Photon Statistics of Filtered Resonance Fluorescence

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Spectral filtering of resonance fluorescence is widely employed to improve single photon purity and indistinguishability by removing unwanted backgrounds. For filter bandwidths approaching the emitter linewidth, complex behaviour is predicted due to preferential transmission of components with differing photon statistics. We probe this regime using a Purcell-enhanced quantum dot in both weak and strong excitation limits, finding excellent agreement with an extended sensor theory model. By changing only the filter width, the photon statistics can be transformed between antibunched, bunched, or Poissonian. Our results verify that strong antibunching and a sub-natural linewidth cannot simultaneously be observed, providing new insight into the nature of coherent scattering.

Resonance fluorescence (RF) of two-level emitters (TLEs) is integral to numerous important proposals for optical quantum technologies such as single photon sources [1–3], spin-photon entanglement [4, 5] and entanglement of remote spins [6, 7]. The emitted spectrum is well-suited to these applications, exhibiting strong single-photon antibunching and, under appropriate excitation conditions, a dominant coherently scattered component with a sub-natural linewidth inherited from the laser coherence. Indeed, for an ideal TLE in the limit of vanishing driving strength, the coherent fraction tends towards unity whilst retaining perfect antibunching. It is thus perhaps intuitive to assume that this coherent component must itself be antibunched. However this is not the case; in this work we exploit spectral filtering to demonstrate that strong antibunching and a sub-natural linewidth are mutually exclusive properties, in accordance with theoretical predictions [8, 9].

In experimental quantum optics, spectral filtering around the zero phonon line (ZPL) of a TLE is widely employed to remove unwanted backgrounds from the driving laser [1, 3], other transitions [10] or phonon sidebands [10–12], improving the measured single photon purity and indistinguishability. Considering only indistinguishability, reducing the filter bandwidth always gives an improvement (at the cost of efficiency) as more background is removed [13]. However, as the filter bandwidth approaches the natural linewidth (γ) of the ZPL, theory predicts strongly modified photon statistics in both weak (coherent scattering) [9, 14] and strong (Mollow triplet) [15] driving regimes, an effect generally overlooked in experiments to date. Here, we experimentally verify these predictions, combining our results with a theoretical model to develop a thorough understanding of the complex photon statistics associated with spectrally filtered resonance fluorescence. These concepts are equally applicable to the broad assortment of atomic and atom-like TLEs used in current quantum optics research.

The sample is studied in a liquid helium bath cryostat at 4.2 K and incorporates a self-assembled InGaAs quantum dot (QD) into an H1 photonic crystal cavity (PhCC) with coupled W1 waveguides (Fig.1(a)). Resonant continuous wave (CW) laser excitation and collection of emission is made from directly above the cavity whilst laser back-scattering is rejected using a cross-polarisation technique. A p-i-n diode structure allows the QD neutral exciton to be electrically tuned. At the cavity resonance, a maximum Purcell factor of 43 shortens the QD’s radiative lifetime (T1) to 22.7 ps and results in lifetime-limited coherence [3]. Here, the QD is slightly detuned from the cavity, giving a Purcell factor of ∼30 and a broad natural linewidth (γ) of 20 µeV. This large γ enables exploration of filter bandwidths (Γ) ≤ γ using a combination of diffraction grating and etalon filters.

For resonant CW excitation and a lifetime-limited emitter coherence time (T2 = 2T1), the weak excitation limit is defined as ΩR < (γ2/2) where ΩR is the Rabi frequency and γ = 1/T1 [16]. This is often termed the Resonant Rayleigh Scattering (RRS) or Heitler regime [17–19]. The RF spectrum in this regime includes contributions from both coherent RRS and an incoherent part originating from spontaneous and stimulated emission [20, 21]. For coherent scattering, excitation and emission become a single coherent event where the elastically scattered photons inherit the laser coherence, leading to a sub-natural linewidth [3, 18–20, 22, 23] that illustrates the long coherence times possible in this regime. Meanwhile, the natural linewidth of the incoherent com-
ponent is given by $\gamma = 1/T_1$. Theory suggests that for weak excitation, interference between these different components is the origin of the observed photon antibunching [9]. Owing to the discrepancy in linewidth between coherent and incoherent components, filtering with width $\Gamma < \gamma$ inevitably alters the ratio of the different components, modulating the interference between them and thus the observed photon statistics.

To explore this, Hanbury Brown and Twiss correlation measurements (HBT) [24] of the second-order correlation function $g^{(2)}(t)$ were performed in the continuous weak driving regime. A value of $g^{(2)}(0) < 1$ corresponds to antibunched emission whilst a value of $g^{(2)}(0) = 1$ corresponds to the Poissonian statistics of a coherent source such as a laser. The QD is resonantly excited at laser energy $\hbar \omega_L$, inducing a Rabi frequency $\Omega_R = 0.5 \gamma$. The emission is collected in cross-polarisation and passed through a filter centred on the ZPL before being split by a 50:50 fiber beamsplitter to a pair of superconducting nanowire single photon detectors (SNSPD) connected to a time-correlated single photon counting module (TCSPC), shown schematically in Fig. 1(a). The SNSPDs have a Gaussian instrument response function (IRF) with $37.5 \pm 0.1$ ps full-width half-maximum.

Fig. 1(b) illustrates the theoretical total spectrum (purple) of the QD under these conditions, comprising an incoherent peak with $\gamma = 20$ meV (blue) and a coherent peak with a linewidth of $\sim 10$ neV inherited from the laser (red). The area of the coherent peak relative to the total spectrum is the coherent fraction $F_{CS}$ [16]:

$$F_{CS} = \frac{1}{1 + 20\Omega_R^2 / \gamma^2}$$  \hspace{1cm} (1)

which gives $F_{CS} = 2/3$ for $\Omega_R = 0.5 \gamma$. The transmission coefficients of the coherent and incoherent parts through an ideal Lorentzian filter with bandwidth $\Gamma$ are plotted in Fig. 1(c). As $\Gamma$ is reduced, the transmission of the incoherent component decreases much faster than the coherent component owing to the large (2000$\times$) linewidth difference. Spectral filtering can thus manipulate this ratio up to a limiting case where narrow filtering removes the incoherent component almost entirely.

The variation of $g^{(2)}(0)$ with $\Gamma$ is shown in Fig. 2(a) for $\Omega_R = 0.5 \gamma$. As expected for an unfiltered ideal TLE, strong antibunching is observed where the filter bandwidth exceeds the natural linewidth ($\Gamma > \gamma$). At $\Gamma = 150 \gamma$, $g^{(2)}(0) = 0.09 \pm 0.01$ limited only by the detector IRF ($= 1.14 \gamma^{-1}$). However, as the filter bandwidth becomes $\ll \gamma$, the antibunching is lost and $g^{(2)}(0)$ tends towards 1. The experiment agrees well with theoretical predictions (black lines in Fig. 2(a)) derived using the sensor formalism [25–27] with (solid line) or without (dashed line) convolution with the detector IRF. It is interesting to note that when moving from $\Gamma = 150 \gamma$ to $\Gamma = 23 \gamma$, nearly the entire phonon sideband [31–33] is removed with no appreciable change in $g^{(2)}(0)$. The nature of these measurements mean that electron-phonon interaction processes such as excitation induced dephasing [34] and phonon sideband emission [31–33] have negligible impact on $g^{(2)}(t)$. Discussions of the sensor formalism, its extension to include laser background (see below) and consideration of phonon effects are given in the Supplemental Material [27].

Comparing Fig. 1(c) to Fig. 2(a) illustrates that the loss of antibunching occurs in the regime ($0.1 \gamma \lesssim \Gamma \lesssim 10 \gamma$) where the reduced filter bandwidth removes almost
the entire incoherent component. Indeed, the inset to Fig. 2 shows that in this region, the filtered coherent fraction approaches unity. This demonstrates that without both coherent and incoherent contributions, strong antibunching cannot be observed, indicating in accordance with theoretical predictions [8, 9] that the antibunching originates from interference between these components.

Fig. 2(b) shows some of the individual $g^{(2)}(t)$ measurements from which Fig. 2(a) is derived. As the filter bandwidth narrows, the central dip in $g^{(2)}(t)$ broadens in width. This can be interpreted according to the uncertainty relation $\Delta E \Delta t > \frac{\hbar}{2}$ which implies that a narrower filter $(\Delta E)$ inevitably increases the associated timing uncertainty of the photon. Considering that passing a photon through a narrow filter is equivalent to detecting a narrow linewidth, this illustrates that it is impossible to simultaneously observe both a sub-natural linewidth and strong antibunching from a TLE.

Beyond the weak excitation limit, the strong driving regime is defined as $\Omega_R \gg \frac{1}{\Gamma_0}$. Fig. 3 shows the resulting AC Stark effect transformation of the “bare” states of the TLE into “dressed” states split by the Rabi energy $(\hbar \Omega_R)$. This splitting gives four possible transitions between upper and lower manifolds, as two of the transitions are degenerate, the result is the purple Mollow triplet spectrum shown in Fig. 3 for $\Omega_R = 2\gamma$. The central peak is often termed the Rayleigh peak (blue), flanked by two Mollow side peaks (green). The width of the individual peaks is governed by $\gamma$ [35, 36]. In addition to these incoherent peaks, a contribution from coherent scattering remains (red). As $\Omega_R$ increases, the Mollow splitting between side peaks increases whilst the coherent fraction decreases according to Eq. 1.

Frequency-resolved studies of Mollow triplet photon correlations have revealed a rich assortment of physics. An unfiltered Mollow spectrum exhibits antibunching whilst isolating individual peaks results in $g^{(2)}(0) = 1$ for the central Rayleigh peak and antibunching for the side peaks [37–39]. Cross-correlation measurements between the Rayleigh peak and either side peak exhibit antibunching [40] whilst a cross-correlation between side peaks exhibits bunching $(g^{(2)}(0) > 1)$ [37, 38]. In addition, filtering half-way between the central and side peaks has revealed the existence of weak “leapfrog” two-photon transitions that exhibit strong bunching [41, 42].
The aforementioned studies were performed with broad filtering ($\Gamma > \gamma$), aside from Ref. [41] where weak bunching ($g^{(2)}(t) \sim 1.2$) was observed when the Rayleigh peak was filtered at $\Gamma \sim 0.25 \gamma$. Here, the large $\gamma$ of our sample facilitates thorough exploration of this regime. We begin by measuring $g^{(2)}(0)$ as a function of $\Omega_R$, filtering centered on the Rayleigh peak with $\Gamma = 0.29 \gamma$. The results (Fig. 4(a)) illustrate a surprising transition from antibunching to strong bunching with increasing $\Omega_R$.

To understand this result requires careful consideration of the relationship between the Rabi frequency $\Omega_R$ and the amplitude and filter transmission of the various components of the RF spectrum. The fraction ($\mathcal{F}$) of the filtered ($\Gamma = 0.29 \gamma$) spectrum arising from each Mollow triplet component is plotted against $\Omega_R$ in Fig. 4(b). At small $\Omega_R$, Eq. 1 dictates a large coherent fraction. Thus, the behavior in this region corresponds to Fig. 2(a); only weak antibunching is observed as the filter bandwidth $< \gamma$ removes the majority of the incoherent component. As $\Omega_R$ increases, the coherent fraction falls and the splitting of the Mollow triplet increases, reducing the transmission of the side peaks (green) through the filter. It is thus intuitive to expect a transition to the Poissonian statistics of the Rayleigh peak (blue) [37–39] that now dominates the filtered spectrum.

However, in the limit $\Gamma < \gamma$, the additional effect of “indistinguishability bunching” [15] also becomes relevant. This phenomena originates in the quantum fluctuations of the light field [43, 44] and has been observed to lead to photon bunching when filtering at less than the natural linewidth of a light source, even for a classical input state such as a laser [45]. In the case of the RF spectrum considered here, the filtering is narrow compared to the incoherent Rayleigh peak but still broad compared to the coherent component. As such, for larger $\Omega_R$ where side peak contributions are negligible, the filtered $g^{(2)}(0)$ of Fig. 4(a) is determined by competition between the Poissonian statistics of the coherent part (see Fig. 2(a)) and bunching originating from the narrowly filtered incoherent part. Therefore, as $\Omega_R$ increases, the decreasing coherent fraction allows the indistinguishability bunching effect to dominate, leading to the strong bunching observed for large $\Omega_R$ in Fig. 4(a).

Our theoretical model (solid line in Fig. 4(a)) reproduces well the experimental results and predicts a maximum bunching of $g^{(2)}(0) \sim 2.1$ for these parameters. Experimentally, measurements cannot accurately be made at $\Omega_R > 4\gamma$ owing to increasing laser background. We note that theoretical studies [15] predict an ultimate upper limit of $g^{(2)}(0) = 3$ reached at $\Omega_R = 150\gamma$ and...
Γ = 0.005γ. For solid-state emitters such as the QD studied here, this value may not be reached owing to phonon-mediated interactions that cause the coherent fraction to revive at large Ω_R [36].

To further investigate filtering in the strong driving regime, Fig. 4(c) presents a filter width dependence at constant Ω_R = 2γ. At Γ ≪ γ, antibunching is observed in accordance with the expectation for unfiltered RF. The antibunching in this region is degraded due to the period of the Rabi oscillations in g^2(t) (see Fig. 4(c) inset) being shorter than the detector IRF. Fig. 4(d) shows the fraction (F) of the filtered spectrum arising from each component for Ω_R = 2γ. As Γ becomes comparable to γ in the central region of Fig. 4(c), there is a transition to bunched photon statistics in accordance with Fig. 4(a). This transition originates in the removal of the Mollow side peaks (green) from the filtered spectrum as Γ decreases, combined with the onset of the indistinguishability bunching effect previously described.

As Γ ≪ γ is approached on the left-hand side of Fig. 4(c), g^2(0) transitions again towards the Poissonian statistics that were observed for Γ < γ in Fig. 2(a). The interpretation here is also the same; for such small Γ the filtered spectrum contains almost solely coherent scattering (red line in 4(d)) which exhibits Poissonian statistics when spectrally isolated. Ultimately, for very narrow filters of bandwidth comparable to the laser linewidth (∼0.005γ), bunching would be expected to return due to indistinguishability bunching associated with the coherent part of the spectrum. Our theoretical model (green area in Fig. 4(c)) successfully reproduces this behaviour, incorporating both the detector IRF and lower and upper bounds corresponding to the uncertainty (0–20%) in the laser background contribution to the total signal. It is interesting to note that the upper bound incorporating a 20% background exhibits stronger bunching than the lower bound, indicating the non-trivial effect of introducing an additional Poissonian background.

In summary, we have demonstrated that the resonance fluorescence spectrum of a two-level emitter comprises multiple interfering components that each exhibit distinct photon statistics. Without filtering, these components always interfere to produce the strong antibunching expected from single quantum emitters. However, when spectrally filtering with bandwidth comparable to the natural linewidth (γ) or Rabi frequency (Ω_R), the ratio of these components is modified in the filtered spectrum, leading to strongly modified photon statistics. For weak resonant driving, a suitably narrow filter removes nearly the entire incoherent component, destroying the antibunching and illustrating that a sub-natural linewidth and strong antibunching cannot be simultaneously measured. For strong resonant driving, a pronounced bunching effect is observed at filter bandwidths comparable to the natural linewidth before the system ultimately trends towards Poissonian statistics for the narrowest filters. These results illustrate a potential new approach to manipulate the photon statistics of quantum light. In addition, we emphasise that care is required to preserve antibunching when filtering the spectrum of quantum emitters, an important consideration for future high throughput quantum networks where techniques such as wavelength-division multiplexing will be required.

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THE SENSOR FORMALISM

The sensor theory approach to calculating frequency-filtered N-photon correlation functions relies on enlarging the system Hilbert space to include N auxiliary two-level systems that act as sensors of the emitted photons. The formalism presented in this Supplement is based on Refs. [25, 26], extended to include the effects of laser background.

We consider a laser-driven quantum dot (QD) as a two-level system with ground state $|g⟩$ and excited state $|e⟩$, split by energy $\epsilon$. In a frame rotating at the laser driving frequency and within the rotating-wave approximation on the driving, the QD Hamiltonian may be written (we take $\hbar = 1$ throughout)

$$H_{QD} = \nu |e⟩⟨e| + \frac{\Omega_R}{2} \sigma_x,$$

(S1)

where $\Omega_R$ is the Rabi frequency, $\nu = \epsilon - \omega_l$ is the QD-laser detuning, and $\sigma_x = |e⟩⟨g| + |g⟩⟨e|$. The driven QD is coupled to the electromagnetic field, which has the free Hamiltonian

$$H_{EM} = \sum_k \omega_k a_k^{\dagger} a_k.$$  

(S2)

The QD-field interaction, again in the rotating frame and within the rotating-wave approximation, is written

$$H_{QD-EM} = \sum_k (g_k \sigma^+ a_k e^{i\omega_l t} + g_k^* \sigma^- a_k^{\dagger} e^{-i\omega_l t}).$$

(S3)

Here, $a_k$ annihilates a photon in mode $k$ of the electromagnetic field, which couples to the QD with strength $g_k$, and $\sigma = |g⟩⟨e|$.  

We may trace out the electromagnetic environment following the standard Born-Markov-secular approach assuming an electromagnetic field at zero temperature [30].

This allows us to derive an optical master equation governing the QD dynamics in the rotating frame:

$$\dot{\rho}_{QD} = -i[H_{QD}, \rho_{QD}] + \gamma \mathcal{L}_\sigma (\rho_{QD}).$$

(S4)

Here, $\rho_{QD}$ is the QD reduced density operator after tracing out the electromagnetic field, $\gamma = 1/T_1$ is the QD spontaneous emission rate, and

$$\mathcal{L}_\sigma (O) = \alpha O a^{\dagger} - \frac{1}{2} \{ a^\dagger a, O \},$$

(S5)

is the dissipator with anti-commutator $\{\cdot, \cdot\}$. We have neglected small energy shift terms that can anyway be absorbed into the definitions of $\nu$ and $\Omega_R$. The optical master equation is valid close to resonance, $\omega_l \sim \epsilon$, and for $\Omega_R \ll \epsilon$, both of which are fulfilled within the experiments.

To include the two-level sensors we enlarge our system Hilbert space, such that the system Hamiltonian now becomes (once more in the rotating frame and rotating-wave approximation)

$$H_S = H_{QD} + \sum_{i=1}^2 \nu_i \theta_i^\dagger \theta_i + \eta_i (\sigma \theta_i^\dagger + \sigma^\dagger \theta_i) + b_i \eta_i (\theta_i^\dagger + \theta_i),$$

(S6)

where $\theta_i = |g_i⟩⟨e_i|$ is the lowering operator for sensor $i$, which is centred at (and thus filters around) frequency $\omega_i$, with $\nu_i = \omega_i - \omega_l$. The QD-sensor coupling is given by $\eta_i$, which will eventually be taken to be vanishingly small. We also include laser background terms with strengths $b_i$, modelled phenomenologically as direct excitation of the sensors. These terms are necessary due to the imperfect polarisation rejection of the excitation laser within the experiments. We consider two sensor systems, as our primary interest is in second-order photon correlations.

Given that the QD-sensor couplings are vanishingly small, the form of the QD emission dissipator $\mathcal{L}_\sigma$ is un-
affected by the presence of the sensor systems. Likewise, broadening of the sensors, and hence the filter width, can be described by dissipators acting individually on each sensor system, such that our master equation now becomes [25, 26]

$$\dot{\rho}_S = -i[H_S, \rho_S] + \gamma L_\sigma(\rho_S) + \Gamma_1 L_{\theta_1}(\rho_S) + \Gamma_2 L_{\theta_2}(\rho_S),$$

(S7)

where $\Gamma_i$ is the filter width for sensor $i$. The frequency- and time-resolved two-photon correlation function is given by [26]

$$g^{(2)}_{\nu_1, \nu_2}(\nu_1, T_1; \nu_2, T_2) = \lim_{\eta_1, \eta_2 \to 0} \frac{\langle : n_1(T_1)n_2(T_2) : \rangle_T}{\langle n_1(T_1)\rangle \langle n_2(T_2) \rangle},$$

(S8)

where $n_1(t) = \theta_1(t)\theta_i(t)$, and $\langle : \cdot : \rangle_T$ indicates normal and time ordering.

The experiments are performed under resonant continuous driving conditions, such that $\nu = 0$ in Eq. S1, with the filters centred on the QD transition, meaning that $\nu_1 = 0 = \nu_2$ as well. In the steady-state, the second-order photon correlation function then becomes

$$g^{(2)}_{\nu_1, \nu_2} = \lim_{\eta \to 0} \frac{\text{tr}\{n_2(\nu)\theta_i(0)\rho_S(\infty)\theta_i(0)\}}{\langle n_1(0) \rangle_{ss} \langle n_2(0) \rangle_{ss}},$$

(S9)

where $\rho_S(\infty)$ is the steady-state density operator of the combined system of the QD and sensors, $\langle n_i(0) \rangle_{ss} = \text{tr}[n_i(0)\rho_S(\infty)]$, we set $\Gamma_1 = \Gamma_2 = \Gamma$ as the filter width, and $n_1 = n_2 = \eta$. Multi-time correlation functions are calculated using regression in the standard way [30].

The parameters $\Omega$, $\gamma$, and $\Gamma$ are known in the experiments, and so take fixed values within the theoretical calculations. The only free parameter is the background level, $b_1 = b_2 = b$, which is used as a fitting parameter for the insets of Fig. 4 (a) and (c), as well as to give the confidence bounds in the main panel of Fig. 4 (c). When considering the detector instrument response we convolve $g^{(2)}_\nu(\tau)$ with the Gaussian function

$$I(t) = \frac{2}{\delta t} \sqrt{\ln 2/\pi} e^{-4\ln 2(t/\delta t)^2},$$

(S10)

where $\delta t$ gives the full-width half-maximum.

**ELECTRON-PHONON COUPLING AND $g^{(2)}(t)$**

Usually, phonon coupling plays a prominent role in determining the characteristics of QD emission and the associated photon correlation functions [31, 32]. This is through three mechanisms, Rabi frequency renormalisation [28], excitation induced dephasing [34], and phonon sideband emission [31, 32].

In the first of these mechanisms, the dipole of the quantum emitter is dressed by modes of the phonon environment [28]. This results in the effective Rabi frequency being reduced from the value expected in the absence of phonons. It is thus this renormalised Rabi frequency which is observed in the experiments presented in the main manuscript, and so the renormalisation effect is already implicitly included in the model by our use of the observed Rabi frequencies in the theoretical calculations.

In contrast, excitation induced dephasing has negligible impact on the measured values of $g^{(2)}$. This is due to the nature of such correlation measurements; the second order optical coherence of a quantum emitter principally probes the level structure of the emitter, and is naturally insensitive to coherence and dephasing. To emphasise this point, Fig. S1 compares $g^{(2)}$ as a function of time in the absence of filtering as calculated using a full phonon theory based on the polaron formalism [29], and a simple atomic theory using the renormalised Rabi frequency as outlined above. As can be seen, the resultant dynamics are identical, suggesting that excitation-induced phonon effects are negligible for the quantities of interest in this work.

Finally, a phonon sideband is also present in the emission of the QD [31, 32]. From the measured data in the regions where $\Gamma > 10 \gamma$ in Figs. 2(a) and 4(c), it is clear that filtering the sideband has no impact on the phonon statistics. We interpret this as being a consequence of the nature of the sideband. Each photon emitted through the sideband is naturally correlated with a phonon, with the state becoming mixed when one traces

![FIG. S1. The second order correlation function calculated including phonons through a polaron theory approach (solid), and a simplified atomic theory in the absence of phonons. In keeping with the measured sample, the spontaneous emission rate is $\gamma = 20 \mu eV$ and the renormalised Rabi frequency is $\Omega_R = 2\gamma$. The phonon parameters were found in Ref. [32] through fitting to the emission spectra of the same QD used in the present experiments. The temperature is set to 4 K.](image-url)
out the phonon degrees of freedom. This prevents interference between the sideband and the zero-phonon line scattering of the quantum emitter, which would be necessary to see changes in the photon statistics.