Magneto-optical cavity quantum electrodynamics effects in quantum dot - micropillar systems

S. Reitzenstein¹, S. Münch¹, P. Gold¹, P. Franeck¹, A. Rahimi-Iman¹, A. Löffler¹, S. Höfling¹, L. Worschech¹, I. V. Ponomarev², T. L. Reinecke² and A. Forchel¹

¹Technische Physik, University of Würzburg, Am Hubland, D-97074 Würzburg, Germany
²Naval Research Laboratory, Washington, DC 20375, USA
E-mail: stephan.reitzenstein@physik.uni-wuerzburg.de

Abstract. We report on magneto-optical studies of strongly coupled quantum dot – micropillar cavity systems. Large In₀.₃Ga₀.₇As quantum dots (QDs) in the active layer of the micropillar facilitate the observation of strong coupling. In addition, they exhibit a particular large diamagnetic response which is exploited to demonstrate magneto-optical resonance tuning in the strong coupling regime. The magnetic field employed in Faraday configuration induces a transition from strong coupling towards the critical coupling regime which is explained in terms of a magnetic field dependent oscillator strength of the In₀.₃Ga₀.₇As QDs. We further study the coherent interaction between spin resolved states of the QDs and microcavity photon modes. A detailed oscillator model is used to extract the associated coupling parameters of the individual spin and cavity modes and reveals an effective coupling between photon modes that is mediated by the exciton spin states.

1. Introduction
Following the first demonstration of strongly coupled quantum dot (QD)-microcavity systems enormous effort has been devoted to the field of cavity quantum electrodynamics (cQED) [1, 2]. For instance cQED experiments in semiconductor nanostructures have been motivated by an interest in demonstrating quantum optics effects such as the coherent exchange of energy between quantum emitters and resonant modes of optical cavities on a solid state platform, which can open opportunities for large scale integration in technologies such as quantum information[3, 4]. To date most experimental studies involving strong coupling in QD-microcavity systems have relied on temperature tuning [1, 2] or electro-optical resonance tuning based on the quantum confined Stark effect [5].

Here we demonstrate that an external magnetic field provides an additional degree of freedom to more fully explore the potential of coherently coupled QD-microcavities. A magnetic field induced diamagnetic shift and Zeeman splitting as well as magnetic localization of carriers present a means of controlling the transition energy [6, 7], the spin-state and the oscillator strength [8], respectively, of a QD interacting with a microcavity. In this sense the magnetic field not only acts as a tuning parameter but also opens a way of modifying in situ the coupling strength of the interacting system. A prerequisite for this in situ manipulation is the presence of In₀.₃Ga₀.₇As QDs that are extended laterally in the active layer of the micropillar. Their large diamagnetic coefficient of up to 30 μeV/T² facilitates the exploitation of the diamagnetic shift as a tuning
The additional confinement introduced by the external magnetic field controls the oscillator strength of the large QDs from values close to 50 down to about 25 for the highest applied magnetic field (5 T) which induces a transition from the strong coupling regime towards the critical coupling regime when the magnetic field is increased. In addition, we address spin related cQED effects in a coherently coupled QD-micropillar system. The spin degree of freedom is accessed when a non-zero magnetic field is applied so that the spin degeneracy of the QD exciton is lifted. This allows us to study the coupling of individual spin states to cavity photon modes.

2. Technology
We have investigated high quality (Q) factor micropillar cavities based on planar GaAs/AlAs microcavities grown by molecular beam epitaxy. The planar distributed Bragg reflector (DBR) structure consists of 23 and 27 λ/4 thick GaAs/AlAs mirror pairs on the surface and the substrate side of an one-λ thick GaAs cavity layer, respectively. At the center of the GaAs cavity, a single layer of \text{In}_{0.3}\text{Ga}_{0.7}\text{As} QDs is introduced during the epitaxial growth. The growth of the QDs was optimized in order to realize large QDs (in-plane extension of about 40 nm), providing increased dipole moments and oscillator strengths, which facilitate the observation of the strong coupling regime. Subsequent to the growth of the planar microcavities, high Q-factor micropillar cavities were realized by means of high resolution electron beam lithography and plasma etching in an electron cyclotron resonance system. We have realized micropillar cavities with diameters ranging from 1.0 µm to 2.0 µm, providing Q factors \( Q = E_c/\gamma_c \) up to 15,000, where \( E_c \) denotes the mode energy and \( \gamma_c \) the full width at half-maximum (FWHM) of the mode. We refer to Refs. [9, 10] for details on the fabrication of the micropillar cavities.

Magneto-optical studies were performed at low temperature using a high resolution microphotoluminescence (\( \mu \)PL) setup. The sample was mounted to the cold-finger of a He flow cryostat providing magnetic fields up to 5 T in Faraday configuration. This means that the magnetic field was applied collinearly to the symmetry axis of the micropillars as sketched in Fig. 1. The sample was excited by a frequency doubled Nd:Yag laser emitting at 532 nm. The PL was detected using a nitrogen cooled Si charge coupled device camera.
Figure 2. Magnetic field dependent $\mu$PL spectra of In$_{0.3}$Ga$_{0.7}$As quantum dots. (a) Magnetic response of a quantum dot exciton line $X_1$. The QD line exhibits a large diamagnetic constant of 29 $\mu$eV/T$^2$ and a Landé g-factor close to zero. (b) Magnetic response of another QD exciton $X_2$ showing a slightly smaller diamagnetic constant (22 $\mu$eV/T$^2$) if compared to the exciton lines shown in panel (a). The QD exciton line splits into two Zeeman split components at large magnetic fields. From the splitting a Landé g-Factor $g_L = 0.46$ is determined.

3. Experimental

3.1. Magnetic Response of Large In$_{0.3}$Ga$_{0.7}$As Quantum Dots

Magneto-optical studies allow one to probe the diamagnetic shift as well as the Zeeman-splitting of QDs, where the magnitude of the diamagnetic shift depends on the lateral extension of the electron and hole wavefunction. In fact, provided that the cyclotron energy $\hbar\omega_c$ is much smaller than the exciton binding energy, which is the case in the present experiment, the diamagnetic shift of QD excitons $\Delta E_{dia}$ can be expressed via [11]

$$\Delta E_{dia} = \kappa_x B^2 = \frac{e^2}{8} \left( \frac{\langle r^2_e \rangle}{m_e} + \frac{\langle r^2_h \rangle}{m_h} \right) B^2,$$

(1)

where $\langle r^2_{e,h} \rangle$ denotes the average quadratic extension of the electron (heavy hole) wavefunction and $m_{e,h}$ represents the in-plane mass of the electron (hole). In Fig. 2 we present magnetic field dependent $\mu$PL spectra of large In$_{0.3}$Ga$_{0.7}$As QDs. The QD exciton line in panel (a) shows a pronounced blue shift of emission when the magnetic field increases from 0 to 5 T. From an lineshape analysis of the emission lines we determine a diamagnetic constant $\kappa_{X1} = 29$ $\mu$eV. The large diamagnetic shift of these type of QDs associated with diamagnetic constants between about 20 and 30 $\mu$eV/T$^2$ if compared to standard In$_{0.6}$Ga$_{0.4}$As QDs exhibiting diamagnetic shifts close to 10 $\mu$eV/T$^2$ reflects clearly their enhanced lateral extension. While no significant Zeeman splitting can be observed of the exciton lines presented in Fig. 2(a), a clear splitting is observed for the exciton line investigated in Fig. 2(b) showing a slightly smaller diamagnetic constant $\kappa_{X2} = 22$ $\mu$eV. From the Zeeman splitting $\Delta E_Z$ we determine a Landé g-factor $g_X = 0.46$ according to $\Delta E_Z = g_X \mu_B B$. Comparing these diamagnetic constant and Landé g-factors of these QD lines suggests that QDs with a smaller diamagnetic shift exhibit larger Landé g-factors. Indeed, an statistical evaluation of the magnetic response of In$_{0.3}$Ga$_{0.7}$As QDs revealed an anti-correlation between the Landé g-factor and the diamagnetic shift.
Figure 3. Magneto-optical tuning in the strong coupling regime. (a) Magnetic field dependent \( \mu \text{PL} \) spectra of a QD - micropillar with a diameter of 1.6 \( \mu \text{m} \) and a Q factor of about 10000 at 10 K. The coherent interaction between the QD exciton X and the photon mode C is reflected in a normal mode splitting on resonance at \( B = 2.75 \text{ T} \). (b) Energy dispersion of the strongly coupled QD - micropillar system. On resonance a vacuum Rabi-splitting of 110 \( \mu \text{eV} \) is observed.

### 3.2. Magneto-Optical Resonance Tuning

In this section we address magneto-optical resonance tuning in the strong coupling regime. The large QDs are very suitable for this study as they provide large light-matter coupling constant and also feature a large \( \kappa_x \) which allows for a sufficiently large tuning range of almost 1 meV for magnetic fields between 0 and 5 T. This tuning range is similar to those obtained typically for temperature or electro-optical resonance tuning [1, 2, 5].

Magneto-optical tuning is presented in Fig. 3(a) which shows magnetic field dependent \( \mu \text{PL} \) spectra of a 1.6 \( \mu \text{m} \) diameter micropillar at 10 K. At \( B = 0 \) the exciton is located at the low energy side of the cavity mode. With increasing magnetic field the exciton experiences a diamagnetic blue shift and resonance with the cavity mode is achieved at \( B = 2.75 \text{ T} \). The avoided crossing of the excitonic and the photonic mode is a clear signature of the strong coupling regime. The avoided crossing of the interacting modes can also be seen in Fig. 3(b) which shows the corresponding energy dispersions. The energy of the cavity mode is not affected by the magnetic field. In contrast, the exciton line shows a blue shift of 780 \( \mu \text{eV} \) between 0 and 5 T which corresponds to \( \kappa_x = 31 \mu \text{eV/T}^2 \). The coherent interaction of the exciton and photon mode is reflected in a vacuum Rabi-splitting of \( \Delta E_r = 110 \mu \text{eV} \) at \( B = 2.75 \text{ T} \).

### 3.3. Magnetic Field Control of the Coupling Strength

The magnetic field introduces an additional confinement of the carriers in the elongated QDs which is expected to change to dipole moment of the QD excitons and, thus, the light-matter coupling strength. In order to investigate the light-matter coupling strength as a function of magnetic field we recorded PL emission spectra at resonance for magnetic fields between 0 and 5 T. The corresponding spectra are depicted in Fig. 4. The mode splitting decreases with increasing magnetic field and for magnetic field exceeding 3 T a single, broad emission peak evolves at the resonance energy \( E_{\text{res}} \). Lorentzian lineshape fitting the experimental allows us to
extract the vacuum Rabi splitting $\Delta E_R$ from which the light-matter coupling constant $g$ can be determined via the relation $\Delta E_R = 2\sqrt{\left(g^2 - \frac{\gamma_c^2 - \gamma_a^2}{16}\right)}$, where $\gamma_c$ ($\gamma_a$) denote the cavity (exciton) mode linewidth. For small magnetic fields $\Delta E_R$ the QD exciton is confined within the extended potential of the QD and the coupling with the cavity mode results in a vacuum Rabi splitting of about 100 $\mu$eV and a coupling constant of about 60 $\mu$eV. For magnetic field exceeding 3 T the exciton wavefunction becomes localized and squeezed by the additional magnetic confinement when the magnetic length falls below the characteristic size of the QD potential [12]. This leads to a reduction of the dipole moment and the oscillator strength of the QD exciton which in turn explains the significant decrease of the coupling constant and the vacuum Rabi splitting to values of $g = 44 \mu$eV and $E_R = 65 \mu$eV, respectively. The reduction of the coupling strength is related to a decrease of the QD oscillator strength from about 45 at 0 T down to about 25 at 5 T which was confirmed independently by time resolved PL measurements [12]. This results show that the interaction strength of the strongly coupled QD-microcavity system can be controlled in situ by an external magnetic field.

3.4. Exciton Mediated Photon-Photon Coupling

In this section we study the interaction of spin degree of freedom of excitons in InGaAs quantum dots with the photon cavity modes in pillar microcavities, and we address the question of how the polarization of the modes affects their coupling. The spin states are split with a magnetic field in Faraday configuration, and their interactions with individual cavity modes are identified.
Figure 5. (a) Temperature-dependent µPL spectra of micropillar with a diameter of 1.6 µm 3 T. The zeeman split QD exciton lines $X_1$ and $X_2$ interact coherently at 24.5 K and 26.6 K, respectively, with the two linearly polarized components $C_1$ and $C_2$ of the fundamental cavity modes. (b) Corresponding energy dispersions obtained from Lorentzian lineshape fitting.

in magneto-photoluminescence experiments.

Spin resolved light-matter interaction is explored by temperature dependent µPL measurements at 3 T. Fig. 5(a) shows sets of µPL spectra of a 1.6 µm diameter micropillar at $B = 3$ T. The cavity peak has a distinctive left shoulder that can be deconvolved into two unequal Lorentzians ($C_1$ and $C_2$). Both mode components are split by $\alpha \approx 50 \mu$eV and have a Q factor of about 15000. The splitting of the fundamental cavity mode into two orthogonal linearly polarized mode components originates from the processing of the micropillars [9].

At $B = 0$ T the exciton shows a fine structure splitting of $\beta = 20 \mu$eV. Spin related effects come into play when a magnetic field splits the excitonic line into two components $X_1$ and $X_2$. The magnetic field also changes their polarizations, and a total Zeeman splitting of $\Delta E_{Zeem} = 117 \mu$eV at $B = 3$ T can be seen in the lowest trace of Fig. 5(a). Magnetic field dependent measurements at constant temperature give a diamagnetic coefficient of $\kappa = 22 \mu$eV/T$^{-2}$ for the QD exciton, which reflects again the extended electron-hole wavefunction of these large oscillator strength QDs [12].

The coupling of spin-related excitonic emission lines with photonic modes of a microcavity is studied at 3 T. Both exciton lines shift through resonance with the cavity modes when the sample temperature is increased from 20 K to 29 K (cf. Fig. 5(a)). Minima of the splitting in energy of the outermost emission lines are observed at about 24.5 K and 26.5 K, respectively. Significant broadening and splitting (marked by an arrows) of the associated emission lines occurs when the exciton components enter the interaction regime. The corresponding energy dispersions are plotted in Fig. 5(b). At both resonance temperatures anticrossings of all involved modes indicate unambiguously the coherent cQED coupling with the Zeeman split exciton components in the spin resolved cQED regime.

In order to analyze the interacting system we employ a detailed coupled oscillator model of the exciton and cavity modes described in Ref. [13]. The model considers a basis of cylindrically symmetric cavity modes and cylindrically symmetric QD excitonic states and experimental parameters $\alpha$ and $\beta$ which determine the mixings of the two cavity modes and the two exciton
modes, respectively. The only free parameter is the coupling constant $g$ between cavity and exciton states. It is interesting to note that a strong magnetic field adds an additional splitting to the fine structure splitting $\beta$ and tends to restore the circular polarization. At intermediate fields excitons are neither linearly nor circularly polarized, with a polarization governed by ratio of parameters $\delta/\beta$, where $\delta = g_L \mu_B B$ is the bare Zeeman splitting, and $g_L$ is the Landé $g$-factor. In the present experiment $\delta/\beta = 6.25$ for $B = 3$ T. The total splitting of two excitons is determined by the parameter $\tilde{\beta} = \sqrt{\beta^2 + \delta^2}$.

The real and imaginary parts of the complex eigenenergies of the coupled modes as functions of $T$ are calculated from the four oscillator model and are potted Fig. 5(b) and Fig. 6, respectively, along with the experimental data. The best agreement was obtained for $g = 38 \mu$eV for which the theory (solid lines) describes the experimental data (dots) very well. Additional insight into the physics of the coupled exciton spin-photon system can be obtained from the mode linewidths as a functions of temperature. The calculated imaginary parts of the energy-eigenvalues are shown in Fig. 6 together with the experimental results obtained fitting of the data in Fig. 5(a). Pronounced mixing of the lines occurs for temperatures between $T \approx 24.5$ K and $T \approx 26.5$ K where the exchange of linewidths between the $X$ transitions and the $C$ cavity modes indicates coherent interaction in the strong coupling regime.

Interestingly, the linewidths show considerable involvement of the $C_2$ ($C_1$) mode near the anticrossing at 24.5 K (26.5 K). Indeed, the crossing point of the $X$ and the $C$ linewidths is shifted upwards compared to the expected value $(\Gamma_C + \Gamma_X)/2 = 64 \mu$eV (cf. dashed line in Fig. 6) for strong coupling with only a single photonic mode. This effect is clearly observed for the experimental data the 26.5 K and means that the photonic character of the coupled modes is increased when two photon modes are involved in the spin related light-matter interaction. These observations demonstrate that there is an effective interaction between the two photon states mediated by the quantum dot exciton. The physical mechanism for this coupling is the photon-exciton interaction given by the parameter $g$.

4. Conclusions

In summary, we have presented magnetooptical QD-micropillar systems in the strong coupling regime. The large diamagnetic coefficient of large In$_{0.3}$Ga$_{0.7}$As QDs with values up to $30 \mu$eV/$T^2$ facilitate the exploitation of the diamagnetic shift as a tuning parameter. The additional
confinement introduced by the external magnetic field allows us to control the oscillator strength of the large QDs from values close to 50 down to about 25 for $B = 3$ T which induces a transition from the strong coupling regime towards the weak coupling regime. Moreover, we have addressed spin related cQED effects in solid state by demonstrating strong (coherent) coupling between excitonic spin states and high quality factor photon modes of a pillar microcavity structure. We find a coupling of independent photonic modes via the excitonic transition in addition to the coupling between the spin states of the excitons and the cavity modes. Such a coupling is interesting from a fundamental point of view, and nonlinear interactions mediated by quantum dots are also of considerable interest in connection with quantum communications and quantum information.

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