





**Photon statistics of resonantly driven spectrally diffusive quantum emitters**Aymeric Delteil <sup>\*</sup>, Stéphanie Buil , and Jean-Pierre Hermier   
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In the solid state, a large variety of single-photon emitters present high-quality photophysical properties together with a potential for integration. However, in many cases, the host matrix induces fluctuations of the emission wavelength in time, limiting the potential applications based on indistinguishable photons. A deep understanding of the underlying spectral diffusion processes is therefore of high importance for improving the stability of the light emission. Here, we theoretically investigate the photon statistics of an emitter driven by a resonant laser, and subject to either of two qualitatively different stationary spectral diffusion processes—a continuous diffusion process and a process based on discrete spectral jumps, both of which are known to model the spectral diffusion of various solid-state emitters. We show that the statistics of light emission carries several experimentally accessible signatures that allow us to discriminate between the two classes of models, both at short times in the intensity correlation function, and at long times in the fluctuations of the integrated intensity. These results establish that resonant excitation combined with photon statistics offers a rich access to the spectral diffusion processes, yielding information that goes beyond the bare characterization of the inhomogeneous shape and noise correlation time. Incidentally, our findings shed light on recent experimental results of spectral diffusion of B centers in hexagonal boron nitride, providing more insight into their spectral diffusion mechanisms.

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Solid-state single-photon sources, such as self-assembled quantum dots and color centers in wide gap crystals, are widely seen as major actors in the emerging quantum technologies [1–3]. They can act like individual atoms trapped in a solid matrix, which enables integration into devices. In turn, these quantum emitters are also sensitive to the solid-state environment, which can manifest itself in decoherence and spectral diffusion (SD) of the emitter transition. The investigation of SD processes is therefore crucial for understanding and improving the performance of solid-state single-photon emitters for applications. In particular, fluctuations of the emitter wavelength limit the number of indistinguishable photons emitted by an artificial atom.

SD is associated with a stochastic process where the random variable  $\hbar\omega(t)$  describes the spectral position of the center energy. This varying emission line can be characterized using a variety of experimental techniques, depending on the amplitude and timescale of the fluctuations. When the characteristic time of SD is faster than the inverse count rate, spectroscopy falls short, such that more complex techniques have to be envisaged. Among the possible strategies, photon correlation Fourier spectroscopy [4–6] and sublinewidth filtering [7,8] can be used, albeit with intrinsic limitations in either temporal or spectral resolution. Delay-dependent Hong-Ou-Mandel interference can also be performed [9], but is limited to SPEs emitting close-to-indistinguishable photons

and require us to integrate coincidences separately for each time delay, which in many cases can turn out experimentally impractical.

Recently, we have experimentally demonstrated that a combination of resonant laser excitation and photon correlations allows us to establish the presence of SD as well as some of its characteristics [10]. Indeed, this technique simultaneously provides very high spectral and time resolutions. The resonant laser drive converts spectral fluctuations into intensity fluctuations, yielding photon bunching that can be measured through the intensity autocorrelation function  $g^{(2)}(\tau)$ . It offers the advantage to probe more than ten orders of magnitude of timescales in a single take and does not need indistinguishable photons, nor is it limited to zero-phonon-line emission. Such experimental work would benefit from a detailed theoretical investigation of the statistics of the emitted light and its connection with the microscopic process causing SD. Photon correlations are indeed well-known as a powerful tool to reveal rich signatures of a large variety of fundamental microscopic phenomena involving quantum emitters, such as single photon emission [11], indistinguishability [12], entanglement [13], superradiance [14], blinking [15], and phase transitions [16].

In the present paper, we investigate the relation between the SD process of a quantum emitter and the associated photon statistics under resonant excitation. We consider two emblematic stationary Markovian processes that yield Gaussian inhomogeneous broadening: the Ornstein-Uhlenbeck (OU) process and the Gaussian random jump (GRJ) process. While the former depicts a continuous SD of some solid-state emitters, the latter models emitters undergoing discrete spectral

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jumps. Section II describes the theoretical framework of these two SD mechanisms. Section III is devoted to the analysis of relevant statistical properties of resonantly scattered light when the emitter is subject to either stochastic process. We identify several experimentally measurable signatures that allow us to identify and characterize the diffusion process. Finally, in Sec. IV we discuss the obtained results and provide some insights into the SD process of B centers in hBN [17–19]. The framework and methods developed in the present paper can be straightforwardly extended to other SD models and other experimental protocols, as we also discuss in Sec. IV and in the Supplemental Material. Note that some quantum emitters can, in principle, also be affected by blinking. In most of the present paper, we disregard such cases. However, blinking can also affect photon statistics and should therefore be included in the model when applicable, as discussed in the Supplemental Material (Sec. S4).

## II. MODELS FOR SPECTRAL DIFFUSION PROCESSES

In the solid state, the frequency fluctuations of an emitter can have various origins depending on the nature of the emitter, the properties of the host material and its dimensionality. When it comes to processes yielding a static Gaussian inhomogeneous distribution, two main classes of noise are commonly identified, depending on whether the variations of the transition energy are continuous in time or experiences discrete spectral jumps at random times. These two models are described in the following.

### A. Continuous spectral diffusion: The Ornstein-Uhlenbeck process

In a wide range of physical systems, continuous diffusion can be well described by a OU process. The OU process is a Markovian and stationary Gaussian process and, as such, is completely characterized by its correlation function  $\langle \omega(t + \tau)\omega(t) \rangle = \Sigma^2 e^{-\tau/\tau_{SD}}$ , where  $\Sigma$  is the standard deviation of the associated Gaussian probability distribution function. Originally derived in the frame of nuclear magnetic resonance by Kubo and Anderson [20,21], it has been shown to also describe both charge noise and spin noise in self-assembled quantum dots [22–24] as well as SD in other condensed matter systems, such as ions or molecules embedded in solid-state matrices [25,26]. This process emerges when the light source is coupled to a large ensemble of identical and independent two-level fluctuators [Fig. 1(a)] and is equivalent to the Wiener model of Brownian motion at short times.

Figure 1(b) shows an energy trajectory generated numerically using the stochastic differential equation  $d\omega = \omega - dt(\omega - \omega_0)/\tau_{SD} + \Sigma\sqrt{t/2\tau_{SD}}W_t$ , where  $\hbar\omega_0$  is the center energy of the inhomogeneous distribution,  $\tau_{SD}$  the spectral diffusion correlation time, and  $W_t$  is the Wiener process (the continuous-time stochastic process that describes standard Brownian motion [27]). The numerical integration is performed based on the Euler-Maruyama method [28]. An example of a sample path is shown Fig. 1(b). The trajectories exhibit a continuous drift such that, after a time larger than the correlation time  $\tau_{SD}$ , the energy position is randomly found within a Gaussian envelope of standard deviation  $\Sigma$ .

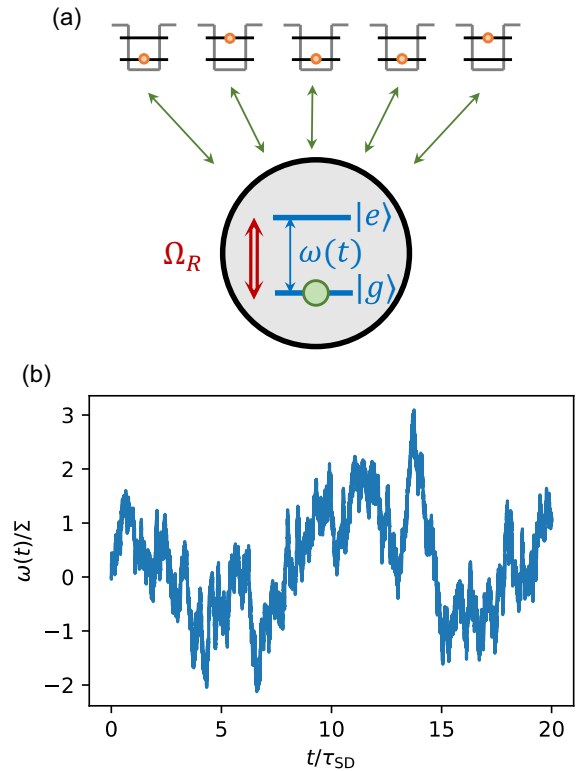


FIG. 1. (a) Sketch of an emitter coupled to a fluctuating environment modeled by an ensemble of identical two-level systems, yielding a OU diffusion process. (b) Numerically generated spectral trajectory.

### B. Gaussian random jump model

Spectral diffusion can also take the form of discrete jumps, occurring at random times governed by a Poisson process, and sampling a Gaussian distribution of standard deviation  $\Sigma$ . Such stochastic process is termed GRJ model by Spokoyny *et al.* [29]—we also adopt this terminology in the present paper. This process is also a Markovian and stationary process with correlation function  $\langle \delta\omega(t)\delta\omega(0) \rangle = \Sigma^2 e^{-t/\tau_{SD}}$ , and therefore cannot be distinguished from the OU model on the sole basis of its probability density function (PDF) and its correlation function. It has been used to describe spectral fluctuations of nitrogen-vacancy centers in nanodiamonds [30], molecules in crystalline matrices [31], color centers in hBN [29], nitride quantum dots [32], perovskite [33], and CdSe nanocrystals [6]—although, in the latter case, it is the spectral shifts (and not the spectral positions) that sample a Gaussian distribution, which makes little difference at short times but yields an additional long-time diffusion. Discrete jumps in SD typically emerge when an emitter is coupled to a low number of carriers migrating in a manifold of heterogeneous trap states [Fig. 2(a)] or a low number of neighboring systems whose configuration alternates within a continuum of states, such as ligand configurations in nanocrystals [6].

The generation of a spectral trajectory is conceptually simple. The emitter energy  $\hbar\omega(t)$  has a constant probability  $dt/\tau_{SD}$  to undergo a spectral jump during  $dt$ . When a jump occurs, the new frequency position is randomly drawn based on a Gaussian distribution of center energy  $\hbar\omega_0$  and standard

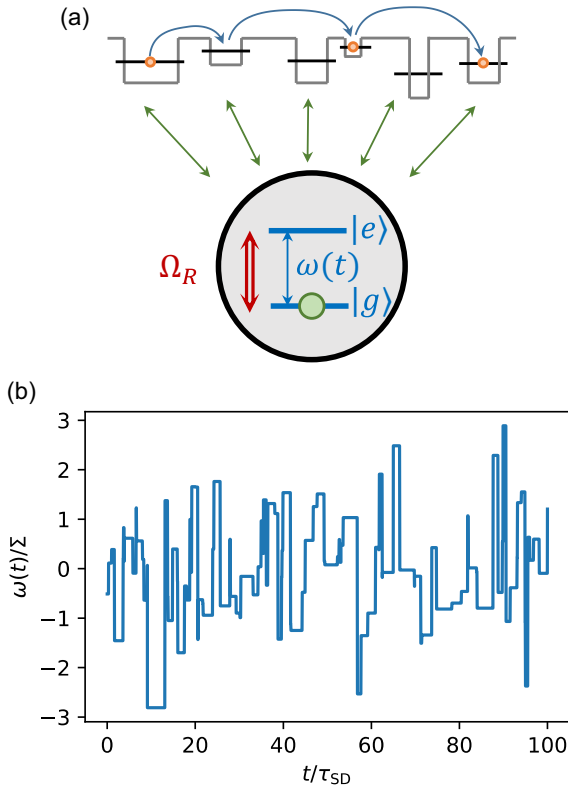


FIG. 2. (a) Sketch of an emitter coupled to a fluctuating environment modeled by a charge hopping between various trap states. (b) Numerically generated spectral trajectory.

deviation  $\Sigma$ . An example of a numerically generated trajectory is shown Fig. 2(b), where the discrete character of the spectral fluctuations is clear:  $\omega(t)$  is piecewise constant and changes values at intervals of order  $\tau_{SD}$ .

In the following section, we investigate the impact of these microscopic SD processes on the photon correlations of resonantly driven two-level emitters.

### III. PHOTON STATISTICS OF RESONANCE FLUORESCENCE

In the absence of SD, the population of a two-level atom driven by a laser close to resonance reaches a steady-state given by [34]

$$\rho_{ee} = \frac{1}{2} \frac{\Omega_R^2 T_1 / T_2}{(\omega - \omega_L)^2 + T_2^{-2} + \Omega_R^2 T_1 / T_2}, \quad (1)$$

where  $\Omega_R$  is the laser Rabi frequency,  $\hbar\omega_L$  its position in energy,  $T_1$  the emitter lifetime,  $T_2$  its coherence time, and  $\omega$  its center frequency.

The rate of scattered photons in the steady-state  $C_{ss}(\omega)$  is proportional to  $\rho_{ee}$ , such that the intensity response to the laser excitation is a Lorentzian of linewidth  $\Delta\omega_{\text{hom}} = 2\sqrt{1 + \Omega_R^2 T_1 T_2 / T_2}$ . Conveniently, the laser power provides an external knob to tune the linewidth of the homogeneous line shape through  $\Omega_R$ , a phenomenon known as power broadening.

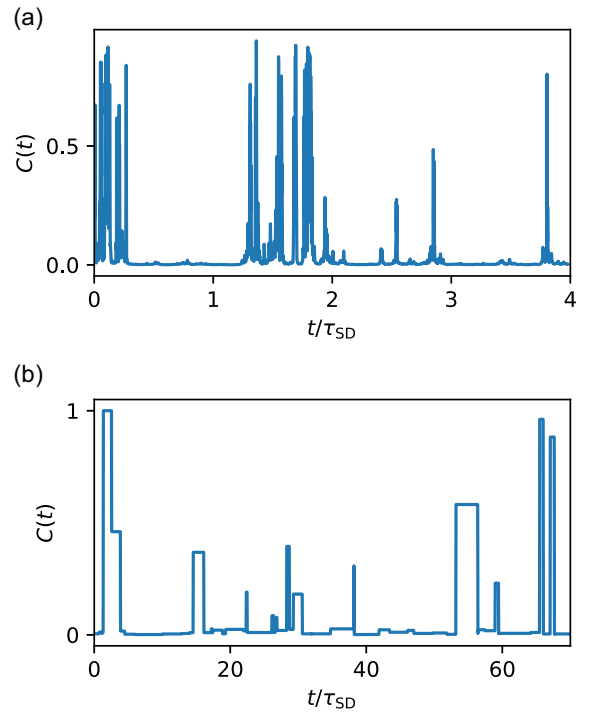


FIG. 3. (a) Intensity time trace generated with the OU model. (b) Intensity time trace generated with the GRJ model.

In the following, unless explicitly stated, we suppose  $\tau_{SD} \gg T_1, 1/\Sigma$ . This corresponds to cases where the spectral fluctuations are slower than both the emitter lifetime and the inverse inhomogeneous linewidth. Most experimental studies of solid-state emitters are consistent with these conditions, at the exception of some recent work demonstrating the achievement of fast SD that yields phenomena such as motional narrowing [22,35–37]. Out of these specific regimes, a continuously driven emitter is supposed to reach its steady state faster than the spectral fluctuations occur. This justifies the adiabatic approximation, in which the time-dependent intensity writes  $C(t) = C_{ss}[\omega(t)]$ .

Figure 3 shows two examples of intensity time traces generated with both a continuous and a jump process. In both cases, the signal exhibits an alternation of bright periods [when  $\omega(t) \approx \omega_L$ ] and dark periods, albeit with a qualitatively different time dependence. Experimentally, these time-intensity curves are impossible to measure as long as the count rate is smaller than the inverse timescale of the intensity fluctuations, which is the case in many practical situations. This justifies a general approach based on photon statistics.

#### A. Intensity autocorrelation

In this section, we focus on the intensity autocorrelation function  $g^{(2)}(\tau)$ , defined as

$$g^{(2)}(\tau) = \frac{\langle C(t)C(t+\tau) \rangle_t}{\langle C(t) \rangle_t^2} = \frac{\langle C_{ss}[\omega(t)]C_{ss}[\omega(t+\tau)] \rangle_t}{\langle C_{ss}[\omega(t)] \rangle_t^2}, \quad (2)$$

where  $\langle \dots \rangle_t$  denotes the time averaging and quantum effects at  $\tau \lesssim T_1$  are neglected. As evidenced in prior work [10], resonantly driving a spectrally diffusive emitter leads to bunching,

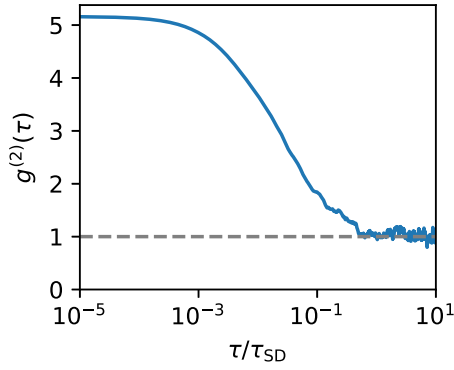


FIG. 4. Example of  $g^{(2)}(\tau)$  generated from a time trace shown Fig. 3. The gray dashed line depicts the Poissonian limit  $g^{(2)}(\tau) = 1$ .

owing to the fact that detection of a photon at time  $t_1$  informs on a close proximity between the emitter center frequency  $\omega(t_1)$  and the laser frequency  $\omega_L$ . Therefore, the photon detection rate at time  $t_2 \approx t_1$  is enhanced with respect to an uncorrelated case where  $|t_2 - t_1| \gg \tau_{SD}$ , which translates into bunching.

Figure 4 shows an example of the  $g^{(2)}$  function generated with an OU process. As expected, we observe  $g^{(2)}(0) > g^{(2)}(\tau)$ , indicating a bunching behavior resulting from the intensity fluctuations that originate from the SD. The asymptotic limit  $g^{(2)}(t \rightarrow \infty)$  is equal to 1 in the absence of additional random processes affecting the intensity stability, such as blinking. We define the degree of bunching  $B(\tau) = g^{(2)}(\tau) - 1$  and the normalized degree of bunching  $\tilde{B}(\tau) = B(\tau)/B(0)$ , which decays from 1 to 0 as  $\tau$  increases. We also define the bunching time  $\tau_b$  such as  $\tilde{B}(\tau_b) = 1/e$ . In the following, we will establish the relation between  $\tau_b$  and  $\tau_{SD}$  for the two classes of SD processes, after having reviewed some useful properties of the short-time bunching  $B(0)$ .

### 1. Short-time bunching

The short-time bunching can be deduced from the steady-state distribution of the emitter frequencies, independently of the underlying SD model [10]:

$$g^{(2)}(0) = \frac{\langle I^2 \rangle}{\langle I \rangle^2} = \frac{\int d\omega \mathcal{P}(\omega) C_{ss}(\omega)^2}{\left[ \int d\omega \mathcal{P}(\omega) C_{ss}(\omega) \right]^2}, \quad (3)$$

where  $\mathcal{P}(\omega)$  is the PDF associated with the inhomogeneous distribution. In the case of an inhomogeneous linewidth largely exceeding the natural linewidth, this further simplifies to

$$g^{(2)}(0) = \frac{1}{\mathcal{P}(\omega_L)} \frac{\int d\omega C_{ss}(\omega)^2}{\left[ \int d\omega C_{ss}(\omega) \right]^2} = \frac{1}{\mathcal{P}(\omega_L)} \frac{1}{\pi \Delta\omega_{\text{hom}}}, \quad (4)$$

where we have used Eq. (1) to express the count rate  $C_{ss}$  of a resonantly driven two-level system. When the PDF  $\mathcal{P}(\omega_L)$  is a Gaussian distribution, as is the case in the two models considered, we then obtain

$$g^{(2)}(0) = \frac{1}{2\sqrt{\pi \ln 2}} \frac{\Delta\omega_{\text{inhom}}}{\Delta\omega_{\text{hom}}} \exp \left[ 4 \ln 2 \left( \frac{\omega_L - \omega_0}{\Delta\omega_{\text{inhom}}} \right)^2 \right], \quad (5)$$

where  $\omega_0$  denotes the center of the inhomogeneous distribution and  $\Delta\omega_{\text{inhom}}$  its full width at half maximum ( $\Delta\omega_{\text{inhom}} = 2\sqrt{2 \ln 2} \Sigma$ ). At zero detuning, we then simply obtain  $g^{(2)}(0) = \frac{1}{2\sqrt{\pi \ln 2}} \frac{\Delta\omega_{\text{inhom}}}{\Delta\omega_{\text{hom}}}$ . This expression can be interpreted intuitively since, in two-photon correlations, the first detection informs that the spectrally diffusing emitter is close to resonance—which at random times has a likelihood given by the ratio of homogeneous to inhomogeneous linewidth. This leads to an increased likelihood of detecting a second photon, which is maintained until SD shifts the emitter spectral position out of resonance. Remarkably, this result shows that it is possible to infer the homogeneous linewidth—and therefore the coherence time of the emitter—from a simple measurement of the inhomogeneous (static) line shape and the amount of bunching at a fixed laser detuning. Moreover, this estimation does not require any assumptions about the nature of the microscopic mechanism yielding SD. This result is all the more interesting as it is generally difficult to extract information about homogeneous properties in inhomogeneously broadened quantum systems, which can require complex experimental procedures such as spectral hole burning [38] and dynamical decoupling [39,40]. We note that, while Eq. (5) is derived in the limit of Gaussian inhomogeneous spectra, Eq. (3) provides  $\Delta\omega_{\text{hom}}$  in the general case, where any experimental time-averaged line shape can be accounted for numerically. We further discuss some of these considerations in the Supplemental Material (Sec. S2).

To illustrate the dependence of the short-time bunching on the homogeneous linewidth, in Fig. 5(a), we plot  $B(0)$  as a function of the laser power. The decrease of the amount of bunching as power increases, already observed experimentally [10], is a consequence of power broadening yielding a larger homogeneous response, as discussed in the previous section. The dependence of  $B(0)$  on the laser detuning is plotted on Fig. 5(a). In this situation, bunching increases with the detuning due to the decreasing likelihood for the emitter to be resonant with the laser when the detuning increases. More examples of calculations are given in the Supplemental Material (Sec. S2). In both cases, we verify numerically that, as expected, the observed behavior is independent of the SD mechanism and correlation time. This is, however, not the case of the bunching decay, as we expose in the following section.

### 2. Bunching decay

Since the amount of bunching  $B(0)$  is independent of the model and only depends on the laser detuning and power, we now focus on the normalized bunching  $\tilde{B}(\tau)$  and we fix the laser frequency to the center of the inhomogeneous distribution ( $\omega_L = \omega_0$ ). We first investigate the normalized intensity correlation function in the case of a continuous SD governed by a OU process. Figure 6 shows a numerical calculation of  $\tilde{B}(\tau)$  for various laser powers above saturation. Two important characteristics can be observed: First, the decay is not exponential. Second, the characteristic time  $\tau_b$  increases with the power. This latter observation demonstrates that the bunching timescale  $\tau_b$  is, in general, not equal to the correlation time  $\tau_{SD}$ . Since the considered model is continuous,  $\tau_b$  represents the characteristic time during which the emitter stays near resonance with the laser before SD detunes it significantly.

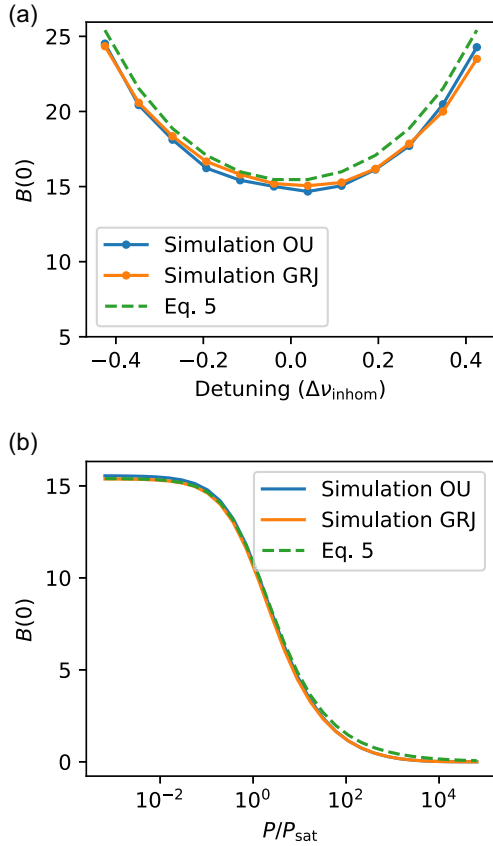


FIG. 5. (a) Zero-time bunching  $B(0)$  as a function of the laser detuning. (b)  $B(0)$  as a function of the laser power. In both simulations,  $\Delta\omega_{\text{hom}}/\Delta\omega_{\text{inhom}} = 2.2 \times 10^{-2}$ .

In presence of power broadening, it is therefore expected that the increased linewidth prolongs this duration when the laser power increases.

To gain more quantitative insight, we calculate the correlation function analytically, based on the following expression:

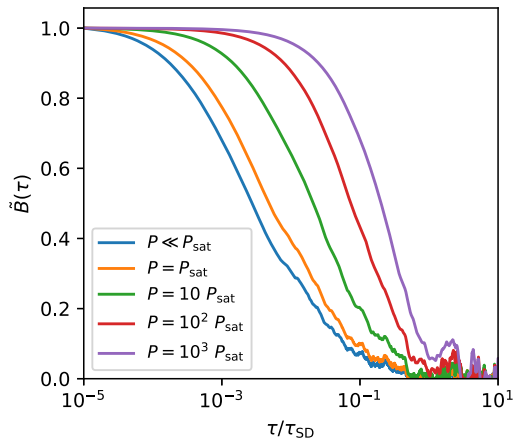


FIG. 6. Normalized bunching  $\tilde{B}(\tau)$  as a function of  $\tau$  in units of  $\tau_{\text{SD}}$  for various laser powers.

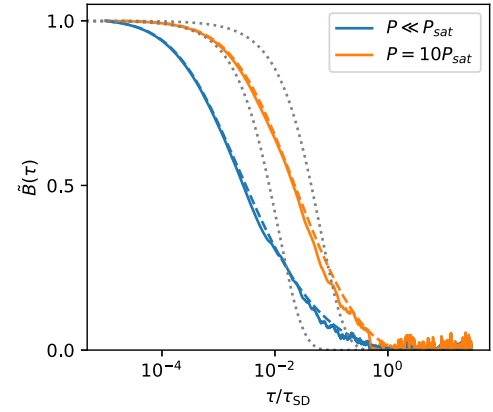


FIG. 7. Plain curves (dashed curves): Numerical simulation [analytical calculation using Eq. (8)] of  $\tilde{B}(\tau)$  as a function of  $\tau$  in units of  $\tau_{\text{SD}}$  for two laser powers. Dotted gray lines: Exponential fits of the simulations.

$$g^{(2)}(\tau) = \frac{1}{N} \iint d\omega d\omega' \mathcal{P}(\omega) C_{\text{ss}}(\omega) P(\omega', t + \tau | \omega, t) C_{\text{ss}}(\omega'), \quad (6)$$

where  $N = [\int \mathcal{P}(\omega) d\omega C_{\text{ss}}(\omega)]^2$ . We have introduced the SD kernel [41]

$$P(\omega', t' | \omega, t) = \frac{\exp\left(-\frac{\omega' - \omega e^{-(t'-t)/\tau_{\text{SD}}}}{2\Sigma^2(1 - e^{-2(t'-t)/\tau_{\text{SD}}})}\right)}{\sqrt{2\pi}\Sigma\sqrt{1 - e^{-2(t'-t)/\tau_{\text{SD}}}}}, \quad (7)$$

which expresses the conditional PDF for the emitter frequency to be found at position  $\omega'$  at time  $t'$  given position  $\omega$  at time  $t$ . In the limit where  $\Delta\omega_{\text{inhom}} \gg \Delta\omega_{\text{hom}}$ , the intensity correlation then reads

$$g^{(2)}(\tau) = \frac{1}{2\sqrt{\pi} \ln 2} \frac{\Delta\omega_{\text{inhom}}}{\Delta\omega_{\text{hom}}} f\left(\frac{2\Delta\omega_{\text{hom}}\sqrt{\ln 2}}{\Delta\omega_{\text{inhom}}\sqrt{1 - e^{-2\tau/\tau_{\text{SD}}}}}\right), \quad (8)$$

with

$$f(x) = \sqrt{\pi} x e^{x^2} \text{erfc}(x). \quad (9)$$

Figure 7 displays the analytical result of Eq. (8) together with the numerical simulation for two different powers (and therefore two different values of  $\Delta\omega_{\text{hom}}/\Delta\omega_{\text{inhom}}$ ). An exponential fit is also plotted to indicate the clear deviation from the exponential behavior. Based on Eq. (8), we can infer an approximate expression for  $\tau_b$  in the limit  $\Delta\omega_{\text{inhom}} \gg \Delta\omega_{\text{hom}}$ , which provides

$$\tau_b = K \left(\frac{\Delta\omega_{\text{inhom}}}{\Delta\omega_{\text{hom}}}\right)^2 \tau_{\text{SD}}, \quad (10)$$

with  $K \approx 18$ . This expression for  $\tau_b$  can be understood intuitively. At short times, the OU process is equivalent to the Wiener process of Brownian motion. Therefore, an emitter close to resonance at  $t = 0$  probes a spectral range growing as  $\sigma(t) \approx \Sigma\sqrt{t}/\tau_{\text{SD}}$ . It then escapes resonance when the standard deviation of SD becomes comparable to its homogeneous linewidth, i.e., for  $\sigma(t = \tau_b) \sim \Delta\omega_{\text{hom}}$ , yielding  $\tau_b \propto \tau_{\text{SD}}(\Delta\omega_{\text{hom}}/\Delta\omega_{\text{inhom}})^2$ .

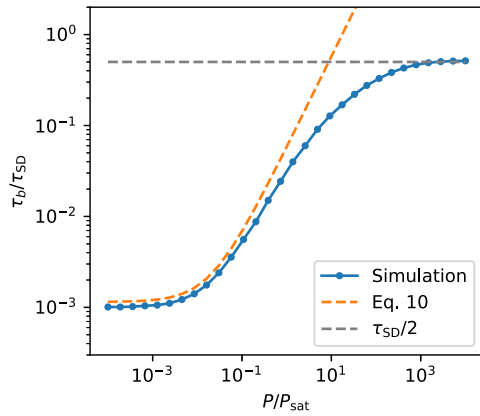


FIG. 8. Bunching time  $\tau_b$  as a function of the laser power for  $\Delta\omega_{\text{hom}}/\Delta\omega_{\text{inhom}} \approx 130$ . Blue line: Numerical simulation. Orange dashed line: Calculation from Eq. (10). Gray dashed line: High power asymptote  $\tau_b = \tau_{\text{SD}}/2$ .

Figure 8 shows the bunching time  $\tau_b$  as calculated numerically as a function of the laser power. At low powers, it is constant, given by Eq. (10). Above saturation,  $\tau_b$  increases due to power broadening prolonging the duration of bright periods, and then saturates when the homogeneous linewidth exceeds the width of the inhomogeneous distribution, with in this case  $\tau_b = \tau_{\text{SD}}/2$ . The factor 1/2 originates from the fact that, in the limit  $\Delta\omega_{\text{hom}} \gg \Delta\omega_{\text{inhom}}$ , the system is first-order insensitive to SD [24]. Therefore, at second order, fluctuations of  $\omega(t)$  at a frequency  $f$  yield fluctuations of  $C(t)$  at a frequency  $2f$  and thus the associated decay time is halved.

We now consider the GRJ model. Figure 9 shows the numerically calculated  $\tilde{B}(\tau)$  at various laser powers. Contrarily to the OU case, here the bunching decay time and shape do not vary with the power. This can be seen intuitively: if the emitter is on resonance with the laser at  $t = 0$ , it stays so on average during  $\tau_{\text{SD}}$ , independently of the laser power, before a spectral jump detunes the emitter transition energy. The laser power only acts on the bare probability to be close to resonance at a given time, and therefore on  $B(0)$ , as discussed in the previous section. As a consequence,  $\tilde{B}(\tau)$  directly inherits its time dependence from the SD correlation function  $\langle\omega(t + \tau)\omega(t)\rangle$

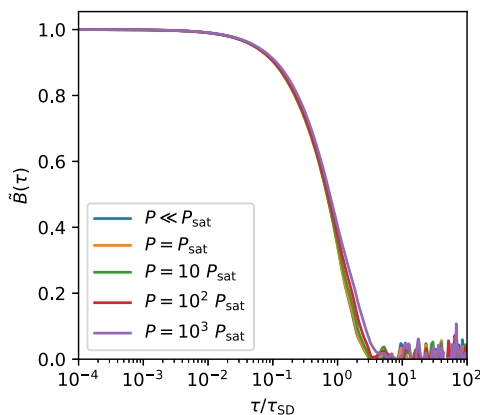


FIG. 9. Normalized bunching  $\tilde{B}(\tau)$  as a function of  $\tau$  in units of  $\tau_{\text{SD}}$  for various laser powers.

and exhibits an exponential decay of characteristic time  $\tau_b = \tau_{\text{SD}}$ , as verified in our simulations.

These observations establish that the long-time  $g^{(2)}(\tau)$  carries clear experimentally accessible signatures of the underlying SD stochastic process: In the case of the OU model,  $g^{(2)}(\tau)$  decays nonexponentially according to Eq. (8), and the decay time increases with the laser power above saturation. In the case of a GRJ mechanism,  $g^{(2)}(\tau)$  decays exponentially and the bunching time is independent of the power. Only in the latter case do we have  $\tau_b = \tau_{\text{SD}}$ .

We observe that a power-independent decay of  $g^{(2)}(\tau)$  is a general signature of discrete jumps of the emitter to uncorrelated spectral positions within the inhomogeneous distribution. The decay of  $g^{(2)}(\tau)$  is then directly inherited from the time statistics of the jumps and is exponential in the case of Poissonian jumps. On the opposite, a power-dependent decay time indicates that intermediate spectral positions of the emitter are probed increasingly as power broadens the emitter, and therefore constitutes a general signature of continuous SD mechanisms.

In the Supplemental Material (Secs. S4 and S5), we provide examples of emitters subject to more complex mechanisms, such as a combination of two SD components, as well as a contribution from blinking, thereby showing that our approach can, in principle, be generalized to a wide range of experimental situations.

## B. Intensity fluctuations

In this section, we investigate additional consequences of SD on the photon statistics of resonance fluorescence. We show that the long-time photon statistics are affected by SD in a way that is inherited from the microscopic model.

### 1. Standard deviation of the intensity distribution

At a fixed laser detuning, the average intensity—or count rate—that the emitter scatters is simply given by  $\langle C(\omega_L) \rangle = \int d\omega \mathcal{P}(\omega) C(\omega)$ . The count rate is typically measured by integrating the photon number over a macroscopic time  $T$ . This measured intensity fluctuates because of shot noise, with a relative intensity noise given by  $\Delta I_T / \langle I \rangle_T = 1/\sqrt{\langle N \rangle_T}$ , where  $\langle N \rangle_T$  is the average number of photons detected during  $T$ . These Poissonian fluctuations can be mitigated by optimizing the photon collection efficiency. Additional sources of noise can cause the variance  $V_T$  of the photon number during  $T$  to exceed the Poisson limit  $V_T = \langle N \rangle_T$ . The Mandel parameter  $Q = V_T / \langle N \rangle_T - 1$  relates the long-time intensity fluctuations to the second-order intensity correlation via the following relation [42–45]:

$$Q = \frac{\langle N \rangle_T}{T} \int_{-T}^T d\tau \left(1 - \frac{|\tau|}{T}\right) (g^{(2)}(\tau) - 1). \quad (11)$$

If  $g^{(2)}(\tau) = 1$ , we have  $Q = 0$  and the intensity noise is given by the shot noise. As can be seen from Eq. (11), the presence of finite bunching increases the intensity noise above the Poisson limit. Since SD is associated with photon bunching, as discussed in the previous sections, it is also expected to yield additional intensity fluctuations at macroscopic times ( $T \gg \tau_{\text{SD}}$ ).

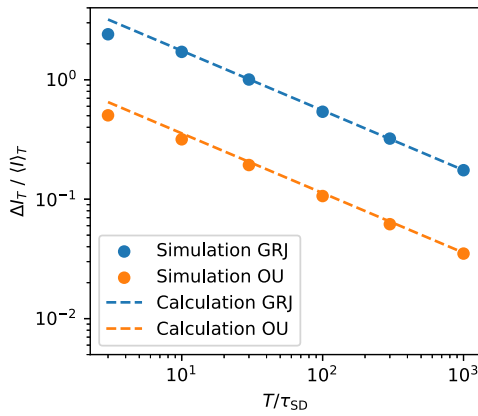


FIG. 10. Relative intensity noise as a function of the integration time  $T$ . Blue (orange) dots: Simulations for the GRJ (OU) model. Blue (orange) dashed lines: Calculation for the GRJ (OU) model using Eq. (14) [Eq. (13)].

To isolate the contribution of SD to the intensity noise, we now consider the high intensity (or high photon collection) limit where the shot noise can be neglected ( $\langle N \rangle_T \gg 1$ ). Since the latter is statistically independent from the SD noise, it can be reintegrated at a later step (see Supplemental Material Sec. S3 [46]). In this limit, the relative intensity fluctuations take the form

$$\frac{\Delta I_T}{\langle I \rangle_T} = \left[ \frac{1}{T} \int B(\tau) d\tau \right]^{\frac{1}{2}}, \quad (12)$$

which, when the laser is fixed at the center of the inhomogeneous distribution, writes

$$\frac{\Delta I_T}{\langle I \rangle_T} = \left[ \frac{1}{\sqrt{\pi \ln 2T}} \frac{\Delta \omega_{\text{inhom}}}{\Delta \omega_{\text{hom}}} \int \tilde{B}(\tau) d\tau \right]^{\frac{1}{2}}. \quad (13)$$

In the case of GRJ model, Eq. (13) further simplifies to

$$\frac{\Delta I_T}{\langle I \rangle_T} = \sqrt{\frac{1}{\sqrt{\pi \ln 2}} \frac{\Delta \omega_{\text{inhom}} \tau_{\text{SD}}}{\Delta \omega_{\text{hom}} T}}. \quad (14)$$

In the case of SD based on OU process, the integral in Eq. (13) has, to the best of our knowledge, no analytical solution. We therefore perform a numerical evaluation of this expression. Figure 10 shows the result of these calculations together with a numerical simulation for varying integration time, at low power. The simulations and the analytical calculations are in good agreement, in particular, for  $T \gg \tau_{\text{SD}}$ . The intensity fluctuations for the OU process are smaller than in the GRJ case, due to the fact that  $B(\tau)$  decays much faster in the former case. On the other hand, both have identical dependence on the integration time  $T$ . Therefore, the intensity noise at a given power is not sufficient to identify the SD process without prior knowledge of  $\tau_{\text{SD}}$ .

Figure 11 shows the power dependence of the relative intensity noise, as calculated from Eq. (13). In both cases, the fluctuations decrease with increasing power. However, in the case of the GRJ model, the reduction occurs around the saturation power, while in the OU model, the reduction occurs at a power such that  $\Delta \omega_{\text{hom}} \sim \Delta \omega_{\text{inhom}}$ , which is much higher in many practical situations where  $\Delta \omega_{\text{inhom}} \gg 1/T_2$ . The

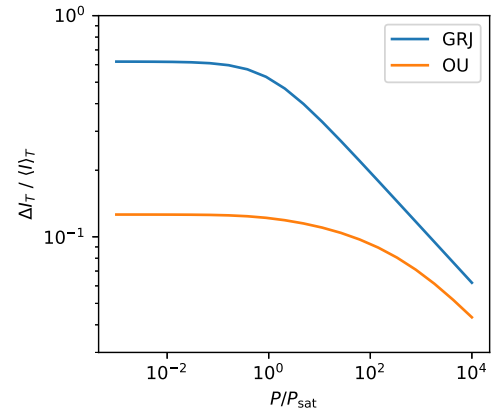


FIG. 11. Relative intensity noise as a function of the excitation laser power.

example of Fig. 11 is taken with  $\Delta \omega_{\text{inhom}}/\Delta \omega_{\text{hom}} \approx 50$ , leading to a twofold decrease at  $P \approx 10^3 P_{\text{sat}}$  in the OU case. Therefore, knowing the saturation power, the power dependence of the intensity fluctuations constitutes a macroscopic-time manifestation of the underlying microscopic SD mechanism.

## 2. Skewness of the intensity distribution

Another informative aspect carried in the long-time intensity fluctuations is the skewness of the intensity distribution. The skewness quantifies the asymmetry of a distribution and is defined as

$$\gamma_1 = \left\langle \left( \frac{I - \langle I \rangle_T}{\Delta I_T} \right)^3 \right\rangle. \quad (15)$$

For a Poisson distribution,  $\gamma_1 = \Delta I_T / \langle I \rangle_T$ . Figure 12 plots the skewness of the time trace used in the previous section, normalized by the intensity noise. In the OU model, we obtain a close to Poissonian skewness  $\gamma_1 \approx \Delta I / I$  for all bin sizes. On the contrary, in the GRJ model, we observe super-Poissonian skewness  $\gamma_1 \approx 2\Delta I / I$ , demonstrating an additional impact of the SD mechanism on the statistics of the integrated intensity.

The super-Poissonian skewness of the intensity distribution in the case of the GRJ model can be well understood

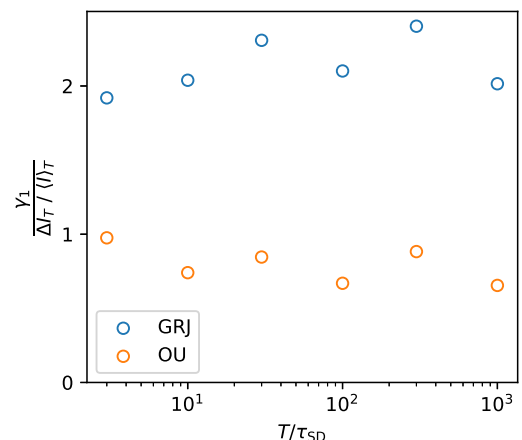


FIG. 12. Relative skewness as a function of the integration time.

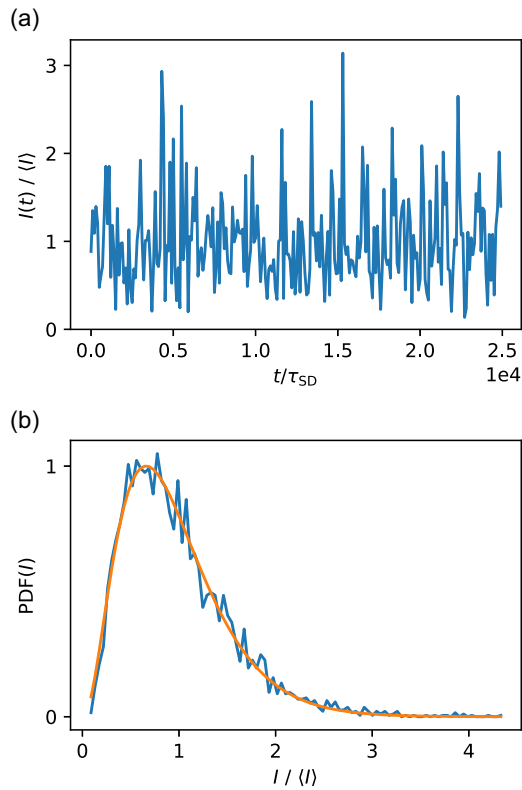


FIG. 13. (a) Intensity time trace with integration time  $T = 100\tau_{\text{SD}}$ . (b) Blue curve: Histogram of the intensities. Orange curve: Fit using a gamma distribution.

considering that the duration of the stable periods—as defined as the time between two jumps—is exponentially distributed, an elementary property of Poisson processes. During the time  $T_{\text{bin}}$ , there are, on average,  $T_{\text{bin}}/\tau_{\text{SD}}$  stable periods. The fraction of them which are bright (i.e.,  $\omega \approx \omega_L$ ) is  $\sim \Delta\omega_{\text{hom}}/2\sqrt{\pi \ln 2} \Delta\omega_{\text{inhom}}$ , where we have supposed a Gaussian PDF and zero detuning ( $\omega_L = \omega_0$ ). The average number of bright stable periods during  $T_{\text{bin}}$  is  $N_b = (T_{\text{bin}}/\tau_{\text{SD}})\Delta\omega_{\text{hom}}/2\sqrt{\pi \ln 2} \Delta\omega_{\text{inhom}}$ . By making the approximation that the number of bright periods is independent of their duration—which is only valid asymptotically—the intensity can then be written as a sum of exponentially distributed random variables. Therefore, the intensity is itself a random variable whose distribution is a gamma distribution of parameters  $N_b$  and  $\tau_c$ . This distribution has a skewness of  $\gamma_1 = 2\Delta I/I$ . Figure 13(a) shows an intensity trace, where the occurrence of short periods of brightness sizably higher than average can be observed as a manifestation of the asymmetry of the intensity distribution. Figure 13(b) shows the histogram of the integrated intensities over a long period, together with a fit using the gamma distribution, showing the excellent agreement with the simulation. Such a histogram can provide an estimation of the SD time, based on prior knowledge of  $\Delta\omega_{\text{hom}}/\Delta\omega_{\text{inhom}}$ . By using Eq. (14) together with the relation  $\gamma_1 = 2\Delta I/I$  valid for any gamma distribution, we obtain  $\tau_{\text{SD}} = \sqrt{\pi \ln 2} \frac{\Delta\omega_{\text{inhom}}}{\Delta\omega_{\text{hom}}} T (\gamma_1/2)^2$ , which we have verified provides back the input value used in the simulation, within 10%. For comparison, we also simulate a time

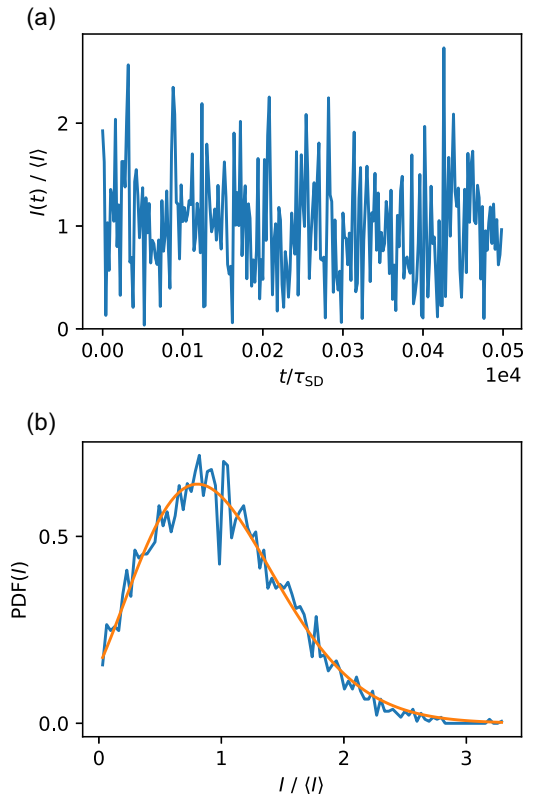


FIG. 14. (a) Intensity time trace generated using the OU model. (b) Blue curve: Histogram of the intensities. Orange curve: Fit using a Poisson distribution.

trace [shown in Fig. 14(a)] and the corresponding intensity histogram [Fig. 14(b)] in the case of the OU model. The latter can be best fitted with a Poisson distribution, consistent with the Poissonian skewness discussed above. Note the nonzero intercept in this case as opposed to the GRJ case. These observations suggest that measurement of super-Poissonian skewness in intensity traces provides evidence for a jump-based SD process, as well as an estimation of the SD time.

#### IV. DISCUSSION AND CONCLUSION

We have provided a general framework to describe and analyze the photon statistics of spectrally diffusive emitters resonantly driven by a cw laser. Our simulations, supported by analytical calculations, have evidenced several experimentally accessible signatures of the discrete or continuous nature of the stochastic process governing the variations of the emitter transition energy in time. Two of these characteristic signatures are associated with the intensity correlation function  $g^{(2)}$ . As a general rule, a decrease in the amount of bunching that is not accompanied by an increase in the bunching decay time is characteristic of discrete jumps to uncorrelated positions. In this regard, a recent experimental study has established the presence of power-dependent bunching in the  $g^{(2)}$  of a B center in hBN [10]. In light of the results exposed in the present paper, we can attribute the SD mechanism of B centers to a discrete jump process, owing to the fact that the bunching timescale is almost independent of the power above



saturation (up to  $P \approx 100P_{\text{sat}}$ ). This observation is consistent with what has been demonstrated with other color centers in hBN based on photon correlation Fourier spectroscopy [29]. In the latter reference, the GRJ process is evidenced by a direct measurement of the spectral correlation function, which would not be possible in the case of B centers due to their much narrower homogeneous and inhomogeneous widths. As a property of the GRJ model that we have established in Sec. III, the bunching time is equal to the SD time, which in the case of B centers has been measured to be  $\sim 30 \mu\text{s}$  [10]. The present paper provides guidelines for a more in-depth investigation of SD in these systems, as well as in a large variety of other single-photon sources. It can be readily extended to other potential microscopic models, as well as to other experimental processes. As an example, an alternative method that does not require resonant excitation is to filter the emitter fluorescence with a bandwidth narrower than the inhomogeneous linewidth. In the Supplemental Material (Sec. S1) [46], we investigate the statistical properties of light filtered from nonresonantly excited emitters. We show that it shares some similarities with resonance fluorescence, where the spectral selectivity is implemented by the resonant laser. It therefore carries analogous signatures. For instance, observation of a bunching time independent of the filter width implies discrete jumps to the whole inhomogeneous distribution. In

addition, the bunching time associated with continuous diffusion is also shorter than the SD time, which possibly modifies the conclusions of prior work using this technique [8]. We also present examples of more complex situations, where the emitter subject to blinking (Sec. S4), or where the emitter subject to more than one SD mechanism (Sec. S5) illustrating thereby the versatility of our approach. We note that, in the presence of blinking, more detailed studies will be needed to properly disentangle the effect of both mechanisms in the photon statistics.

Given the ubiquity of SD processes in condensed matter as well as the importance of resonance fluorescence in the study and control of solid-state quantum emitters, we expect our paper to play a helpful role in the general understanding and technological development of quantum dots and color centers, with applications to quantum computing and quantum networks.

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