Dephasing of Triplet-Sideband Optical Emission of a Resonantly Driven InAs/GaAs Quantum Dot inside a Microcavity

S. M. Ulrich,[1,*] S. Ates,[1] S. Reitzenstein,[2] A. Löffler,[2] A. Forchel,[2] and P. Michler[1]

[1]*Institut für Halbleiteroptik und Funktionelle Grenzflächen Research Center SCoPE, Universität Stuttgart, D-70569, Stuttgart, Germany*

[2]*Technische Physik, Wilhelm Conrad Röntgen Research Center for Complex Material Systems, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany*

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Detailed properties of resonance fluorescence from a single quantum dot in a micropillar cavity are investigated, with particular focus on emission coherence in the dependence on optical driving field power and detuning. A power-dependent series over a wide range reveals characteristic Mollow triplet spectra with large Rabi splittings of $|\Omega| \leq 15$ GHz. In particular, the effect of dephasing in terms of systematic spectral broadening $\propto \Omega^2$ of the Mollow sidebands is observed as a strong fingerprint of excitation-induced dephasing. Our results are in excellent agreement with predictions of a recently presented model on phonon-dressed quantum dot Mollow triplet emission in the cavity-QED regime.

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Exploiting the quantum properties of the light which is emitted from semiconductor quantum dots (QDs) has the potential of enabling various new applications in the field of photonics and quantum information technology [1]. Many of these applications require single-photon [2,3] and entangled-photon [4–7] light sources. The implementation of single-photon based quantum logic algorithms critically relies on photon indistinguishability [8] which is directly related to the coherence properties of the emitted light. The coherence time T_2 can be defined via the excited state's dephasing rate $(T_2)^{-1} = (2T_1)^{-1} + (T_2^*)^{-1}$, with T_1 as the radiative emitter lifetime and T_2^* as the pure dephasing time. True resonant s-shell excitation appears beneficial to avoid pure dephasing, and is therefore highly anticipated to approach the ideal Fourier transform limit [9,10] given by $T_2/(2T_1) = 1$. The observation of the Mollow triplet is the characteristic hallmark of resonance fluorescence from a strongly driven and dressed two-level system. Resonance fluorescence emission from a single QD was first reported by Muller *et al.* [11]. Recently, they also directly observed Mollow triplets [12] with Rabi splittings up to $\Omega \approx 4.4$ GHz (18 μeV), where a distinct pump-power independent dephasing rate of $\Gamma_2^* = (T_2^*)^{-1} \approx 3.1$ GHz has been found [13]. In another remarkable work by Vamivakas *et al.* [14] the resonance fluorescence and spin-resolved characteristics of a trionic QD state have been investigated. The Mollow triplet-sideband emission showed a high degree of coherence with $\Gamma_2^* \approx 18$ MHz. It is important to note that all these investigations have been performed under moderate pumping levels ($\Omega < 6$ GHz) and investigations under stronger s-shell excitation have not been reported so far.

In this Letter, we report on detailed studies of resonance fluorescence from single-QD neutral excitonic X^0 recombination in a high-Q micropillar cavity. The influence of (a) resonant pump-power and (b) frequency detuning of the driving laser (-3 GHz $< \Delta <$ 4 GHz) on the characteristic Mollow sideband emission is investigated, with particular focus on the emission coherence properties.

The sample structure under study is grown by molecular beam epitaxy on GaAs substrate. The initial planar cavity consist of 30 (26) AlAs/GaAs distributed Bragg reflector period pairs as the bottom (top) mirrors, respectively. Spacers of 2×130 nm GaAs form a λ cavity around a single centered layer of (In,Ga)As QDs with a spatial density of $\sim 6 \times 10^9$ cm^{-2}. Ordered fields of different diameter (1.5–4 μm) high-quality micropillars have been finally processed by combined electron-beam lithography and plasma-induced reactive ion etching [15].

Our investigations are performed on a confocal low-temperature ($T \geq 4$ K) microphotoluminescence (μ-PL) setup [10], using a special orthogonal symmetry between lateral optical laser excitation of individual micropillar structures close to the cleaved [110] sample edge and vertical QD emission detection along the pillar axis. A narrow-band (≈ 500 kHz) continuous-wave (cw) Ti: Sapphire ring laser or a mode-locked Ti:Sa pulse laser (~ 2 ps pulses at 76.2 MHz) for time-resolved μ-PL (TCPC: time-correlated photon counting) are used for excitation. In addition to μ-PL detection by a spectrometer/CCD system ($\Delta E_{\mathrm{res}} \sim 35$ μeV), a scanning Fabry-Pérot interferometer [10] (Finesse $F \sim 150$; FSR $= 15$ GHz $\sim$ 62.035 μeV free spectral range) provides high-resolution PL (HRPL) with $\Delta E_{\mathrm{res}}^{\mathrm{HRPL}} < 1\,\mu$eV.

The presented measurements have been performed on a 1.75 μm diameter micropillar. As shown in Fig. 1(a), this microcavity reveals single-QD neutral exciton X^0 s-shell emission at $\sim$1.3571 eV close to the fundamental mode (FM) ($\sim$ 1.3568 eV) at $T = 10$ K under selective *quasiresonant* QD p-shell excitation $\sim$22 meV above the ground

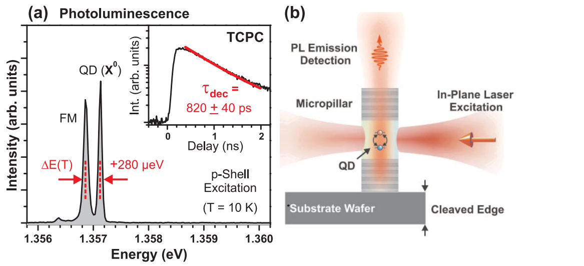

FIG. 1 (color online). (a) Single-QD neutral exciton (X^0) emission spectrally close to the fundamental mode (FM) of a 1.75 μm micropillar cavity, observed at quasiresonant p-shell excitation ($P_0 \approx 50\ \mu$W; $\Delta E(T)$: temperature-dependent QD-mode detuning). Inset: Time-resolved X^0 emission. (b) Schematic geometry of orthogonal excitation and detection.

state. At this QD-FM detuning $\Delta E = +280\ \mu$eV, the spontaneous radiative X^0 decay time has been measured as $\tau_{\text{dec}} = 820 \pm 40$ ps by TCPC under pulsed p-shell excitation [Fig. 1(a) inset]. Because of very fast (ps scale) p- to s-shell carrier relaxation in those QDs, the pure radiative lifetime T_1 obeys $T_1 \approx \tau_{\text{dec}}$. From the FM emission energy and linewidth δE, we derive a high-quality factor of $Q \approx E/\delta E = 13\,500 \pm 500$. The prominent effect of FM emission despite its distinct spectral *detuning* from the dominantly "feeding" single QD is due to nonresonant QD-mode coupling in such solid-state emitter-cavity systems [16–19].

As was first theoretically described by B. R. Mollow [12], a strong resonant light field (detuning $\Delta = 0$) "dresses" the electronic states of a two-level system into a quadruplet, which reflects in characteristic Mollow triplet emission spectra composed of the bare emitter frequency ν_0 and two symmetric satellite peaks at $\nu_0 \pm \Omega$. $\Omega = \mu E_0/h$ denotes the bare Rabi frequency (in Hz) with transition dipole moment μ and local field strength E_0. For $\Delta = 0$ and sufficient pump power P_0, the energetic center-to-sideband Rabi splitting obeys $|\Omega| \propto P_0^{1/2}$.

Figure 2(a) depicts a systematic HRPL series of single-QD exciton s-shell emission under strictly resonant ($\Delta = 0$) power-dependent cw excitation. The emission detuning relative to the bare X^0 line is denoted as δ in units of frequency (GHz) and energy (μeV). Because of the Fabry-Pérot technique in HRPL, all spectral features appear periodically with an offset equal to the interferometer FSR. With increasing power, the evolution of two symmetrically spaced sidebands is clearly observed around the central line which is composed of QD resonance fluorescence and residual scattered laser light. By varying the pump power over more than 2 orders of magnitude, large Rabi splittings of up to $\Omega \approx \pm 15$ GHz ($\pm 62\ \mu$eV) are traced, limited only by the FSR due to the overlap of sidebands with adjacent FPI transmission orders. Extracted Rabi splittings $|\Omega|$ from Fig. 2(a) are plotted in Fig. 2(b) as a function of the square root of laser power, clearly confirming the theoretically expected proportionality. We emphasize that in our studies the investigated excitation

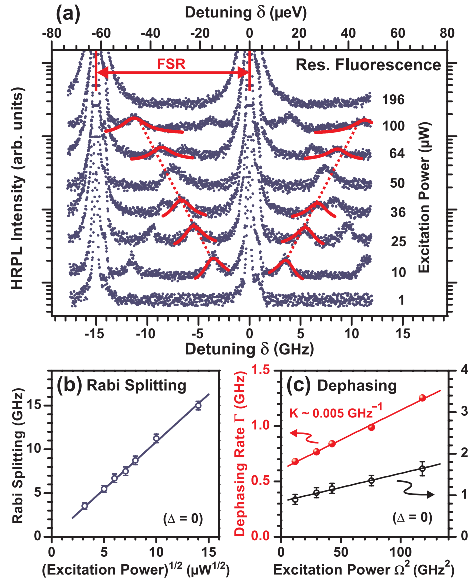

FIG. 2 (color online). (a) Power-dependent high-resolution spectra of X^0 emission at resonant ($\Delta = 0$) cw excitation. (b) Mollow sideband splitting $|\Omega| \sim P_0^{1/2}$ extracted from (a). (c) Power-dependent Mollow sideband FWHM $\Delta\nu$ and total dephasing rate $\Gamma \propto \Omega^2$ from theoretical analysis of data in (a), indicative of excitation-induced dephasing (EID).

power range and consequently the regime to observe Mollow triplets of Rabi splitting $|\Omega|$ are significantly larger than in previous investigations [13,14] using direct detection of QD dressed state fluorescence.

A more detailed inspection of the Mollow triplet series in Fig. 2(a) also reveals a systematic distinct spectral broadening of the Rabi sidebands with increasing power. The spectral line widths $\Delta\nu$ and their gradual broadening have been deduced from Lorentzian least-squares fits to the HRPL data (not shown). Figure 2(c) (black line) depicts corresponding FWHM values (GHz) of the Rabi sidebands, revealing an overall line width increase by a factor of $\sim$1.8 over the observed power range. In particular, a clear linear dependence $\Delta\nu \propto \Omega^2$ on the squared Rabi frequency is traced as a strong indication of excitation-induced dephasing (EID) as an important additional effect accompanying the 'dressed' character of resonant QD emission. As is shown below, our experimental findings are in full quantitative agreement with predictions of a recently presented model on phonon-dressed QD Mollow triplet emission in the cavity-QED regime [20].

Prior to detailed data analysis, we note here that other studies on pulsed resonant single-QD excitation have interpreted EID in terms of (time-domain) Rabi rotation damping to originate from coherent energy exchange between the emitter and a resonant LA-phonon bath in the barrier matrix [21,22]. In addition to pulsed broadband emission, their model addressed InGaAs QDs in n-i-Schottky diode structures *without* a microcavity—in clear contrast to our experiments. Alternatively, carrier scattering between a resonantly *pulsed* single QD in a photodiode structure (without cavity coupling) and off-resonant wetting layer and multiexciton states were theoretically discussed as the origin of Rabi oscillation damping by EID [23]. For resonance fluorescence under pure cw conditions, photon statistics and emission dephasing via phonon-bath coupling of a single QD without cavity coupling have also been analyzed theoretically [24]. In qualitative conformity with all previous studies [21–24], EID rates proportional to the squared Rabi frequency Ω^2 were concluded. Nevertheless, the particular regime of cavity-QED with distinct emitter-mode coupling between a single dressed QD and a high-Q 3D microcavity has not been addressed by either of the previous studies, thus hindering a direct numerical interpretation of our data in Fig. 2. In contrast, we compare our results to a new recently presented theory by Roy and Hughes [20] on cw-excited single-QD resonance fluorescence in a high-Q cavity, which explicitly considers the effects of electron-acoustic phonon-bath coupling and electron-photon coupling to model a full *cavity-QED system* based on a polaron dynamics master equation approach [25].

To quantitatively interpret the observed spectral sideband broadening [Fig. 2(a)], our Mollow triplet spectra were modeled on the theoretical basis of Ref. [13]. Although the authors report no EID effect, the influence of pure dephasing was already accounted for by independent rates of radiative decay $(2T_1)^{-1}$ and decoherence $\Gamma = T_2^{-1}$ (Ref. [13], Eqn. 3). In our analysis, the total dephasing rate was substituted as $\Gamma = (2T_1)^{-1} + K\Omega^2$ to compute the side peak spectra in the dependence on *radiative* and power-dependent *pure dephasing* (EID) with emission broadening $\Delta\nu \propto \Omega^2$. For various fixed values of K (GHz^{-1}), the whole set of Mollow triplet side peak spectra was repeatedly calculated [26]. High consistency over the full power series could be achieved for a dephasing parameter $K = 0.005 \pm 0.001$ GHz^{-1} ($T = 10$ K), well reproducing all side peak spectra [Fig. 2(a), bold red (medium gray) lines] and their overall broadening, i.e., the according dephasing rate Γ extracted in Fig. 2(c). Apart from radiative dephasing no extra constant pure dephasing needed to be included, which anticipates close to ideal Fourier transform-limited resonance fluorescence $T_2/(2T_1) \approx 1$ in the limit $\Omega^2 \to 0$, in full agreement with our previous studies [10].

To numerically compare the observed Mollow side peak broadening with the predictions of the QED model and the coupling regime considered in Ref. [20], we deduced the emitter-cavity coupling strength g for our QD-cavity system from $g \approx (\frac{F_P \kappa_{\text{cav}}}{4\tau_X})^{1/2}$. For a measured Purcell emission enhancement factor of $F_P \approx 13$, a FM cavity-photon loss rate of $\kappa = \frac{2\pi c_0}{Q\cdot\lambda_X} \approx 104$ μeV (25.2 GHz) and an exciton lifetime of $\tau_X = 820 \pm 40$ ps at $\Delta E = +280$ μeV QD-FM detuning [Fig. 1(a)], we obtain $g \approx 16.2 \pm 0.5$ μeV (or 3.9 ± 0.1 GHz), consistent with the regime described by Roy and Hughes [20]. Their calculations, considering pure cw-resonant s-shell excitation at zero detuning ($\Delta = 0$), clearly predict the effect of EID due to combined emitter phonon-bath coupling and cavity-photon bath interaction especially in those cavity-QED systems. Distinct Mollow triplet-sideband broadening $\Delta\nu \propto \Omega^2$ is expected for small and moderate emitter-cavity couplings 15 μeV $\le g \le$ 50 μeV. For parameters very similar to our experimental conditions, Mollow side peak bandwidth enhancement by $K \approx 0.005$–0.007 GHz^{-1} (i.e., $K \approx 0.001$–0.002 μeV^{-1}) is anticipated, in high quantitative conformity with our observations [Fig. 2(c)].

Furthermore, as a direct consequence of phonon-mediated nonresonant cavity feeding from the detuned QD into the detuned FM ($\Delta E = +280$ μeV here), an asymmetry between the Mollow triplet-sideband intensities is expected [20] due to their slightly different spectral detuning from the microcavity mode. Clear indications of this effect could be consistently traced from the theoretical fit of our HRPL data in Fig. 2(a) (bold lines), reflecting $\sim$15% higher intensity from the low frequency Mollow triplet sidebands at $\nu_0 - \Omega$ (i.e., spectrally closer to the FM) with respect to their counterparts at $\nu_0 + \Omega$.

The second part of our studies focused on single-QD Mollow triplet spectra in explicit dependence on laser frequency detuning. According to theory [14], spectral detuning Δ between the driving field and the bare emitter resonance ν_0 characteristically modifies the dressed emission. Besides the center transition at $\nu_0 + \Delta$, the two sideband frequencies become $\nu_0 + \Delta \pm \Omega'$, where $\Omega' = \sqrt{\Omega^2 + \Delta^2}$ denotes the generalized Rabi frequency, with Ω as the (zero detuning) bare Rabi frequency at a given excitation strength. Figure 3(a) depicts HRPL spectra at a fixed pump power of $P_0 \approx 16$ μW, taken under systematic laser frequency detuning from the QD s shell over a large range of -3 GHz $\le \Delta \le +4$ GHz (see arrow markers). The center peak composed of QD resonance fluorescence and intense laser stray light has been removed from the spectra for clarity. Under variation of Δ, a gradual shift of the whole Mollow triplet and a significant increase of sideband splittings with increasing detuning are observable. Figure 3(b) extracts the peak positions of each emission component in Fig. 3(a) as a function of laser detuning Δ, where the horizontal dashed line denotes the symmetry point $\Omega' = \Omega$ of $\Delta = 0$. Fits of the sideband positions according to $\nu(\Delta) = \nu_0 + \Delta \pm \sqrt{\Omega^2 + \Delta^2}$ [Fig. 3(b), bold red (medium gray) lines] reveal high consistence with theory. For a more convenient analysis of the detuning series, the full sideband splittings as a function of Δ are plotted in Fig. 3(c), revealing a minimum for $\Delta \to 0$. From a least-squares fit $\propto 2\sqrt{\Omega^2 + \Delta^2}$ to the data (bold line) at this fixed power level, we derive a bare Rabi frequency of $\Omega = 3.99 \pm 0.06$ GHz. Taking into account this value of Ω together with idealized beam geometry and spatial QD-field overlap for an estimation of the local field strength $|E_{\text{loc}}|$, we can deduce the electric dipole moment of this particular QD as $\mu_{\text{el}} = h\Omega/|E_{\text{loc}}| = 18 \pm 2$ Debye. This

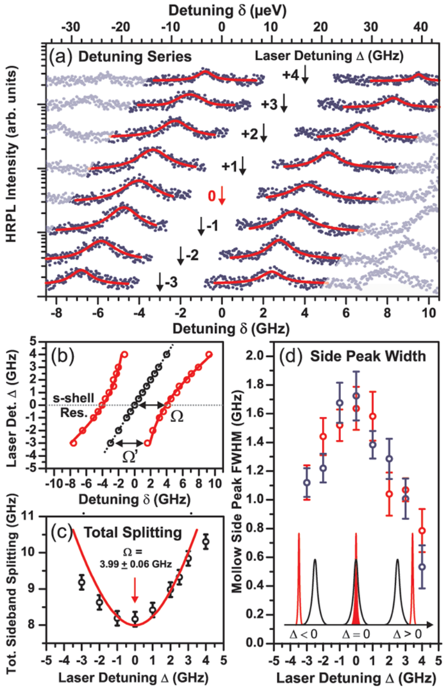

FIG. 3 (color online). (a) HRPL of QD resonance fluorescence under excitation detuning Δ (see arrows) relative to the QD s shell, taken at a fixed power of $P_0 = 16\ \mu$W. Red (medium gray) traces: Lorentzian FWHM fits. (b) Spectral evolution of Mollow sidebands (red [medium gray]) with laser detuning Δ (black center trace), derived from (a). (c) Total sideband splitting in plot (b), together with a theory fit to evaluate the bare Rabi frequency Ω. (d) Sideband FWHM vs laser detuning Δ from plot (a), revealing the phenomenon of FWHM reduction with increasing Δ. Please note a reduced QD-FM detuning $\Delta E = +230\ \mu$eV, yielding $T_1 \sim 410 \pm 20$ ps and correspondingly larger FWHM by a factor of $\sim$1.6 for $\Delta = 0$ conditions with respect to Fig. 2(c).

is only somewhat smaller than recently reported values for neutral X^0 states in similar types of InAs/GaAs QDs [14].

Another interesting phenomenon is traced from an evaluation of the sideband FWHM in the HRPL series of Fig. 3(a), extracted in Fig. 3(d) (color-coded for the red [medium gray] (blue [dark gray]) detuned sidebands). Under increasing laser detuning from the QD s-shell resonance but fixed excitation power ($P_0 \approx 16\ \mu$W), we observe a distinct spectral *narrowing* of either sideband with increasing Δ. This behavior appears unexpected in terms of the cavity-QED-based EID model [20], as dressed emission with increased sideband splittings equivalent to the generalized Rabi frequency $\sqrt{\Omega^2 + \Delta^2}$ should reflect also in increased dephasing [27], i.e., line broadening $\propto \Omega'$—in contrast to our experiment. Though, first indications of aberrations from the theoretically expected detuning dependence of emission are found by detailed inspection of the extracted total sideband splitting with Δ in Fig. 3(c). With increasing laser detuning we observe an increasing deviation between the measured splittings and the theoretical model, which might tentatively be interpreted to result from a systematically reducing bare Rabi frequency independent of the increasing value of Δ. Nevertheless, a final interpretation of this effect has to be left for further ongoing in-depth analysis.

In conclusion, resonance fluorescence emission properties of a single InAs/GaAs QD in a high-quality micropillar cavity have been investigated in detail with particular focus on *excitation power* and/or *excitation detuning* relative to the s-shell ground state. Wide-range excitation power series could trace characteristic Mollow triplet spectra with large Rabi splittings of $\Omega \approx \pm 15$ GHz, accompanied by the effect of systematic spectral sideband broadening $\propto \Omega^2$ as a strong indication of excitation-induced dephasing (EID).

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*Corresponding author
s.ulrich@ihfg.uni-stuttgart.de

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