Emission-Frequency Separated High Quality Single-Photon Sources Enabled by Phonons

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We demonstrate theoretically that the single-photon purity of photons emitted from a quantum dot exciton prepared by phonon-assisted off-resonant excitation can be significantly higher in a wide range of parameters than that obtained by resonant preparation for otherwise identical conditions. Despite the off-resonant excitation, the brightness stays on a high level. These surprising findings exploit the fact that the phonon-assisted preparation is a two-step process where phonons first lead to a relaxation between laser-dressed states while high exciton occupations are reached only with a delay to the laser pulse maximum by adiabatically undressing the dot states. Due to this delay, possible subsequent processes, in particular multiphoton excitations, appear at a time when the laser pulse is almost gone. The resulting suppression of reexcitation processes increases the single-photon purity. Due to the spectral separation of the signal photons from the laser frequencies this enables the emission of high quality single photons not disturbed by a laser background while taking advantage of the robustness of the phonon assisted scheme.

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On-demand single-photon sources continue to gain attention as key building blocks in quantum technological applications, ranging from novel metrology over quantum communication to quantum computing. Semiconductor quantum dots (QDs) have proven to be suitable single-photon emitters [1–8] that, due to their high compatibility with existing semiconductor technology, are promising candidates for device applications. In contrast to atomic systems, these nanoscale structures are prone to the influence of the surrounding solid state crystal matrix. Longitudinal acoustic (LA) phonons are the main source of decoherence of excitons in semiconductor QDs even at cryogenic temperatures of a few kelvin [9–13]. Nevertheless, phonon-assisted off-resonant QD excitations have been shown to provide a robust alternative to resonant exciton preparation schemes [14–18]. In this Letter, we demonstrate theoretically that, quite unexpectedly, the coupling to LA phonons combined with off-resonant driving can be extremely beneficial for a single-photon source based on a QD-cavity system, allowing for the generation of high-quality single-photons that are easily detectable due to their spectral separation from the laser pulses used for the excitation of the QD.

Placing a QD in a cavity strongly enhances the photon emission by enlarging the effective dot-cavity coupling and by setting a preferable emission axis. When exciting the QD exciton resonantly, the frequencies of the excitation and the signal are identical—separating the two is a formidable experimental challenge. In fact, spectral separability is achievable, e.g., by wetting layer excitation or by exciting the biexciton via the two-photon resonance and subsequently exploiting the biexciton-exciton cascade [8,19]. But while the former introduces a time jitter that reduces the on-demand character of the photon source, the latter is sensitive to small fluctuations of excitation parameters such as the laser energy and the pulse area. Both problems are overcome by an off-resonant excitation of the quantum dot, which is thus extremely advantageous. Indeed, it has recently been shown that the robustness of off-resonant excitation schemes paves the way to excite two spatially separated QDs with different transition energies simultaneously with the same laser pulse, which is a milestone towards the scalability of complex quantum networks [20].

The quality of a QD-cavity system as an on-demand single-photon source is typically quantified by several key figures of merit, such as the single-photon purity $\mathcal{P}$ and the brightness $\mathcal{B}$. While the former measures whether indeed a single photon is emitted by the source, the latter characterizes its total photon yield [5]. When $\mathcal{P} = \mathcal{B} = 1$, the source emits a single photon with a probability of unity at every excitation pulse via the cavity. The single-photon purity (SPP) can be extracted from a Hanbury Brown–Twiss coincidence experiment [3,7,8,21–24], which gives a conditional probability to detect a second photon when a first one has already been detected. Suppressing this probability is possible, e.g., by parametric down-conversion, which enhances the SPP, albeit at the cost of a severely reduced brightness of the photon source [25]. Maximizing both SPP and brightness is of utmost importance to create efficient single-photon emitters.

Simultaneously large $\mathcal{P}$ and $\mathcal{B}$ in a QD-cavity system can be achieved by exciting the dot resonantly by ultrashort

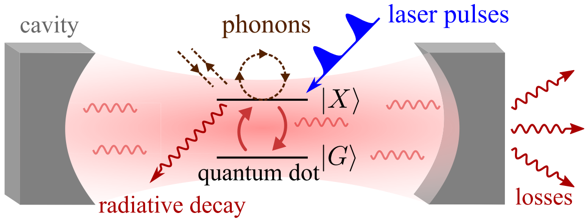

FIG. 1. Sketch of the system under consideration. A two-level QD with a ground state $|G\rangle$ and an exciton state $|X\rangle$ is coupled to a lossy single-mode microcavity. The $|G\rangle \to |X\rangle$ transition is driven by external laser pulses and the exciton state is coupled to LA phonons in a pure-dephasing manner. Finally, the dot can decay radiatively.

laser pulses [3,4,7]. However, shortening the pulse duration is equivalent to widening it spectrally. The detrimental influence of exciting higher-lying states, especially the biexciton state of the QD by short pulses is discussed in Ref. [26]. In view of the various advantages of phonon-assisted off-resonant excitations listed above, the question arises how photonic characteristics such as SPP and brightness perform under off-resonant schemes. In short, we want to explore whether all of the advantages of phonon-assisted off-resonant schemes come at the cost of severely reduced photonic properties.

It is expected that driving a QD off-resonantly is much less efficient. For longer and stronger pulses the resulting quantum state of a QD-cavity system contains an admixture of multiphoton states, which reduces the SPP. Phonon-induced dephasing is expected to degrade the quantum state even further. But paradoxically quite the opposite can take place: a combination of off-resonant driving with the phonon-induced relaxation between laser-dressed QD states leads eventually to high exciton occupations in a subsequent adiabatic undressing process [27]. In this Letter, we demonstrate that the delay of the exciton creation caused by the undressing suppresses the probability for multiphoton generation. Therefore, comparing off-resonant and resonant excitation with otherwise same conditions may, quite unexpectedly, yield enhanced SPPs in the off-resonant case. The best values predicted in this Letter are even comparable to the best values obtained so far within resonant schemes addressing the exciton.

We model the QD-cavity system as a laser-driven two-level system with a ground state $|G\rangle$ and an excited state $|X\rangle$, $H_{\mathrm{DL}} = -\hbar\Delta\omega_{\mathrm{LX}}|X\rangle\langle X| - (\hbar/2)f(t)(|X\rangle\langle G| + |G\rangle\langle X|)$, coupled to a single-mode microcavity (cf., Fig. 1), $H_{\mathrm{C}} = \hbar\Delta\omega_{\mathrm{CL}}a^\dagger a + \hbar g(a^\dagger|G\rangle\langle X| + a|X\rangle\langle G|)$, which is on resonance with the QD exciton. Here, $\Delta\omega_{\mathrm{LX}}$ and $\Delta\omega_{\mathrm{CL}}$ are the laser-exciton and cavity-laser detuning, respectively, and a is the photon annihilation operator in the cavity, which is coupled to the dot by the coupling constant g. A train of Gaussian pulses is assumed represented by the laser envelope function $f(t)$. The excitation can leave the system either via radiative decay or cavity losses modeled by Lindblad rates γ and κ, respectively. Finally, the exciton is coupled to a continuum of LA phonons in a pure-dephasing manner [28], $H_{\mathrm{Ph}} = \hbar\sum_{\mathbf{q}}\omega_{\mathbf{q}}b^\dagger_{\mathbf{q}}b_{\mathbf{q}} + \hbar\sum_{\mathbf{q}}(\gamma^{\mathrm{X}}_{\mathbf{q}}b^\dagger_{\mathbf{q}} + \gamma^{\mathrm{X}*}_{\mathbf{q}}b_{\mathbf{q}})|X\rangle\langle X|$. $b_{\mathbf{q}}$ annihilates a phonon in the mode $\mathbf{q}$ coupled to the dot by the coupling constant $\gamma^{\mathrm{X}}_{\mathbf{q}}$. Full details of the model and of our numerical approach are given in the Supplemental Material [29]. It is worthwhile to note that we use path-integral methods for our simulations that allow us to perform all simulations without approximation to the model [29,36–38].

For the calculations, standard GaAs parameters are used [39] for a QD of 6 nm diameter (for details on the phonon coupling consider the Supplemental Material [29]). If not stated otherwise, the excitation pulse full width at half maximum is set to 7 ps, the cavity mode is resonant with the QD transition, the dot-cavity coupling is $\hbar g = 50\ \mu$eV, the radiative decay rate is $\hbar\gamma = 20\ \mu$eV, and the cavity loss rate is $\hbar\kappa = 50\ \mu$eV. This corresponds to a Purcell factor of $F_{\mathrm{P}} = g^2/(\gamma\kappa) = 2.5$. The initial phonon distribution is assumed to be thermal with a temperature of $T = 4.2$ K.

The main target quantities of interest in this Letter, the SPP $\mathcal{P}$ and the brightness $\mathcal{B}$, are obtained from path-integral simulations of the two-time photonic correlation function $G^{(2)}(t,\tau) = \langle a^\dagger(t)a^\dagger(t+\tau)a(t+\tau)a(t)\rangle$ and the time dependent photon occupation $\langle a^\dagger a\rangle(t)$, respectively. In order to express the SPP in terms of $G^{(2)}(t,\tau)$ one first needs to take the average over the first time argument t, i.e., $G^{(2)}(\tau) = \int_{-\infty}^{\infty} dt G^{(2)}(t,\tau)$, which yields a function with the delay time τ of the coincidence measurement as its single argument. The probability p of detecting a second photon during the same excitation pulse after a first one has already been emitted thus can be obtained by

$$p = \frac{\int_{-T_{\mathrm{Pulse}}/2}^{T_{\mathrm{Pulse}}/2} d\tau G^{(2)}(\tau)}{\int_{T_{\mathrm{Pulse}}/2}^{3T_{\mathrm{Pulse}}/2} d\tau G^{(2)}(\tau)}, \tag{1}$$

where T_{Pulse} is the separation of the pulses in the pulse train. The SPP is then defined as $\mathcal{P} = 1 - p$. Note that $-\infty < \mathcal{P} \leq 1$, where the lack of a lower bound is due to the possibility of bunching instead of antibunching.

In this Letter, the brightness of the source is modeled as the integrated leakage of the average photon number during the duration of one pulse, i.e., $\mathcal{B} = \kappa\int_{-T_{\mathrm{Pulse}}/2}^{T_{\mathrm{Pulse}}/2} dt\langle a^\dagger a\rangle(t)$. Due to the definition, this quantity formally ranges in $0 \leq \mathcal{B} < \infty$ without an upper bound since in principal infinitely many photons can exist in a single electromagnetic field mode.

In Fig. 2(a) the brightness simulated without phonons is shown as a function of the detuning $\Delta\omega_{\mathrm{LX}}$ between the central laser frequency and the transition frequency connecting the ground and the exciton state of the QD as well as the pulse area Θ. An oscillatory behavior as a function of the pulse area with maxima at odd multiples of π is observed [cf., Fig. 2(a)]. This is a consequence of the well-known Rabi rotation of the exciton occupation since

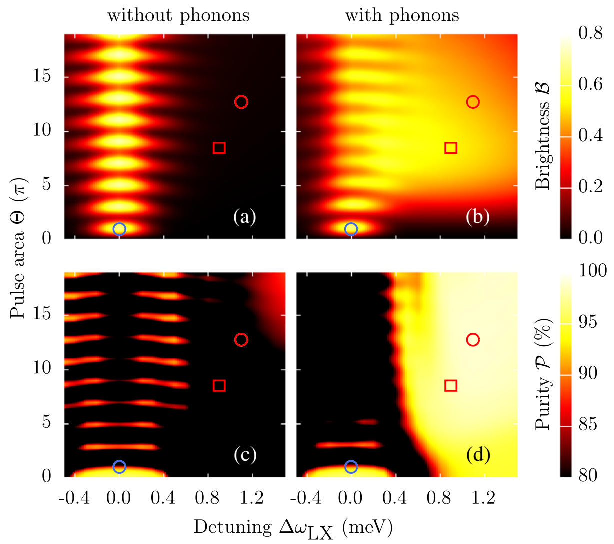

FIG. 2. Brightness $\mathcal{B}$ [panels (a) and (b)] and SPP $\mathcal{P}$ [panels (c) and (d)] as a function of the laser-exciton detuning $\Delta\omega_{\mathrm{LX}}$ and the excitation pulse area Θ of a pulse in the pulse train. The left column (a), (c) is the result of a phonon-free calculation, the right column (b), (d) includes the coupling to a continuum of LA phonons. Blue circle: resonant π-pulse excitation. Red circle: maximal SPP (with phonons). Red square: optimal SPP and brightness for off-resonant excitation (with phonons).

the exciton feeds the cavity photons, which in turn are measured by the brightness. As a function of the detuning, the regions of high brightness are confined to a fairly small range around resonance. The inclusion of phonons drastically changes this picture [cf., Fig. 2(b)]. Through off-resonant excitation with detunings that can be bridged by the emission of LA phonons, a nonvaninshing brightness can be obtained in a previously dark region. Note that the asymmetry with respect to the sign of the detuning is due to the low temperature of $T = 4.2$ K considered here where phonon absorption is largely suppressed.

The SPP in the phonon-free case [cf., Fig. 2(c)] also displays Rabi rotational behavior but decreases with rising pulse area close to resonance, which is due to a reexcitation of the QD during the same laser pulse. This leads to the emission of more than one photon per pulse, thus diminishing the SPP. Although a SPP can always be calculated, one should be aware that it constitutes a physically meaningful quantity only for finite brightness. Therefore, the area of increased SPP in the upper right corner of Fig. 2(c) is of no physical relevance.

It is intuitively expected that the continuum of LA phonons reduces the quantum correlations of the system and thus the SPP by inducing a manifold of transitions between its quantum states. However, contrary to these expectations Fig. 2(d) reveals a huge systematic increase in $\mathcal{P}$ at $\Delta\omega_{\mathrm{LX}} \gtrsim 0.5$ meV. Moreover, the maximum $\mathcal{P}_{\max} = 98.8\%$ (red circle) is even larger than 90.7% obtained for the resonantly driven system (blue circle). Combined with an appreciably large $\mathcal{B}$, this indicates a possibility to have a good quality single-photon source in the off-resonant excitation regime. Note that $\mathcal{B} = 0.46$ observed at the point of $\mathcal{P}_{\max}$ [cf., red circle in Fig. 2(b)] is not much smaller than the maximal value of 0.67 achieved in the resonantly driven case [cf., blue circle in Fig. 2(b)]. It is also noteworthy that it is possible to obtain a significantly larger brightness at the cost of a slight decrease in the SSP. For example, if we choose a trade-off by maximizing the sum of the squares of the two figures of merit in the off-resonant regime, we obtain $\mathcal{B} = 0.53$ and $\mathcal{P} = 98.1\%$ (red square). This value for $\mathcal{P}$ is close to typical experimental values obtained for resonant excitation of the quantum dot exciton (98.8% [4], 99.1% [7]) even though the pulse lengths in Refs. [4,7] have been slightly shorter [40].

To explain the mechanism behind this observation, one needs to consider the dynamics of the QD-cavity states. In Fig. 3, the time dependent occupations in the resonant and the off-resonant case (cf., the blue and red circles in Fig. 2, respectively) are compared. The considered states are product states between the QD states and a photon state with photon number n. After resonant π-pulse excitation [cf., Fig. 3(a)], the exciton state $|X, 0\rangle$ without photons is occupied (blue curve). The cavity coupling rotates the dot back to its ground state and produces one photon in the cavity (orange curve). Because the driving is still nonzero at this point, the dot is reexcited to produce an occupation of the state $|X, 1\rangle$ (green curve), which is shown in the inset of Fig. 3(a). Finally, the cavity coupling leads to an occupation of the ground state with two photons $|G, 2\rangle$ (red curve), such that the SPP is diminished.

In contrast to the π-pulse induced rotation of the Bloch vector, the off-resonant excitation scheme exploits an effect called adiabatic undressing [27]. Switching on the laser transforms the dot states to a new energy eigenbasis commonly known as laser-dressed states, the gap between which can be bridged by LA phonons with typical energies of a few meV. At low temperatures, the lower dressed state becomes occupied via phonon emission. However, the phonon-induced relaxation is only efficient when both dressed states have roughly equal exciton components. Thus, the exciton state exhibits typically occupations of the order of 50% after the relaxation is completed [27]. When the laser is turned off adiabatically, the lower dressed state is subsequently transformed to the exciton state in the original basis provided the detuning is positive (otherwise the ground state is reached [27]). This adiabatic undressing of the dot states therefore boosts the exciton occupation only at the end of the pulse [cf., the blue curve in Fig. 3(b)]. This in turn means that during the phase of phonon relaxation no photon can be put into the cavity efficiently [cf., the orange curve in Fig. 3(b)]. When finally the adiabatic undressing-induced exciton boost occurs, the occupation of $|G, 1\rangle$ follows [cf., Fig. 3(b)]. Since the excitation pulse is basically gone by then, the reexcitation of the QD is strongly

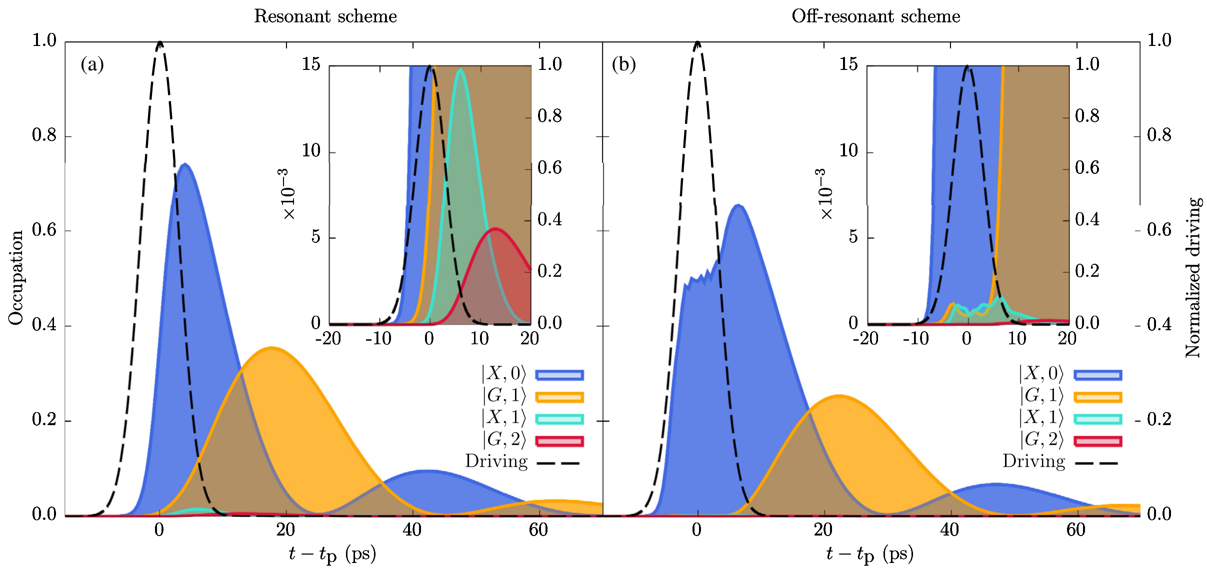

FIG. 3. Time-dependent occupations: (a) after resonant π-pulse excitation (cf., blue circle in Fig. 2) and (b) in the off-resonant phonon-assisted case (cf., red circle in Fig. 2). The occupations of the states $|X,0\rangle$, $|G,1\rangle$, $|X,1\rangle$, and $|G,2\rangle$ are shown as colored filled curves. The Gaussian envelope of the laser driving pulse normalized to its maximum value centered at t_p is shown as a black dashed line. The insets show the same curves, respectively, on an enlarged scale for the occupations.

suppressed (green curve), such that effectively no second photon can be put into the cavity (red curve). This implies a far higher SPP than in the resonant counterpart, as is observed in Fig. 2(d). In summary, the delay of the exciton occupation caused by the two-step procedure of first relaxing to a dressed state via phonon emission and then reaching the exciton by adiabatic undressing is responsible for the enhancement of the SPP.

To quantify the robustness of the phonon-induced SPP enhancement against variations of other system parameters,

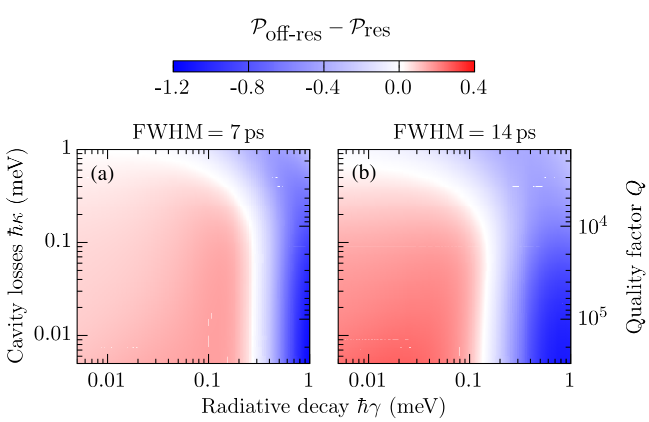

FIG. 4. The difference between the SPP after off-resonant phonon-assisted excitation $\mathcal{P}_{\text{off-res}}$ and after resonant π-pulse rotation $\mathcal{P}_{\text{res}}$ is shown for two different pulse lengths (FWHM), namely: (a) 7 ps and (b) 14 ps, as a function of radiative decay $\hbar\gamma$ and cavity losses $\hbar\kappa$. The cavity quality factor $Q = \omega_c/\kappa$ is obtained via the cavity losses assuming a cavity single-mode energy of $\hbar\omega_c = 1.5$ eV. The pulse area is set to 12.75π and $\Delta\omega_{LX} = 1.1$ meV.

the difference between the SPP after off-resonant excitation and after the resonant one is shown as a function of the radiative decay γ and the cavity loss rate κ in Fig. 4. A positive value (reddish shade) indicates a set of parameters where the SPP is enhanced for off-resonant excitation. We find such an enhancement for a wide parameter regime in κ and γ that is experimentally well accessible. Also, changing the pulse length from 7 ps in Fig. 4(a) to 14 ps in Fig. 4(b) does not change the phonon-induced SPP enhancement qualitatively. The reason why the SPP for off-resonant excitation falls below the resonant one in the bad cavity limit and/or in the limit of high radiative losses is that relaxation processes limit the time available for the adiabatic undressing which eventually becomes incomplete.

In conclusion, we have presented a seemingly paradoxical scheme for the phonon-assisted operation of a QD-cavity system as a single-photon source, where the excitation is spectrally separated from the generated photons. Two factors that would separately lead to a quality degradation—off-resonant driving and dot-phonon coupling—in combination result in a huge boost in critical characteristics of a single-photon source. We have demonstrated that the achievable single-photon purity can be noticeably higher than for resonant excitation while the brightness is still at an acceptable level. The physical mechanism of this enhancement—the adiabatic undressing—is realized in a wide interval of physically accessible parameters.

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