Hybrid resonators for light trapping and emission control

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Chapter 7

Observation of strong and tunable fluorescence enhancement in hybrid systems

The promise of hybrid antenna-cavity systems relies to a large extent on the prediction that one can benefit simultaneously from the high cavity quality factor and the low cavity mode volume, leading to highly enhanced local density of states (LDOS). In this chapter, we experimentally verify this symbiotic behaviour. We study hybrid antenna-cavity systems with fluorescent quantum dots positioned at the antenna apex. Fluorescence spectra show asymmetric Fano lineshapes at the hybridized cavity mode frequencies that go from a strong peak to a dip, depending on antenna size. We discuss the role of LDOS and collection efficiency on the emission spectra and show that these measurements can be used to obtain a lower bound on the LDOS in these systems, which can be as much as 14 times higher at the hybrid mode than for the bare antenna. Finally, a study of quantum dot decay rates reveals a strong increase that correlates with the antenna resonance.
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7.1 Introduction

Hybrid antenna-cavity systems are attractive candidates for single-photon sources, quantum logic gates or particle sensors, owing to their potential for large enhancement of the local density of states (LDOS) and their large bandwidth tuneability. Several theoretical studies have predicted very high LDOS in a variety of antenna-cavity geometries [93, 104–106, 109]. In Chapters 2 and 3, we discussed under which conditions these systems can work symbiotically and achieve such high LDOS, and when, in contrast, destructive interference causes LDOS to be reduced. Despite their great potential for light-matter interaction, to date very few experimental works exist that study LDOS in these systems, possibly due to the difficulty of deterministically integrating an emitter, an antenna and a cavity. Experiments have been done on hybrid structures where intrinsic fluorescence of the cavity material was used to study emission [110], or a gain material was embedded inside the cavity [97, 98]. To benefit from the plasmonic field enhancement, however, the emitters need to be placed at an antenna ‘hot-spot’, which was not the case for these studies. Without this field enhancement, the antenna merely acts as a source of loss, invariably decreasing LDOS.

In this chapter, we present the first experimental study of fluorescent emitters placed at the antenna hotspot inside a hybrid antenna-cavity system. We study fluorescence spectra and emitter decay rates from colloidal quantum dots, observing strongly enhanced emission at the hybridized cavity modes. Spectra assume asymmetric Fano-type lineshapes, matching LDOS spectra predicted by coupled-oscillator theory and full-wave simulations. We explain our results by showing that, for broadband emitters coupled to narrow photonic resonances, emission spectra trace the product of collection efficiency and LDOS, which indeed shows excellent agreement with the data. Our results give a lower bound on the LDOS increase at the hybrid resonance — relative to the bare antenna — which can assume values up to 14 for the smallest antennas used. Furthermore, we find fluorescent decay rates to be strongly increased in the hybrid system, with average decay rates peaked near the antenna resonance condition. These results constitute the first observation of strong LDOS enhancements in a hybrid antenna-cavity system.

Section 7.2 describes our experimental methods. The obtained fluorescence spectra and their analysis are then discussed in Section 7.3, after which we discuss the results from the quantum dot decay rate measurements in Section 7.4.

7.2 Experimental methods

Samples consisted of silicon nitride (Si$_3$N$_4$) disks with 5 different diameters between 3960 and 4120 nm. Each disk contained an antenna placed 300 nm...
from the disk edge. We used 5 antenna lengths between 88 and 168 nm. Colloidal quantum dots (Invitrogen Qdot 800 ITK Organic) were positioned in an area approximately 120 nm in diameter around the antenna apex. Images of a typical hybrid and of the quantum dots (QD) near the antenna are shown in Fig. 5.5c and Fig. 5.12b, respectively. Further details of the sample fabrication are given in Chapter 5. Note that this procedure leads to multiple QDs present per antenna. We measure one structure for each of the 25 different combinations of disk size and antenna size. Out of these 25 measurements, three were discarded because the fluorescence signal was too low.

Figure 7.1: Experimental setup. (a) Sketch of the fluorescence microscope used in this study. Samples are illuminated by a pump laser, and fluorescence is collected either on a camera, a fiber-coupled spectrometer or two avalanche photodiodes (APDs) for lifetime measurements. The pump and fluorescence are linearly polarized by polarizers (LP). (b-c) Camera images of a hybrid, pumped by the 532 nm laser (b) or by the 640 nm laser (c). The antenna is located at the bottom of the disk, and is clearly visible in fluorescence. In both cases, the pump is defocused, illuminating an area much larger than the disk. For 532 nm illumination, strong background fluorescence from the disk is visible, which is absent for 640 nm illumination. (d) Fluorescence spectra pumped by the 532 nm (blue) and 640 nm (red) laser. The 532 nm spectrum is taken from a part of the disk edge without quantum dots, and shows a broad peak from the intrinsic silicon nitride fluorescence. Sharp peaks correspond to enhanced fluorescence at the cavity modes. The 640 nm spectrum is taken from a single quantum dot on the disk, not near the antenna. No silicon nitride fluorescence is visible. The 532 nm (640 nm) spectrum is normalized to its maximum of $1.4 \cdot 10^3$ (1.8 $\cdot 10^3$) counts, acquired with 10 (20) MHz pulsed excitation at $\approx 0.1$ mW (1.8 mW) average power and 60 s (120 s) integration time.

To investigate the modification of spontaneous emission by the quantum dots, we perform fluorescence spectroscopy and lifetime measurements. Fig. 7.1a shows the experimental setup, which is an adapted version of the
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setup reported in [270]. The sample is illuminated through an objective (Olympus MPlan IR, 100x, NA 0.95) by a pulsed pump laser. In the detection path, this pump is removed by a dichroic beamsplitter and a long-pass filter. Pump and detection polarization are controlled by linear polarizers. Fluorescence is sent either to a camera, to a fiber-coupled spectrometer (Andor Shamrock 303i, equipped with an iVac DR316B-LDC-DD detector) or two avalanche photodiodes (APD) in a Hanbury-Brown-Twiss configuration [271]. The spectrometer uses a multimode fiber of 10 \( \mu \text{m} \) core size, which translates to a detection area of \( \sim 1 \mu \text{m} \) on the sample. We performed broadband (\( \Delta \lambda = 314 \text{ nm}, \text{ resolution } \sim 0.4 \text{ nm } = 0.2 \text{ THz} \)) or high-resolution (\( \Delta \lambda = 62 \text{ nm}, \text{ resolution } \sim 0.1 \text{ nm } = 0.05 \text{ THz} \)) measurements using a 300 lines/mm or a 1200 lines/mm spectrometer grating, respectively. By correlating the arrival times of photons on the APDs (Excelitas SPCM-AQRH-14-FC single photon counting modules, 350 ps timing resolution) to the timing of the pump pulses, we can measure emitter lifetime [272]. Fig. 5.7b shows a typical decay curve of a single quantum dot on a glass substrate. Correlating the events on the two APDs to each other corresponds to a measurement of the second order correlation function \( g^{(2)} \), which shows antibunching if a single emitter is probed [233]. APD counts and pump pulse events are recorded on a Becker & Hickl DPC 230 timing card.

To avoid background signal from intrinsic silicon nitride fluorescence [273], we use a pump laser of 640 nm wavelength (PicoQuant LDH-P-C-640B pulsed diode laser, <500 ps pulse width) with repetition rate variable between 2.5 and 40 MHz. Unless stated otherwise, measurements were done with the pump beam focused on the sample. Fig. 7.1b and c show fluorescence images of a hybrid pumped by a 532 nm pulsed laser (Time-Bandwidth Products, 10 MHz, <10 ps pulse width) and by the 640 nm laser, respectively. With the 532 nm illumination, we observe fluorescence from the entire disk. With the 640 nm laser, in contrast, only fluorescence from the location of the antenna (and quantum dots) is visible. Fig. 7.1d compares spectra obtained under 532 nm and 640 nm illumination. In the first case, we see a broad spectrum peaking around 650 nm, typical of silicon nitride fluorescence [273]. In the second case, this background is absent and only a quantum dot fluorescence peak at \( \sim 780 \text{ nm} \) is visible.

All spectra are acquired using a 60 or 120 second camera integration time, with the laser set to a 20 MHz repetition time. Background spectra, taken without pumping, are subtracted. Pixels with anomalously high counts (usually attributed to cosmic rays) are removed in post-processing. The high-resolution spectra may show intensity fringes due to an etalon effect in the camera chip itself, with amplitudes up to 50% of the signal. As they occur at a specific frequency, we remove them by suppressing the corresponding frequency components in a Fourier transform of the signal. Fluorescent decay

*Bandwidth \( \Delta \lambda \) and resolution specified at 780 nm wavelength.
7.3 Observation of LDOS boosts from hybrid emission spectra

7.3.1 Experimental results

Fig. 7.2: Polarization of QD emission. (a) Example of a typical emission spectrum, for QDs at the antenna apex. The broad QD emission peak is modulated with sharp Fano-type resonances. (b) Emission spectra recorded for in- and output polarization set to vertical-vertical (blue, same spectrum as shown in (a)), vertical-horizontal (purple), horizontal-horizontal (green) and horizontal-vertical (red). Fano resonances are only clearly visible for in- and outputs vertically polarized, i.e. along the antenna main axis. All spectra measured on the same hybrid, with antenna length 168 nm.

Fig. 7.2a shows a typical fluorescence spectrum from quantum dots at the antenna apex in a hybrid system. We recognize a broad fluorescence peak from the intrinsic QD emission spectrum. Remarkably, this emission spectrum is modulated at regular intervals by an asymmetric, Fano-type resonance. Emission can be reduced by as much as ~70% at the dips of these resonances. These lineshapes are strongly reminiscent of the asymmetric LDOS resonances predicted by coupled-oscillator theory in Chapter 3 and shown for example in Fig. 3.1b. The notion that hybrid LDOS could be the cause behind these lineshapes matches the fact that the resonances appear only when both pump and detection polarization are chosen along the antenna main axis, as shown in Fig. 7.2b. In fact, when detection polarization is chosen horizontally, i.e. along the antenna short axis, entirely different spectra are observed. These peak at a shorter wavelength, and no Fano resonances are observed. This corresponds to the fact that near the antenna apex, no significant LDOS enhancement is
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expected for emitters polarized transversely to the antenna main axis.

Figure 7.3: Emission spectra for different antenna lengths. (a-e) Broadband emission spectra from hybrids with equal disk size and 5 different antenna lengths L (indicated). The dispersive Fano resonances change gradually from a peak to a dip as L increases. (f-j) High-resolution emission spectra, zoomed in on the mode near 800 nm wavelength, for the same hybrids as in (a-e). The shape of the Fano resonances is more clearly visible. We show data (blue) and a fit (red).

If the resonances in the quantum dot spectra do indeed originate from the LDOS resonances in the hybrid system, we would expect from our calculations in Chapter 3 that the lineshape depends strongly on cavity-antenna detuning. Fig. 7.3a-e show broadband spectra for hybrids with 5 different antenna lengths, and consequently different cavity-antenna detunings. We indeed observe a gradual change from a resonance that is mostly peaked for short antennas (i.e. cavity modes far red-detuned) to complete destructive interference for an antenna length of 148 nm. At even larger length, the Fano lineshapes take on opposite asymmetry to that of the short antennas, i.e. first a peak, then a dip. This change of phase can be observed even more clearly in
the high-resolution spectra shown in Fig. 7.3f-j. This behaviour corresponds remarkably well to that of the Fano resonances in Fig. 3.1b.

To quantitate these results, we fit the resonances in high-resolution spectra with the expression for a Fano lineshape [12]

$$I(\omega) = |E_2 e^{i\theta} + E_1 \frac{\kappa/2}{-i\Delta + \kappa/2}|^2.$$  

(7.1)

Here, $\Delta$ and $\kappa$ are frequency detuning and linewidth, respectively, and $\theta$ is the Fano phase. Fig. 7.3f-j show examples of resonances near 800 nm wavelength, fitted with Eq. (7.1). For each hybrid, we apply the fit routine to three different modes between 720 and 800 nm wavelength. The resulting resonance wavelengths are visible in Fig. 7.4. We find that resonances shift linearly with disk diameter, further confirming that these correspond to the hybridized whispering-gallery modes (WGM). To first approximation, WGMs are waves that fit an integer times within an effective disk circumference. As such, their resonance wavelengths depend approximately linearly on disk diameter [274]. By comparing these resonance wavelengths to those obtained from simulations, we can estimate them to be the WGMs of azimuthal order $m_{\phi}$ 21, 22 and 23. Note that these disks are too small to support high-$Q$ modes of higher radial order than $m_r = 0$ in this wavelength range.

As we have seen in Chapter 6, hybrid linewidth depends strongly on antenna length. A similar dependence is visible for the Fano resonance linewidths, as can be seen in Fig. 7.5a. As for the linewidths measured in taper-coupled spectroscopy on larger disks, shown in Fig. 6.5, we observe a strong linewidth increase up to antenna lengths of 148 nm. At this length,
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Figure 7.5: Dependence of hybrid resonance properties on antenna length. (a) Fano resonance linewidth, showing a strong increase as the antenna is tuned to resonance (around 150 nm length). (b) Fano phase $\theta$ transitions from a peak ($\theta \approx 0$) for short antennas to a dip ($\theta \approx \pi$) for antennas at resonance. For the largest antennas, the resonance takes on a Fano shape with opposite asymmetry, i.e. $\theta < \pi$. (c) Hybrid boost factor is strongest for small antennas, where cavity modes are far from antenna resonance. All experimental data (blue markers) are obtained from fits as shown in Fig. 7.3. Results are compared to values from the LDOS (green line) profiles or collected LDOS (LDOSC, red dashed line) profiles predicted by coupled-oscillator theory, as well as LDOS obtained from finite element simulations on the full hybrid system (purple line). Theory and simulations are discussed in Section 7.3.2.

antennas are on resonance with the cavity modes and induce maximum broadening. When increasing length further, linewidth drops slightly as the antenna is brought away from resonance. A plot of Fano phase $\theta$ in Fig. 7.5b confirms that the behaviour seen in Fig. 7.3 is generic to all cavity modes. The resonances change from nearly Lorentzian peaks ($\theta \approx 0$) for short antennas, to dips ($\theta \approx \pi$) at antenna resonance, and to opposite asymmetry $\theta < \pi$ at 168 nm antenna length.

All our observations suggest that the resonances lineshapes in the emission spectra follow the LDOS spectra of the hybrid systems. It is therefore interesting to compare the peak heights of the resonances to the background at the same frequency. This ratio, which we call hybrid boost, would then measure the peak LDOS, or Purcell factor, of the hybrid mode relative to the LDOS provided by the bare antenna at that frequency. From Chapter 6, we know that the broadband LDOS resonance corresponds to the bare antenna mode. Fig. 7.5c shows this hybrid boost for different antenna lengths. We see that for antennas near resonance (lengths near 140 nm), hybrid peak heights are not far above the background. This is not surprising, as for these resonances $\theta \approx \pi$, i.e. they are dips rather than peaks. In contrast, for short antennas, peak
7.3 Observation of LDOS boosts from hybrid emission spectra

heights can be as much as 14 times higher than the background. Again, this trend qualitatively matches that of the LDOS peaks in Fig. 3.1b, which extend far above the broad antenna background only if the antenna is significantly detuned. It is tempting to ascribe the observed boosts directly to a boost in LDOS at the hybrid modes. However, it is not straightforward to see why emission spectra should follow LDOS. Therefore, a more subtle analysis is required to correctly interpret this data, which is the topic of the next section.

7.3.2 Relation between emission spectra and LDOS

The manner in which emitters report on LDOS depends strongly on the emitter bandwidth. We can distinguish two different regimes. The first is the case of emitters that are narrowband compared to the LDOS spectrum. At unit efficiency, each emitter produces one photon per excitation, with a fluorescent decay rate proportional to LDOS as dictated by Fermi’s golden rule [11]. Hence, intensity is independent of LDOS. This is the regime discussed in most literature concerning LDOS measurements, particularly in plasmonics [68, 270, 275]. If the emitters are very inefficient, intensity may report on LDOS through an effective increase in radiative efficiency [76]. The second regime, often referred to as the ‘bad emitter’ regime, is that of emitters with bandwidths much larger than the LDOS features. Particularly, to be in this regime the emitters are required to be individually broadband on time scales shorter than the fluorescence decay time. Hence, an emitter showing slow spectral diffusion [276, 277] or a polydisperse ensemble of individually narrowband emitters can be classified under the first regime. In this limit of broadband emitters, the decay rate averages over all decay channels (i.e. energies), while the emission spectrum shows differences that are proportional to LDOS. For efficient emitters, the total, spectrally integrated intensity remains independent of LDOS, as is the case for narrowband emitters. An obvious example are emitters that can decay into a multitude of electronic levels. In this case, Fermi’s golden rule states that each transition probability is proportional to the LDOS at that energy difference. This fact was recently used to alter the branching ratio of multilevel emission lines from Eu$^{3+}$ ions [236, 278]. A similar example are dye molecules at room temperature, which typically support multiple excited state and ground state levels due to coupling of the vibrational to the electronic states, leading to broad emission spectra. In such cases, it was shown that LDOS can cause strong changes in the emission spectrum [279–281]. Beside multilevel decay, another reason for broad emission spectra can be spectral diffusion, which has been observed for organic emitters [277] and quantum dots [276, 282, 283]. We will show that, if diffusion happens on time scales much faster than the lifetime, this leads to the same behaviour as with multi-level decay, i.e. the emission spectrum traces (collected) LDOS and the decay rate measures the spectral average of LDOS weighted by the intrinsic emission spectrum.
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The quantum dots in our experiment belong to the second category. Individual dots show high quantum efficiency [243] and emission bandwidths of ∼50 nm (see Fig. 5.7), much broader than the LDOS linewidths in the hybrid system (< 2 nm). We do not know whether our QDs are spectrally broad due to decay into multiple electronic or vibrational modes, or due to rapid spectral diffusion. Both mechanisms have been suggested for CdSe and CdSe/ZnSe QDs [284–286]. Slow diffusion, however, can be excluded as it was shown for similar (CdSe) QDs that emission spectra were broad (∼30 nm) even on ∼100-fs time scales [287]. This indicates that if spectral diffusion takes place, it is much faster than the lifetime. Hence, even if the exact mechanism of linewidth broadening is disputed, the resulting dependencies of emission spectra and decay rates on LDOS are the same. For simplicity, we will discuss here the case of a multilevel emitter, such as Eu³⁺. The derivation of a spectrally diffusing emitter is given in Section 7.A.

Emission spectra of broadband emitters in a narrowband photonic environment

Consider an emitter with a intrinsic emission spectrum \( p(\omega) \) in a photonic environment with frequency-dependent local density of states LDOS(\( \omega \)). The observed emission spectrum can be described as [279]

\[
I(\omega) = N_{\text{ex}} \frac{p(\omega) \gamma(\omega)}{\int p(\omega) \gamma(\omega) d\omega} \eta(\omega),
\]

(7.2)

where \( N_{\text{ex}} \) is the number of emitter excitations, the fraction represents the probability of decay to a state with frequency difference \( \omega \), \( \gamma(\omega) = \gamma_0 \text{LDOS}(\omega) \) is the frequency-dependent decay rate, and \( \eta(\omega) \) is the frequency-resolved quantum efficiency, given as [66]

\[
\eta(\omega) = \eta_C(\omega) \frac{\gamma_r(\omega)}{\gamma(\omega)} = \eta_C(\omega) \frac{\gamma_0 r \text{LDOS}(\omega)}{\gamma_{0,\text{nr}} + \gamma_{0,\text{r}} \text{LDOS}(\omega)}.
\]

(7.3)

Here, \( \eta_C(\omega) \) is the collection efficiency and \( \gamma_{0,\text{r}} \) and \( \gamma_{0,\text{nr}} \) are the intrinsic radiative and non-radiative decay rates of the emitter, which obey \( \gamma_{0,\text{r}} + \gamma_{0,\text{nr}} = \gamma_0 \). Material absorption and finite numerical aperture are captured in \( \eta_C(\omega) \), whereas intrinsic emitter losses are captured in \( \gamma_{0,\text{nr}} \). Eq. (7.2) holds for any two-level or multilevel emitter, and even for a rapidly diffusing emitter, as shown in Section 7.A. In the case that LDOS(\( \omega \)) varies much more rapidly with frequency than \( p(\omega) \) in the vicinity of a frequency \( \omega_1 \) (e.g. a broadband emitter coupled to a narrow LDOS resonance), we may simplify Eq. (7.2) to

\[
I(\omega) \approx N_{\text{ex}} p(\omega_1) \gamma_{0,\text{r}} \text{LDOS}(\omega) \eta_C(\omega).
\]

(7.4)

From this we see that the spectral shape is entirely determined by LDOS and the collection efficiency \( \eta_C(\omega) \), a fact that has been used to quantify LDOS
using emission spectra of cavity-coupled emitters in the ‘bad emitter’ limit [288]. We can define the ‘collected LDOS’ as

$$\text{LDOSC}(\omega) \equiv \text{LDOS}(\omega)\eta_C(\omega), \quad (7.5)$$

which represents the portion of LDOS leading to light emitted into the far-field and collected by the detector. The detected emission spectrum traces LDOSC(\omega). The emitter decay rate \(\gamma\) observed in a fluorescence decay trace, in contrast, is given by spectrally averaging \(\gamma(\omega)\) as

$$\gamma = \langle \gamma(\omega) \rangle = \int p(\omega)\gamma(\omega)d\omega. \quad (7.6)$$

Moreover, from Eq. (7.2) we see that total, spectrally integrated intensity depends only on \(N_{ex}\) and the collection efficiency \(\eta_C\) for an emitter with unit quantum efficiency, in agreement with literature [76, 289].

**Emission spectra in a hybrid system**

Eq. (7.4) maps onto our hybrids, since the intrinsic QD emission spectra have linewidths of 50 nm, while hybrid linewidths are below 1 nm. Let us therefore find explicit expressions for LDOS and collection efficiency in a hybrid system, using the coupled-oscillator model from Chapter 2. Cavity parameters \((Q = 3 \cdot 10^4, V_{eff} = 21\lambda^3, \omega_c/2\pi = 377.57 \text{ THz})\) are obtained from a finite element simulation (COMSOL v5.1) of a 200-nm thick Si\(_3\)N\(_4\) microdisk with a diameter of 4 \(\mu\)m.\(^\dagger\) Bare antenna dipole moment \(\alpha_{hom}\) and antenna-emitter coupling strength as captured in \(G_{bg}\) are obtained from the simulations of aluminium rod antennas on a Si\(_3\)N\(_4\) substrate used in Section 6.5 (spectra shown in Fig. 5.4). Considering the quantum dot diameter of \(~10\) nm, we assume the emitter to be 5 nm from the antenna apex and 5 nm above the substrate. Hybrid LDOS relative to vacuum can then be found from a slightly modified version of Eq. (2.46)

$$\text{LDOS}(\omega) = \text{LDOS}_{bg} + \frac{6\pi \epsilon_0 c^3}{\omega^3} \text{Im} \left\{ \alpha_H G_{bg}^2 + 2G_{bg} \alpha_H \chi_{hom} + \chi_H \right\}. \quad (7.7)$$

Here, \(\text{LDOS}_{bg} = 1.62\) is the relative LDOS of the background environment, which we find from the bare antenna simulations and which corresponds well to the LDOS of 1.6 felt by an in-plane dipole 15 nm from a Si\(_3\)N\(_4\)-air interface. Collection efficiency is assumed to be given by the fraction of power emitted as dipole radiation by the source and antenna (see Section 2.4.4). This assumption uses the fact that practically all radiation from the microdisk WGM

\(^\dagger\)Note that at such small diameters, bending losses dominate over the surface scattering and absorption that limit \(Q\) for the 8, 12 and 15 \(\mu\)m disks. This is evident from the fact that \(Q\) as obtained from a linear extrapolation of the \(Q\) of these larger disk is higher \((Q \approx 6 \cdot 10^4)\) than the \(Q\) obtained from the simulations, which only contains bending losses.
is emitted in the in-plane direction, which is not collected by the objective. Our simulations show that \( \sim 3\% \) of the radiation by a 4 \( \mu \)m disk is collected by a NA=0.95 objective. Given that the spectrometer fiber also selects a \( \sim 1 \) \( \mu \)m detection area on the sample, this fraction will be even lower in practice.

![Graphs showing LDOS and LDOSC](image)

**Figure 7.6: LDOS and LDOSC in a hybrid system.** LDOS (blue) and collected LDOS (LDOSC, red), as well as collection efficiency \( \eta_C(\omega) \) (green) in hybrid systems, for different antenna lengths \( L \) and bare cavity \( Q \) (both indicated above plots). The theoretical quality factor of the WGM cavities in this study is \( 3 \cdot 10^4 \), corresponding to panels (c,d,i,j). The black dashed line indicates the bare antenna albedo \( A \), which determines the collection efficiency away from the hybrid mode. For sufficiently high \( Q \), or if the antenna is close enough to resonance to be the dominant source of loss, LDOS and LDOSC take on mostly the same lineshape. LDOS and LDOSC are normalized to their maxima, to facilitate comparison. Note that (a-f) and (g-l) have different x-axes.

Let us first compare the LDOS and LDOSC spectra for several examples of cavities and antennas, to learn what influences their spectral shape. Fig. 7.6 shows calculated spectra of LDOS, LDOSC and \( \eta_C \) for two different antenna lengths \( L \) and three different cavity quality factors. For both LDOS and LDOSC, we observe the familiar Fano-type lineshapes discussed in Chapter 3.
For a short antenna, far from resonance, LDOS resonances are narrow and lineshapes are close to Lorentzian peaks, while for an antenna closer to resonance (L=128 nm), resonances are broadened by antenna losses and the lineshapes are more asymmetric. This is the same behaviour as observed for the Fano resonances in the experimental hybrid spectra.

Interestingly, the lineshapes of LDOS and of LDOSC can be very similar under certain conditions. For the antenna near resonance, LDOS and LDOSC are very similar for all values of $Q$ shown. For the short antenna, this is only true for the highest $Q$. This behaviour is best explained by considering the collection efficiency $\eta_C$. Collection efficiency is determined by bare antenna albedo $A$ away from resonance. However, at the minimum of the Fano feature in LDOS, it drops significantly. This is due to destructive interference between field coupled directly from emitter to antenna, and field that travels via the cavity. This interference depolarizes the antenna, causing a drop in dipole radiation $P_r$. LDOS, however, remains finite at this frequency due to the contribution of the cavity mode, i.e. the $\chi_H$ term in Eq. (7.7). In other words, at the Fano dip, nearly all emission is transferred into the cavity decay channel, which is not collected by the objective, causing a dip in collection efficiency. This phenomenon was also observed recently in theoretical work on a different hybrid system [109]. In general, the dip in $\eta_C$ causes a difference in lineshape between LDOS and LDOSC. However, if antenna losses dominate the hybrid linewidth, lineshapes are far broader than the dip in $\eta_C$, such that LDOSC lineshape remains mostly unaffected, except for a decrease of the minimum at the Fano dip. This explains why LDOSC and LDOS are similar for the long antenna, whereas for the short antenna, where antenna losses are lower, this is only the case if $Q$ is high.

We have shown that in general, emission spectra of broadband emitters coupled to a narrowband LDOS resonance follow the spectral shape of the collected LDOS. However, our results suggest that these lineshapes can, under the right circumstances, be nearly equivalent to the lineshapes in LDOS. Let us therefore compare the lineshapes expected from our theoretical model to those measured in the hybrid systems. Fig. 7.5 shows this comparison for the linewidth, Fano phase and hybrid boost factor. Data is compared to theory values obtained from LDOS and from LDOSC, where we assumed $Q = 3 \cdot 10^4$ following the simulation result. For linewidth, LDOS and LDOSC give the same prediction, which corresponds very well to the trend in the data. This linewidth is just the hybrid linewidth also studied in Chapter 6, i.e. the cavity linewidth broadened according to the Bethe-Schwinger perturbation formulas Eqs. (2.28) and (2.29). Predictions of Fano phase $\theta$ and hybrid boost from LDOS and LDOSC differ for the smallest antennas, since there antenna losses are not very dominant. For $\theta$, the difference is small and data agrees reasonably well with both curves. For the hybrid boost, however, we find overall good agreement of the data to the predictions from LDOSC, which is markedly different from that by LDOS at small antennas. A few datapoints,
however, show much higher boosts, lying closer to the prediction from LDOS. Since we would not expect our cavities to have quality factors higher than the theoretical limit set by bending losses, this likely indicates that for these hybrids, part of the cavity radiation is collected by the objective. Our calculation of LDOSC assumes no cavity radiation is collected, yet in reality we do collect a finite fraction, as we have seen in the taper-coupled measurements in Chapter 6 (see for example Fig. 6.3f). For the smallest antennas the majority of fluorescence is emitted as cavity radiation, so collecting just a small fraction of it can make a large difference to collection efficiency, causing LDOSC to be closer to LDOS in lineshape. This could explain why these hybrid modes show such high boosts, close to the boosts in LDOS. Finally, Fig. 7.5 also shows curves obtained directly from finite element simulations of the complete hybrid system, similar to those discussed in Section 3.5, where we take aluminium antennas rather than gold ellipsoids and place the constant current source again 5 nm from the antenna apex and the substrate. We see that resulting LDOS lineshapes show excellent agreement with those from our coupled-oscillator model, confirming again the validity of our analysis.

These results show that LDOSC can be significantly boosted in a hybrid system, with respect to a bare antenna. Owing to the geometry of our sample, in which cavity radiation is very poorly collected, the measured boosts in LDOSC are always lower than boosts in LDOS. Therefore, we may interpret these boosts as a lower bound on the LDOS boosts experienced by the emitters in a hybrid system. This indicates the great potential of hybrid systems for emission control: at hybrid resonances far detuned from an antenna resonance, LDOS is boosted by more than an order of magnitude with respect to the antenna. Tuning closer to antenna resonance allows increasing the bandwidth of operation. Hybrid boost decreases, but since antenna LDOS increases simultaneously, overall LDOS should remain similar.

### 7.4 LDOS enhancements measured from quantum dot decay rates

Fluorescence decay curves were measured for each hybrid system. Additionally, for comparison we measured decay curves of 5 different individual QDs, dilutely dispersed on a glass substrate, as well as a decay curve of a QD ensemble attached to a large Al pad of 30 nm thickness. During the hybrid measurements, both pump and detection polarizers are aligned to the antenna long axis. Fig. 7.7g-h,j-k show two examples of fluorescent decay traces measured in hybrid systems. We can see that fluorescence decays significantly faster than for a bare QD on glass, which is shown in Fig. 7.7a,b. Within the first 3 ns, signal has decayed by approximately two orders of magnitude. Beside this rapid decay, a slow component similar to that of the QD on glass appears present as well.
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Figure 7.7: Fluorescence decay in hybrid systems. Fluorescence decay curves, showing just the first 10 ns or the full 100 ns, as well as fitted decay rate distributions $P(\gamma)$. We show data for a single QD on glass (a-c), an ensemble of QDs on a large Al pad (d-f), and for two hybrid systems with antenna lengths $L$ of 108 nm (g-i) and 148 nm (j-l). Data (blue line) is fitted (red dashed line) with a bimodal distribution of decay rates. Fractions of light in the slow decay rates $F_2$ are indicated. In the rightmost panels, the dashed green (red) lines indicate the expectation values $\langle \gamma_1 \rangle \ (\langle \gamma_2 \rangle)$ of decay rate in the fast (slow) modes of the distributions. Note that, although these distributions may have finite amplitude outside the range of rates shown here, those rates are not taken into account when calculating decay curves, due to the finite integration bounds $\gamma_{\min}$ and $\gamma_{\max}$.

To extract decay rates and relative weights of the slow and fast components, we fit the data with a bimodal distribution of decay rates. Since the signal comes from more than a single QD, a bi-exponential decay is not suited for the hybrid data. A practical solution that is frequently employed for analysing decay curves of emitter ensembles, is to use a distribution of decay rates.
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[290, 291]. The decay curve $f(t)$ is then described by

$$f(t) = \int_0^\infty P(\gamma)e^{-\gamma t}d\gamma,$$

(7.8)

with $P(\gamma)$ the decay rate distribution. With respect to the conventional description [290], we added the extra factor $\gamma$ in the integrand. Provided that $P(\gamma)$ is normalized such that $\int_0^\infty P(\gamma)d\gamma = 1$, this ensures that $f(t)$ obeys $\int_0^\infty f(t)dt = 1$. Therefore, the weight of a particular rate in $P(\gamma)$ can be interpreted as the contribution of that rate to the integral of the decay trace, i.e. the total signal. In practice, we do not let the integral in Eq. (7.8) run from 0 to $\infty$ but use realistic integration bounds $\gamma_{\text{min}}$ and $\gamma_{\text{max}}$, given respectively by the inverse of 20 times our measurement time window (100 ns) and by $1/\Delta t$, where $\Delta t$ is the time resolution of our timing card (0.16 ns).

Part of our signal will likely come from emitters positioned very close to the antenna apex, yet there is also a contribution from emitters positioned further from the apex. The emitters near the apex should experience large LDOS, being in the antenna hotspot, while the emitters that are not attached to the apex will experience significantly lower LDOS. Remember that, although LDOS at the hybrid mode peaks may be high, what counts for the observed decay rate $\gamma$ is the spectrally averaged LDOS as given in Eq. (7.6). We thus expect $\gamma$ to be determined mostly by the bare antenna, which offers broadband LDOS enhancement, and not by the hybrid modes. The contribution of both fast and slow emitters to the decay curve, as well as the bi-exponential decay observed for individual QDs, rationalizes a fit using a bimodal decay rate distribution

$$P(\gamma) = \frac{(1-A_2)}{\sigma_1\sqrt{2\pi}} \exp \left[ \frac{(\gamma - \mu_1)^2}{2\sigma_1^2} \right] + \frac{A_2}{\sigma_2\sqrt{2\pi}} \exp \left[ \frac{(\gamma - \mu_2)^2}{2\sigma_2^2} \right],$$

(7.9)

where $\mu_1$ and $\mu_2$ are the peak lifetimes in respectively the first and second mode, $\sigma_1$ and $\sigma_2$ are their respective variances and $A_2$ is the weight of the second mode to the total distribution. From the fitted distributions, we calculate expectation values $\langle \gamma_1 \rangle$ and $\langle \gamma_2 \rangle$ for decay rates in the first and second mode, respectively, as $\langle \gamma_1 \rangle = \int_{\gamma_{\text{min}}}^{\gamma_{\text{max}}} \gamma P_{(A_2=0)}(\gamma) d\gamma$ and similarly for $\langle \gamma_2 \rangle$. Moreover, we calculate fractions $F_1$ and $F_2$ of the detected light coming from mode 1 and mode 2 as the integrated area under the respective modes, relative to the total area under $P(\gamma)$. Note that if the modes are sufficiently narrow and centered far from $\gamma_{\text{min}}$ and $\gamma_{\text{max}}$, we simply obtain $\langle \gamma_1 \rangle = \mu_1$, $\langle \gamma_2 \rangle = \mu_2$, $F_1 = 1 - A_2$ and $F_2 = A_2$. From here on, we choose $\mu_1 > \mu_2$ and refer to the first and second mode as the fast and slow decay rate modes. Note that we do not claim that the underlying distributions are perfectly described by a bimodal distribution. However, this serves as a parametrization of the results and allows a comparison between measurements.

Fig. 7.7g-l show exemplary fits to the decay curves of the hybrid systems using the bimodal decay rate distribution, as well as the fitted decay rate
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distributions \(P(\gamma)\). The same fit is performed on measurements of a single QD on glass and of a QD ensemble on a large Al patch, with results shown in Fig. 7.7a-c and d-f, respectively. The model fits the data well. The accelerated decay observed in the hybrid data is visible in the decay rate distribution as an increase of the \(\langle \gamma_1 \rangle\). We also observe a decrease of \(F_2\) compared to the bare QD, indicating that more light is coming from the rapid decays. One could argue that the accelerated decay rates in the hybrid systems arise simply from nonradiative quenching, as the QDs are placed very close to a metal. However, a comparison with QDs on a large Al patch shows that these QDs do not have fast decay rates as high as those on the hybrids. In fact, \(\langle \gamma_1 \rangle\) is comparable to that in QDs on glass.

Figure 7.8: Fitted decay rates \(\gamma\) and slow decay fractions \(F_2\). (a) Fast-mode decay rate expectation values \(\langle \gamma_1 \rangle\) obtained from fits on hybrid systems, as function of antenna length. We compare values for the hybrids (blue markers) to the value for a individual QDs on glass (green dashed line), and for QDs on an Al substrate (red dashed line). The \(1/e\) time of the instrument response function (IRF) is indicated by the black dashed line. (b) Fractions of light in the slow decay rates \(F_2\). Again, hybrid data is compared to QDs on glass (green) and QDs on Al (red). In both (a) and (b), error bars and shaded areas indicate standard deviations or, for QDs on Al in panel (a), the width of the lifetime distribution.

Fig. 7.8 shows \(\langle \gamma_1 \rangle\) and \(F_2\) for all the hybrid systems, as function of antenna length. We observe a clear trend of decay rate peaking around an antenna length of 128 nm at 2.2(6) ns\(^{-1}\), approximately 2.8 times faster than for QDs on glass ( 0.8(0.5) ns\(^{-1}\)). Moreover, the lower values of \(F_2\) in the hybrid systems as compared to the QDs on glass indicate that more light is coming from the fast decays in these systems. Whereas a QD on glass emits 97(4)% of its light with a slow decay rate (associated to the bright state, see Section 5.5), in the hybrids this fraction starts at 76(11)% for the lowest antennas and drops to 55(6)% for antennas of 128 nm length. These trends show that there is a

\[1\text{In (e) and (k), small peaks are visible around 30 ns, which are attributed to APD afterpulsing. To ensure that they do not influence the fit, we disregard the 10-ns time window around 30 ns containing most of the peaks during the fit procedure.}\]
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decay rate enhancement in these hybrids. This enhancement is likely to be an effect of the bare antenna rather than fluorescence quenching, for two reasons. Firstly, decay rates peak when the antenna length is tuned approximately to resonance with the QDs, and are significantly higher than for QDs on an Al patch. If quenching would be dominant, we would expect these rates to be roughly equal. Secondly, if the fast decay component in the hybrid systems would come from strongly quenched quantum dots, we should expect only a small contribution of these decays in $P(\gamma)$. Instead, this contribution is much larger in the hybrids than in the bare QDs. Whether to assign the enhanced values of $\langle \gamma_1 \rangle$ to an acceleration of the bright or dark state lifetime is unclear. The fact that $F_2$ is lower than for the QDs on glass, suggests that at least part of the fast decay mode can be attributed to quantum dots in the bright state. However, we cannot be sure, as we do not know if, for example, the presence of the Al has affected the QD blinking statistics. For this, experiments on hybrids containing only a single QD per antenna would be required. In that case, one can use only the counts from the bright state in a decay trace, which has been shown to yield a single exponential [242, 246]. This would allow an unambiguous retrieval of bright state lifetime. Combining this information with the hybrid boosts measured on the QD spectra, one could then determine a lower boundary for the LDOS at the hybrid peak. Finally, we note that no significant difference was found between $\langle \gamma_2 \rangle$ on the hybrids and the QDs on glass. In all cases, $\langle \gamma_2 \rangle \approx 0.008$ ns$^{-1}$. On the hybrids, this can be understood as the contribution of QDs that are too far from the antenna to experience significant LDOS.

7.5 Conclusions and outlook

We have fabricated antenna-cavity hybrids dressed with fluorescent quantum dots positioned at the antenna hotspots. Quantum dot fluorescence spectra and fluorescence decay curves were measured. Spectra showed Fano-type resonances, which we associated to the hybridized whispering-gallery modes of the cavity. We showed that in systems of broadband emitters coupled to narrowband LDOS resonances, fluorescence spectra take on the spectral shape of the collected LDOS (LDOSC). This was supported by the fact that measured Fano linewidth and lineshape were found to vary with antenna length, in good agreement with theoretical values for LDOSC resonances. Furthermore, we observed strong fluorescence boosts at the peaks of hybrid modes, which indicates a strong enhancement of LDOSC in the hybrid system with respect to the bare antenna alone. These hybrid boosts grow with decreasing antenna length, up to a maximum of $\sim 14$ for the shortest antennas, in good agreement with the prediction from LDOSC. These boosts provide a lower bound on LDOS boosts experienced by the emitters in a hybrid system. Our results demonstrate the symbiotic behaviour in antenna-cavity hybrids. For cavities
red-detuned from the antenna resonance, LDOS can be significantly enhanced with respect to that of the antenna alone.

Fluorescence decay measurements showed a strong decay rate enhancement in hybrid systems, as compared to quantum dots on glass. We ascribe this enhancement to an effect mainly of the antenna alone, since the hybrid modes offer only LDOS enhancements over a small fraction of the emitter linewidth. Decay rates peaked when antennas were near resonance with the quantum dots, a strong indication of antenna effects.

These results constitute the first experimental observation of tunable Fano-type lineshapes and strongly enhanced LDOS for single-photon emitters in hybrid antenna-cavity systems. Earlier works have observed fluorescence in hybrid systems [97, 110], though not from single-photon emitters. Moreover, emitters were never placed at the antenna hotspot, thus not making optimal use of the confinement provided by antenna, while still suffering from the losses it induces. In this work, instead, emitters benefit optimally from the antenna confinement, being right at the antenna hotspot.

In future experiments, we propose to use hybrids containing a single quantum dot per antenna, which is feasible using the fabrication techniques employed in this work. This would allow the unambiguous retrieval of LDOS from the bright state lifetime. Combined with the hybrid boosts measured from the quantum dot spectra, one could then determine a lower boundary for the LDOS at the hybrid peak. Moreover, employing emitters with emission bandwidths below the hybrid linewidths, possibly at cryogenic temperatures, would allow a direct measurement of LDOS through the emitter lifetime. This would also open up the possibility of bringing hybrid systems towards the strong coupling regime, which was suggested to be theoretically feasible in a different antenna-cavity geometry [109].
Appendices

7.A Spectrum and decay rate of a spectrally diffusing emitter

Here we derive the emission spectrum and decay rate of a spectrally diffusing emitter in a structured photonic environment. We emphasize that our derivation is fully classical. Quantum-mechanical models of emitters with dephasing coupled to a single optical cavity mode can be found for example in [292–294].

Consider a system as depicted in Fig. 7.9. An emitter is prepared at \( t = 0 \) in the excited state \( |e_0\rangle \). It can make the transition to a ground state \( |g\rangle \), at energy difference \( \hbar \omega_0 \), emitting a photon of frequency \( \omega_0 \). Suppose the emitter wanders around with a characteristic time scale \( \Delta t \), hopping to a new state \( |e\rangle \). Assuming temporally uncorrelated hopping probability, it has a spectrum of new states to choose from at each hopping event, governed by the spectral probability density \( p(\omega) \) of hopping to a state with energy \( \hbar \omega \) relative to \( |g\rangle \), which is normalized such that \( \int_{-\infty}^{\infty} p(\omega) d\omega = 1 \). We could interpret \( p(\omega) \) as the intrinsic emission spectrum of the emitter (i.e. its spectrum if it were in an environment with frequency-independent photonic density of states), when measured on timescales much longer than the hopping time \( \Delta t \).

\[ \chi_{\phi \tau}(\phi \sigma) \]

\[ \chi_{\phi \lambda}(\phi \sigma) \]

\[ \Delta \mu \]

\[ \hbar \omega_0 \]

\[ \hbar \omega_1 \]

\[ \hbar \omega_2 \]

\[ \Delta t \]

\[ |e_0\rangle \]

\[ |e_1\rangle \]

\[ |e_2\rangle \]

\[ |g\rangle \]

\[ \gamma(\omega_0) \]

\[ \gamma(\omega_1) \]

\[ \gamma(\omega_2) \]

\[ \text{Figure 7.9: Sketch of an emitter showing spectral diffusion. After every time } \Delta t \text{ it hops to a new excited state } |e\rangle. \text{ In each state, it has a probability of decaying to the groundstate } |g\rangle, \text{ emitting a photon at energy } \hbar \omega. \]

\[ \text{Note that we might as well have chosen to describe the system as hopping between available groundstates } |g_j\rangle. \text{ Although this seems like a less physical scenario, for the analysis this choice is irrelevant.} \]
7. Spectrum and decay rate of a spectrally diffusing emitter

7.1 Emission spectrum

One may now ask: what is the chance that an emitter decays with transmission frequency $\omega_e$ precisely after $N$ jumps. Suppose that we first examine a specific trajectory that does this - and that previously visited the specific list of frequencies $\omega_1, \omega_2, \ldots, \omega_{N-1}$. Suppose also that the probability to survive a time span $\Delta t$ at a frequency $\omega_i$ is written as $s_{\omega_i}$. Our specifically chosen trajectory first needs that you survive the first $N-1$ jumps, and then precisely in the $N$th bin, you require decay. The likelihood of this particular trajectory is

$$
\mathcal{P}^{N,\omega_e}_{\omega_1,\omega_2,\ldots} = \left( \prod_{i=1}^{N-1} p(\omega_i) s_{\omega_i} \right) p(\omega_e) (1 - s_{\omega_e}).
$$

(7.10)

Now note that the likelihood to somehow end up decaying at $\omega_e$ precisely after $N$ jumps is obtained by summing over all possible trajectories. Assuming that the draws from time bin to time bin are uncorrelated, this means that

$$
\mathcal{P}^{N,\omega_e} = \left[ \int \ldots \int \left( \prod_{i=1}^{N-1} p(\omega_i) s_{\omega_i} \right) d\omega_1 d\omega_2 \ldots d\omega_{N-1} \right] p(\omega_e) (1 - s_{\omega_e}).
$$

(7.11)

simplifies to

$$
\mathcal{P}^{N,\omega_e} = \langle s \rangle^{N-1} p(\omega_e) (1 - s_{\omega_e}).
$$

(7.12)

Here we introduced the spectrally averaged survival probability

$$
\langle s \rangle = \int p(\omega_i) s_{\omega_i} d\omega.
$$

(7.13)

For instance, suppose $\Delta t$ is much smaller than any $\gamma$ in the system, then we can take \[11\]

$$
\begin{align*}
    s_{\omega} &= 1 - \gamma(\omega) \Delta t \\
    \langle s \rangle &= 1 - \langle \gamma(\omega) \rangle \Delta t
\end{align*}
$$

(7.14)

with $\langle \cdot \rangle$ the spectral averaging. Alternatively, if $\Delta t$ is not taken short, we have to assume single exponential decay, with a rate dependent on the frequency bin, and find instead

$$
\begin{align*}
    s_{\omega} &= e^{-\gamma(\omega) \Delta t} \\
    \langle s \rangle &= e^{-\langle \gamma(\omega) \rangle \Delta t}.
\end{align*}
$$

(7.15)

From Fermi’s golden rule we know that $\gamma(\omega)$ is proportional to the optical local density of states (LDOS) \[11\].

Now, the probability to finally end up with a decay favouring a particular color $\omega_e$ after any number of steps, is obtained from

$$
\mathcal{P}(\omega_e) = \sum_{N=1}^{\infty} \mathcal{P}^{N,\omega_e} = \left[ \sum_{N=1}^{\infty} \langle s \rangle^{N-1} \right] p(\omega_e) (1 - s_{\omega_e}) = \frac{p(\omega_e)(1 - s_{\omega_e})}{1 - \langle s \rangle}.
$$

(7.16)
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It is easily verified that this expression is properly normalized, that is, \( \int_\omega \mathcal{P}(\omega) d\omega = 1 \). Note that here the quantity \( s \) strictly relates to total decay rates, and \( \mathcal{P}(\omega_e) \) relates to the chance to decay, not necessarily that this results in a photon. To this end we introduce the frequency resolved instantaneous quantum efficiency \( \eta(\omega) \) as [66]

\[
\eta(\omega) = \eta_C(\omega) \frac{\gamma_\text{r}(\omega)}{\gamma(\omega)} = \eta_C(\omega) \frac{\gamma_0,\text{r} \text{LDOS}(\omega)}{\gamma_{0,\text{nr}} + \gamma_0,\text{r} \text{LDOS}(\omega)},
\]

(7.17)

with \( \eta_C(\omega) \) the frequency-dependent collection efficiency and \( \gamma_0,\text{r} \) and \( \gamma_{0,\text{nr}} \) the intrinsic radiative and non-radiative decay rates of the emitter. Material absorption and finite numerical aperture are captured in \( \eta_C(\omega) \), whereas intrinsic emitter losses are captured in \( \gamma_{0,\text{nr}} \). The expected photon count rate per emitter excitation is then

\[
\mathcal{C}(\omega_e) = \frac{p(\omega_e)(1 - s_{\omega_e})}{1 - \langle s \rangle} \eta(\omega_e).
\]

(7.18)

You can now expand this as

\[
\mathcal{C}(\omega_e) = \frac{1 - e^{-\gamma(\omega_e) \Delta t}}{1 - \langle e^{-\gamma(\omega) \Delta t} \rangle} p(\omega_e) \eta(\omega_e).
\]

(7.19)

The photon count rate traces the product of the intrinsic emitter spectrum and the spectrally resolved quantum efficiency times a factor dependent on \( \Delta t \). If \( \Delta t \) is large (slow diffusion), one finds a ratio unity, i.e. the spectrum does not trace LDOS variation (unless through \( \eta(\omega_e) \), which is proportional to LDOS only if \( \gamma_{0,\text{nr}} \gg \gamma_0,\text{r} \)). In contrast, for \( \Delta t \) very small, one can use \( 1 - e^{-x} \approx x \), and interchange Taylor expansion and averaging to obtain

\[
\mathcal{C}(\omega_e) \approx \frac{\gamma(\omega_e)}{\gamma(\omega)} p(\omega_e) \eta(\omega).
\]

(7.20)

Now the spectrum traces the ratio of total LDOS to the spectrally averaged total LDOS.

### 7.A.2 Decay rate

The emitter decay curve \( f(t) \) is described by the probability that the emitter survives \( N - 1 \) hopping events and then decays precisely after \( N \) jumps, where \( N = t/\Delta t \). Analogous to Eq. (7.10), the likelihood of a particular trajectory is

\[
\mathcal{P}_{\omega_1,\omega_2,\ldots} = \left( \prod_{i=1}^{N-1} p(\omega_i) s_{\omega_i} \right) p(\omega_N)(1 - s_{\omega_N}),
\]

(7.21)

and consequently the likelihood of somehow decaying precisely after \( N \) jumps, i.e. \( f(t) \), is

\[
f(t) = \mathcal{P}^N = \langle s \rangle^{N-1} \langle 1 - s \rangle.
\]

(7.22)
7. A Spectrum and decay rate of a spectrally diffusing emitter

If we assume again that $\Delta t$ is much shorter than any $\gamma$ in the system, we obtain

$$f(t) = \left[ 1 - \langle \gamma(\omega) \rangle \Delta t \right]^{N-1} \langle \gamma(\omega) \rangle \Delta t.$$  \hspace{1cm} (7.23)

It is readily verified that this is properly normalized, such that $\sum_{N=0}^{\infty} f(t) = 1$. If $\Delta t$ is short,

$$f(t) \approx e^{-\langle \gamma(\omega) \rangle t \langle \gamma(\omega) \rangle \Delta t},$$  \hspace{1cm} (7.24)

where we used $N \Delta t = t$ and the power series expression for an exponential. This shows that the emitter exponentially decays with a decay rate given by the spectral average of $\gamma(\omega)$. 