Phonon-Induced Dephasing in Quantum Dot-Cavity QED

A. Morreau and E. A. Muljarov
School of Physics and Astronomy, Cardiff University, Cardiff CF24 3AA, United Kingdom
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We present a semi-analytic exact solution to the problem of phonon decoherence in a quantum dot embedded in an optical microcavity. Our approach is based on the Trotter decomposition and takes into account the effects of the exciton-cavity and exciton-phonon coupling on equal footing, thereby providing access to regimes of comparable polaron and polariton timescales. We show that in these regimes, the optical decoherence is determined by acoustic phonon-induced transitions between different polariton states of the quantum dot-cavity system. We also provide purely analytic approximations which accurately describe the limit of longer polariton timescales.

A quantum dot (QD) embedded in a solid-state optical microcavity presents a fundamental system within cavity quantum electrodynamics (cavity-QED) [1]. The QD exciton couples to an optical mode of the cavity in a manner well described by the exactly solvable Jaynes-Cummings (JC) model [2–4]. Within the strong coupling regime well described by the exactly solvable Jaynes-Cummings (JC) model [2–4], there is a partly reversible exchange of energy, with a (JC) model [2–4]. Within the strong coupling regime there is a partly reversible exchange of energy, with a period τJC, between the exciton and the cavity mode, which gives rise to polaron formation and characteristic vacuum Rabi splitting [3–4].

Whilst not accounted for in the JC model, there is significant experimental and theoretical evidence [5–11] to suggest that phonons play a crucial role in the optical decoherence of the QD-cavity system. The general phenomenon of phonon-induced dephasing in semiconductor QDs is well studied; it has been successfully explained and quantified by the exactly solvable independent boson (IB) model [21]. This model describes a polaron, formed from a QD exciton coupled to bulk acoustic phonons [22], with a characteristic polaron formation time τIB. The IB model accounts for the major effect of the non-Markovian pure dephasing but is known to fail treating the exciton zero-phonon line (ZPL) broadening [23].

It is natural to draw upon the JC and IB models when addressing the problem of phonon-induced dephasing in the QD-cavity system. However, the combination of the two models presents a significant challenge. Various approaches to the QD-cavity problem have been suggested in the literature, ranging from Born-Markov approximations [5–9] to path-integral methods [12–14] and non-equilibrium Greens function techniques [12–14]. These approaches can be broadly divided into perturbative and non-perturbative methods.

The perturbative methods employ a polaron transformation followed by a perturbative treatment of the coupling of the phonon-dressed exciton to the cavity mode, carried out in the 2nd order Born approximation [5–9] or beyond [12–15]. These approaches are usually valid for (i) exciton-phonon interactions much stronger than exciton-cavity coupling, and (ii) polaron formation time much quicker than the polariton dynamics, τIB ≪ τJC.

Non-perturbative techniques based on a quasi-adiabatic Feynman path-integral scheme [24] enable accurate numerical solutions but are computationally expensive and provide little insight into the underlying physics. Nahri et al. [14] apply a tensor multiplication scheme [24] to the case of a QD-cavity system with superohmic spectral density. This technique requires a complex algorithm with an “on-the-fly path selection” optimization [23]. Glassl et al. [12] present a real-time path-integral scheme [24] adapted for a QD in a lossless cavity. Cavity and QD dampings are included in later work [27], but in this case the exciton-phonon coupling is added phenomenologically.

In this Letter, we present a semi-analytic exact solution of the long-standing problem of the phonon-induced decoherence of the QD-cavity system. Our approach is based on the Trotter decomposition with a subsequent use of the cumulant expansion technique [21, 23, 28], which provides a computationally straightforward and physically intuitive formulation. Being non-perturbative, our approach treats the effects of the exciton-photon and exciton-phonon couplings on equal footing, thereby rendering the technique appropriate across the full range of both coupling strengths, as well as timescales τIB and τJC. Furthermore, the solutions we present become fully analytical in the limit τIB ≪ τJC.

A key principle of the present method is a separation of the system Hamiltonian into two exactly solvable parts, \(H = H_{JC} + H_{IB}\), described by the JC and IB models respectively. The JC Hamiltonian has the form (\(\hbar = 1\)):

\[H_{JC} = \omega_X d\dagger d + \omega_C a\dagger a + g(a\dagger d + d\dagger a),\]  

where \(d\dagger (a\dagger)\) is the exciton (cavity photon) creation operator, \(g\) is the exciton-cavity coupling strength, and \(\omega_X (\omega_C)\) is the exciton (cavity photon) complex frequency, including the long-time ZPL exciton dephasing (radiative decay) rate \(\gamma_X (\gamma_C)\). Without the ZPL, which is already included in Eq. (1), the IB Hamiltonian \(H_{IB}\) is \(H_{ph} + d\dagger dV\) consists of the free phonon bath Hamiltonian \(H_{ph}\) and the exciton-phonon interaction \(V\),

\[H_{ph} = \sum_q \omega_q b\dagger_q b_q, \quad V = \sum_q \lambda_q (b_q + b\dagger_{-q}),\]  

where \(b\dagger_q (\omega_q)\) is the creation operator (frequency) of the
where \( \Delta \) and \( \hat{a} \) denote a pulsed exciton (cavity) excitation.

The polarization is then given by a 2 \times 2 matrix \( \hat{P}(t) \) with the matrix elements \( P_{jk}(t) \) expressed in terms of the time evolution operator \( \hat{U}(t) \)

\[
P_{jk}(t) = \langle \langle j | \hat{U}(t) | k \rangle \rangle_{ph}, \quad \hat{U}(t) = e^{i H_{ph} t} e^{-i \hat{H} t},
\]

where \( \langle \langle . \rangle \rangle_{ph} \) denotes the expectation value over all phonon degrees of freedom in thermal equilibrium and \( j, k = X, C, \) see [29] for details. Here, \( j \) indicates the initial exciton mode of the system and \( k \) the mode in which the polarization is measured. For example, \( P_{XX} \) (PCC) denotes the excitonic (photonic) polarization under a pulsed exciton (cavity) excitation.

Using Trotter’s decomposition theorem, the time evolution operator \( \hat{U}(t) \) can be re-expressed as

\[
\hat{U}(t) = \lim_{\Delta t \to 0} e^{i H_{ph} \Delta t} (e^{-i H t} e^{-i H_{JC} \Delta t})^N,
\]

where \( \Delta t = t/N \). We introduce two new operators, \( \hat{M} \) and \( \hat{W} \), associated with the JC and IB Hamiltonians, respectively,

\[
\hat{M}(t_n - t_{n-1}) = \hat{M}(\Delta t) = e^{-i H_{JC} \Delta t},
\]

\[
\hat{W}(t_n - t_{n-1}) = e^{i H_{ph} t_n} e^{-i H_{IB} \Delta t} e^{-i H_{ph} t_{n-1}},
\]

where \( t_n = n \Delta t \). Exploiting the commutativity of \( H_{JC} \) and \( H_{ph} \) enables us to express the time evolution operator as a time-ordered product of pairs \( \hat{M} \hat{W} \):

\[
\hat{U}(t) = \mathcal{T} \prod_{n=1}^{N} \hat{W}(t_n, t_{n-1}) \hat{M}(t_n - t_{n-1}),
\]

where \( \mathcal{T} \) is the time ordering operator. Noting that both \( \hat{W} \) and \( \hat{M} \) are 2 \times 2 matrices in the \( |X, C \rangle \) basis and that \( \hat{W} \) is diagonal with diagonal elements \( W_j \), the polarization Eq. 3 takes the form

\[
P_{jk}(t) = \sum_{i_{n-1}=X,C} \sum_{i_1=X,C} \ldots \sum_{i_{n}X,C} M_{i_{n}i_{n-1}} \ldots M_{i_2i_1} M_{i_1i_0} \times \langle W_{i_n}(t, t_{n-1}) \ldots W_{i_2}(t_2, t_1) W_{i_1}(t_1, 0) \rangle_{ph},
\]

where \( i_n = j, i_0 = k, M_{i_ni_m} = [\hat{M}(\Delta t)]_{i_ni_m} \), and

\[
W_{i_n}(t_n, t_{n-1}) = \mathcal{T} \exp \left\{ -i \delta_{i_nX} \int_{t_{n-1}}^{t_n} V(\tau) d\tau \right\}
\]

with \( \delta \) the Kronecker delta and \( V(\tau) = e^{i H_{ph} \tau} V e^{-i H_{ph} \tau} \), see [29] for details.

It is instructive at this point to introduce the concept of a “realization” of the system as a particular combination of indices \( i_n \) within the full summation of Eq. (13). We associate with each realization a step-function \( \hat{\theta}(\tau) \) being equal to 0 (1) over the time interval \( t_n - t_{n-1} \) if \( i_n = C \) (\( i_n = X \)) [the system is in state \( |C \rangle \) (\( |X \rangle \)]. An example realization is given in [29]. The product of \( W \)-operators for a particular realization can be written as

\[
W_{i_0}(t, t_{N-1}) \ldots W_{i_n}(t_n, 0) = \mathcal{T} \exp \left\{ -i \int_0^t \hat{V}(\tau) d\tau \right\},
\]

where \( \hat{V}(\tau) = \hat{\theta}(\tau) V(\tau) \). Now, applying the linked cluster theorem [21] for calculating the trace of Eq. (10) over all phonon states, we obtain

\[
\langle W_{i_0}(t, t_{N-1}) \ldots W_{i_n}(t_n, 0) \rangle_{ph} = e^{\hat{K}(t)},
\]

where

\[
\hat{K}(t) = -\frac{1}{2} \int_0^t d\tau_1 \int_{t_{n-1}}^t d\tau_2 \langle \mathcal{V} \hat{V}(\tau_1) \hat{V}(\tau_2) \rangle
\]

is the linear cumulant for the particular realization. Its explicit dependence on the specific indices \( i_n \) of the realization is given by

\[
K_{[n-m]} = \sum_{n=1}^{N} \sum_{m=1}^{N} \delta_{i_nX} \delta_{i_mX} K_{[n-m]},
\]

where

\[
K_{[n-m]} = -\frac{1}{2} \int_{t_{n-1}}^{t_n} d\tau_1 \int_{t_{n-1}}^{t_n} d\tau_2 \langle \mathcal{V} \hat{V}(\tau_1) \hat{V}(\tau_2) \rangle.
\]

Note that \( K_{[n-m]} \) depends only on the time difference \( |t_n - t_m| \). Furthermore, all \( K_{[n-m]} \) can be efficiently calculated from the standard IB model cumulant \( K(t) = \mathcal{T} \exp \left\{ -i \int_0^t V(\tau) d\tau \right\} \), as shown in [29].

Having in mind an application of this theory to semiconductor QDs coupled to bulk acoustic phonons, we use the conditions of the super-Ohmic coupling spectral density and a finite phonon memory time [29]. This allows us to reduce dramatically the number of terms in the double summation of Eq. (13). Indeed, we need to take into account only instances in which \( |t_m - t_n| \leq \tau_{IB} \) or equivalently, \( |n - m| \leq \tau_{IB}/\Delta t \). When selecting \( \Delta t \), we must also be mindful of the requirement imposed by the Trotter decomposition method: \( \Delta t \to 0 \), which is carefully addressed below. We initially consider the most straightforward application of the technique, which will be referred to as the nearest neighbors (NN) approach.

In the NN approach, we limit our consideration to \( |n - m| \leq 1 \), selecting \( \Delta t \approx \tau_{IB} \) so as to best satisfy both
Crucially, this reduction to a single summation allows us to re-express Eq. (8) as

\[ P_{jk}(t) = e^{\delta_i x K_0} \sum_{i_{N-1}} \ldots \sum_{i_1} G_{i_{N-1}i_{N-2} \ldots i_1} M_{i_{N-1}j} \ , \]

where

\[ G_{i_{n-1}i_{n-1}} = M_{i_{n-1}i_{n-1}}, e^{\delta_i x (K_0 + 2\delta_{i_{n-1}x}K_1)} \ . \]

Equation (10) can be compactly written in a 2x2 matrix form in the \|X\|, \|C\| basis:

\[ \dot{\hat{P}}(t) = \begin{pmatrix} P_{XX} & P_{XC} \\ P_{CX} & P_{CC} \end{pmatrix} = \begin{pmatrix} e^{K_0} & 0 \\ 0 & 1 \end{pmatrix} \hat{G}^{N-1} \tilde{M} \]

(18)

with \( \hat{G} \) given by

\[ \hat{G} = \begin{pmatrix} M_{XX}e^{K_0+2K_1} & M_{XC} \\ M_{CX}e^{K_0} & M_{CC} \end{pmatrix} \ . \]

(19)

It should be noted that our time step \( \Delta t \approx \tau_B \) is too large to capture the initial rapid phonon-induced decay of the polarization associated with the phonon broadband. There is, however, a simple solution to this problem: For all \( t < \tau_B \), we allow \( \Delta t \) to vary as \( \Delta t = t/2 \) (corresponding to \( N = 2 \)), as in the NN approach this does not neglect any part of \( K \).

From the NN result Eq. (18), one can extract a simple analytic expression that describes the long-time behavior of the linear optical response. We use the asymptotic behavior of the standard IB model cumulant \( K(t) \) in the long-time regime:

\[ \dot{\hat{P}}(t) \approx e^{-S/2} e^{-i\tilde{H}t} e^{-S/2} \quad (t > \tau_B) \ , \]

where \( \tilde{H} = \begin{pmatrix} \omega_X + \Omega_p & ge^{-S/2} \\ ge^{-S/2} & \omega_C \end{pmatrix} \) and \( \tilde{S} = \begin{pmatrix} S & 0 \\ 0 & 0 \end{pmatrix} \).

(21)

Comparing the long-time analytics for \( P_{jk}(t) \), given by Eqs. (21) and (22), with the exact linear polarization in the JC model (no phonons), \( \langle j | e^{-iH_{JC}t} | k \rangle \), we see that the effect of acoustic phonons in this limit \( (\tau_B \ll \tau_C) \) is a reduction of the exciton-cavity coupling strength \( g \) by a factor of \( e^{S/2} \) and the ZPL weight of the excitonic polarization by a factor of \( e^{S} \). Additionally the bare exciton frequency is polaron-shifted: \( \omega_X \rightarrow \omega_X + \Omega_p \). These facts are consistent with the analytic results of the IB model and in agreement with previous experimental and theoretical works. Also, the form of the modified Hamiltonian \( \hat{H} \) given by Eq. (22) is exactly the same as obtained after making the polaron transformation of the full Hamiltonian \( \hat{H} \) and then neglecting any interactions which appear in the polaron frame.

We now address a general case in which the polaron and polarization time scales can be comparable, \( \tau_B \sim \tau_C \), for example, in the case of a much larger exciton-cavity coupling \( g \). This implies that we must find a way to reduce the time-step \( \Delta t \) in the Trotter decomposition. We achieve this by going beyond the NN regime to the \( L \)-neighbor (LN) regime, where \( L \) indicates the number of “neighbors” that we consider, corresponding to the condition \( |n - m| \ll L \) in Eq. (13). In the LN regime, we are bound by a constraint \( L \Delta t \lesssim \tau_B \), which allows us to make \( \Delta t \) arbitrarily small through an increase in \( L \).

In the LN approach we define a quantity \( F_{L_1 \ldots i_l}^{(n)} \) which is generated via a recursive relation:

\[ F_{L_1 \ldots i_l}^{(n+1)} = \sum_{l' = X,C} G_{i_{L_1} \ldots i_{l-1} l'} F_{L_1 \ldots i_{l-1} l'}^{(n)} \]

(23)

using \( F_{L_1 \ldots i_1}^{(1)} = M_{i_{L_1}i_1} \) as the initial value, where \( \tilde{M} \) is defined as before by Eq. (5), while \( G_{i_{L_1} \ldots i_l} \) is the LN analog of Eq. (17):

\[ G_{i_{L_1} \ldots i_l} = M_{i_{L_1}i_1} e^{\delta_i x (K_0 + 2\delta_{i_{L_1}x}K_1 + 2\delta_{i_lx}K_l)} \ . \]

(24)

The polarization is then given by

\[ P_{jk}(t) = e^{\delta_i x K_0} F_{C_1 \ldots C_j}^{(n)} \]

(25)

Equations (23) – (25) present an asymptotically exact solution for the linear polarization. By extending the matrix size of the operators involved, it is straightforward to generalize this result to other correlators, such as the photon indistinguishability or to other elements of the density matrix, such as the four-wave mixing polarization.

Figure I shows the exact linear excitonic polarization \( |P_{XX}(t)\rangle \) and absorption \( \text{Re} P_{XX}(\omega) \) \( |P_{XX}(\omega)\rangle \) is the Fourier transform of \( P_{XX}(t) \), alongside the NN and analytic results, Eqs. (18) and (21), respectively. In this case of a rather small QD-cavity coupling \( (g = 50 \mu eV) \), the absorption spectrum consists of a well-resolved polaron doublet, described by the eigenvalues \( \omega_j = \Omega_j - i\Gamma_j \) \((j = 1, 2)\) of the effective Hamiltonian Eq. (22), superimposed with a phonon broadband (BB). At lower temperatures, the BB is more asymmetric and the ZPL weight is increased, in agreement with the IB model.
parameters selected, $\tau_B \approx 3.2$ ps (at $T = 50$ K) and $\tau_{JC} \approx 50$ ps, so that the NN approach presents a good approximation in this regime. As expected, the analytic calculation Eq. (21) describes the long-time dynamics well but fails at short times, as it is clear from Fig. 1(a). This is manifested in the absorption spectrum in Fig. 1(b) as an absence of the BB. To improve on this shortcoming, we have developed a refined, fully analytical solution which captures the BB and reproduces the whole spectrum to very good accuracy, see the green dotted line in Fig. 1(b) and Sec. IX in [29] for details of the model.

In regimes of comparable polaron and polariton times $\tau_B \sim \tau_{JC}$ [achieved by increasing the QD-cavity coupling constant to $g = 0.6$ meV while keeping all other parameters the same], the NN approach and the analytic approximations all fail, so that we have only the LN results. For large values of $g$, the long-time dynamics of the polarization remains bi-exponential,

$$\hat{P}(t) \approx \sum_{j=1}^{2} \hat{A}_j e^{-i\Omega_j t - \Gamma_j t} \quad (t > \tau_B),$$

(26)

where $\Omega_j$ ($\Gamma_j$) are the polariton frequencies (linewidths) and $\hat{A}_j$ are the amplitudes. The polariton Rabi splitting is $\Omega_2 - \Omega_1 \approx 2g$, see [29].

Both polarizations, $|P_{XX}(t)|$ and $|P_{CC}(t)|$, which are very similar in this case, are shown in Fig. 2(a). There is a clear damping of the beating of the two exponentials, even for zero detuning (shown). This implies that the lines of the polariton doublet now have quite different linewidths, as it is clear from Fig. 2(b). The temperature dependence of these linewidths, shown in the inset of Fig. 3, is a clear evidence of the phonon-induced

![Fig. 1](image1.jpg)

**(a)** Excitonic linear polarization and (b) absorption for $T = 0$ and 50 K, calculated in the LN approach with $L = 15$ (red thick solid lines), NN approach with $L = 1$ (black thin solid lines), analytic approximation Eq. (21) (blue dashed lines) and refined analytics (29) (green dotted line). We use the realistic parameters of InGaAs QDs studied in [23, 28] and micropillars studied in [4, 5] (see also [24] for details) including $g = 50$ $\mu$eV, $\omega_X = \Omega_X - \tau_X$ with $\Omega_X = 1329.6$ meV and $\tau_X = 2$ $\mu$eV; $\omega_C = \Omega_C - \tau_C$ with $\Omega_C = \Omega_X + \Omega_p$, $\Omega_p = -49.8$ $\mu$eV and $\gamma_C = 30$ $\mu$eV. Inset: linear plot of the absorption with limited frequency range.

![Fig. 2](image2.jpg)

**(a)** As Fig. 1 but for $g = 0.6$ meV and only LN result shown, for $T = 0$ (red lines) and 50 K (black lines). The photon polarization and absorption are also shown (dashed lines). Insets: (a) the initial polarization dynamics; (b, left) linear plot of the absorption, Re $P_{XX}(\omega)$; (b, right) the relative error for the parameters of the long-time bi-exponential dependence of $P_{XX}(t)$, Eq. (26), as a function of the number of neighbors $L$, using $L = 15$ for the exact solution.
of the Rabi doublet. For small $g$, this solution reveals the dephasing mechanism in terms of the coupling strength $g$, calculated in the $L$ approach with $L = 15$. Inset: temperature dependence of $\Gamma_{1,2}$ for $g = 50 \mu eV$ and 0.6 meV.

broadening mechanism of the polariton lines. Indeed, at $T = 0$, the high-energy polariton state (2) is significantly broader than the low-energy state (1), due to the allowed transition $2 \rightarrow 1$, accompanied by emission of an acoustic phonon, see the left inset in Fig. 2(b). At elevated temperatures both transitions $2 \rightarrow 1$ and $1 \rightarrow 2$, with phonon emission and absorption respectively, are allowed, giving rise to more balanced linewidths.

Looking more closely at the decoherence mechanism caused by real phonon assisted transitions between the lines of the polariton doublet, we see in Fig. 2 that the phonon-induced line broadening increases with $g$, reaching its maximum at a value corresponding to the typical energy of acoustic phonons surrounding the QD [28], which is in the range 1 – 2 meV for these QDs. This corresponds to the polariton line splitting at around $g = 0.6 \mu eV$.

The right inset in Fig. 2(b) demonstrates the quality of the present calculation. Even for large $g$, the LN result quickly converges to the exact solution. Indeed, the error for the parameters of the long-time dependence Eq. (26) decreases exponentially as $2^{-L/2}$. The computational time $t_c$ is $\propto 2^L$, giving an error that scales as $1/\sqrt{t_c}$. Also, Fig. 3 shows that for $L = 15$, the relative error reaches small values of $10^{-5}$ for $g = 50 \mu eV$ and scales as $g^3$ up to $g = 0.5 \mu eV$ where it saturates at the level below 1%.

In conclusion, we have provided an asymptotically exact semi-analytic solution for the linear optical response of a QD-microcavity system coupled to an acoustic-phonon environment, valid for a wide range of system parameters. For large cavity-QD coupling strength $g$, this solution reveals the dephasing mechanism in terms of real phonon-assisted transitions between polariton states of the Rabi doublet. For small $g$, our method simplifies to an accurate analytic solution which provides an intuitive physical picture in terms of polaron-transformed polariton states superimposed with the phonon broad-band, known from the independent boson model.

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* Electronic address: morreauai@cardiff.ac.uk

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**Supplemental Material for**

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A. Morreau and E. A. Muljarov

School of Physics and Astronomy, Cardiff University, Cardiff CF24 3AA, United Kingdom

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**DERIVATION OF EQ. (3) FOR THE LINEAR POLARIZATION**

We take as our starting point the standard definition of the optical polarization,

\[ P = \text{Tr} \{ \rho(t)c \}, \quad (S1) \]

where the annihilation operator \( c \) stands either for the exciton operator \( d \) or for the cavity operator \( a \). Consequently, Eq. (S1) has the meaning of the full excitonic or photonic polarization, respectively. Here \( \rho(t) \) is the full density matrix of the exciton, cavity, and phonon degrees of freedom.

To obtain the linear polarization from Eq. (S1), we first need to assume a pulsed excitation of the system at time \( t = 0 \), which is described by the following evolution of the density matrix:

\[ \rho(0_+) = e^{-i\mathcal{V}} \rho(-\infty) e^{i\mathcal{V}}, \quad (S2) \]

where \( \rho(-\infty) \) is the density matrix of a fully unexcited system, with its exciton-cavity part being in the absolute ground state \( |0\rangle \) and phonons being in thermal equilibrium,

\[ \rho(-\infty) = |0\rangle \langle 0| \rho_0, \quad \rho_0 = e^{-\beta \mathcal{H}_{ph}}/\text{Tr} \{ e^{-\beta \mathcal{H}_{ph}} \}_\text{ph}. \quad (S3) \]

Here, \( \beta = (k_B T)^{-1} \), and the trace is taken over all possible phonon states. The perturbation \( \mathcal{V} \) due to the pulsed excitation has the form:

\[ \mathcal{V} = \alpha (\tilde{c} \dagger + \tilde{c}), \quad (S4) \]

where \( \alpha \) is a constant, and again, \( \tilde{c} \) is either \( d \) or \( a \), depending on the excitation (feeding) channel. In the linear polarization, we keep in the full polarization only the terms which are linear in \( \alpha \). Looking closer, this implies keeping only \( |X\rangle \langle 0| \) and \( |C\rangle \langle 0| \) elements of the density matrix.

We assume that the evolution of the full density matrix of the exciton-cavity-phonon system after its optical pulsed excitation is given by the following standard Lindblad master equation

\[ i\dot{\rho} = H \rho - \rho H^* + 2i\gamma_X d \rho d^\dagger + 2i\gamma_C a \rho a^\dagger, \quad (S5) \]

where \( H \) is full Hamiltonian of the exciton-phonon-cavity systems, defined on the first page of the main text, and \( H^* \) is its complex conjugate. When the density matrix is reduced to only \( |X\rangle \langle 0| \) and \( |C\rangle \langle 0| \) elements, the last two terms in Eq. (S5) vanish, which yields an explicit solution of Eq. (S5):

\[ \rho(t) = e^{-iH^* t} e^{-iH^* t} \rho(0_+) e^{iH^* t}, \quad (S6) \]

in which \( H^* \) can actually be replaced by \( \mathcal{H}_{ph} \). The linear polarization then takes the form

\[ P_L(t) = -i\alpha \text{Tr} \left\{ e^{-iH^* t} |0\rangle \langle 0| \rho_0 e^{i\mathcal{H}_{ph} t} c \right\} \quad (S7) \]

Now, dropping the unimportant constant factor \(-i\alpha\) and introducing indices \( j, k = X, C \) to replace the operators \( \tilde{c} \dagger \) and \( c \), we arrive at Eq. (3) of the main text.

**TROTTER DECOMPOSITION OF THE EVOLUTION OPERATOR**

Using the Trotter decomposition, the evolution operator is presented in Eq. (4) as \( \hat{U}(t) = \lim_{N \to \infty} \hat{U}_N(t) \), where

\[
\hat{U}_N(t) = e^{i\mathcal{H}_{ph} t} e^{-i\mathcal{H}_{ph} (t-t_{N-1})} e^{-iH_{JC} (t-t_{N-1})} \cdots e^{-i\mathcal{H}_{ph} (t_{n-1})} e^{-iH_{JC} (t_{n-1})} \cdots e^{-i\mathcal{H}_{ph} t_1} e^{-iH_{JC} t_1} \\
= e^{i\mathcal{H}_{ph} t} e^{-i\mathcal{H}_{ph} (t-t_{N-1})} e^{-iH_{ph} t_{N-1}} e^{-iH_{JC} (t-t_{N-1})} \cdots \\
\times e^{i\mathcal{H}_{ph} t_1} e^{-i\mathcal{H}_{ph} (t_{n-1})} e^{-iH_{ph} t_{n-1}} e^{-iH_{JC} (t_{n-1})} \cdots e^{iH_{ph} t_1} e^{-iH_{ph} t_1} e^{-iH_{JC} t_1} \\
= \hat{W}(t, t_{N-1}) \hat{M}(t - t_{N-1}) \cdots \hat{W}(t_n, t_{n-1}) \hat{M}(t_n - t_{n-1}) \cdots \hat{W}(t_1, 0) \hat{M}(t_1), \quad (S8)
\]
where we have used the fact that the operators $H_{\text{ph}}$ and $H_{\text{IC}}$ commute. From the definition of $H_{\text{IB}}$ we note that

$$W(t_n, t_{n-1}) = e^{iH_{\text{ph}}t_n} e^{-iH_{\text{IB}}(t_n-t_{n-1})} e^{-iH_{\text{ph}}t_{n-1}}$$

(S9)

is a diagonal operator in the 2-basis state matrix representation in terms of $|X\rangle$ and $|C\rangle$:

$$W(t_n, t_{n-1}) = \begin{pmatrix} W_X(t_n, t_{n-1}) & 0 \\ 0 & W_C(t_n, t_{n-1}) \end{pmatrix}$$

(S10)

with

$$W_X(t_n, t_{n-1}) = e^{iH_{\text{ph}}t_n} e^{-i(H_{\text{ph}}+V)(t_n-t_{n-1})} e^{-iH_{\text{ph}}t_{n-1}},$$

(S11)

$$W_C(t_n, t_{n-1}) = 1.$$  

(S12)

Using the time ordering operator $\mathcal{T}$, Eq. (S11) for $W_X(t_n, t_{n-1})$ can be written as

$$W_X(t_n, t_{n-1}) = \mathcal{T} \exp \left\{ -i \int_{t_{n-1}}^{t_n} V(\tau) d\tau \right\},$$

(S13)

where $V(\tau) = e^{iH_{\text{ph}}\tau} V e^{-iH_{\text{ph}}\tau}$ is the interaction representation of the exciton-phonon coupling $V$, which is given by Eq. (2) of the main text.

Substituting the evolution operator Eq. (S8) into Eq. (3) for the polarization $P_{jk}(t)$ and explicitly expressing the matrix products gives

$$P_{jk}(t) = \sum_{i_N=1}^{X,C} \cdots \sum_{i_1=1}^{X,C} \langle W_{i_N M_{i_N} i_{N-1}} W_{i_{N-1} M_{i_{N-1} i_{N-2}}} \cdots M_{i_{N-2} i_{N-3}} W_{i_{N-3} M_{i_{N-3} i_{N-4}}} \cdots W_{i_1 M_{i_1 i_0}} \rangle_{\text{ph}}$$

(S14)

with $i_N = j$ and $i_0 = k$. From here, we note that only $W$ elements contain the phonon interaction and through a simple rearrangement of Eq. (S14) we arrive at Eq. (3) of the main text.

**CALCULATION OF $K_{|n-m|}$ FROM THE IB MODEL CUMULANT**

As is clear from the definition given in Eq. (14) of the main text, the integral $K_{|n-m|}$ depends only on the difference $|n-m|$. To find $K_0$, we set $m = n = 1$:

$$K_0 = -\frac{1}{2} \int_0^{t_1} d\tau_1 \int_0^{t_1} d\tau_2 \langle TV(\tau_1) V(\tau_2) \rangle = K(\Delta t).$$

(S15)

Here, $K(t)$ is the IB cumulant, which is calculated explicitly in Sec. below, see Eq. (S27).

Analogously, to find $K_1$ we may set $m = 1$ and $n = 2$ which gives

$$K_1 = -\frac{1}{2} \int_0^{t_2} d\tau_1 \int_0^{t_1} d\tau_2 \langle TV(\tau_1) V(\tau_2) \rangle,$$

(S16)

or, by setting $m = 2$ and $n = 1$ instead, we obtain the same result:

$$K_1 = -\frac{1}{2} \int_0^{t_1} d\tau_1 \int_0^{t_2} d\tau_2 \langle TV(\tau_1) V(\tau_2) \rangle.$$  

(S17)

Eqs. (S16) and (S17) correspond to the squares labeled as $K_1$ in Fig. S1. In order to calculate $K_1$ from the IB cumulant, we note that

$$K(2\Delta t) = 2K_0 + 2K_1.$$  

(S18)

Therefore,

$$K_1 = \frac{1}{2} [K(2\Delta t) - 2K_0].$$

(S19)

In general, all the integrals $K_p$ can be found recursively:

$$K_{p>0} = \frac{1}{2} \left[ K((p+1)\Delta t) - (p+1)K_0 - \sum_{q=1}^{p-1} 2(p+1-q)K_q \right].$$

(S20)
THE IB MODEL CUMULANT AND ITS LONG-TIME BEHAVIOR EQ. (20)

The IB model cumulant \( K(t) \) can be conveniently written in terms the standard phonon propagator \( D_q \),

\[
K(t) = -\frac{i}{2} \int_0^t d\tau_1 \int_0^t d\tau_2 \sum_q |\lambda_q|^2 D_q(\tau_1 - \tau_2), \tag{S21}
\]

where

\[
iD_q(t) = \langle T [b_q(t) + b_{-q}^\dagger(0)] [b_{-q}(0) + b_{-q}(0)] \rangle = N_q e^{i\omega_q |t|} + (N_q + 1)e^{-i\omega_q |t|} \tag{S22}
\]

and \( N_q \) is the Bose distribution function,

\[
N_q = \frac{1}{e^{\beta\omega_q} - 1}. \tag{S23}
\]

Performing the integration in Eq. (S21), we obtain

\[
K(t) = \sum_q |\lambda_q|^2 \left( \frac{N_q}{\omega_q^2} [e^{i\omega_q t} - 1] + \frac{N_q + 1}{\omega_q^2} [e^{-i\omega_q t} - 1] + \frac{it}{\omega_q} \right). \tag{S24}
\]

Converting the summation over \( q \) to an integration \( \sum_q \rightarrow \frac{V}{(2\pi)^3 v_s^2} \int d^3\omega \), where \( V \) is the sample volume, we then have

\[
K(t) = \frac{V}{(2\pi)^3 v_s^2} \int d^3\omega |\lambda_q|^2 \left( \frac{N_q}{\omega^2} [e^{i\omega t} - 1] + \frac{N_q + 1}{\omega^2} [e^{-i\omega t} - 1] + \frac{it}{\omega} \right) \tag{S25}
\]

\[
= \frac{4\pi V}{(2\pi)^3 v_s^2} \int_0^\infty d\omega \omega^2 |\lambda_q|^2 \left( \frac{N_q}{\omega^2} [e^{i\omega t} - 1] + \frac{N_q + 1}{\omega^2} [e^{-i\omega t} - 1] + \frac{it}{\omega} \right). \tag{S26}
\]

Noting that \( |\lambda_q|^2 \) may be expressed in terms of the spectral density function \( J(\omega) \) [see Eq. (S36) in Sec. below], we re-write Eq. (S26) as

\[
K(t) = \int_0^\infty J(\omega) \left( \frac{N_q}{\omega^2} [e^{i\omega t} - 1] + \frac{N_q + 1}{\omega^2} [e^{-i\omega t} - 1] + \frac{it}{\omega} \right). \tag{S27}
\]

In the long-time limit, Eq. (S27) simplifies to

\[
K(t \rightarrow \infty) = -i\Omega_p t - S, \tag{S28}
\]

with the polaron shift

\[
\Omega_p = -\int_0^\infty d\omega \frac{J(\omega)}{\omega}. \tag{S29}
\]
FIG. S2. IB model cumulant $K(t)$, with its long-time asymptotics $-i\Omega_p t - S$ subtracted, as a function of time $t$ for different temperatures as given. The parameters used are listed at the end of Sec. Inset: the phonon memory time $\tau_{IB}$ playing the role of the cut-off parameter in calculation of the cumulants for different realizations in the $LN$ approach.

and the Huang-Rhys factor

$$S = \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} \left( 2N_\omega + 1 \right) = \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} \coth \left( \frac{\omega}{2k_B T} \right).$$  \hspace{1cm} (S30)

Figure S2 shows the cumulant function $K(t)$ of the IB model at different temperatures. It demonstrates, in particular, that the asymptotics Eq. (S28) (subtracted) accurately describe the long-time behavior, but at the same time $K(t)$ strongly depends on temperature. This results in temperature dependent cut-off times $\tau_{IB}$ shown in the inset. Physically, the cut-off time relates to the phonon memory time required for the polaron cloud to form around an optically excited quantum dot. In our calculation, it determines the choice of the minimal time step in the NN approximation ($\Delta t \approx \tau_{IB}$) and the $LN$ approach ($L\Delta t \approx \tau_{IB}$). The definition of $\tau_{IB}$ depends on the required accuracy of the calculation; in the present calculation we define $\tau_{IB}$ as the time at which the quickly varying part of the cumulant, $K(t) + i\Omega_p t + S$, shown in Fig. S2 drops down to approximately $10^{-4}$. $\tau_{IB}$ also depends on a number of model parameters, such as the exciton confinement radius $l$ and the deformation potential constants for the valence and conduction bands, $D_v$ and $D_c$. For $l = 3.3$ nm, $D_c - D_v = -6.5$ eV, and $T = 50$ K, the polaron timescale is approximately 3.25 ps; it increases to 10 ps at $T = 0$.

**EXCITON-PHONON COUPLING MATRIX ELEMENT $\lambda_q$ AND THE SPECTRAL DENSITY FUNCTION $J(\omega)$**

At low temperatures, the exciton-phonon interaction is dominated by the deformation potential coupling to longitudinal acoustic phonons. Assuming (i) that the phonon parameters in the confined QD do not differ significantly from those in the surrounding material, and (ii) that the acoustic phonons have linear dispersion $\omega_q = v_s |q|$, where $v_s$ is the sound velocity in the material, the matrix coupling element $\lambda_q$ is given by

$$\lambda_q = \frac{qD(q)}{\sqrt{2\rho_m \omega_q V}},$$  \hspace{1cm} (S31)

where $\rho_m$ is the mass density of the material. Assuming a factorizable form of the exciton wave function, $\Psi_X(r_e, r_h) = \psi_e(r_e)\psi_h(r_h)$, where $\psi_{e(h)}(r)$ is the confined electron (hole) ground state wave function, the form-factor $D(q)$ is given by

$$D(q) = \int dr \left[ D_e|\psi_h(r)|^2 - D_c|\psi_e(r)|^2 \right] e^{-iq\cdot r},$$  \hspace{1cm} (S32)
with $D_{c(v)}$ being the material-dependent deformation potential constant for the conduction (valence) band. We choose for simplicity spherically symmetric parabolic confinement potentials which give Gaussian ground state wave functions:

$$
\psi_{c(h)}(r) = \frac{1}{(\sqrt{\pi}l_{c(h)})^{3/2}} \exp \left( -\frac{r^2}{2l_{c(h)}^2} \right),
$$

(S33)

and thus

$$
\lambda_q = \sqrt{\frac{q}{2\rho_m v_s^3}} \left( D_v e^{-\frac{q^2}{4v_s^2}} - D_c e^{-\frac{q^2}{4v_s^2}} \right) = \sqrt{\frac{q}{2\rho_m v_s^3}} (D_v - D_c) e^{-\frac{q^2}{4v_s^2}},
$$

(S34)

using also $l_c = l_h = l$ for simplicity.

The spectral density $J(\omega)$ is defined as

$$
J(\omega) = \sum_q |\lambda_q|^2 \delta(\omega - \omega_q).
$$

(S35)

This is equivalent to taking the product of $|\lambda_q|^2$ with the density of states in $\omega$-space. Switching from the summation to an integration, as in Eq. (S25), the spectral density becomes

$$
J(\omega) = |\lambda_q|^2 \frac{2V}{(2\pi)^2 v_s^2} \omega^2 = \frac{\omega^3(D_v - D_c)^2}{4\pi^2 \rho_m v_s^5} e^{-\frac{\omega^2}{\omega_0^2}}.
$$

(S36)

where $q = \omega/v_s$ and $\omega_0 = \sqrt{2}v_s/l$ is the so-called “cut-off” frequency; it is approximately equal to the inverse phonon memory time: $\tau_{ph} \sim 1/\omega_0$.

In all calculations, we use $l = 3.3$ nm, $D_c - D_v = -6.5$ eV, $v_s = 4.6 \times 10^4$ m/s, and $\rho_m = 5.65$ g/cm$^3$.

**LINEAR POLARIZATION IN THE NN APPROXIMATION, INCLUDING AN EXAMPLE REALIZATION**

The single summation in the cumulant Eq. (15) allows us to express, for each realization, the expectation value in Eq. (3) as a product

$$
\langle W_{i_1} (t, t_{N-1}) \ldots W_{i_2} (t, t_1) \rangle_{ph} = e^{\Delta N x K_0} e^{\Delta N x (K_0 + 2\delta_N x K_1)} \ldots e^{\Delta N x (K_0 + 2\delta_N x K_1)}.
$$

(S37)

It is convenient to introduce

$$
R_{i_{n-1}i_n} = e^{\Delta N x (K_0 + 2\delta_N x K_1)},
$$

(S38)

enabling us to express the expectation values of the product of $W$-operators for a given realization as

$$
\langle W_{i_1} (t, t_{N-1}) \ldots W_{i_2} (t_1, t_0) \rangle_{ph} = e^{\Delta N x K_0} R_{i_{N-1}i_N} \ldots R_{i_2i_1}.
$$

(S39)

Substituting Eq. (S39) into Eq. (3) for $P_{jk}$, we find

$$
P_{jk}(t) = e^{\Delta N x K_0} \sum_{i_{N-1}=X,C} \ldots \sum_{i_1=X,C} (M_{i_{N-1}i_N} \ldots M_{i_2i_1} M_{i_1i_0}) (R_{i_{N-1}i_N} \ldots R_{i_2i_1}).
$$

(S40)

We then join together corresponding $M_{i_{n-1}i_n}$ and $R_{i_{n-1}i_n}$ elements through the definition of a matrix

$$
G_{i_{n-1}i_n} = M_{i_{n-1}i_n} R_{i_{n-1}i_n},
$$

(S41)

which transforms Eq. (S40) to

$$
P_{jk}(t) = e^{\Delta N x K_0} \sum_{i_{N-1}=X,C} \ldots \sum_{i_{n-1}=X,C} \ldots \sum_{i_1=X,C} G_{i_{N-1}i_N} \ldots G_{i_{n-1}i_n} \ldots G_{i_2i_1} M_{i_1i_0}.
$$

(S42)
FIG. S3. Example realization for the NN implementation with \( N = 5 \). In this realization, \( i_1 = X, i_2 = C, i_3 = X, i_4 = X, i_5 = C \), as is clear from the step function \( \hat{\theta}(t) \) associated with the given realization, shown on the top.

Using the fact that \( i_N = j \) and \( i_0 = k \), we arrive at Eq. (16), which is compactly represented in Eq. (18) as a product of matrices.

To illustrate this idea by way of an example, we take a particular realization for \( N = 5 \), provided for illustration in Fig. S3. In this realization, \( i_1 = X, i_2 = C, i_3 = X, i_4 = X, i_5 = C \). Each exponential \( e^{\delta_{i-1}x(K_0 + 2\delta_{i}xK_1)} \) in Eq. (18) can be visualized as an L-shaped portion of the time grid (color coded in the figure). In the illustrated realization we have,

\[
\begin{align*}
R_{i_2i_1} &= e^{\delta_{i}X(K_0 + 2\delta_{i}xK_1)} = e^{K_0}, \\
R_{i_3i_2} &= e^{\delta_{i}X(K_0 + 2\delta_{i}xK_1)} = e^{0} = 1, \\
R_{i_4i_3} &= e^{\delta_{i}X(K_0 + 2\delta_{i}xK_1)} = e^{K_0 + 2K_1}, \\
R_{i_5i_4} &= e^{\delta_{i}X(K_0 + 2\delta_{i}xK_1)} = e^{K_0}, \\
R_{i_5i_4} &= e^{\delta_{i}X(K_0 + 2\delta_{i}xK_1)} = e^{0} = 1.
\end{align*}
\]

We then find

\[
\begin{align*}
G_{i_2i_1} &= G_{CX} = M_{CX}e^{K_0}, \\
G_{i_3i_2} &= G_{XC} = M_{XC}, \\
G_{i_4i_3} &= G_{XX} = M_{XX}e^{K_0 + 2K_1}, \\
G_{i_5i_4} &= G_{CX} = M_{CX}e^{K_0},
\end{align*}
\]

which contributes to the total polarization Eq. (S42).

Note that the condition for the NN approximation to be valid is also illustrated in Fig. S3. All the time moments of integration for which \( |\tau_2 - \tau_1| < \tau_B \) should be located within the colored squares, which are taken into account in the NN calculation of the cumulant.

**LONG-TIME ANALYTICS FOR THE LINEAR POLARIZATION**

In this section, we derive the approximate analytic result Eqs. (21) and (22) for the linear polarization \( \hat{P}(t) \) in the long-time limit. This approximation is valid for small values of the exciton-cavity coupling strength \( g \), which guarantees that the polariton timescale is much longer than the phonon memory time, \( \tau_J \gg \tau_B \). As a starting point,
we take the result for $\hat{P}(t)$ in the NN approach, Eqs. [18] and [19], and use it for $\Delta t \geq \tau_{IB}$. This condition implies that we can take both $K_0$ and $K_1$ in the long-time limit, using the asymptotic formula Eq. (20):

$$K_0 = K(\Delta t) \approx -i\Omega_p \Delta t - S,$$  \hspace{1cm} (S43)

$$K_1 = \frac{1}{2}(K(2\Delta t) - 2K(\Delta t)) \approx \frac{S}{2},$$  \hspace{1cm} (S44)

We would now like to replace the product of $N$ matrices in Eq. (18) by an approximate analytic expression, taking the Trotter limit $N \to \infty$. To do so, we initially derive explicit expressions for $\hat{M}$ and $\hat{G}$ in the two-state basis of $|X\rangle$ and $|C\rangle$. From Eq. [20] we obtain

$$(M_{XX} M_{XC}) = e^{-i\Omega C \Delta t} = \begin{pmatrix} \alpha & \beta \\ -\beta & \alpha \end{pmatrix} \begin{pmatrix} e^{-i\omega_1 \Delta t} & 0 \\ 0 & e^{-i\omega_2 \Delta t} \end{pmatrix} \begin{pmatrix} \alpha & -\beta \\ \beta & \alpha \end{pmatrix} = e^{-i\omega_1 \Delta t} \begin{pmatrix} 1 - \beta^2 & -\alpha \beta \delta \\ -\alpha \beta \delta & 1 - \alpha^2 \delta \end{pmatrix},$$  \hspace{1cm} (S45)

where $\omega_{1,2}$ are the eigenvalues of the Jaynes-Cummings Hamiltonian $H_{JC}$, $\delta = 1 - e^{-i(\omega_2 - \omega_1)\Delta t}$, and $\alpha$ and $\beta$ make up the unitary matrices that diagonalize $H_{JC}$:

$$H_{JC} = \begin{pmatrix} \omega_X & g \\ g & \omega_C \end{pmatrix} = \begin{pmatrix} \alpha & \beta \\ -\beta & \alpha \end{pmatrix} \begin{pmatrix} \omega_1 & 0 \\ 0 & \omega_2 \end{pmatrix} \begin{pmatrix} \alpha & -\beta \\ \beta & \alpha \end{pmatrix}.$$  \hspace{1cm} (S46)

Substituting the expression for $\hat{M}$ given by Eq. (S45) into Eq. (19), and using Eqs. (S43) and (S44), we find

$$\hat{G} = \begin{pmatrix} M_{XX} e^{K_0 + 2K_1} M_{XC} e^{K_0} \\ M_{XX} e^{K_0} \end{pmatrix} \approx e^{-i\omega_1 \Delta t} \begin{pmatrix} e^{-i\Omega_p \Delta t} & 1 - \beta^2 \delta \\ -\alpha \beta \delta & 1 - \alpha \beta \delta \end{pmatrix}.$$  \hspace{1cm} (S47)

Now we use the fact that $\Delta t \ll \tau_{JC}$ (which is equivalent to $|\omega_2 - \omega_1| \Delta t \ll 1$). We also assume that the polaron shift $\Omega_p$ is small, so that $|\Omega_p| \Delta t \ll 1$. Working within these limits is equivalent to taking the Trotter limit $\Delta t = t/N \to 0$. Keeping only the terms linear in $\Delta t$ in the matrix elements, we obtain

$$\hat{G} \approx e^{-i\omega_1 \Delta t} \begin{pmatrix} 1 - i\Delta t (\Omega_p + \beta^2 (\omega_2 - \omega_1)) & -i\Delta t \alpha \beta (\omega_2 - \omega_1) \\ -i\Delta t \alpha \beta (\omega_2 - \omega_1) e^{-S} & 1 - i\Delta t \alpha^2 (\omega_2 - \omega_1) \end{pmatrix},$$  \hspace{1cm} (S48)

From Eq. (S46) and the fact that $\alpha^2 + \beta^2 = 1$ we find

$$\beta^2 (\omega_2 - \omega_1) = \omega_X - \omega_1,$$  \hspace{1cm} (S49)

$$\alpha^2 (\omega_2 - \omega_1) = \omega_C - \omega_1,$$  \hspace{1cm} (S50)

$$\alpha \beta (\omega_2 - \omega_1) = g.$$  \hspace{1cm} (S51)

This allows us to re-write Eq. (S48) in the following way

$$\hat{G} = e^{-i\omega_1 \Delta t} \left[ 1 + i \omega_1 \Delta t - i \Delta t \begin{pmatrix} \omega_X + \Omega_p & g \\ g e^{-S} \omega_C \end{pmatrix} \right],$$  \hspace{1cm} (S52)

where $1$ is a $2 \times 2$ identity matrix. Now, we diagonalize $\hat{G}$:

$$\hat{G} = \hat{Y} \hat{\Lambda} \hat{Y}^{-1},$$  \hspace{1cm} (S53)

where the transformation matrix has the form

$$\hat{Y} = \begin{pmatrix} e^{S/2} & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \hat{\alpha} & \hat{\beta} \\ -\hat{\beta} & \hat{\alpha} \end{pmatrix},$$  \hspace{1cm} (S54)

in which the second matrix diagonalizes a phonon-renormalized JC Hamiltonian $\tilde{H}$ [see Eq. (22)]:

$$\tilde{H} = \begin{pmatrix} \omega_X + \Omega_p & g e^{-S/2} \omega_C \\ g e^{-S/2} \omega_C & \omega_C \end{pmatrix} = \begin{pmatrix} \hat{\alpha} & \hat{\beta} \\ -\hat{\beta} & \hat{\alpha} \end{pmatrix} \begin{pmatrix} \omega_1 & 0 \\ 0 & \omega_2 \end{pmatrix} \begin{pmatrix} \hat{\alpha} & -\hat{\beta} \\ \hat{\beta} & \hat{\alpha} \end{pmatrix}.$$  \hspace{1cm} (S55)

The matrix of the eigenvalues $\hat{\Lambda}$ in Eq. (S53) then takes the form

$$\hat{\Lambda} = e^{-i\omega_1 \Delta t} \begin{pmatrix} 1 - i \Delta t (\tilde{\omega}_1 - \omega_1) & 0 \\ 0 & 1 - i \Delta t (\tilde{\omega}_2 - \omega_1) \end{pmatrix}.$$  \hspace{1cm} (S56)
Coming back to the NN expression for the polarization Eq. (18),

$$\hat{\mathcal{P}}(t) = \left( e^{K_0} 0 \\ 0 1 \right) \hat{G}^N \hat{G}^{-1} \hat{M}, \quad \text{(S57)}$$

we note that $\hat{G}^{-1} \approx 1$ and $\hat{M} \approx 1$ in the limit $\Delta t \to 0$, and also $e^{K_0} \approx e^{-S}$ (still keeping the condition $\Delta t \gtrsim \tau_B$). We then obtain in the long-time limit $t \gtrsim \tau_B$:

$$\hat{\mathcal{P}}(t) = e^{-i\omega t} \left( e^{-S} 0 \\ 0 1 \right) \hat{N}^{\text{AN}} \hat{N}^{-1} \hat{Y}. \quad \text{(S58)}$$

Finally, we take the limit $N \to \infty$ in the expression $\hat{N}^{\text{AN}}$, using an algebraic formula

$$\lim_{N \to \infty} \left( 1 + \frac{x}{N} \right)^N = e^x. \quad \text{(S59)}$$

Introducing

$$x = -i(\tilde{\omega}_1 - \omega_1)t, \quad \text{(S60)}$$
$$y = -i(\tilde{\omega}_2 - \omega_1)t, \quad \text{(S61)}$$
we find

$$\lim_{N \to \infty} \hat{N}^{\text{AN}} = \lim_{N \to \infty} \left( 1 + \frac{x}{N} \cdot 0 \cdot 1 + \frac{y}{N} \right)^N = e^{i\omega_1 t} \left( e^{-i\omega_1 t} 0 \\ 0 e^{-i\omega_2 t} \right). \quad \text{(S62)}$$

So finally,

$$\hat{\mathcal{P}}(t) = \left( e^{-S} 0 \\ 0 1 \right) \left( e^{S/2} 0 \\ 0 1 \right) \left( \hat{\alpha} \hat{\beta} \hat{\beta} \hat{\alpha} \right) \left( e^{-i\omega_1 t} 0 \\ 0 e^{-i\omega_2 t} \right) \left( \hat{\alpha} - \hat{\beta} \right) \left( 0 1 \right) \quad \text{(S63)}$$

$$= \left( e^{-S/2} 0 \\ 0 1 \right) e^{-i\hat{C}t} \left( e^{-S/2} 0 \\ 0 1 \right), \quad \text{(S64)}$$

which is Eq. (21) of the main text.

**POLARITON RABI SPLITTING, BI-EXPONENTIAL FIT PARAMETERS AND TIMESCALES**

Figure S4 illustrates the dependence of the polariton Rabi splitting $\Delta \omega = \Omega_2 - \Omega_1$ on the exciton-cavity coupling strength $g$ for zero detuning. As it follows from the analytic approximation Eqs. (21) and (22), valid for small $g$, the Rabi splitting in this regime is roughly given by

$$\Delta \omega \approx 2ge^{-S/2}, \quad \text{(S65)}$$

where $S$ is the Huang-Rhys factor defined by Eq. (S30). This dependence is indeed observed in the full calculation for coupling strength $g$ below 0.2 meV (0.5 meV) for $T = 50$ K ($T = 0$) (compare the full and dashed lines in Fig. S4). A minor deviation from the analytic formula Eq. (S65) at small $g$ is due to finite exciton and cavity lifetimes used in the calculation: $\gamma_X = 2 \mu$eV and $\gamma_C = 30 \mu$eV.

To better understand this behavior and the range of applicability of the phonon-induced Rabi splitting renormalization Eq. (S65), we first note that the temporal period of the polariton Rabi oscillations is given by

$$\tau_{JC} = \frac{2\pi}{\Delta \omega}. \quad \text{(S66)}$$

The derivation of Eq. (S65) provided in Sec. is based on the NN approximation which is using $\Delta t \approx \tau_B \ll \tau_{JC}$. As $g$ increases the polariton time reduces according to Eqs. (S65) and (S66), and hence $\Delta t$ becomes comparable to, or even greater than $\tau_{JC}$. Note that this limitation only affects the NN expression for $\hat{P}(t)$ given by Eq. (18) and the analytic expression for $t \gg \tau_B$ given by Eq. (21). For $L$ neighbors we are not limited to $\Delta t \approx \tau_B$ and may reach infinitesimal $\Delta t$ as $L \to \infty$ according to the constraint $L \Delta t \approx \tau_B$.

The imaginary part $\Gamma_j$ ($j = 1, 2$) of the complex polariton frequencies $\omega_j$ are shown in Fig. 3 of the main text as functions of the coupling strength $g$ and temperature $T$. For completeness, we provide in Fig. S5 the amplitudes of the bi-exponential fit Eq. (26) of the full calculation in the LN approach with $L = 15$ (symbols) and the analytic approximation Eq. (21) which is strictly bi-exponential (lines). Figure S5 addresses both excitonic and photonic polarization, $P_{XX}$ and $P_{CC}$ (black and red respectively).
FIG. S4. Deviation of the polariton Rabi splitting $\Delta \omega = \Omega_2 - \Omega_1$ (calculated via the LN model with $L = 15$) from the nominal Rabi splitting $2g$ (solid lines), as a function of the exciton-cavity coupling strength $g$ for zero effective detuning ($\omega_C = \omega_X + \Omega_p$) and two different temperatures, $T = 0$ and $T = 50$ K. The deviation of the phonon renormalized Rabi splitting from the nominal Rabi splitting $2g[e^{-S/2} - 1]$ is shown by dashed lines. The inset demonstrates the calculated full Rabi splitting $\Delta \omega$ (solid line) for $g = 50 \mu$eV as a function of the temperature $T$, in comparison with $2ge^{-S/2}$ (dashed lines).

FIG. S5. Polariton amplitude coefficient $|A_1|$ ($|A_2|$) as a function of the quantum dot-cavity coupling strength $g$ for (a) $T = 0$ and (b) $T = 50$ K shown for the full calculation by full squares (open circles) and for the long-time analytic model by full (dashed) lines. Insets zoom in the region of small $g$, where the analytic model predicts significant changes of the amplitudes with $g$.

REFINED FULL TIME ANALYTIC APPROACH

The analytic solution derived in Sec. is suited only for describing the optical polarization at long times $t \gtrsim \tau_{IB}$, so that any information on the evolution at short times, which is responsible for the so-called phonon broadband observed in the optical spectra of quantum dots, is missing. To improve on this, we derive a refined, purely analytic approach which properly takes into account both the short and long time dynamics, providing a smooth transition between the two regimes.

We again start with the general formula Eq. \ref{eq:refined} for the linear polarization, writing it in a matrix form using the two basis states $|X\rangle$ and $|C\rangle$:

$$\hat{P}(t) = \langle \hat{U}(t) \rangle .$$  
(S67)
Note that the expectation value in Eq. (S67) is taken over the phonon system in thermal equilibrium, and the $2 \times 2$ evolution matrix operator $\tilde{U}(t)$ has the form:

$$\tilde{U}(t) = e^{iH_{ph}t}e^{-iHt} = e^{-iH_{3C}t}e^{iH_{1}t}e^{-iHt},$$

where

$$H_{JC} = \left( \begin{array}{cc} \omega_X & g \\ g & \omega_C \end{array} \right), \quad H_1 = H_{JC} + H_{ph1}, \quad H = H_1 + \left( \begin{array}{c} 1 \\ 0 \end{array} \right) V,$$

and $H_{ph}$ and $V$ are defined in Eq. 2 of the main text. We apply to the evolution operator the polariton transformation Eq. (S40) which can be compactly written as $H_{JC} = \tilde{Y}H_{0}\tilde{Y}^{-1}$, where

$$\tilde{Y} = \left( \begin{array}{cc} \alpha & \beta \\ -\beta & \alpha \end{array} \right), \quad H_0 = \left( \begin{array}{cc} \omega_1 & 0 \\ 0 & \omega_2 \end{array} \right).$$

We then obtain

$$\tilde{U}(t) = \tilde{Y}e^{-iH_{0}t}e^{iH_{1}t}e^{-iHt}\tilde{Y}^{-1} = \tilde{Y}e^{-iH_{0}t}\tilde{U}(t)\tilde{Y}^{-1},$$

where

$$\tilde{H}_1 = \tilde{Y}^{-1}H_1\tilde{Y} = H_0 + H_{ph1},$$

$$\tilde{H} = \tilde{Y}^{-1}H\tilde{Y} = H_0 + H_{ph2} + \tilde{Q}V,$$

and

$$\tilde{Q} = \tilde{Y}^{-1}\left( \begin{array}{cc} 1 \\ 0 \end{array} \right) \tilde{Y} = \left( \begin{array}{cc} \alpha^2 & \alpha\beta \\ \alpha\beta & \beta^2 \end{array} \right).$$

The reduced evolution operator can then be written as an exponential series

$$\tilde{U}(t) = e^{i\tilde{H}_1t}e^{-i\tilde{H}t} = T\exp\left\{-i\int_0^t H_{int}(t')dt'\right\},$$

where

$$H_{int}(t) = e^{i\tilde{H}_1t}(\tilde{H} - \tilde{H}_1)e^{-i\tilde{H}_1t} = \tilde{Q}(t)V(t),$$

with individual interaction representations of the polariton and phonon operators: $\tilde{Q}(t) = e^{iH_{0}t}\tilde{Q}e^{-iH_{0}t}$ and $V(t) = e^{iH_{ph2}t}e^{-iH_{ph2}t}$. The expectation value of $\tilde{U}(t)$ then becomes an infinite perturbation series:

$$\langle \tilde{U}(t) \rangle = 1 + (-i)^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \tilde{Q}(t_1)\tilde{Q}(t_2)(V(t_1)V(t_2))$$

$$+ (-i)^4 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \tilde{Q}(t_1)\tilde{Q}(t_2)\tilde{Q}(t_3)\tilde{Q}(t_4)(V(t_1)V(t_2)V(t_3)V(t_4)) \ldots$$

Using Wick’s theorem, all of the expectation values split into pair products. For example,

$$\langle V(t_1)V(t_2)V(t_3)V(t_4) \rangle = \langle V(t_1)V(t_2) \rangle \langle V(t_3)V(t_4) \rangle + \langle V(t_1)V(t_3) \rangle \langle V(t_2)V(t_4) \rangle + \langle V(t_1)V(t_4) \rangle \langle V(t_2)V(t_3) \rangle$$

$$= D(t_1 - t_2)D(t_3 - t_4) + D(t_1 - t_3)D(t_2 - t_4) + D(t_1 - t_4)D(t_2 - t_3),$$

where

$$D(t-t') = \langle V(t)V(t') \rangle = \sum_q |\lambda_q|^2 iD_q(t-t')$$

is the full phonon propagator, see Eq. (S22).

It is convenient to introduce the bare polariton Green’s function

$$\tilde{G}^{(0)}(t) = \left( \begin{array}{cc} G_{1}^{(0)}(t) & 0 \\ 0 & G_{2}^{(0)}(t) \end{array} \right) = \theta(t) e^{-iH_{0}t} = \theta(t) \left( \begin{array}{cc} e^{-i\omega_1 t} & 0 \\ 0 & e^{-i\omega_2 t} \end{array} \right),$$

where

$$G_{1}^{(0)}(t) = \left( \begin{array}{cc} \alpha^2 & \alpha\beta \\ \alpha\beta & \beta^2 \end{array} \right).$$
where $\theta(t)$ is the Heaviside step function. Then the full phonon-dressed polariton Green’s function $\hat{G}(t)$, which is related to the polarization matrix \( \sum \) via

\[
\hat{P}(t) = \hat{Y} \hat{G}(t) \hat{Y}^{-1},
\]

satisfies the following Dyson’s equation

\[
\hat{G}(t) = \hat{G}^{(0)}(t) + \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \hat{G}^{(0)}(t - t_1) \hat{\Sigma}(t_1 - t_2) \hat{G}(t_2). \tag{S82}
\]

Note that this equation is equivalent to the perturbation series Eq. (S77). Here, the self energy $\hat{\Sigma}$ is represented by all possible connected diagrams such as the 2nd and 4th order diagrams sketched in Fig. S6, which are given by the following expressions:

\[
\hat{\Sigma}(t - t') = \hat{Q} \hat{G}^{(0)}(t - t') \hat{Q} \hat{D}(t - t') + \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \hat{Q} \hat{G}^{(0)}(t - t_1) \hat{Q} \hat{G}^{(0)}(t_1 - t_2) \hat{Q} \hat{G}^{(0)}(t_2 - t') \hat{Q}
\]

\[
\times [D(t - t_2)D(t_1 - t') + D(t - t')D(t_1 - t_2)] + \ldots \tag{S83}
\]

Equations (S82) and (S83) are exact provided that all the connected diagrams are included in the self energy. No approximations have been used so far.

In the case of isolated (phonon-decoupled) polariton states, all of the matrices are diagonal and the problem reduces to the IB model for each polariton level, having an exact analytic solution which we exploit in our approximation. For the two phonon-coupled polariton states treated here, the exact solvability is hindered by the fact that the matrices $\hat{Q}$ and $\hat{G}^{(0)}(t)$ do not commute for any finite time $t$. However, in the timescale $|\omega_1 - \omega_2| t \ll 1$, Eq. (S80) may be approximated as $\hat{G}^{(0)}(t) \approx \theta(t) e^{-\omega_1 t} \hat{F}$ and thus $\hat{G}^{(0)}(t)$ approximately commutes with $\hat{Q}$, so for example,

\[
\hat{Q} \hat{G}^{(0)}(t - t_1) \hat{Q} \hat{G}^{(0)}(t_1 - t_2) \hat{Q} \hat{G}^{(0)}(t_2 - t') \hat{Q} \approx \hat{Q} \hat{G}^{(0)}(t - t_1) \theta(t - t_1) \theta(t_1 - t_2) \theta(t_2 - t'), \tag{S84}
\]

using $\hat{Q}^2 = \hat{Q}$. Clearly, this approximation is valid if $\tau_C \gg \tau_B$. In this case we obtain

\[
\hat{\Sigma}(t) = \hat{Q} \begin{pmatrix} \Sigma_1(t) & 0 \\ 0 & \Sigma_2(t) \end{pmatrix}, \tag{S85}
\]

where $\Sigma_j(t)$ is the self energy of an isolated polariton state $j$, which contributes to the corresponding IB model problem

\[
G^{IB}_j(t) = G_j^{(0)}(t) + \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 G_j^{(0)}(t - t_1) \Sigma_j(t_1 - t_2) G^{IB}_j(t_2), \tag{S86}
\]

having the following exact solution:

\[
G^{IB}_j(t) = G_j^{(0)}(t) e^{K(t)}, \tag{S87}
\]

where the cumulant $K(t)$ is given by Eq. (S21). Equation (S86) then allows us to find the self energies in frequency domain:

\[
\Sigma_j(\omega) = \frac{1}{G^{(0)}_j(\omega)} - \frac{1}{G^{IB}_j(\omega)}, \tag{S88}
\]

where $\Sigma_j(\omega)$, $G_j^{(0)}(\omega)$, and $G^{IB}_j(\omega)$ are the Fourier transforms of $\Sigma_j(t)$, $G_j^{(0)}(t)$, and $G^{IB}_j(t)$, respectively. The full matrix Green’s function (and hence the polarization) is then obtained by solving Dyson’s equation (S82) in frequency domain:

\[
\hat{G}(\omega) = \left[ 1 - \hat{G}^{(0)}(\omega) \hat{\Sigma}(\omega) \right]^{-1} \hat{G}^{(0)}(\omega), \tag{S89}
\]
replacement $\alpha$ in Eq. (S90) with its symmetrized version $\bar{\alpha}$. Note that the first and last matrices of Eq. (S91) arise as a result of the replacement of the adjusted Hamiltonian $\hat{\Sigma}$ with $\hat{\Sigma}/\bar{\alpha}$. As in Eq. (S81), we can express the Fourier transform of the polarization as

$$g \hat{P}(\omega) = \begin{pmatrix} e^{-S/2} & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \bar{\alpha} & \bar{\beta} \\ -\bar{\beta} & \bar{\alpha} \end{pmatrix} \hat{G}(\omega) \begin{pmatrix} \bar{\alpha} & -\bar{\beta} \\ \bar{\beta} & \bar{\alpha} \end{pmatrix} \begin{pmatrix} e^{S/2} & 0 \\ 0 & 1 \end{pmatrix},$$

where the matrices containing $\bar{\alpha}$ and $\bar{\beta}$ diagonalize a symmetrized Hamiltonian $\hat{H}_{JC}$:

$$\hat{H}_{JC} = \begin{pmatrix} \bar{\alpha} & \bar{\beta} \\ -\bar{\beta} & \bar{\alpha} \end{pmatrix} = \begin{pmatrix} \bar{\alpha} & 0 \\ 0 & \bar{\beta} \end{pmatrix} \begin{pmatrix} \omega_1 & 0 \\ 0 & \omega_2 \end{pmatrix} \begin{pmatrix} \bar{\alpha} & -\bar{\beta} \\ \bar{\beta} & \bar{\alpha} \end{pmatrix}.$$

Note that the first and last matrices of Eq. (S91) arise as a result of the replacement of the adjusted Hamiltonian in Eq. (S90) with its symmetrized version $\hat{H}_{JC}$. We see that $\hat{G}(\omega)$ in Eq. (S91) is the analog of Eq. (S81) with a replacement $\alpha \rightarrow \bar{\alpha}$, $\beta \rightarrow \bar{\beta}$, $\omega_1 \rightarrow \bar{\omega}_1$, $\omega_2 \rightarrow \bar{\omega}_2$.

For $P_{XX}(\omega)$ and $P_{CC}(\omega)$ the solution Eq. (S91) gives the following simple explicit expressions:

$$P_{XX}(\omega) = \frac{\alpha^2 \bar{G}_{1}(\omega) + \bar{\beta}^2 \bar{G}_{2}(\omega)}{D(\omega)}, \quad P_{CC}(\omega) = \left( \frac{\alpha^2}{G_{1}^{IB}(\omega)} + \frac{\bar{\beta}^2}{G_{2}^{IB}(\omega)} \right) \frac{\bar{G}_{1}(\omega) \bar{G}_{2}(\omega)}{D(\omega)},$$

where

$$D(\omega) = \alpha^2 \bar{G}_{1}(\omega) \bar{G}_{1}^{IB}(\omega) + \bar{\beta}^2 \bar{G}_{2}(\omega) \bar{G}_{2}^{IB}(\omega).$$

FIG. S7. Absorption spectra for $g = 50 \mu eV$, $T = 5 K$, and zero detuning, calculated in the LN approach with $L = 15$ (red thick solid lines), NN approach with $L = 1$ (black thin solid lines), long-time analytic approximation (blue dashed lines) and refined analytics (green dotted line). Other parameters used: $\Omega_X = 1329.6 \text{meV}$, $\gamma_X = 2 \mu eV$, $\Omega_C = \Omega_X + \Omega p$ with $\Omega p = -49.8 \mu eV$, and $\gamma_C = 30 \mu eV$.

where $\hat{G}^{(0)}$ and $\hat{\Sigma}$ are given, respectively, by Eqs. (S80) and (S84), with self energy components provided via Eq. (S88) by the IB model solution Eq. (S77).

An obvious drawback of the above analytic model is that it does not show any phonon-induced renormalization of the exciton-cavity coupling due to the interaction with the phonon bath. This is a consequence of the present approach not properly taking into account the cumulative effect of self-energy diagrams of higher order, for which the approximate commutation of matrices $\hat{Q}$ and $\hat{G}^{(0)}(t)$ is not valid. But we know from the IB model that its exact solution in the form of a cumulant includes a nonvanishing contribution of all higher-order diagrams of the self energy series (for realistic phonon parameters of semiconductor quantum dots). This significant problem can, however, be easily healed through use of the large time asymptotics obtained in Sec. . We introduce by hand one minor correction: we replace the exciton-cavity coupling $g$ in the bare JC Hamiltonian by the renormalized coupling strength $ge^{-S/2}$ in the following way

$$H_{JC} = \begin{pmatrix} \omega_X & g \\ g & \omega_C \end{pmatrix} \rightarrow \begin{pmatrix} \omega_X & ge^{-S} \\ ge^{-S} & \omega_C \end{pmatrix}.$$

As in Eq. (S81), we can express the Fourier transform of the polarization as

$$P(\omega) = \begin{pmatrix} e^{-S/2} & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \bar{\alpha} & \bar{\beta} \\ -\bar{\beta} & \bar{\alpha} \end{pmatrix} \hat{G}(\omega) \begin{pmatrix} \bar{\alpha} & -\bar{\beta} \\ \bar{\beta} & \bar{\alpha} \end{pmatrix} \begin{pmatrix} e^{S/2} & 0 \\ 0 & 1 \end{pmatrix},$$
FIG. S8. (a) Excitonic linear polarization and (b) absorption for $g = 50 \mu eV$, $T = 5 K$, and nonzero detuning, calculated in the $LN$ approach with $L = 15$ (red thick solid lines), $NN$ approach with $L = 1$ (black thin solid lines), analytic approximation (blue dashed lines) and refined analytics (green dotted line). Other parameters used: $\Omega_X = 1329.6 \text{ meV}$, $\gamma_X = 2 \mu eV$, $\Omega_C = 1329.45 \text{ meV}$, and $\gamma_C = 30 \mu eV$.

FIG. S9. As Fig. S8 but for $T = 50 K$.

and $\bar{G}_j^{(0)}(\omega)$ and $\bar{G}_j^{\text{IB}}(\omega)$ are, respectively, the Fourier transform of $\bar{G}_j^{(0)}(t) = \theta(t)e^{-i\Omega_j t}$ and $\bar{G}_j^{\text{IB}}(t) = \bar{G}_j^{(0)}(t)e^{\mathbf{K}(t)}$.

Figures S7, S8(b) and S9(b), as well as Fig. 1(b) of the main text, demonstrate a very good agreement between the refined analytic solution and the exact result provided by the full $LN$ approach (with $L = 15$). In addition to the case of zero detuning at low temperature ($T = 5 K$) presented in Fig. S7, we also show in Figs. S8 and S9 both low and high temperature results for a non-zero detuning of 0.1 meV (the exact parameters are given in the captions).