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DETUNING-DEPENDENT NARROWING OF MOLLOW TRIPLET LINES OF DRIVEN QUANTUM DOTS

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We study the two-time correlation function and the resonance fluorescence spectrum of a semiconductor quantum dot excited by a strong off-resonant laser pulse. The obtained analytic expressions exhibit a specific detuning-dependent damping of Rabi oscillations of the dressed quantum dot as well as a detuning-dependent width of Mollow triplet lines. In the absence of pure dephasing, the central peak of the triplet is broadened upon increasing detuning, but the blue- and red-side peaks are narrowed. We demonstrate that the pure dephasing processes can invert these dependences. A crossover between the regimes of detuning-dependent narrowing and broadening of the side and central peaks is identified. The predicted effects are consistent with recent experimental results and numerical calculations.

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Emission properties of driven semiconductor quantum dots (QDs) have attracted much interest recently due to their potential applications in the fields of photonics and quantum information technology [1–3]. Semiconductor QDs provide an atomlike light–matter interaction demonstrating typical quantum dynamical features of isolated natural atoms. In particular, artificial atoms such as QDs excited by a strong resonant continuous-wave optical field exhibit a resonance fluorescence spectrum containing three peaks, known as the Mollow triplet [4]. Under pulsed resonant excitation in time-resolved experiments, QDs undergo the Rabi oscillations [5, 6]. In the dressed-atom approach [7], these effects are understood as resulting from the quantum transitions in the total coupled system of the atom and driving photons. In contrast with natural atoms, QDs interact with their solid-state environment in a more complicated manner. There is a variety of loss mechanisms for quantum dots [5]. The damping of the Rabi oscillations as well as the width of the Mollow triplet lines can be used for identifying these mechanisms. One of the main consequences of the solid-state character of QDs is a specific dephasing caused by coupling to acoustic phonons. A well-known signature of phonon coupling is revealed in an excitation-induced dephasing with a rate proportional to the square of the effective Rabi frequency. Experimental evidence of the excitation-induced dephasing effects was recently observed as oscillation damping in pulsed photocurrent measurements on a resonantly driven QD [5]. The effect of the excitation-induced dephasing has also been observed as the Mollow triplet sideband broadening under resonant continuous-wave excitation of a single QD in a microcavity [1]. In that paper, the phenomenon of the spectral Mollow sideband narrowing depending on the laser-excitation detuning from the bare emitter resonance was also demonstrated, but that effect had to be left open for further theoretical analysis. Previous numerical studies using the polaron master equation approach with cavity coupling did not reveal the narrowing effect [8]. Recently, it has been shown that a crossover between the regimes of detuning-dependent sideband narrowing and broadening can be qualitatively understood from a theoretical model based on the polaron ME without inclusion of the QD–cavity coupling [3]. Numerical calculations presented in [9] have also shown that for off-resonant driving, narrowing in the spectral sideband width can occur under cer-

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tain conditions. In [10], it was shown that the decay rate of the Rabi oscillations in QDs can decrease as the detuning increases.

In this paper, we analytically calculate the two-time correlation function and resonance fluorescence spectrum of a semiconductor QD excited by an off-resonant laser pulse taking the exciton–phonon interaction into account. Our analytic results allow us to give a clear physical treatment of the detuning-dependent narrowing and broadening of the Mollow triplet lines. We show that pure dephasing processes radically influence the character of these phenomena and can cause a crossover between the detuning-dependent narrowing and broadening of the triplet peaks.

We model the QD as a two-level system with an energy splitting ω_0 between the ground $|1\rangle$ and excited $|2\rangle$ states (we set $\hbar = 1$). The QD is driven by a coherent laser field of a frequency ω_L and is coupled to a phonon bath. The master equation for the density matrix ρ of the QD [3] in the Markovian approximation can be written as

$$i\frac{\partial \rho}{\partial t} = [H, \rho] + i\Lambda\rho, \tag{1}$$

where

$$H = \Delta s^z + \frac{\Omega}{2}(s^+ + s^-) \tag{2}$$

is the Hamiltonian of the QD in the frame rotating at the frequency ω_L, and

$$\Lambda\rho = \frac{\gamma_{21} + \gamma_{ph}^-}{2} D[s^-]\rho + \frac{\gamma_{12} + \gamma_{ph}^+}{2} D[s^+]\rho + \\ + \frac{\eta}{2} D[s^z]\rho - \gamma_{ph}^{cd}(s^+\rho s^+ + s^-\rho s^-) \tag{3}$$

is the relaxation superoperator. Here, $s^{\pm,z}$ are components of the pseudospin operator, describing the QD state and satisfying the commutation relations $[s^+, s^-] = 2s^z$ and $[s^z, s^\pm] = \pm s^\pm$, $\Delta = \omega_0 - \omega_L$, $\Omega = \Omega_0 \exp[-W(T)/2]$, where Ω_0 is the bare Rabi frequency that describes the coherent exciton pumping from the laser field, and $\exp[-W(T)]$ is the Debye–Waller factor, which takes the effect of acoustic phonons on the coherent laser–QD interaction into account and depends on the bath temperature T and the electron–phonon coupling. This factor describes the so-called elastic processes in which the momentum received by the QD from the exciting photon is transmitted to the whole crystal without emission or absorption of a phonon. In addition, $D[O]\rho = 2O\rho O^+ - O^+O\rho - \rho O^+O$, γ_{21} and γ_{12} are the rates of photon radiative processes from the excited state $|2\rangle$ of the QD to its ground state $|1\rangle$ and vice versa, the γ_{ph}^- process corresponds to enhanced radiative decay, while the γ_{ph}^+ process represents the incoherent excitation caused by the exciton–phonon interaction (nonelastic photon–QD scattering processes are realized through multiphonon transitions), γ_{ph}^{cd} is the cross-dephasing rate induced by phonons, and η is the dephasing rate introduced phenomenologically. As is shown in Ref. [3], the relaxation rate $\gamma_{ph} = \gamma_{ph}^- + \gamma_{ph}^+$ weakly depends on Δ in the detuning range considered below and can be approximated as $\gamma_{ph} = k\Omega^2$, where k is a coefficient. For sufficiently large driven strengths, $\Omega \gg \gamma_{ph}^{cd}$, the cross-dephasing term in Eq. (3) can be neglected. This term rapidly oscillates in the interaction representation with Hamiltonian (2) and gives only corrections of the second and higher orders in γ_{ph}^{cd}/Ω.

After the canonical transformation $\rho_1 = u^+\rho u$, where $u = \exp[-\theta(s^+ - s^-)/2]$, the master equation (1) is transformed into

$$\begin{gathered} i\frac{\partial \rho_1}{\partial t} = [H_1, \rho_1] + i\Delta_1\rho_1, \\ H_1 = u^+Hu - iu^+\frac{\partial u}{\partial t} = \varepsilon s^z, \\ \Lambda_1 = u^+\Lambda u = \frac{\Gamma_\downarrow}{2} D[s^-] + \frac{\Gamma_\uparrow}{2} D[s^+] + \frac{\Gamma_\varphi}{2} D[s^z], \end{gathered} \tag{4}$$

where $\varepsilon = \sqrt{\Delta^2 + \Omega^2}$,

$$\Gamma_\downarrow = \frac{1}{4}(\gamma_{21} + \gamma_{12} + \gamma_{ph}^- + \gamma_{ph}^+)(1 + \cos^2\theta) + \\ + \frac{1}{2}(\gamma_{21} + \gamma_{ph}^- - \gamma_{12} - \gamma_{ph}^+)\cos\theta + \frac{1}{4}\eta\sin^2\theta,$$

$$\Gamma_\uparrow = \frac{1}{4}(\gamma_{21} + \gamma_{12} + \gamma_{ph}^- + \gamma_{ph}^+)(1 + \cos^2\theta) - \\ - \frac{1}{2}(\gamma_{21} + \gamma_{ph}^- - \gamma_{12} - \gamma_{ph}^+)\cos\theta + \frac{1}{4}\eta\sin^2\theta,$$

$$\Gamma_\varphi = \eta\cos^2\theta + (\gamma_{21} + \gamma_{12} + \gamma_{ph}^- + \gamma_{ph}^+)\sin^2\theta,$$

$$\cos\theta = \Delta/\varepsilon, \quad \sin\theta = \Omega/\varepsilon.$$

We obtain the relaxation superoperator in the rotating wave approximation (RWA). In this case, because $\Gamma_\downarrow, \Gamma_\uparrow, \Gamma_\varphi \ll \varepsilon$ for the strong laser–QD interaction, the nondiagonal terms that contain the products of spin operator pairs $(s^\pm, s^z)$, (s^+, s^+), and (s^-, s^-) are neglected in the structure of the operator. The analogous form of the relaxation superoperator was obtained in [11–13] in describing lasing and amplification effects in superconducting qubits.

The solution of Eq. (4) can be written as

$$\rho_1(t) = \exp[(-iL_1 + \Lambda_1)t]\,\rho_1(0). \tag{5}$$

The superoperator L_1 acts in accordance with the rule $L_1 X = [H_1, X]$. The density matrix $\rho(t)$ in the laboratory frame is given by

$$\rho(t) = u_1 \rho_1(t) u_1^+, \tag{6}$$

where $\rho_1(t)$ is defined by Eq. (4) and $\rho_1(0) = u_1^+ \rho(0) u_1$. Moreover, if the QD is in the ground state, then $\rho(0) = 1/2 - s^z$ and we have

$$\rho_1(0) = \frac{1}{2} + \frac{1}{2}(s^+ + s^-)\sin\theta - s^z \cos\theta.$$

The following relations can be verified by direct calculation:

$$\begin{gathered}
\exp\left[(-iL_1 + \Lambda_1)t\right] s^\pm = \exp\left[(\mp i\varepsilon - \Gamma_\perp)t\right] s^\pm, \\
\exp\left[(-iL_1 + \Lambda_1)t\right] s^z = \exp(-\Gamma_\parallel t) s^z, \\
\exp\left[(-iL_1 + \Lambda_1)t\right] a = \\
= \left\{1 + 2\sigma_0 \left[1 - \exp(-\Gamma_\parallel t)\right] s^z\right\} a,
\end{gathered} \tag{7}$$

where $a = \text{const}$,

$$\begin{gathered}
\sigma_0 = -\frac{\Gamma_\downarrow - \Gamma_\uparrow}{\Gamma_\parallel} = \\
= -\frac{\gamma_{21} + \gamma_{ph}^- - \gamma_{12} - \gamma_{ph}^+}{\Gamma_\parallel}\cos\theta, \\
\Gamma_\parallel = \Gamma_\downarrow + \Gamma_\uparrow = \tilde{\gamma}_\parallel + \left(\tilde{\gamma}_\perp - \tilde{\gamma}_\parallel\right)\sin^2\theta, \\
\Gamma_\perp = \frac{1}{2}(\Gamma_\downarrow + \Gamma_\uparrow + \Gamma_\varphi) = \tilde{\gamma}_\perp - \frac{1}{2}\left(\tilde{\gamma}_\perp - \tilde{\gamma}_\parallel\right)\sin^2\theta, \\
\tilde{\gamma}_\parallel = \gamma_\parallel + \gamma_{ph}, \quad \tilde{\gamma}_\perp = \gamma_\perp + \frac{\gamma_{ph}}{2}, \\
\gamma_\parallel = \gamma_{12} + \gamma_{21}, \quad \gamma_\perp = \frac{1}{2}(\gamma_{12} + \gamma_{21} + \eta), \\
\gamma_{ph} = \gamma_{ph}^- + \gamma_{ph}^+.
\end{gathered} \tag{8}$$

Using Eqs. (4)–(8), we obtain the density matrix in the laboratory frame

$$\begin{gathered}
\rho_{lab}(t) = \frac{1}{2} + \frac{1}{4}\{[\sin\theta(\cos\theta + 1) \times \\
\times \exp\left(-i(\omega_L + \varepsilon)t - \Gamma_\perp t\right) + \sin\theta(\cos\theta - 1) \times \\
\times \exp\left(-i(\omega_L - \varepsilon)t - \Gamma_\perp t\right)] s^+ + \text{H.c.}\} - \\
-\frac{1}{2}\sin\theta\left[\cos\theta\exp(-\Gamma_\parallel t) + \frac{\Gamma_\downarrow - \Gamma_\uparrow}{\Gamma_\parallel}\left[1 - \exp(-\Gamma_\parallel t)\right]\right] \times \\
\times\left[s^+\exp(-i\omega_L t) + \text{H.c.}\right] - \Bigg\{\exp(-\Gamma_\perp t)\sin^2\theta\cos\varepsilon t + \\
+ \cos\theta\left[\cos\theta\exp(-\Gamma_\parallel t) + \frac{\Gamma_\downarrow - \Gamma_\uparrow}{\Gamma_\parallel} \times \right. \\
\left. \times \left[1 - \exp(-\Gamma_\parallel t)\right]\right]\Bigg\} s^z.
\end{gathered} \tag{9}$$

We now find the two-time correlation function

$$\begin{gathered}
g^{(1)}(t) = \langle s^-(t)s^+(0)\rangle = \langle s^-(t)\rangle_{s^+(0)\rho(0)} = \\
= \frac{1}{2}\sin^2\theta\exp(-i\omega_L t - \Gamma_\parallel t) + \frac{1}{4}(1 + \cos\theta)^2 \times \\
\times \exp\left[-i(\varepsilon + \omega_L)t - \Gamma_\perp t\right] + \frac{1}{4}(1 - \cos\theta)^2 \times \\
\times \exp\left[-i(\varepsilon - \omega_L)t - \Gamma_\perp t\right]
\end{gathered} \tag{10}$$

and the spectral density of the emitted radiation

$$\begin{gathered}
S(\omega) = \frac{1}{\pi}\,\text{Re}\int\limits_0^\infty dt\, e^{i\omega t}\langle s^-(t)s^+(0)\rangle = \\
= \frac{1}{2\pi}\left[\frac{\Gamma_\parallel\sin^2\theta}{\Gamma_\parallel^2 + (\omega - \omega_L)^2} + \frac{\Gamma_\perp(1 + \cos\theta)^2/2}{\Gamma_\perp^2 + (\omega - \varepsilon - \omega_L)^2} + \right. \\
\left. + \frac{\Gamma_\perp(1 - \cos\theta)^2/2}{\Gamma_\perp^2 + (\omega + \varepsilon - \omega_L)^2}\right].
\end{gathered} \tag{11}$$

It follows from Eqs. (8) and (10) that the respective decay rates of the oscillations at the side ($\omega_L \pm \varepsilon$) and central (ω_L) frequencies are

$$\begin{gathered}
\Gamma_\perp = \frac{1}{2}(\gamma_\parallel + \gamma_{ph} + \eta) + \frac{1}{4}(\gamma_\parallel + \gamma_{ph} - \eta)\frac{\Omega^2}{\Delta^2 + \Omega^2}, \\
\Gamma_\parallel = \gamma_\parallel + \gamma_{ph} - \frac{1}{2}(\gamma_\parallel + \gamma_{ph} - \eta)\frac{\Omega^2}{\Delta^2 + \Omega^2}.
\end{gathered} \tag{12}$$

According to Eq. (11), the full width at half maximum (FWHM) of the side and central lines of the Mollow triplet are respectively equal to $2\Gamma_\perp$ and $2\Gamma_\parallel$.

We conclude from Eqs. (8) and (12) that the rates of the transverse $\tilde{\gamma}_\perp$ and longitudinal $\tilde{\gamma}_\parallel$ relaxations determine the character of the detuning dependences of the decay rates for oscillations at the side ($\omega_L \pm \varepsilon$) and central (ω_L) frequencies. If $\tilde{\gamma}_\perp > \tilde{\gamma}_\parallel$, for a fixed driven strength, the function $\sin^2\theta = \Omega^2/(\Delta^2 + \Omega^2)$ decreases with the increasing detuning Δ, which results in increasing the decay rate $\Gamma_\perp$ of the side oscillations. At the same time, the decay rate $\Gamma_\parallel$ of the central oscillation decreases. If $\tilde{\gamma}_\perp < \tilde{\gamma}_\parallel$, the decay rate $\Gamma_\perp$ of the side oscillations decreases with the increasing detuning, while the decay rate $\Gamma_\parallel$ of the central oscillation increases. Obviously, the narrowing of the Mollow sidebands with increasing detuning can be realized without the exciton–phonon interaction (see Eq. (12)). In that case ($\gamma_{ph} = 0$), the condition $\gamma_\perp < \gamma_\parallel$ must be fulfilled. This inequality can be realized when the contribution of pure dephasing processes to the linewidth is smaller than the natural linewidth. We note that at $\tilde{\gamma}_\perp = \tilde{\gamma}_\parallel$ (or $\gamma_\perp = \gamma_\parallel$ for $\gamma_{ph} = 0$), the linewidths of the sidebands as well as the central peak do not depend on detuning.

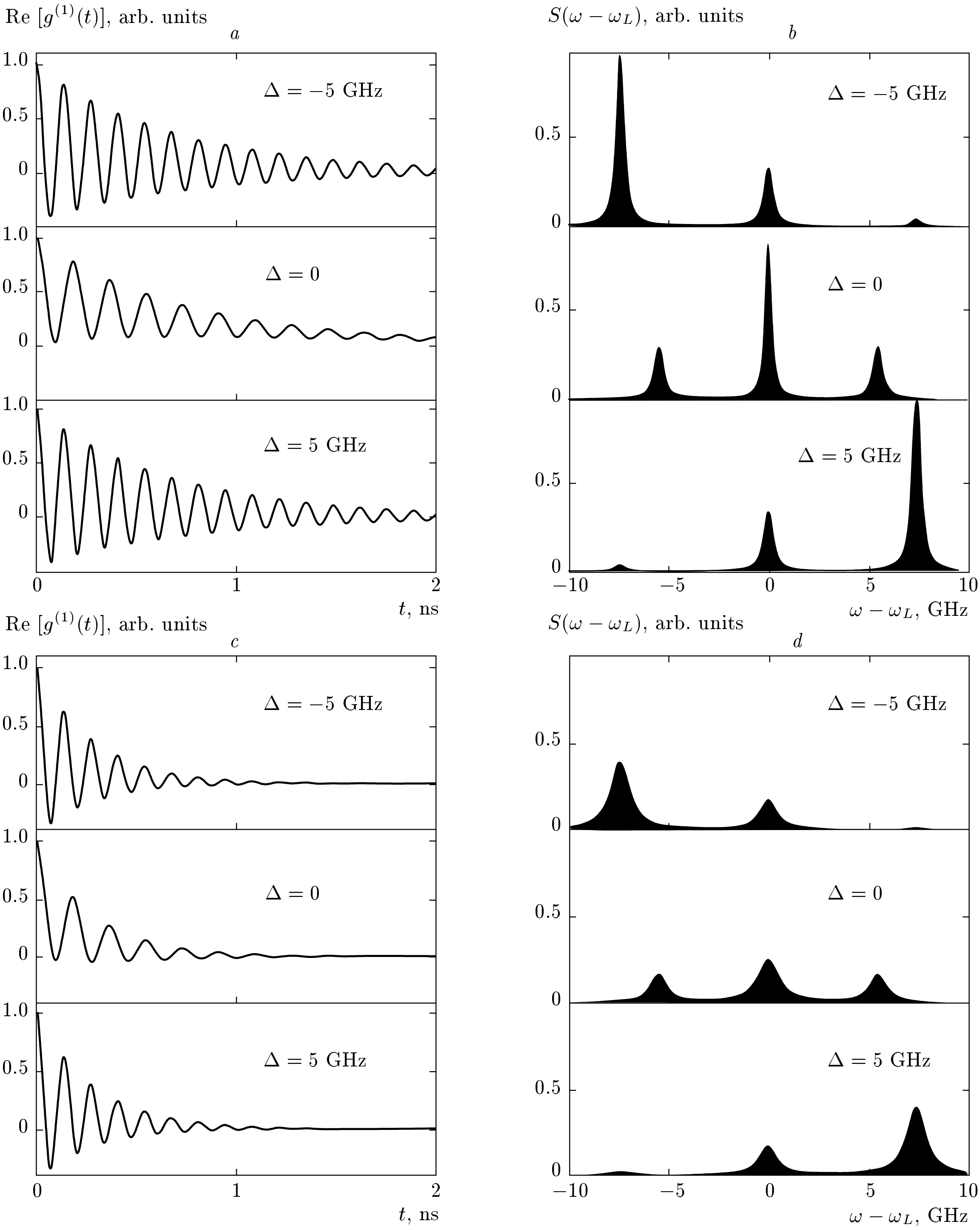

Fig. 1. (*a* and *c*) The two-time correlation functions and (*b* and *d*) the QD emission spectra for different values of the laser–QD detuning at a fixed $\Omega/2\pi = 5.5$ GHz. The parameters employed are $\gamma_{\|} = 0.2$ GHz, $\gamma_{ph} = 0.12$ GHz, (*a* and *b*) for $\eta = 0$, (*c* and *d*) for $\eta = 0.94$ GHz

Thus, in the absence of pure dephasing, the narrowing of the Mollow sidebands with the increasing laser–QD detuning is realized. Pure dephasing processes can bring about a crossover between the narrowing and broadening regimes. If phonon–QD interactions do not result in pure dephasing processes and the relaxation rate $\gamma_{ph} = k\Omega^2$ is constant in the considered detuning range, then phonon–QD interactions do not influence the detuning dependence and only change the condition of the crossover, $\eta = \gamma_{\parallel} + \gamma_{ph}$. Cross-relaxation processes are usually too weak to change this behavior noticeably.

Figure 1 displays the two-time correlation functions and the QD emission spectra versus the laser–QD detuning at a fixed driven strength for two dephasing rates. The correlation functions are shown in the rotating frame and represent the Rabi oscillations. The off-resonant laser excitation modifies the intensity and the linewidth of each peak in the Mollow triplet differently. Moreover, the pure dephasing strongly influences the modification. Figures 1*a* and 1*b* display the emission properties in the absence of pure dephasing. At $\eta = 0$, the central peak undergoes broadening and decreasing of its intensity with the increasing laser detuning. By contrast, we observe narrowing of the sidebands and different changes in intensities for the red and blue sidebands. With the increasing positive (negative) detuning, the amplitude of the blue sideband increases (decreases), whereas the amplitude of the red sideband decreases (increases). Our results demonstrate a systematic narrowing of the Mollow sidebands and a broadening of the central peak with the increasing laser detuning. As we can infer from Figs. 1*c* and 1*d*, pure dephasing processes invert these dependences.

In Fig. 2, we plot the dependences of the amplitudes of the side and central peaks on the laser detuning for the spectra presented in Fig. 1. Figure 3 depicts the FWHM values of these spectral lines. We can see that the sideband narrowing and the central-peak broadening occur when $\eta < \gamma_{\parallel} + \gamma_{ph}$. By contrast, for $\eta > \gamma_{\parallel} + \gamma_{ph}$, the strong pure dephasing results in the broadening of the sidebands and narrowing of the central peak under off-resonant excitation. Moreover, at $\eta = \gamma_{\parallel} + \gamma_{ph}$, Fig. 3 shows a crossover between the narrowing and broadening regimes. In that case, the FWHM values of all peaks of the Mollow triplet are equal and do not depend on detuning.

In Fig. 4, we compare our analytic description with the experimental results for sideband broadening with the increasing laser–QD detuning [3]. The parameters used are the same as in Ref. [3] except that the cross-dephasing term is neglected and the value of η is larger

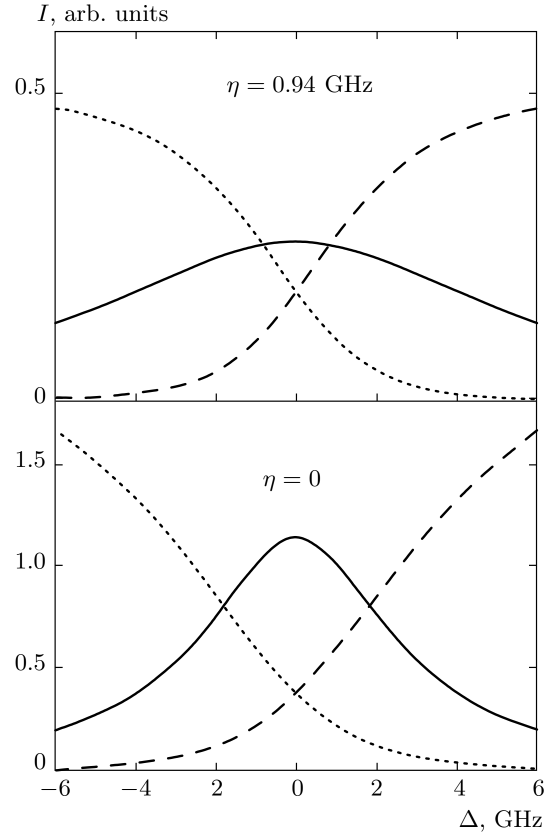

Fig. 2. The amplitudes of the blue (dot line), red (dashed line), and central (solid line) peaks of the Mollow triplet versus the laser–QD detuning at a fixed $\Omega/2\pi = 5.5$ GHz for (*a*) $\eta = 0$ and (*b*) $\eta = 0.94$ GHz. The relaxation rates $\gamma_{\parallel}$ and γ_{ph} are the same as in Fig. 1

(by about 10 %). We note that except for the cross-dephasing term, Eqs. (12) coincide with the expressions in Ref. [3] for the on-resonance ($\Delta = 0$) FWHM of the side and central lines. The analytic curve is in good agreement with the experimental data and lies between the values obtained numerically [3]. According to the theoretical prediction in Ref. [3], the FWHM values of the blue and red Mollow sidebands should have distinct broadening with the increasing laser–QD detuning. The precision of experimental data is not sufficient to confirm this prediction.

As one can see from Eq. (12) and Fig. 3, the sideband narrowing occurs when $\eta < \gamma_{\parallel} + \gamma_{ph}$. In this regime, our analytical results for the sideband FWHM values are consistent with the recent numerical calculations [9] obtained at identical parameters and with the observed experimental trend [1] (Fig. 5). In a more detailed analysis of the observed narrowing effect, the

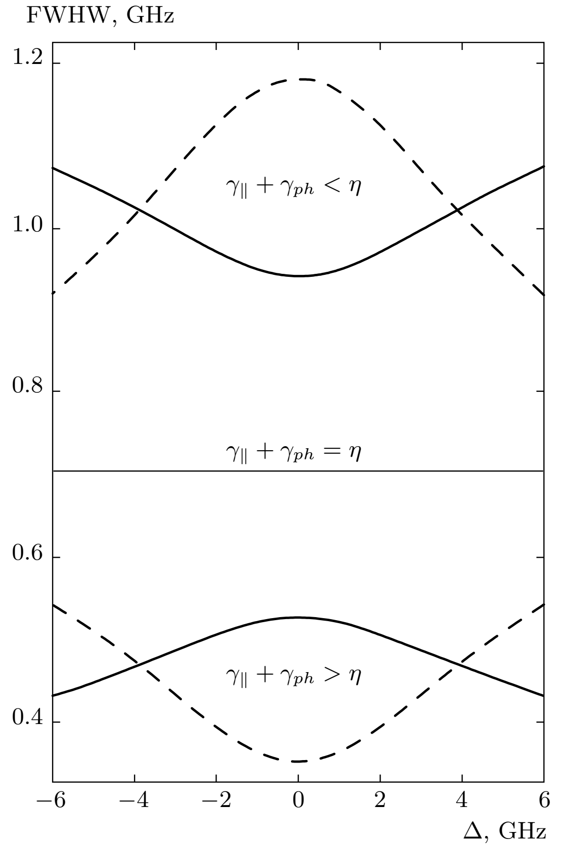

Fig. 3. The widths of the side (solid line) and central (dashed line) peaks of the Mollow triplet as functions of the laser–QD detuning at a fixed $\Omega/2\pi = 5.5$ GHz for $\eta = 0$, $\eta = \gamma_{\parallel} + \gamma_{ph}$, and $\eta > \gamma_{\parallel} + \gamma_{ph}$ ($\eta = 0.94$ GHz). The relaxation rates $\gamma_{\parallel}$ and γ_{ph} are the same as in Fig. 1

systematical reducing of the Rabi frequency Ω with the increasing value of Δ as well as the cavity effects should be taken into account [1]. The Ω-reducing can strongly intensify the sideband narrowing effect with the increasing laser–QD detuning.

In the model used, the strong laser field drastically modifies the eigenstates of a two-level QD, resulting in the additional splitting of their energy levels. Because the splitting is much smaller than the energy difference between the bare states, the appearance of low-frequency transitions (at the Rabi frequency) in the dressed QD enables the effective interaction between the QD and phonon excitations of environment. The participation of phonons in the elastic processes of the QD–photon interactions only renormalizes the constant of the excition–photon interaction. On the other hand, participating in the nonelastic processes, phonons provide an additional channel of energetic and phase re-

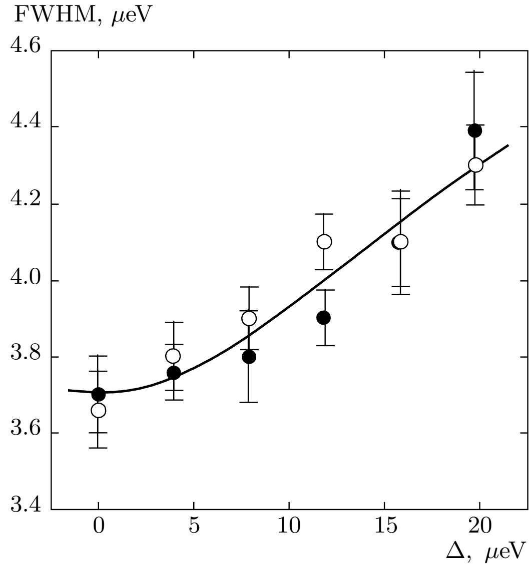

Fig. 4. The FWHM of the Mollow sidebands versus the laser–QD detuning. Circles denote the experimental results adopted from [3]. The solid line is calculated using Eq. (12) with the parameters $\Omega = 22.7$ μeV, $\gamma_{\parallel} = 0.84$ μeV, $\gamma_{ph} = 0.34$ μeV, and $\eta = 3.89$ μeV

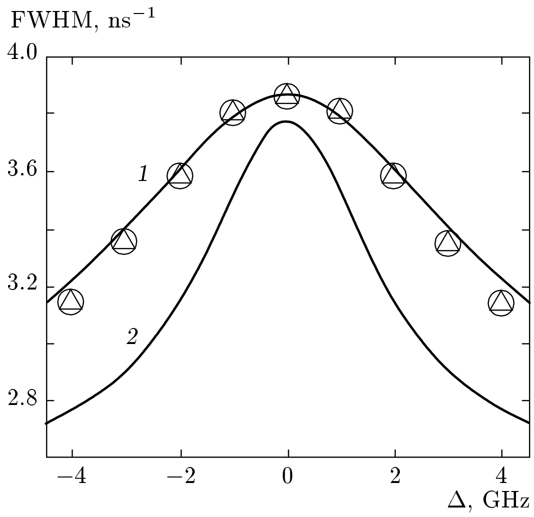

Fig. 5. The FWHM of the Mollow sidebands as a function of the laser–QD detuning at two fixed Rabi frequencies $\Omega/2\pi = 4$ GHz, $\gamma_{ph} = 0.08$ ns^{-1} (line *1*) and $\Omega/2\pi = 2$ GHz, $\gamma_{ph} = 0.02$ ns^{-1} (line *2*) for $\eta = 0$ and $\gamma_{\parallel} = 2.5$ ns^{-1}. The symbols correspond to the numerical calculations in Ref. [9] for the red (○) and blue (△) sidebands

laxations. Due to the specific interaction between the QD dressed states and phonons, the relaxation rates are characterized by the quadratic dependence on the driving strength. The high-field limit used means that the Rabi frequency is much larger than the relaxation rates of the QD. Therefore, the non-RWA terms in the relaxation operator in the representation of the bare and dressed QD states can be neglected. That fact allows obtaining the approximate analytical expressions for the time-resolved resonance fluorescence and the spectrum of scattering emission. The obtained simple explicit expressions show that the dressing of the QD states by the strong driven field mixes the relaxation rates. For example, the phase relaxation of the dressed states turns out to depend on both the phase and energetic relaxations of the bare QD. The degree of such mixing depends on the exciton–photon interaction and the laser–QD detuning.

One should note that we demonstrate the detuning-dependent sideband narrowing without cavity coupling. Moreover, this effect is not specific only for QDs and it can be observed for any two-level system when dephasing processes do not significantly broaden the natural line.

In conclusion, we have analytically described the time-resolved and spectral features of photon emission from a QD excited by an off-resonant laser pulse. We have shown that pure dephasing processes influence radically the detuning-dependent damping of the Rabi oscillations of the dressed QD as well as the width of the Mollow triplet lines. At weak pure dephasing, the central peak of the triplet is broadened with the increasing detuning, but the blue and red side peaks are narrowed. For stronger pure dephasing, the crossover between the narrowing and broadening regimes is realized. We have found a good agreement between our approximate analytical description and the recent experimental results and numerical calculations. Our approach can be applied to describe the off-resonant emission features of a wide range of two-level systems.

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