Strong Photon-Photon Interactions
in Solid State Cavity QED

A dissertation submitted to the
ETH ZURICH
for the degree of
Doctor of Sciences

presented by
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2013
Summary

This dissertation reports experiments investigating photon-photon interactions in a solid-state cavity-quantum electrodynamics (cavity-QED) setting. We study low-dimensional quantum-photonic systems, namely single quantum dots (QDs) and quantum wells (QWs) strongly-coupled to nano- and microcavities, respectively.

A device consisting of a single self-assembled QD strongly-coupled to a photonic-crystal nanocavity is investigated under resonant-scattering spectroscopy. Unlike cold atoms in dilute gases self-assembled QDs are mesoscopic objects embedded in a semiconductor host material. Decoherence mechanisms such as charge fluctuations (blinking) in the QD occur, when probing the system optically. To counteract the blinking effect an optical re-pump technique is implemented.

Photon correlation measurements reveal strong antibunching of photons scattered off the system demonstrating the photon-blockade effect. This proves that our device enables strong photon-photon interactions. For particular laser-light detunings, significant photon bunching is observed, due to a direct two-photon transition to the second Jaynes-Cummings manifold.

These single-photon nonlinearities enable the implementation of a single-photon all-optical switch where a single control photon resonant with one of the fundamental polariton states enables the scattering of signal photons resonant with a transition from the first to the second Jaynes-Cummings manifold. By using appropriately tailored laser pulses, the switching speed of the device is shown to be on ultrafast timescales of about 50 ps. As an application, a single-photon pulse correlator working on these timescales is presented.

The second part of the thesis is devoted to a different cavity-QED setting based on the coupling of two-dimensional QW excitons to Fabry-Pérot cavities. In contrast to monolithic cavities with little flexibility of tuning parameters, we present a new type of semi-integrated system consisting of a fiber-end mirror, which is approached to the surface of a chip-integrated semiconductor mirror with a QW layer on top. The curved fiber mirror ensures photonic-mode confinement without degrading the cavity-quality factor. In this fully-tunable system unprecedented polariton lifetimes of up to 100 ps are achieved. Moreover, when driving such a system with an off-resonant laser signatures of polariton lasing are observed for high powers.
Zusammenfassung


Insbesondere erforschen wir spektroskopisch die resonante Lichtstreuung an einem Bauteil, bestehend aus einem einzelnen selbstorganisierten Quantenpunkt, welcher stark an einen Nanoresonator in einem photonischen Kristall gekoppelt ist. Im Gegensatz zu Atomen in kalten Gasen sind selbstorganisierte Quantenpunkte mesoskopische, in einen Halbleiterkristall eingebettete Objekte. Dekohärenzmechanismen, wie zum Beispiel Ladungshfluktuationen im Quantenpunkt, treten zum Vorschein, sobald das System optisch untersucht wird. Hier wird eine Technik vorgestellt, wie diesen Ladungsschwankungen mit Hilfe eines Rückpumplasers entgegengewirkt werden kann.


Im zweiten Teil der Dissertation behandeln wir ein anderes Gebiet der Resonator-Quantenelektrodynamik, in dem die Exzitonen von zweidimensionalen Quantentöpfen an Fabry-Pérot Resonatoren gekoppelt werden. Im Gegensatz zu monolithischen Resonatoren, welche in ihren Eigenschaften kaum verstellbar sind, wird hier ein neuartiges halbintegriertes System vorgestellt, das einerseits aus einem Hohlspiegel an einem Glasfaserende und andererseits aus einem Chip-integrierten Halbleiterspiegel mit darüberliegendem Quantentopf besteht. Werden die beiden Systeme nah aneinandergebracht, entsteht durch die Krümmung des Hohlspiegels eine räumlich stark eingeschränkte photonische Resonatormode mit unvermindert hohem Qualitätsfaktor. In diesem vollkommen verstimmmbaren System werden beispiellos lange Polariton-Lebenszeiten von bis zu 100 ps gemessen. Zudem weist das System Anzie-
chen von Polaritonlaser-Emission auf, wenn es mittels eines nichtresonanten Lasers stark angeregt wird.
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<tr>
<td>$k$</td>
<td>Wave vector</td>
</tr>
<tr>
<td>$m$</td>
<td>Mass</td>
</tr>
<tr>
<td>$n$</td>
<td>Refractive index</td>
</tr>
<tr>
<td>$Q$</td>
<td>Cavity-quality factor</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>Exciton spontaneous-decay rate</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>Cavity-energy dissipation rate</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>Light wavelength</td>
</tr>
<tr>
<td>$\omega$</td>
<td>Light (angular) frequency</td>
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<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
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<tbody>
<tr>
<td>1 eV</td>
<td>$1.602 \cdot 10^{-19}$ J 1 electronvolt</td>
</tr>
<tr>
<td>$e$</td>
<td>$1.602 \cdot 10^{-19}$ As Electron charge</td>
</tr>
<tr>
<td>$c$</td>
<td>$299 \ 458 \ 792 \ \text{m} \ \text{s}^{-1}$ Speed of light</td>
</tr>
<tr>
<td>$\hbar$</td>
<td>$1.0546 \cdot 10^{-34}$ Js Reduced Planck’s constant</td>
</tr>
<tr>
<td>$k_B$</td>
<td>$86 \ \mu\text{eV} \ \text{K}^{-1}$ Boltzmann constant</td>
</tr>
<tr>
<td>$m_e$</td>
<td>$9.109 \cdot 10^{-31}$ kg Free electron mass</td>
</tr>
<tr>
<td>$\epsilon_0$</td>
<td>$8.854 \cdot 10^{-12}$ $\text{Fm}^{-1}$ Vacuum permittivity</td>
</tr>
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<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>AFM</td>
<td>Atomic force microscopy</td>
</tr>
<tr>
<td>As</td>
<td>Arsenic</td>
</tr>
<tr>
<td>AlAs</td>
<td>Aluminum arsenide</td>
</tr>
<tr>
<td>AlGaAs</td>
<td>Aluminium gallium arsenide</td>
</tr>
<tr>
<td>AOM</td>
<td>Acousto-optic modulator</td>
</tr>
<tr>
<td>APD</td>
<td>Avalanche photodiode</td>
</tr>
<tr>
<td>BEC</td>
<td>Bose-Einstein condensation</td>
</tr>
<tr>
<td>BS</td>
<td>Beam splitter</td>
</tr>
<tr>
<td>CCD</td>
<td>Charge-coupled device (camera)</td>
</tr>
<tr>
<td>CdTe</td>
<td>Cadmium telluride</td>
</tr>
<tr>
<td>c</td>
<td>Cavity</td>
</tr>
<tr>
<td>cw</td>
<td>Continuous wave</td>
</tr>
<tr>
<td>DBR</td>
<td>Distributed Bragg reflector</td>
</tr>
<tr>
<td>DPSSL</td>
<td>Diode-pumped solid-state laser</td>
</tr>
<tr>
<td>EIT</td>
<td>Electromagnetically induced transparency</td>
</tr>
<tr>
<td>FC</td>
<td>Fiber coupler</td>
</tr>
<tr>
<td>Symbol</td>
<td>Abbreviation</td>
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<tr>
<td>--------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>FDTD</td>
<td>Finite-difference time domain</td>
</tr>
<tr>
<td>Ga</td>
<td>Gallium</td>
</tr>
<tr>
<td>FSR</td>
<td>Free spectral range</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full width at half maximum</td>
</tr>
<tr>
<td>GaAs</td>
<td>Gallium arsenide</td>
</tr>
<tr>
<td>HBT</td>
<td>Hanbury-Brown and Twiss</td>
</tr>
<tr>
<td>HF</td>
<td>Hydrofluoric acid</td>
</tr>
<tr>
<td>HH</td>
<td>Heavy hole</td>
</tr>
<tr>
<td>In</td>
<td>Indium</td>
</tr>
<tr>
<td>InAs</td>
<td>Indium arsenide</td>
</tr>
<tr>
<td>JC</td>
<td>Jaynes-Cummings</td>
</tr>
<tr>
<td>LA</td>
<td>Longitudinal-acoustic</td>
</tr>
<tr>
<td>LED</td>
<td>Light-emitting diode</td>
</tr>
<tr>
<td>LH</td>
<td>Light hole</td>
</tr>
<tr>
<td>LP</td>
<td>Longpass (filter)</td>
</tr>
<tr>
<td>LP</td>
<td>Lower polariton</td>
</tr>
<tr>
<td>MBE</td>
<td>Molecular beam epitaxy</td>
</tr>
<tr>
<td>MCWF</td>
<td>Monte Carlo wavefunction</td>
</tr>
<tr>
<td>MO</td>
<td>Microscope objective</td>
</tr>
<tr>
<td>N₂</td>
<td>Molecular nitrogen</td>
</tr>
<tr>
<td>NA</td>
<td>Numerical aperture</td>
</tr>
<tr>
<td>PBS</td>
<td>Polarizing beam splitter</td>
</tr>
<tr>
<td>PC</td>
<td>Photonic crystal</td>
</tr>
<tr>
<td>PD</td>
<td>Photodiode</td>
</tr>
<tr>
<td>PID</td>
<td>Proportional-integral-derivative</td>
</tr>
<tr>
<td>PL</td>
<td>Photoluminescence</td>
</tr>
<tr>
<td>QD</td>
<td>Quantum dot</td>
</tr>
<tr>
<td>QE</td>
<td>Quantum efficiency</td>
</tr>
<tr>
<td>QED</td>
<td>Quantum electrodynamics</td>
</tr>
<tr>
<td>QW</td>
<td>Quantum well</td>
</tr>
<tr>
<td>RS</td>
<td>Resonant scattering</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning electron microscopy</td>
</tr>
<tr>
<td>Si</td>
<td>Silicon</td>
</tr>
<tr>
<td>SIL</td>
<td>Solid immersion lens</td>
</tr>
<tr>
<td>SM</td>
<td>Single-mode (fiber)</td>
</tr>
<tr>
<td>TCSPC</td>
<td>Time-correlated single-photon</td>
</tr>
<tr>
<td>TEM</td>
<td>Transverse Electromagnetic</td>
</tr>
<tr>
<td>Ti:sa</td>
<td>Titanium-sapphire (laser)</td>
</tr>
<tr>
<td>TTL</td>
<td>Transistor-transistor logic</td>
</tr>
<tr>
<td>UP</td>
<td>Upper polariton</td>
</tr>
<tr>
<td>X</td>
<td>Exciton</td>
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1. Introduction

Cavity-quantum electrodynamics (cavity-QED) studies the quantum limit of light-matter interaction where one or more two-level quantum emitters are coupled to the field of a single cavity mode [1]. In the strong-coupling regime of cavity-QED the coherent interaction strength between the emitter(s) and the cavity mode exceeds all the dissipative rates in the system, and the energy oscillates coherently back and forth between the emitter and cavity. As a consequence, new dressed eigenstates form as a superposition of quantum emitter and cavity-photon states. These new dressed states, spectrally separated by the so-called Vacuum-Rabi splitting, can be treated as quasi-particles, termed polaritons, with half-matter / half-light character. While probing the system via the photonic channel, the matter-part of the polaritons ultimately enables effective interactions between single photons, leading to exciting new applications ranging from quantum information processing to the study of strongly-correlated many-body systems of light.

In single-emitter cavity-QED systems, the nonlinearity is inherited from the intrinsic anharmonicity of the single quantum emitter. Various implementations of cavity-QED systems in the strong-coupling regime have been reported with atoms in high-finesse cavities [2], with quantum dots (QDs) in different types of monolithic cavities [3–7] and in the microwave domain using superconducting qubits in stripline resonators [8–10].

A different approach to realizing a nonlinear cavity-QED system is based on an ensemble of bosonic quantum emitters inside a two-dimensional layer strongly-coupled to a Fabry-Pérot cavity mode. To first order, such a system is linear with a harmonic energy diagram. However, if a confinement is chosen small enough, dipolar or contact interactions between emitters will become relevant and introduce an effective system nonlinearity.

Single-emitter cavity-QED systems: A fundamental feature in strongly-coupled single-emitter cavity systems is the anharmonic energy level diagram (Jaynes-Cummings ladder): Due to the two-level nature of the emitter and the quantization of the cavity-radiation field, the energy cost for the generation of a polariton is strongly dependent on the number of polaritons that already occupy the system. Such a device therefore embodies the ultimate nonlinear optical device enabling the observation of photon-photon interactions at the single-photon level [11].

A way to confirm single-photon nonlinearities in strongly-coupled cavity-QED systems is the observation of the photon-blockade effect: When a single photon from a resonant laser is absorbed, the system makes a transition into an excited state. A second photon of the same color is blocked and cannot be scattered off the system anymore, due to the additional energy-shift of the transition.

A few experiments in the optical domain using a single atom coupled to Fabry-Pérot [12] or toroidal cavities [13] and cavity-QED implementations in the microwave
domain [14] demonstrated the photon-blockade effect by observing photon antibunching in correlation measurements. In quantum-dot cavity-QED systems, early results indicating optical nonlinearities were reported [15].

Unlike in photon-blockade experiments where the population of the second Jaynes-Cummings manifold is blocked, explicit probing of the higher-energy rung enables conditional quantum dynamics: The excitation of a fundamental polariton using a first laser controls the outcome of a measurement done on a transition between the first and second Jaynes-Cummings manifold using a second signal laser of a different color. Conditional dynamics at the single-photon level is at the heart of classical and quantum information processing, enabling devices with new functionalities such as low-energy or even single-photon switches or transistors [16–18] or controlled-phase gates [19].

Even if a cavity-QED implementation exhibits a Vacuum-Rabi splitting, the possibility of probing the second and higher Jaynes-Cummings manifolds is not guaranteed, due to the decreasing anharmonicity of the energy diagram at higher occupation states. To spectrally resolve these states, a cavity-QED system that is well in the strong-coupling regime is required, in which the coherent coupling strength exceeds the dissipation rates by at least a factor of 2. Extensive studies of the second rung of the Jaynes-Cummings energy diagram have been performed either spectroscopically or through photon-correlation measurements in a few cavity-QED implementations, including atoms coupled to Fabry-Pérot or toroidal microcavities [20, 21], superconducting qubits in strip-line resonators [8–10] or quantum dots in nanocavities [22]. A key feature of conditional quantum dynamics is the timescale of single-photon nonlinearities, which ultimately determines how fast such a device can be controlled. Since the switching times are limited by the reciprocal emitter-cavity coupling strength, QDs strongly-coupled to nanocavities emerge as ideal candidates for the realization of ultrafast single-photon nonlinear devices, due to their record-high coupling strengths at high cavity-photon decay rates.

In the light of applications, the implementation of cavity-QED systems in the optical domain placed onto a semiconductor chip is clearly desirable. However, a major drawback of devices based on self-assembled QDs is the random nucleation of dots both spatially and spectrally. Consequently, the study of several tunnel-coupled nonlinear cavity-QED systems based on self-assembled QDs is too challenging at present. Albeit, when considering arrays [23, 24] or lattices [25] of such nonlinear photonic dots separated by tunnel-barriers, polaritons could hop between neighboring lattice sites and would constitute the elementary excitations in a many-body setting.

The dynamics of such a bosonic lattice is described by the Bose-Hubbard model [26, 27]. When denoting the hopping energy by $J$ and the system nonlinearity by $U$, tuning the ratio $U/J$ induces a quantum phase transition from a superfluid to a so-called Mott-insulating many-body state. Such a transition was experimentally observed using ultracold Rubidium atoms arranged in an optical lattice [28]. In the field of cavity-QED, one-dimensional arrays of strongly-correlated photons were proposed for the extreme case of $U/J \gg 1$, forming a so-called Tonks-Girardeau gas [29] representing a ‘traffic jam’ of impenetrable photons [30] or systems showing crystallization of strongly interacting photons via electromagnetically induced transparency (EIT)-type interactions in optical waveguides [31]. Recently, arrays
of coupled nonlinear cavities were proposed to exhibit exotic Majorana-like modes yielding fractionalized photons [32].

**Many-emitter cavity-QED systems:** Prominent examples for atomic systems exhibiting ultra-strong dipole-dipole interactions over length scales of several $\mu$m are Rydberg atoms. In the Rydberg-blockade effect [33] a single Rydberg atom was shown to block the excitation of a second atom around a radius corresponding to the dipolar interaction length scale [34, 35]. The same idea could be applied to quantum well (QW)-cavity polaritons: If confined to a radius that is comparable to the polariton-polariton scattering length, e.g. by engineering the photonic part of the polariton wavefunction, strong polariton-polariton interactions could be induced. When driving such a system with a resonant laser, the generation of a single cavity polariton would block the subsequent generation of a second polariton, due to the additional nonlinear-energy cost $U$. In analogy to the photon-blockade effect discussed previously, this effect was termed polariton blockade [36].

Semiconductor-cavity polaritons based on QW excitons coupled to planar Fabry-Pérot cavities [37, 38] have been studied extensively in the last two decades. The main challenge in studying strongly-correlated photon states in this environment lies in the tiny QW-exciton Bohr radius of around 10 nm preventing significant exciton-exciton interactions to happen over larger length scales. Exciton confinement to comparable dimensions is non-trivial. A confinement to much larger length scales was achieved by exploiting naturally occurring defects [39], controlled application of sample stress [40], and very recently by light-induced creation of spatially-modulated excitonic reservoirs [41–43]. Fabricating cavities with sub-$\mu$m spatial dimensions like micropillars [44], photonic-crystal cavities [45] or epitaxy-overgrown mesas [46] could provide a solution for stronger confinement. However, these methods typically suffer from additional excitonic (or photonic) loss channels, once the spatial dimensions approach the micron scale, for which surface-interactions become increasingly important. Despite all these ideas, to date an experimental demonstration of strong polariton-polariton interactions showing the polariton-blockade effect has remained elusive.

Conversely, cavity polaritons in the weak-confinement regime with polariton-polariton interactions playing a minor role for the system dynamics have emerged as a test bed for exploring non-equilibrium dynamics of quantum fluids in an integrated solid-state setting [47]. Due to their bosonic nature, polaritons can undergo condensation at elevated temperatures of a few Kelvin [48]. The demonstration of Bose-Einstein condensation of cavity polaritons a few years ago by several groups [39, 40, 49] stipulated a whole series of experiments including the demonstration of superfluid flow [50], Bogoliubov excitations [51, 52], Josephson oscillations [53], vortices [54, 55] and solitons [56].

So far, all experiments have relied on fully-integrated devices with little to no flexibility for modification of device properties. A novel approach followed in this thesis for realizing confined-cavity polaritons is a versatile semi-integrated fiber-cavity platform [57–60] that enables in-situ tuning of the cavity length and thereby of the polariton energy and lifetime. These systems exhibit better cavity-quality factors than those reported fully-integrated systems to date.


Scope of this thesis

This thesis is structured as follows:

Chapter 2 discusses the theoretical framework of the interaction between a two-level quantum emitter with the electromagnetic radiation field of a single cavity mode. The concrete system based on a self-assembled QD coupled to a photonic-crystal nanocavity is presented. Furthermore, the implementation of a maximally-coupled system is discussed.

In Chapter 3, the experimental tools for probing a single-QD cavity system are presented. The method of photoluminescence spectroscopy is introduced. The main experimental tool in this work, namely resonant-scattering spectroscopy, is presented. When resonantly probing the system, the QD is subject to charge fluctuations, leading to an interruption of the optical response. This surprising blinking effect is phenomenologically introduced and an optical re-pump technique to counteract the blinking is presented.

Our main findings of strong single-photon nonlinearities in a single-QD cavity system including the photon-blockade effect and a two-photon resonance into the second Jaynes-Cummings manifold are presented in Chapter 4.

Based on the strong photon-photon interactions of our device we demonstrate the realization of a single-photon all-optical switch on the same Jaynes-Cummings system in Chapter 5.

Chapter 6 introduces a cavity-QED system in which QW excitons are strongly-coupled to the field of a Fabry-Pérot cavity mode. The collective interaction of excitons to optical fields is analyzed along with the effect of system disorder. An introduction to polariton lasing and condensation is given. Moreover, theoretical considerations of polariton-polariton interactions in confined polariton systems are presented.

Finally, Chapter 7 discusses our implementation of a confined cavity-polariton system using a semi-integrated fiber-cavity platform with full tunability of the cavity-mode resonance and the photon lifetime. Results on long-lived polaritons and signatures of polariton lasing with off-resonant optical excitation are discussed.

Part of the results presented in Chapter 4 and Chapter 5 were published in References [61] and [62], respectively. Part of the results presented in Chapter 7 were published in Ref. [60], others are currently being prepared for publication [63]. These references were separately listed at the end of this thesis.
Chapter 2: Cavity-QED with quantum dots in nanocavities

This chapter describes the interaction between a two-level quantum emitter with the electromagnetic radiation field of a single cavity mode. In Section 2.1, the Jaynes-Cummings model is introduced and its implications discussed. The rest of the chapter presents the implementation of the particular solid-state cavity-QED system studied in this work: A single self-assembled quantum dot coupled to a photonic-crystal nanocavity. The basic underlying physics and the growth and fabrication of the two building blocks are discussed in Sections 2.2 and 2.3. A brief introduction to a maximally-coupled QD-cavity system is given in Section 2.4, along with a detailed description of the optical setup and the cryogenics. The technique of photoluminescence spectroscopy is explained in Section 2.5, which enabled the observation of strong QD-cavity coupling.

2.1. The Jaynes-Cummings model

Atoms or quantum dots are multi-level quantum emitters that are intrinsically well separated in frequency. Hence we can reduce an atom or QD to an ideal two-level system. We will constrict the following considerations to the electric-dipole approximation of optical transitions and study a single transition with linear optical polarization. Moreover, we consider a single optical cavity mode with linear polarization. The atom is located in the cavity-mode electric field with maximum strength, as illustrated in Figure 2.1.

The atomic ground state is denoted by $|g\rangle_a$ and the excited state by $|e\rangle_a$. The cavity mode can be occupied by an arbitrary number $m$ of photons, denoted by the Fock states $|m\rangle_c$. A general system state is then

$$|\Psi\rangle = \sum_{j=g,e} \sum_{m=0}^{\infty} c_{jm} |j\rangle_a \otimes |m\rangle_c \equiv \sum_{j,m} c_{jm} |j,m\rangle_c.$$  \hspace{1cm} (2.1)

We will introduce the angular momentum (or excited state) lowering operator $\hat{\sigma}_- = |g\rangle_a \langle e|_a$, with the corresponding raising operator $\hat{\sigma}_+ = \hat{\sigma}_-^\dagger = |e\rangle_a \langle g|_a$. The electromagnetic cavity field annihilation operator will be denoted as $\hat{a}$, with $\hat{a} |m\rangle_c = \sqrt{m} |m-1\rangle_c$ and $\hat{a} |0\rangle_c = 0$. The free system Hamiltonian is therefore given by

$$\hat{H}_0 = \hbar \omega_a |e\rangle_a \langle e|_a + \hbar \omega_c \hat{a}^\dagger \hat{a}.$$  \hspace{1cm} (2.2)
A single two-level atom is coherently coupled to a cavity mode with coupling strength $g$. The system is subject to losses: The stored cavity energy is lost at a rate $\kappa$, due to imperfectly reflective mirrors or absorption in the material. The atom in its excited state spontaneously decays at a rate $\gamma$ and thereby emits photons into free space.

The interaction Hamiltonian in the electric-dipole approximation is obtained by using the Power-Zienau-Wooley transformation [64] and applying the long-wavelength-approximation, which takes into account that the atomic Bohr radius is much smaller than the optical wavelength, resulting in

$$\hat{H}_{\text{int}} = -e \hat{\mathbf{r}} \cdot \hat{\mathbf{E}}(\mathbf{r}_a),$$  \hspace{1cm} (2.3)$$

where $e$ is the unit charge, $e \hat{\mathbf{r}}$ is the electric-dipole operator and $\hat{\mathbf{E}}(\mathbf{r}_a)$ is the cavity-electric field operator at the atom position $\mathbf{r}_a$ in the canonical-quantization picture, given by [65]

$$\hat{\mathbf{E}}(\mathbf{r}) = i \sqrt{\frac{\hbar \omega_c}{2\varepsilon_0 n^2 V_{\text{eff}}}} \left( \phi(\mathbf{r}) \hat{a} - \phi^*(\mathbf{r}) \hat{a}^\dagger \right),$$  \hspace{1cm} (2.4)$$

where $\phi(\mathbf{r}) = \frac{n(\mathbf{r})E(\mathbf{r})}{nE(\mathbf{r}_{\text{max}})}$ is the complex electric-field vector, normalized to the maximum cavity-electric field at position $\mathbf{r}_{\text{max}}$. $n(\mathbf{r})$ is the position-dependent optical refractive index (e.g. inside a solid-state crystal), and we write $n = n(\mathbf{r}_{\text{max}})$ for simplicity. The pre-factor in the square root is obtained when satisfying the normalization condition of the single-photon electromagnetic cavity field, whereby the effective cavity-mode volume was introduced, according to

$$V_{\text{eff}} = \iiint d^3r |\phi(\mathbf{r})|^2 = \iiint d^3r \frac{n(\mathbf{r})^2 |E(\mathbf{r})|^2}{n^2 |E(\mathbf{r}_{\text{max}})|^2}. $$  \hspace{1cm} (2.5)$$

The electric-dipole operator in Equation 2.3 can be written as

$$e \hat{\mathbf{r}} = d_{eg} |e\rangle_a \langle g|_a + d_{ge} |g\rangle_a \langle e|_a,$$  \hspace{1cm} (2.6)$$

with $d_{eg} = \langle e|_a e \mathbf{r} |g\rangle_a$ being the atomic transition-dipole element. When applying the rotating wave approximation and introducing the coherent atom-cavity coupling constant

$$g = \sqrt{\frac{\omega_c}{2\hbar \varepsilon_0 n^2 V_{\text{eff}}}} |\phi(\mathbf{r}_a) \cdot d_{eg}|,$$  \hspace{1cm} (2.7)$$

we obtain the interaction Hamiltonian ($\text{Jaynes-Cummings Hamiltonian}$) [66]
\[
\hat{H}_{\text{int}} = \hbar g \left( |e\rangle_{a} \langle g|_a \hat{a} + \hat{a}^\dagger |g\rangle_{a} \langle e|_a \right).
\] (2.8)

For a maximal coupling constant \( g \) in Equation 2.7, the atom has to be placed into the electric-field maximum, to obtain \( |\phi (r_a)\rangle = 1 \). The polarization axis of the electric field \( \phi (r_a) \) has to be parallel to the transition-dipole vector \( d_{eg} \). Note that the smaller the effective cavity-mode volume \( V_{\text{eff}} \) the larger the coupling is.

The eigenvalues of the system Hamiltonian \( \hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} \) are

\[
E_0 = 0
\]
\[
E_{\pm m} = \hbar \left( m\omega_a + \frac{\delta}{2} \pm \sqrt{mg^2 + \frac{\delta^2}{4}} \right) \quad m \geq 1,
\] (2.9)

where \( \delta = \omega_c - \omega_a \) is the detuning between the two-level transition energy and the cavity-photon energy. The corresponding eigenstates are

\[
|g\rangle = |g,0\rangle
\]
\[
|m, +\rangle = \cos \theta_m |e, m - 1\rangle + \sin \theta_m |g, m\rangle
\]
\[
|m, -\rangle = \sin \theta_m |e, m - 1\rangle - \cos \theta_m |g, m\rangle,
\] (2.10)

where \( \tan 2\theta_m = \frac{2g\sqrt{m}}{\delta} \). For zero detuning \( \delta = 0 \), the energies \( E_{\pm m} \) are separated by \( 2\hbar \sqrt{mg} \), as shown in Figure 2.2(b). The eigenenergies of the fundamental states \( (m = 1) \) are shown in Fig. 2.2(a). They exhibit the characteristic anticrossing signature. The splitting of \( 2g \) between excited states at \( \delta = 0 \) is referred to as Vacuum-Rabi splitting.

At this point we can make an important observation. It is not possible to factorize the state \( |m, \pm\rangle \) into cavity and atom orbitals. The eigenstates are therefore of hybrid atom-cavity character and are called dressed states or polaritons.

Calculating the time evolution of a polariton state in the basis of the atom and the cavity mode, one finds that the energy oscillates coherently between the two systems. These Rabi oscillations with oscillation frequency \( \sqrt{mg} \) are expressed by the energy sidebands of \( \pm \hbar \sqrt{mg} \).

An important consequence of the Jaynes-Cummings Hamiltonian is the arising anharmonicity of the level scheme. Obviously, the energies between different manifolds are not equally spaced, as illustrated in Figure 2.2(b). This anharmonicity is the central theme of most chapters of this thesis.

### 2.1.1. Weak and strong coupling

A real-world system is typically subject to losses and dissipation. The origins are mainly the leakage of light out of the cavity and light absorption in the material at a total rate \( \kappa \), and the spontaneous decay of the excited atom or quantum dot at rate \( \gamma \). The cavity leakage counteracts the system coherence, but also provides a coupling to the environment and thus allows us to probe and manipulate the system.

For the characterization of the cavity losses, \( \kappa \) is often expressed in terms of the dimensionless cavity-quality factor \( Q \). It is defined as the energy loss per optical cycle versus the stored energy. In particular, if pumping is abruptly stopped, the energy stored in the cavity decays exponentially, as given by
Figure 2.2.: Vacuum-Rabi splitting of a coupled atom-cavity system. (a) Close to zero cavity-atom detuning $\delta$, the new eigenstates of the fundamental Jaynes-Cummings manifold, i.e. upper and lower polaritons (blue lines) anti-cross. The dashed red and green curves denote the free space energies of the atomic transition and the cavity mode. (b) The two lowest manifolds of the Jaynes-Cummings ladder on exact cavity-atom resonance.

\[ \frac{I(t)}{I(0)} = e^{-\kappa t}, \text{ with } \kappa = \frac{\omega_c}{Q}. \] (2.11)

In analogy to the cavity-dissipation rate $\kappa$, the rate $\gamma$ characterizes the atomic exponential decay. Here, we will neglect pure-dephasing rates of the excited electronic states, although these turn out to be relevant in our system (see Section 3.2.1).

For an understanding of the lossy system, we restrict our discussion to the low-excitation limit, in which only the subspace spanned by $\{|g, 0\rangle, |e, 0\rangle, |g, 1\rangle\}$ is considered. The master equation

\[
\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [\hat{H}, \rho] + L(\rho),
\] (2.12)

describes the evolution of the system density matrix $\rho$ and has a standard Lindblad term $L(\rho)$ characterizing system losses and dephasing,

\[
L(\rho) = \frac{\gamma}{2} (2\hat{\sigma}_- \rho \hat{\sigma}_+ - \hat{\sigma}_+ \hat{\sigma}_- \rho - \rho \hat{\sigma}_+ \hat{\sigma}_-) + \frac{\delta}{2} (2\hat{a} \rho \hat{a}^\dagger - \rho \hat{a}^\dagger \hat{a} - \hat{a}^\dagger \hat{a} \rho). \] (2.13)

The expression results in a linear differential equation describing the time evolution of the expectation values $\langle \hat{\sigma}_+ (t) \rangle$ and $\langle \hat{a}^\dagger (t) \rangle$,

\[
\frac{\partial}{\partial t} \begin{pmatrix} \langle \hat{\sigma}_+ (t) \rangle \\ \langle \hat{a}^\dagger (t) \rangle \end{pmatrix} = \begin{pmatrix} \frac{\kappa + \gamma}{4} + i\frac{\delta}{2} - i\omega_a \\ ig \end{pmatrix} \begin{pmatrix} \frac{\kappa}{4} - i\frac{\delta}{2} + ig \\ ig \end{pmatrix} \cdot \begin{pmatrix} \langle \hat{\sigma}_+ (t) \rangle \\ \langle \hat{a}^\dagger (t) \rangle \end{pmatrix}. \] (2.14)
2.1. The Jaynes-Cummings model

The real part of the complex matrix eigenvalues yields the excited eigenenergies of the system

\[ E^\pm = \hbar \left( \omega_a + \frac{\delta}{2} \pm \Re \sqrt{g^2 - \left( \frac{\kappa - \gamma}{4} - i\frac{\delta}{2} \right)^2} \right), \quad (2.15) \]

whereas the imaginary part describes the corresponding dissipation rates

\[ \frac{\Gamma^\pm}{2} = - \left( \frac{\kappa + \gamma}{4} \pm \Im \sqrt{g^2 - \left( \frac{\kappa - \gamma}{4} - i\frac{\delta}{2} \right)^2} \right). \quad (2.16) \]

In typical solid-state implementations like ours, the cavity-dissipation rate \( \kappa \) is about two orders of magnitude larger than the two-level spontaneous-decay rate \( \gamma \), a condition that is valid for the rest of this discussion. For such a situation, Figure 2.3 shows the eigenenergies of the lowest excited states as a function of the coupling strength \( g \) at zero detuning. The graphs clearly suggest a classification of the system into two regimes: the weak-coupling regime, if \( g < \frac{\kappa - \gamma}{4} \) and the strong-coupling regime, if \( g > \frac{\kappa - \gamma}{4} \).

Figure 2.3.: Eigenenergies and loss rates. The fundamental eigenenergies and dissipation rates of the lossy system on cavity-atom resonance are displayed as a function of the coupling strength \( g \). (a) The lowest excited eigenenergies of the system become non-degenerate, forming the upper and lower-polariton states, if \( g \) exceeds the rate \( \frac{\kappa - \gamma}{4} \): The system enters the strong-coupling regime. (b) The decay rates \( \kappa \) and \( \gamma \) of the isolated systems merge into the joint polariton-decay rate \( \frac{\kappa + \gamma}{2} \) as the system approaches the strong-coupling regime. The parameters are \( \kappa/\gamma = 80 \).

In the weak-coupling regime – also referred to as the Purcell regime – the system-dissipation rate is too large for Rabi oscillations to occur. Eigenstates of the system are the bare cavity and emitter states. For zero detuning, the states are therefore degenerate. However, in this regime, the relaxation rates are modified, as shown in Figure 2.3(b). In particular, the atomic-decay rate increases considerably, as \( g \) is raised. This so-called Purcell-enhanced emission [67] can be understood in the following way: A ‘bad’ cavity serves as a narrow reservoir of states with a large density \( \rho \propto 1/\kappa \) around the cavity-center frequency. The number of final states into
which the excited atom can decay is thus increased. According to Fermi’s Golden rule, the atomic relaxation rate is therefore enhanced. This intuitive argument is quantified when linearizing Equation 2.16 for the case $g \ll \kappa$, yielding

$$\Gamma_P \approx \gamma (1 + F_P), \text{ with } F_P = \frac{4g^2}{\kappa \gamma}, \quad (2.17)$$

where $F_P$ is known as the Purcell factor. It can be much larger than unity, such that the spontaneous-emission rate is significantly increased.

In the strong-coupling regime, the condition $g > \frac{\kappa - \gamma}{4}$ is fulfilled and the new eigenstates – the polaritons – split in energy by

$$\Delta E^\pm = 2\hbar \sqrt{g^2 - \left(\frac{\kappa - \gamma}{4}\right)^2}, \quad (2.18)$$

for zero cavity-exciton detuning, as shown in Fig. 2.3(a). The system-dissipation rate is $\Gamma^\pm = \frac{\kappa + \gamma}{2} \approx \frac{\kappa}{2}$ for both polariton states. Another expression for the strong-coupling condition is found when replacing $g$ by the value given in Equation 2.7, leading to

$$\frac{Q}{\sqrt{V_{\text{eff}}/ (\lambda/n)^3}} > \sqrt{\frac{\hbar c_0 \pi^3 c^3}{n \omega_c^2 |\phi (r_a) \cdot d_{\text{eg}}|^2}}. \quad (2.19)$$

The experimental challenge for the implementation of a strongly-coupled single-atom cavity system arises in the reduction of the cavity-mode volume $V_{\text{eff}}$, while keeping the cavity-quality factor $Q$ large.

### 2.1.2. Anharmonicity

A system is anharmonic, if the transition energies between different states in the energy ladder are distinct from each other. In the case of a single emitter strongly-coupled to a cavity mode, the level diagram is intrinsically anharmonic – a fact that has its origin in the quantized nature of the radiation field inside the cavity and the intrinsic anharmonicity of the two-level quantum emitter. The anharmonicity becomes evident when looking at the Jaynes-Cummings transition energies (see Fig. 2.2(b)). Considering e.g. the lower-polariton state of each manifold, the transition energy from the $n^{\text{th}}$ to the $(n+1)^{\text{th}}$ manifold at zero detuning $\delta = 0$ can be calculated using Equation 2.9,

$$E_{n \to n+1} = \hbar \left(\omega_{\text{cav}} - \left(\sqrt{n+1} - \sqrt{n}\right) g\right). \quad (2.20)$$

A degree of anharmonicity can be attributed to the system when considering how well the two lowest-transition energies $E_{0 \to 1}^-$ and $E_{1 \to 2}^-$ can be spectrally distinguished from each other. If the polariton linewidth is $\hbar \kappa/2$, the condition is satisfied, if

$$\frac{|E_{0 \to 1}^- - E_{1 \to 2}^-|}{\hbar \kappa/2} = 2 \left(2 - \sqrt{2}\right) \frac{g}{\kappa} \approx 1.17 \frac{g}{\kappa}. \quad (2.21)$$

\(1\) The use of Equation 2.9 instead of Equation 2.15 is justified if $g^2 \gg \left(\frac{\kappa - \gamma}{4}\right)^2$ is fulfilled.
is clearly larger than 1. A useful figure of merit in cavity-QED is therefore given by

$$\xi = \frac{g}{\kappa}. \tag{2.22}$$

With $\hbar \kappa \approx 53 \, \mu\text{eV}$ and $\hbar g \approx 141 \, \mu\text{eV}$, the single-QD cavity system that we use in the experiments throughout this thesis exhibits $\xi \approx 2.7$. Notably, this model does not take into account additional broadening effects, which we find to be relevant (see Section 3.2.1).

### 2.2. Self-assembled InGaAs quantum dots

**Self-assembled quantum dots** (QDs) provide a strong spatial confinement of the electronic movement in a solid-state crystal. In the system studied here, lens-shaped clusters of indium arsenide (InAs) with spatial in-plane dimensions $< 20 \, \text{nm}$ are embedded in a gallium arsenide (GaAs) crystal lattice. The formation of dots is **self-assembled**, due to an elastic strain-relaxation process when combining the two semiconductors of different lattice constants. The clusters of InAs form a confining potential for the wavefunctions of both electrons and holes, due to the considerably smaller band gap of InAs ($0.36 \, \text{eV}$) compared to the host material GaAs ($1.43 \, \text{eV}$).

The QD-emission and absorption spectra consist of discrete narrow lines, featured by transitions between various electron and hole-energy levels. Furthermore, a single QD behaves like a quantum emitter, i.e. the emitted photon stream exhibits perfect antibunching [68]. Due to these properties, self-assembled QDs are often referred to as artificial atoms.

Despite the resemblance, self-assembled QDs remain mesoscopic objects containing about $10^4 - 10^5$ atoms. More involved aspects, such as the interaction with the host material in which the dot is embedded, strain in the material, not well defined dot boundaries and other effects make an understanding of those systems much more challenging.

### 2.2.1. Quantum-dot growth

Molecular beam epitaxy (MBE) allows for the growth of materials with ultimate control of the composition, by exposing a semiconductor wafer with atomic or molecular beams of different materials. During InGaAs QD growth, a GaAs wafer is exposed to particle beams of indium (In) and arsenic (As). Due to a lattice constant mismatch between GaAs and InAs of about 7%, an InAs layer is intrinsically strained when grown on a GaAs matrix. For a properly chosen growth temperature and material composition, the strain is released by the spontaneous formation of small islands of InAs [69], already before the completion of the second monolayer. These clusters are formed at random positions on the wafer. The technique of self-assembled QD growth is called **Stranski-Krastanow method**, named after I. N. Stranski and L. Von Krastanow who developed a model for the nucleation process based on an interplay between binding energies on layers, edges and volume as well as mechanical strain, when combining two materials of different lattice constants [70].
Figure 2.4.: Growth of QDs with reduced thickness. (a) Clusters of InAs are spontaneously formed when exposing the wafer to an overpressure of As. (b) A partial capping layer is grown. (c) During the annealing process, material from the InAs clusters is evaporated, which makes the QDs thinner. (d) A protection layer of GaAs is grown. The strain is released by the formation of shallow bumps.

Here, for the growth of QDs, a GaAs wafer is put in ultra-high vacuum and kept at a high temperature of around 600°C. Initially, a buffer layer with a thickness of a few 100 nm of GaAs is grown on the wafer by exposing it to an overpressure of As and a particle beam of gallium (Ga). Its purpose is the preparation of a clean surface for the QD formation. The Ga beam is subsequently replaced by a beam of In [71]. QDs of a size on the order of 5 – 20 nm in plane and a height of ≈ 4 nm are formed, as depicted in Figure 2.4(a). These dots exhibit optical transitions at wavelengths longer than ≈ 1100 nm. For practical purposes, mostly due to the lack of efficient silicon (Si)-based charge-coupled device (CCD) detectors covering this spectral range, it is desirable to blue-shift the QDs to a more accessible wavelength range between approximately 900 and 980 nm. Therefore, a capping layer of GaAs is grown on the substrate (Fig. 2.4(b)). At the same, in a process called annealing [72], the temperature is increased, resulting in evaporation of material from the QDs, such that their height is decreased, as shown in Figure 2.4(c).

In the last step, a layer of GaAs is grown in order to avoid surface effects, and to protect the dots from diffusion or oxidation, as depicted in Fig. 2.4(d). If this protection layer is sufficiently thin (< 100 nm), a useful side-effect emerges: Rather large (150 – 300 nm) bumps with a height of 1 – 2 nm are formed on top of each QD, due to cluster-induced strain in the capping layer. Using atomic force microscopy (AFM) QDs can be precisely located on the wafer.

Due to the incomplete formation of InAs droplets during the growth process, the remaining material forms a so-called wetting layer. The spectral position of this quantum-well structure is however far in the blue (between ≈ 840 and 860 nm) as compared to the QD spectra.

2.2.2. The quantum-dot level scheme

For a better understanding of the electronic structure in a QD, it is useful to consider the bulk semiconductors GaAs and InAs first, and introduce the potential confinement in a later step.

The band structures of InAs and GaAs have similar shape, though with large band-gap energy differences, as shown in Figure 2.5, so the following discussion is valid for both materials. The lowest-energy Bloch-orbitals of electrons in the conduction band have s-type character. Consequently, the total angular momentum of an electron occupying the conduction band is provided by its spin, yielding a
2.2. Self-assembled InGaAs quantum dots

Figure 2.5.: The band structure of InAs and GaAs in the [100] and [111] directions.

Twofold degeneracy of $S_z = \pm 1/2$. According to semiconductor theory, an effective electron mass $m_{e,GaAs}^* \approx 0.066 m_e$ for GaAs and $m_{e,InAs}^* \approx 0.026 m_e$ for InAs can be attributed to the band curvatures at the Γ-point (electron wave vector $k = 0$) [73].

The highest-energy holes (quasi-particles corresponding to missing electrons with charge $+e$) in the valence band have p-shell symmetry, i.e. the orbital angular momentum is 1. An angular momentum coupling with the hole spin of 1/2 results in a total angular momentum $J$, which can take the values 3/2 and 1/2. Due to spin-orbit coupling, the 3/2 and 1/2 bands are widely separated in energy by 0.34 eV. The $J = 1/2$ states form the so-called split-off (SO) band. Due to the large energy separation from the $J = 3/2$ states, the SO band can safely be neglected in the rest of this discussion.

The hole-angular momentum in quantization direction (growth axis, defined as $z$) in the band with $J = 3/2$ can take the half-integer values between $J_z = +3/2$ and $J_z = -3/2$. In bulk GaAs and InAs these states exhibit fourfold degeneracy at the Γ-point. For non-zero $k$ close to the Γ-point the band structure can be calculated perturbatively using the Luttinger model [74]. These calculations show that the degeneracy is lifted for $k \neq 0$, resulting in two separated bands with different curvatures. These holes are therefore denoted heavy holes (HH) and light holes (LH) respectively, with effective masses $m_{HH,GaAs}^* \approx 0.5 m_e$, $m_{HH,InAs}^* \approx 0.41 m_e$ and $m_{LH,GaAs}^* \approx 0.082 m_e$, $m_{LH,InAs}^* \approx 0.025 m_e$ respectively [73].

If a confinement in growth direction is introduced, such as that from an InAs quantum well sandwiched between bulk GaAs, the modification in the electron-density of states results in an increase of the lowest conduction-band energy. In the case of holes, the situation is more complicated. Due to their different effective masses, HHs and LHs split into separate energy bands differing by more than 10 meV for typical QW thicknesses. In the rest of this discussion, only the higher-lying HHs will be considered. It should be noted that HHs and LHs hybridize away from the Γ-point, due to an anticrossing of the two bands for large $k$ [75].

In the case of self-assembled QDs, the system is additionally confined in $x$- and $y$-directions. This confinement is more gentle than in $z$-direction, yielding lens-shaped QDs with a height of a few nm and in-plane dimensions of $\approx 10$ nm. In analogy
to atoms, the electron and hole orbitals arrange in shells, quantized by the orbital angular momentum. The energy splittings between orbitals of different fundamental quantum numbers correspond to temperatures of several 100 K. Here, I will restrict the discussion to the highest energy (accessible) hole state and the lowest energy electron state.

If an electron is lifted from the valence band to the conduction band in the QD potential (for example by absorbing a photon), the resulting electron-hole pair forms a compound called exciton, due to their mutual Coulomb attraction (see Figure 2.6). In the reverse process, upon recombination of the exciton, a photon is emitted from the dot.

Figure 2.6.: Schematic of an exciton in the QD potential. A confining potential for both, electrons in the conduction band and holes in the valence band results in discrete carrier orbitals. The electron and hole form a bound exciton. Upon the recombination of the electron-hole pair, the transition energy is emitted in form of a photon.

The wavefunction of an exciton is analogous to that of the hydrogen atom. With a Bohr radius of \( \approx 10 \text{ nm} \), excitons are two orders of magnitude larger than their atomic counterparts. This mainly arises from the considerably smaller effective masses and the large dielectric constant of the semiconductor (\( \epsilon \approx 12.5 \)). Due to the twofold spin degeneracy of each carrier, there are four different possible combinations for the formation of excitons. The total angular momentum in \( z \)-direction \( M_z = S_z + J_z \) can take the values \( M_z = \pm 1 \) and \( M_z = \pm 2 \). According to the selection rules for optical transitions in the electric-dipole approximation the total angular momentum difference between initial and final states has to be \( \pm 1 \). The so-called dark excitons with \( M_z = \pm 2 \) therefore cannot recombine optically. Recombinations of excitons with \( M_z = \pm 1 \) can on the other hand occur by the emission of a photon with right-hand or left-hand circular polarization. These compounds are called bright excitons.

Besides those neutral excitons (\( \text{X}^0 \)), other exciton types can be formed, if extra carriers are confined in the QD, for example an extra electron: The generation of an electron-hole pair with the proper spin orientation accordingly leads to the formation of an electron-spin singlet in the conduction band and the occupation of one hole state in the valence band. Such a trion complex is called negatively charged exciton, denoted by \( \text{X}^- \). A theoretical treatment of this trion state is difficult, due to the complicated Coulomb interactions between the carriers. Typically, the recombination energy of \( \text{X}^- \) is by a few meV lower than that of \( \text{X}^0 \).
The case is similar for the positively charged exciton $X^+$. An additional hole in the dot is responsible for the effective positive charge. The respective electron-hole recombination energy is strongly dot-dependent and can be a few meV larger or smaller than the neutral-exciton recombination energy.

If an electron singlet and two holes with angular momenta $\pm 3/2$ occupy a QD, the compound is called neutral biexciton, denoted by $XX^0$. The recombination of one electron with one hole emits a photon that is typically slightly red-shifted with respect to the neutral exciton recombination energy.

### 2.2.3. Exciton Fine Structure

The charge carriers in exciton orbitals of each type can occupy different spin states. The exchange interaction between the spins of electrons and holes thus results in distinct binding energies for the various spin configurations that can be separated by tens, up to hundreds of $\mu$eV. This lifting of degeneracies results in the creation of new optical transitions, due to different angular-momentum states. In combination with dot symmetries some important conclusions concerning optical transitions can be made.

Here, only the fine structure of the neutral exciton will be discussed. An effective Hamiltonian for the electron-hole exchange is given by [76]

$$H_{\text{exchange}} = -\sum_{i=x,y,z} \left( a_i J_i S_i + b_i J_i^3 S_i \right), \quad (2.23)$$

where $S_i$ stands for the electron-spin and $J_i$ for the hole-spin components. $a_i$ and $b_i$ are dot parameters, defined by QD geometry. In the basis $(|+1\rangle, |-1\rangle, |+2\rangle, |-2\rangle)$ – the number in the ket referring to $M_z$ – one finds

$$H_{\text{exchange}} = \frac{1}{2} \begin{pmatrix} \delta_0 & \delta_1 & 0 & 0 \\ \delta_1 & \delta_0 & 0 & 0 \\ 0 & 0 & -\delta_0 & \delta_2 \\ 0 & 0 & \delta_2 & -\delta_0 \end{pmatrix}, \quad (2.24)$$

where $\delta_0 = 1.5 (a_x + 2.25 b_z)$, $\delta_1 = 0.75 (b_x - b_y)$ and $\delta_2 = 0.75 (b_x + b_y)$. For a lens-shaped QD with $D_{2d}$ symmetry, i.e. with equal spatial dimensions in $x$ and $y$ directions, the relation $b_x = b_y$ is fulfilled. Thus, $\delta_1 = 0$ and the bright exciton matrix sub-block in the exchange Hamiltonian is diagonal. Thus, this subspace is twofold degenerate in energy. The transition of each bright exciton into the dot ground state occurs by the emission of a photon of right or left-hand circular polarization, respectively. Thus, the emission of the combined state is unpolarized.

Self-assembled QDs however are slightly elongated along a crystal axis, so the circular symmetry is broken and $b_x \neq b_y$. The exchange Hamiltonian therefore has non-diagonal terms. As a consequence, the excitons with $M_z = +1$ and $M_z = -1$ are not eigenstates of the Hamiltonian anymore. The new eigenstates are $\frac{1}{\sqrt{2}} \left( |+1\rangle \pm |-1\rangle \right)$, spectrally separated by a typical $xy$-splitting of a few tens of $\mu$eV. The photon emission resulting from recombination of the carriers is thus linearly polarized along the symmetry axes of the dot. In this thesis, I will denote these states with $X_{0x}^0$ and $X_{0y}^0$. 

2.3. Photonic-crystal nanocavities

Using an optical cavity, the rapid expansion of light can be prohibited and light can be confined to a certain volume in between two mirrors – at least for a typically very short amount of time, before the light leaks through the mirrors. Motivated by prospects of many applications in physics, like nonlinear optics, lasing or cavity-QED, a very intense field of research is the engineering of cavities with small effective mode volume and a high $Q$-factor, with a single quantum emitter placed inside. Prominent examples for such *microcavities* are micropillars having an active region sandwiched between two highly-reflective dielectric mirrors, or microdisks with so-called optical *whispering gallery modes* along the outermost edge of a dielectric disk. In particular, in the case of cavity-QED, a large figure of merit $Q/\sqrt{V_{\text{eff}}}$ ensures an efficient coherent emitter-cavity coupling, while keeping a long average cavity-photon lifetime $\tau = Q/\omega$. This turned out to be challenging, since the ratio between the cavity surface area and effective mode volume gets very large for decreasing cavity dimensions, which makes absorption at surfaces more prominent.

In 1987, the field of photonics gained a lot of momentum, when a new mechanism for strong photon localization by the use of dielectric superlattices was proposed [77]. At the same time, an inhibition of the spontaneous optical decay of solid-state emitters was proposed when placing them spatially and spectrally properly into such a superlattice [78], nowadays usually called a *photonic crystal* (PC) [79]. The inhibited decay is a result of an optical band gap in the energy diagram, in analogy to electronic band gaps in atomic lattices. Artificial *defect regions* in PC membranes were fabricated, forming cavities with mode volumes on the order of a cubic wavelength and $Q$-factors up to several thousands [80]. Further engineering of the shape of the defect region resulted in even larger $Q$-factors approaching $10^6$ [81].

While three-dimensional PCs [82] with high-$Q$ defect regions [83] were successfully fabricated very recently, cavities with equally promising optical properties can be made with a technologically much simpler approach using thin two-dimensional membranes with a periodic lattice of holes drilled through the semiconductor material. The defect region is defined by omitting one or more holes in the crystal design. Although the PC membrane is two-dimensional, three-dimensional light confinement is achieved by an interplay between Bragg reflection from the lattice of holes and total internal reflection at the membrane surfaces.

Photonic-crystal cavities have been used in many experiments, including lasing [84–88] and cavity-QED [1, 3, 7, 25, 89–91] experiments, and are still subject of intense research.

2.3.1. Light confinement in photonic-crystal nanocavities

Photonic crystals consist of a regular lattice of holes with diameters on the order of 100 nm drilled into a dielectric membrane with a typical thickness of about 100 nm. The propagation of the electromagnetic field in such a structure can be calculated by numerically solving Maxwell’s equations. Similar to electron wavefunctions in atomic lattices, the magnetic component of the electromagnetic field exhibits a Bloch-form that is invariant under certain crystal-lattice translations [79]. Analogously, a band structure describes the frequency distribution $\omega(\mathbf{k}_\parallel)$ of photons with in-plane wave
2.3. Photonic-crystal nanocavities

There are several parameters defining the band structure and thus the band gap of a 2D photonic-crystal lattice. The most important ones are the membrane material and thickness, the type of lattice, the lattice constant $a$ and the radius of the holes $r$.

Due to the Bragg-condition, light with a frequency within the optical band gap cannot propagate in the plane. Hence, a cavity can be constructed in the following way: By e.g. changing the size of one or more holes or even leaving them out at all, a defect region is created. Light cannot propagate in any direction away from the defect region and is consequently confined to a very small volume with only the evanescent field penetrating into the crystal. The bound state lies in the band gap, very much like bound states due to impurities in atomic crystals.

The 2D design of the defect region is such that the majority of the cavity field components have large in-plane wave vectors $k_\parallel$, such that a total internal reflection condition at the membrane surfaces is fulfilled. As a result, a fully three-dimensional optical confinement is achieved. The cavity leaks mainly via the small in-plane wave vectors $k_\parallel \ll \frac{2\pi}{a}$. Such rays instead have large out of plane $k_\perp$ vectors and leave the cavity in a cone shape perpendicular to the membrane surface. An active suppression of the leaky region by slightly modifying the design of the defect region increases the cavity-quality factor $Q$ by at least another order of magnitude [92].

The geometry of the defect region accounts for the resonance frequency, mode structure and polarization of the cavity field, as well as for its $Q$-factor. Typically, a defect supports one or more cavity modes with symmetries that are comparable with the nature of the defect and the surrounding PC lattice [93].

In this work, so-called L3 cavities [94], consisting of three missing air holes in a triangular lattice are used. Whereas PC lattices with one missing hole turn out to have $Q$-factors below 1 000, L3 cavities yield a compromise between a small mode volume $V_{\text{eff}} \approx 0.7 (\lambda/n)^3$ and a high $Q$-factor. Displacing and shrinking the two adjacent holes along the long cavity axis results in a smoother confinement and increases the $Q$-factor up to a theoretical value of 300 000 [92]. Cavities based on silicon and wavelengths in the telecom-C band on the order of 1550 nm reach such high $Q$-factors, whereas GaAs cavities with wavelengths on the order of 900 – 1000 nm exhibit a $Q$-factor that is about one order of magnitude smaller than what is expected from calculations. The reason for this was attributed to surface-state absorption [95]. A scanning electron microscope (SEM) image of an L3 cavity is shown in Figure 2.7(a).

The band gap of L3 cavities supports more than one cavity mode, however, the second lowest mode is spectrally separated by about 25 nm from the fundamental mode. Using a numerical method for solving Maxwell’s equations, which calculates the time propagation of the electromagnetic field in a dielectric structure using the finite-difference time domain (FDTD) method [96], the fundamental mode of an L3 cavity can be calculated [94, 97] in a straightforward manner. The resulting intensity distribution of the mode field in the plane is shown in Fig. 2.7(b). The intensity is clearly maximal in the defect center, where the electric field exhibits an in-plane polarization normal to the long axis of the defect. Moreover, the mode is non-degenerate in frequency, which is important for our applications. All photonic-crystal experiments presented here were performed with the fundamental mode.
2.3.2. Fabrication of photonic-crystal nanocavities

For the PC fabrication, a heterostructure is grown with MBE. First, a sacrificial layer of Al$_{0.7}$Ga$_{0.3}$As with a thickness of $\approx 1$ $\mu$m is grown directly on a GaAs wafer. A layer of GaAs with the desired cavity-membrane thickness (126 nm in this work) follows. If desired, a layer of self-assembled QDs is grown sandwiched in the center of the GaAs layer. Such a heterostructure is illustrated in Figure 2.8(a).

The 2D cavity pattern is written with electron beam lithography: To this end, an electron-beam resist is applied on top of the substrate. The electron-beam breaks the molecular chains in the exposed parts of the polymer with nm resolution. With a developer the exposed material is removed. Then, an inductively coupled plasma (ICP) out of Ar, Cl$_2$ and BCl$_3$ is used to etch the material GaAs, while the resist is hardly affected. The plasma energy is provided by strong oscillating electric
2.4. A maximally-coupled single-QD cavity system

In order to obtain the largest possible coherent-coupling strength $g$, a QD should be coupled to the electric-field maximum of a photonic-crystal cavity. However, self-assembled QD nucleation occurs at random positions on the wafer. Together with the fact that the central lobe of the PC field is about 100 nm wide in diameter, the probability of obtaining a reasonable overlap between the PC mode and a single QD by chance, is small.

Various different approaches have been implemented to overcome this problem. The most common one has been the fabrication of many PC cavities on a QD wafer with low dot densities of around 1 $\mu$m$^{-2}$ [3, 87, 90, 99]. A few well-coupled QDs are then selected for further experiments. However, most QDs are still significantly displaced from the cavity-field maximum. Moreover, two or more QDs are often positioned in the electric-field region of a PC membrane. Even if the excitons of additional dots are spectrally detuned, electronic continuum states [100] can influence the system behavior.

Self-assembled QDs are invisible by SEM-imaging techniques, which are the standard imaging and aligning methods in electron beam lithography. A technique to make QDs visible under SEM is the growth of QD stacks [101, 102] that were used to position PC cavities [89, 103]. However, a degradation of the cavity Q-factor was observed in these structures. As confirmed by FDTD simulations [94], the QD stack modifies the cavity field. Moreover, the cavity-feeding effect (see Section 2.5.2) is stronger, due to the multiple of marker dots, which presumably additionally degrades the $Q$-factor.

Recently, pyramidal site-controlled QDs [104, 105], grown at deterministic positions on the wafer, allowed for precise spatial matching with PC cavities [106]. But these comparably large dots (100 nm – 1 $\mu$m) have so far been of inferior optical quality, exhibiting a poor coupling efficiency to the electromagnetic field on the one hand, and large exciton linewidths on the order of 100 – 200 $\mu$eV on the other hand. This is possibly a result of fluctuating charge distributions close to the dot, or due to impurities incorporated during the growth by metalorganic chemical vapour deposition (MOCVD) [107].

For the experiments presented in this thesis, a QD-cavity system is used that was fabricated with an AFM technique for the deterministic coupling of a single QD to the mode of PC cavity. The devices were fabricated on wafers with QDs grown by Antonio Badolato at the University of California in Santa Barbara (UCSB) and by Stefan Fält at ETH Zürich. AFM-mapping was performed by Martin Winger...
Figure 2.9: An L3 cavity with a deterministically positioned QD. (a) AFM image, with clearly visible strain-induced deformation, indicating the location of the QD (compare Fig. 2.4(d)). (b) Position of the QD, relative to the electric-field intensity distribution obtained from FDTD simulations. The images are taken from Ref. [7].

and Antonio Badolato at ETH Zürich and the cavities were fabricated by Kevin Hennessy at the UCSB.

As described in Ref. [108], the fabrication process requires five separate steps that are briefly summarized in the following:

1. A wafer with an appropriate QD density of $10^{-2} - 10^{-1} \text{ cm}^{-2}$ is chosen. This is achieved by investigating the chip with photoluminescence (PL) imaging (see Section 2.5) and detecting the light with a CCD camera.

2. A course grid of labeled Ni / Au markers as well as thin cross and stripe markers are fabricated around the chosen QD low-density region. This is achieved by electron-beam lithography in combination with a lift-off process.

3. In the labeled region, PL spectroscopy is carried out to scan the chip for isolated QDs with a distance to the markers that is appropriate for cavity fabrication. A subset of QDs with narrow and well identifiable emission lines is chosen.

4. Using Atomic force microscopy (AFM), the relative coordinates of these QDs with respect to gold markers are registered. Although the gold-marker lines and the shallow QD hills (described in Section 2.2.1) have extents of a few 100 nm, the precision is an order of magnitude better by choosing the center of mass coordinates.

5. Cavities with dimensions adapted to the excitonic wavelength of each dot (lithographic tuning) are fabricated around the QDs. For fine-tuning of the cavity-resonance frequencies, the digital-etching technique described in Section 2.4.3 is used.
An AFM image of a final QD-cavity system processed with this method is shown in Fig. 2.9(a). The result of a corresponding FDTD simulation, with the QD position denoted by the filled circle, is shown in Figure 2.9(b). A typical mismatch in the position of the QD with respect to the cavity-electric field maximum of less than 30 nm was achieved [7, 94].

To ensure that there is one and only one QD inside the photonic-crystal membrane, PL spectroscopy together with correlation measurements (Section B.4.1) were carried out.

### 2.4.1. The optical setup for QD cavity-QED

For optical studies on a single-QD cavity system as here I just described, the sample is mounted in a flow cryostat with good optical access (see next section). A description of the confocal optical setup used for both PL and resonant-scattering spectroscopy (see Section 3.1) is provided in the present section. The setup is schematically shown in Figure 2.10. The optics is mounted on an elevated breadboard, with the microscope objective directed onto the cryostat window from the top (see Figure 2.11(a)). Laser light and photons collected from the system are transported via single-mode fibers. As a result, the optical setup, together with the cryostat, forms an independent unit.

In addition to the purpose of confocal excitation and detection (illustrated in Fig. 2.10), the microscope objective (MO) is part of an imaging setup to monitor the sample position during the experiments. The illumination is done using the de-focused light of a near-infrared light-emitting diode (LED) and a beamsampler (BS7), while a CCD camera reveals the image. A photograph of the sample is shown in Figure 2.11(b). The use of a motorized flip mirror (M6 in Fig. 2.10) allows for easily changing between CCD illumination and fiber coupling (FC4) of the light reflected from the sample.

The PL excitation path is defined by the fiber coupler FC1. A linear polarizer (POL1) ensures vertical light polarization to achieve well-defined reflection from the polarization-dependent beamsampler BS1. The measured power with the photodiode PD1 is used for a feedback loop to control the laser power (see Section A.2). A longpass filter (LP) with a cutoff wavelength of 900 nm – mounted before the fiber coupler FC4 – transmits the luminescence light but extincts back-reflected excitation light.

In resonant-scattering experiments, the resonant lasers enter the optical setup through the fiber couplers FC2 and FC3. Again, the resonant lasers are actively power stabilized. A good strategy to achieve an efficient resonant laser-cavity coupling is to initially overlap it with the PL laser spot centered to the PC membrane.

The collected light (FC4 & C) is either directed to a spectrometer when carrying out PL measurements (see Section B.1) or to an avalanche photodiode (APD, see Section B.2) or Hanbury-Brown and Twiss (HBT) setup consisting of two APDs (see Section B.4.1) in resonant-scattering experiments.
2.4.2. Cryogenics

All experiments in this work are conducted at cryogenic temperatures of around 4 – 7 K. If the temperature is considerably raised, phonon-scattering rates increase in the semiconductor crystal, leading to system decoherence. At temperatures above 50 K, thermal relaxation starts to dominate over optical QD recombinations, which is clearly not desired. In order to cool the sample, a liquid-helium flow cryostat is used. In a flow cryostat, the cooling is provided by a laminar flow of helium through the sample environment and relies on the large latent heat of liquid helium near the boiling point of $T = 4.2$ K.

In our experiments, we use the Konti-Cryostat-Mikro from CryoVac Low Temperature Technologies Inc. It consists of a chamber that provides a high-vacuum environment of typically $10^{-7}$ down to $10^{-8}$ mbar. For the purpose of nitrogen tuning (compare Section 2.4.3) the cryostat-top cover was home-designed, with a steel-feedthrough system including a small nozzle directed towards the sample surface. A glass window on top of the sample enables optical access.

Cooling of the sample holder is achieved by its thermal connection to a flexible tubing system containing the flowing helium. For a good thermal connection, the sample is glued onto the sample holder with high-vacuum grease (Fig. 2.12). The
Table 2.1.: Detailed list of the most important elements of the optical setup.

<table>
<thead>
<tr>
<th>Item</th>
<th>Description</th>
<th>Manufacturer, product</th>
</tr>
</thead>
<tbody>
<tr>
<td>L1</td>
<td>SM fiber for PL laser input</td>
<td>Thorlabs P3-830A-FC-5</td>
</tr>
<tr>
<td>L2,3</td>
<td>SM fibers for resonant laser inputs</td>
<td>Thorlabs P3-830A-FC-5</td>
</tr>
<tr>
<td>C</td>
<td>SM collecting fiber</td>
<td>Thorlabs P3-830A-FC-5</td>
</tr>
<tr>
<td>FC1-3</td>
<td>Fiber couplers</td>
<td>Aspheric lenses Thorlabs C280TME-B, ( f = 18.4 \text{ mm}, \text{ NA = 0.15} ) and ( z )-axis translation stages Thorlabs SM1Z</td>
</tr>
<tr>
<td>BS1-3, BS6-7</td>
<td>90 : 10 beamsamplers (V-polarization)</td>
<td>Newport 10B20NC.2, wedged 30 ± 15 arcmin, (99 : 1 for H-polarization)</td>
</tr>
<tr>
<td>BS4-5</td>
<td>50 : 50 beam splitters</td>
<td>Thorlabs BSW08, wedged 30 ± 10 arcmin</td>
</tr>
<tr>
<td>POL1-4</td>
<td>Linear polarizers</td>
<td>Thorlabs LPVIS050, sodium-silicate glass</td>
</tr>
<tr>
<td>LP</td>
<td>900 nm longpass filter</td>
<td>Thorlabs FEL0900</td>
</tr>
<tr>
<td>M1,M2</td>
<td>Dielectric mirrors</td>
<td>Newport 10D20BD.2 and Thorlabs BB1-E03</td>
</tr>
<tr>
<td>M3,M4</td>
<td>Silver coated mirrors</td>
<td>Thorlabs P01</td>
</tr>
<tr>
<td>M5</td>
<td>Silver coated mirror with xy Piezo actuators</td>
<td>Radiant Dyes MDI-H and Thorlabs P01</td>
</tr>
<tr>
<td>M6</td>
<td>Flip mirror for imaging sample on CCD camera</td>
<td></td>
</tr>
<tr>
<td>PD1-3</td>
<td>Si switchable-gain detectors</td>
<td>Thorlabs PDA100A-EC</td>
</tr>
<tr>
<td>PD4</td>
<td>Optical power meter</td>
<td>Newport 1830-C</td>
</tr>
<tr>
<td>CCD</td>
<td>CCD camera, focused to infinity</td>
<td>Watec 120N and lens Thorlabs LBF254-200-B, ( f = 200 \text{ mm} )</td>
</tr>
<tr>
<td>MO</td>
<td>50× microscope objective, NA = 0.55, ( f = 4 \text{ mm} )</td>
<td>Nikon CF Plan 50X CR, EPI; correcting for imaging errors from the 0.7 mm thick cryostat window; mounted on a ( z ) translation stage with a piezo actuator</td>
</tr>
</tbody>
</table>

sample holder only has a weak mechanical link to the outside chamber, and can be moved laterally with sub-\( \mu \text{m} \) resolution by means of a stepper motor.

In the experiments, we connect a helium-dewar vessel to the cryostat via a transfer tube. A self-sustaining laminar helium flow sets in, due to a growing overpressure in the helium dewar. If the pressure is not sufficient, it can be either artificially increased by connecting a compressed helium bottle to the dewar, or alternatively, a pump can be connected to the cryostat exhaust. The cool down of the sample to the final temperature typically takes between half an hour and an hour. The helium that is consumed during the experiment is collected and fed back into a helium recycling system.

For the fine regulation of the sample-holder temperature, we use the mechanical valve of the transfer tube. After a thermalization period of about 1 h, the temperature is stable enough (to about 0.1° C) for performing experiments. Alternatively,
Figure 2.11.: Sample illumination. (a) Photograph showing the microscope objective directed onto the cryostat window from the top. The small pipe for the nitrogen gas inlet is visible in the lower right corner. (b) Photograph of the sample taken with the CCD camera. The PC membrane is visible as a darker square of about $5 \times 5 \, \mu m^2$. A resonant laser illuminates the center of the membrane (white spot).

Figure 2.12.: The cryostat. Photograph of the cryostat chamber without top cover, showing the sample holder with the semiconductor chip on top.

A proportional-integral-derivative (PID) controller can be used for stabilizing the temperature of the system.

Typically, the helium flow is chosen such that the final temperature is about 7 K. This reduces the helium consumption on the one hand, and makes the helium flow less turbulent on the other hand, leading to an increased sample stability.

Using a liquid-helium dewar with 100 liters, the helium supply lasts for about 36 h. For longer experiments, the dewar can be refilled from a second dewar without
2.4. A maximally-coupled single-QD cavity system

The interruption of sample cooling. For this purpose, we designed a special adaptor with two feedthroughs for filling that is directly mounted on the head of the first helium dewar.

2.4.3. Control of the cavity-mode wavelength

One of the drawbacks of most monolithic cavities is their lack of spectral tunability. Whereas the tuning of cavity-mode frequencies is straightforward in Fabry-Pérot cavities (compare Chapter 7), by simply modifying the distance between the two mirrors, there is no degree of freedom in monolithic structures, as for example in micropillars or microdisks. Tuning these cavities close to the resonance of a QD thus requires an extremely precise control of the dimensions during fabrication.

In the case of a photonic crystal, the FDTD simulations for the written PC design predict rough values for the mode wavelength. Nevertheless, the size and shape of the actually processed structure can deviate from the model by errors in the percent range, resulting in spectral distributions of a few tens of nm. In order to overcome this issue, a technique called wet chemical digital etching is used for fine-tuning the mode wavelength [109]. To this end, the sample surface is exposed to air, resulting in a ≈ 1.5 nm deep oxidation of the cavity surface area, as illustrated in Figure 2.13(a). This layer can then be removed by immersing the sample into citric acid.

![Figure 2.13: Tuning a photonic-crystal nanocavity](image)

**Figure 2.13:** Tuning a photonic-crystal nanocavity. (a) A full cycle of the wet chemical etching technique: The sample is first exposed to air, resulting in the oxidation of the membrane-surface area. The ablation of the oxidized material is achieved by immersing the structure into citric acid. (b) Nitrogen-gas adsorption on a photonic-crystal cavity. The effective PC-refractive index increases through the formation of an N$_2$-ice layer.

Consequently, the membrane has slightly shrunk to a smaller thickness on the one hand, and exhibits larger holes on the other hand. The effective refractive index is therefore smaller, such that the cavity mode is shifted by ≈ 2 nm towards shorter wavelengths. This self-limiting etching process can be repeated many times, and the desired wavelength can be set with the number of etching cycles up to a total shift of several tens of nm while hardly degrading the cavity Q-factor. Initially, the cavity pattern is thus designed with some red-shift with respect to the desired wavelength, in order to have precise control using the digital tuning technique.
The wet-etching of the structure is an irreversible process. However, for an extensive study of the system, it is necessary to have a knob for the relative detuning of emitter and cavity frequencies during the experiments. In different monolithic QD-cavity implementations, this is typically achieved by tuning the QD-transition frequency, either by applying a static electric field in between electrical gates, inducing a QD-frequency shift due to the Stark effect [91, 110, 111] or by increasing the temperature, resulting in a shift of the electronic band-gap energy [3, 4, 90]. Both methods have a very narrow tuning range and bring along other problems, such as optical losses at the gates or increased phonon-scattering rates at higher temperatures.

In this work, we apply a tuning method that does not have all these drawbacks, and that is based on the adsorption of a thin layer of additional material on the PC membrane [87, 112], which is exactly the opposite mechanism to the one behind the wet chemical etching technique. Additional dielectric material results in a thicker membrane and smaller air holes (see Fig. 2.13(b)), and therefore raises the effective PC-refractive index, whereby the cavity mode is shifted to longer wavelengths.

In practice, the adsorption of dielectric material can easily be achieved in a cryogenic environment, since the cold chip acts as a getter pump for particles in the near environment. In fact, a relatively fast intrinsic cavity-mode drift of about 1 nm h$^{-1}$ towards longer wavelengths was observed in the initial stages of the experiments. This uncontrolled mode walking arose from a bad vacuum and a resulting adsorption of dirt on the PC membrane. For a study of the system under temporally stable cavity-mode detunings, a spontaneous mode walk is not desirable and should be suppressed. Setting up a clean vacuum environment was thus indispensable and yielded a considerable decline of the mode-walking speed in our experiment. Further improvements could be achieved by baking the chamber for a week and mounting the sample as close as possible to the clean window surface – yielding a mode-walking speed of about 0.007 nm h$^{-1}$.

Whereas this intrinsic cavity-mode walking turned out to be useful in a few long-lasting experiments, controlled cavity-mode tuning is a must for precision experiments. To this end, a controlled scheme was designed and developed by Martin Winger and Thomas Volz, based on the adsorption of molecular nitrogen (N$_2$) gas, as schematically depicted in Figure 2.13(b). A feedthrough for pure N$_2$ gas was fabricated into the lid of the cryostat chamber, as visible in Fig. 2.11(a). The nozzle needs to be directed towards the sample with more or less line-of-sight for efficient tunability. To tune the cavity mode in a controlled way, a small reservoir chamber between the N$_2$ gas bottle and the cryostat feedthrough is filled with gas and subsequently pumped down to a low pressure of about 15 mbar in a first step. Then, a high precision needle valve is carefully opened for gas injection into the cryostat. By monitoring the cavity-mode frequency using in-situ PL spectroscopy, the mode can be placed at the desired detuning with a precision of the spectrometer resolution.

It turned out that a higher tuning rate with less overall gas injection is achieved, if the tuning process is conducted at a sample temperature of about 20 K. Most likely due to a higher surface mobility at this temperature, the grown film is rougher in this case. As soon as the final mode wavelength is reached, the valve is closed and the structure re-cooled to 7 K. The nitrogen tuning enables a cavity shift towards longer wavelengths of up to 10 nm without degradation of the $Q$-factor.
In few cases, the tuning process did not work nicely. This is indicated by the formation of a ‘leopard fur pattern’, i.e. arrays of dark spots on the sample surface, that are clearly visible on the CCD camera image. Typically, this resulted in a rather small cavity-signal contrast in resonant-scattering experiments and a bad suppression efficiency (compare Section 3.1.1). This unwanted effect can be eliminated by pre-heating the sample to 40 K for about 5 minutes before starting the tuning process.

The reverse process – shifting the cavity to shorter wavelengths – can be done by simply heating the sample to temperatures larger than 25 K. However, the heating can not be controlled very well and the shift in wavelength is larger and more abrupt.

### 2.5. Photoluminescence spectroscopy

Photoluminescence (PL) spectroscopy is a very useful technique to characterize optical transitions in a semiconductor environment. The basic operation principle is shown in Fig. 2.14: A laser with a frequency above or around the band gap of GaAs generates electron-hole pairs, forming bulk excitons that migrate in the near vicinity of the QD-trap potential (a). If an exciton is trapped by the QD potential, a non-optical relaxation process ensures the occupation of the lowest-energy state of the dot within ps timescales (b). This fast non-energy conserving decay mechanism relies on polaron states involving longitudinal-optical phonons [113]. After a lifetime of about 1 ns, the QD exciton spontaneously recombines and the energy is released by the emission of a photon (c).

![Figure 2.14.](image)

**Figure 2.14.: Schematic of a photoluminescence experiment.** (a) An electron-hole pair is generated in the bulk semiconductor upon absorption of a photon from the pump laser. (b) The carriers relax into the QD-potential trap by fast non-optical relaxations. (c) By spontaneous recombination, a luminescence photon is emitted.

In this work, we carry out PL experiments on a QD embedded in a PC cavity. The presence of the cavity mode modifies the QD spectrum, as we will demonstrate in Section 2.5.1. Notably, the measured PL spectra turn out to be strongly dependent on the excitation laser wavelength (see Section 3.2.1). In fact, if the system is excited above the GaAs band gap (wavelengths shorter than 819 nm), we observe primarily luminescence of the cavity resonance (due to cavity feeding, see Section 2.5.2), while the QD lines are weak. Therefore, we typically perform PL spectroscopy using pump wavelengths slightly below the GaAs band gap, at around 835 nm (close to carbon
impurity acceptor states) or at around 857 nm (on the wetting-layer resonance). The corresponding spectra exhibit narrower QD lines and less efficient cavity feeding. We typically use titanium-sapphire (Ti:sapphire) lasers for PL excitation (a MIRA 900 from Coherent® Inc. for continuous-wave (cw) and pulsed excitation or a TIS-SF07 from Teknoscan for cw excitation; see Section A.1).

A practical implementation of high-resolution PL spectroscopy of single semiconductor QDs [114] is micro-PL in a confocal excitation and detection scheme. As shown in the simplified scheme in Figure 2.15, the light is guided from a single-mode (SM) fiber coupler via a beam splitter (BS) and through a microscope objective with a high numerical aperture (NA) to the sample surface. The objective corrects for aberrations from the cryostat window, resulting in a diffraction limited spot on the chip. The luminescence is collected with the same objective and coupled into a SM fiber. The confocal scheme with point-like excitation and collection (achieved by the SM-fiber core and NA-matched couplers) ensures high spatial resolution on the one hand and efficient detection on the other hand. The light, coupled into the SM fiber, is guided to a high-resolution grating spectrometer for analysis. A more detailed description of the optics is found in Section 2.4.1 and in Section B.1.

![Diagram of the confocal setup](image)

**Figure 2.15.:** Scheme of the confocal setup with fiber cores as point apertures. Fiber couplers (FC) ensure collimation of the beams. A BS reflects 10% of the laser light, which is then focused onto the sample using a microscope objective (MO). The luminescence is collected through the out-coupling fiber port. The high (90:10) BS ratio ensures efficient collection of the emitted light.

While here photoluminescence is mainly utilized as a QD re-pump mechanism (see Section 3.2.1) or for spectrally localizing the cavity and exciton resonances, the study of a single-QD cavity system with PL spectroscopy itself revealed beautiful, but also unexpected results that led to active discussions in the whole QD-cavity community in the past years. These observations will be shortly discussed in the following sections.

### 2.5.1. Observation of strong coupling

When the system is excited with an off-resonant laser around the band-gap frequency of GaAs, the charge configuration of the QD is random in time and as a consequence a multitude of spectral lines are detected by the spectrometer, originating from optical recombinations of the different exciton configurations (see Section 2.2.2). Figure 2.16 displays a spectrum recorded using cw-laser light at a wavelength of $\lambda = 857$ nm.
and a power just below QD saturation. The different charge configurations can be identified by several characteristic features, such as their spectral positions and dependence of the emission intensities on pump power. An in-depth study of the optical properties of InGaAs QDs in cavities, ranging from the characterization of the \( X^0 \) finestructure to the observation of neutral dark excitons, was carried out previously [115]. This work paved the way for a thorough understanding of our system.

**Figure 2.16.** A typical PL spectrum of a QD-cavity system. The off-resonant luminescent cavity mode is clearly visible. Prominent transitions are labeled. The off-resonant excitation power is 65 nW.

In addition to the various exciton lines, the fundamental cavity mode lights up in all spectra. Even when the cavity mode is not resonant with any exciton line, as is the case in Fig. 2.16, it luminesces strongly—an effect that remained mysterious for a long time, but could recently be explained by a feeding mechanism based on multi-excitonic QD states and acoustic phonons (see Section 2.5.2). Before explaining this mechanism, we first describe the photoluminescence in the case, when the cavity-mode frequency is close to an exciton line in more detail.

In this thesis, we mainly study the coupling of the cavity mode to the neutral exciton \( X^0 \) transition (the coupling energy \( h\gamma \approx 141 \mu eV \) to the \( X^0 \) exciton state is slightly larger than the coupling to the \( X^+ \) state of 99 \( \mu eV \)). Strong coupling between the neutral exciton and the cavity mode through an avoided-level crossing is evident, as the cavity mode is tuned across the exciton line (Figure 2.17(a)). The feature observed in the experiment exhibits an additional vertical line, corresponding to the other exciton line of the finestructure doublet with linear polarization perpendicular to that of the cavity mode (see Figures 2.17(a) and (b)). The emission from this line cannot be suppressed entirely by polarization filters, due to a slight mismatch of the cavity polarization axis \( V \) with the polarization axes of the QD (see inset of Fig. 2.17(a)).

Besides the polariton lines and the weakly-coupled exciton line, the uncoupled-cavity mode is visible as well. This is at first sight a totally surprising observation, since the uncoupled-cavity mode is not an eigenstate of the strongly-coupled system.
Figure 2.17.: Observation of strong coupling. False-color plots of PL spectra near cavity-X\(^0\) resonance. (a) Besides the avoided-level crossing, the uncoupled-cavity mode is clearly visible, as a straight line crossing the X\(^0\) line. In addition, a dark exciton state lights up at around 937.1 nm. The emitted light was filtered with a polarizer aligned to the cavity polarization V. Some light from the x-polarized exciton state (see inset) is detected as well, due to the slight mismatch of the cavity polarization axis V with the exciton polarization axis y. (b) With the polarizer aligned to the H-axis, only the weakly-coupled exciton is visible. The displayed angular mismatch in the inset in (a) is exaggerated.

However, during a finite observation time, the QD charge state changes many times and leads to off-resonant feeding of the then uncoupled-cavity mode.

### 2.5.2. Off-resonant cavity feeding

The results presented in this section were measured by Martin Winger and Thomas Volz, et al. and were published in Ref. [100]. As already mentioned in the previous section, the cavity luminescence is maintained in PL experiments, even if the cavity mode is not in resonance with any QD-spectral line. Luminescence is observed at a cavity-mode blue-detuning of up to 8 nm with respect to the s-shell exciton-transition manifold and at a red-detuning of up to 30 nm. Cavity feeding via other channels than the QD, such as bulk excitons or tails of the broad wetting-layer resonance play a negligible role, since cavities that contain a wetting layer but no QD and even cavities with a QD located at a cavity-electric field node do not show significant cavity luminescence. An early explanation of the effect was pure exciton dephasing [116], however, this mechanism could not explain feeding at detunings of more than 10 nm.

The observations suggest that there is a quasi-continuous density of QD states that feeds the cavity mode very efficiently, due to strong Purcell-enhancement. This background was explained taking into account the transitions between all possible highly-excited shell states (p, d, etc.) for both electrons and holes. Notably, these series of higher-order exciton states are energetically located above the stable exciton states (see the green levels in the calculated level scheme in Figure 2.18(a)). Consequently, the lowest-order exciton transition that is able to couple to the cav-
Photoluminescence spectroscopy

2.5. Photoluminescence spectroscopy

ity mode is a biexciton (XX) recombination. The experimental observations were well reproduced with numerical simulations based on the model by the authors of Ref. [100].

For cavity red-detunings less than 3 nm longitudinal-acoustic (LA) phonons can assist relaxation from the exciton into the cavity mode leading to cavity feeding as well [108] (see Figure 2.18(b)), an observation that has been seen by many other groups in experiments [117–119] and was modeled theoretically [120, 121]. Off-resonant cavity feeding is very well understood meanwhile and clearly demonstrates that a semiconductor QD is not a simple $N$-level atom. Instead, the mesoscopic environment of the QD plays a significant role. Ref. [122] provides an overview of the present states of the field in the investigation of the cavity-feeding phenomena.

**Figure 2.18.** Off-resonant cavity feeding mechanisms. (a) Calculated QD spectrum showing a series of possible transitions between higher exciton manifolds, to which the cavity couples. The image is taken from Ref. [108]. (b) Cavity feeding by acoustic phonons. The exciton energy is de-excited via the cavity mode by the emission of a phonon carrying away the difference in energy.
3. Experimental methods

In this chapter, I discuss the experimental and technological requirements for the experimental results presented in Chapters 4 and 5. A detailed description of resonant-scattering spectroscopy is provided in Section 3.1 including a discussion of QD-charge blinking (Section 3.2). Section 3.3 describes the preparation and control of tailored resonant pulses in the ps-range.

All new results in QD cavity-QED described in this thesis were performed using a sample with a maximally cavity-coupled QD (see Section 2.4). The thickness of the photonic-crystal membrane is 126 nm, the lattice constant is $\approx 260$ nm and the hole radius $\approx 70$ nm. The cavity-quality factor is $Q \approx 25\,000$, corresponding to a linewidth $\hbar \kappa \approx 53$ $\mu$eV. The neutral exciton of the InAs QD luminesces at a wavelength of 937.25 nm at a working temperature of $T \approx 7$ K. Without active N$_2$ tuning, the cavity-mode wavelength is at $\approx 935$ nm. As extracted from resonant-scattering experiments (see Section 3.1), the QD-cavity coherent coupling constant is $\hbar g = 141$ $\mu$eV. The system is well in the strong-coupling regime and therefore strongly anharmonic; i.e. the figure of merit $\xi = g/\kappa \approx 2.7$ is clearly larger than 1 (see Section 2.1.2).

3.1. Resonant-scattering spectroscopy

As demonstrated in the previous sections, photoluminescence is an extremely useful technique to study QD-cavity devices. However, the excitation of the QD is intrinsically incoherent, since it relies on non-optical electronic decay and relaxation mechanisms from bulk GaAs or impurity exciton states. In light of future applications, e.g. in quantum information processing, a resonant and coherent excitation of the system from the vacuum state is clearly favorable.

Another drawback of PL excitation is the fact that the charge configuration in the dot is randomized. Although, QD-charge control is possible by sandwiching the QD layer in between electrical gates [123, 124], even inside photonic-crystal cavities [111, 125, 126], we do not make use of this technique here, since it significantly degrades the cavity-quality factor. As a result, the exciton lines of many different charge configurations are present in the time-integrated spectrum. In contrast, using resonant spectroscopy, single spectral lines can be addressed individually.

Resonant spectroscopy can be performed using a narrow-bandwidth laser that is tunable across the system resonance. In order to obtain a signal from the system, the transmitted or reflected light is recorded using a photodiode (PD). While PL spectroscopy directly excites the QD, resonant probing accesses the photonic channel, i.e. the cavity mode [6]. Whereas Fabry-Pérot cavities exhibit an almost ideal absorption contrast of 100 %, the free-space coupling contrast to photonic-crystal
Experimental methods

The coupling contrast to PC cavities can be significantly enhanced using photonic-crystal waveguides [127, 128] or fiber tapers [6, 129, 130]. In these cases, light is coupled into and out of the cavity via near-field coupling. Here, we used a free-space design in a reflection geometry. In the following, we will call the resonant probing of our QD-cavity device resonant-scattering (RS) spectroscopy. Notably, in our device the direct resonant laser-coupling to a QD is orders of magnitude smaller than the laser-cavity coupling.

For recording continuous spectra, a narrow bandwidth laser that is tunable in a mode-hop free manner is essential. We use a TLB-6319 Velocity® diode laser from New Focus (Newport). The laser wavelength is controlled by an active feedback loop using an optical wavelength meter (Ångtrom WS Ultimate 30 MC4) for measuring the wavelength. The scattered light is recorded with APDs (see Section B.2). The laser power has to be stable over the whole tuning range. This is achieved with active power stabilization, described in Section A.2.

We use the same confocal setup as for PL experiments, discussed in Section 2.5. Thus, the reflected light is detected, in contrast to typical transmission geometries used for Fabry-Pérot cavities (see also Fig. 2.10 and Table 2.1).

In order to suppress any undesired laser light back-reflected from the sample surface, we use a so-called crossed-polarization method [90], which is described in the following section.

3.1.1. The crossed-polarization technique

As previously mentioned, the laser-PC coupling efficiency is at least an order of magnitude better than the laser coupling to the QD. Consequently, we predominantly couple to the single-QD cavity system via the cavity mode. Nevertheless, the contrast of the cavity resonance within the laser-light background is less than 10 %, which makes a suppression of the non-interacting laser light crucial.

Using a polarization filter (POL4 in Figure 2.10), the beam is linearly polarized to a very high degree. The polarizer axis is chosen +45° with respect to the polarization axis (0°) of the cavity mode. Whereas non-interacting light reflected from the photonic-crystal surface remains in this polarization state, scattered light exhibits a polarization direction of 0°. Using a second polarizer (analyzer) with its axis at −45°, the non-interacting light can thus be extinguished. A schematic of the polarization of the different light fields is shown in Fig. 3.1(a). The method comes at a moderate cost of losing a factor of 2 when coupling the light into the cavity, and another factor of 2 when collecting the scattered light.
3.1. Resonant-scattering spectroscopy

Figure 3.1.: Crossed-polarized detection. (a) The cavity mode is excited under $+45^\circ$ using a polarizer, and the light collected through an analyzer at $-45^\circ$ with respect to the polarization axis of the far-field cavity mode. (b) RS spectrum taken with the crossed-polarization technique.

In principle, one could use a free-space detection scheme of the scattered light, with the analyzer placed directly in front of the detector. Instead, we first couple all the light into a single-mode fiber (via the fiber coupler FC4 in Fig. 2.10, NA-matched with the couplers FC2 and FC3). The out-coupled light then traverses the analyzer and is subsequently detected by an APD. This intermediate fiber passage has two advantages: Firstly, the fiber is equipped with polarization paddles (see Section A.3) that allow for precise polarization control, making the use of expensive optical waveplates obsolete. The second benefit is even more important: When reflected from the photonic-crystal lattice, the non-interacting laser light is subject to interference effects induced by the PC holes. The beam-cross section consequently no longer remains completely Gaussian, but gathers components of higher-order transverse-electromagnetic (TEM) modes [131]. Since these modes propagate differently within the collection path (including lenses and mirrors), they are subject to small rotations of the polarization and therefore no longer efficiently suppressed in a free-space detection scheme. Instead, if the fundamental TEM mode is coupled into a SM fiber in a mode-matched manner, all higher-order TEM modes are efficiently extinct. This-fiber purification step improves the spectral cavity-mode contrast by about one order of magnitude, resulting in a reflected laser-light suppression of up to $1:50,000$.

Figure 3.1(b) shows the RS spectrum of a PC-cavity mode, recorded with this method. A Lorentzian function fits the data very well, yielding a cavity-quality factor of $Q \approx 23,000$.

In the experiment, the implementation of a crossed-polarization scheme is by no means a trivial task and brings along a few difficulties that are discussed in the following:

1. As mentioned previously, the cross section of the optical laser mode with the PC-cavity field is very weak, whereas the PC membrane acts like a mirror for
direct laser-light reflection. Consequently, we measure a non-interacting laser power that is about 400 times larger than the power of scattered cavity light. Including the additional loss by a factor of 4 as discussed before, a laser-light suppression of 1 600 would be necessary to obtain a signal-to-background ratio of only 1. A precise alignment of the collected light to the analyzer axis is thus mandatory.

2. The optical elements in the setup are not perfectly achromatic over a spectral range of several nm, resulting in a bad laser suppression outside a fairly narrow spectral window, for which the polarization suppression was optimized. This limitation is less important in the actual experiment, since we are typically interested in a much smaller spectral range of about 0.5 - 1 nm. However, another spectral interference effect turns out to be more severe: Possibly due to an etaloning effect from optical elements with plane-parallel surfaces that guide the light through the setup, the resonant spectra exhibit in some cases power modulations with a peak-to-peak wavelength of about 0.3 nm. During the different experimental runs (cool-downs), these modulations were more or less pronounced - ranging from a complete absence up to more than 50 % in the background. A possible explanation involves the shapes of the nitrogen-ice layer grown on the PC surface. The presence of interference fringes made meaningful experiments impossible and led to repeated cycles of warming-up and cooling-down again.

3. As described in Section 2.4.2, the chip is mounted on a sample holder that can be positioned with an integrated two-axis motor. This construction however is subject to drifts on the order of several µm h⁻¹, possibly due to small temperature changes inside the cryostat. Since the beam spot is diffraction-limited, a misalignment of a few 100 nm results in a considerably inferior coupling efficiency to the cavity mode. Moreover, the background-laser light-suppression efficiency suffers from drifts of the PC-defect region away from the laser spot. A realignment of the sample position after about every 15 min is thus necessary. In addition to re-alignments using the motor, we correct x and y drifts with a piezo-controlled mirror mount (element M5 in Figure 2.10). The microscope objective is equipped with a piezo actuator as well for corrections in focal distance.

4. The background-laser light can be suppressed by a factor of up to 1 : 50 000, resulting in a power ratio of about 20 between scattered cavity light and non-extinct laser light. This moderate ratio results in interference effects between the two light fields, which modify the RS spectrum. The exact nature of the interferences relies on their phase difference. To give an example, for precise constructive interference, the signal-to-background ratio is $(E_1 + E_2)^2 = (\sqrt{20} + 1)^2 \approx 30$, whereas it is $(\sqrt{20} - 1)^2 \approx 12$ for destructive interference. For intermediate phase differences, the cavity-mode spectral line becomes strongly asymmetric. In the experiment, we are able to control this phase difference by the use of fiber-polarization paddles, allowing us to observe spectral lineshapes ranging from constructive to intermediate and destructive interference patterns. The tuning for perfect constructive interference and sub-
sequent subtraction of the background delivers clear and low-noise RS spectra. However, interferences with laser light clearly influence the measurements of second-order correlations (compare Section 4.3) and should therefore be suppressed in the best possible manner.

Additional necessary requirements for a successful experiment are high stability of the optical setup, high lab-temperature stability, helium-flux stability and motional stability of all optical fibers.

3.1.2. RS spectroscopy of a single-QD cavity system

In the experiment, the scattered cavity-peak power is very low – around 25 fW (femtowatt) for a laser input power of 1 nW. This power is below the noise level of p-i-n photodiodes. We therefore use APDs from PerkinElmer, Inc. (SPCM-AQR-14 APD) in the single-photon counting mode with a quantum efficiency of about 20% in the wavelength range of the QD emission (compare Section B.2). The cavity-peak power thus corresponds to a photon-count rate of about 50 000 s⁻¹.

The cavity mode is initially prepared by spectrally tuning it in resonance with the X⁰ line (see Section 2.4.3), such that polaritons are formed. When a resonant laser is scanned across the spectral range around the X⁰ resonance, the light is expected to be scattered off the cavity, when the laser frequency is resonant with one of the two polariton states, which both have half-cavity and half-exciton character. However, it turns out that even for low resonant-excitation powers of 1 nW, we only observe the uncoupled-cavity mode. But, if the excitation laser power is decreased to only 40 pW, the polariton states appear in addition to the uncoupled-cavity peak, as shown in Fig. 3.2(a). A full scan of the cavity-mode resonance across the exciton line, taken at an excitation power of 22.5 pW, is shown in Fig. 3.3. The outer peaks exhibit the expected anticrossing signature and can therefore clearly be identified as the upper and lower-polariton states. In contrast, the central peak follows a straight line, as expected for the uncoupled-cavity mode.

![Figure 3.2](image)

**Figure 3.2.** RS spectra of the three-peak structures at low laser power. (a) Strong coupling of the cavity mode with the X⁰ state. (b) Strong coupling with the X⁺ transition.
**Figure 3.3.: Avoided-level crossing in RS spectroscopy.** False-color plot of RS scans taken at a power of 22.5 pW, while the cavity-mode wavelength is scanned across the X0 resonance. In addition to the polariton states, a third central peak stemming from RS at the uncoupled-cavity mode is observed.

Although this three-peak structure resembles the structure observed in PL spectroscopy (see Figure 2.17), its origin is different and unexpected. In PL spectroscopy, the uncoupled-cavity peak is a result of cavity feeding. In RS spectroscopy however, we address a single two-level system strongly-coupled to a cavity mode and we would a priori not expect any influence of higher-lying QD states. A saturation effect (or Mollow-triplet signature [132]) can be excluded, since the system always remains in the low-excitation limit: The power, at which the average polariton-occupation number would reach $\langle n \rangle = 1$, is given by

$$P_{(1)} = 2 \frac{E_{\text{phot}}}{\eta \tau} = \frac{4\pi^2 \hbar c^2}{\eta Q \lambda^2} \approx 860 \text{ nW}, \quad (3.1)$$

that is orders of magnitudes larger than the laser powers in the experiment. Here, $E_{\text{phot}}$ is the energy of a photon with wavelength $\lambda = 937$ nm, $\eta = 0.02$ is the laser-cavity coupling-efficiency and $\tau = Q \lambda / \pi c$ is the polariton lifetime on cavity-exciton resonance.

For a further study of the unexpected third peak and the polariton-saturation mechanism, we recorded the system spectrum for different resonant laser-excitation powers down to a few pW. The traces shown in Figure 3.4(a) demonstrate that the third peak completely vanishes for a laser power of 3.2 pW and only the two polariton modes remain. For increasing power, the uncoupled-cavity peak grows quadratically, whereas the upper and lower-polariton peaks saturate against a value of about 200 detected photon events per second.

A possible explanation for the observed blinking effect is the occurrence of an electron-spin flip, once a polariton has been generated by a laser photon, yielding a
3.1. Resonant-scattering spectroscopy

![Figure 3.4](image)

**Figure 3.4.: Nonlinear-power dependence of the cavity mode and polariton states.** (a) RS traces taken at different laser powers. (b) Peak-count rate on the uncoupled-cavity resonance as a function of laser power. (c) Level scheme of a phenomenological model for the system dynamics. The first entry in the kets describes the QD state, the second one the cavity-photon number. A decay into a charged state, denoted by $|h, 0\rangle$, occurs with a branching ratio $b$, when a polariton state $|1, +\rangle$ or $|1, -\rangle$ is excited. The QD-recovery rate into the neutral ground state $|g^0, 0\rangle$ is denoted by $R$.

dark and long-lived exciton and an uncoupled-cavity mode. However, we excluded this effect from being mainly responsible for the interrupted polariton signal, since an expected recovery of the RS signal, when the lower polariton is resonant with the dark exciton (see Fig. 2.17), was not observed. Another explanation for the third peak is a laser-induced charging of the QD, resulting in a de-coupling of the QD transitions from the cavity mode. The assumption that additional charge carriers could play a role in the dynamics is confirmed by the fact that a similar three-peak structure is observed, when the cavity mode is scanned across the $X^+$-transition (see Figure 3.2(b)). With no spontaneous or induced charging present, the QD should always be neutral and no polaritons should be visible in this case. Their observation gives a hint that the QD ‘sometimes’ captures an additional hole.


3.2. Quantum-dot blinking

The observed effect of blinking is present in many QD systems [133] and is routinely reported from other solid-state based emitters, such as nitrogen-vacancy centers [134] and nanowires [135]. It typically gives rise to an interruption or modification of the optical response, when the emitter changes its internal state by acquiring an additional charge carrier or by a spin flip to a dark state (or off-state). These off-states can exhibit lifetimes that exceed the original state (on-state) lifetime by far, leading to a long-term interruption of the actual signal. Blinking can occur spontaneously or can be induced by the excitation laser.

It was demonstrated that excitonic transitions of higher manifolds ($n \geq 2$) hybridize with wetting-layer continuum states [136] and could therefore play a role in QD blinking. In our system, the rate at which two-photon processes occur at resonant powers of 10 pW, can be estimated in the following way: Accounting for the 2% laser-cavity coupling, the 45° polarization angle and the half-cavity content of a polariton, the rate at which polaritons are generated is $r_{\text{pol}} \approx 2.4 \cdot 10^5 \text{ Hz}$. The rate, at which two photons with arrival times $t_1$ and $t_2$ enter the system jointly (i.e. $|t_2 - t_1| < \frac{\kappa}{2}$, where $\frac{\kappa}{2}$ is the polariton lifetime), is given by $r_{\text{pol}}^2 \cdot \frac{\kappa}{2} \approx 1.4 \text{ Hz}$. Since the experimentally observed blinking rates are orders of magnitude larger, two-photon processes can be excluded from being responsible for the blinking effect.

The observation of cavity feeding (see Section 2.5.2 and Ref. [100]), led to the conclusion that the cavity mode does not resonantly couple to excitonic continuum states, when the QD is in the neutral ground state. Hence, the origin of the blinking effect remains unknown. A possible explanation involves a fluctuating charge environment in the vicinity of the QD, due to impurity states, to which the polaritons resonantly couple, opening additional polariton-decay channels (compare Fig. 3.4). As a result, a polariton decay does not always result in a neutral QD-ground state $|g^0, 0\rangle$. Instead – with a small branching ratio $b$ – an additional charge carrier is acquired, which results in the cavity mode being uncoupled from the QD transitions. A spectroscopic investigation of the charged states when driving the system resonantly could confirm that assumption. However, this experiment turned out to be too difficult, due to low photon rates.

We will make the following assumptions: If an extra carrier has been captured by the dot, the neutral ground-state recovery rate $R$ is low compared to all other involved rates, and the system most of the time remains off-resonant. In the rest of the discussion, we will refer to the resonant QD state, if the QD is neutral and therefore coupled to the cavity mode, and the off-resonant state, if the dot is charged.

In the following, we will denote $\tau_b$ as the average neutral ground-state lifetime that is limited by laser-induced blinking and $\tau_{\text{off}}$ as the average time the QD remains in a charged off-resonant state. It turns out that these blinking times depend on the resonant-laser power, as we will find out. Our assumptions are substantiated experimentally by measuring the statistics of the resonantly scattered light from the polaritons and the uncoupled-cavity mode. We measured the time-dependent second-order autocorrelation functions $g^{(2)}(\tau)$ (see Section B.4.1) on the polaritons and the uncoupled-cavity mode for different excitation powers between 5 and 20 pW. The results for a power of 15 pW are shown in Figure 3.5(a), demonstrating a clear bunching effect of both the lower-polariton photon emission (black trace) and the
uncoupled-cavity photon emission (red trace). Exponential fits on the bunching peaks reflect the typical lifetimes of the two system ground states $\tau_b$ and $\tau_{\text{off}}$. Note that these lifetimes are different from the polariton or cavity excited-state lifetimes. In Fig. 3.5(b) the extracted lifetimes $\tau_b$ (black dots) and $\tau_{\text{off}}$ (red dots) are shown. Two important conclusions can therefore be made:

1. $\tau_b$ considerably decreases for increasing laser power, whereas $\tau_{\text{off}}$ exhibits a slight increase.

2. Even for a low power of 15 pW, $\tau_{\text{off}}$ is about an order of magnitude larger than $\tau_b$, which implies that the QD remains charged for most of the time.

![Figure 3.5.](image)

**Figure 3.5.:** Lifetimes of the neutral QD ground state and the charged ground state. (a) Autocorrelation traces on the lower-polariton resonance (black trace) and the uncoupled-cavity resonance (red trace) at a probe power of 15 pW. (b) Extracted $\tau_b$ (black dots) and $\tau_{\text{off}}$ (red dots) as a function of excitation power.

With a simple classical rate-equation model, the blinking dynamics can be simulated. A rough estimate of typical orders of magnitude of the involved lifetimes can be estimated.

We define the rate $F$, at which photons are injected into the cavity on exact resonance, as

$$ F \approx \frac{\eta p}{2E_{\text{phot}}}. $$

(3.2)

Here, the factor 2 stems from the $45^\circ$ angle between the cavity-mode and laser polarization. First, we assume that the laser is resonant with one of the polariton states (for example the lower polariton $|1, -\rangle$) and injects photons into the system at a rate $F/2$ (due to only half cavity content of the polariton state). It is sufficient to consider the occupation probabilities of the three states $|g, 0\rangle$, $|1, -\rangle$ and $|h, 0\rangle$.
only (compare Fig. 3.4(c)); denoted here as $\rho_0$, $\rho_1$ and $\rho_2$. The rate equations in the steady state thus are

$$
\begin{align*}
0 &= \partial_t \rho_1 = \frac{F}{2} \rho_0 - \frac{1}{2} (F + \kappa) \rho_1 \\
0 &= \partial_t \rho_2 = \frac{b \kappa}{2} \rho_1 - R \rho_2 \\
1 &= \rho_0 + \rho_1 + \rho_2,
\end{align*}
$$

(3.3)

where $\frac{\kappa}{2}$ is the polariton-decay rate and $b$ the previously mentioned branching ratio for capturing an additional charge carrier. The low-excitation limit ensures that $F \ll \kappa$. Together with the assumption, $R \ll \{ \frac{F}{2}, \frac{b \kappa}{2} \}$, the solutions are easily calculated,

$$
\begin{align*}
\rho_0 &\approx \frac{1}{1 + \frac{F b}{2 R}}, \quad \rho_1 \approx \frac{\frac{F}{2} \rho_0}{1 + \frac{F b}{2 R}}, \quad \rho_2 \approx \frac{\frac{F b}{2} \rho_0}{1 + \frac{F b}{2 R}}.
\end{align*}
$$

(3.4)

In this model, the correlation traces follow – to first order$^1$ – exponential decay curves at rates at which the system relaxes from $|g^0, 0\rangle \rightarrow |h, 0\rangle$ (via $|1, -\rangle$) and $|h, 0\rangle \rightarrow |g^0, 0\rangle$, so

$$
\tau_b = \left( \frac{b \kappa \rho_1}{2 \rho_0} \right)^{-1} = \left( \frac{F b}{2} \right)^{-1}, \quad \tau_{\text{off}} = R^{-1}.
$$

(3.5)

At a laser power of $p = 15$ pW, the photon-injection rate is $F = 7.1 \cdot 10^5$ s$^{-1}$. With the measured values $\tau_b \approx 240$ µs and $\tau_{\text{off}} \approx 3$ ms (see Figure 3.5(b)) we obtain

$$
R \approx 300 \text{ s}^{-1}, \quad b \approx 1.2 \cdot 10^{-2}.
$$

(3.6)

This means that in roughly 1 % of the cases, a polariton decay does not result in the neutral QD-ground state. Instead, the energy is released by the decay into a system state with an additional charge, which results in a ‘blackout’ of the polariton line, i.e. no photons are subsequently scattered on the polariton transition. In the steady state, the power-dependent rate, at which photons are scattered off a polariton resonance, is given by

$$
\tilde{f}_{\text{pol}} = \frac{F}{2} \rho_0 = \frac{F}{1 + \frac{F b}{2 R}}.
$$

(3.7)

Saturation sets in as soon as the term $\frac{F b}{2 R}$ in the denominator exceeds 1. Using Equation 3.2, the laser power at which the polaritons start to saturate is thus given by

$$
p_{\text{sat}} = \frac{4E_{\text{phot}} R}{\eta b} \approx 1.1 \text{ pW}.
$$

(3.8)

This result is consistent with the observations in Fig. 3.4(a). Our model can also explain the quadratic increase of the photon-emission rate from the uncoupled-cavity mode, when the laser wavelength is resonant with this mode. Depending on whether the QD is in the on- or off-resonant ground state, we have to distinguish between two different photon-injection rates $F_{\text{on}}$ and $F_{\text{off}}$. In the resonant ground state, the system eigenstates are the polaritons, therefore the laser light is far detuned and

\footnote{neglecting higher-order processes, such as e.g. $|h, 0\rangle \rightarrow |g^0, 0\rangle \rightarrow |h, 0\rangle \rightarrow |g^0, 0\rangle$}
the injection rate thus strongly suppressed. Due to a finite spectral overlap $\epsilon$ of the laser line with the polariton resonances, the polariton-injection rate is given by

$$\frac{F_{\text{on}}}{2} \approx \epsilon \frac{F}{2}, \quad (3.9)$$

with an estimated $\epsilon = 2 \left(1 + \left(\frac{4g}{\kappa}\right)^2\right)^{-1} \approx 0.0175$ (using the experimental parameters as input). If the QD is in the off-resonant state, the laser light couples to the bare cavity mode and hence

$$F_{\text{off}} = F. \quad (3.10)$$

The associated photon-scattering rate, when the laser excites the uncoupled-cavity peak, is thus given by

$$f_{\text{cav}} = F_{\text{off}} \rho^2 = F_{\text{off}} \frac{F_{\text{onb}}}{2R} = \frac{\epsilon b}{2R} \frac{F^2}{1 + \frac{F_{\text{onb}}}{2R}} \quad (3.11)$$

with $\rho^2$ obtained in Equation 3.4. As expected, the rate increases quadratically, up to a power, when the term $\frac{F_{\text{onb}}}{2R}$ in the denominator exceeds 1, and then linearizes. The power at which the cavity-mode signal becomes linear is thus given by

$$p_{\text{lin}} = \frac{4E_{\text{phot}} R}{\eta e b} \approx 62 \text{ pW}, \quad (3.12)$$

consistent with the experiment.

To obtain admissible count rates, probe powers on the order of 1 nW are desirable. However, based on the above model, this power is so large that a resonant QD-ground state occupation probability of only $\rho_0 \approx 0.1 \%$ is predicted, which is far too low to perform meaningful experiments. The ground-state lifetime at this power is $\tau_b \approx 4 \mu s$, as predicted from the equations above. In order to overcome this limitation, we developed a QD re-pump scheme at a rate larger than $\tau_b^{-1}$. It relies on the application of a re-pump laser that shuffles the QD back into its neutral ground state.

### 3.2.1. Off-resonant quantum dot re-pumping

As described in the previous section, the QD-exciton transitions become off-resonant from the cavity mode, if an additional carrier is captured by the dot. A single electron or single hole remains in the dot for several ms, as estimated in Equation 3.6. To observe polaritons, the laser power is therefore limited to a maximum of about 40 pW. At this power and on exact cavity-$X^0$ resonance, the photon-count rate on the polariton peaks is about 100 s$^{-1}$. This is clearly too low for recording second-order correlation measurements within a realistic time frame.

A way to solve this problem, would be to ‘flush’ the dot with carriers and hope that the cascade of recombination events eventually would leave the QD absolutely empty. This indeed can be done in our system by additionally shining in an off-resonant laser with a frequency above or around the band gap of GaAs, in analogy to PL spectroscopy (see Section 2.5).
As a matter of fact, the single-QD cavity PL spectrum exhibits a strong dependence on the excitation wavelength. The total photon-emission rate by different excitonic transitions is of particular interest in this regard, since it gives a hint about the distribution of carriers charging the QD. As shown in Fig. 3.6, the emission of photons by recombinations of the $X^-$ transition is strongly suppressed between 838 and 847 nm, whereas $X^+$ transitions are the brightest and thus most frequent recombinations. These imbalances could have their origin in the presence of charge (impurity) acceptors in the vicinity of the QD [137], such as carbon-acceptor states.

Since the recombination of a neutral-exciton state leaves the QD empty, it is desirable to choose an excitation wavelength at which the $X^0$ emission dominates. According to Figure 3.6, excitations in the narrow band between 835 and 836 nm, or above 850 nm result in the preferential formation of $X^0$ states (see spectrum taken at 857 nm in Fig. 2.16).

In order to implement the idea of QD re-pumping, we apply a laser pump-probe scheme at a repetition rate between 0.5 and 1 MHz. The case of 1 MHz is shown in Fig. 3.7(a): First, we probe the system for 500 ns with RS spectroscopy. Then, the APD readout is interrupted (see Figure 3.7(b)), while the QD is re-pumped using an additional PL laser pulse of 250 ns duration. The APD readout remains in the OFF state for the rest of the period to ensure that light from carrier recombination is not detected. During the ON state, the residual rate of PL photons is less than 1 % of the count rate of resonantly scattered photons. The PL signal recorded during the re-pump pulse (OFF state) can be used for monitoring sample drifts. The re-pump pulses are derived from a cw Ti:sapphire laser (TIS-SF07 from Tekhnoscan, see Section A.1) and modulated using an acousto-optic modulator (AOM; see Section A.2).

For a re-pump wavelength in the range of 836 nm, only a tiny increase of the resonant polariton-scattering rate was observed. On the other hand, at a wavelength of around 857 nm (which is close to the wetting-layer resonance), the polariton states can be very well recovered. Figure 3.8 shows RS spectra without a re-pump laser (black trace) and with applied re-pump laser (red trace), when the cavity mode is...
3.2. Quantum-dot blinking

Figure 3.7: Scheme for QD re-pumping. (a) Pulsing sequence for QD re-pumping at 1 MHz. The RS laser (red rectangle) is continuously switched on. The blue pulses indicate the re-pump laser pulses with a duty cycle of 0.25. During the re-pump time and an additional buffer time for optical PL relaxations (ruled area), the APD signal is gated to the OFF line (shown in (b)). (b) Gating the APD signal. The electronic APD pulses (corresponding to single-photon detection events) are temporally split in two parts using an analog switch (DG419) and a pulse-delay generator (DG645 digital delay generator by Stanford Research Systems) that is synchronized with the PL pulses. The signal is thus split into resonantly scattered light and photoluminescence.

on exact resonance with the $X^0$ transition. When the re-pump scheme is applied, the polariton peaks are clearly resolved and overlap only slightly with the tail of the uncoupled-cavity resonance.

An important conclusion can be made when comparing the two signals at the uncoupled-cavity peak position. If the off-resonant re-pump pulse always yielded a neutral QD, the middle peak would vanish almost completely. However, as can be seen in Fig. 3.6, a certain percentage of the light emitted by the dot stems from other charge configurations, and we expect a non-perfect suppression of the uncoupled-cavity peak. The ratio between the photon-count rates of the uncoupled-cavity mode on the two traces displayed in Figure 3.8 directly yields the probability

\footnote{A small residual uncoupled-cavity peak would remain, due to resonant-laser induced charging of the dot within the resonant-probing time of 500 ns. However, the chance for this to happen is small, since the neutral-QD lifetime is about 6 $\mu$s (compare Sections 3.2 and 3.2.2)}
(of about 60 – 70 %) of a re-pump pulse to leave behind a charged QD state after recombination. The re-pump success probability is therefore about $r \approx 30 – 40 \%$.

**Figure 3.8.: On-resonance cw RS spectra.** The black trace was recorded without the additional re-pump laser. With applied re-pump laser (peak power of 400 nW and a wavelength of 857 nm), the resonant signal from the polaritons is partially recovered (red trace). The probe power is 1 nW.

A surprising observation is made when measuring the polariton linewidths: According to the calculations conducted in Chapter 2 (see Equation 2.16) the linewidth of the upper and lower polaritons is expected to be $\hbar \Gamma^\pm = \hbar \frac{\kappa + \gamma}{2} \approx \hbar \frac{\kappa}{2}$, since the exciton-decay rate $\gamma$ is negligible. Instead, we observe a linewidth of both upper and lower polaritons that is identical to the uncoupled cavity-mode linewidth $\hbar \kappa$ (see Figure 3.8). A possible reason for the broadening of the polariton resonances is pure exciton dephasing that may stem from phonon-mediated coupling of the two dressed polariton states. In addition, fluctuations of the charge environment of the QD, partly due to the re-pump laser, might also contribute. A simple simulation of the system using a Master-equation ansatz reproduces the experimental data, if a pure exciton-dephasing rate $\hbar \gamma_{\text{deph}} = 13 \mu$eV is introduced.

With the additional re-pump laser applied, we tuned the cavity-mode wavelength across the neutral-exciton transition, and took RS spectra at the same time. An avoided crossing of the polariton states is clearly visible, as shown in Fig. 3.9. We observe photon-count rates on the two polariton peaks of up to 5000 s$^{-1}$. This is sufficient for carrying out autocorrelation measurements with pulsed resonant spectroscopy (see Section B.4.1 and Chapter 4).

### 3.2.2. Estimating typical blinking times

As discussed in Section 3.2, $\tau_b$ (corresponding to the lifetime of the neutral-QD state in the presence of laser light) depends strongly on the resonant-laser power, with an estimated value of $\tau_b \approx 4 \mu$s for 1 nW excitation power using the numbers worked out in Section 3.2.

To experimentally determine the blinking times, we performed autocorrelation measurements on the upper-polariton resonance, using the re-pump method. Fig. 3.10 displays such a histogram for pulsed resonant excitation at a mean power of 1 nW and a re-pump pulsing rate of $1/T_{\text{repump}} = 0.5$ MHz. As a consequence of
3.2. Quantum-dot blinking

**Figure 3.9:** Avoided-level crossing using a re-pump laser. False-color plot of RS spectra for different cavity-mode detunings from the neutral-exciton resonance, taken at a resonant-laser power of 1 nW. The off-resonant re-pump laser on the wetting-layer transition was applied, resulting in clearly visible polariton states. The cavity mode was tuned by exploiting an intrinsic cavity-mode walking of about 0.007 nm h$^{-1}$.

the periodic gating of the APD signal, the autocorrelation function exhibits a periodic triangular shape as a function of delay. The triangle at zero time delay is about a factor of 3 larger than triangles at other times. Assuming that photon emission from the polariton is observed, only if the QD is in the neutral ground state, the height is proportional to the re-pump success probability $r$ (compare Section 3.2.1), whereas the height of all other triangles is expected to be proportional to $r^2$. This (classical) bunching effect thus indicates that the system, on average, is in the desired neutral ground state after every third re-pump pulse, so $r \approx 0.33$, consistent with the estimation of $r$ provided in Section 3.2.1.

To find out the neutral ground-state lifetime, the slightly curved trailing slope of the triangle at zero delay is investigated. Using a simple classical model, the detected polariton emission after a re-pump pulse is given by $I(t) = I_{pol} e^{-t/\tau_b}$, if $0 < t < T_{\text{repump}}/2$ and $I(t) = 0$, if $T_{\text{repump}}/2 < t < T_{\text{repump}}$ (when the APD is gated OFF). The classical autocorrelation function therefore reads as follows

$$G^{(2)}(\tilde{t}) = \int_0^{T_{\text{repump}}} I(t) I(t + \tilde{t}) \, dt = \frac{1}{2} I_{\text{pol}}^2 \tau_b e^{-\tilde{t}/\tau_b} \left( e^{-2\tilde{t}/\tau_b} - e^{-T_{\text{repump}}/\tau_b} \right)$$

for $0 < \tilde{t} < T_{\text{repump}}/2$.

We fit the trailing slope of the central triangle to the formula above, resulting in a blinking time

$$\tau_b (1 \text{ nW}) = (6.2 \pm 1.2) \mu s.$$  \hspace{4cm} (3.14)

In all RS experiments, we did not excite the system with powers much larger than 1 nW. Higher powers would result in a shorter blinking time $\tau_b$. Therefore, a higher re-pump frequency would be required to work with a reasonable neutral-QD ground-state occupation. The re-pump-cycle frequency of our system is limited by the AOM-rise and fall time of about 50 ns (see Section A.2). Using a re-pump
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Figure 3.10.: Measuring the blinking time. Correlation histogram for pulsed-laser excitation of the upper-polariton resonance at an mean power of 1 nW, recorded with the re-pump laser present at a rate of 0.5 MHz and with gated APD-readout mode (see Section 3.2.1). The inset demonstrates that the autocorrelation of periodic square gating-pulses with duty cycle 0.5 results in a triangular-shaped histogram, in analogy to the depicted overlapping area as a function of the delay. The triangle around zero delay is clearly larger than the others, as a result of classical photon bunching due to charge blinking. The red line on top of the histogram is a fit of the slightly curved trailing slope of the zero-delay triangle, while the dashed straight line is shown for reference. The vertical axis specifies the total photon coincidences time-integrated over a laser pulse.

frequency of 1 MHz, the off-resonant pulses have durations of 250 ns (compare Fig. 3.7). Re-pump frequencies much larger than 1 MHz are therefore impractical.

3.3. Resonant correlation measurements with short pulses

The second-order autocorrelation function $g^{(2)}(\tau)$ provides a direct measure of the statistics of the emitted light (see Section B.4.1). The degree of antibunching or bunching is given by the value at zero time delay $g^{(2)}(0)$. The width of the dip / peak in $g^{(2)}(\tau)$ is determined by the polariton-state lifetime, which is in our case on the order of tens of ps. Since our APDs (SPCM-AQR-14 from PerkinElmer, Inc.) have a much larger timing jitter, they cannot be used to measure $g^{(2)}(0)$ under cw-laser excitation. Instead, state-of-the-art superconducting single-photon detectors (SSPDs) [138, 139] with quantum efficiencies of about 20 % and timing jitters of 40 ps could be considered. However, the detector-count rates would simply be too low in the case of cw excitation at 1 nW: If we record a correlation histogram with
3.3. Resonant correlation measurements with short pulses

time channels of width $\delta t = 25$ ps (corresponding to the polariton lifetime), the count rate of a single time channel is given by

$$f_{\text{ch}} = f_1 f_2 \delta t,$$

(3.15)

where $f_1$ and $f_2$ are the photon-count rates of the two detectors in the Hanbury-Brown and Twiss setup. They are at most $f_1 = f_2 \approx 5000$ s$^{-1}$ in our system, when the laser is resonant with a polariton state, resulting in $f_{\text{ch}} \approx 6 \cdot 10^{-4}$ s$^{-1}$. The observation of non-Poissonian photon statistics is indicated by the photon-count rate of the time channel at $\tau = 0$ ($G^{(2)} (\tau = 0)$) being either smaller or larger than the count rates of the other channels. In order to obtain a 10 % confidential result for $G^{(2)} (\tau = 0)$, an average number of counts per time channel of at least 100 is required. Using Equation 3.15, the corresponding integration time would thus be $\approx 44$ hours.

In order to overcome this limitation, we measure the autocorrelation function by exciting the system with short resonant pulses with a pulse duration similar to the polariton lifetime (see e.g. the correlation traces in Figures 4.2(b-e)). We define $g^{(2)}_{\text{pulsed}} (0)$ as the area of the central peak divided by the average area of peaks at other times. Correlations with statistical significance hence require a lower total number of photon coincidences and can be recorded in a shorter period of time. It should be mentioned here that the blinking effect occurs on much longer timescales than the pulse repetition time of 13.1 ns: This becomes intuitive when considering that about 100 polariton-scattering events occur on average, before the QD falls into an off-resonant state (according to calculations in Section 3.2). If the system is excited with laser pulses that contain one or less photons per pulse on average ($\approx$ nW excitation level), the blinking time $\tau_b$ is expected to be on the $\mu$s scale.

Consequently, the spectrum measured upon pulsed resonant excitation is expected to be very similar to the three-peak structure recorded with cw excitation (Fig. 3.8) for a comparable average power. This is confirmed in the experiment, as shown in Figure 3.11.

Photon coincidences thus occur at times corresponding to $N \cdot T_{\text{rep}}$, with $T_{\text{rep}}$ being the elapsed time between two laser pulses and $N$ being an integer (see e.g. Fig. 4.2(c)). In this case, $G^{(2)}_{\text{pulsed}} (\tau)$ is defined by the integrated count rate of a whole pulse. Thus, $G^{(2)}_{\text{pulsed}} (\tau = 0)$ describes the rate at which a single pulse generates a scattered photon pair. Using a MIRA 900 laser from Coherent® Inc. (see Section A.1) for the generation of laser pulses with $T_{\text{rep}} = 13.1$ ns (still at an average excitation power of 1 nW), the integrated coincidence rate of a pulse is

$$f_{\text{pulse}} = f_1 f_2 T_{\text{rep}},$$

(3.16)

and thus orders of magnitudes larger than the coincidence rate of a time channel in cw excitation. For photon-count rates of $f_1 = f_2 \approx 5000$ s$^{-1}$, the coincidence rate is $f_{\text{pulse}} \approx 0.33$ s$^{-1}$ and a histogram can be recorded in a few minutes.

In the case of two-color spectroscopy (see Chapter 5), a pulsed excitation scheme is preferred to cw excitation as well: The light of a control laser lifts the system onto an excited-energy state whereas a second probe laser is used to investigate a higher system eigenstate. To efficiently do so, the control laser should operate close to the power at which the average excited-state population is 1 – which corresponds
Figure 3.11.: Pulsed RS spectra. The center wavelength of the laser pulses is scanned by rotating the mirror that couples the pulses into a single-mode fiber after leaving the grating spectrometer (see Section 5.2). The black trace is recorded without the re-pump laser and the red trace with the re-pump laser at 1 MHz repetition frequency. The mean probe power is 2 nW and the pulse duration 86 ps.

to 860 nW in the case of polaritons with 25 ps lifetime (see Equation 3.1). At a cw power of 1 nW, the polariton population is negligible – making two-color spectroscopy too inefficient.

If, on the other hand, short resonant pulses are used, larger polariton populations at moderate average excitation powers can be achieved.

3.3.1. Resonant-pulse generation

In experiment, the spectral linewidth of resonant pulses should be clearly smaller than the QD-cavity coupling constant $g \left( \frac{\lambda^2}{2\pi} g \approx 0.1 \text{ nm} \right)$. Otherwise, optical transitions cannot be addressed individually. We use close to Gaussian pulses with a full width at half maximum (FWHM) linewidth between 0.085 nm and 0.018 nm, corresponding to (Fourier-limited) pulses of durations between 15 ps and 72 ps.

The laser pulses are derived from a mode-locked Ti:sapphire laser (MIRA 900 from Coherent® Inc.; see Section A.1) with a pulse-repetition rate of 76.3 MHz and an intrinsic pulse duration of 3 – 4 ps. The center wavelength of these broadband pulses is approximately matched with the desired wavelength in a first step. The pulses are then sent through a grating spectrometer (Acton SP2750 spectrograph from Princeton Instruments) in the monochromator mode (see Figure B.1(b)) with both, entrance and exit slits fully open. After passing the monochromator, the light is coupled into a single-mode optical fiber (P3-830A-FC-5 from Thorlabs) for spectral filtering, as illustrated in Figure 3.12. A slit in front of the spectrometer adjusts the effective NA of the beam entering the monochromator to gain control over the spectral bandwidth that is coupled into the fiber. The center wavelength of the generated pulses is recorded with a wavelength meter (Angstrom WS Ultimate 30 MC4 from High Finesse GmbH) and adjusted by rotating the grating of the spectrometer. To check the filtered spectrum, we sent the light of a tunable cw laser through the setup and recorded the transmitted power. Figure 3.13(a) shows the
3.3. Resonant correlation measurements with short pulses

spectra, when the slit is fully open (blue trace) and partly closed (green and red traces). For a fully open slit, the overall transmission of the center wavelength is about 30%. Pulse shapes and delays are monitored by sending the light reflected from the sample surface to a streak camera (C5680 from Hamamatsu Photonics K.K.) with approximately $2 - 4$ ps time resolution. Figure 3.13(b) shows the temporal profile of the pulses corresponding to the filters applied in Fig. 3.13(a). Pulse durations with FWHM between 19 and 86 ps are measured.

![Diagram of grating spectrometer](image)

**Figure 3.12:** Tailoring short pulses of $19 - 86$ ps duration. A broadband laser pulse of 0.5 nm linewidth corresponding to $3 - 4$ ps duration, is first sent through a grating monochromator for color dispersion and subsequently coupled into a single-mode optical fiber that is mode-matched with part of the beam corresponding to a tiny spectral window of about 0.015 nm. If the beam is artificially cropped using a variable slit in front of the monochromator entrance, the NA of the beam entering the monochromator is decreased, resulting in a partial illumination of the diffraction grating. Consequently, the spectral resolution drops, whereby the bandwidth of the fiber-coupled light increases up to 0.04 nm.

For investigating our single-QD cavity system, on the one hand we require a narrow pulse spectrum in order to selectively address optical transitions, but the laser pulses should be short to avoid multiple photon-scattering events for correlation measurements on the other hand. Using Fourier-limited pulses, the relation between temporal and spectral widths, $\text{FWHM}_T$ and $\text{FWHM}_\lambda$, is given by

$$\text{FWHM}_T \cdot \text{FWHM}_\lambda = \frac{2 \ln(2) \cdot \lambda^2}{\pi c} \approx 1.29 \text{ ps} \cdot \text{nm} \quad (3.17)$$

for a wavelength $\lambda = 937$ nm. To check how close we come to this limit, we calculated the Fourier-limited pulse durations (red open circles in Figure 3.13(c)) from the measured spectral widths (blue filled circles), using Equation 3.17. The comparison with the actually measured pulse durations using the streak camera (red filled circles) shows that the deviation is small, especially for short pulses below 50 ps and for the slit fully open. The maximum deviation from the ideal Fourier-limited pulse durations is about 30%.
Figure 3.13: Characterizing the filtered pulses. (a) Filtered spectrum of wavelength-tuned cw-laser light guided through the monochromator and coupled into a SM fiber. The blue trace is taken with the slit fully open, the green curve with a slit width of 0.5 mm and the red curve with a width of 0.2 mm. The spectra are approximately Gaussian, but exhibit small side lobes due to diffraction at the rectangular slit opening. (b) Temporal intensity profile of initially 3 – 4 ps long pulses, after sending them through the monochromator with the slit openings as described in (a). The traces, recorded with a streak camera, show a clear reduction of the pulse duration, as the spectrum is broadened by closing the slit. (c) Performance of the pulse filtering technique. The blue data points show the FWHM$_\lambda$ of the transmitted spectrum as a function of the slit width. The red open circles depict the theoretically expected Fourier-limited pulse duration (FWHM$_T$). The red filled circles display the measured pulse durations.
4. Strongly correlated photons on a chip

In this chapter, I describe our findings of strong quantum correlations between photons scattered off a strongly-coupled QD-cavity system. We observe (a) photon antibunching upon resonant excitation of the lowest-energy polariton state, proving that the first cavity photon blocks the subsequent injection of a photon of the same color, and (b) photon bunching, when the laser field is in two-photon resonance with the polariton eigenstates of the second Jaynes-Cummings manifold. The main findings presented in this chapter are published in Nature Photonics [61].

4.1. Strong photon-photon nonlinearities

A single quantum emitter constitutes the ultimate nonlinear system, as witnessed by the so-called photon-blockade effect: By absorbing a photon the system makes a transition into an excited state. Due to the anharmonicity of the energy diagram (see Figure 4.1), a second photon of the same color is blocked and cannot be scattered off the system. Even if the emitter is driven by a photon stream with Poissonian statistics (e.g. by a narrow-bandwidth laser), only one photon at a time can be absorbed. Consequently, a train of single photons is re-emitted by the system and the photon stream is antibunched. This behavior can be confirmed by second-order correlation measurements with a HBT setup (see Section B.4.1). Photon-correlation measurements have therefore emerged as an indispensable tool in many experiments conducted in our group to test the quantum nature or dynamics of a given system [7, 68, 100, 140–143].

The photon-blockade effect was demonstrated in the optical domain using a single atom coupled to the modes of Fabry-Pérot [12] or toroidal cavities [13] by observing photon antibunching in correlation measurements and in the microwave domain using superconducting qubits coupled to stripline resonators [14]. Using semiconductor-quantum dots, early results on non-classical light generation were reported [15, 22]. However, the observed quantum correlations were rather weak.

In the device studied in this thesis, the strong coupling of a single quantum dot to the field of a single photonic-crystal cavity mode clearly results in the anharmonic Jaynes-Cummings energy-level diagram (see Fig. 4.1). Transition energies from the ground state $|g\rangle$ to the first Jaynes-Cummings manifold ($|1, +\rangle$, $|1, -\rangle$) are distinct from transition energies higher up in the ladder (see Section 2.1.2). As a consequence, the system is intrinsically nonlinear at the single-photon level.
**Figure 4.1.** Resonant-scattering spectroscopy of a single emitter strongly-coupled to a cavity mode. The Jaynes-Cummings level scheme is shown up to the second manifold: The kets $|n, +/−⟩$ denote the upper / lower-polariton states of the $n^\text{th}$ manifold, where $n$ is the number of energy quanta in the system. When exciting a state of the first manifold with a resonant laser (blue arrows), the second manifold cannot be accessed (photon blockade). For a slight blue-detuning of the laser wavelength however, the second Jaynes-Cummings manifold can be excited via a two-photon resonance (red arrows).

### 4.2. Photon blockade and a two-photon resonance

We carried out correlation measurements (see Section B.4.1) using resonant pulses at a FWHM duration of $T_{\text{pulse}} \approx 72 \text{ ps}$ (see Section 3.3) and APDs (SPCM-AQR-14 from PerkinElmer, Inc.). Figure 4.2 displays correlation histograms for different laser-wavelength / cavity-mode detunings$^1$. The respective detunings are indicated in the RS spectrum shown in Fig. 4.2(a). We define the correlation function for pulsed excitation at zero time delay $g^{(2)}_{\text{pulsed}}(0)$ as the area of the central peak divided by the average area of peaks at other times (see Section 3.3). First, we chose the frequency of the pulsed laser to match the upper-polariton transition and obtained significant antibunching with $g^{(2)}_{\text{pulsed}}(0) = 0.75 \pm 0.06$ (Fig. 4.2(c)). For comparison, we turned off the re-pump laser and verified that photons scattered from the uncoupled-cavity mode had Poissonian statistics (Fig. 4.2(b)). As a cross-check, we also confirmed that the scattered photons from a resonantly driven cavity mode that was far detuned from all QD transitions exhibited Poissonian statistics. These experiments jointly demonstrate that the applied re-pump laser can be used to switch on the single-photon nonlinearity and to control the statistics of the scattered photons.

One of the principal limitations in the experiment is the pulse duration being longer than the polariton lifetime. The ratio between pulse duration and polariton lifetime is $T_{\text{pulse}} \cdot \frac{\kappa}{\bar{\gamma}} \approx 2.9$, enabling multiple-photon absorption events within a

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$^1$In the whole chapter, detunings are specified in terms of the wavelength; i.e. a positive detuning refers to a detuning towards longer wavelengths.
4.2. Photon blockade and a two-photon resonance

single pulse. Accordingly, a higher degree of antibunching on the lower polariton 
\( g^{(2)}_{\text{pulsed}}(0) = 0.55 \pm 0.03 \) is observed, when the cavity mode is blue-detuned from 
the exciton resonance, as shown in Figure 4.2(d); here the polariton-state lifetime 
is prolonged, making multiple-photon absorption events within a single pulse less 
likely. Experimentally, we do not observe a difference between correlations recorded 
on the upper and lower-polariton resonance, if the respective laser-wavelength and 
cavity-mode detunings from the exciton change their sign.

When tuning the laser-photon energy to half the energy of the lower-polariton 
eigenstate of the second Jaynes-Cummings manifold, we observe photon-bunching. 
Figure 4.2(e) displays a corresponding correlation histogram with 
\( g^{(2)}_{\text{pulsed}}(0) = 1.5 \pm 0.1 \). The bunching has its origin in a two-photon transition from the ground 
state to the second Jaynes-Cummings manifold [21, 144]. We emphasize that the 
non-vanishing correlations at zero time delay in the case of photon blockade, and 
the moderate bunching-feature on the two-photon transition have their origin mostly 
in the particular implementation of the measurement using pulsed-laser excitation 
and slow single-photon detectors (see Section 3.3); in order to confirm that and to 
explain the principal experimental features, we carried out numerical simulations 
of \( g^{(2)}_{\text{pulsed}}(0) \) using a Monte Carlo wavefunction (MCWF) approach (see Ref. [145] 
and Section 4.3). Here, we directly accounted for the pulsed-laser excitation, the 
non-zero laser background due to imperfect extinction of the laser reflection, and the 
uncoupled cavity-mode resonance due to the blinking of the dot (see Section 3.2). 
For the experimental settings corresponding to Figures 4.2(c),(d) and (e), we estimate 
the temporal maximum of the average number of energy quanta in the Jaynes- 
Cummings system to be 0.12, 0.086 and 0.020, based on these MCWF simula-
tions. Fig. 4.3(a) displays the calculated autocorrelation function at zero time delay 
\( g^{(2)}_{\text{pulsed}}(0) \) for varying cavity-mode and laser-wavelength detunings. The blue and red 
dashed lines denote the expected positions of the polariton frequencies and the two-
photon resonances, respectively. The latter correspond to the optimal second-rung 
excitation conditions.

In Fig. 4.3(b) and (c), we probed the upper and lower-polariton branches for con-
stant cavity-mode detunings of \((-0.33 \pm 0.04) g \) and \((1.04 \pm 0.03) g \), respectively, 
by tuning the laser wavelength. The continuous change in photon correlations from 
strong antibunching to bunching maps out the (anharmonic) spectrum of the Jaynes-
Cummings ladder. In addition, we performed correlation measurements for a vary-
ing cavity-exciton detuning at a constant laser-exciton detuning of \((0.94 \pm 0.05) g \), 
demonstrating that the nature of strong photon correlations can be tuned by chang-
ing either the laser or the cavity-mode frequency. The dashed vertical lines in 
Figures 4.3(b)–(d) correspond to the polariton energy (blue) and the energy of the 
two-photon transition to the second manifold (red), indicating the origin of both 
the antibunching and the bunching features.
Figure 4.2.: From Poissonian light to antibunched and bunched photon streams. (a) RS spectra for different cavity-mode detunings close to the neutral-exciton resonance. The circles indicate the detunings for which the correlation histograms in (b)–(e) were taken. The vertical axes in the histograms specify the photon coincidences per time channel of 192 ps. (b) Autocorrelation histogram recorded on exact cavity-exciton resonance without applied re-pump laser for a probe-laser wavelength corresponding to the upper-polariton resonance. (c) Autocorrelation with the same detunings, but with a re-pump laser applied. The scattered photons in this case exhibit photon antibunching. (d) Autocorrelation on the lower-polariton resonance for a blue-detuning of the cavity mode of $-0.68 \, g$, where the longer polariton lifetime ensures stronger photon antibunching. (e) Pronounced photon-bunching on the lower two-photon resonance. Note that the small features at $\approx \pm 10 \, \text{ns}$ in (b)–(e) originate from cross-talk between the APDs, due to secondary photon-emission events.
Figure 4.3.: Calculated and measured autocorrelation functions $g_{\text{pulsed}}^{(2)}(0)$ for different cavity-mode and laser-wavelength detunings. (a) Results of a MCWF simulation close to the neutral exciton-cavity resonance using experimental parameters obtained from resonant-spectroscopy data (compare Fig. 3.8). The blue regions correspond to detunings with sub-Poissonian statistics, whereas the red regions indicate photon-bunching. (b)–(d) Measured $g_{\text{pulsed}}^{(2)}(0)$ for different cavity-mode and laser-wavelength detunings, as indicated by the bold-dotted lines in (a). The red dotted traces are the theoretical expectations from the MCWF simulations.
4.3. Theoretical estimate of correlations

In order to have a realistic comparison between theory and our experimental data, we use a MCWF approach to obtain an estimate of \( g_{\text{pulsed}}^{(2)}(0) \). The previously defined pulsed correlations can be written as

\[
g_{\text{pulsed}}^{(2)}(0) = \frac{2}{T_{\text{rep}}/2} \int_{-T_{\text{rep}}/2}^{T_{\text{rep}}/2} dt_1 \int_{-T_{\text{rep}}/2}^{T_{\text{rep}}/2} dt_2 \ G^{(2)}(t_1, t_2) I(t_1) I(t_2),
\]

where \( T_{\text{rep}} \) is the time difference between two laser pulses. Here, \(|g\rangle\) denotes the system ground state, before a laser pulse has excited the system. The cavity-photon collapse operator \( \hat{C}(t) = \sqrt{\kappa} \hat{a}(t) \) evolves according to the non-Hermitian effective Hamiltonian [145] (in the interaction picture with the rotating frame of the laser center frequency)

\[
H_{\text{eff}}(t) = H_{\text{JC}} + H_{\text{int}}(t) - \frac{i\hbar}{2} \left( \kappa \hat{a}^\dagger \hat{a} + \gamma \hat{\sigma}_+ \hat{\sigma}_- + \gamma_{\text{deph}} \right),
\]

where \( \hat{a} \) is the cavity-photon annihilation operator and \( \hat{\sigma}_+, \hat{\sigma}_- \) are the exciton creation and annihilation operators. \( H_{\text{JC}} \) is the Jaynes-Cummings Hamiltonian and \( H_{\text{int}} = \frac{\hbar \Omega(t)}{2} \left( \hat{a} + \hat{a}^\dagger \right) \) denotes the interaction with the Gaussian laser pulse,

\[
\Omega(t) = \Omega_0 \exp \left( -2 \ln(2) t^2 / T_{\text{pulse}}^2 \right).
\]

\( \kappa \) denotes the cavity-photon dissipation rate and \( \gamma \) the exciton spontaneous-recombination rate. The term \( \gamma_{\text{deph}} \hat{\sigma}_+ \hat{\sigma}_- = \gamma_{\text{deph}} \) in Equation 4.2 describes the pure exciton-dephasing rate, discussed in Section 3.2.1. In order to determine \( G^{(2)}(t_1, t_2) \) and \( I(t) \), we use a Monte-Carlo wavefunction approach, as described in Ref. [145]. The calculations are performed with the experimentally determined values, \( \lambda_{\text{exciton}} = 937.25 \text{ nm}, \hbar g = 141 \mu\text{eV}, \hbar \kappa = 53 \mu\text{eV}, \hbar \gamma = 0.66 \mu\text{eV}, \) and \( \hbar \gamma_{\text{deph}} = 13 \mu\text{eV} \). For a typical pulse duration of 72 ps (FWHM), a total pulse energy of 82 eV and an estimated laser-cavity coupling efficiency of \( \eta \approx 2\% \), we obtain \( \hbar \Omega_0 \approx 24 \mu\text{eV} \).

In the experiments, the suppression of the non-interacting reflected laser light is not perfect (\( \approx 1:50000 \)), and consequently, our measurements are subject to interference effects between cavity-photon emission and laser light (see Section 3.1.1). Possible reasons for incomplete suppression are a mixing of polarization states, when the light traverses the collection fiber or the analyzer, or depolarization effects, due to the reflection at the photonic-crystal membrane. In the simulations, we superimpose the detected optical field with a laser-background term. The phase between the laser light that drives the Jaynes-Cummings system and the non-interacting (reflected) laser light is chosen such that the (experimentally optimized) ratio between
4.3. Theoretical estimate of correlations

detected cavity photons and laser background is the largest. We emphasize that we have a control over both the amount of laser suppression and the phase difference, by the use of fiber-polarization controllers.

As described in Section 3.2.1, the system is in the desired neutral ground state (on-state) in only \( r = 30 - 40 \% \) of the cases on average and in an off-resonant ground state (off-state) otherwise. If the system is in the on-state, the coincidence rate of the center peak is \( G_{\text{on}}^{(2)} (0) = r I_{\text{on}}^{\text{g}} g_{\text{pulsed}}^{(2)}(0) \), whereas the coincidence rate of an off-center peak (e.g. \( \tau = T_{\text{rep}} \)) of the correlation function is given by \( G_{\text{on}}^{(2)} (T_{\text{rep}}) = r I_{\text{on}}^{\text{g}} g_{\text{pulsed}}^{(2)} \). In contrary, if the system is in the off-state, the coincidence rate is given by \( G_{\text{off}}^{(2)} (0) = G_{\text{off}}^{(2)} (T_{\text{rep}}) = (1 - r) I_{\text{off}}^{2} \).

Experimentally, we cannot distinguish between on- and off-states, so the resulting correlation function is a sum of both on-resonant and the Poissonian off-resonant correlation functions. Thus, at zero time delay

\[
g_{\text{pulsed}}^{(2)} (0) = \frac{G_{\text{on}}^{(2)} (0) + G_{\text{off}}^{(2)} (0)}{G_{\text{on}}^{(2)} (T_{\text{rep}}) + G_{\text{off}}^{(2)} (T_{\text{rep}})} = \frac{r I_{\text{on}}^{\text{g}} g_{\text{pulsed}}^{(2)} (0) + (1 - r) I_{\text{off}}^{2}}{r I_{\text{on}}^{\text{g}} + (1 - r) I_{\text{off}}^{2}}, \tag{4.4}
\]

where \( I_{\text{on}} \) is the average detected energy per pulse of the Jaynes-Cummings system, as described above, and \( I_{\text{off}} \) the respective pulse energy of the uncoupled-cavity emission, together with the non-interacting laser-light background. For the rest of the discussion here, and in the whole chapter, we will omit the tilde and refer to Equation (4.4) as the definition of correlations at zero time delay.

As previously discussed, a limitation in the experiment is the pulse duration being longer than the polariton lifetime. Simulations of the correlation function, when a laser probes a polariton resonance, reveal that \( \left| g_{\text{pulsed}}^{(2)} (0) - 1 \right| \) is reduced, if a pulsed laser (instead of a cw laser) is used. The reasons are repeated photon-absorption / re-emission events within the duration of a single laser pulse, excitations of more than one resonance, due to the broad pulse spectrum, and the large laser-peak power with \( \Omega_{0} \lesssim \kappa \). For Fourier-limited Gaussian laser pulses, we find a minimum of \( g_{\text{pulsed}}^{(2)} (0) = 0.75 \) for a laser pulse duration of about 50 ps, as shown in Figure 4.4(a).

In the experiments, we measure the spectrally filtered pulses to have slightly larger durations of \( T_{\text{pulse}} \approx 72 \) ps. This corresponds to a ratio between pulse duration and polariton lifetime of \( \frac{T_{\text{pulse}}}{\kappa} \approx 2.9 \). For increasing pulse energy, the calculated \( \left| g_{\text{pulsed}}^{(2)} (0) - 1 \right| \) decreases, as shown in 4.4(b). We typically work with pulse energies between 40 and 100 eV, which corresponds to \( 0.6 - 1.5 \) scattered photons per pulse on laser-cavity resonance for the estimated coupling efficiency of \( \eta \approx 2 \% \).

The efficiency of the generation of non-classical light improves with a slight cavity-exciton detuning. A blue-detuning of the cavity mode by \( g \) with respect to the exciton resonance yields \( g_{\text{pulsed}}^{(2)} (0) = 0.63 \) when probing the lower-polariton resonance. This reduction in \( g_{\text{pulsed}}^{(2)} (0) \) originates predominantly from the prolongation of the polariton lifetime, which reduces the likelihood of multiple-photon absorption events within a single laser pulse.
Figure 4.4.: Simulation results of $g_{\text{pulsed}}^{(2)}(0)$ for different pulse parameters. In both panels (a) and (b), blue traces indicate probing of the lower polariton and red traces the lower two-photon resonance. (a) $g_{\text{pulsed}}^{(2)}(0)$ as a function of pulse duration for a fixed pulse energy of 82 eV. (b) $g_{\text{pulsed}}^{(2)}(0)$ as a function of pulse energy for 72 ps pulses.

4.4. Probing the exciton resonance

In addition to the bunching features originating from two-photon resonances, we would normally expect strong bunching, when the external laser that drives the cavity mode is resonant with the bare exciton [21], due to a quantum interference based on an electromagnetically induced transparency (EIT) effect that appears, when the system is weakly probed via the cavity mode (at a laser-cavity coupling strength that is much smaller than the cavity-exciton coupling strength $g$). Since the quantum interference only appears on the transition from the ground state to a fundamental-polariton state, whereas the photon-scattering rate on higher-order Jaynes-Cummings transitions remains finite, the detected photon stream is expected to be strongly bunched.

For small cavity-exciton detunings ($\lesssim$ 0.5 $g$), and for negligible QD blinking, a MCWF simulation reveals a pronounced bunching peak, when the laser frequency matches the one of the bare exciton (see Figure 4.5(a)).

Unlike in previous experiments [15], we do not observe a bunching behavior. In the real system exhibiting blinking we measure $g_{\text{pulsed}}^{(2)}(0) \approx 1$ (see simulations in Figure 4.3(a) and Fig. 4.5(b)), due to the predominant detection of photons scattered from the uncoupled-cavity mode.

In the case of larger cavity-exciton detunings ($\gtrsim$ 0.5 $g$), the influence of the uncoupled-cavity mode resonance is weaker, and we expect correlations along the zero laser-exciton detuning line to exhibit bunching, as predicted by the simulations shown in Fig. 4.3(a). Experimentally, the count rate is very low (due to the EIT-type quantum interference) and we do not have sufficient coincidences to obtain reliable results for $g_{\text{pulsed}}^{(2)}(0)$. 
4.4. Probing the exciton resonance

Figure 4.5.: Results from MCWF simulations for cavity-exciton resonance. (a) Assuming that there is no QD-blinking, the spectrum of the strongly-coupled system exhibits only the two polariton peaks (black line). There is a strong bunching peak for zero laser-exciton detuning in $g_{\text{pulsed}}^{(2)}(0)$ (red line). (b) Simulations of the real system showing QD-blinking ($r = 0.4$) yield a (time-averaged) spectrum with a three-peak feature (black line); the uncoupled-cavity mode peak in the middle causes the bunching feature in $g_{\text{pulsed}}^{(2)}(0)$ (red line) to vanish.

In conclusion, our experiments demonstrate that a strongly-coupled QD-cavity device realizes the anharmonic Jaynes-Cummings model in the solid state. These results elevate the system to the ultimate optical nonlinear building block in more complex structures that could be used to investigate strongly-correlated photonic systems in non-equilibrium settings, such as an optical Josephson interferometer [146] or coupled arrays of nonlinear cavities [24, 25]. Apart from these prospects, I will demonstrate in the next chapter that our device as a single unit can be used to switch photons of one color with a single photon of a different color.
5. Ultrafast all-optical switching by single photons

In this chapter, I present our results from pulsed two-color spectroscopy and demonstrate that in a strongly-coupled QD-cavity system the presence of a single photon on one of the fundamental-polariton transitions can turn on light scattering on a transition from the first to the second Jaynes-Cummings manifold. We demonstrate the overall switching time of this single-photon all-optical switch to be about 50 ps. In addition, we use the single-photon nonlinearity to implement a pulse-correlator. The main findings presented in this chapter are published in Nature Photonics [62].

5.1. A single-photon Jaynes-Cummings switch

In the previous chapter, we demonstrated the photon-blockade effect on our single-QD cavity system proving that laser photons resonant with a fundamental-polariton transition enter the cavity only one at a time preventing the population of manifolds lying higher than the first one. However, when photons of a second laser at a color that matches the energy difference between the levels of the first and second rung are injected into the system simultaneously (see Fig. 5.1(a)), scattering into the second manifold is possible provided a photon is absorbed on a fundamental-polariton transition. As a result, the device constitutes a single-photon switch where a single optical gate (or control) photon controls the propagation (or scattering) of incident signal photons through nonlinear-optical interactions, as illustrated in Figure 5.1(b).

Single-photon switches / transistors were realized in the optical domain, based on surface plasmons [16] or single molecules [17] strongly-coupled to a light field and in the microwave domain, by coupling a superconducting qubit to a stripline resonator [8]. In a GaAs-based single-QD cavity system, a switch triggered by low photon numbers was demonstrated [18]. However, the speed of devices that are based on single-photon nonlinearities were not addressed in those experiments. Since the ultimate switching times are limited by the reciprocal emitter-cavity coupling strength, quantum dots strongly-coupled to nanocavities [3–5, 7] emerge as good candidates for the realization of ultrafast single-photon nonlinear devices, due to their record-high coupling strengths.

Our QD-cavity system is well in the strong-coupling regime. The figure of merit for the anharmonicity of the coupled system is $\xi = g/\kappa \approx 2.7$ (see Section 2.1.2). Notably, due to the observed pure exciton dephasing (see Section 3.2.1), this number is lowered by a factor of two, but is still larger than unity – which is the fundamental
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5.2. Pulse preparation

In the described experiments in this chapter, two pulses with slightly different center wavelengths and mutual synchronization in time are required. Since the upper and lower polariton states are spectrally separated by about 0.2 nm, the wavelength separation of the two required pulses should be within this range. Fulfilling all these requirements simultaneously can be achieved by sending a single spectrally broad laser pulse through two different narrow-bandpass filters (e.g. using a grating monochromator). The two pulses are intrinsically synchronized and can be separately delayed using mechanical delay stages.

In the experiment, pulse preparation is achieved in the same way as described in Section 3.3.1. Figure 5.2 provides an overview of the optical setup for the preparation of two laser pulses.

The center wavelength of the pulse controlled by the piezo-driven mirror is monitored using a wavelength meter (Ångstrom WS Ultimate 30 MC4 from High Finesse GmbH), and a computer-controlled feedback loop allows for relative tuning of the center wavelengths of the pulses. The average power of both control and signal-laser beams is stabilized using acousto-optic modulators (see Section A.2).

Pulse durations can be tuned by changing the NA of the beam coupled into the spectrometer. The pulse shapes, durations and delays are monitored by sending the
Figure 5.2.: Setup for the preparation of two pulses of different color. A broadband laser pulse of $3 - 4$ ps duration is first sent through a grating spectrometer for color dispersion and subsequently split by a 50 : 50 beam splitter. Both beams are then partially coupled into single-mode optical fibers selecting a small spectral window by spatial mode-matching (see Section 3.3.1 for details). In addition to rotating the grating of the spectrometer for tuning the colors of both pulses simultaneously, a mechanically tunable piezo-driven mirror holder (Motorized Stability™ Mount 8816-6 from Newport) in front of one of the fiber couplers allows for individual spectral tuning. The relative delay between signal and control pulses is adjusted by a continuous delay stage supporting in principle sub-ps time resolution. In addition, a discrete delay line, corresponding to a time delay of $\approx 5$ ns, can be added to the path of the signal pulse using a micro-electro-mechanical system (MEMS) fiber switch (OSW22-830E from Thorlabs). The laser-power stabilization is conducted for both pulses separately (not displayed).

Light reflected from the sample surface to a streak camera with approximately 4 ps time resolution.

5.3. Resonant nonlinear spectroscopy

We first carried out resonant spectroscopy of the strongly-coupled QD-cavity system by scanning the center wavelength of the pulsed laser across the system spectrum, when the QD resonance and cavity mode were very close to resonance, and recording the scattered power with an APD. The resulting count rate $N_{\text{signal}}(\delta)$ as a function of the signal-laser detuning\(^1\) $\delta$ is displayed as open circles in Fig. 5.3(a) for a pulse

\(^1\)In the whole chapter, detunings are specified in terms of the wavelength; i.e. a positive detuning refers to a detuning towards longer wavelengths.
duration of 86 ps and an average signal power of 1 nW. As described in Section 3.2.1, we employed an off-resonant re-pump laser at a repetition rate of 1 MHz to partially counteract the laser-induced quantum-dot blinking present in the system.

Next, we performed pulsed two-color spectroscopy of the strongly-coupled QD-cavity system. We tuned the control-laser wavelength, with average power 2 nW, to the upper-polariton resonance and scanned the signal-laser pulse across the spectrum. Note that the power of the control laser corresponded to about 120 photons per pulse. With a laser-cavity coupling mode matching efficiency $\eta \approx 2 \%$ and the 50 % photon content of the polaritons, this yields on average about 1.2 photons per pulse scattered off the strongly-coupled system. Having demonstrated the photon-blockade effect (see Chapter 4) on both upper and lower-polariton transitions to the first Jaynes-Cummings manifold, we are sure that the switching operation of our device is enabled by the presence of a single photon in the cavity.

For the scan, the delay of the signal pulse with respect to the control pulse was chosen to be $\tau = 25$ ps corresponding to the polariton lifetime. The resulting spectrum $N_{\text{both on}}(\delta, \tau) - N_{\text{control}}$ is displayed in Fig. 5.3(a): the filled (open) circles show the system response with (without) control laser present. The difference between the two data sets $N_{\text{nl}}(\delta, \tau) = N_{\text{both on}}(\delta, \tau) - N_{\text{control}} - N_{\text{signal}}(\delta)$ directly reflects the nonlinear response of the QD-cavity system, with the reduction of the polariton signal being the most obvious effect of the presence of the control laser. We plotted this difference in Fig. 5.3(b) (red data points). In addition to the fast Jaynes-Cummings (subindex JC) photon-photon interactions of interest, the coupled system also exhibits a slow nonlinearity caused by the laser-induced QD blinking (subindex b). Consequently, in the experiment we measure

$$N_{\text{nl}}(\delta, \tau) = N_{\text{nl,JC}}(\delta, \tau) + N_{\text{nl,b}}(\delta, \tau).$$  \hspace{1cm} (5.1)

To distinguish between the two nonlinear effects, we acquired the same spectra as in Fig. 5.3(a), but now with an intermediate time-delay of about $\tau_{\text{int}} \approx 5$ ns between the control and signal pulse. This can be achieved by switching an additional delay line into the path of the signal laser (see Figure 5.2). The difference signal $N_{\text{nl}}(\delta, \tau_{\text{int}})$ is again displayed in Figure 5.3(b) as blue bullets.

Having chosen $\tau_{\text{int}}$ much longer than the polariton lifetime and the laser-pulse durations, but clearly shorter than the blinking time $\tau_b \approx 6.2 \mu$s (see Section 3.2.2), the fast nonlinearity due to the Jaynes-Cummings dynamics vanishes, $N_{\text{nl,JC}}(\delta, \tau_{\text{int}}) \approx 0$ and the slow system nonlinearity due to the blinking effect remains unchanged, $N_{\text{nl,b}}(\delta, \tau_{\text{int}}) \approx N_{\text{nl,b}}(\delta, \tau)$. As a result, the desired Jaynes-Cummings nonlinearity is given by

$$N_{\text{nl,JC}}(\delta, \tau) \approx N_{\text{nl}}(\delta, \tau) - N_{\text{nl}}(\delta, \tau_{\text{int}}).$$  \hspace{1cm} (5.2)

A more detailed description of how we measured this quantity is provided in Section B.3. The resulting fast nonlinear-optical response $N_{\text{nl,JC}}(\delta, \tau)$ from the strongly-coupled QD-cavity system is depicted in Fig. 5.3(c): the data show that the largest nonlinear effect occurs at the spectral position of the polaritons (vertical blue lines). Here, the change in the scattering rate induced by the control laser is negative ($\approx -15 \%$), due to saturation of the corresponding transitions. At the transition wavelength from the first to the second manifold (vertical red line), this
change is positive ($\approx +6\%$), since the absorption of the control photon enables the
subsequent scattering of a signal photon\(^2\). The relative increase in reflection signal is rather moderate, due to the presence of the uncoupled-cavity peak. In the absence of blinking and background laser light, we expect theoretically a switching contrast of +110% (see the discussion about the ON/OFF ratio in Section 5.7).

In order to compare our experimental data with theoretical expectations, we performed numerical simulations based on a MCWF approach [145] with the experimental parameters as input and only the absolute amplitude of the nonlinear signal determined from a least-square fit to the data.

The simulations were performed in the same way as described in Section 4.3, but this time we added a second laser pulse to the Hamiltonian,

\[
\begin{align*}
\Omega(t) &= \Omega_{\text{control}} \exp \left(-i\omega_{\text{control}}t - 2 \ln (2) t^2 / T_{\text{pulse}}^2 \right) + \Omega_{\text{signal}} \exp \left(-i\omega_{\text{signal}}t - 2 \ln (2) (t + \tau)^2 / T_{\text{pulse}}^2 \right). \tag{5.3}
\end{align*}
\]

Since the phase relation between the two laser pulses in the experiment is not known and also unlikely to be preserved during a single measurement, we calculated the MCWF simulations with the different phase relations 0, \(\pi/2\), \(\pi\) and \(3/2\pi\) and averaged over the four cases. In the simulations, a fixed phase relation between the two pulses would result in a false nonlinearity, due to interference effects, when the two pulses overlap in wavelength.

Figure 5.4 depicts the calculated spectral nonlinearity, when the control laser is resonant with the upper polariton and the cavity-mode wavelength is scanned across the exciton transition. Blue regions show negative nonlinearities, red regions correspond to positive nonlinearities.

The excellent agreement between theory (black line in Figure 5.3(c)) and experiment clearly demonstrates that the observed positive nonlinearity is indeed a result of the two-color transition to the second Jaynes-Cummings manifold, ensuring the single-photon nature of the observed nonlinearities. We emphasize here, that due to the finite linewidth of the coupled-system eigenstates as well as the finite bandwidth of the laser pulses, there is some overlap between transitions from the first to the second and from the second to the third manifold and so forth. Hence, we expect a non-negligible contribution to the nonlinear signal stemming from transitions higher up in the Jaynes-Cummings ladder.

In addition to the contribution from the transition of interest, we also observe a non-negligible positive response at slightly positive detunings (see Fig. 5.3(c)) that we attribute to pure exciton dephasing (discussed in Section 3.2.1). A consequence of exciton dephasing is a population exchange of the upper and lower-polariton states. Thus, as we excite the upper-polariton resonance with the control laser, also the lower-polariton state is populated occasionally. As a result, the signal laser can subsequently inject a photon via the transition from the lower fundamental polariton to the lower polariton of the second Jaynes-Cummings manifold. The additional positive nonlinear response at \(\approx +0.3\ g\) in Fig. 5.3(c) most likely has its origin in

\(^2\)Note that we probed only the transition \(|1, +\rangle \rightarrow |2, +\rangle\). The transition \(|1, +\rangle \rightarrow |2, -\rangle\) is expected to appear at a signal laser detuning of +2.5 \(g\). However, this transition is hard to detect, since the scattering rate is reduced by a factor of \((\sqrt{2} - 1)^4 \approx 0.03\), compared to the transition \(|1, +\rangle \rightarrow |2, +\rangle\).
5.4. Ultrafast nonlinear response

In order to demonstrate ultrafast switching by single photons, i.e. conditional scattering of signal photons on ultrafast timescales, we varied the delay between control and signal pulses (see Fig. 5.6(b)) while recording the (positive) nonlinearity for fixed laser-detunings. As depicted in Fig. 5.1(a), we chose the control-laser pulse to be resonant with the fundamental upper-polariton transition and the signal pulse to be resonant with the transition from the first to the second manifold. The result is plotted in Figure 5.6(a) for a pulse duration of 33 ps; the black dots depict experimental data, while the red curve was obtained from a MCWF simulation with the absolute amplitude extracted from a least-square fit to the data. Note that the nonlinearity is displayed as the relative increase / decrease of the detected signal-

Figure 5.4.: Monte-Carlo simulation of the nonlinear system response as a function of cavity-mode detuning and laser center-wavelength detuning. For each spectrum (horizontal traces), the center wavelength of the control laser is chosen to be resonant with the upper-polariton transition and excites it at a mean power of 2 nW. The probe laser has a mean power of 1 nW and is delayed by 25 ps with respect to the control pulse. Both laser pulses have a duration of 86 ps. The color scale indicates the average number of additionally scattered signal photons per pulse, due to the presence of the control laser. The black dotted line shows the spectral position of the uncoupled-cavity mode. The regions with the largest nonlinearities appear about $0.4 \, g$ blue-detuned with respect to the uncoupled-cavity mode resonance.

this mechanism. As a countercheck, we performed a MCWF simulation for the same system, but without pure exciton dephasing. The result is shown in Figure 5.5 and clearly demonstrates that the positive nonlinearities at positive detunings vanish, if there is no pure exciton dephasing present.

5.4. Ultrafast nonlinear response
Figure 5.5.: Monte-Carlo wavefunction simulation of the system dynamics with the experimental parameters as input. The corresponding experimental data that exhibit pure dephasing are shown in Fig. 5.3(c) and resemble very well the black trace, calculated with the QD pure-dephasing rate $\hbar \gamma_{\text{deph}} = 13 \mu eV$. The red curve shows the nonlinearity, when no pure dephasing is present. The cavity-mode detuning is $-0.11 g$.

... photon scattering rate as compared to the case without control laser present. As the data shows, our single-photon switch combines both ultrafast turn-on and turn-off times, which is typically hard to achieve in other quantum-emitter-cavity systems. The sharp turn-on of the nonlinear system response around zero time delay is directly related to the parameters of the laser pulses. We find that in this particular case the corresponding turn-on time (the time the signal takes to rise from 10 % to 90 % of the maximum) is about 20 ps. This is very close to the ultimate switch-on time, which is limited by the anharmonicity of the Jaynes-Cummings spectrum, and is given by $\left(2 - \sqrt{2}\right) g^{-1} \approx 20$ ps. For shorter pulse durations the spectral selectivity of the control laser and therefore the performance of the switch would degrade.

At delay times longer than $\approx 30$ ps, the nonlinear signal of Fig. 5.6(a) exhibits a fast decay with a 1/e-time of about 30 ps, close to the polariton lifetime of 25 ps, which in turn sets the fundamental limit for the turn-off time of our device. The asymmetry in the pulse-delay dependence is a direct consequence of the cascaded nature of the underlying two-photon transition (see Figure 5.1(a)) and most pronounced for pulse durations on the order of the polariton lifetime. Note that both experimentally and theoretically, the nonlinear signal is negative for probe-pulse delays between $-70$ ps and $-20$ ps. An explanation is found in a finite overlap of the spectrum of the (earlier arriving) signal pulse with the transition $|g\rangle \rightarrow |1, +\rangle$, leading to a non-zero excitation probability of the fundamental upper-polariton state. Photons of the later arriving control pulse therefore have a reduced probability to scatter on the $|g\rangle \rightarrow |1, +\rangle$ transition, yielding a negative nonlinearity.

Besides the switching speed, a quantity of interest is the transfer characteristic, i.e.
5.4. Ultrafast nonlinear response

Figure 5.6.: Ultrafast nonlinear response. (a) Measured nonlinear response of the QD-cavity system (black points) as a function of delay between control and signal-laser pulses for a FWHM of the pulses of 33 ps. Here, the nonlinearity is given as the relative increase / decrease of the detected photon-scattering rate compared to the case without control laser. The red curve was obtained from a numerical simulation using a Monte-Carlo wavefunction approach with the experimental parameters as input. (b) By the use of a fiber-coupled delay stage, the control pulses are temporally shifted with respect to the probe pulses. (c) Transfer characteristic of the single-photon switch. The nonlinear signal was recorded as a function of control power for a signal-pulse delay of 25 ps. For average powers larger than 1 nW, the system saturates. The dashed line is a guide to the eye. The error bars shown on the first datapoints in (a) and (c) denote photon-shot noise.

The output signal as a function of control power, which is plotted in Fig. 5.6(c). Here, we recorded the number of signal photons scattered from the system as a function of input power of the control pulse. As expected, we first observe a linear signal increase with control-laser power and then saturation, when the average polariton number reaches $\approx 0.5$. 
5.5. A single-photon pulse correlator

Besides the realization of single-photon switching, the strong nonlinearity of the QD-cavity system can be applied for measuring pulse durations of ultrafast optical pulses with pulse energies down to the single-photon level. Pulse-correlator operation is demonstrated in Figure 5.7, where we mapped out the nonlinear system response as a function of pulse delay for pulse durations of 51 ps and 86 ps. Since the pulse durations are significantly longer than the polariton lifetime of 25 ps, the system response is more symmetric than in the case of the 33 ps pulses of Figure 5.6(a). We find good agreement between the pulse-delay-dependent nonlinearity and the numerical convolutions of the independently obtained streak-camera images (red lines, recorded using a C5680 streak camera from Hamamatsu Photonics K.K.). Figure 5.7(c) compares the FWHM of the nonlinear system response with the FWHM of the incident (Gaussian) laser pulses. The open circles indicate the pulse durations calculated with Monte-Carlo simulations and the blue circles show the values extracted from the experiments. The red dashed line corresponds to the correlator width of the pulses. Above 50 ps, the deviation of the simulated width from this line is less than 5 % – in this range our device works nicely as a pulse correlator.

5.6. Cross-correlation of upper and lower polaritons

A single-photon switch and / or pulse-correlator operation can also be realized by centering the signal pulse on the other (lower) polariton transition, which yields a larger magnitude for the nonlinearity. We carried out this experiment by exciting the upper-polariton transition with the control laser and the lower polariton with the signal laser. In this case, the optical nonlinearity is negative (compare Figure 5.3(c)), since the ground-state population is decreased with the control / signal laser present, preventing the other laser to scatter photons on the lower / upper-polariton transition, respectively. Without post-filtering the scattered photons (corresponding to the situation in our experiment), the control and signal pulses are inter-changeable without having any effect on the nonlinear system response. The negative nonlinear response on the polaritons is about a factor of two larger than the positive nonlinearity on the transition into the second Jaynes-Cummings manifold, as can be seen in Figure 5.3(c). Figure 5.8 displays the correspondingly measured negative nonlinear system responses as a function of signal-pulse delay for pulse durations of (a) 33 ps and (b) 51 ps.

5.7. Transistor operation

Our single-QD cavity system could in principle be operated as a single-photon transistor [16, 17]. Compared to a switch, a transistor should exhibit gain, which enables the operation in potentially lossy circuits. At the level of single light quanta one can define the gain $G$ as the number of scattered signal photons per incident control photon before the spontaneous relaxation of the device back into its ground (or off) state. One might naively expect that in a Jaynes-Cummings system the gain should
Figure 5.7.: Pulse correlator. The strength of the nonlinearity, as defined in the caption of Fig. 5.6, as a function of delay for pulse durations of (a) 51 ps and (b) 86 ps. The longer the pulse, the more symmetric the nonlinear response. The red curves are the numerical convolutions of the streak-camera images shown as insets. The absolute amplitudes and the peak positions of the convoluted signals were fitted to the data. In both cases, the control and signal laser had an average power of 1 nW. This power corresponds to about 60 photons per pulse impinging on the sample surface or a pulse energy of 80 eV, which means that on average 0.6 photons per pulse are scattered off the system. (c) The width of the nonlinear system response versus laser-pulse duration obtained from MCWF simulations (open circles) and from experimental data (blue circles). For pulse durations larger than 50 ps, the simulated width of the nonlinear signal approaches that of the correlator width of the laser pulses (red dashed line). For very short pulses, the polariton lifetime sets the lower limit for the system response. The error bars shown on the first datapoints in (a) and (b) denote photon-shot noise.

be determined by the ratio of the excited state lifetimes of the first compared to the second manifold.

However, due to the finite overlap of transitions between higher-lying manifolds (see Figure 5.9), multiple transitions are involved in the system dynamics. If $g/\kappa > 2$, the fundamental-polariton transitions are to a certain extent spectrally separated from the rest, i.e. from the higher-lying transitions. If a weak control-laser pulse is resonant with a transition from the ground state to a polariton state, while a stronger signal pulse addresses the higher-lying transitions jointly, the gain
Figure 5.8.: Ultrafast negative nonlinear system response. Nonlinear system response of the QD-cavity device as a function of the signal-pulse delay with the control laser set to the upper-polariton and the signal laser on the lower-polariton transition for pulse durations of (a) 33 ps and (b) 51 ps. Both lasers have an average power of 1 nW. Since here the role of control and signal lasers are interchangeable, the system response is symmetric. The red curves are results from MCWF simulations with the absolute amplitude determined from a least-square fit to the data. The error bars shown on the first datapoints denote photon-shot noise.

Figure 5.9.: Possible scheme for transistor operation on exact cavity-exciton resonance. The x-axis describes the lower-polariton transition energies (labeled as the detuning from the cavity-mode resonance in units of the coupling strength $\hbar g$) from the $n^{th}$ to the $(n+1)^{th}$ manifold (circles), as derived from Equation 2.20. Due to the large spectral separation of all higher-order Jaynes-Cummings transitions (red circles), a selective addressing of the fundamental transition (blue circle) with a weak control-laser pulse is possible. All higher-order transitions are addressed with the much stronger signal pulse.

can be larger than unity, due to the decreasing anharmonicity for higher photon numbers. We define the gain as

$$G = \frac{N_{ON} - N_{OFF}}{N_{control\ in}},$$

where $N_{ON} / N_{OFF}$ is the average number of scattered photons per cycle when the control laser is ON / OFF respectively (the signal laser is ON in both cases);
and $N_{\text{control in}}$ is the average number of input-photons per control pulse. Another transistor-key property is the ON/OFF ratio, defined here as

$$R_{\text{ON/OFF}} = \frac{N_{\text{ON}}}{N_{\text{OFF}}}$$  \hspace{1cm} (5.5)

In a simple simulation using the MCWF method, we find that our Jaynes-Cummings system can have a gain of $G \approx 2.0$ and $R_{\text{ON/OFF}} \approx 2.1$ for the following parameters: pulse durations, 50 ps; signal-pulse delay, 30 ps; $N_{\text{control in}} = 2.5$; $N_{\text{signal in}} = 10$; control-laser detuning, $g$ (resonant with the UP); signal-laser detuning, 0.35 g. This poor gain and ON/OFF ratio could be improved, if $g/\kappa$ were increased by a factor of 4 (as could be achieved with a larger $Q$-factor in state-of-the-art nanocavities). If the pure QD-dephasing rate is set zero in addition, a reasonable gain $G \approx 7.2$ and an ON/OFF ratio $R_{\text{ON/OFF}} \approx 16$ could be obtained. In this case, the input parameters are: $Q = 100 000$; pulse durations, 50 ps; signal-pulse delay, 70 ps; $N_{\text{control in}} = 8$; $N_{\text{signal in}} = 30$; control-laser detuning, $g$ (resonant with the UP); signal-laser detuning, 0.35 g.

Obviously, the calculated gain rapidly decreases for the experimentally observed exciton-dephasing rates. To investigate this behavior, we calculated the time dependence of the diagonal density-matrix elements corresponding to the occupation probabilities of the ten lowest upper and lower-polariton states. The evolution of the real system with the input variables as described above, is shown in Fig. 5.10(a). When looking at the fundamental-polariton evolutions $\rho_{1,+}$ and $\rho_{1,-}$, it appears that the system gains an undesirable significant lower-polariton population after a few 10 ps, due to pure QD dephasing. As a result, higher-order upper-polariton transitions are less frequently accessible, which lowers the system gain. In addition, pure QD dephasing smears out the levels, such that signal-laser photons scatter on polariton transitions, even if there is no control laser applied. Hence, the ON/OFF ratio is reduced as well. For a higher signal-laser power, the ON/OFF ratio is even further reduced.

In the hypothetical system shown in Figure 5.10(b) on the other hand, a clean system evolution without any lower-polariton contribution is found. The input powers are allowed to be higher without a significant reduction of the ON/OFF ratio. Consequently, the system shows non-negligible excitations up to the 10\textsuperscript{th} Jaynes-Cummings manifold, yielding the mentioned high gain $G \approx 7.2$.

In conclusion, while increasing the ratio $g/\kappa$ would already increase $G$, high-gain ($G \gg 1$) transistors may be realized in combination with EIT schemes [11, 147–149]. Finally, combining our device with state-of-the-art waveguide technology [150–152] and implementing QD-charge control using p-i-n structures [126] to suppress the blinking effect would enable high-contrast all-optical switching of single-photon pulses. This might enable the demonstration of preserved quantum coherence during the nonlinear interaction, which could in turn pave the way for the realization of an ultrafast controlled-phase gate between two single-photon pulses [19].
Figure 5.10.: MCWF simulation of the system evolution in the transistor mode. The time evolution of the $n^{th}$ upper (blue) and lower (red) polariton state occupation probability $\rho_{n,\pm}$ is shown. On the left hand side, the real system with $Q = 25\,000$ and $\hbar\gamma_{\text{deph}} = 13\, \mu\text{eV}$ is simulated. On the right hand side, the evolution of a hypothetical system without QD dephasing and a cavity-quality factor $Q = 100\,000$ is shown.
6. Cavity-QED with quantum wells in Fabry-Pérot cavities

This chapter discusses the physics of cavity-QED in quantum-well microcavities. The basic light-matter interaction of a cavity-coupled many-body quantum system is explained in Section 6.1. Section 6.2 discusses fundamental properties of semiconductor Fabry-Pérot cavities, while Section 6.3 presents the implications for the coupling with quantum wells. Confined polariton systems are considered in Section 6.4, along with a brief discussion about exciton disorder. The last part of the chapter, Section 6.5, looks at polariton-polariton interactions.

Exciton-polaritons, first predicted by Hopfield [37] in 1958, are coherent superpositions of cavity photons and quantum-well (QW) excitons, when the two systems are coherently coupled in a single device. In the late 1970’s, incoherent (weak) coupling of QWs to microcavities led to the fabrication of vertical-cavity surface-emitting lasers (VCSELs) [153] that revolutionized the field of semiconductor lasers. With increasing quality of semiconductor-material growth during the following years, the experimental era of microcavity polaritons officially began in 1992, when the first Vacuum-Rabi splitting was observed in a Fabry-Pérot microcavity containing QWs [38]. Since then, many different inorganic and organic materials have been coherently coupled to microcavities. Interesting applications like polariton LEDs [154] and low-threshold polariton lasers [155] were realized recently.

The optically active semiconductor-quantum wells consist of layers made of a low-energy band-gap semiconductor, sandwiched between a higher-energy band-gap semiconducting host material. The thickness is typically close to the exciton Bohr radius of about 10 nm to ensure two-dimensional motion of the carriers inside the QW plane. Quantum-well excitons are quasi-stable compounds consisting of a heavy hole and a conduction-band electron. In a QW, the distance between two excitons is typically much larger than the exciton Bohr radius. Excitons can therefore be considered ‘point like’ and constitute bosonic particles, which can occupy the same momentum state inside a single QW. In stark contrast to QD-cavity polaritons, discussed in the previous chapters of this thesis, QW microcavities therefore are linear systems, with the transition energies independent of the number of polaritons in the system.

As the cavity-mode frequency is tuned across the exciton resonance, the spectral signature is an avoided crossing, in analogy of what is observed in a single-QD cavity system (see Chapter 2). However, the Rabi splitting between the upper and lower-polariton branches is more than an order of magnitude larger than in the case of a single QD, due to the collective coherent coupling of many excitons to the cavity.
mode. The harmonic energy diagram and spectral features of cavity polaritons are derived in the following Section.

6.1. Interaction between N two-level emitters and a single cavity mode

For a basic understanding of the linear system dynamics, we can assume that the emitters do not interact with each other, and all couple – for simplicity with the same coupling strength $g$ – to a single radiation-field mode of a cavity. If the atomic occupation state is mapped to a spin-$\frac{1}{2}$ angular-momentum basis (using the notation $g = -\frac{1}{2}$, $e = \frac{1}{2}$), the collection of atoms is described by the Dicke states [65]

$$|\frac{-N}{2}\rangle_a = |g\rangle_1 |g\rangle_2 \cdots |g\rangle_N, \quad |\frac{-N}{2} + 1\rangle_a = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |g\rangle_1 |g\rangle_2 \cdots |e\rangle_j \cdots |g\rangle_N, \quad \ldots$$

$$|\frac{N}{2}\rangle_a = |e\rangle_1 |e\rangle_2 \cdots |e\rangle_N,$$

where $|s\rangle_j$ describes the angular-momentum state of the atom located at the lattice site with index $j$. $|\frac{-N}{2}\rangle_a$ describes the system, when all atoms are in the ground state, $|\frac{-N}{2} + 1\rangle_a$ the situation with one atom in the excited state, and so forth.

If each atom is coupled to a cavity mode with coupling strength $g$, the Hamiltonian is written

$$\hat{H} = \hbar \omega_a \hat{J}_z + \hbar \omega_c \hat{a}^\dagger \hat{a} + \hbar g (\hat{a} \hat{J}_+ + \hat{a}^\dagger \hat{J}_-),$$

where $\{ \hat{J}_z, \hat{J}_+, \hat{J}_- \}$ are the collective angular-momentum operators, defined as

$$\hat{J}_z |m\rangle_a = m |m\rangle_a, \quad \hat{J}_+ |m\rangle_a = \sqrt{\frac{N}{2} \left( \frac{N}{2} + 1 \right) - m (m + 1)} |m + 1\rangle_a, \quad \hat{J}_- |m\rangle_a = 0,$$

$$\hat{J}_- |m\rangle_a = \sqrt{\frac{N}{2} \left( \frac{N}{2} + 1 \right) - m (m - 1)} |m - 1\rangle_a, \quad \hat{J}_- |\frac{N}{2}\rangle_a = 0.$$

Including the Fock states of the cavity degree of freedom $|n\rangle_c$ and writing $|m, n\rangle \equiv |m\rangle_a \otimes |n\rangle_c$, it is straightforward to show that the system eigenstates with one energy quantum (at zero detuning $\omega_c = \omega_a$) and the corresponding eigenenergies are given by

$$|\Psi^{+1/2}_1\rangle = \frac{1}{\sqrt{2}} (|\frac{-N}{2}, 1\rangle + |\frac{-N}{2} + 1, 0\rangle), \quad E^{+1/2}_1 = \hbar \left( \omega_a + \sqrt{N}g \right),$$

$$|\Psi^{-1/2}_1\rangle = \frac{1}{\sqrt{2}} (|\frac{-N}{2}, 1\rangle - |\frac{-N}{2} + 1, 0\rangle), \quad E^{-1/2}_1 = \hbar \left( \omega_a - \sqrt{N}g \right).$$

The Vacuum-Rabi splitting of a collection of $N$ atoms coupled to a cavity mode is hence

$$\Delta E = 2\sqrt{N} \hbar g,$$

and therefore scales with the square root of the number of coupled atoms (see Figure 6.1). An important difference to the single-atom case is revealed, when
the number of quanta $q$ in the system is larger than 1: Instead of two dressed levels, there are $q + 1$ dressed eigenstates, formed by superpositions of the vectors \( \{ -\frac{N}{2}, q \}, \{ -\frac{N}{2} + 1, q - 1 \}, \ldots, \{ -\frac{N}{2} + q, 0 \} \}. \) The eigenvalues turn out to be

\[
E_s^q = \hbar \left( q\omega_a + 2s\sqrt{Ng} \right), \quad \text{with} \quad s = -\frac{q}{2}, -\frac{q}{2} + 1, \ldots, \frac{q}{2}.
\]

(6.6)

Figure 6.1.: The energy-level diagram of a collection of $N$ atoms coupled to a cavity mode at zero cavity-atom detuning. The lowest-lying two excitation manifolds ($q = 1, 2$) are depicted. The two outermost transitions between the first and second manifolds are forbidden, ensuring a harmonic energy ladder.

However, not all the transitions between two successive manifolds are optically accessible. Calculating the transition matrix elements,

\[
\langle \Psi_q^s | \hat{J}_- | \Psi_{q+1}^{s'} \rangle = 0, \quad \text{if} \quad |s - s'| \neq \frac{1}{2},
\]

(6.7)

reveals that only those transitions are allowed that exhibit a transition energy of either $\hbar \left( \omega_a + \sqrt{Ng} \right)$ or $\hbar \left( \omega_a - \sqrt{Ng} \right)$, as illustrated in Figure 6.1. Therefore, the energy ladder is harmonic (for $q < N$).

6.1.1. Interaction with a two-dimensional gas of quantum emitters

In this thesis, we are particularly interested in the coupling of a two-dimensional Bose gas to a Fabry-Pérot cavity mode. We assume that the emitters do not interact with each other and are all located in a two-dimensional layer in the middle of a cavity-field antinode close to the beam waist, as depicted in Figure 6.2. The number of lattice sites $N$ coupled to the cavity mode is in a simplified description given by $N = \frac{1}{2} \omega_0^2 \pi \rho$, where $\omega_0$ is the minimum cavity-waist radius, $\frac{1}{2} \omega_0^2 \pi$ is the waist area defined by the 2D-integration over the normalized cavity-field intensity and $\rho$ the area density of lattice sites.

The effective cavity-mode volume writes

\[
V_{\text{eff}} = \frac{1}{4} \omega_0^2 \pi L_{\text{eff}},
\]

(6.8)
where $L_{\text{eff}}$ is the effective cavity length. The additional factor $\frac{1}{2}$ is due to the integration over the intensity nodes and antinodes. Substituting this value into the expression of the coupling strength $g$ (Equation 2.7), and calculating the system Rabi splitting according to Equation 6.5, one obtains

$$\Delta E = 2\sqrt{N} \hbar g = \sqrt{\frac{8\hbar \omega_c \rho}{\epsilon_0 n^2 L_{\text{eff}}}} \left| \phi \left( \mathbf{r}_a \right) \cdot \mathbf{d}_{\text{el}} \right|. \quad (6.9)$$

The Rabi splitting is hence independent from the cavity-spot size $\omega_0$, but scales with $\Delta E \propto 1/\sqrt{L_{\text{eff}}}$. In this work, we implement a system like the one displayed in Fig. 6.2 by placing an indium gallium arsenide (InGaAs) QW at the position of the beam waist in a Fabry-Pérot microcavity. Here, the Bose gas is formed by QW excitons that can move freely in the plane. Written in terms of the oscillator strength $f_{2D}$ between a two-dimensional QW exciton and the light field, the Rabi splitting is given by [156]

$$\Delta E = 2\hbar \Omega_R = 2\hbar \sqrt{\frac{4\pi \omega_c f_{2D}}{L_{\text{eff}}}} \left| \phi \left( \mathbf{r}_a \right) \right|. \quad (6.10)$$

Here, the effective cavity length is the optical distance between the two mirrors, with the penetration into the mirrors included, $L_{\text{eff}} = n_{\text{eff}} L_{\text{phys}} + L_{\text{eff}}^{\text{mirror}}$, where $L_{\text{phys}}$ is the physical distance between the two mirrors and $n_{\text{eff}}$ the effective refractive index (see Section 6.2.1).

Besides the minimization of the cavity length (down to the lower limit of a few wavelengths), a way to increase the Rabi splitting is to place many QWs at the antinodes of the cavity field. In this case, the Rabi splitting additionally scales with the square root of the number of QWs

$$\Delta E(N_{\text{QWs}}) = \sqrt{N} \Delta E, \quad (6.11)$$

assuming that all QWs are maximally coupled to the cavity-mode field.
6.2. Dielectric Fabry-Pérot microcavities

Fabry-Pérot cavities consist of two planar or curved mirrors to confine light to a small volume that is either empty or filled with a transparent dielectric material. For certain frequencies of light, a resonance condition allows the build-up of a large optical field inside the cavity.

In this thesis and in the context of cavity-QED, two quantities are of particular importance: Firstly, the cavity $Q$-factor that is directly connected to the linewidth of a cavity-mode resonance $\kappa = \omega/\mathcal{Q}$ and to the average cavity-photon lifetime, given by its inverse, $\tau = \mathcal{Q}/\omega$. The second quantity of great interest is the cavity length $L$, emerging as the main tuning parameter for the coherent coupling strength between the cavity-light field and a sandwiched two-dimensional optical emitter, $\Omega_R \propto 1/\sqrt{L}$ (see Equation 6.10).

Neglecting optical absorption and aberrations, the $Q$-factor of a Fabry-Pérot cavity consisting of two identical mirrors is given by

\[
\mathcal{Q} = \frac{2\pi nL}{\lambda \ln \frac{1}{R}} \tag{6.12}
\]

where $n$ is the refractive index of the spacer material and $R$ denotes the reflectivity of both mirrors. To achieve the largest possible coupling strength $\Omega_R$, cavity lengths on the order of a few wavelengths are desired. Within the last two decades, the ability of fabricating high-quality dielectric mirrors with reflectivities larger than 99.999% enabled high $Q$-factors, even for cavities with lengths on the order of an optical wavelength. These achievements heralded the extremely active field of cavity-QED with semiconductor microcavity polaritons.

Ultra-high reflectivities larger than 99.999% can only be achieved using dielectric distributed Bragg reflectors (DBRs), whereas the best metallic mirrors reflect less than 99% of the incident light. A DBR consists of an alternating series of thin layers of two materials with different refractive indices $n_1$ and $n_2$. If the thickness of each layer is chosen to be $d_1 = \lambda_0/(4n_1)$ and $d_2 = \lambda_0/(4n_2)$, respectively, the reflections from all layers interfere constructively, as light at wavelength $\lambda_0$ hits the structure perpendicularly. If the number of layer pairs is sufficiently large, the light cannot penetrate the structure and is almost totally reflected. However, this condition is only valid for a finite optical frequency-band around $\lambda_0$, the so-called stopband of the mirror. We define the stopband as the wavelength range over which $1 - R < 2(1 - R_{\text{max}})$. Figure 6.3 depicts the wavelength-dependent DBR reflectivity that was calculated using a transfer-matrix approach [157]. The bandwidth of the stopband is determined by the refractive-index contrast of the two materials. In our experiments, we use gallium arsenide (GaAs) and aluminium arsenide (AlAs), yielding an about 40 nm wide stopband, as illustrated in the figure.

Monolithic Fabry-Pérot microcavities typically consist of a thin semiconductor-spacer layer that is sandwiched between two DBRs. The dielectric structure can be characterized by the mirror and spacer materials and the number of pairs $N$ of the two mirror materials. A common notation for microcavities refers to the cavity spacer; for instance, a GaAs-3/2$\lambda$ cavity consists of a spacer layer made of GaAs with a thickness $3/2 \cdot \lambda_0/n_{\text{GaAs}}$. Figure 6.4(a) shows the center part of the dielectric heterostructure of a GaAs-$\lambda$ cavity with GaAs/AlAs mirrors and $N = 25$ for both
Figure 6.3.: Wavelength-dependent reflectivity of a DBR. The stopband is centered at $\lambda = 900$ nm and consists of 25 GaAs/AlAs-layer pairs. The yellow part indicates the mirror stopband.

mimics. The field was calculated using a transfer-matrix approach. In contrast to a cavity consisting of metallic mirrors, the cavity-mode field penetrates deeply into the mirrors. The shortest possible effective cavity length is therefore limited by the optical penetration depth into the mirrors. A reasonable way of defining this length is to account for the phase the light acquired upon reflection at the mirrors. The corresponding optical length in the middle of the stopband is then given by

$$L_{\text{DBR}} = \frac{\lambda_0}{2} \frac{n_1 n_2}{n_1 - n_2},$$

(6.13)

where $n_1 > n_2$. For a GaAs cavity with GaAs/AlAs mirrors and a center wavelength of $\lambda_0 = 900$ nm, the penetration depth is about $L_{\text{DBR}} \approx 8.5 \lambda_0$, using the refractive indices $n_{\text{GaAs}} \approx 3.54$ and $n_{\text{AlAs}} \approx 2.93$. In the rest of this discussion, we refer to the effective cavity length as

$$L_{\text{eff}} = n_s L_{\text{phys}} + L_{\text{DBR}},$$

(6.14)

where $L_{\text{phys}}$ is the physical thickness and $n_s$ the refractive index of the spacer layer. For a wavelength $\lambda$ that is within the stopband, the cavity-mode resonance condition is met, if

$$\left( m + \frac{2L_{\text{DBR}}}{\lambda_0} \right) \lambda = 2L_{\text{eff}},$$

(6.15)

where $m$ is a positive integer. The cavity-transmission spectrum of the structure shown in Fig. 6.4(a) is depicted in Figure 6.4(b). The transmission is almost identical to $(1 - R(\lambda))^2$, where $R(\lambda)$ is the reflection curve shown in Fig. 6.3, except for a sharp cavity-mode resonance at $\lambda = 900$ nm.
6.2. Dielectric Fabry-Pérot microcavities

Figure 6.4.: The electric field and spectrum of a DBR-cavity. (a) Heterostructure of a typical microcavity consisting of two materials with different refractive indices. A GaAs-spacer layer of $\lambda_0/n_{\text{GaAs}}$ thickness is sandwiched between two distributed Bragg reflectors made of GaAs and AlAs. The red trace depicts the calculated square of the electric field, when the incident light is resonant with the cavity mode ($\lambda = 900$ nm), normalized to the square of the input electric field. The field shows a major increase within a confined region of a few $\mu$m length. The $z$-axis denotes the growth direction. (b) The calculated transmission spectrum shows almost zero transmission for light wavelengths within the stopband, except for a sharp cavity-mode resonance at the center of the stopband at 900 nm.

6.2.1. Cavity dispersion

The photon dispersion in free space is given by $\omega = c|k|$, indicating that a photon has no mass. However, if light is confined between two plane mirrors, the photon wave vector perpendicular to the plane ($z$-axis) is quantized, according to

$$k_m = m \frac{\pi n_{\text{eff}}}{L_{\text{eff}}},$$

where $n_{\text{eff}}$ is the effective (mean) refractive index of a photonic round trip and $m$
denotes the quantum number of the respective longitudinal cavity mode. A cavity-mode resonance with in-plane wave vector \( \mathbf{k} \) has the energy

\[
\hbar \omega_{m,\mathbf{k}} = \frac{\hbar c}{n_{\text{eff}}} \sqrt{k_m^2 + |\mathbf{k}|^2} = \hbar \omega_{m,0} \sqrt{1 + \frac{c^2 |\mathbf{k}|^2}{n_{\text{eff}}^2 \omega_{m,0}^2}}
\]

and therefore has a hyperbolic dispersion. From the band curvature, an effective cavity-photon mass can be defined [73]

\[
m_c = \frac{n_{\text{eff}}^2}{c^2} \hbar \omega_{m,0},
\]

where \( \omega_{m,0} = \frac{ck_m}{n_{\text{eff}}} \). We can re-write Equation 6.17, which results in the relativistic expression for the photon energy

\[
\hbar \omega_{m,\mathbf{k}} = \sqrt{\left( m_c \frac{c^2}{n_{\text{eff}}^2} \right)^2 + \left( \hbar |\mathbf{k}| \frac{c}{n_{\text{eff}}} \right)^2} \approx \hbar \omega_{m,0} + \frac{\hbar^2 |\mathbf{k}|^2}{2m_c},
\]

with the reduced speed of light \( \frac{c}{n_{\text{eff}}} \). The last expression approximates the dispersion curve by a parabola around \( \mathbf{k} = 0 \) (for \( |\mathbf{k}| \ll \frac{m_c c}{\hbar n_{\text{eff}}} \)).

For a GaAs cavity and a wavelength \( \lambda = 900 \text{ nm} \), the effective cavity-photon mass is orders of magnitude smaller than the free electron mass, \( m_c \approx 3.3 \cdot 10^{-5} m_e \).

### 6.3. Microcavity polaritons

Having discussed the basics of semiconductor microcavities, this section investigates the coupling of a QW to a cavity-mode field. To achieve the largest optical coupling, the QW layer is placed at an antinode of the field. As we demonstrated in Section 6.1, the collective coupling of an ensemble of emitters to a single cavity mode results in an increased coupling strength \( \Omega_R \), as compared to the coupling of a single quantum emitter. It was found that \( \Omega_R \propto \sqrt{\frac{N}{L_{\text{eff}}}} \), where \( N \) is the number of quantum wells coupled to the cavity mode. An important finding is also the fact that microcavity polaritons constitute a harmonic system and can therefore be described by bosonic operators.

In order to describe the coupling of a two-dimensional ensemble of emitters to a cavity mode, the discussed in-plane momentum of the cavity photons as well as the exciton momentum must be included, resulting in the Hamiltonian

\[
\hat{H} = \int \frac{d^2 \mathbf{k}}{(2\pi)^2} \sum_{\sigma} \left\{ \sum_{j=c,X} \left[ \hbar \omega_j(\mathbf{k}) \hat{a}_{j,\sigma}^\dagger(\mathbf{k}) \hat{a}_{j,\sigma}(\mathbf{k}) \right] + \hbar \Omega_R \left[ \hat{a}_{X,\sigma}^\dagger(\mathbf{k}) \hat{a}_{c,\sigma}(\mathbf{k}) + \hat{a}_{c,\sigma}^\dagger(\mathbf{k}) \hat{a}_{X,\sigma}(\mathbf{k}) \right] \right\}
\]

where \( \mathbf{k} \) is the in-plane wave vector. The operators \( \hat{a}_{X,\sigma}, \hat{a}_{c,\sigma} \) are the bosonic annihilation operators of an exciton (cavity photon) with wave vector \( \mathbf{k} \) and exciton spin \( \sigma \) (cavity-photon polarization \( \sigma \)), which is usually given in a circular basis.
Since the circular symmetry in our system is broken, however, a linear basis turns out to be more appropriate. The eigenvalues of the Hamiltonian are found to be

\[ E_{\sigma}^{\pm}(k) = \frac{\hbar}{2} [\omega_{c,\sigma}(k) + \omega_{X,\sigma}(k)] \pm \hbar \sqrt{\Omega_{R} + \frac{1}{4} [\omega_{c,\sigma}(k) - \omega_{X,\sigma}(k)]^2}. \]  
(6.21)

For an InGaAs QW embedded in GaAs and having an effective cavity length of about \( L_{\text{eff}} \approx 6 \lambda \), the coupling strength is measured \( \hbar \Omega_{R} \approx 2.1 \text{ meV} \) (see Figure 7.8), and is therefore about an order of magnitude larger than the coupling strength \( \hbar g \) between an InAs QD coupled to the electric-field maximum of a photonic-crystal nanocavity (see Chapter 3).

The cavity-mode dispersion is rather steep, due to the small photonic mass \( m_{c} \), whereas the excitonic dispersion can be considered as being flat. As an example, a calculated polariton dispersion (using Equations 6.17 and 6.21) is shown in Figure 6.5, for a cavity-exciton detuning of \( \omega_{c} - \omega_{X} = -0.5 \Omega_{R} \).

![Figure 6.5: Calculated dispersion curves of the upper and lower polaritons. The cavity-exciton detuning is \(-0.5 \Omega_{R}\). The curvature of both upper and lower-polariton branches is inherited from the photonic-dispersion relation.](image)

Since only the cavity part is responsible for the bending of the polariton dispersion at \( k = 0 \), the effective polariton masses on exact cavity-exciton resonance are given by \( m_{LP} = m_{UP} = 2m_{c} \), where the sub-indices LP and UP stand for lower polariton and upper polariton, respectively.

Experimentally, the polariton dispersion of a planar microcavity can be measured by angle-resolved spectroscopy. The emission angle \( \theta' \) inside the material is given by \( \sin \theta' = \frac{k_{c}}{\omega_{n_{\text{eff}}}} \). So the wave vector \( k \) can be obtained by measuring the free space angle \( \theta \),

\[ k = \frac{\omega}{c} \sin \theta. \]  
(6.22)

The above calculated dispersion curve was experimentally confirmed by PL measurements [158, 159].
6.3.1. Polariton lasing and condensation

An important aspect of cavity polaritons is their bosonic nature. Bose-Einstein condensation (BEC) in a gas of identical bosonic particles sets in once the thermal de Broglie wavelength exceeds the atomic Bohr radius. BEC of atoms requires the trapping of atoms and their cooling to ultralow temperatures on the order of $\mu K$ or below to fulfill this condition [160]. Condensation of cavity polaritons was first proposed by İmamoğlu et al. in 1996 [161]: A coherent macroscopic population of polaritons in their lowest energy ($k = 0$) state was predicted to build up upon incoherent optical pumping, via exciton-phonon scattering. However, a polariton condensate is not a BEC in standard terms, for mainly two reasons: It is generated by final-state stimulation through phonon scattering and is therefore not necessarily in thermal equilibrium with the phonon bath. Moreover, a polariton condensate is of intrinsically non-equilibrium nature. The polariton lifetime is on the order of $1 - 100$ ps, since spontaneous ‘particle’ losses occur via photon leakage through the cavity mirrors.

The idea of condensing polaritons came together with the proposal of a new kind of laser, namely a polariton laser, that has a fundamentally different working principle than a conventional laser, which operates under population inversion, setting in above a critical pump threshold. In contrast, the photon emission from a polariton condensate is intrinsically coherent and therefore indistinguishable from the coherent photon stream of a conventional laser. Following this idea, a polariton laser does not necessarily exhibit a lasing threshold. In reality, it turned out that an efficient population of the polariton ground state using non-resonant light is prohibited, due to a bottleneck effect [162, 163] that prevents polaritons with large in-plane momentum to relax into the $k = 0$ ground state: Due to the tiny effective polariton mass, the slope of the polariton dispersion is so large that the final density of states is too low for supporting efficient phonon-mediated relaxation channels. When the system is nevertheless driven strongly, a saturation effect sets in instead: Non-relaxed carriers form an electron-hole plasma that screens the Coulomb potentials of low-momentum polariton states. Consequently, these states are bleached and the strong coupling to the cavity mode is lost [164, 165]. As a result, the system emits predominantly via the weakly-coupled cavity mode. Lasing can occur, if the electron-hole plasma exceeds a critical density, at which a system-population inversion is attained. Stimulated emission then starts to dominate over spontaneous electron-hole recombinations and the system behaves like a conventional laser, which is in this context typically termed a photon laser, in contrary to a polariton laser [47].

The first convincing demonstration of stimulated polariton generation was presented in an experiment performed under resonant parametric excitation [166]. The authors of this publication generated a large population of non-ground state polaritons at wave vector $k_0$ by pumping the system with a resonant short laser pulse under a certain ‘magic’ angle. After a delay of 1 ps, they applied a probe-laser pulse at normal incidence to generate a seed population for the formation of a condensate. For a properly chosen pump angle, an amplification of the ground-state polariton population by a factor 100 was observed. The mechanism is explained by optical parametric oscillation (OPO): Through polariton-polariton interaction, two (pump) polaritons with momenta $k_0$ each generate a ground-state (signal) polariton with
6.3. Microcavity polaritons

$k = 0$ and an almost exciton-like (idler) polariton with wave vector $2k_0$. Very similar results were obtained when exciting the system in cw [167]. As theoretically discussed in Ref. [168], long-range coherence and phase stability of the ground-state (signal) polaritons is achieved when pumping the system (under the magic angle) above the parametric-oscillation threshold $P_{th}$. The final state is hence described by a non-equilibrium condensate. In contrast, the signal light remains incoherent below $P_{th}$.

Due to the bottleneck effect, the polariton condensation under incoherent pumping turned out to be more tricky. According to both experiments and calculations, the polariton-bleaching effect sets in at pump powers below the critical density that supports polariton condensation in systems based on GaAs [169]. To overcome this limitation, materials with higher exciton-binding energies were used and several $(N)$ QWs were placed into the antinodes of a microcavity. The power at which polariton bleaching occurs is therefore multiplied by a factor $N$. Moreover, the Rabi frequency $\Omega_R$ is larger by a factor of $\sqrt{N}$, such that the strong-coupling condition is more robust against bleaching. Strong evidence for polariton condensation or lasing was observed by several groups. The authors of Ref. [170] studied a system based on 16 cadmium telluride (CdTe) QWs and observed an exponential increase of the photon-emission rate above an incoherent pump-power threshold that was attributed to polariton stimulation. A second threshold at even higher input power was identified as the photon-lasing transition. A deeper insight into the system dynamics was provided by the study of an AlGaAs microcavity with 12 embedded GaAs QWs. The dispersion relation exhibits the effective polariton mass above the lasing transition. Moreover, a gentle transition from thermal to coherent photon emission above the lasing threshold was demonstrated by second-order correlation measurements [171]. In a CdTe-based system with 4 QWs, stimulated emission from several bright spots on the sample surface was attributed to spontaneous symmetry breaking and formation of condensates of different phases [172].

The first non-ambiguous demonstration of polariton condensation was performed on a 16 QW CdTe-based system [39]: The authors directly measured the ground-state occupation as a function of incoherent pump power, deduced from angle-resolved spectroscopy. Above a threshold power $P_{th}$, the emission was confined to a small solid angle and hence clearly attributed to recombinations of ground-state polaritons. Other signatures of polariton condensation were observed: A phase transition from unpolarized emission below threshold to linear emission above threshold (and hence emission of a single mode) was found. Using a Michelson interferometer, the authors measured a fourfold increase of the temporal coherence from the polariton lifetime of 1.5 ps to 6 ps. Another striking feature was the observation of long-range spatial coherence of the condensate over the whole pump spot, whereas it was determined to match with the thermal de Broglie wavelength below $P_{th}$. Polariton condensation was also demonstrated on GaAs-based microcavities [49] and for polaritons in a trap [40].

The field of polariton condensation blossomed, when superfluid hydrodynamics was proposed [173] and experimentally measured [50]: The diffusionless flow of a macroscopically degenerate state of polaritons was observed, along with the polariton flow crossing an obstacle without resistance. Many more spectacular experiments followed, demonstrating e.g. Bogoliubov excitations [51, 52], Josephson
oscillations [53], quantized vortices [54, 55] and dark solitons [56].

In the next section, I will treat systems with additional polariton confinement in the plane. Without further discussing lasing in confined systems, it must be noted that the breaking of translation invariance in such systems lifts the momentum conservation rule for polariton-polariton scattering processes [174]. In order to populate the polariton ground state, a remaining condition for efficient parametric scattering processes is to have a harmonic trap potential [43]. Consequently, a macroscopic population of the polariton ground state is more easily achieved in confined polariton systems.

6.4. Confined polariton systems

So far, we have considered two-dimensional polariton systems that ideally spread over the whole semiconductor chip. Additional confinement in the plane results in the quantization of the in-plane wave vector $k$. If the confinement is so tight that the energy separation of the $k = 0$ and the next higher $k$-state is larger than the polariton linewidth, well-defined and discrete higher-order transverse-polariton modes emerge. In this thesis, I will refer to zero-dimensional polariton systems as polariton boxes. I will briefly treat such a confined microcavity system theoretically for a rough understanding of the resulting system eigenmodes using a quantum Hamiltonian model [36, 173]. The system Hamiltonian is given by

$$
\hat{H} = \int d^2r \left[ \sum_{i,j=X,c} \hat{\Psi}_i^\dagger (r) \hat{h}_{ij} \hat{\Psi}_j (r) + \left( \hbar F_p (r, t) e^{-i\omega_L t} \hat{\Psi}_c (r) + \text{h.c.} \right) \right],
$$

(6.23)

where

$$
\hat{h} = \hbar \left( \frac{\omega_X - \hbar \frac{2m_X}{\Omega_R} \nabla^2}{\omega_X} - \hbar \frac{2m_c}{\Omega_R} \nabla^2 + \frac{\Omega_R}{2\hbar} \left( \Xi_x^2 \hat{x}^2 + \Xi_y^2 \hat{y}^2 \right) - i\kappa \right). \tag{6.24}
$$

Here, we neglected any nonlinear dynamics, such as polariton-polariton interactions or exciton-saturation effects and considered only one polarization / spin state. The field operators $\hat{\Psi}_i (r)$ annihilate an exciton ($i = X$) or a cavity photon ($i = c$) as a function of the position $r = (x, y)$ in the QW plane and satisfy the standard Bose-commutation rules $[\hat{\Psi}_i (r), \hat{\Psi}_j^\dagger (r')] = \delta^2 (r - r') \delta_{i,j}$. The parabolic elliptic cavity-confining potential is described by the term $\frac{\Omega_R}{2\hbar} \left( \Xi_x^2 \hat{x}^2 + \Xi_y^2 \hat{y}^2 \right)$. The coherent external pump laser that drives the cavity mode at frequency $\omega_L$ is described by $F_p (r, t)$. System losses result from the cavity-photon loss rate $\kappa$. Note that we neglected exciton-spontaneous decay or dephasing events in this model. This approximation is reasonable, since excitons with low in-plane momentum only have a radiation channel perpendicular to the plane and hence only decay via the cavity mode. Moreover, the exciton-phonon scattering rate (of the lower polariton) is weak as well, due to the steep polariton dispersion around $k \approx 0$ (see the more detailed discussion in Ref. [157]).

Introducing the cavity-spot sizes in $x$ and $y$-directions $\omega_{0x} = \sqrt{\frac{2\hbar}{m_c \Xi_x}}$, $\omega_{0y} = \sqrt{\frac{2\hbar}{m_c \Xi_y}}$, the dimensionless variables $\tilde{x} = \frac{\sqrt{\Xi_x}}{\omega_{0x}} x$, $\tilde{y} = \frac{\sqrt{\Xi_y}}{\omega_{0y}} y$ and the transverse-mode
lowering operators \( \hat{c}_x = \frac{1}{\sqrt{2}} (\hat{x} + \partial_x) \) and \( \hat{c}_y = \frac{1}{\sqrt{2}} (\hat{y} + \partial_y) \) and neglecting the exciton kinetic-energy term (since \( m_X \gg m_e \)), \( \hat{h} \) can be re-written as

\[
\hat{h} = \left( \frac{\omega_X}{\Omega_R} \omega_c + (\hat{m} + \frac{i}{2}) \Xi_x + (\hat{n} + \frac{i}{2}) \Xi_y - i \frac{\delta_{mn}}{2} \right),
\]

(6.25)

where we wrote \( \hat{m} = \hat{c}_x \hat{c}_x \) and \( \hat{n} = \hat{c}_y \hat{c}_y \). With \( H_m \) being the Hermite polynomial of \( m \)th order [131] we introduce the two-dimensional normalized Hermite functions

\[
\phi_{mn}(\hat{x}, \hat{y}) = \frac{1}{\sqrt{\pi^m n! 2^{m+n}}} H_m(\hat{x}) H_n(\hat{y}) e^{-\frac{1}{2}(\hat{x}^2 + \hat{y}^2)}
\]

(6.26)

that are the eigenfunctions of \( \hat{m} \) and \( \hat{n} \), i.e. \( \hat{m}\phi_{mn} = m\phi_{mn} \) and \( \hat{n}\phi_{mn} = n\phi_{mn} \). Writing

\[
\hat{\Psi}_i^{\mp} = \sqrt{\frac{\omega_{0x}}{2}} \int \int d\hat{x} \, d\hat{y} \, \phi_{mn}^*(\hat{x}, \hat{y}) \hat{\Psi}_i \left( \frac{\omega_{0x}}{\sqrt{2}}, \frac{\omega_{0y}}{\sqrt{2}} \right) \hat{\Psi}_i
\]

(6.27)

for the exciton wavefunction \( (i = X) \) and the cavity-mode field \( (i = c, \text{ representing the cavity transverse-electromagnetic (TEM) modes [131]} \), the transverse eigenoperators (transverse upper and lower-polariton annihilation operators) are easily found to be

\[
\hat{p}_m^+ = \cos \theta_{mn} \hat{\Psi}_m^X + \sin \theta_{mn} \hat{\Psi}_m^c, \quad \hat{p}_m^- = \sin \theta_{mn} \hat{\Psi}_m^X - \cos \theta_{mn} \hat{\Psi}_m^c,
\]

(6.28)

with the (complex) eigenenergies

\[
E_{mn}^+ = \hbar \left( \frac{\omega_X}{2} - i \frac{\kappa}{4} + \sqrt{\frac{\Omega_R^2}{4} + \left( \frac{\delta_{mn}}{2} - i \frac{\kappa}{4} \right)^2} \right).
\]

(6.29)

Here, \( \delta_{mn} = \omega_c - \omega_X + (m + \frac{1}{2}) \Xi_x + (n + \frac{1}{2}) \Xi_y \) is the TEM\(_{mn}\) cavity-mode detuning from the exciton resonance, and \( \tan(2\theta_{mn}) = \frac{2\Omega_R}{\delta_{mn}} \).

The lateral shape of the TEM\(_{00}\) (lowest energy) polariton wavefunction inside the QW plane is Gaussian with the amplitude \( \propto e^{-\left( \frac{x^2}{\omega_{0x}^2} + \frac{y^2}{\omega_{0y}^2} \right)} \). All TEM\(_{mn}\) modes exhibit the same Rabi splitting of \( 2\Omega_R \) between the upper and lower-polariton modes on exact respective cavity-exciton resonance \( (\delta_{mn} = 0) \). When driving the system with a resonant laser, a TEM\(_{mn}\) mode gets excited, if the overlap integral of \( \phi_{mn}(\hat{x}, \hat{y}) \) with the pump spot \( F_p \left( \frac{\omega_{0x}}{\sqrt{2}}, \frac{\omega_{0y}}{\sqrt{2}} \right) \) is non-zero.

Experimentally, three-dimensional polariton confinement has been achieved by introducing potential traps for QW excitons, induced by applying external stress to a thinned chip [175, 176]. Another way for providing confinement is the engineering of the photonic part of the polariton wavefunction, e.g. by using photonic-crystal nanocavities [45] or micropillars [177]. However, these systems are typically subject to additional excitonic or photonic losses, due to absorption at the semiconductor-air interfaces. A more gentle confinement is achieved using an epitaxial-overgrowth technique [178]. The authors of Ref. [46, 179] grew a 6 nm shallow circularly shaped mesa on top of the cavity-spacer layer, before growing the top mirror. Thus, the
cavity-mode energy is locally red-shifted by about 9 meV. For mesa diameters of a few \( \mu m \), quantization of higher-order transverse-mode polaritons was observed.

A very similar method for potential engineering was demonstrated by depositing metal stripes on top of a microcavity, which locally blue-shifted the cavity-mode energy by up to 400 \( \mu eV \) \[180\]. Very recently, ways to modulate the potential landscape by utilizing the repulsive potential induced by background excitons created through the off-resonant excitation laser were demonstrated \[41–44, 181\].

A major drawback of all these monolithic QW microcavities presented above, is the lack of in-situ tunability of their photonic part, i.e. the cavity-photon resonance energy. In this thesis, we present a novel semi-integrated design for a polariton microcavity, consisting of a movable semiconductor DBR with a QW grown on top, and a second DBR deposited on the concave end-face of an optical fiber. This system allows for the implementation of (a) tight polariton confinement and (b) tunability of the cavity-mode energy and also polariton lifetime. This system will be discussed in detail in Chapter 7.

### 6.4.1. Exciton disorder

The lineshape of QW excitons emerges from an interplay between different forms of structural disorder, for instance from local QW-thickness fluctuations or inhomogeneities in the alloy that consists of two different semiconductor materials. This inhomogeneous exciton broadening (with a typical width of about 500 – 1500 \( \mu eV \) in InGaAs QWs) is about an order of magnitude larger than the homogeneous broadening stemming from QW-exciton spontaneous recombinations and dephasing, due to inelastic exciton-phonon scattering. Measured lifetimes are about 10 ps \[182\], corresponding to a lifetime-broadening of 66 \( \mu eV \).

In this section, we investigate what impact disorder has on the linewidth of polariton states. In experimental implementations (for example in Ref. \[38\]), the polariton linewidths on cavity-exciton resonance are routinely found to be smaller than half the inhomogeneous linewidth of the uncoupled QW excitons. Surprisingly, the upper-polariton resonance is often found to be broader than the lower-polariton line.

An interpretation of these reduced linewidths was found in motional narrowing \[183\]: The polariton effective mass \( m_c \) is so small that the energy of the orbital averages over a diameter of about 1 \( \mu m \), whereas an uncoupled QW exciton of the size of a Bohr radius directly ‘feels’ the local QW potential.

The authors in Ref. \[184\] discuss this problem by studying the collective coupling of an inhomogeneously-broadened exciton system (exhibiting Gaussian disorder) to a cavity mode, described by Dicke states (compare Section 6.1). But in contrary to the previous derivation in this thesis (see Section 6.1), they treat the excitons as distinguishable particles with varying transition energies and find two important conclusions: Firstly, the Rabi frequency emerges from the collective coupling of all inhomogeneously distributed excitons and yields \( \sqrt N \) times the root mean square of the individual coupling strengths,

\[
\Omega_R = \sqrt{\sum_{j=1}^{N} g_j^2} = \sqrt N \text{ RMS } \{g_j\}, \tag{6.30}
\]
and secondly, the linewidths of both upper and lower-polariton states are solely determined by the homogeneous broadening of the two systems, the cavity mode and the emitter transitions, weighted by their content in the polariton wavefunction. In particular, for zero cavity-exciton detuning, the polariton broadening is predicted to be

\[ \delta_{UP} = \delta_{LP} = \frac{1}{2} \left( \delta_X^{\text{hom}} + \frac{\omega_c}{Q} \right). \]  

(6.31)

In addition, the authors of Ref. [184] claim that a residual broad absorption peak (of width corresponding to the exciton inhomogeneous broadening) appears at the exciton resonance, which is, however, clearly less prominent than the upper and lower-polariton states.

To gain a deeper insight into the role of exciton disorder we simulated a microcavity system with an additional spatially-fluctuating exciton potential. To this end, a defining function \( \tilde{u}(|k|) \) (see Figure 6.6(a)) was chosen to support the scattering of excitons with certain wave vectors \( k \).

The function is normalized according to

\[ u(|k|) = \frac{\sqrt{8\pi} \tilde{u}(|k|)}{\sqrt{\sum_{k_i,k_j} |G(k_i,k_j)|^2 \tilde{u}(\sqrt{k_i^2 + k_j^2})}}. \]  

(6.32)

where \( G(k_i,k_j) \) is a Gaussian distribution of random complex numbers around the zero-point with variance \( \text{Var}(G) = 1 \). The wave vectors \( k_i, k_j \) span the whole two-dimensional momentum space with a sufficiently fine sampling interval. The exciton potential is written

\[ V(r) = \frac{1}{2\pi} \Re \left[ \sum_{k_i,k_j} G(k_i,k_j) e^{i(k_i x + k_j y)} u(\sqrt{k_i^2 + k_j^2}) \right]. \]  

(6.33)

The normalization condition (Equation 6.32) guarantees that \( \text{Var}(V) = \langle V(r)^2 \rangle - \langle V(r) \rangle^2 = 1 \), where \( \langle \cdot \rangle \) is the average over the real 2D-space. A graph of a potential \( V \) generated in this way is shown in Fig. 6.6(b).

To incorporate the exciton-disorder potential in the system Hamiltonian we add an additional term in the matrix \( \hat{h} \) (see Equation 6.25),

\[ \hat{h} = \left( \begin{array}{cc} \omega_X + \sigma V(\hat{x},\hat{y}) & \Omega_R \\ \Omega_R & -\omega_c + (\hat{n} + \frac{1}{2}) \Xi_x + (\hat{n} + \frac{1}{2}) \Xi_y - i \frac{\kappa}{2} \end{array} \right), \]  

(6.34)

where \( \hbar \sigma \) is the standard deviation of the energy distribution of excitons in the QW. Obviously, the exciton-disorder potential leads to coupling between different transverse-exciton modes, since the matrix elements

\[ \langle \hat{\Psi}_{m'n'}^X | V(\hat{x},\hat{y}) | \hat{\Psi}_{mn}^X \rangle = \frac{1}{2\pi} \Re \sum_{k_i,k_j} G(k_i,k_j) u\left(\sqrt{k_i^2 + k_j^2}\right) \langle \hat{\Psi}_{m'n'}^X | e^{i(k_i \hat{x} + k_j \hat{y})} | \hat{\Psi}_{mn}^X \rangle \]  

(6.35)

are non-zero.

We performed numerical simulations on a polariton box for different inhomogeneous QW-exciton linewidths (FWHM) \( \delta_X^{\text{inh}} = \sqrt{8 \ln(2)} \sigma \) and calculated the
The defining function for the random exciton potential was chosen \( \tilde{u} = |k|^4 \ e^{-|k|^2 \omega_0^2/40} \) (using \( \omega_0 = 2.5 \ \mu m \), being the cavity-spot size), supporting potential traps and hills with lateral extensions larger than 1 \( \mu m \).

(b) False-color image of the real-space exciton potential \( V \) with in-plane variance \( \text{Var}(V) = 1 \).

Resonant-transmission spectra, as the cavity-mode resonance was tuned across the QW-exciton center frequency, by numerically diagonalizing the system Hamiltonian. The laser-pump spot was chosen to overlap with the TEM\(_{00}\) cavity mode. The results are shown in Figure 6.7 for QW linewidths between \( \delta_{\text{inh}}^X = 0 \) and \( \delta_{\text{inh}}^X = \Omega_R \).

Clearly, the TEM\(_{00}\) lower-polariton mode hybridizes with higher-order transverse-polariton modes in the presence of disorder. For increasing QW linewidth, the mode experiences a blue-shift, while an ensemble of higher-order transverse modes appears, when the laser frequency is close to the exciton resonance. The laser-transmission intensity is separately shown in Fig. 6.7(d), when the TEM\(_{00}\) cavity mode is resonant with the exciton center frequency, for zero disorder (blue trace), \( \delta_{\text{inh}}^X = 0.5 \ \Omega_R \) (green trace) and for \( \delta_{\text{inh}}^X = \Omega_R \) (red trace). For an inhomogeneous exciton broadening

\[
\delta_{\text{inh}}^X < 0.5 \ \Omega_R
\]  

(6.36)

the blue-shift becomes negligible. We argue that this is the critical condition for the observation of pure and narrow lower-polariton lines. A few theoretical studies on polariton systems with Gaussian exciton disorder [185, 186] came to similar conclusions that the disorder potential is almost ‘transparent’, if the polariton resonance is not resonant with localized-exciton modes, i.e. the polariton Rabi splitting is clearly larger than the tail of the Gaussian distribution of QW-exciton states.

In the previously derived theoretical model we neglected a momentum dependence of the cavity-photon loss rate. In a real semi-planar DBR cavity the probability that a photon leaks through the plane mirror increases, if the photon has a large in-plane momentum. Based on a transfer-matrix approach simulating a cavity with the plane mirror consisting of 30 GaAs/AlAs DBR pairs with the stopband centered at the exciton resonance, we calculated the cavity-photon loss rate \( \kappa \) as a function of \(|k|\). The result is shown in Fig. 6.8. Clearly, the cavity-photon losses significantly increase...
for $|k| > 7 \mu m^{-1}$. This means that a disorder potential with domain sizes much smaller than $\frac{2\pi}{7} \mu m \approx 0.9 \mu m$ leads to polariton losses via coupling to higher-order transverse-polariton modes, which exhibit large in-plane momentum components\(^1\). In reality, the typical disorder-potential domain size (correlation length) in a QW is smaller than 100 nm [187]. Accordingly, the disorder-induced polariton hybridization of the TEM\(_{00}\) mode with higher-order transverse modes automatically leads to losses and a broadening of the resonance\(^2\).

Moreover, in the above model we assumed a symmetric (Gaussian) energy distribution of exciton states. However, the density of states of uncoupled QW excitons at frequencies $\omega > \omega_X$ is finite, leading to an asymmetric lineshape of the inhomogeneously-broadened exciton resonance in real systems. This additional

---

\(^1\)Note that in our implementation of a fiber-cavity system (see Chapter 7) a semiconductor-vacuum interface does not support the propagation of photons with transverse momentum $|k| > \frac{2\pi}{\lambda} \approx 7 \mu m^{-1}$, due to total internal reflection at the interface, leading to losses.

\(^2\)In the performed simulations above, we chose domain sizes larger than 1 $\mu m$, since the numerical calculations with smaller domain sizes would require more computer memory than available.
Figure 6.8.: Cavity losses as a function of the photon in-plane wave vector $k$. The plane mirror consists of 30 GaAs/AlAs DBR pairs, with the stopband centered at the exciton resonance at $\lambda_X = 900$ nm. The curved mirror is assumed to be perfectly reflecting. The curve is derived using a transfer-matrix model.

Polariton-scattering amplitude leads to a more pronounced broadening of the upper-polariton resonance in a cavity-coupled system \[188\]. In summary, the condition for small lower-polariton absorption rates lies in a far spectral detuning of the lower-polariton resonance from the Gaussian distribution of localized excitons. Increasing the Vacuum-Rabi splitting therefore yields a narrow lower-polariton linewidth. With these arguments and provided that Equation 6.36 is fulfilled, the lower-polariton linewidth on exact cavity-exciton resonance is predicted to be half the cavity-mode linewidth

$$\delta_{LP} = \frac{\omega_c}{2Q}. \quad (6.37)$$

### 6.5. Polariton quantum blockade in a photonic dot

Cavity polaritons are composite neutral bosons. While so far we have largely neglected polariton-polariton interactions, we have to take them into account, when polaritons are brought together so closely that the electronic substructure of the exciton part becomes relevant. If this typically repulsive dipolar or contact exciton-exciton interaction energy $U$ is so large that it exceeds the polariton linewidth $\delta$, the system becomes nonlinear at the level of single polaritons. If a single polariton is injected into such a polariton box using a resonant laser, the generation of a second polariton is no longer supported by the laser photon energy, due to the additional blue-shift of the transition. This effect was termed polariton blockade \[36\]. We will denote such microcavity systems in the strong polariton-polariton interaction regime photonic dots in the rest of this discussion.

In a simple approximation considering the optimal case of exactly parallel electron-hole dipoles, the repulsive interaction energy is given by
where $a_X \approx 10$ nm is the exciton Bohr radius and $r$ the mean distance between the excitons. In a micron-sized cavity ($r \approx 1 \mu m$) the interaction energy is only about $U_{\parallel} \approx 0.14 \mu eV$. Achieving $U/\delta > 1$ in QW microcavities therefore is a highly non-trivial task and to date, none of the implemented polariton boxes discussed in Section 6.4 has shown the polariton-blockade effect. To get a deeper insight into the difficulties for its observation, we calculate the interaction energy $U$ in a quantum description and assume a polariton-polariton contact interaction instead of the unrealistic case of parallel dipoles. This discussion primarily follows the argumentation in the paper by Verger et al. [36] and is based on resonant excitation via the cavity mode using a narrow-bandwidth cw laser. Second-order correlations of the emitted photon stream are analyzed for various different system parameters. As a starting point, the description of the QW microcavity system in a canonical-quantization picture, as already used in the previous section, is the most intuitive. The Hamiltonian is given by [36]

$$\hat{H} = \int d^2r \sum_{i,j=X,c} \hat{\Psi}^\dagger_i(r) \hat{h}_{ij} \hat{\Psi}_j(r)$$

(6.39)

$$- \left[ \frac{\hbar \Omega_R}{n_{sat}} \int d^2r \hat{\Psi}^\dagger_c(r) \hat{\Psi}^\dagger_X(r) \hat{\Psi}_X(r) \hat{\Psi}_X(r) + \text{h.c.} \right]$$

(6.40)

$$+ \frac{\hbar \epsilon}{2} \int d^2r \hat{\Psi}^\dagger_X(r) \hat{\Psi}^\dagger_X(r) \hat{\Psi}_X(r) \hat{\Psi}_X(r)$$

(6.41)

$$+ \int d^2r \ h F_p(r,t) e^{-i\omega_L t} \hat{\Psi}^\dagger_c(r) + \text{h.c.},$$

(6.42)

with the field operators $\hat{\Psi}_X(r)$ and $\hat{\Psi}_c(r)$ as defined in Section 6.4. The linear expression 6.39 describes the free Hamiltonian and the kinetic-energy terms, as well as the exciton-photon interaction, derived in Section 6.4, with $\hat{h}_{ij}$ given by Equation 6.24. A nonlinear term that corresponds to polariton bleaching, as discussed in Section 6.3.1, is described by expression 6.40: If the number of excitons in the potential approaches $n_{sat}$, the Rabi splitting vanishes. This term is much smaller than the polariton-polariton interaction term in typical microcavities [189] and will be neglected in the rest of this discussion. The interesting nonlinear term arising from exciton-exciton interactions is described by expression 6.41, where $\hbar \epsilon$ defines the repulsive contact-interaction potential. Finally, expression 6.42 contains the external-pump field $F_p(r,t)$, oscillating at the optical laser frequency $\omega_L$.

We found the cavity eigenmodes to be the TEM$_{mn}$ modes with the bosonic annihilation operators $\hat{\Psi}^e_{mn}$ (Equation 6.27) and Hermite wavefunctions $\phi_{mn}(\sqrt{2\pi} r/\omega_0)$ (assuming here that $\omega_0 \equiv \omega_{0x} = \omega_{0y}$). Due to the flat exciton dispersion, we expressed the exciton modes in the same basis with the annihilation operators $\hat{\Psi}^X_{mn}$. As previously discussed, the transverse-polariton modes are well resolved in energy. We refer to the fundamental cavity-mode frequency as $\omega_c$. For convenience, the fundamental eigenmodes $\Psi_{00}$ and $\hat{\Psi}^X_{00}$ will be referred to as $\hat{a}$ and $\hat{b}$.
\[ \hat{\Psi}_X(r) = \frac{\sqrt{2}}{\omega_0} \sum_{mn} \phi_{mn} \left( \frac{\sqrt{2}r}{\omega_0} \right) \hat{\Psi}_{mn}^X, \]

and restricting the calculations to the fundamental mode \( \phi_{00} \left( \frac{\sqrt{2}r}{\omega_0} \right), \) we find that

\[ \int d^2r \ \hat{\Psi}_X^†(r) \hat{\Psi}_X^†(r) \hat{\Psi}_X(r) \hat{\Psi}_X(r) = \frac{1}{\omega_0^2} \hat{b}^† \hat{b}^† \hat{b} \hat{b}. \] (6.43)

Defining \( \hbar \omega_{nl} = \frac{\hbar \epsilon}{\omega_0^2} \) and \( F_0(t) = \int d^2r \ F_p(r, t) \phi_{00}^* \left( \frac{\sqrt{2}r}{\omega_0} \right) \) and rewriting the Hamiltonian in the rotating frame described by the unitary transformation \( R(t) = e^{i\omega_L t(\hat{a}^† \hat{a} + \hat{b}^† \hat{b})} \) yields the effective Hamiltonian

\[ \hat{H}_{\text{eff}} = \hbar \left[ (\omega_c - \omega_L) \hat{a}^† \hat{a} + (\omega_X - \omega_L) \hat{b}^† \hat{b} + \Omega_R \left( \hat{a}^† \hat{b} + \hat{b}^† \hat{a} \right) \right. \]

\[ \left. + \frac{\omega_{nl}}{2} \hat{b}^† \hat{b} \hat{b} + F_0(t) \hat{a}^† + F_0^†(t) \hat{a} \right]. \] (6.44)

The system nonlinearity becomes evident from the term \( \hbar \omega_{nl} \hat{b}^† \hat{b} \hat{b}^† \hat{b} \), yielding the extra interaction energy \( \hbar \omega_{nl} \), if two excitons are present in the dot. Notably, the strength of the exciton-exciton interaction is proportional to the inverse of the cavity-spot area.

Using a master-equation ansatz including the damping terms corresponding to both the cavity-photon and exciton-spontaneous losses, the authors of Ref. [36] calculate the steady-state values of the second-order correlation function of the photon stream emitted by the cavity, \( g_2^{(\text{phot})}(t, t') \), for various different detunings and pump powers. They find that the cavity photons are antibunched, if the laser is slightly red-detuned with respect to the lower-polariton resonance \( E_{00}^- \). For a photon energy higher than \( E_{00}^- \), the emitted light is bunched. The effect is strongest, when the cavity mode is blue-detuned from the exciton resonance (red line), since the exciton content of the lower-polariton state is larger in this case.

Ultimately, the ratio of the system nonlinearity to the lower-polariton linewidth \( \omega_{nl}/\gamma_{LP} \) determines the strength of the antibunching effect. As shown in Ref. [36], the correlations at zero delay become significantly less than unity, once \( \hbar \omega_{nl} \) exceeds the polariton linewidth \( \hbar \gamma_{LP} \). The authors of this paper also show that second-order correlations remain more or less equal in pulsed excitation mode, if the pulses are Fourier-limited and the pulse duration coincides with the polariton lifetime.

From an engineering point of view, the goal for setting up a photonic dot lies in the implementation of a QW microcavity with strong mode confinement on the one hand, and a narrow lower-polariton linewidth on the other hand. Assuming that the system is on exact cavity-exciton resonance, the lower-polariton wavefunction with two quanta in the dot is given by \( |\Psi_{LP2}\rangle = \frac{1}{2} |2, 0\rangle - \frac{1}{\sqrt{2}} |1, 1\rangle + \frac{1}{2} |0, 2\rangle \) [65], and therefore

\[ \frac{\hbar \omega_{nl}}{2} \langle \Psi_{LP2}\rangle |\Psi_{LP2}\rangle |\Psi_{LP2}\rangle \approx \frac{\hbar \omega_{nl}}{4}. \]

The system nonlinearity is thus given by

\[ U = \frac{\hbar \epsilon}{4 \omega_0^2 \pi}. \] (6.45)

The exciton-exciton interaction potential is on the order of \( \hbar \epsilon \approx Ra_X^2 \), where \( R \) is the exciton Rydberg energy of about 10 meV and \( a_X \) is the exciton Bohr radius.
of about 10 nm [190]. Based on a realistic value $\hbar \epsilon = 1.5 \cdot 10^{-2} (\mu m)^2$ meV [36, 189] the condition $\hbar \gamma_{LP} < U$ writes

$$\frac{\omega_0^2}{\mu m^2} \cdot \frac{\hbar \gamma_{LP}}{\mu eV} \lesssim 1.2$$

(6.46)

Using the measured spot size of $\omega_0 \approx 2.5 \mu m$ of our presently implemented fiber-cavity box (see Chapter 7), the nonlinearity is $U \approx 0.2 \mu eV$ and the unrealistic cavity $Q$-factor of about $3.4 \cdot 10^6$ would be required, assuming the ideal case that the polariton linewidth is only determined by the homogeneous system broadening (see the discussion in Section 6.4.1).

### 6.5.1. Feshbach blockade

As derived in the previous section, the implementation of a system exhibiting a significant polariton-energy shift due to exciton-exciton interactions, turns out to be very demanding. Here, a well-known technique widely used in cold-atom experiments could help to increase the nonlinearity: When the sum of the energies of two colliding atoms is equal to the energy of a bound diatomic-molecular state, their scattering-cross section is significantly enhanced, provided there is a coupling-mechanism between the free atoms and the molecular bound state. Using a magnetic field, the energy of the molecular state can be tuned to this so-called Feshbach resonance [191]. In a very similar manner, the energy of microcavity-polariton states could be tuned to the QW-biexciton resonance and in this way enhance polariton-polariton interactions [190, 192]. The following discussion is based on the publication by Carusotto et al. [190].

On the basis of a Hamiltonian in a second-quantized form, the authors derive the coupling strength for the exciton-photon-biexciton coupling, describing the formation of a biexciton by capturing a photon within the distance of the biexciton-Bohr radius $a_b$ to an existing exciton. The calculation is restricted to the TEM$_{00}$ polariton state. Furthermore, the method relies on a well-resolved linear-polarization splitting of the fundamental polaritons. In the following, only one linear-polarization state will be considered. Writing $\hat{p} \equiv \hat{p}^0_0$ (see Equation 6.28) and expressing a biexciton-annihilation operator $\hat{b}$ in the same basis $\phi_{00}(\mathbf{r})$, the resulting Hamiltonian is given by

$$\hat{H} = E^- \hat{p}^+ \hat{p} + E_b \hat{b}^+ \hat{b} + \frac{U}{2} \hat{p}^+ \hat{p}^+ \hat{p} \hat{p} + G \left( \hat{b}^+ \hat{p} \hat{p} + \hat{p}^+ \hat{p} \hat{b} \right),$$

(6.47)

where $E^-$ is the lower-polariton energy and $E_b$ the biexciton energy. The additional nonlinear term scales with the polariton-pair biexciton-coupling strength $G$, given by

$$G \approx \frac{1}{2} \hbar \Omega_{Ra_b} u^X_{LP} u^c_{LP} \overline{\hbar} \left( \int d^2r \left| \phi_{00}(\mathbf{r}) \right|^4 \right)^{1/2}$$

(6.48)

where $u^X_{LP}$ and $u^c_{LP}$ are the Hopfield coefficients describing the excitonic and photonic content of the polariton state, and $\overline{\hbar}$ is a form factor of the biexciton-scattering potential that is on the order of unity. In a cavity-tunable polariton box, the lower-polariton resonance can be tuned in energy, whereas the biexciton-transition energy
is a constant number, that is about 2 meV lower than the exciton energy. When tuned into resonance (see Figure 6.9(a)), the biexciton is expected to hybridize with a lower-polariton pair, leading to a splitting of the polariton-pair state, as illustrated in Fig. 6.9(b).

![Figure 6.9. Polariton Feshbach resonance.](image)

(a) Level dispersion of the lower and upper polaritons for a cavity-mode blue-detuning of $E_c - E_X = 1.26$ meV, using $\hbar \Omega_R = 1.5$ meV. The lower polariton is in Feshbach-resonance with the biexciton transition. (b) The polariton-pair state is split into two new system states, due to the hybridization with the biexciton state. The levels are split by $2\sqrt{2} G$ and additionally shifted by the polariton-polariton interaction energy $U$. As a result, the energy ladder is anharmonic.

A value for $G$ is found to be about $1.5 \, \mu$eV, using the realistic values $\hbar \Omega_R = 1.5$ meV, $a_b = 20$ nm, $u_{LP}^X u_{LP}^c = 0.4$, $\hbar = 1$ and the measured waist radius $\omega_0 \approx 2.5 \, \mu$m of our fiber-cavity box. The additional line-shift of the polariton-pair state is $\sqrt{2} G \approx 2 \, \mu$eV and thus a factor of 10 larger than the nonlinear contribution $U$ from direct polariton-polariton interactions.

In the ideal case of pure homogeneous line broadening (see Section 6.4.1), a cavity $Q$-factor of about 300 000 would be required to observe optical signatures of polariton blockade. This value could be achieved using high-quality mirrors.
7. All-optical polariton boxes in a tunable fiber cavity

In this chapter, our approach to the realization of a photonic dot using a semi-integrated fiber-based cavity system is described. I discuss first results of QW cavity-QED in the strong-coupling regime and the observation of long-lived polaritons. Moreover, signatures of polariton lasing are presented. Part of the results discussed in this chapter are currently being prepared for publication [63].

7.1. Towards the implementation of a photonic dot

In Chapters 4 and 5, we demonstrated a system showing single-photon nonlinearities based on the strong coupling of a single QD to the mode of a photonic-crystal cavity. The QD is itself a two-level system showing perfect antibunching; the nonlinearity of the coupled device therefore is inherited from the nonlinearity of the quantum emitter.

In contrast, QW-cavity polaritons are per-se linear systems with a harmonic energy ladder, as discussed in Chapter 6. To activate sizable polariton-polariton interactions, a strong three-dimensional confinement of the polariton wavefunction has to be introduced, e.g. by incorporating a strong photonic confinement, as we figured out in Section 6.5. Such a system could enter the strong-confinement regime featuring the polariton-blockade effect.

A motivation behind implementing such a type of nonlinear system is the great flexibility of photonic engineering. Here, we demonstrate a new method for the implementation of tight polariton confinement, which at the same time allows us to tune both energy and lifetime of the polaritons. Our system consists of a concave dielectric mirror at the end of a fiber tip and a semiconductor Bragg mirror with an optically active quantum-well layer on top, as illustrated in Figure 7.1. The Fabry-Pérot-type cavity defines the light modes and thereby imprints the resulting polariton wavefunction. Tuning the length of the cavity allows both for adjustment of the polariton lifetime and the precise tuning of the polariton energy at one and the same spot on the sample. The spot size on the sample surface was measured to be about $\omega_0 \approx 2.5 \, \mu m$. Using a single InGaAs quantum well, we observed an avoided-level crossing as a function of cavity length giving a vacuum Rabi splitting of 4.2 meV. Using a 4-QW system, we measured polariton lifetimes up to 100 ps. In a device with 9 coupled QWs, we observed signatures of polariton lasing when pumping the system strongly using an off-resonant laser.
Figure 7.1.: Tunable fiber-cavity boxes. Semi-integrated microcavity for the creation of confined polaritons consisting of a curved fiber-end mirror and an planar sample mirror. The inset illustrates part of the two Bragg mirrors. The quantum well is sandwiched within the surface layer of the semiconductor DBR, in a way that the electric field at its position is maximum. The curvature of the fiber mirror is exaggerated.

Several groups have previously employed a similar setup [193] in connection with single quantum emitters, such as quantum dots [59, 60, 194] or dye molecules [195], however, these experiments so far have not reached the single-emitter strong-coupling regime.

7.2. Experimental setup and methods

The heart of our microcavity setup (displayed in Figure 7.1) is a concave highly-reflective mirror fabricated by CO$_2$-laser ablation at the tip of a single-mode optical fiber [58] and coated with a DBR made of the two materials silicon dioxide (SiO$_2$) and tantalum pentoxide (Ta$_2$O$_5$) with the stopband centered at 900 nm. These fiber-based mirrors were fabricated in collaboration with the research group of Prof. Jakob Reichel at the École normale supérieure (ENS) in Paris. Figure 7.2(a) shows an SEM image of a laser-machined fiber-end surface, as published in Ref. [58]. The shallow depression due to laser ablation has approximately Gaussian shape (see Figure 7.2(b)) and is only a few $\mu$m deep. The radii of curvature at the center of the depression of the fiber mirrors used in our experiments were between $R \approx 60 \mu$m and $R \approx 75 \mu$m.

The sample part of the semi-integrated system was grown by molecular beam epitaxy and consists of a Bragg mirror containing 30 pairs of AlAs/GaAs layers (see Section 6.2) with a single InGaAs quantum well on top ($\lambda_X \approx 891$ nm), sandwiched between two GaAsspacer layers optimized for maximum coupling of the QW to the cavity mode.

When the two systems are mounted a few micrometers away from each other, 3D-confined cavity modes form. The electric-field component of the cavity mode,
7.2. Experimental setup and methods

Figure 7.2.: Laser-machined fiber end. (a) SEM image of a laser-machined fiber-end surface with visible concave depression. (b) The depression has approximately Gaussian shape. When approximating the center part by a sphere, the depression can be characterized by a radius of curvature $R$. The total depth $z_t$ is only a few $\mu$m. The images are taken from Ref. [58].

when the two components almost touch, is shown in the inset of Fig. 7.1.

7.2.1. Optical design

A standard SM fiber (P3-830A-FC-5 from Thorlabs) was spliced to the ‘non-mirror’ end of the cavity fiber. It enters a 2 inch wide dip-stick filled with helium exchange gas at a pressure of 20 mbar through a Teflon® feedthrough [196] (see Figure 7.3(a)) and is guided down to a scanner unit. As displayed in Fig. 7.3(b), the fiber makes a U-turn, such that the fiber mirror faces upwards. The fiber end was glued to a special holder in a way that only about the last 100 $\mu$m of the fiber end sticks out of the holder, as can be seen in the photograph (Figure 7.3(c)). Using a slip-stick positioner (linear positioner ANPx101/RES from attocube systems AG) the vertical position of the fiber-end surface can be adjusted with sub-nm resolution (positioner $z$ in Fig. 7.3(b)), by applying a voltage between 0 and 150 V to the piezo actuator, yielding a total travel range of about 1.5 $\mu$m. For a coarse positioning of the vertical fiber position, the stepping mode of the positioner is applied, allowing for a total movement over 5 mm.

The samples are glued to a sample holder using vacuum grease, such that the mirror side is facing downwards. The illuminated spot on the sample is chosen using two linear positioners in $x$ and $y$ direction (ANPx101/RES from attocube systems AG). Having an all-over travel range of 5 mm per positioner, several samples can be mounted on the holder. In this way, we can switch between samples during a single cool-down. All positioners are equipped with a resistive read-out with sub-$\mu$m precision.

The dip-stick is directly inserted into a wide-neck dewar filled with liquid helium, giving a system temperature of $T = 4.2$ K for 2 – 3 weeks. For mechanical damping and acoustic-noise rejection, the dewar sits on vibration-absorbing feet and is enclosed in a soundproof box. In this way, the cavity length is stable to less than 10 pm, without applying active-stabilization schemes.

The whole system is based on our own design and was machined at ETH Zürich.
Figure 7.3.: The fiber-microscope setup. (a) The dip-stick containing the fiber-cavity setup is immersed into liquid helium. Arrows indicate the optical paths in reflection and transmission geometries. Drawings are not to scale. (b) The fiber scanner (to-scale image). The laser light enters the system through a SM fiber. The distance between fiber mirror and sample surface is precisely adjusted using the $z$-positioner. The backside-polished sample is glued at the bottom of the sample holder, with the DBR facing downwards. The position of the lens can only be adjusted, before the system is cooled down. (c) Microscope image showing the fiber end positioned close to the sample surface. The mirror image of the fiber tip is clearly visible.
7.2.2. Photoluminescence spectroscopy

Photoluminescence (PL) measurements are carried out by sending off-resonant excitation light (typically at 780 nm, see Section A.1) through a fiber-based beam splitter (BS) and guiding it into the cryostat (see Figure 7.3(a)). The light, which is spectrally clearly outside the stopband of the fiber mirror (compare Section 6.2), is transmitted through the mirror and subsequently absorbed by the bulk GaAs that is close to the surface of the semiconductor chip. Electron-hole pairs are generated and form bulk excitons, whereby a few excitons diffuse into the QW over a wide area of several µm diameter. If a cavity-mode resonance is close to the exciton resonance, high-momentum excitons relax into the lower-polariton ground state via exciton-phonon and exciton-exciton interactions. Moreover, due to cavity-feeding mechanisms, other (lower-lying longitudinal) cavity modes, which are red-detuned by up to about 20 nm from the exciton resonance, are populated as well, in analogy to cavity feeding with QDs (see Section 2.5.2). Upon recombination of ground-state polaritons, part of the luminescence is emitted via the far-field of the cavity mode. Since the reflectivity of the semiconductor mirror is about an order of magnitude lower than the one of the dielectric fiber mirror, most of the photons leave the polariton box through the upper (semiconductor) mirror. A smaller fraction of the luminescence leaks through the fiber mirror and is partly coupled to the guiding mode of the SM fiber (see Figure 7.3(a)). After passing the fiber-based BS, these photons are sent through a long-pass filter to a high-resolution spectrometer (see Section B.1).

We also performed PL measurements in transmission geometry, yielding a larger collection efficiency. These measurements however require a backside-polished sample. In this case, the light transmitted through the sample is collimated by a lens. The beam is directed out of the chamber in a free-space geometry through a wedged window (see Figure 7.3(a)), mounted in the top-part of the dip-stick.

We also use PL measurements to characterize the cavity length. By measuring the free spectral range between two fundamental cavity modes at wavelengths $\lambda_1$ and $\lambda_2$, the cavity length can be determined using the basic relation $L = \frac{\lambda_1 \lambda_2}{2(\lambda_1 - \lambda_2)}$. For a more precise determination of the length that takes into account the penetration of the field into the mirrors, we simulated the cavity-mode field using a transfer-matrix approach [157].

7.2.3. Resonant transmission measurements

Using a resonant light source (either laser light or broad-band white light) the system transitions can be directly probed and complicated relaxation mechanisms that occur in off-resonant excitation schemes can be avoided. To this end, we couple the light into the system via the fiber port, as is done in PL experiments. If not resonant with a cavity (or polariton) mode, the light is simply back-reflected from the fiber mirror and does not interact with the microcavity system at all. Light that is resonant with the cavity (polariton) mode is injected into the cavity and mainly leaks out of the system through the upper (semiconductor) mirror, after a short delay corresponding to the lifetime of the cavity photon (polariton).

Figure 7.4(a) displays the resonant-transmission spectrum of one linear polarization state of a mostly cavity-like fundamental polariton transition in a sam-
ple with a single embedded QW. The data was recorded using the method described in Section B.2. The narrow lineshape of the resonance reveals a cavity-quality factor of \( Q \approx 74,000 \), which corresponds to a finesse of about 3700.

A method to retrieve the full transmission spectrum at once is the use of a broadband light source. To this end, the excitation light is generated using a pulsed white-light source (SC-500-2 from Fianium Ltd.) and a band-pass filter (here, a Thorlabs FB900-10 is used), resulting in a continuous spectrum of about 10 nm bandwidth at a center wavelength of 897 nm (see inset of Fig. 7.4(b)). We measured the transmission spectrum of the single-QW sample in this way using a spectrometer for detecting the transmitted light, while scanning the cavity-mode resonance across the exciton resonance. The results are shown in Fig. 7.4(b). The fundamental lower-polariton line vanishes for wavelengths shorter than 892 nm, as the exciton content of the polariton wavefunction increases (compare Fig. 7.8(a)).

![Figure 7.4: Resonant-transmission measurements on a single-QW sample. (a) Resonant-transmission signal from one of the two polarization-split lower-polariton resonances. The spectrum was recorded using a tunable cw laser. Here, the cavity mode was far red-detuned from the exciton resonance and the polariton therefore almost cavity-like. The line-shape is close to a Lorentzian and corresponds to a cavity \( Q \)-factor of about 74,000. (b) Resonant-transmission measurements using a pulsed broad-band light source. The transmitted light was detected with a spectrometer, while the cavity length was decreased by tuning the piezo voltage of the \( z \)-positioner. The prominent line corresponds to the fundamental lower-polariton resonance. The inset shows a spectrum of the broad-band light that excited the system through the fiber. The effective cavity length in both (a) and (b) was about 9 \( \mu \)m.](image)

### 7.3. Characterization of the fiber-based cavity

Many of the intrinsic properties of the cavity can be inferred from PL spectra, when the cavity mode is only weakly coupled to an emitter and therefore luminesces

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1In contrast to the results obtained in PL spectroscopy (compare Fig. 7.8(a)), the first-order (TEM\(_{01/10}\)) modes are hardly visible, whereas the second-order modes again appear in the spectrum. This behavior could be due to the node in the center of the TEM\(_{01/10}\) orbitals, preventing these modes from being efficiently coupled to the SM fiber outside the cryostat.
through the cavity-feeding effect (see Section 2.5.2). Figure 7.5(a) displays a typical spectrum of the device based on a substrate with many coupled InAs QDs, recorded for a cavity length of \( L_{\text{eff}} \approx 43 \, \mu m \). Besides two fundamental TEM\(_{00}\) modes, spectrally separated by a free spectral range (FSR), we also observed higher-order transverse modes (here, up to the second order), due to their finite spatial overlap with the guiding fiber mode. The fact that we detected odd cavity modes (i.e. TEM\(_{mn}\) modes with \( m + n \) being an odd integer) suggests that the core of the SM fiber is significantly misaligned with the center of the spherical depression. For a semi-planar cavity, the higher-order mode frequencies are given by [131]

\[
\omega_{qmn} = \frac{\pi c}{L_{\text{eff}}} \left[ q + \left(m + \frac{1}{2}\right) \pi^{-1} \cdot \arccos \left( \sqrt{1 - \frac{L_{\text{eff}}}{R_x n_{\text{eff}}}} \right) \right. \\
\left. + \left(n + \frac{1}{2}\right) \pi^{-1} \cdot \arccos \left( \sqrt{1 - \frac{L_{\text{eff}}}{R_y n_{\text{eff}}}} \right) + \frac{2L_{\text{DBR}}}{\lambda_0} \right], \quad (7.1)
\]

where \( q \geq 0 \) is the integer longitudinal-mode quantum number and \( m, n \geq 0 \) are the integer transverse-mode quantum numbers. \( L_{\text{eff}} \), \( L_{\text{DBR}} \) and \( \lambda_0 \) are the effective cavity length, the mirror-penetration depth and the DBR center wavelength, as defined in Section 6.2. The quantities \( R_x \) and \( R_y \) are the radii of the curved mirror along the \( x \)- and \( y \)-axes and \( n_{\text{eff}} \) is an effective refractive index that depends on the geometry of all the combined materials. Clearly, we observed a splitting \( \hbar \Delta \omega_{\text{TM}} \approx 280 \, \mu eV \) of the higher-order transverse modes with a constant sum \( m + n \) (e.g. by looking at \( \Delta \omega_{\text{TM}} = |\omega_{11} - \omega_{20}| \)), suggesting that the curved mirror is slightly elliptic. Assuming that \( n_{\text{eff}} \approx 1 \), since the light travels mainly in vacuum for a cavity length \( L_{\text{eff}} = 43 \, \mu m \), we extracted the radii of curvature \( R_x \approx 93 \, \mu m \) and \( R_y \approx 106 \, \mu m \).

All the cavity modes moreover exhibit a polarization splitting that is significantly larger than the cavity-mode linewidth. Figure 7.5(b) shows the polarization splitting of the TEM\(_{00}\) modes, when the upper and lower mirrors are almost touching (\( L_{\text{eff}} \approx 7 \, \mu m \)). The observed polarization splitting of \( \hbar \Delta \omega_{\text{pol}} \approx 122 \, \mu eV \) could be the result of a slight birefringence of the semiconductor (GaAs, InAs and AlAs) material, yielding a slight difference of the effective optical cavity length \( L_{x_{\text{eff}}}^x \) and \( L_{y_{\text{eff}}}^y \) for light polarized along the \( x \) and \( y \) axis,

\[
\frac{L_{x_{\text{eff}}}^x}{L_{y_{\text{eff}}}^y} = \frac{\omega_{y_{00}}}{\omega_{x_{00}}} = 1 + \frac{\Delta \omega_{\text{pol}}}{\omega_{00}} \approx 1 + 9 \cdot 10^{-5}. \quad (7.2)
\]

Note that the polarization splitting decreases for longer cavities, since the optical path-length difference is merely a result of the GaAs-spacer layer, the QW and the semiconductor DBR.

The quality factor of the cavity modes increases with the length of the gap between the fiber tip and the substrate. A more relevant figure of merit for this type of cavity is the finesse, which becomes length-independent for long enough cavities. Depending on the sample, we measured a finesse ranging from 3500 to 6500. The transverse confinement of the exciton is related to the Gaussian waist of the TEM\(_{00}\) mode. We measured spot sizes as small as 2.5 \( \mu m \) with a fiber mirror having a radius of curvature of \( R \approx 75 \, \mu m \), as demonstrated in the following section.
Figure 7.5: Typical cavity spectrum. (a) A PL spectrum of the cavity modes ($L_{\text{eff}} = 43 \, \mu\text{m}$), coupled to InAs QDs under off-resonant excitation. In addition to the fundamental transverse modes (TEM$_{00}$ modes) of two different longitudinal cavity modes separated by a FSR, higher-order transverse cavity modes are visible. (b) Polarization splitting of the TEM$_{00}$ modes for a cavity length $L_{\text{eff}} \approx 7 \, \mu\text{m}$. A differential-transmission measurement of the cavity modes is shown as a function of laser-photon energy, yielding a polarization splitting of about 122 $\mu$eV.

7.3.1. Cavity spot size

The Gaussian beam waist is located directly at the sample surface. To determine the cavity-spot size, the same QD chip as used for determining the cavity spectrum was mounted. QDs have lateral dimensions much smaller than the cavity-mode waist and can therefore be used to probe the local optical intensity. We set the cavity length to $L_{\text{eff}} = 43 \, \mu\text{m}$ and performed PL spectroscopy at a laser power above QD saturation. The resulting spectrum is displayed in the bottom of Fig. 7.6(a) and shows three different cavity-mode resonances, corresponding to the TEM$_{00}$ mode ($\approx 977 \, \text{nm}$), the TEM$_{01}$/10 modes ($\approx 974 \, \text{nm}$) and the TEM$_{02}$/11/20 modes ($\approx 972 \, \text{nm}$). We then modulated the cavity length (in $z$-direction) in a 'zig-zag' waveform at a rate of 4 Hz and with a peak-to-peak displacement of about 58 nm. As a result, the cavity-mode resonances were ‘smeared out’ over a spectral bandwidth of about 1.3 nm, as indicated by the yellow arrows. Consequently, when recording PL spectra during an integration time of 1 s, all the QDs that have a resonance within this bandwidth light up, due to Purcell-enhanced spontaneous emission. While taking PL spectra, the sample was laterally displaced (using the slip-stick $x$-positioner), such that various QDs were moved through the cross section of the cavity spot. PL spectra as a function of sample displacement are displayed in the false-color plot in Fig. 7.6(a). Within the plotted area, indicated by the red boundaries, several bright spots can clearly be identified as single QDs that were moved in and out of the cavity-spot area.

Plotting the integrated luminescence of the labeled QD1 as a function of sample displacement $x$ yields the cavity-spot profile in this direction. As shown in
Figure 7.6.: Measuring the cavity-spot size. (a) PL spectra of a sample containing self-assembled InAs QDs, recorded while displacing the sample horizontally (in $x$-direction). Bright spots indicate QDs moving in and out of the cavity mode, as the sample is moved. (b) Cavity luminescence as a function of sample displacement at three different wavelengths (vertical cuts in (a) corresponding to three different QDs). The different transverse-mode profiles can clearly be distinguished. The fundamental cavity mode is Gaussian with a waist radius of $\omega_0 \approx 2.7 \, \mu m$, extracted from a fit (red trace). The cavity length is 43 $\mu m$.

Figure 7.6(b), the shape of the spot is perfectly Gaussian with a fitted waist radius\(^2\) of $\omega_0 \approx 2.7 \, \mu m$. A very similar result was measured when displacing the sample in $y$-direction. The profiles of higher-order cavity modes look more complicated (see mode profiles of QD2 and QD3). We attribute these shapes to cuts through Hermite-Gaussian cavity modes. Note that the red ‘boxes’ in Figure 7.6(a) are ‘tilted’, due to a slight wedge of the sample: When displacing the chip in $x$-direction, the cavity length slightly increases, shifting the cavity-mode resonances towards longer wavelengths.

The method for measuring cavity-mode intensity profiles relies on the fact that (a), the carriers generated by the off-resonant laser diffuse over an area that is much larger than the cavity-spot size (we assume the migration length of the carriers to be around 10 $\mu m$) and (b), that the laser-pump power is above saturation, such that each dot gets re-populated immediately after a spontaneous decay. Provided that these conditions are satisfied, the rate at which luminescence photons are collected via the cavity mode is $\gamma_c (x) = 4g (x)^2 / \kappa$ (see Equation 2.17). The coupling strength $g (x)$ of a QD is proportional to the spatial-field overlap $g (x) \propto |\phi (x)|$ with the cavity mode (see Equation 2.7), hence $\gamma_c (x) \propto |\phi (x)|^2$, so the photon-collection rate of a QD is proportional to the cavity-mode intensity at the dot position.

The measured waist size $\omega_0 \approx 2.7 \, \mu m$ slightly deviates from the predicted value of $\omega_0 \approx 3.3 \, \mu m$ using the relation

\(^2\)The transverse intensity profile at the beam waist is given by $I (r) = I_{max} e^{-2r^2/\omega^2}$, where $r$ denotes the distance from the beam axis.
with the experimentally determined values of the cavity length \( L = 43 \, \mu m \) and the mirror-radius of curvature \( R = 75 \, \mu m \) (inferred from interferometric images of the fiber-end surface) as input (see blue trace in Figure 7.7). For \( R = 58 \, \mu m \) (red trace) the measured data point (red bullet) would agree with the result from Equation 7.3. A prediction of the spot size for short cavities in our typical working range between 6.5 \( \mu m \) and 10 \( \mu m \) yields a value of \( \omega_0 \approx 2.5 \, \mu m \), as shown in Fig. 7.7.

**Figure 7.7.: Cavity-spot sizes.** The spot size \( \omega_0 \) as a function of cavity length for a semi-planar cavity, derived from Equation 7.3. The spot-size dependence for two different radii of curvature of \( R = 75 \, \mu m \) (blue trace) and \( R = 58 \, \mu m \) (red trace) are shown. The red bullet corresponds to the measured value. The shaded area denotes the cavity length at which most of the experiments were conducted. The shortest possible cavity length of about 4.2 \( \mu m \) is indicated by the dashed line and yields a waist size of about 2 \( \mu m \). The inset illustrates the semi-planar cavity with the beam waist at the position of the planar mirror, where the QW is located.

From the theoretical prediction for the spot size (see Equation 7.3), we estimate that a minimal waist of about 1.2 \( \mu m \) at a wavelength of 900 nm is within reach, corresponding to \( R = 10 \, \mu m \) and a cavity length \( L_{\text{eff}} = 5 \, \mu m \). This value for the confinement-length scale of polaritons is comparable to or even better than currently available in mesas [46] and micropillars [44].

### 7.4. Observation of microcavity polaritons

In order to demonstrate strong coupling of quantum-well excitons to the modes of the semi-integrated cavity, we approached the fiber tip close to the sample surface (less than 500 nm from touching) and recorded the PL emission upon excitation with a 780 nm laser while varying the cavity length around this offset position. Resulting
PL spectra using two different samples containing 1 QW and 9 QWs respectively, are plotted in Figure 7.8.

**Figure 7.8.: Avoided-level crossings.** (a) False-color plot of cavity-length dependent PL spectra of the strongly-coupled system with a single QW, centered at 890.9 nm under off-resonant excitation at 780 nm. The fundamental cavity mode and a higher-order mode were tuned across the exciton resonance resulting in different pairs of LP and UP branches. The red dashed line indicates the position of the z-actuator where the TEM$_{00}$ cavity mode is on exact resonance with the exciton wavelength. The corresponding cut is shown in (b), clearly demonstrating the upper (UP$_{00}$) and lower (LP$_{00}$) polariton modes. A spectral line corresponding to the mutually overlapping higher-order upper-polariton resonances UP$_{01/10}$ is visible as well. The Rabi splitting between the polariton modes is about $2 \hbar \Omega_R \approx 4.2$ meV. (c) PL spectra of a sample with 9 embedded QWs, centered at 899.8 nm and coupled to several cavity modes. The resonance condition between the fundamental cavity mode and the QW excitons is indicated by the red dashed line. (d) The corresponding PL spectrum reveals a Rabi splitting of $2 \hbar \Omega_R \approx 8.7$ meV between the fundamental upper and lower-polariton modes. The photoluminescence was collected through the excitation fiber in both cases. The intensities are given on a logarithmic scale.

The data displayed in Fig. 7.8(a) clearly exhibit an avoided-level crossing as a function of effective cavity length for all the transverse cavity modes, demonstrating the formation of polariton branches. The higher-order transverse modes couple to the QW excitons with approximately the same strength as the fundamental TEM$_{00}$ mode as is theoretically expected, due to the very weak dependence
of the coupling strength on the excitonic momentum. Figure 7.8(b) shows a cut through the 2-dimensional map in (a), corresponding to the case, when the fundamental cavity mode is exactly in resonance with the exciton (red dashed line in (a)). The Rabi splitting between the upper and lower-polariton modes is about $2\Omega_R \approx 4.2$ meV. We also confirmed the existence of strong exciton-cavity coupling in resonant-transmission spectroscopy directly probing the available density of states.

Figure 7.8(c) displays a PL spectrum, taken in the same configuration as the one shown in (a), but using a sample with 9 cavity-coupled QWs. Transverse lower-polariton modes up to the third order are visible as well. The spectrum on cavity-exciton resonance (denoted by the red dashed line in (c)) is shown in (d). The upper and lower fundamental polariton modes can clearly be identified and are split by $2\Omega_R \approx 8.7$ meV. Theoretically, we expect a splitting that is $\sqrt{9} = 3$ times larger than the Rabi splitting of the single-QW sample (see discussion in Section 6.1.1). In the real system however, the design is such that there are three triplets of QWs coupled to three different cavity-field antinodes. The two outermost QWs of each triplet are not maximally coupled to the field and therefore contribute less to the Rabi splitting. Moreover, the spacer layer of the 9-QW sample is one optical wavelength thicker than the one of the single-QW sample, yielding a longer cavity. These two effects together result in a Rabi splitting that is only about a factor of 2 larger than the single-QW Rabi splitting.

### 7.5. Long-lived polaritons

Long polariton lifetimes are e.g. beneficial in achieving a considerable ground-state population when pumping the system off-resonantly, such that the polariton lasing transition is shifted to lower pump powers (see Section 7.6). In the light of our goal to observe the polariton-blockade effect, a long lifetime is accompanied with a narrow homogeneous linewidth that is necessary to resolve the polariton-polariton interaction energy $U$. Ultimately, the coherence of the system is limited by the polariton lifetime.

To measure polariton lifetimes we excited the system via the fiber part with pulsed light of about 10 nm bandwidth in the same way as discussed in Section 7.2.3. First, we made sure that only one (the fundamental) polariton line appears in the transmitted spectrum. We then sent the light to an APD (id100-20 APDs from ID Quantique SA) to measure the ring-down time of the polariton mode that repeatedly got populated by the light pulses at a rate of 20 MHz. The average power of the broad-band pulses was about 160 nW. Notably, most of the light was simply reflected back by the fiber mirror. Based on the narrow polariton linewidth and on the fact that the fiber mirror is about an order of magnitude more reflective than the semiconductor mirror, we estimated the number of photons per light pulse injected into the cavity to be less than one. A detailed description of the conduction of lifetime measurements is provided in Section B.4.2.

We measured the exponential ring-down time of the lower polariton in a sample consisting of four QWs and an effective cavity length of 14.5 $\mu$m for different cavity-mode detunings from the exciton. At this length, the Vacuum-Rabi splitting was
7.5. Long-lived polaritons

$\Omega_R \approx 5.8$ meV. The results are shown in Fig. 7.9(a). Probably due to polariton scattering at localized QW excitons, the lifetimes at lower-polariton wavelengths shorter than 905 nm are clearly reduced. Obviously, a long lifetime and a high exciton content cannot be achieved simultaneously. Lifetimes of about 60 ps were measured at a cavity-mode red-detuning of about 1.05 $\Omega_R$ with respect to the exciton, yielding an exciton content$^3$ of the lower polariton of about 27%. For reference, Fig. 7.9(b) displays PL spectra recorded close to the cavity-exciton resonance.

**Figure 7.9.** Polariton lifetimes as a function of cavity-mode detuning from the exciton. (a) Using a pulsed white-light source, the lifetime of the fundamental lower polariton was recorded, yielding lifetimes up to 74 ps (blue bullets). The red line illustrates the exciton content of the lower polariton that was estimated based on the PL spectrum shown in panel (b). The gray dotted line denotes the spectral position of the LP, for which the lifetime data in Figure 7.10 were recorded. (b) False-color plot of cavity-length dependent PL spectra recorded in transmission geometry. The dashed ellipse indicates the fundamental lower-polariton resonance, for which the lifetimes in (a) were measured. The sample contains 4 QWs, spectrally centered at around 901.4 nm. The effective cavity length was about 14.5 $\mu$m. The error bars in (a) correspond to 95% confidence bounds for the fits of the polariton lifetimes.

The central feature of our system compared to other polariton experiments is its tunability. This enables the in-situ control of the cavity-exciton detuning, making the observation of an avoided-level crossing in one and the same spot of the sample possible. More importantly, when changing the cavity length over a much wider tuning range, the polariton lifetime can be significantly prolonged, while only moderately reducing the coupling strength. To this end, we measured the lower-polariton lifetime for different cavity lengths, but made sure that the LP-resonance wavelength was 904.8 nm in each measurement corresponding to the compromise between low disorder scattering and high exciton content, as discussed previously. Figure 7.10 illustrates this behavior. The results demonstrate ultra-long polariton lifetimes of up to 100 ps for an overall cavity length of about 28 $\mu$m. We confirmed

$^3$The exciton content of the lower-polariton wavefunction is given by $c_{LP}^{X} = \frac{1}{2} \left( 1 + \frac{\delta}{\sqrt{\Omega_R^2 + \delta^2}} \right)$, where $\delta = \omega_c - \omega_X$. 

that the device was still in the strong-coupling regime at this length, yet with a reduced Rabi splitting\(^4\) of about \(2\Omega_R \approx 4.3\) meV. Accordingly, we estimated the exciton content of the resulting polariton to be about 16\%. Figure 7.10(b) displays an example of the measured ring-down times of the LP, in this case at a length of 32.5 \(\mu\)m (blue solid trace). The data is ‘washed out’, due to the finite APD timing jitter of about 40 ps. The decay was accordingly fitted taking into the account the APD response-time function (dashed trace), yielding a polariton lifetime of 101.1 ps (red trace).

\[\Omega_R \propto \frac{1}{\sqrt{L_{eff}}},\]

The coupling strengths as a function of length follow the relation \(\Omega_R \propto \frac{1}{\sqrt{L_{eff}}},\) which is confirmed by PL measurements.

\[4\]The coupling strengths as a function of length follow the relation \(\Omega_R \propto \frac{1}{\sqrt{L_{eff}}},\) which is confirmed by PL measurements.

Figure 7.10.: Tuning the lower-polariton lifetime. (a) Lifetime of the lower-polariton resonance in the 4-QW sample as a function of cavity length. The lower-polariton resonance was chosen at \(\lambda_{LP} = 904.8\) nm (compare Fig. 7.5(a)). Up to a length of 28 \(\mu\)m, the lifetime increased linearly with length, as expected. (b) Measured lifetime trace (blue solid line) at a cavity length of 32.5 \(\mu\)m using pulsed broad-band light for excitation. The dashed line shows the time response when directing the broad-band light directly to the APD, and thus represents the time-response function of the APD. The red line shows the convolution of the APD response function with an exponential decay at lifetime \(\tau\) that was fitted to the polariton-decay curve yielding a polariton lifetime of \(\tau = 101.1\) ps. The inset displays the spectrum of the transmitted light, recorded with the spectrometer.

### 7.6. Signature of polariton lasing

The measured long lifetimes elevate our system into a new domain that should allow for the study of equilibrated long-lived bosonic quantum fluids in an all-solid-state setting. In order to demonstrate the potential of the setup in this respect, we went back to the 9-QW sample and recorded the emitted PL intensity of the LP\(_{00}\) mode as a function of off-resonant excitation power at an effective cavity length of about 10 \(\mu\)m. We chose a cavity-mode red-detuning of about 0.4 \(\Omega_R\) with respect to the exciton resonance, corresponding to an exciton content of about 40\%. As is illustrated in Figure 7.11(a), we observe a clear threshold behavior with a sharp non-linear increase in the emitted number of photons at around 1 mW of cw pump power.
In addition, the linewidth of the LP\textsubscript{00} mode quickly drops by almost an order of magnitude (Figure 7.11(b)), while the energy shift versus pump power exhibits a kink (Figure 7.11(c)). The logarithmic blue-shift above threshold was observed in other systems before and was attributed to polariton lasing or condensation [174, 181].

**Figure 7.11.: Signatures of polariton lasing in a sample with 9 QWs.**

(a) The intensity of the light emitted from the lowest-polariton mode LP\textsubscript{00} shows a highly non-linear behavior as a function of pump power with an increase by four orders of magnitude just above threshold. (b) The width of the LP\textsubscript{00} mode decreases sharply around threshold, while the mode energy (panel (c)) displays a continuous blue-shift over the whole range of pump powers with distinctly different behavior above and below threshold. Notably, the uncoupled-cavity mode was estimated to be about 3.5 meV blue-shifted (white dashed line) with respect to the polariton mode at low power. After the onset of lasing, the polariton mode clearly blue-shifts still towards the cavity mode. This suggests that the strong-coupling condition is preserved. The recorded spectra are normalized with respect to the polariton-intensity maximum.

### 7.7. Towards single-polariton nonlinearities

The ultimate goal of this project has been the observation of polariton blockade. To this end, second-order correlation measurements were performed on photons resonantly scattered from the fundamental lower-polariton transition using a HBT setup (see Section B.4.1). Correlation measurements were carried out in resonant-transmission measurements using a tunable cw diode laser and different samples with
one, four or nine quantum wells strongly-coupled to the cavity mode, each centered at around 900 nm. The measurements were conducted with different settings, e.g. different detunings of the cavity mode with respect to the exciton resonance and for varying cavity lengths. However, so far we have not observed any statistically significant deviation from Poissonian photon statistics.

Besides a too large cavity-spot size and insufficient cavity-quality factors to resolve the polariton-polariton interaction energy $U$, we attribute the main hindrance to observe the polariton-blockade effect to excitonic disorder. The QWs, grown at a center wavelength of about 900 nm, have inhomogeneous linewidths $\hbar\gamma_{QW} > 1.5$ meV, hence the condition for narrow lower-polariton lines $2\Omega_R > 4\gamma_{QW}$ cannot be fulfilled (see Section 6.4.1). In grown multi-QW samples, even larger linewidths were observed, due to an imperfect overlap of the different QW resonances. A way to decrease the QW linewidth and thereby reduce exciton disorder is to use a lower Indium content in the QW alloy. Consequently, the QW-exciton resonance is shifted towards shorter wavelengths. We grew samples with one or three QWs spectrally centered at about 850 nm. As shown in Figure 7.12, the upper and lower-polariton modes are more clearly resolved than in the case of QWs centered at 900 nm, and the spectral lines of polaritons with large exciton contents are narrower. However, in these samples, we measured cavity-finesse values $F < 1000$, which again limits the system from the observation of sufficiently narrow polariton lines. So far, we could not observe any signatures of polariton blockade in correlation measurements.

We attribute the reduced cavity quality to non-negligible absorption of 850 nm light in bulk GaAs. To solve this problem, the pure GaAs layers in the sample could be replaced by an alloy with some additional Al content, resulting in a material with a higher-energy band gap and therefore a higher degree of transparency at $\lambda = 850$ nm.

**Figure 7.12.:** PL spectra of a sample with 3 QWs centered at 848.2 nm. (a) Avoided-level crossing, as the cavity mode is tuned across the QW-exciton resonance. Several higher-order transverse modes are visible as well. The multiple of faint lines are higher-order transverse modes belonging to an adjacent fundamental mode one FSR lower. (b) PL spectrum on resonance (red dashed line in (a)) showing clearly-resolved upper and lower-polariton lines. All spectra were recorded in transmission geometry. The intensities are shown on a logarithmic scale.
In summary, we realized a new method for tight polariton confinement using a semi-integrated system consisting of a planar semiconductor mirror with a QW grown on top and a curved mirror deposited on the fiber-end surface of a single-mode optical fiber. Full tunability of the cavity length allows for in-situ control of the detuning between the exciton and cavity-mode resonance. Moreover, a cavity-length change over several $\mu m$ allows for the tunability of the cavity-photon lifetime. We measured a cavity-spot size of $\omega_0 \approx 2.5 \mu m$, and observed discrete higher-order Hermite-Gaussian cavity modes. With PL spectroscopy we confirmed the strong coupling between QW-excitons and cavity photons close to cavity-exciton resonance, by the observation of the upper and lower-polariton peaks, showing a Rabi splitting of about 4.2 meV. When tuning the cavity length to 30 $\mu m$, polaritons with low exciton content are measured to exhibit lifetimes up to 100 ps. Using a sample with 9 coupled QWs, we observe clear signatures of polariton lasing, as the above-band gap pump-laser power exceeds a certain threshold. On exact cavity-exciton resonance, we measure lower-polariton linewidths between 0.2 meV and 0.7 meV, which is a factor of $100 - 1000$ too broad for the observation of the polariton-blockade effect. The main hindrance of realizing narrow-linewidth lower polaritons with a high exciton content is assumed to be found in exciton-disorder scattering. Resonant correlation measurements in transmission geometry using a narrow-bandwidth cw laser so far have not shown any deviation from Poissonian statistics.
8. Conclusion and Outlook

The main experimental results of this thesis were obtained on a strongly-coupled QD-cavity system, which is well in the nonlinear regime. Previous studies using resonant probing techniques on similar devices faced difficulties due to the blinking effect dominating the system response. Although the mechanism for blinking is not fully understood yet, we demonstrated that an optical re-pump laser could be used to efficiently counteract the effect. As a result, sophisticated resonant-scattering experiments on a cavity-QED system integrated in a chip were made possible, despite the various dephasing mechanisms present in a semiconductor environment.

An immediate extension of the present work on single-photon nonlinear optics would be the demonstration of transistor operation in a QD-cavity device [16]. Ultimately, a strongly-coupled system, like the one studied in this thesis, could be used as a nonlinear-optical building block in future high-bandwidth photonic networks operating in the quantum regime [150–152, 197], or for studying strongly-correlated photonic systems in non-equilibrium settings, such as an optical Josephson interferometer [146] or coupled arrays of nonlinear cavities [24, 25] that would lead to the observation of exciting effects, such as the fermionization of photons [30] or to the realization of fractional quantum Hall states of light [198]. Given the recent progress on the fabrication of site-controlled QDs within arrays of photonic-crystal cavities [106, 107] and the ability to tune QD transitions by up to 25 meV in p-i-n structures [199], the present work demonstrates the great potential of QD-cavity systems as candidates for photonic quantum simulators [200].

In the second part of this thesis, a different cavity-QED system based on the coupling of QW excitons to Fabry-Pérot cavities was investigated. Our semi-integrated cavity platform provides tight cavity-waist sizes down to about 2.5 µm. This however turns out not to be narrow enough for the demonstration of strong polariton-polariton interactions leading to polariton blockade [36], as was confirmed by numerical estimates.

Ways to improve the confinement could be achieved by additionally engineering the semiconductor part of the semi-integrated system. Using e.g. a solid immersion lens (SIL), the NA of the cavity field that is inside the semiconductor could be enhanced – theoretically by a factor of $n_{GaAs} \approx 3.5$. The resulting cavity-spot area would be reduced by a factor of $n_{GaAs}^2 \approx 12$, leading to 12-times stronger polariton-polariton interactions. The use of mesas [46] instead of SILs could result in a similar effect.

An alternative approach to increasing polariton-polariton interactions would be to enhance the interaction length of excitons, e.g. by making use of indirect excitons, which are spatially separated electron-hole pairs with larger and direction-aligned electric-dipole moments. However, these compounds suffer from weaker optical oscillator strengths due to the tunnel barrier between the electron and hole. Based on ideas presented in Ref. [201], coherent superposition states of direct and indi-
rect excitons strongly coupled to a cavity field could form tunneling polaritons with interacting electric-dipole moments.

The observation of the polariton-blockade effect would open the door towards the study of strongly-correlated polariton systems with similar prospects to the nonlinear QD-cavity system.

We anticipate that with system parameters optimized for the particular purpose in mind, many exciting other directions of research are within reach with the present fiber-cavity setup. As an example, a high-efficiency spin-photon interface could be realized [60, 142]. Our semi-integrated platform combined with electrical injection of carriers could serve as a fiber-coupled tunable low threshold polariton laser. Finally, for material systems, where the growth of high-quality mirror substrates is not as advanced, our approach might provide a viable alternative route towards achieving high-$Q$ systems.
A. Light generation and manipulation

In this chapter, short descriptions of the optical equipment and devices, used for generation and manipulation of light, are provided. Section A.1 gives a list of all light sources, Sections A.2 and A.3 describe the power and polarization control of the light used in the experiments.

A.1. Light sources

This section provides a list of all coherent and incoherent light sources used in the experiments, with a short description of their applications. The light of all lasers is coupled into SM fibers (P3-830AFC-5 from Thorlabs) and power stabilized (see Section A.2).

- For various experiments presented in this work, we use a Ti:sapphire laser (MIRA 900), pumped by a 532 nm diode-pumped solid-state laser (DPSSL), a Verdi V10, both from Coherent® Inc. The operation wavelength can be tuned from 770 to 1000 nm, at an optical power of 500 – 1000 mW. The laser can be operated in cw or pulsed mode. The generation of pulses is based on passive Kerr-lens mode locking. In pulsed ps-mode, the pulses exhibit a duration of 3 – 4 ps, at a repetition rate of 76.3 MHz. In cw mode, the light emission is subject to a competition between several longitudinal modes, resulting in power fluctuations of a few %.

- A cw single-frequency Ti:sapphire laser (TIS-SF07 ring laser from Tekhnoscan, operated in the linear mode in this work), pumped by a 532 nm DPSSL (Verdi V18 from Coherent® Inc.) is used for QD re-pumping to counteract the QD-blinking effect. It provides up to 1500 mW optical power and is tunable over the range 695 – 1050 nm.

- For resonant cw measurements, we use narrow-bandwidth external-cavity diode lasers in the Littman-Metcalf configuration that are tunable over a bandwidth of 15 – 20 nm in a mode-hop free manner.
  - A New Focus TLB-6319 Velocity® with an operating wavelength range of 930 – 946 nm, used for RS experiments on the single-QD PC-cavity system.
  - A New Focus TLB-6318 Velocity® with an operating wavelength range of 890 – 910 nm, that we use for differential transmission measurements on the fiber-cavity system.
- A Newport TLB-6316 Velocity® with an operating wavelength range of 838 – 853 nm, also used for differential transmission measurements on the fiber-cavity system.

- A fiber-coupled diode laser S1FC780 from Thorlabs, with an operating wavelength of 780 nm is used for cw PL experiments on the fiber-cavity system.

- For ringdown measurements of polariton lines, we use a pulsed broadband white-light source (SC-500-2 from Fianium Ltd.) with a bandwidth of 450 – 1800 nm, a pulse duration of 400 fs and a repetition rate of 50 MHz. The light is filtered using bandpass filters with \( \approx 10 \) nm passband and various different center wavelengths (e.g. Thorlabs FB900-10 with a center wavelength of 900 nm) before entering the fiber cavity.

### A.2. Laser-power stabilization and modulation

All the lasers used in the experiments exhibit temporal power fluctuations. Also, mechanical drifts and vibrations of the optical setup result in deviations of the desired power. Furthermore, the optical elements are slightly chromatic, resulting in power modulations when scanning the laser wavelength. Therefore, a stabilization of the optical power reaching the sample is necessary.

Power stabilization is achieved by using an acousto-optic modulator (AOM; we use AOM 3080-125 deflectors with 80 MHz drivers 1080AF-AIFO-2.0, both from Crystal Technology Inc.): A piezo element driven at a radio frequency of 80 MHz generates a standing wave refractive index pattern inside a quartz crystal, capable of deflecting part of an optical beam when traversing the crystal. The signal is power-modulated by an applied signal voltage \( V_{\text{signal}} \) between 0 and 1 V. Consequently, the total deflected optical power is proportional to \( V_{\text{signal}} \) and can easily be controlled. Figure A.1 illustrates a fiber-coupled power modulation bench used in our experiments. A beam double-pass configuration ensures efficient fiber-coupling of the deflected light for different optical wavelengths that exhibit different deflection angles.

For laser power stabilization, part of the light that has already passed the setup is directed to a reference photodiode (compare parts PD1 – PD3 in Fig. 2.10) prior to sample illumination, using a beam splitter. A home-built PID controller monitors the detected light and controls the laser power in a feedback loop. Thus, a constant and computer-controlled laser power is provided, with reaction times of about 1 ms.

We set up three power-control setups for the simultaneous power control of up to three lasers.

In addition to ensuring a constant laser power, we used the controllers for the generation of periodic square laser pulses, that were e.g. necessary for QD re-pumping (see Section 3.2.1). As depicted in Figure A.2, this can be achieved by modulating the AOM input signal using a transistor-transistor logic (TTL) pulse-delay generator (DG645 digital delay generator by Stanford Research Systems) and an analog switch (DG419). The power stabilization is maintained in this configuration and ensures pulses of equal peak power.
A.2. Laser-power stabilization and modulation

**Figure A.1.** A fiber-coupled laser-power controller using an AOM. The power of the light out-coupled from the single-mode fiber can be pre-adjusted using a $\lambda/2$ plate in combination with a polarizing beam splitter (PBS). A beam expander reduces the diameter of the beam traversing the AOM crystal. The non-deflected light is blocked, whereas the deflected light is back-passed through the AOM using a plane mirror and a lens. Again, only the deflected light is coupled into a single-mode fiber. The double-passed $\lambda/4$ plate rotates the light polarization by $90^\circ$ to ensure total reflection at the PBS.

**Figure A.2.** Power modulation of a laser beam. To stabilize the mean laser power, the signal from the photodiode is first low pass filtered, with a cut-off frequency much smaller than the TTL-oscillator frequency $f$ and serves as the PID reference signal. The PID controller must be set to have a longer response time than $1/f$. The PID-output signal is then mixed (multiplied) with the TTL-oscillator signal, using an analog switch (DG419), and sent to the AOM controller.

We used this modulation scheme for QD re-pumping, but also for the quasi-simultaneous probing of the system with different lasers (see Section B.3). For QD re-pumping, the TTL-oscillator frequency is set between 0.5 and 1 MHz, with a duty cycle of 0.25. The pulses thus have a duration down to 250 ns, such that rise and fall times of the AOM activation / deactivation operations become relevant. These are mainly limited by the time it takes for the radio-frequency signal in the quartz crystal to traverse the Gaussian beam profile, due to a finite group velocity of the sound wave. For this reason, one of the AOM benches is slightly modified, by focused the spot diameter down to $\approx 125$ $\mu$m inside the crystal, resulting in a measured turn-on and turn-off time of about 50 ns.
A.3. Polarization control

A straightforward way to perform an arbitrary unitary polarization rotation of light is to guide it through the series of $\lambda/2 - \lambda/4 - \lambda/2$ optical waveplates and having full control over all three rotation angles. This method is expensive and requires high-precision rotation mounts to fine-tune the polarization.

In this work, the light of all the used lasers, as well as the detected light, is guided through non-polarization maintaining SM fibers (P3-830A-FC-5 from Thorlabs). For polarization control, we utilize the elasto-optic effect inside the fibers: If external strain is applied in the material the light traverses, a slight birefringence is induced. By a controlled twisting of the fiber, achieved by having the fiber winded around a rotatable polarization paddle, the polarization is altered in a unitary way. With three or more (home-built) paddles in series, rotations within the full Poincaré sphere can be achieved, in analogy to the series of waveplates. These fiber-polarization controllers are particularly practical for the crossed-polarization scheme used for non-interacting laser-light suppression (see Section 3.1.1), since fine-tuning of the polarization is more easily accomplished. A condition for precise polarization control is a high motional stability of the optical fibers.
B. Detection apparatus

In this chapter, I describe the most common optical detection techniques used throughout this work, including detection with a grating spectrometer (Section B.1), resonant spectroscopy using APDs (Section B.2) including the measurement of weak optical nonlinearities (Section B.3) and time correlated detection schemes (Section B.4).

B.1. Detection with a grating spectrometer

In PL experiments, the spectral light distribution is analyzed using a grating spectrometer. The working principle is shown in Figure B.1. The luminescence is focused onto the entrance slit of the spectrometer, at an NA that is adapted to the spectrometer $F/D$ number\(^1\). Inside the spectrometer, a concave mirror with focal length $F$ collimates the beam and directs it onto the diffraction grating. Using a second concave mirror with the same focal length, a 1 : 1 image of the light as seen at the entrance slit is generated on the plane of a charge-coupled device (CCD) camera (in the spectroscopy mode, see Fig. B.1(a)). The spectral information is provided by a horizontal position of the spot image. The full spectrum of the luminescence light is therefore reconstructed as a horizontal intensity distribution detected by a CCD array. All PL spectra in this work were recorded in this way.

The monochromator mode (see Fig. B.1(b)) is used to filter small spectral portions out of the incident light, by focusing the diffracted light onto the plane that contains the exit slit. In this case, the spectrometer acts as a very narrow-bandwidth bandpass filter. This technique was used for the generation of resonant pulses with small bandwidth (see Section 3.3.1).

In this work, we use an Acton SP2750 spectrograph from Princeton Instruments with a focal length $F = 750 \text{ mm}$ and $F/D = 9.7$. Two different holographic diffraction gratings with either 300 or 1500 grooves per mm are available. The spectral image is detected by a CCD camera with $1340 \times 100$ pixels of dimension $20 \mu\text{m} \times 20 \mu\text{m}$. The CCD chip is cooled to $-120^\circ \text{ C}$ with liquid nitrogen to obtain single-photon sensitivity and low dark-count rates.

In this work, the PL light from all systems is always coupled into a SM fiber. By focusing the out-coupled light to a point within the spectrometer entrance slit, we obtain the optimal resolution of about $30 \mu\text{eV}$ or $0.02 \text{ nm}$ in the wavelength range around $900 \text{ nm}$, using the grating with 1500 grooves per mm.

\(^{1}\)The $F/D$ number describes the full acceptance angle of the spectrometer; $F$ is the spectrometer focal length and $D$ the width of the grating. The NA is thus chosen slightly smaller than $\frac{D}{2F}$ for optimal grating illumination and therefore spectral resolution.
In both operation modes described above, the investigated spectral portion of the light can be chosen by a computer-controlled rotation of the diffraction grating.

**Figure B.1.: The spectrometer.** (a) In the spectroscopy mode, the spectral-intensity distribution is projected onto a high-sensitivity CCD matrix for a full reconstruction of the light spectrum. (b) In the monochromator mode, a portion of the spectrum is passed through the spectrometer exit slit of variable width. In the case of a fully open exit slit, a spatial separation of the light into its wavelength components is obtained. The images are taken from Ref. [108].

### B.2. Detection in resonant spectroscopy

The resonant spectra in this work are recorded by scanning the wavelength of a laser across the system transitions while detecting the scattered light using a photodiode. To assign an absolute wavelength to each recorded data point, we typically lock the laser to the desired wavelength using a wavelength meter (Ångstrom WS Ultimate 30 MC4 from High Finesse GmbH), and subsequently record the optical power emitted from the system.

In the earlier version of the fiber-cavity scanner, we measured the transmission with a built-in p-i-n diode and amplified the photocurrent with a variable-gain current amplifier (DLPCA-200 from FEMTO Messtechnik GmbH).

In the newer version of the fiber scanner and also in resonant measurements on the single-QD cavity system (see Section 3.1), the transmitted (reflected) light was first coupled into a single-mode fiber and then guided to an APD (SPCM-AQR-14 from PerkinElmer, Inc.) operating in the single-photon counting (Geiger) mode. The photocurrent amplification is based on an avalanche effect due to electronic impact ionization. In the wavelength range we are using (850 – 940 nm) the quantum efficiency (QE, probability that a photon impact is detected) drops from 35 % to 20 % (lower QE for longer wavelengths). Upon detection of a photon, the APD generates a TTL pulse that is registered using an electronic data acquisition (DAQ) card (BNC-2120 from National Instruments™). If \( f \) is the average rate of photon incidences, the optical power is given by
Due to electronic noise that is amplified by the avalanche effect, the APD exhibits a dark-count rate of \( \approx 100 \text{ s}^{-1} \), which has to be subtracted at low photon rates. At large photon-count rates above \( 10^6 \text{ s}^{-1} \), a saturation of the count rate sets in, due to the APD dead time of about 30 ns after a photon impact. Using an APD, optical powers below 100 aW (attowatts) can be detected.

\[ P = \frac{\hbar \omega f}{\text{QE}}. \]  

(B.1)

**B.3. Extraction of optical nonlinearities**

The method for extracting the spectral nonlinearity when exciting a nonlinear system with two lasers of different colors is described in Section 5.3. In practice, we also include the photon rate, detected when both lasers are switched off (a small fraction of residual PL photons from the re-pump laser is still detected). For the extraction of the nonlinearity we therefore calculate

\[
N_{nl}(\tau) = (N_{\text{both on}}(\tau) - N_{\text{control}} - N_{\text{signal}} + N_{\text{both off}}) - (N_{\text{both on}}(\tau_{\text{int}}) - N_{\text{control}} - N_{\text{signal}} + N_{\text{both off}}) \quad (B.2)
\]

with the experimental data as input. Each data point is measured with an integration time of several 10 s. On these timescales, a tiny deviation of the excitation spot positions on the sample surface due to drifts might result in a slight modification of the detected photon rate. Since we have to subtract several separately measured photon numbers from each other, these small deviations would result in huge relative errors of the extracted nonlinearity. To eliminate long-time drifts, we measure \( N_{\text{both on}}(\tau) \), \( N_{\text{control}} \), \( N_{\text{signal}} \) and \( N_{\text{both off}} \) on ms timescale by switching the control and signal lasers on / off with 10 kHz and 5 kHz respectively and sorting the detected photon clicks accordingly (see Figure B.2). The lasers are stabilized by their mean power using the modulation technique shown in Fig. A.2.

![Figure B.2: Method for the elimination of long-time drifts.](image)

The control laser is modulated at 10 kHz and the signal laser at 5 kHz. The recorded photon counts are read at a rate of 10 kHz and sorted into the four signal bins \( \{N_{\text{both on}}, N_{\text{signal}}, N_{\text{control}}, N_{\text{both off}}\} \).
B.4. Time-correlated detection

To figure out the coherence of a photon stream or the characteristic lifetime of an optical emitter, it is necessary to obtain information about the incidence time of the emitted photons. In the following sections, we briefly discuss these concepts and the technical implementations.

B.4.1. Autocorrelation measurements

A useful technique for the investigation of an optical emitter is the measurement of the time dependent intensity (i.e. second order) autocorrelation function. The classical normalized autocorrelation function is given by

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

(B.3)

with \( I(t) \) being the detected optical power at time \( t \) and \( \langle \cdot \rangle \) indicating an ensemble average. In a quantum description, the second-order autocorrelation function of a single-mode light field writes [65]

\[ g^{(2)}(t, \tau) = \frac{\langle \hat{a}^\dagger(t) \hat{a}^\dagger(t + \tau) \hat{a}(t + \tau) \hat{a}(t) \rangle}{\langle \hat{a}^\dagger(t) \hat{a}(t) \rangle \langle \hat{a}^\dagger(t + \tau) \hat{a}(t + \tau) \rangle}, \]  

(B.4)

where \( \hat{a}(t) \) is the photon annihilation operator of the quantum system at time \( t \) and \( \langle \cdot \rangle \) denotes the expectation value. It provides a measure of correlations between two photons emitted at times \( t \) and \( t + \tau \). The value \( g^{(2)}(\tau = 0) \) is of particular interest in this regard, since it describes (phenomenologically) the ability of the system to emit two photons at the same time.

Classical systems (consisting of a multiple of emitters) always fulfill \( g^{(2)}(\tau = 0) \geq 1 \). Photons emitted by a black body, for example, preferentially appear in bunches, with \( g^{(2)}(\tau = 0) = 2 \). A coherent light source (e.g. a laser) on the other hand, emits a photon stream with Poissonian statistics \( g^{(2)}(\tau = 0) = 1 \), in which any two chosen photons do not have any correlation in their time of emission. Conversely, quantum systems can exhibit \( g^{(2)}(\tau = 0) < 1 \) or even \( g^{(2)}(\tau = 0) = 0 \). Such a non-classical or sub-Poissonian system shows photon antibunching, meaning that photons are preferably emitted singly (in time).

To measure \( g^{(2)}(\tau) \) of a quantum system, the statistical distribution of arrival-time differences between two photons is recorded using an APD (in the Geiger mode) and a time-correlated single-photon counter (TCSPC). The time differences are plotted in a histogram (with time channels of the width \( \delta t \)). To normalize the histogram, it is divided by \( f^2 \delta t T \), where \( f \) is the photon-detection rate of each APD and \( T \) the overall integration time. The dead time \( t_{\text{dead}} \) of an APD (the time it takes for restoring the charge balance in the photo-sensitive region after a photon detection) is typically several ns and therefore much larger than the relevant timescales of the quantum systems we study. Nevertheless, two photons at shorter mutual delay can be detected, if the emitted photon stream is divided in two parts with a 50:50 BS and each stream detected using a separate APD. Upon detection of a start photon by APD1, a TCSPC measures the delay until a stop photon is registered by APD2.
This configuration is called *Hanbury-Brown and Twiss* (HBT) setup, illustrated in Fig. B.3(a).

**Figure B.3.: Optical setup for autocorrelation and lifetime measurements.**

(a) A HBT setup consists of a 50:50 BS that splits the photon stream in two parts. Each beam is guided to a separate APD. A TCSPC measures the delay of a stop photon, detected by APD$_2$, after detecting a start photon using APD$_1$. All the events are plotted in a histogram, which reproduces the system autocorrelation function. (b) The setup for lifetime measurements. The studied system is excited with short pulses, and the photons emitted from the system guided to an APD. A TCSPC interprets the TTL-synchronization signal of the laser as the start pulses and the APD signal as stop pulses. The mutual delays are plotted in a histogram, yielding the characteristic shape of the system decay curve. In both setups (a) and (b), variable electronic delay lines in the stop-signal paths enable the registration of *negative* delays.

In this way, $g^{(2)}(\tau)$ of the studied system can be measured, if the following conditions are fulfilled:

1. The APD timing jitter (time resolution) must be smaller than all relevant timescales of the studied quantum system. Otherwise, the correlation function is ‘washed out’.

2. To measure $g^{(2)}(\tau)$, $f \tau \ll 1$ must be satisfied. Otherwise, the probability of detecting a stop photon at time $t + \tau$ after having detected a start photon at $t$ is already lowered, due to the high chance of detecting another photon, before this event can happen. A system that fulfills the above mentioned condition is termed to be measured in the *low detection limit*. 
Autocorrelation measurements can be performed in cw or pulsed-excitation mode. In the latter, the investigated system is excited by a pulsed laser, with pulse durations much smaller than the timescales of interest. In this case, the time information is imprinted on the system by the laser pulses themselves, such that APDs with a large timing jitter can be used; see the discussion in Section 3.3.

For cw experiments presented in this work, we used two id100-20 APDs from ID Quantique SA, with a timing jitter of about 40 ps and a quantum efficiency of \( \approx 4\% \) within the observed wavelength range of 850 – 900 nm.

For pulsed measurements, we used two SPCM-AQR-14 APDs from PerkinElmer, Inc., with a timing jitter of about 300 ps and a QE of \( \approx 20 - 35\% \) within the wavelength range of 850 – 940 nm. The QE is at least a factor of 5 larger than the QE of the id100-20 APDs from ID Quantique SA, so the coincidence rate is more than a factor of 25 larger than in the other case. A dirt effect, observed when recording correlations with the PerkinElmer SPCM-AQR-14 APDs, is the formation of symmetric side-peaks at \( \tau \approx \pm 10 \) ns, which is the more pronounced, the lower the APD-count rate is (see for example Figures 4.2(b-e)). The explanation is found in secondary-photon events, emitted by one APD, and detected by the other APD via scattering at optical elements: Due to the avalanche effect, electron-hole pairs are generated in the active region of the APD in a chain reaction. A few of these pairs recombine optically, before the carriers are separated by the applied voltage. The use of pin-holes at the APD apertures, as well as black tape in the vicinity of the BS reduced this effect significantly in our experiment.

For both, cw and pulsed measurements, a PicoHarp 300 from PicoQuant GmbH with a minimum channel width of 4 ps served as the TCSPC.

### B.4.2. Lifetime measurements

In many experiments, it is important to investigate the characteristic state lifetimes or the delays at which these states are populated upon optical excitation. This is achieved by initially exciting the system with a short optical pulse (with a pulse duration that is considerably shorter than all relevant system timescales) and detecting the optical system response with an APD in the Geiger mode. The statistical distribution of delays between the excitation of the system with the pulse and the emission and detection of a photon offers a great insight into the system dynamics.

Pulsed light sources typically exhibit a voltage output with a TTL-trigger signal synchronized with the generated optical pulses. Here, it was used as the start trigger of the TCSPC, whereas the photons, detected by the APD, constitute the stop signal (see Fig. B.3(b)). The generated histogram reflects the characteristic optical decay curve of the system.

In the experiments presented in this work, we typically used the id100-20 APDs from ID Quantique SA, with a timing jitter of about 40 ps. These APDs are subject to a low afterpulsing probability. After a real photon detection, charge carriers can be trapped in the active semiconductor region of the APD (due to impurities) that can eventually trigger a false pulse – the afterpulse. In the used APDs, the afterpulses occur at average delays on the order of 1 ns after the real photon detection. Therefore, a de-convolution of the recorded system response with respect to the characteristic APD-afterpulsing response is advantageous (in particular, if the
system response occurs on timescales considerably below 1 ns). A de-convolution is performed in the following way: If $I(t)$ is the emitted optical power, $\text{ref}(t)$ is the APD-afterpulsing response function and $D(t)$ is the system response detected with the described method above,

$$D(t_i) = [I * \text{ref}](t_i), \quad \text{(B.5)}$$

where ‘*’ is the convolution operator and $\{t_i, \ i = 1...N\}$ are the time channels. According to the convolution theorem,

$$\mathcal{F}[D](\omega_j) = \mathcal{F}[I](\omega_j) \cdot \mathcal{F}[\text{ref}](\omega_j), \quad \text{(B.6)}$$

where $\mathcal{F}$ is the discrete Fourier transform and $\{\omega_j, \ j = 1...N\}$ are the discrete frequencies of the reciprocal space. Thus, the original optical response $I(t)$ can be reconstructed by calculating

$$I(t_i) = \mathcal{F}^{-1} \left[ \frac{\mathcal{F}[D](\omega_j)}{\mathcal{F}[\text{ref}](\omega_j)} \right](t_i). \quad \text{(B.7)}$$
C. Bibliography


List of publications


Acknowledgment

When I learned to know Ataç and his enthusiasm in teaching quantum optics, my passion for this field was born. I appreciate by heart your confidence in giving me the possibility to contribute to the Quantum Photonics Group and taking part in your enthusiasm. It made the last four years of my life the most inspiring and fruitful ones. During the whole PhD there was no time I could not count on your advice and support, always with the right balance between inspiration and constructive criticism and no matter how little time you had. You offered me to gain wonderful knowledge and also technical skills by your optimism and confidence in what I was trying to do in the lab. Many thanks for having me as a member in your group.

Great thanks to Martin Winger for introducing me to the lab with an infinite portion of enthusiasm, goodwill and patience and teaching me everything about optics and lab work. Thanks for the great collaboration in the initial stages of the experiment. Moreover, from you I learned that experiments work better under the influence of ABBA phonons and loud encouragement. Thanks for your support in everything beyond ETH and for widening my horizon by introducing me to the ‘Gsibergians’.

Extra big thanks to Thomas. When working with you, I learned that every crazy idea is worth a try and that every detail is important for a successful experiment. Without your technical expertise, optimism and confidence it wouldn’t have been possible to squeeze so much out of G8. Thank you for the close collaboration on all the experiments we performed in these years. Although working with tiny eyes and in slow motion, the many night sessions with you were legendary and great fun. It was invaluable to know I could always count on you, both in the lab and as a friend. Great thanks for proofreading this thesis.

My special thanks goes to Antonio and Kevin who I worked with in the very initial stages of my time here. The sample you grew/fabricated – in the meantime an old lady – literally did its job and was the fundament of my work in this group. Great thanks to Martin Kroner, for sharing your technical expertise in all aspects of lab life. Thanks for organizing the legendary movie nights and the kicker table. Many thanks to Parisa for helping me find my way through the clean room. Thanks to Emre Ilgünsatiroglu for the great teamwork in the very beginning of my PhD and for offering us the awesome time in Istanbul.

With Javier, the Latin spirit came into our team. Thank you for your indispensable contribution to the projects and all the help and discussions. Great thanks for growing such awesome samples. The frequent sweets and meats, etc. you brought were always delicious. Thanks to Emre Togan for your helpful contribution in the last stages of the experiment and the inspiring discussions. Many thanks to Weibo, Stefan and Ajit that I could always rely on you whenever I needed your help. Thanks to Steffi, Madeleine, Madeleine, Manuela and Kathi for the great teamwork and the pleasant time you shared with us physicists. I would like to thank all the other
group members for their great company and help. Big thanks to Andi Stuker and his team of the Physics workshop for transforming all our ideas into unimpeachable items that formed the basis of many experiments.

Our teamwork even extended to Paris – as an incredible enrichment for the experiments and our horizon. Many thanks to Jakob, for sharing all your technology, knowledge and experience with us. Your efforts even in the lab accounted for the success of the experiments. Thank you for your pleasant hospitality during our visits in your group. Thanks to Jérôme for the great teamwork on the fiber-cavity project and your faithful dedication to our venture. Thank you Ben, for your indefatigable contribution to the project. All the discussions with you were extremely fruitful, all the night shifts in our lab unbelievably productive, all the time we spent together great fun.

Thanks to Patrick, Jereon and Yves for being the best imaginable office mates. Thanks to Kathi, Mena and Priska for the frequent reminders that there is also life outside ETH, for taking us to the Swiss mountains and for organizing delightful Christmas dinners. Great thanks to Charly, Stephan, Wolf and Florian for the legendary kicker sessions that immediately blew away all forms of frustration.

Specially big thanks go to Iacopo. The time you spent in our group was way too short and I remember it with great pleasure. Thank you for the fruitful discussions and everything I learned from you. Thanks for proofreading the papers and thesis. I deeply appreciate you as my co-Referee.

My studies here in Zurich would have been sad and frustrating without my precious friends that accompanied me on this journey. Great thanks to the Horde, Theo, Philip, Chris, Thomas, Jakob, Raph, Flo, Lars... for all the time we shared and the awesome weekend trips to remote places on this ball that will survive my memory forever. Thanks to Susanne, Andres, Kathi and Bruno for your great company in bouldering trips and other exciting city and road trips. Thanks to Sarah for our great WG and friendship. Thanks to all the Gsibergians, Johanna, Martin, Sigi... for making me feel home here in Zurich and for the ever-delicious Käsknöpfle. Thanks to my Austrian mates Martin, Jan, Christian, Bernhard... for always welcoming me back home.

Finally, I would like to thank my family. I am deeply grateful to my parents who enabled all my way with never ending support, confidence and love. You were always there for me and helped me whenever and wherever you could. Thank you Walter, for your valuable support during my studies. Thanks to Tobias and Michael, simply for your presence when I have been back home. Thank you Seohee, for your patience and love and always making me happy.
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March 10, 2013
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