Spontaneous emission from a quantum dot in a structured photonic reservoir: phonon-mediated breakdown of Fermi's golden rule: supplementary material

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Published 29 April 2015

This document provides supplementary information to "Spontaneous emission from a quantum dot in a structured photonic reservoir: phonon-mediated breakdown of Fermi's golden rule," <http://dx.doi.org/10.1364/optica.2.000434>. First, we show the photon and photon spectral function in the frequency and time domains, using the simple cavity structures introduced in the main text. Second, we show a derivation of the phonon-modified spontaneous emission (SE) rate, equation (2) in our main paper. Third, we derive the cavity-induced SE rate from a previous polaron master equations using cavity-QED equations, and show direct agreement with our results in the high Q limit (i.e., with $\tilde{\gamma}^{\text{mean}}$ in the main paper). Last, we discuss how one can include the influence from a coherent pump field into the quantum master equation and incoherent scattering terms, while accounting for both photon and phonon bath coupling.

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<http://dx.doi.org/10.1364/optica.2.000434.s001>

S1. CAVITY PHOTON AND PHONON BATH FUNCTIONS

For phonon bath temperatures of 4 K and 40 K, Figs. S1(a) and S1(b) show the phonon correlation function versus time and frequency, respectively. For simplicity, a polaron shift Δ_P (defined below in Section S2) is implicitly absorbed in the definition of ω_x , and we also define a phonon correlation function that decays to zero, through $C'_{\text{pn}}(\tau) = e^{\phi(\tau)} - 1$. All phonon parameters and $\phi(\tau)$ are given in our main paper. The time evolution of the real part of the phonon correlation function shows that typical phonon correlation times are very fast ($\tau_{\text{pn}} \leq 3$ ps). For comparison, we also show a photon correlation function in Figs. S1(c) and S1(d), for the three different values of κ used in our paper: $\kappa = 2.4$ meV (thick dark line), 0.6 meV (thin dark line), and 0.06 meV (thin light line). The cavity bath correlation functions $J_{\text{ph}}(t)$ are oscillatory functions damped at the cavity decay rate, with an oscillation frequency is determined by the QD-cavity detuning Δ_{xc} , and we show the case for zero detuning. From equation (2) in our paper, and Fig. S1 below, we expect that (i) phonons should not influence the SE rate γ in

a strongly damped cavity and (ii) phonons will reduce the SE rate to its mean-field limit [1] ($\langle B \rangle^2 \gamma$) only for a weakly damped cavity. Significant deviations from these two limits occur when the damping times of the photon and phonon correlation functions are comparable. This is shown explicitly in Fig. 2 of the main text using the cavity spectral functions shown below. It is also shown explicitly in Fig. 3 for the slow-light photonic crystal waveguide.

S2. DERIVATION OF PHONON-MODIFIED SPONTANEOUS EMISSION RATE

Theoretical descriptions of electron-phonon scattering in QDs systems range from the independent Boson model [2], correlation expansion [3], perturbative master equations (MEs) [4], to polaron MEs [5–8], variational MEs [9] and real-time path integrals [10]. In our Letter we introduce a self-consistent ME approach with both phonon and photon reservoirs included. We model the QD as a two-level system interacting with an inhomogeneous semiconductor based photonic reservoir and

Fig. S1. Phonon and photon bath function in time and frequency domain (a) Time evolution of the real part of phonon bath correlation function, $C'_{pn}(t)$ for $T = 4$ K (solid line) and 40 K (dashed line). (b) Corresponding phonon bath function $\text{Re}[C'(\omega)]$. (c) Time evolution of real part of photon bath correlation function, $J_{ph}(t)$ for cavities with $\kappa = 0.06$ meV (thin light, $Q \approx 23000$), $\kappa = 0.6$ meV (thin dark, $Q \approx 2300$) and $\kappa = 2.4$ meV (thick dark, $Q \approx 600$) at $\Delta_{xc} = 0$ meV. (d) Corresponding $J_{ph}(\omega)$, where ω_0 is ω_c .

an acoustic phonon bath. Assuming a QD of dipole moment $\mathbf{d} = d\mathbf{n}_d$ and position at $\mathbf{r}_d$, the total Hamiltonian of the coupled system in a frame rotating at the QD exciton frequency ω_x , can be written as [11]

$$H = \hbar \int d\mathbf{r} \int_0^\infty d\omega \mathbf{f}^\dagger(\mathbf{r}, \omega) \mathbf{f}(\mathbf{r}, \omega) - [\sigma^+ e^{i\omega_x t} \int_0^\infty d\omega \mathbf{d} \cdot \mathbf{E}(\mathbf{r}_d, \omega) + \text{H.c.}] + \sum_q \hbar \omega_q b_q^\dagger b_q + \sigma^+ \sigma^- \sum_q \hbar \lambda_q (b_q^\dagger + b_q), \quad (\text{S1})$$

where σ^+/σ^- are the Pauli operators of the exciton (electron-hole pair), $b_q/b_q^\dagger$ are the annihilation and creation operators of the acoustic phonon reservoir, and the exciton-phonon coupling strength λ_q is assumed to be real. The operators $\mathbf{f}/\mathbf{f}^\dagger$ are the boson field operators of the photon reservoir and these satisfy the usual commutation rules for boson operators. The interaction between the QD and the photonic reservoir is written using the dipole and the rotating wave approximation. The electric-dipole operator $\mathbf{E}(\mathbf{r}, \omega)$ is given by $\mathbf{E}(\mathbf{r}, \omega) = i \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}'; \omega) \sqrt{\frac{\hbar}{\pi \epsilon_0}} \epsilon_I(\mathbf{r}', \omega) \mathbf{f}(\mathbf{r}', \omega)$ [11], where $\mathbf{G}(\mathbf{r}, \mathbf{r}'; \omega)$ is the electric field Green's function for the medium and $\mathbf{E}(\mathbf{r}, \omega)$ satisfies the Kramers-Kronig relations, with a complex dielectric constant $\epsilon = \epsilon_R + i\epsilon_I$.

To include phonon interactions nonperturbatively, we perform the polaron transform on the Hamiltonian H given by $H' \rightarrow e^P H e^{-P}$, where $P = \sigma^+ \sigma^- \sum_q \frac{\lambda_q}{\omega_q} (b_q^\dagger - b_q)$ [7, 12]. H' consists of the following reservoir and interaction terms,

$$\begin{aligned} H' &= H'_R + H'_I, \\ H'_R &= \hbar \int d\mathbf{r} \int_0^\infty d\omega \mathbf{f}^\dagger(\mathbf{r}, \omega) \mathbf{f}(\mathbf{r}, \omega) + \sum_q \hbar \omega_q b_q^\dagger b_q, \\ H'_I &= -[B_+ \sigma^+ e^{i\omega_x t} \int_0^\infty d\omega \mathbf{d} \cdot \mathbf{E}(\mathbf{r}_d, \omega) + \text{H.c.}], \end{aligned} \quad (\text{S2})$$

where the coherent phonon bath displacement operators $B_\pm$ are defined as $B_\pm = \exp[\pm \sum_q \frac{\lambda_q}{\omega_q} (b_q - b_q^\dagger)]$. A polaron frequency

shift, $\Delta_P = \int_0^\infty d\omega \frac{J_{pn}(\omega)}{\omega}$ is implicitly absorbed in the definition of ω_x , where $J_{pn}(\omega)$ is the phonon spectral function [13]. For our calculations, we use the continuous form of the phonon spectral function $J_{pn}(\omega) = \alpha_p \omega^3 \exp[-\frac{\omega^2}{2\omega_b^2}]$ for longitudinal acoustic (LA) phonon interaction, resulting primarily from deformation potential coupling in InAs QDs [14]. In the above formula, ω_b is the phonon cutoff frequency and α_p is the exciton-phonon coupling strength [13].

We subsequently transform the Hamiltonian H' to the interaction picture using $\tilde{H}' \rightarrow U^\dagger(t) H' U(t)$ with $U(t) = \exp[-iH'_R t/\hbar]$. Assuming weak coupling with the photon bath, a time-convolutionless [15] polaron master equation for the QD reduced density operator ρ is then derived using second-order Born approximation in $\tilde{H}'_I$. The following interaction-picture ME is obtained,

$$\frac{\partial \tilde{\rho}(t)}{\partial t} = -\frac{1}{\hbar^2} \int_0^t d\tau \text{Tr}_{R_{ph}} \text{Tr}_{R_{pn}} \{ [\tilde{H}'_I(t), [\tilde{H}'_I(t-\tau), \tilde{\rho}(t) \rho_R]] \}, \quad (\text{S3})$$

where $\tilde{\rho}$ is the reduced density operator of the QD in the interaction picture and $\text{Tr}_{R_{ph(pn)}}$ denotes trace with respect to the photon (phonon) reservoirs which are assumed to be statistically independent, $\rho_R = \rho_{R_{ph}} \rho_{R_{pn}}$ [16]. The density operator of the photonic reservoir $\rho_{R_{ph}}$ is assumed to be initially in thermal equilibrium. We use the bath approximation, $\text{Tr}_{R_{ph}}[\mathbf{f}(\mathbf{r}, \omega), \mathbf{f}^\dagger(\mathbf{r}', \omega')] = [\tilde{n}(\omega) + 1] \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega')$ and $\text{Tr}_{R_{ph}}[\mathbf{f}^\dagger(\mathbf{r}, \omega), \mathbf{f}(\mathbf{r}', \omega')] = \tilde{n}(\omega) \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega')$ and consider the zero temperature limit ($\tilde{n}(\omega) = 0$). Using the relation $\int d\mathbf{s} \epsilon_I(\mathbf{s}, \omega) \mathbf{G}(\mathbf{r}, \mathbf{s}, \omega) \mathbf{G}^*(\mathbf{s}, \mathbf{r}'; \omega) = \text{Im}[\mathbf{G}(\mathbf{r}, \mathbf{r}'; \omega)]$, transforming to the Schrödinger picture, and carrying out the trace over the photon [17] and phonon reservoir, we derive the following generalised ME: $\frac{\partial \rho}{\partial t} = \mathcal{L}_{ph}(\rho)$, where

$$\begin{aligned} \mathcal{L}_{ph}(\rho) &= \int_0^t d\tau \int_0^\infty d\omega J_{ph}(\omega) [-C_{pn}(\tau) \sigma^+ \sigma^- e^{i\Delta_x \tau} \rho + C_{pn}^*(\tau) \\ &\times \sigma^- \rho \sigma^+ e^{-i\Delta_x \tau} + C_{pn}(\tau) \sigma^- \rho \sigma^+ e^{i\Delta_x \tau} - C_{pn}^*(\tau) \rho \sigma^+ \sigma^- e^{-i\Delta_x \tau}], \end{aligned} \quad (\text{S4})$$

and $\Delta_x = \omega_x - \omega$. The photon-reservoir spectral function $J_{ph}(\omega)$, is given by $J_{ph}(\omega) = \frac{\mathbf{d} \cdot \text{Im}[\mathbf{G}(\mathbf{r}_d, \mathbf{r}_d; \omega)] \cdot \mathbf{d}}{\pi \hbar \epsilon_0}$ and the phonon bath correlation function $C_{pn}(\tau)$ is defined as $C_{pn}(\tau) = e^{[\phi(\tau) - \phi(0)]}$, where $\phi(t) = \int_0^\infty d\omega \frac{J_{pn}(\omega)}{\omega^2} [\coth(\hbar\omega/2k_B T) \cos(\omega t) - i \sin(\omega t)]$. A simple expression for the phonon-modified SE decay rate can be derived from the real part of $\mathcal{L}_{ph}$, so that $\text{Re}(\mathcal{L}_{ph})$ reduces to the Lindblad form, $\tilde{\gamma}(t) L[\sigma^-]$, where $\tilde{\gamma}$ is the phonon-modified SE decay rate of a QD given by

$$\tilde{\gamma}(t) = 2 \int_0^t \text{Re}[C_{pn}(\tau) J_{ph}(\tau)] d\tau, \quad (\text{S5})$$

where $J_{ph}(\tau) = \int_0^\infty d\omega J_{ph}(\omega) e^{i(\omega_x - \omega)\tau}$ is the photon bath correlation function, and $L[O] = \frac{1}{2}(2O\rho O^\dagger - O^\dagger O\rho - \rho O^\dagger O)$. Note that the imaginary part of $\mathcal{L}_{ph}$ yield Lamb shifts [13]. This is Equation (2) in our main paper. In absence of the phonon coupling, the SE decay rate of the QD in a structured photon reservoir reduces to $\gamma(t) = 2 \int_0^t \text{Re}[J_{ph}(\tau)] d\tau$. A similar expression for $\tilde{\gamma}$ was derived by McCutcheon and Nazir [18], who then use a free space photon reservoir function to show no phonon-modification to the SE rate.

S3. CORRESPONDENCE WITH PREVIOUS WORK IN THE MEAN FIELD CAVITY-QED REGIME: DERIVATION OF $\tilde{\gamma}_{\text{WEA}}^{\text{MEAN}}$ USING A POLARON CAVITY-QED MASTER EQUATION

Using the polaron transformed effective Lindblad master equation of an coupled QD-cavity system [13], an expression for the SE rate can be derived in weak excitation approximation (WEA) and bad cavity limit. The effective phonon master equation is defined through,

$$\frac{\partial \rho}{\partial t} = \frac{1}{i\hbar} [H_{\text{sys}}^{\text{eff}}, \rho] + \Gamma^{a^\dagger \sigma} L(a^\dagger \sigma^-) + \Gamma^{\sigma^+ a} L(\sigma^+ a) + \kappa L(a), \quad (\text{S6})$$

where a is the cavity lowering operator and $H_{\text{sys}}^{\text{eff}} = \hbar \Delta_{cx} a^\dagger a + \hbar \langle B \rangle g (a^\dagger \sigma^- + \sigma^+ a)$, in a frame rotating at the exciton frequency ω_x . The QD-cavity coupling strength is g and the cavity damping rate is κ and $\langle B \rangle = \langle B_+ \rangle = \langle B_- \rangle$ is the thermally averaged phonon bath displacement operator [12]. The cavity (exciton) feeding terms are $\Gamma^{a^\dagger \sigma^- / \sigma^+ a} = 2 \langle B \rangle^2 g^2 \text{Re}[\int_0^\infty d\tau e^{\mp i \Delta_{cx} \tau} (e^{\phi(\tau)} - 1)]$ [13].

In the weak excitation approximation, the Bloch equations for $\langle a \rangle$ and $\langle \sigma^- \rangle$ derived from Equation (S6) are given by

$$\begin{aligned} \frac{d \langle a \rangle}{dt} &= - \left(i \Delta_{cx} + \frac{\kappa + \Gamma^{\sigma^+ a}}{2} \right) \langle a \rangle - i \langle B \rangle g \langle \sigma^- \rangle, \\ \frac{d \langle \sigma^- \rangle}{dt} &= - \frac{\Gamma^{a^\dagger \sigma}}{2} \langle \sigma^- \rangle - i \langle B \rangle g \langle a \rangle. \end{aligned} \quad (\text{S7})$$

The coupled equations can be expressed in a matrix form as $\mathbf{V} = \mathbf{M}\mathbf{V}$, where $\mathbf{V} = [\langle a \rangle; \langle \sigma^- \rangle]$. In the bad cavity limit, the real part of the eigenvalues of $\mathbf{M}$ give the decay rates $\langle a \rangle$ and $\langle \sigma^- \rangle$. The SE rate of the QD is twice the decay rate of $\langle \sigma^- \rangle$ and is given by

$$\begin{aligned} \tilde{\gamma}_{\text{WEA}} &= \text{Re} \left[- \left(\frac{\Gamma^{a^\dagger \sigma^-} + \kappa_1}{2} + i \Delta_{cx} \right) \right. \\ &\quad \left. - \sqrt{\left(\frac{\Gamma^{a^\dagger \sigma^-} + \kappa_1}{2} + i \Delta_{cx} \right)^2 - 4 \left(\langle B \rangle^2 g^2 + \frac{\Gamma^{a^\dagger \sigma^-}}{2} \left(\frac{\kappa_1}{2} + i \Delta_{cx} \right) \right)} \right], \end{aligned} \quad (\text{S8})$$

where $\kappa_1 = \kappa + \Gamma^{\sigma^+ a}$. The above expression can be manipulated to give

$$\begin{aligned} \tilde{\gamma}_{\text{WEA}} &= \text{Re} \left[- \left(\frac{\Gamma^{a^\dagger \sigma^-} + \kappa_1}{2} + i \Delta_{cx} \right) \right. \\ &\quad \left. - \sqrt{\left(\frac{\Gamma^{a^\dagger \sigma^-} - \kappa_1}{2} - i \Delta_{cx} \right)^2 - 4 \langle B \rangle^2 g^2} \right], \end{aligned} \quad (\text{S9})$$

which, in the bad cavity limit ($\kappa \gg g/2$), can be further simplified by expanding the second term to the first power of $\frac{4 \langle B \rangle^2 g^2}{\left(\frac{\Gamma^{a^\dagger \sigma^-} - \kappa_1}{2} - i \Delta_{cx} \right)^2}$. The final simplified expression for the QD SE rate is

$$\tilde{\gamma}_{\text{WEA}}^{\text{bad}} = \Gamma^{a^\dagger \sigma^-} + 2 \langle B \rangle^2 g^2 \frac{\left(\frac{\kappa + \Gamma^{\sigma^+ a} - \Gamma^{a^\dagger \sigma^-}}{2} \right)}{\Delta_{cx}^2 + \left(\frac{\kappa + \Gamma^{\sigma^+ a} - \Gamma^{a^\dagger \sigma^-}}{2} \right)^2}. \quad (\text{S10})$$

Moreover, for $\kappa \gg \Gamma^{\sigma^+ a} - \Gamma^{a^\dagger \sigma^-}$,

$$\tilde{\gamma}_{\text{WEA}}^{\text{bad}} = \Gamma^{a^\dagger \sigma^-} + 2 \langle B \rangle^2 g^2 \frac{\left(\frac{\kappa}{2} \right)}{\Delta_{cx}^2 + \left(\frac{\kappa}{2} \right)^2}, \quad (\text{S11})$$

which is equivalent to $\tilde{\gamma}^{\text{mean}}$ in our paper. Using the same polaronic cavity-QED approach, an identical expression is derived in Ref. [8] in the large detuning limit ($\Delta \gg g$) by adiabatically eliminating the cavity. Our paper makes it clear that $\tilde{\gamma}_{\text{WEA}}^{\text{bad}}$, and indeed the entire polaron cavity-QED master equation, is only valid when $\kappa \ll 2\pi/\tau_{\text{pn}}$ (τ_{pn} is around 1 ps for InAs QDs).

S4. COMMENTS ABOUT HOW TO INCLUDE A COHERENT PUMP FIELD

The QD can be excited using a weak coherent pulse of the form $H_{\text{pump}} = \eta_x (\sigma^+ e^{-i\omega_L t} + \sigma^- e^{i\omega_L t})$, where η_x and ω_L are the amplitude and central frequency of the pump pulse. For resonant excitation $\omega_L = \omega_x$. Under this situation, the polaron-transformed system Hamiltonian, H' (Equation (S2)) has the form

$$\begin{aligned} H' &= H'_S + H'_R + H'_I, \\ H'_S &= \hbar \langle B \rangle \eta_x [\sigma^+ + \sigma^-], \\ H'_R &= \hbar \int d\mathbf{r} \int_0^\infty d\omega \mathbf{f}^\dagger(\mathbf{r}, \omega) \mathbf{f}(\mathbf{r}, \omega) + \Sigma_q \hbar \omega_q b_q^\dagger b_q, \\ H'_I &= - [B_+ \sigma^+ e^{i\omega_x t} \int_0^\infty d\omega \mathbf{d} \cdot \mathbf{E}(\mathbf{r}_d, \omega) + \text{H.c.}] + X_g \zeta_g + X_u \zeta_u, \end{aligned} \quad (\text{S12})$$

where $\zeta_g = \frac{1}{2}(B_+ + B_- - 2 \langle B \rangle)$ and $\zeta_u = \frac{1}{2i}(B_+ - B_-)$ are the phonon induced fluctuation operators [7] and X_g and X_u are defined through $X_g = \hbar \eta_x (\sigma^- + \sigma^+)$ and $X_u = i \hbar \eta_x (\sigma^+ - \sigma^-)$. Following the same steps as before (Section II), we subsequently transform the Hamiltonian H' to the interaction picture using $\tilde{H}' \rightarrow U^\dagger(t) H' U(t)$ where $U(t) = \exp[-i(H'_S + H'_R)t/\hbar]$. Assuming weak coupling with the photon bath, a time-convolutionless [15] polaron master equation for the QD reduced density operator ρ is then derived using second-order Born approximation in $\tilde{H}'_I$. After carrying out the trace over the photon and phonon baths, the final ME for the QD reduced density operator ρ in the Schrödinger picture is given by $\frac{d\rho}{dt} = \frac{1}{i\hbar} [H'_S, \rho] + \mathcal{L}_{\text{ph}}(\rho) + \mathcal{L}_{\text{pn}}(\rho)$. The phonon part $\mathcal{L}_{\text{pn}}(\rho)$ remains decoupled from the photon reservoir and, to a very good approximation, can be expressed through the following Lindblad terms (enhanced radiative decay and incoherent excitation [13]):

$$\mathcal{L}_{\text{pn}}(\rho) = \Gamma^{\sigma^-} L[\sigma^-] + \Gamma^{\sigma^+} L[\sigma^+] \quad (\text{S13})$$

where, $\Gamma^{\sigma^\pm} = 2 \langle B \rangle^2 \eta_x^2 \text{Re}[\int_0^\infty d\tau e^{\pm i(\omega_L - \omega_x)\tau} (e^{\phi(\tau)} - 1)]$. The photon part $\mathcal{L}_{\text{ph}}$ on the other hand is modified by the phonons and is given by

$$\begin{aligned} \mathcal{L}_{\text{ph}}(\rho) &= \int_0^t d\tau \int_0^\infty d\omega J_{\text{ph}}(\omega) [-C_{\text{pn}}(\tau) \sigma^+ \sigma^- (-\tau) e^{i\Delta_x \tau} \rho \\ &\quad + C_{\text{pn}}^*(\tau) \sigma^- \rho \sigma^+ (-\tau) e^{-i\Delta_x \tau} + C_{\text{pn}}(\tau) \sigma^- (-\tau) \rho \sigma^+ e^{i\Delta_x \tau} \\ &\quad - C_{\text{pn}}^*(\tau) \rho \sigma^+ (-\tau) \sigma^- e^{-i\Delta_x \tau}], \end{aligned} \quad (\text{S14})$$

where the time-dependent operators $\sigma^\pm(-\tau) = e^{-iH'_S \tau/\hbar} \sigma^\pm e^{iH'_S \tau/\hbar}$. This indicates that the scattering rates are pump-field dependent in general and for strong pumps,

different dressed states ($\omega = \omega_x, \omega_x \pm 2\eta_x$) can sample different regions of the photonic LDOS [5, 9, 19]. Such behavior leads to asymmetric Mollow triplets [20] in the absence of phonons, if the Rabi strength is large enough to sample difference values and an asymmetric LDOS (see Ref. [21] for detailed discussions). Since we assume weak excitation, only the photonic LDOS around ω_x is sampled. Thus $\sigma^\pm(-\tau)$ is replaced by $\sigma^\pm$, which reduces $\mathcal{L}_{\text{ph}}(\rho)$ to Equation Eq. (S4), the form used in the main manuscript. The QD can also be excited by a weak incoherent pump by using higher states of the QD. The precise form of the pump is not important for the study of SE[8], as long as it weakly excites the system. However, one can add in a pump field using our general approach as we show above.

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