Phonon-induced enhancement of photon entanglement in quantum dot-cavity systems

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We report on simulations of the degree of polarization entanglement of photon pairs simultaneously emitted from a quantum dot-cavity system that demand revisiting the role of phonons. Since coherence is a fundamental precondition for entanglement and phonons are known to be a major source of decoherence, it seems unavoidable that phonons can only degrade entanglement. In contrast, we demonstrate that phonons can cause a degree of entanglement that even surpasses the corresponding value for the phonon-free case. In particular, we consider the special situation of vanishing biexciton binding energy and either finite exciton or cavity mode splitting. In both cases, combinations of the splitting and the dot-cavity coupling strength are found where the entanglement exhibits a non-monotonic temperature dependence which enables entanglement above the phonon-free level in a finite parameter range. This unusual behavior can be explained by phonon-induced renormalizations of the dot-cavity coupling $g$ in combination with a non-monotonic dependence of the entanglement on $g$ that is present already without phonons.

The appearance of entangled states is one of the showcase effects that highlights most impressively the dramatic conceptual changes brought forth by going over from classical to quantum physics [1, 2]. Moreover, realizations of entangled states, mostly with photons, have paved the way toward many innovative applications [3], e.g., in quantum cryptography [4, 5], quantum teleportation [6], quantum information processing [7–10] and photonics [11]. In particular, quantum dot (QD) cavity systems have attracted a lot of attention as sources for triggered entangled photon pairs [12–19], not only because these systems hold the promise of a natural integration in solid state devices. Embedding a QD in a micro-cavity enables the manipulation of few-electron and few-photon states in a system with high optical non-linearities, which can be used for realizing a few-photon logic in quantum optical networks [20]. Furthermore, the cavity boosts the quantum yield due to the Purcell effect [14, 21] and for high Q-factors it reduces the detrimental effects of phonons on the photon indistinguishability [22].

The essence of entanglement in a bipartite system is the creation of a state that cannot be factorized into parts referring to the constituent subsystems, which requires the build-up of a superposition state. Polarization entanglement between horizontally ($H$) or vertically ($V$) polarized photon pairs is established, e.g., by creating superpositions of the states $|HH\rangle$ and $|VV\rangle$ with two photons with either $H$ or $V$ polarizations. Obviously, maintaining such a superposition requires stable relative phases between these states. However, in a solid-state system the interaction with the environment unavoidably leads to a loss of phase coherence. In particular, phonons are known to provide a major source of decoherence [23–32] which led to the expectation that phonons should always degrade the entanglement. Indeed, recent simulations [33–35] are in line with this expectation.

In this letter we demonstrate that the destructive effect of phonons on the photon entanglement resulting from phonon-induced decoherence can be overcompensated when phonon-related renormalizations of the QD-cavity coupling shift the system into a regime of higher photon entanglement. A precondition of this mechanism is a decrease of the degree of entanglement with rising QD-cavity coupling $g$ in the phonon-free case in a finite $g$ range. This is realized, e.g., for vanishing biexciton binding energy and finite exciton or cavity mode splitting.

For our studies we adopt a model that comprises the following Hamiltonian [34, 35]:

$$
\hat{H} = \hbar \omega_H |X_H\rangle\langle X_H| + \hbar \omega_V |X_V\rangle\langle X_V| + \hbar (\omega_H + \omega_V - \omega_B)|B\rangle\langle B| + \sum_{\ell=H,V} \hbar \omega_\ell \hat{a}^\dagger_\ell \hat{a}_\ell 
+ \sum_q \hbar \omega_q \hat{b}^\dagger_q \hat{b}_q + \sum_{q,x} n_x (\gamma_q \hat{b}^\dagger_q + \gamma_q \hat{b}_q) |\chi\rangle\langle \chi| + \hat{X},
$$

(1)

where $|B\rangle$ is the biexciton state with energy $\hbar (\omega_H + \omega_V - \omega_B)$ and a biexciton binding energy $E_B = \hbar \omega_B$ while $|X_{H/V}\rangle$ denote the two exciton states with energies $\hbar \omega_{H/V}$ that couple to $H$ or $V$ polarized cavity modes with destruction (creation) operators $\hat{a}_{H/V}$ ($\hat{a}^\dagger_{H/V}$) and mode energies $\hbar \omega_{H/V}$. $\hat{b}^\dagger_q$ ($\hat{b}_q$) are operators that destroy (create) longitudinal acoustic phonons with wave-vector $q$ and energy $\hbar \omega_q$. We consider bulk phonons with a linear dispersion and account for the deformation potential coupling $\gamma_q$. $n_x$ is the number of electron-hole pairs contained in the states $|\chi\rangle \in \{|B\rangle, |X_{H/V}\rangle\}$. Finally, the Jaynes-Cummings type coupling of the cavity modes to the QD with coupling constant $g$ is given by:

$$
\hat{X} = -g \left( \langle G|X_H|\hat{a}^\dagger_H + |X_H\rangle\langle B|\hat{a}^\dagger_V 
+ \langle G|X_V|\hat{a}^\dagger_V - |X_V\rangle\langle B|\hat{a}_V \right) + H.c.,
$$

(2)
FIG. 1. (a) Sketch of the level scheme of a QD-cavity system with finite fine-structure splitting, zero biexciton binding energy and two-photon resonant cavity modes. (b) Concurrence as a function of the temperature for three selected values of the QD-cavity coupling. The corresponding values obtained without phonons are drawn as straight (faded) lines with the same linetype. (c) Concurrence as a function of the QD-cavity coupling for three temperatures together with the phonon-free result. In addition \( C(\tilde{g}(g)) \) is plotted using the phonon-renormalized coupling \( \tilde{g}(g) \) for \( T=30 \) K [indicated on the upper axis], where \( C(g) \) is the phonon-free concurrence. The values of the QD-cavity coupling used in (b) are marked in (c) by horizontal lines.

Parameters: \( \delta = 0.1 \) meV, \( \kappa = 0.025 \) ps \(^{-1} \), electron (hole) confinement length \( a_e = 3 \) nm, \( a_h = a_e / 1.15 \) where we assume a spherical GaAs-type QD with harmonic confinement. All other parameters, e.g., concerning the phonon coupling are taken from Ref. [36].

where \( H.c. \) stands for the Hermitian conjugate and \( |G\rangle \) is the QD ground state, the energy of which is taken as the zero of energy. In addition, we account for cavity losses with a rate \( \kappa \) by the Lindblad operator:

\[
\mathcal{L}_{\text{cav}} [\hat{\rho}] = \sum_{\ell=H,V} \frac{\kappa}{2} \left( 2 \hat{a}_\ell \hat{a}_\ell^\dagger - \hat{\rho} \hat{a}_\ell^\dagger \hat{a}_\ell - \hat{a}_\ell^\dagger \hat{a}_\ell \hat{\rho} \right).
\]

We assume that the system is initially prepared in the biexciton state without photons and that the phonons are initially in equilibrium at a temperature \( T \). The dynamics of the reduced density matrix \( \hat{\rho} \) is determined by the equation:

\[
\frac{d}{dt} \hat{\rho} = \frac{i}{\hbar} \left[ \hat{H}, \hat{\rho} \right] + \mathcal{L}_{\text{cav}} [\hat{\rho}],
\]

where \( [\cdot, \cdot]_- \) denotes the commutator. As in Ref. 35 we evaluate \( \hat{\rho} \) numerically in the subspace spanned by the five states \( |B,0,0\rangle, |X_H,1,0\rangle, |X_V,0,1\rangle, |G,2,0\rangle \) and \( |G,0,2\rangle \), where the numbers \( n_H/V \) in \( |\chi, n_H, n_V\rangle \) denote the number of \( H/V \) photons. We use a path-integral approach which does not introduce approximations to the model. This is made possible by recent methodological advances that allow for a natural inclusion of non-Hamiltonian parts of the dynamics (e.g. represented by Lindblad operators) in the path-integral formalism [37] as well as huge improvements of the performance by iterating a partially summed augmented density matrix [38].

We quantify the degree of entanglement by the concurrence of simultaneously emitted photon pairs that can be calculated directly from the reduced density matrix (explicit expressions may be found in Ref. 35). We focus on simultaneously emitted photon pairs since experiments [39, 40] agree with theory [15, 35] that this case is favorable for the entanglement.

First, we present results for the situation sketched in Fig. 1 (a) where the excitons have a finite fine-structure splitting \( \delta = \hbar (\omega_H - \omega_V) \), the biexciton binding energy is zero and both cavity modes are tuned to the two-photon resonance \( 2\omega_H = 2\omega_V = \omega_H + \omega_V - \omega_B \).

Fig. 1 (b) displays the temperature dependence of the concurrence for three values of the QD-cavity coupling. Only the result for \( g = 130 \) \( \mu \)eV agrees with the common expectation that the entanglement should monotonically decrease with temperature. In contrast, for \( g = 60 \) \( \mu \)eV and \( g = 35 \) \( \mu \)eV unusual non-monotonic \( T \)-dependences are found. Most interestingly, for \( g = 35 \) \( \mu \)eV the concurrence is noticeably higher than the corresponding value obtained without photons in the entire tempera-
FIG. 2. (a) Sketch of the level scheme of a QD-cavity system with finite fine-structure splitting, biexciton binding energy $E_B = 1$ meV and two-photon resonant cavity modes. (b) Concurrence as a function of the temperature for three selected values of the QD-cavity coupling. The corresponding values obtained without phonons are drawn as straight (faded) lines with the same linetype. (c) Concurrence as a function of the QD-cavity coupling for three temperatures together with the phonon-free result. The values of the QD-cavity coupling used in (b) are marked in (c) by horizontal lines. Apart from $E_B$ the same parameters are used as in Fig. 1.

The reason for this remarkable behavior becomes apparent when looking at the $g$ dependence of the concurrence in Fig. 1 (c). First, we note that already without phonons the concurrence is a non-monotonic function of $g$ (purple curve) with a pronounced minimum reached roughly for $g \approx \delta/2$. By dividing Eq. (4) by the coupling strength $g$ and leaving out the coupling to the phonons, the dynamics of the system is described by the rescaled quantities $t' = gt$, $g' = g/g = 1$, $\delta' = \delta/g$ and $\kappa' = \kappa/g$. Since the concurrence is the asymptotic value of the normalized coherence at long averaging times [35] the rescaling of the time is irrelevant. For a large value of $g$ both parameters $\delta'$ and $\kappa'$ tend to zero. This implies that the concurrence approaches one for large coupling strengths because the which-path information disappears for a vanishing splitting and thus the concurrence is one [35, 41]. For very small values of the QD-cavity coupling, $\kappa'$ and $\delta'$ become arbitrarily large. Therefore, the sequential single photon decay via the intermediate exciton states becomes strongly off-resonant and is thus negligible compared with contributions from a direct two-photon transition which is always resonant in the present case. Since the which-path information is contained only in the sequential decay the concurrence approaches again one. But for finite splittings the concurrence is lower than one and thus a minimum must appear at a certain coupling strength $g$.

When phonons are accounted for the minimum is lowered and shifted to a higher coupling strength depending on the temperature. We attribute the shift to the well known effect of phonon-induced renormalization of the light-matter coupling [42]. To support this assignment we have estimated the renormalized coupling $\tilde{g}(g)$ as in Ref. 43 by fitting equations with phenomenological renormalizations of a resonantly driven two-level system to path-integral calculations. The results are shown in the supplement [44]. If the only effect introduced by phonons would be the $g$ renormalization then the value of the concurrence found without phonons at a particular value of $g$ should be shifted by phonons to $\tilde{g}(g)$. Indeed, in Fig. 1 (c) we have plotted $C(\tilde{g}(g))$ using the phonon-renormalized coupling $\tilde{g}(g)$ for $T = 30$ K, where $C(g)$ is the concurrence in the phonon-free case [green curve with circles]. We find that, despite the crudeness of the estimation for $\tilde{g}(g)$, the minimum of the shifted curve agrees even quantitatively well with the minimum found in the full path-integral simulation for this temperature [red dotted curve]. Since the shift is larger for higher temperatures, displacing the phonon-free curve necessar-
ily leads to higher values of the shifted curves in regions where the phonon-free concurrence is monotonically decreasing with $g$.

The total effect of phonons is, however, not merely a shift but also a lowering of the curves with rising temperature which is indeed due to the dephasing action of phonons. It is important for obtaining a phonon-induced entanglement that the gain in entanglement resulting from the shift of the phonon-free curve due the phonon-induced $g$ renormalization is not destroyed by the overall lowering of the concurrence caused by the decoherence. Fig. 1 (c) demonstrates that it is indeed possible that the renormalization-induced shift overcompensates the dephasing action.

It is instructive to contrast the above findings with simulations for the more commonly considered situation sketched in Fig. 2 (a), where the biexciton binding energy has the finite value $E_B = 1$ meV and the cavity modes are in resonance with the two-photon transition to the biexciton. Again, the phonon-free curve exhibits a minimum which is, however, rather flat [purple line in Fig. 2 (c)]. In the limit $g \to \infty$ the concurrence approaches one since the argument given for the case of vanishing biexciton binding energy applies here as well. For the case that both $g/(\frac{1}{2} E_B)$ and $\delta/E_B$ are small parameters it has been shown analytically in Ref. 35 that the phonon-free concurrence approach $E_B^2 - \delta^2/E_B^2$, for which finite $\delta$ is smaller than one. Including phonons, the reduction of the concurrence for small $g$ values is strongly magnified as seen in Fig. 2 (c). Overall, the dephasing action induced by phonons is so strong that the lineshape of the concurrence as a function of $g$ is significantly deformed and effects related to a renormalization of $g$ cannot be identified. As a consequence the concurrence monotonically decreases with rising temperature and always stays below the phonon-free calculation for all values of $g$ as exemplarily shown in Fig. 2 (b). This demonstrates that the phonon-induced enhancement of entanglement described above can only occur when the $g$-renormalization effects dominate over the phonon-induced dephasing.

We note in passing that the situation considered in Fig. 1 is not the only one where the conditions for phonon-induced entanglement are realized. Another configuration where this phenomenon can be observed is provided by a system with vanishing $E_B$ and degenerate excitons where which-path information is introduced by a finite splitting of the cavity modes (corresponding results are shown in the supplement [44]). There, the concurrence calculated without phonons is again a non-monotonic function of $g$, which exhibits even more than one extremum. Also in this case, the phonon-induced renormalization is strong enough to evoke a phonon-induced entanglement.

In conclusion, we have demonstrated that phonon-induced renormalizations of the cavity-dot coupling can overcompensate decoherence effects and shift the system to a region of higher entanglement. In combination with a non-monotonic dependence of the phonon-free concurrence this may result in a non-monotonic temperature dependence of the concurrence. Most interestingly, the concurrence can even reach values above the phonon-free level, manifesting a phonon-induced photon entanglement.

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[40] See Supplemental Material at [URL will be inserted by publisher] for details how the renormalization of the coupling $g$ is estimated and for an additional configuration exhibiting phonon-induced entanglement.