^{(2)}(0) = 1 \) is visible, while for lower pump powers the coherence time of the emission becomes significant smaller than the detector resolution in the HBT setup, preventing the effect of photon bunching \( g^{(2)}(0) = 2 \) from being resolved.

The second structure (right panel in Fig. 6.8) does not show signs of lasing and serves as a reference to distinguish lasing and non-lasing characteristics from the devices. Its output intensity does not follow the characteristics S-bend and \( g^{(2)}(0) \) stays constant and close to one, indicating a low coherence time and emission in the thermal regime. Furthermore, the reference sample shows a power dependence of the intensity that is superlinear for small pump powers, indicated by a slope greater than 1 in Fig. 6.8 d). This hints towards additional loss processes corresponding to an \( A \) term in the RE theory (c.f. Eq. (2.4)). Additional losses are possibly introduced by localized defects in the gain material that are common in nitride-based material and lead to non-radiative recombination of electron-hole pairs.

Theoretical modeling

Similar to the previous section we performed a theoretical modeling of the two device samples following the same general path that was applied to modeling the CNL devices. The output intensity was calculated by use of the laser equations (5.22)–(5.24) but with the addition of a loss term accounting for the influence of non-radiative recombination at lower excitation powers [140]. The additional terms are analogue to the \( A \) term in standard REs and reduce the carrier population functions according to

\[
\frac{d}{dt} f_k^{\text{e}} \bigg|_{\text{nr}} = -A_{\text{nr}} f_k^{\text{e}}, \quad \frac{d}{dt} f_k^{\text{h}} \bigg|_{\text{nr}} = -A_{\text{nr}} f_k^{\text{h}}. \tag{6.2}
\]

In order to verify the transition of the laser emission from thermal to a Poissonian photon statistics, we calculated the \( g^{(2)}(0) \) function for both laser samples (solid lines in Fig. 6.8 c,f)) and compare the results with the measured values. In the first device that showed lasing, we observed a gradual transition to coherent emission that takes place over a range of excitation powers. It is important to note, that the predicted intracavity photon number exceeds the quantum threshold of \( n = 1 \) before \( g^{(2)}(0) \) gets close to the Poisson limit. Furthermore, in agreement with our results in Chapter 3, we observe a delayed onset of coherent emission in
In order to obtain good agreement with the measured $g^{(2)}(0)$ values, we performed the same procedure as for the results for CNL devices. We calculated the first-order correlation function for the device emission and convoluted the theoretical estimate for $g^{(2)}(\tau)$ with the detector response function according to Eq. (6.1). We find good agreement with the experimental results (dashed lines). For the non-lasing reference nanobeam, the calculated $g^{(2)}(0)$ stays close to the thermal value of 2 for all pump powers. However, the coherence time of the emission stays below the detector’s temporal resolution ($\Delta t = 225$ ps) and, as a result, the photon bunching cannot be resolved. This leads to a measured $g^{(2)}(0)$ that appears to be close to the Poisson limit, a behavior that is reproduced by the calculation as well.

Figure 6.8: Room-temperature optical and quantum-optical characterization of a lasing (left) and a reference (right), non-lasing nanobeam cavity. a), d) I/O curves. The I/O curve in a) shows a slight S-bend indicating an intensity threshold. A reference curve with slope 1 is shown as a dashed line. The characteristic in d) is governed by non-radiative losses and does not show a S-bend before saturating. b), e) Emission wavelength (green) and linewidth (FWHM, black). A slight decrease of the linewidth around $P = 5$ kW cm$^{-2}$ is followed by strong line broadening due to thermal fluctuations above 10 kW cm$^{-2}$. c), f) Second-order autocorrelation function $g^{(2)}(0)$ as obtained from experiment (red points) and theory (solid line). The results in c) (together with a)) clearly show the delayed onset of coherence discussed in Chapter 3. Good agreement with experiment is found after convolution of the calculated $g^{(2)}(\tau)$ with the detector response function (dashed lines) accounting for the temporal resolution of 225 ps.
Nanolasers have promising applications as energy efficient sources of coherent light for short- and midrange optical communication and as integral building block for photonic circuits. In the search for higher energy efficiency and further miniaturization as well as integrability, new cavity designs and gain materials are combined and investigated, from which new types of nanolasers emerge. However, characterizing the emission of experimental devices is often challenging. As we have seen, assessing the degree of coherence of a laser’s emission becomes ambiguous in the high-β regime and care must be taking regarding the proper interpretation of the laser’s threshold. We showed, that this is especially true for more recently emerging realizations of nanolasers with extended, two-dimensional gain media.

In this thesis, we contributed to the field by providing semiclassical and quantum-optical models for nanolasers by which we detected and interpreted fingerprints of the photon statistic in the laser emission. We established that high-β nanolasers with extended gain media have significantly different behavior in comparison to more established QD-based nanolasers. In particular, the delayed onset of coherent emission in those devices led to a revised notion of the laser threshold. As a concrete example of a new material class that more recently has been investigated as gain medium, we discussed monolayers of semiconducting TMDs. We presented material-realistic gain calculations in combination with a RE theory, suited for the analysis of TMD-based nanolasers.

Finally, by developing and applying a more extensive quantum-optical semiconductor laser model, we verified the gradual transition to coherent emission in high-β nanolasers with QW-based gain media. This was done in close cooperation with experimental groups. The examination of these nanolasers showed the necessity of employing more sophisticated models beyond standard rate equation approaches and the need for verification of coherent emission by measurement of the photon-correlation function $g^{(2)}(0)$ as an indicator of a changing photon statistics. Over all, the gained understanding of the interplay of spontaneous and stimulated emission in QW-gain nanolasers is crucial for the interpretation of current experiments and provides directions for future device designs in search of further improved efficiency.
Appendix A

Equation of motion and cluster expansion technique

Lasers are many-body systems that operate out of equilibrium. For small systems (e.g. a single atom laser [141]) it is feasible to calculate the dynamics of the full density matrix. This is not possible, however, for the complex interaction of particles and photons in a semiconductor laser and approximate treatments are needed. There are a number of different ways to approach many-body problems, including real-time Green functions [142], path integral formulations [143], and stochastic methods [14]. In this thesis we rely on the equation of motion (EoM) approach that gives direct access to the dynamics of physically relevant quantities. Furthermore, it allows to investigate correlations explicitly up to arbitrary order, which we use extensively for the calculation of photon-correlation functions. In the EoM technique, the inherent complexity of many-body interactions manifest themselves in the form of the arising hierarchy problem [144]. It is characterized by an infinite hierarchy of equations that couple expectation values with increasing number of particles to each other. We apply the cluster expansion technique in order to treat correlations in clusters of up to \( N \) interacting particles and to truncate the system of equations in a consistent way.

The EoM that we use to describe the laser dynamics are formulated for expectation values of operator products involving \( n \) bosonic and \( m \) fermionic particles. They can generally be written as

\[
\langle N \rangle = \langle b_1 \ldots b_n a_1^{\dagger} \ldots a_m^{\dagger} a_m \ldots a_1 \rangle,
\]

where \( N = n + m \) is the total particle number of the interacting cluster and by \( \langle N \rangle \) we denote the generalized expectation values of \( N \) particles with arbitrary combinations of fermions and bosons. Note, that formally 2 fermionic operators \( a_i^{\dagger} a_j \) correspond to a single particle as do single bosonic operators \( b_i \). This correspondence will become more clear when looking at the coupling among the operators via the light-matter interaction [144]. The dynamics of expectation values are derived from the Heisenberg equation with Lindblad terms for open quantum systems. The interaction Hamiltonians that we encountered in Eqs. (3.3) and (5.6) are coupling expectation values of \( N \) particles to those with \( N + 1 \) particles according to

\[
\left. \frac{d}{dt} \langle N \rangle \right|_{H_i} = \langle N \rangle + \langle N + 1 \rangle.
\]

Recursively, this gives rise to the infinite hierarchy of equations that must be truncated in a systematic way in order to obtain a closed set of equations. This is achieved by the cluster expansion technique.
The concept of coupled clusters was initially developed in the context of many-body systems in nuclear physics and was later adapted to become a powerful tool for quantum chemistry. For solid state theory and semiconductor optics the techniques can also be applied [144, 145, 146], in order to reduce the complexity of the many-body problem. The basic idea is to expand any given expectation value \( \langle N \rangle \) as a sum of factorizations involving all possible combinations of \( N - 1 \) particle clusters and the pure correlations among these particles \( \delta \langle N \rangle \). Explicitly, the factorizations for the lowest order clusters are

\[
\begin{align*}
(1) &= \delta(1) \\
(2) &= \langle 1 \rangle \langle 1 \rangle + \delta(2) \\
(3) &= \langle 1 \rangle \langle 1 \rangle \langle 1 \rangle + \langle 1 \rangle \delta(2) + \delta(3) \\
(4) &= \langle 1 \rangle \langle 1 \rangle \langle 1 \rangle \langle 1 \rangle + \langle 1 \rangle \langle 1 \rangle \delta(2) + \delta(2) \delta(2) + \langle 1 \rangle \delta(3) + \delta(4)
\end{align*}
\]

(A.3)

In these expansion scheme, terms are denoted according to the number of the involved particles as singlets, doublets, triplets, quadruplets, etc. A graphical representation of the factorization for \( N = 3 \) is given in Fig. A.1. Distinguished are the full expectation values \( \langle N \rangle \) from their pure correlations denoted as \( \delta \langle N \rangle \). Only for singlets, where no further factorization is possible, the expectation value coincides with the correlations. Furthermore, when summing over all possible combinations of single particle factorizations the anticommuting property of the fermionic operators must be observed by introducing appropriate sign changes due to commuting of fermionic operators.

Figure A.1: Graphical representation of the factorization of a three particle expectation value \( \langle N = 3 \rangle \). Boxes indicate increasing orders of possible approximation (singlet, doublet, etc.) by neglecting all subsequent terms. The pictographic representations account for the general structure of the clusters after factorization. When factorizing explicit terms, all possible combination of factorization must be considered as well as sign changes due to commuting of fermionic operators. As an example consider the three particle expectation value \( \langle b a_1^\dagger a_2^\dagger a_2 a_1 \rangle \). According to the cluster expansion scheme...
given in A.3 it must be factorized as

\[
\langle ba_1^\dagger a_2^\dagger a_2 a_1 \rangle = \langle b \rangle \left( \langle a_1^\dagger a_1 \rangle \langle a_2^\dagger a_2 \rangle - \langle a_1^\dagger a_2 \rangle \langle a_2^\dagger a_1 \rangle \right) \\
+ \left( \langle a_1^\dagger a_1 \rangle \delta \langle ba_2^\dagger a_2 \rangle - \langle a_1^\dagger a_2 \rangle \delta \langle ba_2^\dagger a_1 \rangle \right) + (1 \leftrightarrow 2) \\
+ \langle b \rangle \delta \langle a_1^\dagger a_2 a_2 a_1 \rangle \\
+ \delta \langle ba_1^\dagger a_2^\dagger a_2 a_1 \rangle .
\]

(A.4)

The first line of the right hand side of A.4 corresponds to the singlet factorization with all possible combinations of single particle quantities corresponding to operators from the original expression. Neglecting all other terms in equation A.4 would correspond to an approximation on the lowest (singlet) level of the EoM hierarchy. In terms of the fermionic (electron) operators the singlet factorization of two-particle correlations is equivalent to the Hartree-Fock approximation known from atomic and solid state physics, where the even terms are the Hartree- and the odd terms the Fock-contributions. The terms with the minus sign require an odd permutation of the fermionic operators prior to factorization and reflect their anticommuting relation as exemplified by

\[
\langle a_1^\dagger a_2^\dagger a_2 a_1 \rangle \approx \langle a_1^\dagger a_1 \rangle \langle a_2^\dagger a_2 \rangle - \langle a_1^\dagger a_2 \rangle \langle a_2^\dagger a_1 \rangle .
\]

For larger number of Fermions, a generalization of the factorization scheme must be applied [144]. The lines 2 and 3 in A.4 involve two-particle correlations that are given by, e.g.

\[
\delta \langle ba_1^\dagger a_j \rangle = \langle ba_1^\dagger a_j \rangle - \langle b \rangle \langle a_1^\dagger a_j \rangle .
\]

(A.5)

The last line are the pure triplet correlations \( \delta \langle 3 \rangle \). On the doublet level approximation these triplet correlations are neglected.

It is important to discuss the limitations of the cluster expansion approach. For strongly correlated systems, neglecting correlations at any order will introduce significant errors and the approach is not valid. For initially uncorrelated systems however, correlations of increasing order will build up successively and for small times the cluster expansion approach gives correct results. This can be understood when realizing that the cluster expansion effectively corresponds to an expansion of the dynamics in orders of \( V \Delta t \) where \( V \) describes the interaction of the subsystems and \( \Delta t \) is the time interval of the dynamics. In this thesis we mainly consider the stationary states of laser dynamics, where dissipation terms dampen out higher-order correlations. To assure that this is indeed the case, the system that we simulate has to operates in the regime of weak coupling, i.e. rates of dissipation must be sufficiently large in comparison to the interaction strength.
Appendix B

N-emitter laser model: derivations

B.1 Intensity threshold for a N emitter laser model

In Eqs. (3.9) we derived rate equations for the N emitter laser model studied in Chapter 3. Here we derive an analytic expression for the intensity threshold that corresponds to the center of the threshold jump in the output intensity as would be read off e.g. from Fig. 3.2. In the thermodynamic limit, the intensity threshold corresponds to the pump power at which a phase transition in the system occurs. The corresponding order parameter is the photon density in the cavity, which changes from zero to a finite value. For finite systems, away from the thermodynamic limit, this transition is reflected in a change of scaling behavior of the photon number with the system size as mentioned in Chapter 3.

We start by considering the stationary solutions to Eqs. (3.9)

\[ R_n(2f - 1) + R_f = - \gamma f + P(1 - f), \]  
\[ N[R_n(2f - 1) + R_f] = \kappa n. \]  

(B.1)

These equations describe balance conditions for the stationary state. Their left-hand sides contain the net photon generation rate from a single emitters in Eq. (B.1a) and of all N emitters collectively in Eq. (B.1b). It is made up of spontaneous (\( \propto f \)) and stimulated emission (\( \propto f_n \)), as well as absorption (\( \propto (1 - f)n \)). In Eq. (B.1a) the rate of photon generation is balanced against the rate of the emitter excitation, taking into account both the pumping P and the radiative losses \( \gamma \), while in Eq. (B.1b) the photons generated by all emitters compensate the cavity losses at rate \( \kappa \). Combining these equation we obtain

\[ f_0 - f = \frac{\kappa n}{N(P + \gamma)}, \quad \text{with} \quad f_0 = \frac{P}{P + \gamma}. \]  

(B.2)

In the absence of the Jaynes-Cummings interaction (\( R = 0 \)), \( f_0 \) is the steady-state upper level population. Similarly, Eq. (B.1b) can be rewritten as

\[ f_L - f = \frac{f}{2n}, \quad \text{with} \quad f_L = \frac{\kappa}{2RN} + \frac{1}{2}. \]  

(B.3)

where \( f_L \) is the population for which the gain (stimulated emission minus absorption) exactly compensates the cavity losses. From the last two equations, we find that both \( f_0 \) and \( f_L \) are upper bounds for the true steady-state solution, i.e. \( f \leq f_0, f_L \). We multiply Eqs. (B.2) and (B.3) and obtain a quadratic equation for \( f \)

\[ (f_L - f)(f_0 - f) = \frac{\kappa f}{2(P + \gamma)N}, \]  

(B.4)
whose lower solution turns out to be the physical one. At the same time, the photon number is given by

\[ n = \frac{N(P + \gamma)}{\kappa} (f_0 - f). \]  

(B.5)

These results solve the problem of steady-state populations for the carriers and for the photons at the rate-equation level. However, we can obtain more insight by analyzing the results further. First we observe that (B.4) defines an anti-crossing condition. In the thermodynamic limit \( (N \to \infty) \) the RHS of (B.4) vanishes and, as the pump increases, the solution changes abruptly from \( f = f_0 \) to \( f = f_L \), always following the smaller. The discontinuous change of \( f \) going from one to the other solution indicates the phase-transition-like behavior. This can also be seen from (B.5) where the photon density \( n/N \) changes from 0 to a finite value. For \( f = f_0 \) the energy that is provided by the pump is only spent for exciting the emitters in the cavity, until \( f \) reaches the value \( f_L \) with sufficient inversion to compensate the photon losses. After that, stimulated emission sets in and all excess energy is converted into photons, leading to a macroscopic photon density in the cavity.

For finite values of \( N \) the transition is no longer abrupt. With the RHS of Eq. (B.4) being nonzero, a smooth change (anticrossing) from \( f \approx f_0 \) to \( f \approx f_L \) takes place. The lasing transition becomes gradual, without a well-defined threshold. Now a transition interval around \( P_{th} \) still separates two contrasting behaviors: Above it the photon number grows with system size \( N \), while below it photon numbers stay finite. Even away from the thermodynamic limit we can define the intensity threshold as the pump power that separates the regimes of different scaling behavior. It is given by the crossover point \( f_0 = f_L \), from which we obtain an implicit expression for the threshold pump power satisfying the equation

\[ P_{th} - \gamma - (P_{th} + \gamma) \frac{\kappa}{RN} = 0, \]

(B.6)
as already given in (3.12).

### B.2 Derivation of the rate equations

Using the Heisenberg equation with additional Lindblad terms given in (3.5), we derive here EoM for the \( N \) emitter laser model up to the quadruplet level of the cluster expansion (c.f. Appendix A). This allows us to access two-photon correlations as mentioned in Section 3.3, given by

\[ g^{(2)}(0) = 2 - \frac{\delta (b^\dagger b^\dagger b b)}{(b^\dagger b)^2}. \]

(B.7)

The quadruplet level introduces a set of new expectation values besides the photon number \( n = \langle b^\dagger b \rangle \) and populations \( f = \langle c_i^\dagger c_i \rangle \). The following notations are used below:

\[ c_m = \langle b^\dagger b^m c_i^\dagger c_i \rangle, \quad v_m = \langle b^\dagger b^m v_i^\dagger v_i \rangle, \quad m = 0, 1, 2, \]

(B.8)

\[ p_m = \langle b^\dagger b^m \rangle, \quad \psi_m = -ig \langle b^\dagger b^m v_i^\dagger c_i \rangle, \quad m = 1, 2. \]

(B.9)

We note that with a single carrier in each emitter these averages are not independent since \( c_m + v_m = p_m \), but we keep the notation for clarity. Also \( p_1 \) is the average photon number \( n \), \( c_0 = f \) and \( \psi_1 \) is the same as in Section 3.2. We denote pure correlation (i.e. expectation values minus their factorization) with a \( \delta \), e.g. \( \delta c_1 = c_1 - nf \).

With the notation set up, we generate equations of motion for the dynamics of operator averages from the Hamiltonian (3.3) up to the quadruplet level. Singlet photon expectation
values \( \langle b^{(1)} \rangle \) are zero as they are coupled to coherent polarizations \( \langle v^i_c \rangle \) of the emitters which are not driven by the incoherent system excitation considered here. As a result, we find that

\[
\delta \psi_1 = -ig \delta \langle b^1 v^i_c \rangle = -ig(\langle b^1 v^i_c \rangle - \langle d^1 \rangle \langle v^i_c \rangle) = \psi_1. \tag{B.10}
\]

The smallest order expectation values that are nonzero are the photon number \( n \) and emitter excitation \( f \). The corresponding EoM on doublet level are already given in the main text as

\[
\dot{n} = -\kappa n + 2N \text{Re} \psi_1, \tag{B.11a}
\]
\[
\dot{f} = -\gamma f + P(1 - f) - 2 \text{Re} \psi_1, \tag{B.11b}
\]

and equally for the polarization \( \psi_1 \)

\[
\dot{\psi}_1 = g \left( c_1 - v_2 + \langle c^i_1 v^i_c \rangle + \sum_{i \neq j} \langle c^i_1 v^i_j v^i_c \rangle \right) - \frac{1}{2} \Gamma \psi_1. \tag{B.12}
\]

The on-site carrier-carrier correlations \( \langle c^i_1 v^i_v v^i_c \rangle \) reduce to \( f \) due to the single electron assumption while the correlations among emitters are given in the sum in Eq. (B.12) and factorize to

\[
\langle c^i_1 v^i_v v^i_c \rangle = \langle c^i_1 v^i_c \rangle \langle v^i_v \rangle - \langle c^i_1 \rangle \langle v^i_v \rangle \delta \langle v^i_v \rangle + \delta \langle c^i_1 v^i_v v^i_c \rangle. \tag{B.13}
\]

The first term corresponds to on-site polarizations which are not driven in our system. The second term corresponds to quantities involving different sites which we assume to be less correlated that on-site ones and are thus neglected. The pure correlations \( \delta \langle c^i_1 v_i^i v^i_c \rangle \) among emitters are relevant if we are interested in superradiant effects [82] but they are here omitted, too. As a result the sum in Eq. (B.12) is zero. The carrier-photon correlations \( c_1 \) and \( v_1 \) are factorized according to Eq. (3.8) leading to the EoM for \( \psi_1 \)

\[
\dot{\psi}_1 = |g|^2 (2f^2 - 1 + f + \delta c_1 - \delta v_2) - \frac{1}{2} \psi_1, \tag{B.14}
\]

with \( \Gamma = P + \gamma + \kappa \). We follow this scheme to derive EoM for the correlation functions up to the quadruplet level by neglecting all pure correlation functions of five or more particles

\[
\delta c_1 = \frac{d}{dt}(c_1 - nf) = -2 [\delta \psi_2 + (n + f)\psi_1] - \Gamma \delta c_1, \tag{B.15a}
\]
\[
\delta v_1 = \frac{d}{dt}(v_1 - n(1 - f)) = -2 [\delta \psi_2 + (n + f)\psi_1] - \Gamma \delta v_1 = -\delta \psi_1, \tag{B.15b}
\]
\[
\delta \psi_2 = \frac{d}{dt}(\psi_2 - 2n) = |g|^2 [\delta p_2(2f - 1) + (2n + 1)(\delta c_1 - \delta v_1)] - 4\psi_2^2 - \frac{1}{2} \Gamma' \delta \psi_2, \tag{B.15c}
\]
\[
\delta p_2 = \frac{d}{dt}(p_2 - 2n^2) = 4\delta \psi_2 - 2\kappa \delta p_2, \tag{B.15d}
\]

with \( \Gamma' = P + \gamma + 3\kappa \). We note that these EoM are very similar to the those in Section 5.5 for the dynamics of quadruplet correlations. However, small difference appear. Due to the single electron approximation applied here, upper and lower level of any emitter can not be occupied by an electron at the same time, what results in different factorizations for certain expectation values. Also the consistent description of pump and radiative losses by Lindblad terms in the EoM introduces additional dephasing of the correlation functions.

The EoM for \( n, f \) and \( \psi_1 \) as well as the various pure correlations form a closed system of coupled equations. In the steady state all time derivatives vanish and the system becomes algebraic. From this one obtains an expression of \( \delta p_2 \) and thus of \( g^{(2)}(0) \), by eliminating all the other unknowns, in terms of \( c_0 = f \) and \( p_1 = n \) only as given in Eq. (3.14) in the main text.
Appendix C

Rate equations for TMD-based nanolasers

In Chapter 4 we discuss a laser model for nanolasers with single layers of TMD materials as gain medium. The rate equations that we present there are specifically tailored to account for the particular device geometry of these devices and allow to take results from our microscopic material gain calculations as an input. In the next section we derive a modal gain expression for 2D layers of gain medium interacting with a cavity mode in a semiclassical approach. We proceed along the lines of [79, 115]. In Section C.2 we outline the derivation for the RE theory used in Chapter 4 while in Section C.3 we provide the fit function to the microscopic material gain that served as input to the rate equations.

C.1 Material gain and frequency shift

We consider a cavity that sustains a spatially and spectrally localized cavity mode and a layer of 2D gain material embedded in it (c.f. Fig. 4.4). First we consider the bare cavity without the gain material and derive a relation for its cavity mode, which is the solution to a generalized Helmholtz equation. The wave equation for the electric field $E(r,t)$ of the cavity mode is derived from the macroscopic Maxwell equations and is given as

$$\nabla \times \nabla \times E(r,t) + \mu_0 \epsilon_0 \frac{\partial^2 E(r,t)}{\partial t^2} = -\mu_0 \frac{\partial^2 \tilde{P}(r,t)}{\partial t^2},$$

(C.1)

where $\tilde{P}(r,t) = \epsilon_0 \chi_b(r) E(r,t)$ is the polarization in the bare cavity without the gain medium and $\chi_b(r)$ is the bare cavity susceptibility. The electric field and polarization are assumed to have solutions with a harmonic time dependence but stationary spatial dependence and amplitude

$$E(r,t) = E_0 u(r) e^{-i\omega_0 t},$$

(C.2)

$$\tilde{P}(r,t) = \tilde{P}_0(r) u(r) e^{-i\omega_0 t},$$

(C.3)

again with $\tilde{P}_0(r) = \epsilon_0 \chi_b(r) E_0$. Inserting Eqs. (C.2) and (C.3) into (C.1) yields

$$E_0 \nabla \times \nabla \times u(r) e^{-i\omega_0 t} = (\mu_0 \epsilon_0 \omega_0^2 E_0 + \mu_0 \omega_0^2 \tilde{P}_0(r)) u(r) e^{-i\omega_0 t}$$

$$\Rightarrow \nabla \times \nabla \times u(r) = \mu_0 \epsilon_0 \omega_0^2 (1 + \chi_b(r)) u(r).$$

(C.4)

The mode function $u(r)$ is normalized to one as

$$\frac{1}{V_m} \int \epsilon(r) |u(r)|^2 d^3 r = 1,$$

(C.5)
by definition of the effective mode volume $V_m$

$$V_m = \frac{\int \epsilon(r)|E(r)|^2 d^3r}{\max[\epsilon(r)|E(r)|^2]}.$$  \hspace{1cm} (C.6)

Now, in a second step, we include the gain material assuming that the spatial dependence of the electric field remains unchanged due to the small width of the 2D layer. We write

$$\nabla \times \nabla \times E(r, t) + \mu_0 \epsilon_0 \frac{\partial^2 E(r, t)}{\partial t^2} = -\mu_0 \frac{\partial^2 P(r, t)}{\partial t^2},$$  \hspace{1cm} (C.7)

and

$$E(r, t) = E(t)u(r)e^{-i\omega_0 t},$$  \hspace{1cm} (C.8)

$$P(r, t) = P(r, t)u(r)e^{-i\omega_0 t}.$$  \hspace{1cm} (C.9)

Here, $P(r, t) = \epsilon_0 \chi(r)E(r, t)$ is the polarization of the cavity material plus the gain material layer ($\chi(r) = \chi_b(r)I + \chi_g(r)$). The underscore indicates that $\chi(r)$ is a tensor in general. Furthermore, the amplitudes $E$ and $P$ are now time dependent as well. Inserting the expressions for $E$ and $P$ results in

$$\nabla \times \nabla \times u(r)E(t) + \mu_0 \epsilon_0 \left( \frac{\partial^2 E(t)}{\partial t^2} - 2i\omega_0 \frac{\partial E(t)}{\partial t} - \omega_0^2 E(t) \right) u(r) = -\mu_0 \left( \frac{\partial^2 P(r, t)}{\partial t^2} - 2i\omega_0 \frac{\partial P(r, t)}{\partial t} - \omega_0^2 P(r, t) \right) u(r).$$  \hspace{1cm} (C.10)

We assume that the spatial dependence $u(r)$ is the same as for the bare cavity as a TMD-material layer of atomic thickness will have a negligible effect. Therefore we use Eq. (C.4) in order to eliminate $\nabla \times \nabla \times u(r)$ and write $P_b(r, t) = \epsilon_0 \chi_b(r)E(t)$ for the polarization amplitude due to the cavity background. We get

$$\mu_0 \epsilon_0 \omega_0^2 \left( E(t) + \frac{P_b(r, t)}{\epsilon_0} \right) u(r) + \mu_0 \epsilon_0 \left( \frac{\partial^2 E(t)}{\partial t^2} - 2i\omega_0 \frac{\partial E(t)}{\partial t} - \omega_0^2 E(t) \right) u(r) = -\mu_0 \left( \frac{\partial^2 P(r, t)}{\partial t^2} - 2i\omega_0 \frac{\partial P(r, t)}{\partial t} - \omega_0^2 P(r, t) \right) u(r)$$

$$\Rightarrow \mu_0 \epsilon_0 \left( \frac{\partial^2 E(t)}{\partial t^2} - 2i\omega_0 \frac{\partial E(t)}{\partial t} - \omega_0^2 E(t) \right) u(r) = -\mu_0 \left( \frac{\partial^2 P(r, t)}{\partial t^2} - 2i\omega_0 \frac{\partial P(r, t)}{\partial t} - \omega_0^2 \left( P(r, t) - P_b(r, t) \right) \right) u(r).$$  \hspace{1cm} (C.11)

The slowly varying envelope approximation (SVEA) assumes that the amplitudes of the fields vary slowly in comparison to the harmonic time dependence of the fields. It is expressed as

$$\frac{\partial E(t)}{\partial t} \ll \omega_0 E(t),$$  \hspace{1cm} (C.13)

$$\frac{\partial P(r, t)}{\partial t} \ll \omega_0 P(r, t).$$  \hspace{1cm} (C.14)

We only keep the leading term on both sides of Eq. (C.12) and observe that on the right hand side the influence of the bare cavity susceptibility $\chi_b(r)$ is canceled in the equation

$$-2i\epsilon_0 \omega_0 \frac{\partial E(t)}{\partial t} u(r) = \omega_0^2 \left( P(r, t) - P_b(r, t) \right) u(r)$$

$$\Rightarrow 2i\frac{\partial E(t)}{\partial t} u(r) = -\omega_0 \chi_g(r)E(t)u(r).$$  \hspace{1cm} (C.15)
C.1. MATERIAL GAIN AND FREQUENCY SHIFT

Now we write \( E(t) = \ddot{E}(t) e^{i \phi(t)} \) as a real amplitude times a phase while the SVEA is still valid. We separate real and imaginary part of the equation and obtain

\[
\left( 2i \frac{\partial \ddot{E}(t)}{\partial t} - 2 \dot{E}(t) \frac{\partial \phi(t)}{\partial t} \right) u(r) = -\omega_0 \left( \chi''(r) + i \chi'''(r) \right) \ddot{E}(t) u(r) \tag{C.16}
\]

\[
\Rightarrow \quad \frac{\partial \ddot{E}(t)}{\partial t} u(r) = -\frac{1}{2} \omega_0 \chi''(r) \ddot{E}(t) u(r) \tag{C.17}
\]

\[
\frac{\partial \phi(t)}{\partial t} u(r) = \frac{1}{2} \omega_0 \chi'(r) u(r). \tag{C.18}
\]

We multiply Eq. (C.17) by \( \dddot{E}^*(t) u^*(r) \) from the left and add the complex conjugate of both sides (omitting the tilde on the \( E \)):

\[
\Rightarrow \quad \frac{\partial}{\partial t} |E(t)|^2 |u_1(r)|^2 = -\omega_0 \chi''(r) |E(t)|^2 |u_1(r)|^2. \tag{C.19}
\]

In the last step we used the fact that \( \chi''(r) \) is a 2D susceptibility, i.e. polarizations in the 2D monolayer are only excited along the in-plane directions and as a tensor \( \chi''(r) \) has the form

\[
\chi''(r) = \chi''_i(r) \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{C.20}
\]

In the last step we integrate both sides in (C.19) over \( r \). For that we write the spatial dependence of \( \chi_i(r) \) as

\[
\chi_i(r) = d_i \delta(z - z_i) \chi_i(\omega_0), \tag{C.21}
\]

where \( z_i \) is the position of the layer along the \( z \)-axis and \( d_i \) is the width of the monolayer which acts as a characteristic length scale of the system. Furthermore, we add a cavity decay term \(-2\kappa |E(t)|^2 \) with \( \kappa = \omega_0/(2Q) \), where \( Q \) is the quality factor of the cavity mode. We obtain

\[
\frac{\partial}{\partial t} |E(t)|^2 = -\Gamma \omega_0 \chi''(\omega_0) |E(t)|^2 - 2\kappa |E(t)|^2, \tag{C.22}
\]

with \( \Gamma = \frac{d_i}{\hbar} \int \epsilon(\mathbf{r}) |u_1(\mathbf{r}, z = z_i)|^2 d^2 r \) being the \emph{unitless} confinement factor due to the small extend of the gain material relative to the cavity mode. It is equivalent to the confinement factor introduced in Eq. (4.1)

\[
\Gamma = \frac{\int_{\text{Layer}} \epsilon(\mathbf{r}) |E_{||}(\mathbf{r})|^2 d^3 r}{\int_V \epsilon(\mathbf{r}) |E(\mathbf{r})|^2 d^3 r}, \tag{C.23}
\]

where \( E_{||}(\mathbf{r}) \) is the in-plane electric field in the 2D-layer which is assumed constant along the width (\( z \)-direction) of the layer. The minus sign in front of the stimulated emission term reflects the fact that positive gain is achieved when \( \chi''(\omega_0) \) becomes negative. The photon number density \( N_p \) that is used in standard rate equations is proportional to the electric field intensity, i.e. \( N_p(t) \sim |E(t)|^2 \), resulting in the equation

\[
\frac{\partial}{\partial t} N_p = \Gamma G(N) N_p - 2\kappa N_p, \tag{C.24}
\]
where \( G(N) = -\omega_0 \chi''_l(\omega_0, N) \) defines the material gain (as a function of carrier density) as discussed in Chapter 4. From this relation of the photon density we derive rate equations for TMD-based nanolasers in the next section.

Going back to the equation for \( \frac{d\phi(t)}{dt} \) in (C.18), we see that the change in phase is constant in time and establishes a frequency shift that is proportional to the real part of \( \chi_l(r) \). The shift can be attributed to a change in refractive index, induced by the excitation of the gain medium. Hence, \( \frac{d\phi(t)}{dt} = \Delta\omega_0 = \Gamma \frac{\omega_0}{2} \chi_l(r) \).

Here we do not include this effect in our laser theory as we are considering single mode lasers and assume an optimal spectral position of the cavity mode in the steady state operation of the device (c.f. Section 4.3 on material specific criteria for lasing operation).

### C.2 Rate equations tailored to device geometry

We start from the rate equations for the number of excited carriers in the 2D layer and photon number in the cavity analogue to what is found in [39]

\[
S \frac{d}{dt} N = PS - (R_{sp} + R_{nl})V - R_{st}V, \quad (C.26)
\]

\[
V_p \frac{d}{dt} N_p = R_{st}V + \beta R_{sp}V - \frac{N_p V_p}{\tau_p}, \quad (C.27)
\]

where \( N \) and \( N_p \) are carrier and photon densities related to the monolayer area \( S \) and mode volume \( V_p \), respectively. \( R_{sp}, R_{nl} \) and \( R_{st} \) are the rates of spontaneous, non-radiative and stimulated emission, related to the volume of the 2D gain medium \( V = S \times d \). \( P \) is the rate of carrier generation per volume and \( \tau_p \) is the cavity lifetime. We divide both equations by the respective area and volume and write \( 1/\tau_p = 2\kappa \) to obtain

\[
\frac{d}{dt} N = P - (R_{sp} + R_{nl})d \frac{d}{dt} d_1 - R_{st}d_1, \quad (C.28)
\]

\[
\frac{d}{dt} N_p = R_{st} \frac{V}{V_p} + \beta R_{sp} \frac{V}{V_p} - 2\kappa N_p. \quad (C.29)
\]

From Eq. (C.24) in the previous section we identify the stimulated emission term in the equation for \( N_p \) as \( R_{st} \frac{V}{V_p} = -\Gamma \omega_0 \chi''_l(\omega_0, N) N_p = \Gamma G(N) N_p \). Furthermore we set for the spontaneous emission term \( R_{sp} \frac{V}{V_p} = BN^2 \) with appropriate dimensions for \( B \). The non-radiative losses are given by the standard RE expressions \( R_{nl} = AN + CN^3 \). As a result, the rate equation for the carrier density \( N \) and photon density \( N_p \) have the form that we presented in Eqs. (4.6)

\[
\frac{d}{dt} N = P - \frac{V}{S} \Gamma G(N) N_p - \frac{V_p}{S} (AN + BN^2 + CN^3), \quad (C.30)
\]

\[
\frac{d}{dt} N_p = \Gamma G(N) N_p + \beta BN^2 - 2\kappa N_p. \quad (C.31)
\]

Again, the ratio \( V_p/S \) appearing in the equations reflects the reference volume/area that the carrier and photon densities are related to and reconcile the dimensions accordingly. These can always be absorbed into the other coefficients, which rescales the equations. The confinement factor \( \Gamma \), on the other hand, has a more concrete meaning as it quantifies the spatial overlap of cavity mode and gain medium. It directly influences the device efficiency as it enters the modal gain.
C.3  Fit model for material gain

In Section 4.3 we use a parametrization for the microscopically calculated material gain as input to the presented rate equation theory. The purpose of this parametrization is to provide simple means of using the results of the microscopic gain calculations in the rate-equation approach. This makes our model accessible for other groups to use, rather than to rely on linear or logarithmic gain models that fail to capture the material properties, especially at elevated carrier densities. A particular problem of using such simpler gain models in rate equations is that lasing can always be achieved if the pumping is sufficiently strong, as the only process that limits the increase of gain with increasing carrier density are Auger losses via the $C$ coefficient.

The behavior of the microscopically calculated gain, as shown in Fig. 4.6, is well reproduced by the fit formula

$$G(N) = \omega_0 \left( a(N - N_0)e^{-2b(N + N_1)(N - N_0) - b(N - N_0)^2} \right).$$

for the maximum gain (at optimal mode position) as a function of carrier density. Eq. (C.32) is based on a linear fit model, where $a$ and $N_0$ correspond to the differential gain at low densities and the transparency density. The linear gain is modified by a shifted Gaussian to account for the gain rollover that we have identified as a characteristic for the TMD-monolayer materials. The mode eigenfrequencies enter the material gain together with further parameters that are given in Table C.1. The carrier density $N$ in Eq. (C.32) has units 1/nm$^2$ while the parameters $a$, $N_0$, $b$ and $N_1$ are assumed to have units consistent with $G(N)$ having units of 1/ps. The gain fit is valid for the density ranges $N \in [0, 1.1]$ nm$^{-2}$ (MoS$_2$) and $N \in [0, 0.7]$ nm$^{-2}$ (WS$_2$ and WSe$_2$).

<table>
<thead>
<tr>
<th></th>
<th>$\omega_0$ (ps$^{-1}$)</th>
<th>$a$</th>
<th>$N_0$</th>
<th>$b$</th>
<th>$N_1$</th>
</tr>
</thead>
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<tr>
<td>MoS$_2$</td>
<td>2749</td>
<td>2.025</td>
<td>0.5</td>
<td>2.6</td>
<td>0</td>
</tr>
<tr>
<td>WS$_2$</td>
<td>3001</td>
<td>5.147</td>
<td>0.232</td>
<td>0.85</td>
<td>1.95</td>
</tr>
<tr>
<td>WSe$_2$</td>
<td>2457</td>
<td>2.415</td>
<td>0.25</td>
<td>0.23</td>
<td>9.92</td>
</tr>
</tbody>
</table>

Table C.1: Parameters for the gain fit in Eq. (C.32).
Appendix D

Microscopic light-matter Hamiltonian

Here we derive the explicit form of the light-matter interaction Hamiltonian given in Eq. (5.6) for the semiconductor nanolaser model that we introduce in Chapter 5. The derivation is done along the lines of [117] but we adapt the derivation to the case where we describe the interaction of a single cavity mode with a 2D gain medium of unit cell thickness. We will see that the general form of the resulting Hamiltonian remains unchanged and only the light-matter interaction strength is modified. Thus the derivation is equally applicable to nanolasers with more conventional QW gain media as discussed in Section 6 as well as novel TMD materials with atomic thickness.

D.1 Basic formulation of the Hamiltonian

We start with the full Hamiltonian in the minimal coupling picture [144],

\[
H = \sum_{j=1}^{N} \frac{1}{2m_0} [p_j - eA(r_j)]^2 + U(r_j) + \hbar \omega \left( b^\dagger b + \frac{1}{2} \right) + \sum_{i<j}^{N} V(|r_j - r_i|). \tag{D.1}
\]

The first two terms describe the motion of \(N\) electrons in the periodic crystal potential \(U\), interacting with the vector potential \(A\) of the single cavity mode. We write \(A\) in its quantized form as

\[
A(r) = \frac{E_{ph}}{\omega} \left[ u(r)b + u^*(r)b^\dagger \right], \tag{D.2}
\]

where \(E_{ph} = \sqrt{\frac{\hbar \omega}{2\epsilon_0 V_m}}\) is the cavity field amplitude per photon, \(\hbar \omega\) and \(V_m\) are the cavity mode energy and volume. The eigenmode function is given by \(u(r)\), while \(b^\dagger\) and \(b\) are creation and destruction operators for photons in the cavity mode. The third term in (D.1) is the cavity mode Hamiltonian and the last term accounts for Coulomb interactions, which we neglect at the moment.

Before we perform the 2nd quantization of the charge carriers, we note that expanding the square of the generalized momentum \([p - eA(r)]^2\) will result in a light-matter interaction of the form \(p \cdot A\) and an additional term proportional to \(A^2\). To avoid that, we transform the
Hamiltonian according to the Goeppert-Mayer transformation

\[ H' = e^S H e^{-S}, \quad \text{with} \quad S = -\frac{i}{\hbar} \sum_{j=1}^{N} e \mathbf{r}_j \cdot \mathbf{A}(\mathbf{r}_j). \]  

\[ (D.3) \]

For the transformation of an operator \( O \) we use the expansion

\[ O' = e^S O e^{-S} = O + [S, O]_+ + \frac{1}{2} [S, [S, O]]_+ + \ldots \]  

\[ (D.4) \]

and find for the individual operators

\[ \mathbf{r}_j' = \mathbf{r}_j, \quad b' = b + \frac{i}{\hbar} \sum_{j=1}^{N} e E_{\text{ph}} \mathbf{r}_j \cdot \mathbf{u}^*(\mathbf{r}_j), \]  

\[ \mathbf{p}_j' = \mathbf{p}_j + e (\mathbf{A}(\mathbf{r}_j) + (\mathbf{r}_j \cdot \nabla) \mathbf{A}(\mathbf{r}_j) + \mathbf{r}_j \times (\nabla \mathbf{r}_j \times \mathbf{A}(\mathbf{r}_j))) + \ldots \]  

\[ (D.5) \]

Noting the complicating form of the transformed momentum \( \mathbf{p}_j' \) we use the fact that the wavelength of the cavity mode is much larger than the typical length scales relevant for the electron dynamics. We therefore neglect terms containing spatial derivatives of the vector potential as it will vary only slowly with position. This is a form of the dipole approximation. The transformed momentum then simplifies to

\[ \mathbf{p}_j' = \mathbf{p}_j + e \mathbf{A}(\mathbf{r}_j). \]  

\[ (D.6) \]

We insert all transformed operators in Eq. (D.1) and obtain

\[ H = \sum_{j=1}^{N} \frac{\mathbf{p}_j^2}{2m_0} + U(\mathbf{r}_j) + \hbar \omega \left( b^+ b + \frac{1}{2} \right) \]

\[ - \frac{e}{\epsilon_0} \sum_{j=1}^{N} \mathbf{r}_j \cdot \mathbf{D}(\mathbf{r}_j) + \sum_{i,j}^{e^2/2\epsilon_0 V_m} [\mathbf{r}_i \cdot \mathbf{u}^*(\mathbf{r}_i)] [\mathbf{r}_j \cdot \mathbf{u}(\mathbf{r}_j)]. \]  

\[ (D.7) \]

The dependence of the Hamiltonian on \( \mathbf{A}^2 \) was removed at the expense of introducing an additional two-particle interaction which can be identified as the dipole self energy (last term in (D.7)). The light-matter interaction is now given by a single-particle dipole interaction \(-e \mathbf{r} \cdot \mathbf{D}(\mathbf{r})\), where the transformed electric displacement field \( \mathbf{D}(\mathbf{r}) \) is given by [117]

\[ \frac{1}{\epsilon_0} \mathbf{D}(\mathbf{r}) = i E_{\text{ph}} (\mathbf{u}(\mathbf{r}) b - \mathbf{u}^*(\mathbf{r}) b^+). \]  

\[ (D.8) \]

### D.2 Second quantization

The electronic Hamiltonian contains the bare electron energies and the Coulomb interaction between electrons. The derivation of the electronic Hamiltonian in second quantization leads to the established form [38]

\[ H_{\text{el}} = \sum_{k, \lambda} \epsilon_k a_{\lambda, k}^\dagger a_{\lambda, k} + \sum_{k, k'} V_{k, k'}^{\lambda, \lambda'} a_{\lambda, k}^\dagger a_{\lambda', k'}^\dagger a_{\lambda', k'} a_{\lambda, k}, \]  

\[ (D.9) \]

---

1. Maria Goeppert-Mayer (1906-1972) was the second woman (after Marie Sklodowska Curie) to be awarded the Nobel prize in physics in 1963 (together with J. Hans D. Jensen) "for their discoveries concerning nuclear shell structure." She remained the last woman to receive a Nobel Price in physics for over half a century until 2018, when Donna Strickland was awarded the price "for groundbreaking inventions in the field of laser physics."
D.2. SECOND QUANTIZATION

where \( a_{\lambda,k}^{\dagger} \) are annihilation (creation) operators for electrons with momentum \( k \) in the band \( \lambda \) and energies \( E_{\lambda,k} \). Coulomb matrix elements are given by \( V_{k,k'}^{\lambda,\lambda'} \).

Dipole interaction

We carry out the second quantization of the dipole interaction Hamiltonian explicitly. This requires special attention as it captures the particular geometry of the light field and its overlap with the electronic wave functions. The dipole-interaction Hamiltonian in second quantization is given by

\[
H_1 = -\frac{e}{\varepsilon_0} \int d^3 r \Psi^\dagger(r) [r \cdot D(r)] \Psi(r) ,
\]

where the electron field operators are

\[
\Psi(r) = \sum_{\lambda,k} a_{\lambda,k} \phi_{\lambda,k}(r) ,
\]

with the electronic wave function

\[
\phi_{\lambda,k}(r) = \frac{1}{\sqrt{S}} e^{i k \cdot r} u_{\lambda}(r_\parallel) \xi_{\lambda}(z) .
\]

We only deal with in-plane momenta and write \( k \) instead of \( k_\parallel \) for simplicity but we note that these are two-component momenta living in a 2D plain. The quantization area is \( S \). It is important to note, that the Bloch function \( u_{\lambda}(r_\parallel) \) is only dependent on the in-plane component of the position as the Bloch theorem is only applied in two dimensions. This is due to the atomic thickness of electronic system that extends only over a single unit cell in the \( z \)-direction. Therefore, the only \( z \)-dependence is found in the confinement wave function \( \xi_{\lambda}(z) \). Due to strong confinement in \( z \)-direction we limit the discussion to the lowest subband \((n = 1)\) that arise from the QW potential (c.f. Section 2.2). Inserting the field operators in (D.10) yields

\[
H_1 = \sum_{k,k',\lambda,\lambda'} M_{\lambda,k,\lambda',k'} \hat{a}_{\lambda,k}^{\dagger} \hat{a}_{\lambda',k'} ,
\]

with the matrix elements between Bloch electrons

\[
\hat{M}_{\lambda,k,\lambda',k'} = -\frac{e}{\varepsilon_0 S} \int d^3 r \phi_{\lambda,k}^{\dagger}(r) [r \cdot D(r)] \phi_{\lambda',k'}(r) = -\frac{e}{\varepsilon_0 S} \int d^3 r e^{-i(k-k') \cdot r_\parallel} \xi_{\lambda}^{\dagger}(z) \xi_{\lambda'}(z) u_{\lambda}(r_\parallel) [r \cdot D(r)] u_{\lambda'}(r_\parallel) .
\]

The electronic confinement functions are strongly localized at the layer of the gain material. At the same time the electric field does not vary much on this length scale and can be evaluated at the position of the layer \( z = z_0 \), which we indicate by writing \( D(r_\parallel, z = z_0) = D(r_\parallel) \). The \( z \)-integration can now readily be evaluated as we assume equal and normalized confinement functions for different bands. We furthermore perform a separation of length scales. To that end, the integration is split into a sum of integrals over each unit cell

\[
\hat{M}_{\lambda,k,\lambda',k'} = -\frac{e}{\varepsilon_0 S} \sum_{R_\parallel} e^{-i(k-k') \cdot R_\parallel} \left( D_\parallel(R_\parallel) \cdot \int_{z_0} d^2 r_\parallel u_{\lambda}^{\dagger}(r_\parallel) (R_\parallel + r_\parallel) u_{\lambda'}(r_\parallel) \right) + z_0 D_z(R_\parallel) \int_{z_0} d^2 r_\parallel u_{\lambda}^{\dagger}(r_\parallel) u_{\lambda}(r_\parallel) .
\]
The unit cells are arranged in the 2D plane of the layer. The in-plane area of the unit cells is \( s_0 \). We consider only small momentum transfers due to light matter interaction \( |\mathbf{k} - \mathbf{k}'| \ll 1 \). Thus the plane wave component of the electronic Bloch-functions varies only little over the extent of an unit cell and is therefore evaluated at the lattice position \( \mathbf{R}_\parallel \) of the unit cell. The same holds for the displacement field, that we separate into in-plane and z-components. For different bands \( (\lambda \neq \lambda') \) the Bloch functions \( u_\lambda(\mathbf{r}_\parallel) \) have orbital characters that are mutually orthogonal. Thus the terms containing \( \mathbf{R}_\parallel \) and \( z_0 \) do not contribute to interband transitions. We also neglect contributions to intraband transitions \( (\lambda = \lambda') \), because the small momentum transfer by the photon within one band would lead only to a small change in energy which is smaller than the photon energy. As a result the integral in (D.15) is independent of the unit cell’s position and we define the position independent dipole matrix element

\[
\mathbf{d}_{\lambda\lambda'} = \frac{e}{s_0} \int_{s_0} d^2r_\parallel u_\lambda^*(\mathbf{r}_\parallel)\mathbf{r}_\parallel u_{\lambda'}(\mathbf{r}_\parallel), \quad \lambda \neq \lambda'.
\]  

We proceed by noting that the momentum transfer \( \mathbf{q} \) and also switched the labeling of \( \lambda \) and \( \lambda' \) using the fact that \( \mathbf{d}_{\lambda\lambda'} = (\mathbf{d}_{\lambda'\lambda})^* \). The resulting matrix transition element for the dipole interaction Hamiltonian is given by

\[
M_{\lambda', \lambda}^q = -i\hbar \mathbf{d}_{\lambda\lambda'} \tilde{u}_\parallel(D.20).
\]

The form of the dipole interaction Hamiltonian in Eq. (D.13) by inserting the transition matrix element

\[
H_1 = \sum_{k,q} M_{\lambda\lambda'}^q a_{\lambda', k}^\dagger a_{\lambda, k+q}^- + (M_{\lambda\lambda'}^q)^* a_{\lambda', k}^\dagger a_{\lambda, k}^-.
\]  

In the second sum we interchanged \( -\mathbf{q} \) to \( \mathbf{q} \) and also switched the labeling of \( \lambda \) and \( \lambda' \) using the fact that \( \mathbf{d}_{\lambda\lambda'} = (\mathbf{d}_{\lambda'\lambda})^* \). The resulting matrix transition element for the dipole interaction Hamiltonian is given by

\[
M_{\lambda\lambda'}^q = -i\hbar \mathbf{d}_{\lambda\lambda'} \tilde{u}_\parallel(D.20).
\]

The form of the dipole interaction Hamiltonian in Eq. (D.19) is not the same as in Eq. (5.6). We proceed by noting that the momentum transfer \( \mathbf{q} \) due to absorption or emission of a photon is small in comparison to the carrier momenta \( \mathbf{k} \) under consideration and \( \tilde{u}_\parallel(\mathbf{q}) \) will be strongly centered around some \( q_m \) with \( |q_m| = \omega/c \), where \( \omega \) is the central wavelength of the cavity mode. In Eq. (D.19) we therefore neglect the \( \pm \mathbf{q} \) in the index of the \( a^- \) operators which then are factored out of the summation over \( \mathbf{q} \). Neglecting also other effects like anisotropy of the light-matter interaction we find a \( \mathbf{q} \)-independent effective interaction rate \( g_{\lambda\lambda'} \) by with

\[
g_{\lambda\lambda'} = -i\hbar \mathbf{d}_{\lambda\lambda'} \sum_{\mathbf{q}} \tilde{u}_\parallel(\mathbf{q}).
\]  

The \( \mathbf{k} \)-independent dipole is a further approximation that is relaxed by writing, according to [83],

\[
\mathbf{d}_k = \frac{\mathbf{d}_0}{1 + \frac{k^2}{m_e c^2}},
\]  

\[
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\]
where $d_0$ is the dipole moment at $k = 0$, $m_r$ is the reduced mass of the applied two-band model and $\epsilon_g$ is the band-gap energy of the material at zero excitation. Finally writing for valence- and conduction band operators $a_{v,k}^{(1)} = a_k^{(1)}$ and $a_{c,k}^{(1)} = c_k^{(1)}$, the final form of the Hamiltonian in Eq. (5.6) is obtained.

**Dipole self energy**

In a similar fashion as for the dipole-interaction Hamiltonian we perform the second quantization of the Hamiltonian describing the dipole self energy in Eq. (D.7). We find

$$ H_{\text{dip}} = \sum_{k,q,t,q',\lambda \neq \lambda',\nu \neq \nu'} M_{\lambda \lambda',\nu \nu'}^{q,q'} a_{\lambda,k}^{q} a_{\lambda',k}^{\dagger} t^{-q} a_{\lambda',k-q} $$

and the transition matrix element in this case is

$$ M_{\lambda \lambda',\nu \nu'}^{q,q'} = \frac{1}{2\epsilon_0 V_m} (\tilde{u}_{\parallel}(-q) \cdot d_{\lambda\lambda'}) (\tilde{u}_{\parallel}(q') \cdot d_{\nu\nu'}). \tag{D.24} $$

However, we will neglect the contribution of the dipole self energy in the dynamical equations as it is small compared to the light-matter interaction. This is seen in the following by direct comparison of the transition matrix elements, evaluated with parameters in the range that is typical for our considerations.

The matrix elements given in (D.20) and (D.24) determine the relative strengths of the contribution of $H_1$ and $H_{\text{dip}}$, respectively. In the system dynamics the matrix elements enter as modulus squared (see Section 5.2). We therefore compare using the Cauchy-Schwartz inequality

$$ \left| \frac{M_{\lambda \lambda',\nu \nu'}^{q,q'}}{M_{\lambda \lambda'}^{q,q}} \right|^2 \leq \frac{1}{2\epsilon_0 V_m} \left| \frac{|d_{\lambda\lambda'}|^4}{|d_{\nu\nu'}|^2} |\tilde{u}_{\parallel}(q)|^2 \right| = \frac{|d_{\lambda\lambda'}|^2}{2\hbar\omega_0 V_m} |\tilde{u}_{\parallel}(q)|^2. \tag{D.25} $$

To find a conservative upper bound for this ratio we consider the smallest possible, i.e. diffraction limited, mode volume $V_m = (\frac{\lambda_0}{2\pi})^3$ and write $\hbar\omega = \frac{\hbar}{\pi c_0}$, where $\lambda_0$ is the vacuum wavelength of the cavity mode and $n$ the refractive index of the cavity. Furthermore we find an upper bound for the Fourier transformed mode function

$$ |\tilde{u}_{\parallel}(q)| = \frac{1}{S} \left| \int_S d^2r \parallel u_{\parallel}(r) \parallel e^{-iqr} \right| \leq \frac{1}{S} \int_S d^2r \parallel u_{\parallel}(r) \parallel \leq \max |u(r)| = 1. \tag{D.26} $$

We thus find

$$ \frac{|d_{\lambda\lambda'}|^2}{2\hbar\omega_0 V_m} |\tilde{u}_{\parallel}(q)|^2 \leq \frac{2n^3 |d_{\lambda\lambda'}|^2}{\hbar\pi c_0 \lambda_0^3} \leq 1.1 \cdot 10^{-5}. \tag{D.27} $$

for $|d_{\lambda\lambda'}| \leq 10 \text{ Å e}$, $n = 3$ and $\lambda_0 \geq 380 \text{ nm}$ (near UV). The values for $|d_{\lambda\lambda'}|$ and $n$ correspond to typical values in the material system we consider and the value for $\lambda_0$ maximizes the ratio of the matrix elements in the visible wavelength range. We see, that the influence of the dipole-dipole self energy is order of magnitude smaller than that of the dipole interaction and can thus be neglected in the dynamics.
Bibliography


