Reducing Phonon-Induced Decoherence in Solid-State Single-Photon Sources with Cavity Quantum Electrodynamics

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Solid-state emitters are excellent candidates for developing integrated sources of single photons. Yet, phonons degrade the photon indistinguishability both through pure dephasing of the zero-phonon line and through phonon-assisted emission. Here, we study theoretically and experimentally the indistinguishability of photons emitted by a semiconductor quantum dot in a microcavity as a function of temperature. We show that a large coupling to a high quality factor cavity can simultaneously reduce the effect of both phonon-induced sources of decoherence. It first limits the effect of pure dephasing on the zero-phonon line with indistinguishabilities above 97% up to 18 K. Moreover, it efficiently redirects the phonon sidebands into the zero-phonon line and brings the indistinguishability of the full emission spectrum from 87% (24%) without cavity effect to more than 99% (76%) at 0K (20K). We provide guidelines for optimal cavity designs that further minimize the phonon-induced decoherence.

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Indistinguishable single photons are the building blocks of optical quantum computation protocols and quantum networks [1–4]. This has motivated great efforts to develop devices generating on-demand indistinguishable single photons, using solid-state emitters such as diamond color centers [5,6], molecules [7], or semiconductor quantum dots (QDs) [8–13]. In QDs, understanding the extrinsic sources of decoherence such as spin and charge noise [14] has recently enabled impressive progress in the performances of these sources [12,13]. Yet, acoustic phonons generally remain an intrinsic and limiting source of dephasing.

Indeed, acoustic phonons are responsible for two kinds of dephasing processes. First, acoustic phonons induce a rapid and partial decay of coherence [15–17]. This non-Markovian dephasing dynamics is the time-domain counterpart of the emitter spectrum consisting of a sharp zero-phonon line (ZPL) sitting on top of a broad phonon sideband (PSB) [18–20]. Second, acoustic phonons can assist virtual transitions towards higher energy levels, resulting in a Markovian pure dephasing of the ZPL [21]. Such effects impose two severe limitations to obtain indistinguishable photons: (i) to work at low temperatures, typically below 10 K for QDs, and (ii) to use spectral postselection of the ZPL. Indeed, even at zero temperature, phonon emission processes result in the presence of a PSB on the low energy side, fundamentally limiting the indistinguishability. In practice, the indistinguishability has been measured to rapidly drop with temperature even with spectral selection [22], and without it remains further away from unity [23].

In typical self-assembled QDs, the emission fraction into the ZPL, η_{ZPL}, typically represents 90% of the emission at 4 K, a fraction that rapidly drops with temperature. Moreover, the photons emitted in the PSB are essentially incoherent due to their broadband nature with respect to the natural linewidth. In a Hong-Ou-Mandel (HOM) experiment, only the fraction η_{ZPL} from each of the two photons gives rise to an interference. As a consequence, the indistinguishability (i.e., the degree of two-photon interference) decreases as $\sim\eta_{\mathrm{ZPL}}^2$ [24]. To avoid this problem, a spectral filtering of the collected emission spectrum, corresponding to a spectral postselection, is generally applied to spectrally select the ZPL while suppressing the phonon sideband emission [9–13,22,25]. Yet, such postselection of the emission constitutes a fundamental limitation since it automatically reduces the efficiency of the single-photon source. Besides, the finite efficiency of the spectral filter further decreases the efficiency, shifting away from the sought deterministic characteristic, i.e., the generation of exactly one indistinguishable photon per pulse. It is hence of prime interest to develop solid-state sources of highly indistinguishable photons that do not require postselection.

Here, we study experimentally and theoretically the indistinguishability of photons emitted by QDs inserted in micropillar cavities as a function of temperature. We demonstrate that cavity-quantum electrodynamics (cavity-QED) effects can be used to efficiently reduce the different phonon-induced dephasing effects. Strong Purcell enhancement of the radiative emission rate has long been thought of as a promising strategy to reduce the influence of pure dephasing, but experimental demonstrations have been limited by the fact that increasing the temperature also modifies the QD-cavity coupling [26,27]. Here, we use an electrical tuning of the QD transition to keep the ZPL resonant to the cavity mode and observe indistinguishabilities of the ZPL above 97% up to 18 K. We also show that the coupling to a high quality factor cavity efficiently redirects the PSB emission into the ZPL, leading to a significant increase of η_{ZPL}. At 0K (20K), this redirection allows bringing the indistinguishability from 87% (24%)—limited by phonon emission processes—to more than 99% (76%). Our theoretical study, based on nonequilibrium Green's function calculations, predicts a further improvement of the indistinguishability of postselection-free single photons by reducing the cavity linewidth for a fixed Purcell factor.

The QD-cavity devices are obtained through a deterministic positioning of the QD in the cavity using the *in situ* lithography technique [28]. The micropillar cavity is electrically contacted with four ridges to a surrounding circular structure where the electrical connection is performed [see the sketch in Fig. 1(a)]. The applied bias voltage allows for an accurate control on the detuning of the QD-cavity resonance [12,29] by means of the confined Stark effect. In this Letter, we study two different QD-cavity devices, dubbed device 1 and device 2. The cavity linewidth is 90 μeV (110 μeV) for device 1 (2). In both cases the electronic transition corresponds to a neutral exciton constituted of two orthogonal and linear dipoles showing a fine structure splitting of $\Delta_{\mathrm{FSS}} = 3\ \mu$eV (10 μeV) for device 1 (2). The cavities are excited with a resonant laser using a standard confocal resonant fluorescence setup in a cross-polarization configuration to remove the resonant laser light [10,12,30]. Such strict resonant excitation suppresses the effect of time jitter in the exciton creation process [31], thus isolating the effect of phonon-induced decoherence on indistinguishability.

The sample temperature is controlled between 9 and 20 K in a closed-circuit gas exchange helium cryostat. Measuring the effect of cavity QED on the photon indistinguishability when changing the temperature requires an additional knob to keep the QD-cavity detuning constant since the QD transition shifts faster with temperature than the cavity resonance [26,27]. Here, the QD-cavity resonance is maintained with temperature through the confined Stark effect. Figures 1(b) and 1(c) show two resonant fluorescence maps as a function of the applied bias and energy at 20 and 9 K, respectively, for device 2: a continuous-wave laser resonantly excites one of the cavity modes (energy indicated with the vertical solid line), and the cross-polarized resonant fluorescence is collected. The voltage control allows tuning the QD resonance (the dashed line) and reaching the QD-cavity resonance at 20 and 9 K for an applied bias of −26 and −376 mV, respectively. Note that above 20–23 K, depending on the device, the resonance is reached for a

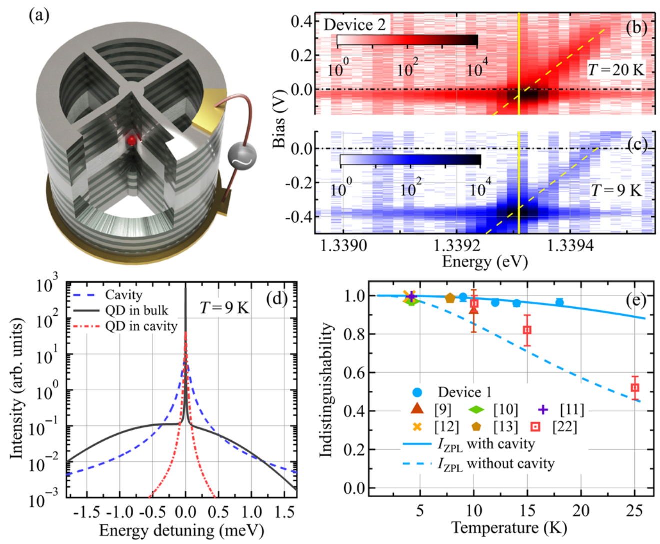

FIG. 1. (a) Schematic of the device. (b),(c) Resonant resonant fluorescence maps as a function of energy and bias voltage on device 2 at temperatures 20 and 9 K, respectively. The cavity and QD spectral positions as a function of the voltage are indicated with full and dashed yellow lines, respectively. (d) Comparison of the calculated emission spectra of a QD in a bulk photonic environment (the black solid line) and coupled resonantly to a cavity (the red dashed-dotted line) at 9 K. The cavity spectrum is also indicated (the blue dashed line). (e) Indistinguishability of the ZPL as a function of temperature. The measurements (device 1) are shown in blue circles. Calculations for the device 1 (the solid line) and without cavity-QED effects (the dashed line) are shown. We also indicate measurements reporting high indistinguishability of the ZPL, as well as the temperature dependence recently reported in Ref. [22] in the absence of the Purcell effect (the red squares).

positive bias for which a non-negligible current is observed, changing the measurement conditions.

Figure 1(c) evidences an asymmetric calculated spectrum at resonance at 9 K, arising from the PSB emission [18–20]. This strongly non-Lorentzian shape is reproduced using a non-Markovian modeling of the phonon bath [32–47]. Accounting for the measured spectrum at 9 K at the QD-cavity resonance allows us to extract the potential deformation constant of $D = 14$ eV for the excitonic transition (see the Supplemental Material [48]). To highlight the influence of the cavity on the spectrum, the exciton spectra are compared in Fig. 1(d) for a bulklike emission (i.e., without cavity) and for the QD in cavity using a logarithmic scale. In the bulk case, the sharp ZPL (width of 0.7 μeV) sits on meV-broad PSBs. When coupling the QD to the cavity, two important changes are observed: (i) the Purcell enhancement of the radiative emission broadens the ZPL, and (ii) the phonon PSBs that fall outside the cavity spectrum (the dashed blue line) have their emission strongly suppressed. The PSB emission is hence effectively redirected towards the ZPL: the reduction of the efficiency of phonon-assisted emission, together with the increase of the ZPL one, strongly modifies the ratio between these two components of the spectrum. The fraction emitted in the ZPL at 9 K is calculated to increase from $\eta_{\mathrm{ZPL}} = 0.81$ in bulk to $\eta_{\mathrm{ZPL}} = 0.98$ in the cavity. The spectra measured at various temperatures are found to be in good agreement with these theoretical expectations (see Fig. S6 in the Supplemental Material [48]). Both cavity-QED effects significantly influence the indistinguishability.

We measure the two-photon coalescence in a fiber based HOM setup. The excitation consists of resonant pulses of 15 ps temporal length with a repetition rate of 82 MHz and a pump power corresponding to the π pulse of the QD transition, in resonance with the cavity mode for every temperature. The single-photon emission is sent to a fiber based Mach-Zehnder interferometer, with a relative temporal delay between the two arms of 12.2 ns corresponding to the repetition rate of the laser. The output signals from the HOM interferometer are temporally correlated to reconstruct the two-photon coincidence histogram which renders the value of the indistinguishability [48].

Using an etalon to filter the PSB (spectral width of 10 μeV, transmission efficiency of 70%), we first study the indistinguishability of photons emitted by the ZPL as a function of temperature [Fig. 1(e)]. The indistinguishably of the ZPL hardly decreases from $0.993^{+0.006}_{-0.024}$ at 9 K to $0.973^{+0.021}_{-0.010}$ at 18 K. Indistinguishabilities above 90% of the ZPL have been reproducibly reported in the 4–10 K range [see the symbols in Fig. 1(e)] [9–13,22]. At higher temperatures, the indistinguishability of the ZPL has been reported to decrease rapidly. This trend was observed both in QDs in microlenses [22] [the red squares in Fig. 1(e)] and in QDs in photonic waveguides where no acceleration of spontaneous emission is implemented [51]. The robust two-photon interferences with the temperature observed here can be attributed to the strong Purcell effect provided by the cavity coupling [26,27,52]. Indeed, for a two-level system with a Markovian dynamics, the indistinguishability is given by [53,54]

$$I_{\mathrm{ZPL}} = \gamma/(\gamma + \gamma^*), \tag{1}$$

where $\gamma = \hbar/T_1$ is the natural linewidth (with T_1 being the exciton lifetime) and $\gamma^* = 2\hbar/T_2^*$ is the additional broadening of the ZPL due to pure dephasing. Increasing the temperature degrades the indistinguishability of the emitted single photons by reducing the pure dephasing time T_2^*. Yet, the Purcell effect has a positive impact on the indistinguishability by accelerating the single-photon radiative lifetime (T_1). Indeed, by placing the QD in resonance with the microcavity, the radiative rate γ increases from $\gamma = \gamma_0$ to $\gamma = (1 + F_{\mathrm{eff}})\gamma_0$, where F_{eff} is the effective Purcell factor describing the reduction from the nominal Purcell factor ($F_P = 24$ for device 1) due to the presence of the PSBs [45,48]. F_{eff} decreases with increasing temperature, from 15 at 0 K to 11 at 20 K. As negligible pure dephasing is observed at 8 K, the broadening of the ZPL is attributed to the phonon-assisted virtual transitions with the higher excitonic states. Such a mechanism involves a scattering from thermal acoustic phonons into another mode, so that the resulting temperature dependence can be approximated by $\gamma^*(T) = \alpha n(\epsilon_p)[n(\epsilon_p) + 1]$, where $n(\epsilon_p)$ is the Bose factor at the wavelength of maximally coupled acoustic phonons ϵ_p [55]. We take $\alpha = 0.1$ μeV and $\epsilon_p = 1$ meV to account for the observed dependence of the ZPL indistinguishability. Figure 1(e) also shows the corresponding indistinguishability without any Purcell effect as $I^0_{\mathrm{ZPL}} = \gamma_0/(\gamma_0 + \gamma^*)$ (the dashed line), evidencing the strong enhancement of the ZPL indistinguishability induced by the large Purcell effect in device 1.

In the following we study the photon indistinguishability without any spectral postselection. For a QD in bulk, the calculated indistinguishability is shown in Fig. 2 (thick solid line) as a function of temperature without spectral postselection and accounting for the pure dephasing measured in Fig. 1(e). It drops to 0.24 as temperature reaches 20 K. This has been recently confirmed by similar measurements on QDs subject to negligible cavity-QED effects [56]. Even in the absence of pure dephasing (the dotted line), the indistinguishability would fall to 0.42 at 20 K, owing to the rapid reduction of the ZPL fraction ($\eta_{\mathrm{ZPL}} = 0.64$ at 20 K) (see Fig. S5 in the Supplemental Material [48]) with increasing temperature. To calculate the indistinguishability in the presence of cavity QED, the coupling to the non-Markovian phonon bath and to the cavity mode are simultaneously accounted for in the calculation of photon correlation [24,57–59]. We use a recently developed non-equilibrium Green's function approach [60]. Its accuracy has been shown by the excellent agreement with the exact diagonalization reported at zero temperature by Kaer *et al.* [57]. Yet, by contrast, our model is numerically tractable at finite temperatures. The calculated indistinguishability is shown in Fig. 2 for the two different devices. The same QD parameters for PSBs and pure dephasing are considered for the two devices [48]. A strong enhancement of indistinguishability with respect to the bulk is predicted due to the combination of (i) the Purcell effect overcoming pure dephasing of the ZPL [as already discussed in Fig 1(e)], and (ii) the cavity redirection of the PSB emission into the ZPL (the relative contribution of these two effects is shown in the Supplemental Material [48]). Such redirection appears first at 0 K, where a close to unity indistinguishability is calculated with cavity QED, when it should be limited to ~0.87 due to phonon emission processes. Considering finite temperatures, the nonpostselected indistinguishability is expected to decrease from ~0.92 (~0.89) at 9 K to ~0.74 (~0.79) at 18 K for device 1 (device 2). The measured indistinguishabilities are also shown as a function of temperature for device 1 (the crosses) along with device 2 (the circles), using voltage to recover the QD-cavity resonance at each temperature. Note that the values are corrected from a residual laser background to extract the emitted photon indistinguishability (see the Supplemental Material [48]). A very good agreement is found between the calculated and measured indistinguishabilities. Note that the dispersion in the measured points mainly arises from the ±5 μeV uncertainty on the QD-cavity detuning, which influences the indistinguishability [48]. Moreover, device 1 does not provide a higher indistinguishability than device 2, in spite of a much larger nominal Purcell factor (24 and 8, respectively). This is due to a larger residual cavity mode splitting, which results in a larger detuning between the exciton and the monitored cavity mode under the cross-polarization resonant excitation scheme adopted here [48]. Such a limitation would vanish under side excitation, and we plot the expected value for a negligible cavity mode splitting, where the indistinguishability at 20 K is predicted to further increase from 0.70 to 0.88 for device 1, as shown by the thin solid line in Fig. 2.

As discussed below, such enhancement of indistinguishability with respect to a bulklike photonic environment [23] results from the small linewidth of the cavities with respect to the PSB width. Indeed, as the PSBs that fall outside the cavity spectrum are efficiently redirected into the ZPL [Fig. 1(d)], a large fraction of incoherent emission is transformed into a coherent one. This second cavity-QED effect differs fundamentally from a filtering effect, for which the PSB emission fraction would simply be lost: the overall cavity-QED effect not only increases the photon indistinguishability but also increases the overall source efficiency. Note that cavity-QED funneling into the ZPL was recently demonstrated for nitrogen-vacancy centers [61,62], but with no measure yet of the full spectrum indistinguishability.

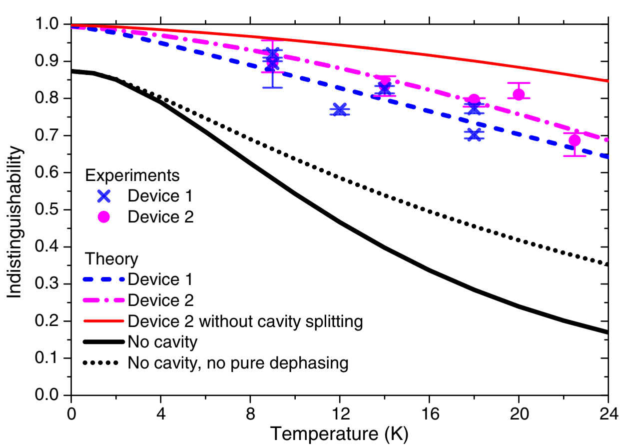

FIG. 2. Indistinguishability of the nonpostselected photons as a function of temperature. The measurements are shown in symbols for the two different QD-cavity devices. Calculations accounting for the coupling to the cavity of device 1 (device 2) are indicated in dashed (dashed-dotted) lines. Calculations displayed by the red thin solid line predict the indistinguishability in the case where the cavity splitting is suppressed (with the other parameters being identical to those of device 1). Calculations in the absence of cavity are also shown (the black thick solid line). The black dotted line represents the calculations in bulk in the absence of pure dephasing of the ZPL.

As shown above, a high Purcell factor allows us to reduce the effect of ZPL pure dephasing. However, as shown by Kaer and co-workers [24,57], reaching the QD-cavity strong coupling regime results in a decrease of indistinguishability due to phonon-assisted scattering processes between the dressed polariton states. Here, we instead propose to reduce the cavity linewidth to enhance the effect of the PSBs being redirected into the ZPL. In Fig. 3(a), we show the calculated indistinguishability as a function of the cavity linewidth for a fixed nominal Purcell factor of 24 (as reported in device 1). The QD-cavity detuning is set to its optimal zero value. Interestingly, the indistinguishability is found to further increase when the cavity linewidth is reduced below the cavity linewidths of our devices ($\sim$100 μeV). This effect is all the more important as the temperature increases since $(1-\eta_{ZPL})$, the nominal fraction of emission in the PSB, increases. The indistinguishability is hence predicted to reach a value of 0.94 at 20 K, and it can be increased up to 0.995 at 4 K. At very small cavity linewidths, the indistinguishability reaches a maximum before decreasing. Indeed, the strong QD-cavity coupling regime is reached for $\kappa \lesssim 15$ μeV and Eq. (1) is then no longer valid [54]. Figure 3(b) shows the corresponding fraction of emission into the cavity mode. Except for very small linewidths below $\sim$20 μeV, the probability of

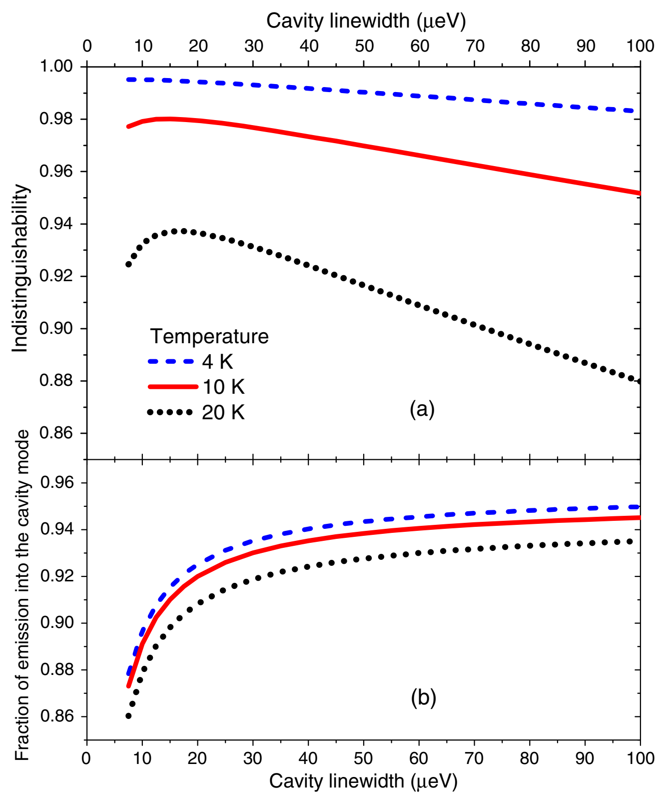

FIG. 3. (a) Calculated indistinguishability of the nonpostselected photons as a function of the cavity linewidth, for a fixed nominal Purcell factor of $F = 24$, for temperatures of 4, 10, and 20 K. (b) Corresponding fraction of the emission into the cavity mode for the same three temperatures. The QD and the cavity mode are assumed to be on resonance.

emission into the cavity mode stays almost constant. Hence, the reduction of the cavity linewidth enables us to enhance the indistinguishability without significantly decreasing the brightness. Experimentally, this can be realized by changing the cavity length of the micropillars from a single wavelength λ to multiple λ wavelengths, while keeping the same Bragg mirrors and hence almost the same Purcell factor (see Fig. S10 in the Supplemental Material [48] for the influence of the Purcell factor). This strategy can also be implemented in other cavity-QED systems where low cavity linewidths and large Purcell effects can be combined, such as open-access cavities [63–65] and photonic crystal cavities [66–69].

Our findings apply to a large variety of solid-state single-photon emitters suffering from phonon-induced decoherence. Various approaches are explored to obtain bright sources of indistinguishable photons from solid-state emitters, some based on broadband spontaneous emission suppression [70,71], others, like the one here, based on the acceleration of spontaneous emission into a narrow-band mode. We have shown that the latter approach allows not only for high extraction efficiency but also for reducing the effect of both phonon decoherence processes. Indistinguishable photons from the ZPL are shown to be less sensitive to thermally activated pure dephasing, thanks to the large Purcell acceleration of photon emission. Concurrently, indistinguishable photon generation without spectral postselection can be obtained at low temperature with high quality factor cavities by effectively reducing the phonon-assisted emission processes.

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Note added.—Recently, a related theoretical investigation was reported by Iles-Smith *et al.* [72].

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