Supporting information for:

Bright phonon-tuned single-photon source

Simone Luca Portalupi,[†] Gaston Hornecker,[‡] Valérian Giesz,[†] Thomas Grange,[‡]
Aristide Lemaître,[†] Justin Demory,[†] Isabelle Sagnes,[†] Norberto D.
Lanzillotti-Kimura,[†] Loïc Lanco,^{†,¶} Alexia Auffèves,^{*,‡} and Pascale Senellart^{*,†,§}

*CNRS-LPN Laboratoire de Photonique et de Nanostructures, Route de Nozay, 91460
Marcoussis, France, CEA/CNRS/UJF joint team “Nanophysics and Semiconductors”,
Institut Néel-CNRS, BP 166, 25 rue des Martyrs, 38042 Grenoble Cedex 9, France,
Département de Physique, Université Paris Diderot, 4 rue Elsa Morante, 75013 Paris,
France, and Département de Physique, Ecole Polytechnique, F-91128 Palaiseau, France*

E-mail: alexia.auffeves@grenoble.cnrs.fr; pascale.senellart@lpn.cnrs.fr

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*To whom correspondence should be addressed

[†]CNRS-LPN Laboratoire de Photonique et de Nanostructures, Route de Nozay, 91460 Marcoussis, France

[‡]CEA/CNRS/UJF joint team “Nanophysics and Semiconductors”, Institut Néel-CNRS, BP 166, 25 rue des Martyrs, 38042 Grenoble Cedex 9, France

[¶]Département de Physique, Université Paris Diderot, 4 rue Elsa Morante, 75013 Paris, France

[§]Département de Physique, Ecole Polytechnique, F-91128 Palaiseau, France

Sample Fabrication

The sample was grown by molecular beam epitaxy with an adiabatic design^{S1}. In order to form the cavity, the thickness W of the GaAs (AlAs) layers is modified adiabatically with respect to the nominal thickness $W_0 = \lambda/4n$, decreasing it progressively by: 0.01, 0.07 (0.04), 0.15 (0.12), 0.19 (0.18), 0.15 (0.12), 0.07 (0.04), 0.01 percent. The central GaAs layer, with decreased thickness of 0.19 %, is the one where the InGaAs quantum dot is inserted. 18 (32) pairs of nominal $\lambda/4n$ GaAs/AlAs Bragg mirrors are placed on top (bottom) of this adiabatic cavity. The *in-situ* lithography technique was then used to fabricate micropillar cavities centered within 50 nm from a selected QD, presenting a cavity mode spectrally matched to the X resonance within 1 nm^{S2}.

Setup description, calibration and brightness

The setup used for the measurements allows us to perform micro-photoluminescence, reflectivity and correlation measurements. Light emission is collected from the top of the micropillar with a 0.54 NA aspheric lens. This signal is sent to a Hanbury-Brown and Twiss setup composed by a non-polarizing 50 : 50 beam splitter, two spectrometers equipped with single photons avalanche detectors. A polarizer and a halfwave plate are placed in the detection line in order to select the emission from one QD axis and to align the incident linear polarization with the direction of maximum efficiency of the spectrometer. The measurement of the maximum brightness is derived by taking the sum of the count rates of the two orthogonal QD axes. In order to quantify the brightness of the source all components of the experimental setup have been carefully calibrated giving the results summarized in table I.

Detection is accounting for the response of the full detection system, i.e. spectrometer, in- and out- coupling lenses and single photons avalanche detector efficiency. It is measured by sending a laser signal of known power and repetition rate of 82 MHz into the detection

Table S1: Transmission and overall efficiency of all setup components.

Component	Transmission/Efficiency	Error bar
Detection	8.8 %	± 0.5 %
HBT NPBS	51 %	± 2 %
Cryostat window	98 %	± 1 %
Polarizer	90 %	± 2 %
Long pass filter	98 %	± 1 %
Half wave plate	98 %	± 1 %
Collection NPBS	40 %	± 2 %
Lens	95 %	± 1 %

line. The ratio between the known photon flux and the detected number of counts gives an efficiency of around 8.8 ± 0.5 %. The overall setup efficiency is $\eta = 1.44 \pm 0.1\%$.

Spontaneous emission of a quantum dot coupled to a continuum of phonons in a cavity

Different approaches have been used to model the spontaneous emission of a quantum dot in a cavity. Of significant interest are the methods based on the second-order Born approximation^{S4} and the time-convolutionless approach^{S5}. As we restrict ourselves to the weak coupling regime, we settle for a simpler model based on first-order perturbation theory. The quantum dot (QD) is modeled as a two-level system coupled to a continuum of longitudinal acoustic (LA) phonons on one hand and to a continuum of electromagnetic (EM) modes on the other hand. The considered EM modes are whether the cavity mode, that leads to an emission in the direction parallel to the pillar axis, or the plane modes, that leads to a perpendicular emission (see Fig.1a in the text). The coupling to the phonons is treated exactly using the independent bosons model. The interaction with the EM field is treated in the rotating wave approximation (RWA) with the Jaynes-Cummings Hamiltonian. The total Hamiltonian of our system is given by

$$H = \hbar\omega_0\sigma_+\sigma_- + H_{\text{ph}} + H_{\text{EM}}, \quad (1)$$

where ω_0 is the frequency of the bare exciton; $\sigma_+ = \sigma_-^\dagger = |e\rangle\langle g|$ where $|g\rangle$ and $|e\rangle$ are respectively the ground and excitonic states of the quantum dot; H_{ph} and H_{EM} stands for the parts of the Hamiltonian involving respectively phononic and EM modes. The phononic part reads

$$H_{\text{ph}} = \int d\omega_{\mathbf{q}} \rho_{\text{ph}}(\omega_{\mathbf{q}}) \hbar \omega_{\mathbf{q}} b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \int d\omega_{\mathbf{q}} \rho(\omega_{\mathbf{q}}) \hbar M(\omega_{\mathbf{q}}) \sigma_+ \sigma_- (b_{\mathbf{q}} + b_{\mathbf{q}}^\dagger), \quad (2)$$

where $b_{\mathbf{q}}$ and $\rho_{\text{ph}}(\omega_{\mathbf{q}})$ are the ladder operator and the density of states (DOS) of the LA phonon mode of wavevector $\mathbf{q}$ respectively; $M(\omega_{\mathbf{q}})$ is the deformation potential coupling between this phonon mode and the exciton. We consider a linear dispersion relation for the LA phonons, and spherical wave functions for the electron (hole) with a gaussian shape: $\psi_{e/h}(\mathbf{r}) = (4\pi\sigma_{e/h}^2)^{-3/4} e^{-\mathbf{r}^2/(8\sigma_{e/h}^2)}$. We denote c_s the speed of sound, ρ_m the mass density, $D_{e/h}$ the deformation potential of the electron/hole, and V the volume of the crystal. The DOS and coupling terms read respectively:

$$\rho_{\text{ph}}(\omega_{\mathbf{q}}) = \frac{V\omega_{\mathbf{q}}^2}{2\pi^2 c_s^3}, \quad (3)$$

$$M(\omega_{\mathbf{q}}) = \sqrt{\frac{\omega_{\mathbf{q}}}{2\hbar c_s^2 \rho_m V}} \cdot \left| D_e e^{-\sigma_e^2 \omega^2 / c_s^2} + D_h e^{-\sigma_h^2 \omega^2 / c_s^2} \right|, \quad (4)$$

The parameters used are: $c_s = 5000$ m/s, $\rho_m = 5320$ kg/m³, $D_e = D_h = 9$ eV, $\sigma_e = \sigma_h = 3$ nm, $V_c = 7.6$ μm^3 , where $D_e, D_h, \sigma_e, \sigma_h$ and V_c have been adjusted to fit the experimental data.

On the other hand, the interaction with the EM modes reads:

$$H_{\text{EM}} = \sum_{\nu=\parallel, \perp} \left[\int d\omega S_\nu(\omega) \hbar \omega_\nu a_\nu^\dagger(\omega) a_\nu(\omega) + \int d\omega S_\nu(\omega) \hbar g_\nu(\omega) (\sigma_+ a_\nu(\omega) + \sigma_- a_\nu^\dagger(\omega)) \right], \quad (5)$$

in which the index $\nu = \parallel, \perp$ stands for the two possible channels for the QD relaxation:

either the cavity mode or the plane modes. $a_\nu(\omega)$ is the ladder operator for the photon mode of pulsation ω . The Jaynes-Cummings coupling $g_\nu(\omega)$ is given by

$$g_\nu(\omega) = d \sqrt{\frac{\omega}{2\hbar\epsilon_0 V_\nu}}, \quad (6)$$

where d is the electric dipole of the quantum dot and V_ν is either the effective volume of the cavity mode or the diverging quantization volume of the free field. The DOS for the cavity mode is given by a Lorentzian of width $\kappa = \omega_c/Q = 2\pi c\gamma_C/\lambda_C^2$, with $Q = 12000$:

$$S_\parallel(\omega) = \frac{1}{\pi} \frac{\kappa/2}{(\kappa/2)^2 + (\omega - \omega_c)^2}. \quad (7)$$

$\omega_c = 2\pi c/\lambda_C$ is the central frequency of the cavity. The DOS for the plane modes is taken as the DOS in free space:

$$S_\perp(\omega) = \frac{\omega^2 V_f}{\pi^2 c^3}. \quad (8)$$

Emission in both channels is well described in first order perturbation theory, such that the spontaneous emission rate $w_\nu(\omega)$ in each mode of pulsation ω and the total rate per channel Γ_ν are

$$w_\nu(\omega) = g_\nu^2(\omega) S_{QD}(\omega), \quad (9)$$

$$\Gamma_\nu = \int d\omega g_\nu^2(\omega) S_\nu(\omega) S_{QD}(\omega), \quad (10)$$

where we have defined the QD spectrum (in absence of any cavity) as

$$S_{QD}(\omega) = \int d\tau e^{-i\omega\tau} \langle \sigma_+(\tau) \sigma_-(0) \rangle, \quad (11)$$

The expression of the correlator $\langle \sigma_+(\tau) \sigma_-(0) \rangle$ is known and derive from an exact resolution of the independent boson model (see^{S3}):

$$\langle \sigma_+(\tau) \sigma_-(0) \rangle = e^{i\omega_{QD}\tau} e^{-\Phi(\tau)}, \quad (12)$$

$$\Phi(\tau) = \int_0^{+\infty} d\omega_{\mathbf{q}} \rho_{ph}(\omega_{\mathbf{q}}) \left(\frac{M(\omega_{\mathbf{q}})}{\omega_{\mathbf{q}}} \right)^2 \left[N(\omega_{\mathbf{q}})(1 - e^{i\omega_{\mathbf{q}}\tau}) + (N(\omega_{\mathbf{q}}) + 1)(1 - e^{-i\omega_{\mathbf{q}}\tau}) \right], \quad (13)$$

where $N(\omega_{\mathbf{q}}) = 1/(e^{\beta\hbar\omega_{\mathbf{q}}} - 1)$ and $\omega_{QD}^X = 2\pi c/\lambda_X = \omega_0 - \int d\omega_{\mathbf{q}} \rho_{ph}(\omega_{\mathbf{q}}) M(\omega_{\mathbf{q}})/\omega_{\mathbf{q}}$ is the frequency of the exciton shifted by the so-called polaron shift. This expression can be rewritten

$$\Phi(\tau) = \int_{-\infty}^{+\infty} d\omega f(\omega)(1 - e^{-i\omega\tau}), \quad (14)$$

$$f(\omega) = \rho_{ph}(\omega) \left(\frac{M(\omega)}{\omega} \right)^2 (N(|\omega|) + \theta(\omega)). \quad (15)$$

The spectrum of the quantum dot is then given by

$$\begin{aligned} S_{QD}(\omega) &= \int d\tau \exp \left[\int d\omega f(\omega) (e^{-i\omega\tau} - 1) \right] e^{-i(\omega - \omega_{QD})\tau} \\ &= 2\pi Z \delta(\omega_{QD} - \omega) + Z \int d\tau \left[\exp \left(\int d\omega f(\omega) e^{-i\omega\tau} \right) - 1 \right] e^{-i(\omega - \omega_{QD})\tau} \\ &= S_{QD}^{ZPL}(\omega) + S_{QD}^{PSB}(\omega), \end{aligned} \quad (16)$$

with $Z = e^{-\int d\omega f(\omega)}$. The first term in (16) corresponds to the zero phonon line, the second takes into account all orders of phonon assisted mechanisms. To account for the finite lifetime of the exciton, the delta function in the ZPL spectrum can be replaced by a lorentzian. This artificial broadening has been used in figures 3., using the experimental width of the ZPL.

Four relaxation channels for the QD can now be defined, the rate of each being

$$\Gamma_{\nu}^{ZPL/PSB} = \int d\omega \rho_{\nu}(\omega) g_{\nu}^2(\omega) S_{QD}^{ZPL/PSB}(\omega). \quad (17)$$

The effective fraction of emission coupled to the mode is

$$\beta^{eff} = \frac{\Gamma_{\parallel}^{ZPL} + \Gamma_{\parallel}^{PSB}}{\Gamma_{tot}}, \quad (18)$$

where Γ_{tot} is the total relaxation rate, *i.e.* it involves the four relaxation processes mentioned above. We define β^{ZPL} and β^{PSB} the same way:

$$\beta^{ZPL} = \frac{\Gamma_{\parallel}^{ZPL}}{\Gamma_{tot}}, \quad \beta^{PSB} = \frac{\Gamma_{\parallel}^{PSB}}{\Gamma_{tot}}. \quad (19)$$

The effective Purcell factor is then given by

$$\begin{aligned} F_P^{eff} &= \frac{\beta^{eff}}{1 - \beta^{eff}} = \frac{\Gamma_{\parallel}^{ZPL} + \Gamma_{\parallel}^{PSB}}{\Gamma_{\perp}} \\ &= \frac{\tau_X^0}{\tau_X} - 1, \end{aligned} \quad (20)$$

where τ_X (τ_X^0) is the decay time of the quantum dot in the pillar cavity (planar cavity). The usual nominal Purcell factor is

$$F_P = \frac{\Gamma_{\parallel}^{ZPL}}{\Gamma_{\perp}^{ZPL}}. \quad (21)$$

Photon correlations in the cavity mode at high power

Here we estimate $g_{C,C}^{(2)}(0)$ when the pump power is high and multiple excitons can be excited. In this regime, the cavity mode peak is fed by the exciton as well as the biexciton. We make the approximation that the total spectrum of the radiative cascade is the sum of the spectrum of the biexciton–exciton transition and exciton recombination. Each line is weighted by the probability of each event. The spectrum of the biexciton–exciton transition is equal to the spectrum of an exciton recombination with a shifted ZPL frequency $\omega_{QD}^{XX} = 2\pi c/\lambda_{XX}$. Both the X and XX spectra are then computed using the model presented in last section. The relative weight of these two events is obtained using :

$$\frac{I_{XX}}{I_X} = \frac{P_{XX} \cdot \beta_{XX}^{ZPL}}{P_X \cdot \beta_X^{ZPL}}, \quad (22)$$

where I_X (I_{XX}) is the measured intensity of the X (XX) peak, which is identified with the intensity of the zero phonon lines, P_X and P_{XX} are the probabilities of the two events and β_X^{ZPL} and β_{XX}^{ZPL} are the fractions of emission coupled to the mode computed with our model. The number of excitons excited inside the dot is assumed to follow a poissonian distribution. P_X (P_{XX}) correspond to the probability that more than one (two) exciton is excited. It is then given by

$$P_X = 1 - e^{-\bar{N}}, \quad (23)$$

$$P_{XX} = 1 - (1 + \bar{N})e^{-\bar{N}}, \quad (24)$$

where $\bar{N}$ is the mean number of excitons in the dot. The ratio of the last two equations gives

$$\frac{\bar{N}e^{-\bar{N}}}{1 - e^{-\bar{N}}} + \frac{P_{XX}}{P_X} - 1 = 0, \quad (25)$$

which can be numerically solved to determine $\bar{N}$, and then P_X and P_{XX} .

The coincidence probability is defined as

$$g_{C,C}^{(2)}(0) = \frac{\int_{-T/2}^{+T/2} dt \int_{-T/2}^{T/2} dt \langle a_1^\dagger(t) a_2^\dagger(t + \tau) a_2(t + \tau) a_1(t) \rangle}{\left| \int_{-T/2}^{T/2} dt \langle a_1^\dagger(t) a_1(t) \rangle \right|^2}, \quad (26)$$

where the ladder operator a_i corresponds to the field in the two branches of the HBT interferometer filtered at the cavity frequency and T is the period between two pulses. The numerator of (26) corresponds to the probability that both the biexciton and exciton emit a photon in the cavity peak in a row, which we identify to $P_{XX} \cdot \beta_{XX}^{PSB} \cdot \beta_X^{PSB}$. The integral in the denominator is the total number of photons sent to the interferometer at the frequency of the cavity peak, given by $P_X \cdot \beta_X^{PSB} + P_{XX} \cdot \beta_{XX}^{PSB}$. Thus, the coincidence probability is

$$g_{C,C}^{(2)}(0) = \frac{P_{XX} \cdot \beta_{XX}^{PSB} \cdot \beta_X^{PSB}}{(P_X \cdot \beta_X^{PSB} + P_{XX} \cdot \beta_{XX}^{PSB})^2}. \quad (27)$$

Note that for very low excitation power, $P_X \approx \bar{N} \ll 1$, $P_{XX} \approx \bar{N}^2 \ll P_X$ so that $g_{C,C}^{(2)}(0) = \frac{\beta_{XX}^{PSB}}{\beta_X^{PSB}}$ is independent on the excitation power as experimentally observed in figure 4a.

Other examples of phonon tuned sources

Figure S1: We present the brightness of other deterministically coupled QD cavity devices as a function of detuning. Both devices present a Q factor of 3000. For QD2 (left), the measured effective Purcell factor at resonance is $F_P^{eff} = 2$. For QD9 (right), it is $F_P^{eff} = 3.5$. Both devices present increased brightness beyond the cavity linewidth shown by the blue dotted line. All observations are well reproduced with Purcell enhanced phonon assisted mechanisms (red lines). Parameters used: for QD2, $V_c = 20\mu\text{m}^3$, $\sigma_e = \sigma_h = 8\text{nm}$, for QD9, $V_c = 15\mu\text{m}^3$, $\sigma_e = \sigma_h = 7\text{nm}$.

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