

Superbunching in cathodoluminescence: A master equation approach

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We propose a theoretical model of a master equation for cathodoluminescence (CL). The master equation describes simultaneous excitation of multiple emitters by an incoming electron and radiative decay of individual emitters. We investigate the normalized second-order correlation function, $g^{(2)}(\tau)$, of this model. We derive the exact formula for the zero-time delay correlation, $g^{(2)}(0)$, and show that the model successfully describes giant bunching (superbunching) in the CL. We also derive an approximate form of $g^{(2)}(\tau)$, which is valid for small excitation rate. Furthermore, we discuss the state of the radiation field of the CL. We reveal that the superbunching results from a mixture of an excited photon state and the vacuum state and that this type of state is realized in the CL.

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I. INTRODUCTION

In electron microscopy, cathodoluminescence (CL) visualizes optical properties beyond the diffraction limit of light. A wide range of materials can be investigated by this approach, for instance, defect or luminescence centers in semiconductors [1–4], quantum-confined structures [5–7], surface plasmon polaritons [8–10], and fluorescent proteins [11–13]. Thus, the electron-microscopy-based CL measurement is a powerful tool to analyze various materials on nanoscale.

The optical state of CL itself has not been in the spotlight for a long time though CL had been used in displays with cathode ray tubes for more than a century. By the recent introduction of Hanbury Brown-Twiss (HBT) interferometry to CL, the quantum character “antibunching” of the emitted states of CL has been revealed with a deep sub-wavelength spatial resolution in the measurement of a single nitrogen-vacancy (NV) center in a nanodiamond [14]. Since antibunching is a result of the particle nature of a photon, this HBT-CL technique opens a way to measure quantum optical phenomena on the nanoscale.

However, the HBT measurement of CL from multiple defect centers has presented strong bunching [15], which has not been observed in photoluminescence (PL) experiments for the same kind of sample. Although there are differences between optical and electron-beam excitations such as absence of the NV[−] spectrum in CL [16], this bunching observation raises a question on the origin of the bunching. In addition, the observed bunching in CL is often huge, where the normalized second-order correlation function, $g^{(2)}(\tau)$, at time

delay $\tau = 0$ is larger than 2, i.e., superthermal values. This is known as superbunching and a peculiar state of light. A representative example of the superbunching is spontaneous parametric down-converted light (a squeezed vacuum), which is the quantum light widely used as heralded single photons and entangled photon pairs [17]. Other examples are super-radiant coupling of the emitters [18–20], quantum dot–metal nanoparticles [21,22], and bimodal lasers [23–27].

The bunching in CL has already enabled us to measure the luminescent lifetime well below the optical diffraction limit without a pulsed electron beam [28]. This time-resolved measurement not only demonstrated the Purcell effect on the nanoscale [29,30] but also quantified excitation and emission efficiencies of optical nanostructures [31,32]. These practical applications prove that the CL photon correlation has great potential to access intrinsic nanophotonic properties in a direct manner and offer important insights into nanophotonic devices. Therefore, it is important to clarify how the strong bunching emerges in CL from both basic and applied aspects. The deeper understanding of CL photon correlation should progress nanoscale optical imaging to the next stage.

There were studies on CL photon statistics about half a century ago [33,34]. These pioneering investigations presented a theoretical description of the photon statistics and an experimental observation of the strong intensity correlation. However, the feature of $g^{(2)}$ in CL was not focused. In the first report of the superbunching in CL [15], Meuret *et al.* assumed that plasmons induce a synchronized excitation of multiple emitters and proposed a stochastic model to perform a Monte Carlo simulation on $g^{(2)}(\tau)$. On the basis of similar assumptions, CL excitation efficiency was estimated [31], and an analytical model was constructed [35]. Feldman *et al.* claimed that the bunching in the nanodiamond CL is mediated by the phonon sidebands and explained $g^{(2)}(\tau)$

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using another Monte Carlo model [36]. Besides these models, Yanagimoto *et al.* derived an expression of $g^{(2)}(\tau)$ using a rate equation for multiple two-level systems [30]. However, the models in all the previous studies are essentially classical, and no quantum model has been proposed that explains the superbunching in CL. Even if photon bunching can be described by classical electromagnetic waves, the lack of photon picture would obscure the understanding of the essence of CL photon correlation.

In this study, we introduce a model of a quantum master equation (QME) to describe the dynamics of multiple emitters in CL. The excitation by incident electrons is incorporated phenomenologically in the QME. We find that the QME is reduced to a semiclassical master equation for the distribution of number of excited emitters. From this master equation, we exactly obtain the stationary distribution and the formula for zero-time delay correlation $g^{(2)}(0)$. We also derive an approximate equation for delay time-dependent correlation $g^{(2)}(\tau)$. We show that these results successfully reproduce several features of CL, in particular, the superbunching and the decaying behavior of $g^{(2)}(\tau)$. Moreover, we extend the model so that it is applicable to the case of large electron-beam current. We also deduce the state of the radiation field from a possible sequence of pulses of the field in the CL. From the model calculation and the deduced argument for the radiation field, we shed light on a universal aspect of the superbunching.

II. MODEL

A. Excitation process in cathodoluminescence

Before describing the quantum master equation of our model, we briefly explain the excitation process in a material by fast incident electrons in CL.

In CL, it is considered that an incident electron excites multiple emitters (defect centers) not directly but via several steps of elementary processes mainly due to bulk plasmons and/or secondary electrons [15,31,37–41]. The timescale required to excite emitters ranges from femtoseconds (bulk plasmon) to picoseconds (secondary electrons). Therefore, when the radiative lifetime τ_{rad} of the emitters is on the order of nanoseconds (we consider this case in the present study), the excitation timescale is sufficiently smaller than the emitter lifetime.

The region excited by the electron beam extends from the beam path. Its size depends on the beam diameter, the generation range of secondary (quasi)particles, and the mean free path and/or diffusion length of secondary (or higher-order) carriers. The typical length scale of the excited region is several tens of nanometers when using a thin sample.

B. Quantum master equation

Considering the above excitation process by electron beam, we propose the following quantum model of CL.

The system of our interest is composed of N emitters, which are located in the excited region. Each emitter is modeled by a two-level system (TLS) with transition energy $\hbar\omega_e$. We assume that the density of emitters is low and thus the interaction among them is absent. Therefore, the system

Hamiltonian is given by

$$\hat{H} = \sum_{j=1}^N \frac{\hbar\omega_e}{2} \hat{\sigma}_j^z. \quad (1)$$

Here $\hat{\sigma}_j^z$ is the z component of the Pauli matrix for the j th TLS. This is expressed as $\hat{\sigma}_j^z = |1\rangle_j\langle 1| - |0\rangle_j\langle 0|$ with the lower-level state $|0\rangle_j$ and the upper one $|1\rangle_j$ of the j th TLS.

In this study, we consider CL emission with a continuous electron beam. The emitters are continuously excited by the incoming electrons and decay with photon emission. The Lindblad-type QME [42–45] is a suitable method for describing dynamics in this situation. The QME has the following form:

$$\frac{d}{dt}\hat{\rho}(t) = \mathcal{L}\hat{\rho}(t), \quad (2)$$

where $\hat{\rho}(t)$ is the state (density matrix) of the system at time t and the Liouvillian \mathcal{L} is given by

$$\mathcal{L}\hat{\rho} = \frac{1}{i\hbar}[\hat{H}, \hat{\rho}] + \mathcal{D}_{\text{rad}}\hat{\rho} + \mathcal{D}_{\text{ex}}\hat{\rho}. \quad (3)$$

The first term represents the unitary part of the time evolution with the system Hamiltonian (1). The second and third terms represent the nonunitary parts due to the decay and excitation, respectively, as explained below.

The second term in the Liouvillian (3) describes the radiative decay of the emitters. We here assume that the dipole moments of the emitters are randomly oriented. In this case, they are independently damped even though the emitters excited by the electron beam are located within the excited region, which is smaller than the wavelength $2\pi c/\omega_e$. Therefore, as in the quantum optical master equation [44–46] under the assumption that the reservoir temperature is sufficiently smaller than $\hbar\omega_e$, the second term is given in the following Lindblad form:

$$\mathcal{D}_{\text{rad}}\hat{\rho} = \frac{1}{\tau_{\text{rad}}} \sum_{j=1}^N \left(\hat{\sigma}_j^- \hat{\rho} \hat{\sigma}_j^+ - \frac{1}{2} \{ \hat{\sigma}_j^+ \hat{\sigma}_j^-, \hat{\rho} \} \right). \quad (4)$$

Here $\hat{\sigma}_j^+ = |1\rangle_j\langle 0|$ and $\hat{\sigma}_j^- = |0\rangle_j\langle 1|$ are the raising and lowering operators of the j th TLS, respectively. And τ_{rad} is the radiative lifetime of each emitter. We note that we can incorporate the nonradiative decay in the same Lindblad form, in which case we should replace the prefactor $1/\tau_{\text{rad}}$ with $1/\tau_{\text{tot}} = 1/\tau_{\text{rad}} + 1/\tau_{\text{nonrad}}$ to include the nonradiative lifetime τ_{nonrad} .

The third term in the Liouvillian (3) describes the excitation of the emitters by an electron beam. As explained in Sec. II A, the excitation timescale for each electron is sufficiently smaller than the radiative lifetime τ_{rad} . Therefore, we can consider that the multiple emitters are excited *simultaneously* by a single incident electron. In this study, for simplicity, we assume that the number of emitters simultaneously excited by an electron is constant and is equal to N that is introduced in Eq. (1). On the other hand, the excitation rate γ is connected to the electron-beam current I . The unit-time number of electrons incident on the sample is (I/e) (e is the

elementary charge). And the excitation occurs $(I/e)p_{\text{ex}}$ times per unit time, where p_{ex} is the probability for the excitation by an electron. Therefore, $\gamma = (I/e)p_{\text{ex}}$. To incorporate this simultaneous excitation of the N emitters at the rate of γ , we introduce the following third term:

$$\mathcal{D}_{\text{ex}}\hat{\rho} = \gamma(\hat{\Pi}^+\hat{\rho}\hat{\Pi}^- - \frac{1}{2}\{\hat{\Pi}^-\hat{\Pi}^+, \hat{\rho}\}), \quad (5)$$

$$\hat{\Pi}^{\pm} = \bigotimes_{j=1}^N \hat{\sigma}_j^{\pm}. \quad (6)$$

The Lindblad operator $\hat{\Pi}^+$ of Eq. (6) raises all the TLSs to the upper levels if all of them are in the lower levels. This expresses the situation that the emitters are simultaneously excited by an incident electron. We note that this term induces no excitation when some of the TLSs are already in the upper levels. However, such a no-excitation event does not occur if the excitation rate γ is much smaller than the radiative damping rate $1/\tau_{\text{rad}}$ since in such cases all the TLSs are in the lower levels for most of the time. Therefore, this model is valid for $\tau_{\text{rad}}\gamma \ll 1$. If τ_{rad} is around 10 ns, this condition is fulfilled for the beam current I less than 10 pA.

Here, we make three remarks on this model. First, the number N of emitters excited by a single incident electron is independent of the current I while the excitation rate γ is proportional to I (as explained above). Instead, N depends on the energy of the electron (acceleration voltage) and on the sample parameters (film thickness, density of emitters, and so on).

Second, as explained for Eq. (5), we assume that the incident electrons always excite the same N emitters. In actual experiments of CL, the number of emitters excited by each electron varies around the average. We give an extension of the model to incorporate this effect in Sec. IV and the Supplemental Material [47] (Refs. [15,44,48–51] are included therein). We note that this effect does not essentially alter the results of the present model if its validity condition $\tau_{\text{rad}}\gamma \ll 1$ is satisfied. Moreover, the extended model can be well approximated by the present model for $\tau_{\text{rad}}\gamma \ll 1$, where N is regarded as the average number of emitters excited by an electron.

Finally, small excitation volume (i.e., high spatial resolution of the electron beam), one of the characteristics of CL, is taken into consideration in the model: the same emitters are excited every time. Combining this with the second remark, the situation we assume in this model is as follows: we consider the emitters located within the excited region (N_{tot} emitters in total), each incident electron in the beam excites a part of them (say, N_{ex} emitters, where N_{ex} varies for each electron), and its average number \bar{N}_{ex} is N .

C. Semiclassical master equation for the number of excited emitters

As seen in the next section, the statistics of the number of excited emitters $\hat{n} = \sum_{j=1}^N \hat{\sigma}_j^+ \hat{\sigma}_j^-$ is useful to investigate the second-order correlation function $g^{(2)}$. The statistics is governed by the probability $P(n, t)$ that the number of excited emitters is n at time t . As derived in Appendix A, QME

(2) is exactly reduced to the following semiclassical master equation for $P(n, t)$:

$$\frac{d}{dt}P(0, t) = \frac{1}{\tau_{\text{rad}}}P(1, t) - \gamma P(0, t), \quad (7)$$

$$\frac{d}{dt}P(n, t) = \frac{n+1}{\tau_{\text{rad}}}P(n+1, t) - \frac{n}{\tau_{\text{rad}}}P(n, t), \quad (8)$$

$$\frac{d}{dt}P(N, t) = \gamma P(0, t) - \frac{N}{\tau_{\text{rad}}}P(N, t), \quad (9)$$

where Eq. (8) is for $1 \leq n \leq N-1$.

III. SECOND-ORDER CORRELATION FUNCTION

A. Steady state

In CL with a continuous beam, the system is in the steady state $\hat{\rho}_{\text{ss}}$, which is determined by $\mathcal{L}\hat{\rho}_{\text{ss}} = 0$. In the following, we write the steady-state average $\text{Tr}[\hat{\rho}_{\text{ss}} \cdots]$ as $\langle \cdots \rangle_{\text{ss}}$.

In the steady state, $P(n, t)$ also becomes the stationary distribution $P_{\text{ss}}(n)$. We obtain the equations that determine $P_{\text{ss}}(n)$ by setting the left-hand sides of Eqs. (7)–(9) to zero. We can exactly solve these equations with the normalization condition $\sum_{n=0}^N P_{\text{ss}}(n) = 1$ to obtain

$$P_{\text{ss}}(0) = \frac{1}{1 + z_N \tau_{\text{rad}} \gamma}, \quad (10)$$

$$P_{\text{ss}}(n) = \frac{\tau_{\text{rad}} \gamma}{n(1 + z_N \tau_{\text{rad}} \gamma)} \quad (1 \leq n \leq N), \quad (11)$$

where $z_N = \sum_{m=1}^N (1/m)$. From $P_{\text{ss}}(n)$, we can calculate the steady-state moments of \hat{n} . The first two are

$$\langle \hat{n} \rangle_{\text{ss}} = \sum_{n=0}^N n P_{\text{ss}}(n) = \frac{N \tau_{\text{rad}} \gamma}{1 + z_N \tau_{\text{rad}} \gamma}, \quad (12)$$

$$\langle \hat{n}^2 \rangle_{\text{ss}} = \sum_{n=0}^N n^2 P_{\text{ss}}(n) = \frac{N(N+1) \tau_{\text{rad}} \gamma}{2(1 + z_N \tau_{\text{rad}} \gamma)}. \quad (13)$$

We use these moments in calculating $g^{(2)}$.

We now investigate the normalized second-order correlation function $g^{(2)}(\tau)$ in the steady state. This is defined by $g^{(2)}(\tau) = \langle \mathcal{T} : \hat{f}_{\text{rad}} \hat{f}_{\text{rad}}(\tau) : \rangle_{\text{ss}} / \langle \hat{f}_{\text{rad}} \rangle_{\text{ss}}^2$, where \hat{f}_{rad} is the intensity operator of the radiation field, \mathcal{T} is the time ordering, and $::$ is the normal ordering [52]. Thanks to the normalization factor $\langle \hat{f}_{\text{rad}} \rangle_{\text{ss}}^2$, collection and detection efficiencies of light and the linear loss of an optical system do not affect the value of $g^{(2)}$, and thus defined $g^{(2)}(\tau)$ describes the second-order correlation that is obtained in the HBT experiments. To proceed further, we again use the assumption that the dipole moments of the emitters are randomly oriented. In this case, the intensity operator reads $\hat{f}_{\text{rad}} \propto \sum_{j=1}^N \hat{\sigma}_j^+ \hat{\sigma}_j^- = \hat{n}$. Therefore, $g^{(2)}(\tau)$ is given by

$$g^{(2)}(\tau) = \frac{\sum_{j_1, j_2} \langle \hat{\sigma}_{j_1}^+ \hat{\sigma}_{j_1}^+(\tau) \hat{\sigma}_{j_2}^-(\tau) \hat{\sigma}_{j_2}^- \rangle_{\text{ss}}}{\langle \hat{n} \rangle_{\text{ss}}^2}. \quad (14)$$

Since the steady-state correlation function is symmetric at $\tau = 0$, we analyze $g^{(2)}(\tau)$ for $\tau \geq 0$ in the following.

B. Zero-time delay correlation: Superbunching

First, we derive the exact formula for the zero-time delay correlation function $g^{(2)}(0)$. At $\tau = 0$, we can rewrite the

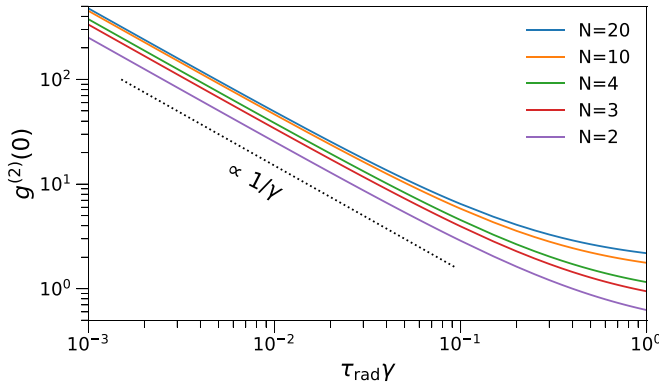


FIG. 1. Zero-time delay correlation $g^{(2)}(0)$ [Eq. (16)] as a function of the excitation rate γ (normalized by the radiative lifetime τ_{rad}). The curves from bottom to top correspond to $N = 2, 3, 4, 10$, and 20 , respectively. The dotted line is a visual guide proportional to $1/\gamma$.

numerator of Eq. (14) as follows:

$$\sum_{j_1, j_2} \langle \hat{\sigma}_{j_1}^+ \hat{\sigma}_{j_2}^+ \hat{\sigma}_{j_2}^- \hat{\sigma}_{j_1}^- \rangle_{\text{ss}} = \sum_{j_1 \neq j_2} \langle \hat{\sigma}_{j_1}^+ \hat{\sigma}_{j_2}^+ \hat{\sigma}_{j_2}^- \hat{\sigma}_{j_1}^- \rangle_{\text{ss}} = \langle \hat{n}^2 \rangle_{\text{ss}} - \langle \hat{n} \rangle_{\text{ss}}, \quad (15)$$

noting that $\hat{\sigma}_{j_1}^{\pm}$ and $\hat{\sigma}_{j_2}^{\pm}$ are commutative only if $j_1 \neq j_2$. Applying Eqs. (12) and (13), we obtain the exact formula for $g^{(2)}(0)$:

$$g^{(2)}(0) = \frac{1}{2} \left(z_N + \frac{1}{\tau_{\text{rad}} \gamma} \right) \left(1 - \frac{1}{N} \right). \quad (16)$$

From this formula, we can easily show that the superbunching, $g^{(2)}(0) \gg 2$, is observed for $\tau_{\text{rad}} \gamma \ll 1$. Figure 1, which shows the γ dependence of $g^{(2)}(0)$ for $N \geq 2$, illustrates this feature clearly. On the other hand, when $N = 1$, we have $g^{(2)}(0) = 0$ indicating the antibunching. This result implies that excitation of multiple emitters ($N \geq 2$) by a single incoming electron are necessary for the superbunching.

We note that, in formula (16), $g^{(2)}(0)$ is proportional to $1/\gamma$ for $\tau_{\text{rad}} \gamma \ll 1$. In Fig. 1, this seems valid for $\tau_{\text{rad}} \gamma \lesssim 0.1$. Since the excitation rate γ is proportional to the electron current I and the excitation efficiency p_{ex} as explained above Eq. (5), this means that $g^{(2)}(0)$ is proportional to $1/I$ and $1/p_{\text{ex}}$ for $\tau_{\text{rad}} \gamma \ll 1$. Therefore, formula (16) reproduces the properties of $g^{(2)}(0)$ discussed in Refs. [30,31,35].

We also make a remark on the limitation of this formula. In experiments, $g^{(2)}(0)$ approaches 1 for large electron current [15,31,35,36]. In comparison, the theoretical formula (16) of $g^{(2)}(0)$ approaches $(1/2)z_N(1 - 1/N) \neq 1$ for large γ (thus for large electron current). This discrepancy is attributed to the limited validity range of $\tau_{\text{rad}} \gamma$ in the present model. As explained below Eq. (6), the model is applicable for sufficiently small $\tau_{\text{rad}} \gamma$ because the excitation term in Eq. (5) does not work for large $\tau_{\text{rad}} \gamma$. However, note that, in Sec. IV and the Supplemental Material [47] (Refs. [15,44,48–51] are included therein), we generalize the present model to apply it even for large $\tau_{\text{rad}} \gamma$ and show that this discrepancy is resolved in the generalized model.

C. Finite-time delay correlation

Next, we derive an approximate form of $g^{(2)}(\tau)$ under the assumption of $N\tau_{\text{rad}}\gamma \ll 1$. To this end, we apply the quantum regression theorem (QRT) [44,45] to the correlation function $\sum_{j_1=1}^N \sum_{j_2=1}^N \langle \hat{\sigma}_{j_1}^+ \hat{\sigma}_{j_2}^+(\tau) \hat{\sigma}_{j_2}^-(\tau) \hat{\sigma}_{j_1}^- \rangle_{\text{ss}} = \sum_{j=1}^N \langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$ in the numerator of Eq. (14). We can derive that this correlation function shows a multiple exponential decay and approaches $\langle \hat{n} \rangle_{\text{ss}}^2$ [thus $g^{(2)}(\tau) \rightarrow 1$] for $\tau \rightarrow \infty$. Furthermore, if we assume $N\tau_{\text{rad}}\gamma \ll 1$, we can show that the lowest decay rate is approximately equal to $\lambda_1 \simeq (1/\tau_{\text{rad}})(1 + N\tau_{\text{rad}}\gamma)$ and the second lowest is $\lambda_2 \simeq (2/\tau_{\text{rad}})[1 - N(N-1)\tau_{\text{rad}}\gamma/4]$ (see Appendix B for derivation). Therefore, the decaying behavior of $\sum_{j=1}^N \langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$ is dominated by $e^{-\lambda_1 \tau}$.

Combining this decaying behavior with the asymptotic value $\lim_{\tau \rightarrow \infty} g^{(2)}(\tau) = 1$, we arrive at an approximate expression for $g^{(2)}(\tau)$:

$$g^{(2)}(\tau) \simeq [g^{(2)}(0) - 1]e^{-\tau/\tau_{\text{rad}}^{\text{eff}}} + 1 = C \left(1 - \frac{1}{N} \right) e^{-\tau/\tau_{\text{rad}}^{\text{eff}}} + \left(1 - \frac{1}{N} e^{-\tau/\tau_{\text{rad}}^{\text{eff}}} \right). \quad (17)$$

In the second line, we used formula (16) for $g^{(2)}(0)$. Here, the effective lifetime $\tau_{\text{rad}}^{\text{eff}}$ is given by

$$\frac{1}{\tau_{\text{rad}}^{\text{eff}}} = \frac{1}{\tau_{\text{rad}}} + N\gamma, \quad (18)$$

and the prefactor C is

$$C = \frac{1}{2} \left(z_N + \frac{1}{\tau_{\text{rad}} \gamma} \right) - 1. \quad (19)$$

We note that our approximate expression (17) has a form similar to that in Ref. [15], which is valid for the small- N region. In their expression, the decay time is the bare lifetime τ_{rad} instead of $\tau_{\text{rad}}^{\text{eff}}$ and the prefactor C' corresponding to C in ours reads $C' = I_0/(I \times P_{\text{el}}^1)$. In the notation of this paper, $I_0 = e/\tau_{\text{rad}}$ and the probability of creating electron-hole pairs by an incoming electron P_{el}^1 should be proportional to p_{ex} . Since $\gamma = (I/e)p_{\text{ex}}$ as explained above Eq. (5), we have $C' \propto 1/(\tau_{\text{rad}}\gamma)$. In the small- N region ($N\tau_{\text{rad}}\gamma \ll 1$), the effective lifetime becomes $\tau_{\text{rad}}^{\text{eff}} \simeq \tau_{\text{rad}}$ and our prefactor C of Eq. (19) yields $C \simeq 1/(2\tau_{\text{rad}}\gamma) \propto 1/(\tau_{\text{rad}}\gamma)$. Therefore, our result from the master equation perspective validates the formula in Ref. [15].

D. Numerical demonstration

To demonstrate the applicability of the approximate expression (17), we compare it with results of numerical simulation. In the simulation, we numerically solve the eigenvalue problem of \mathcal{L} to obtain the steady state $\hat{\rho}_{\text{ss}}$ as the right eigenvector corresponding to its zero eigenvalue. Then, we use the QRT to compute $g^{(2)}(\tau)$. We plot the results for $N = 2$ and $N = 10$ in Fig. 2. We also plot Eq. (17) (solid lines) and its variant where $\tau_{\text{rad}}^{\text{eff}}$ is replaced with τ_{rad} (dotted lines):

$$g^{(2)}(\tau) \simeq C \left(1 - \frac{1}{N} \right) e^{-\tau/\tau_{\text{rad}}} + \left(1 - \frac{1}{N} e^{-\tau/\tau_{\text{rad}}} \right). \quad (20)$$

In the case of $N = 2$, Fig. 2(a) shows that the approximate expression (17) describes the numerical data well. In compar-

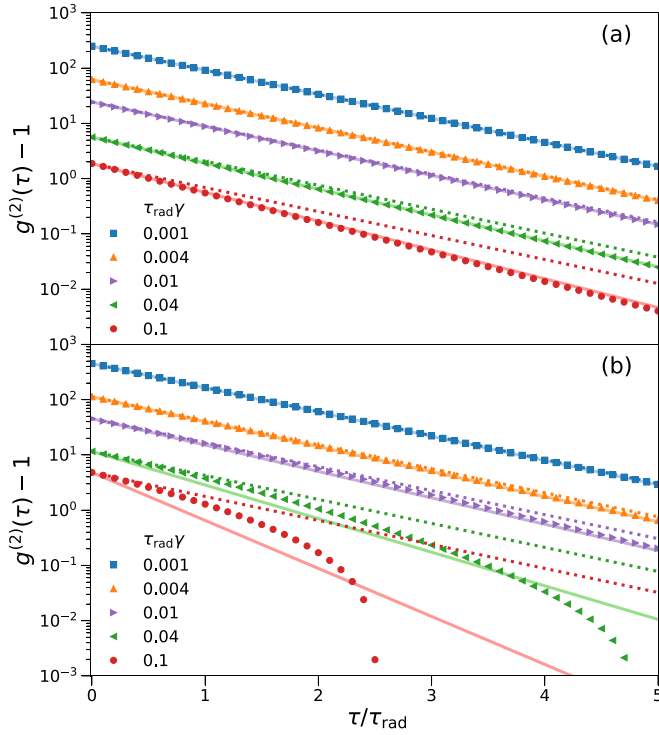


FIG. 2. Normalized second-order correlation function $g^{(2)}(\tau)$ for (a) $N = 2$ and (b) $N = 10$. Semilogarithmic plots of $g^{(2)}(\tau) - 1$ are shown for excitation rates ranging from $\tau_{\text{rad}}\gamma = 0.001$ to 0.1 . The symbols, solid lines, and dotted lines represent the numerical results, approximate formula (17), and decay curves proportional to $e^{-\tau/\tau_{\text{rad}}}$ [Eq. (20)], respectively. For $\tau_{\text{rad}}\gamma = 0.1$ and $N = 10$ [solid circles in (b)], data points for $\tau/\tau_{\text{rad}} \gtrsim 2.5$ are not shown because $g^{(2)}(\tau) - 1$ is extremely small [$g^{(2)}(\tau) \approx 1$] in this region.

ison, although Eq. (20) deviates from the numerical results for $\tau_{\text{rad}}\gamma \geq 0.04$ ($N\tau_{\text{rad}}\gamma \geq 0.08$), it also describes the numerical data well for smaller $\tau_{\text{rad}}\gamma$ because $\tau_{\text{rad}}^{\text{eff}} \simeq \tau_{\text{rad}}$ in this regime.

In the case of $N = 10$, Fig. 2(b) shows that Eq. (17) well approximates the numerical data for $\tau_{\text{rad}}\gamma \leq 0.004$ ($N\tau_{\text{rad}}\gamma \leq 0.04$). As $\tau_{\text{rad}}\gamma$ becomes larger, we observe clearer deviations between the numerical results and Eq. (17) as well as Eq. (20).

From the results of $N = 2$ and $N = 10$, we conclude that the approximation by Eq. (17) is valid for small $N\tau_{\text{rad}}\gamma$. We also note that the cruder approximation by Eq. (20) is valid if $N\tau_{\text{rad}}\gamma$ is sufficiently small, and the decay rate of $g^{(2)}(\tau)$ can be used for estimation of the lifetime τ_{rad} of the emitters. In experiments, one should carefully choose electron-beam current I in order to obtain the lifetime τ_{rad} from the HBT measurement.

IV. GENERALIZATION OF MODEL

In the model in Sec. II, the same N emitters are excited by each incident electron. In experiments, however, the emitters excited are different for each electron. Here, we generalize the model to incorporate this effect.

Let N_{tot} be the number of emitters that the electron beam can excite, so that these emitters are located within the excited region. We assume that N_{tot} is a fixed number. A part of these emitters are excited by an incoming electron. Similarly to

Eq. (5), if j_1 th, j_2 th, \dots , $j_{N_{\text{ex}}}$ th emitters are simultaneously excited by an electron, the excitation term in the QME should read

$$\Gamma_{N_{\text{ex}}} (\hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^+ \hat{\rho} \hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^- - \frac{1}{2} \{ \hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^-, \hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^+, \hat{\rho} \}),$$

where $\hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^{\pm} = \bigotimes_{i=1}^{N_{\text{ex}}} \hat{\sigma}_{j_i}^{\pm}$. $\Gamma_{N_{\text{ex}}}$ is the rate of this excitation; for simplicity, we assume N_{ex} emitter excitations have the same rate (i.e., $\Gamma_{N_{\text{ex}}}$ depends only on the number of emitters, N_{ex} , but not on the indices $j_1, j_2, \dots, j_{N_{\text{ex}}}$). If the emitters are independently excited by an incident electron, the number N_{ex} of emitters excited by the electron follows the binomial distribution $\eta^{N_{\text{ex}}} (1 - \eta)^{N_{\text{tot}} - N_{\text{ex}}}$, where η is the probability that a single emitter is excited by an electron ($0 < \eta < 1$). Therefore, it is reasonable to assume

$$\Gamma_{N_{\text{ex}}} = \gamma_2 \eta^{N_{\text{ex}}} (1 - \eta)^{N_{\text{tot}} - N_{\text{ex}}},$$

where γ_2 is a positive constant that is proportional to the rate of the incoming electron, I/e .

Summing up all the possible excitations, we obtain a generalized excitation term in the QME,

$$\begin{aligned} \mathcal{D}_{\text{ex}}^{(2)} \hat{\rho} = & \gamma_2 \sum_{N_{\text{ex}}=1}^{N_{\text{tot}}} \eta^{N_{\text{ex}}} (1 - \eta)^{N_{\text{tot}} - N_{\text{ex}}} \\ & \times \sum_{j_1 < j_2 < \dots < j_{N_{\text{ex}}}} \left(\hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^+ \hat{\rho} \hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^- \right. \\ & \left. - \frac{1}{2} \{ \hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^-, \hat{\Pi}_{j_1, j_2, \dots, j_{N_{\text{ex}}}}^+, \hat{\rho} \} \right), \end{aligned} \quad (21)$$

where each index in the second sum on the right-hand side runs from 1 to N_{tot} , satisfying the constraint of $j_1 < j_2 < \dots < j_{N_{\text{ex}}}$. We thus obtain a generalized model by replacing $\mathcal{D}_{\text{ex}} \hat{\rho}$ in Eq. (3) with $\mathcal{D}_{\text{ex}}^{(2)} \hat{\rho}$ and N with N_{tot} in Eqs. (1) and (4).

Hereafter, we refer to the model in Sec. II as model 1 and the generalized model in this section as model 2.

Relation between models

We can interpret model 1 as a simplified description of model 2, where N in model 1 corresponds to an average number \bar{N}_{ex} of excitations by an incoming electron in model 2. Since $\bar{N}_{\text{ex}} = \sum_{N_{\text{ex}}=0}^{N_{\text{tot}}} \eta^{N_{\text{ex}}} (1 - \eta)^{N_{\text{tot}} - N_{\text{ex}}} \binom{N_{\text{tot}}}{N_{\text{ex}}} N_{\text{ex}} = \eta N_{\text{tot}}$ in the binomial distribution, N in model 1 is connected to model 2 by

$$N = \eta N_{\text{tot}}. \quad (22)$$

Furthermore, for model 1 to be an effective description of model 2, the average number of emitters excited by the electron beam per unit time must be equal: $\gamma N = \gamma_2 \bar{N}_{\text{ex}}$. Combining this equation with Eq. (22), we have

$$\gamma = \gamma_2. \quad (23)$$

To investigate the condition that model 1 well approximates model 2, we note the relative fluctuation of the

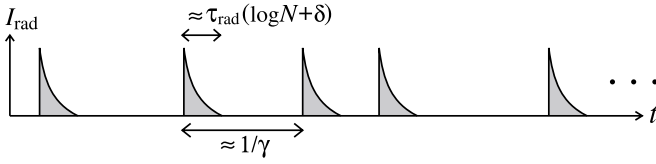


FIG. 3. A sequence of random pulses of radiation intensity I_{rad} . The intensity profile of each pulse is $I_{\text{rad}} \approx N e^{-\tau/\tau_{\text{rad}}}$ on average, so the mean duration of the pulse is roughly equal to $\tau_{\text{rad}}(\log N + \delta)$.

excitation number in the binomial distribution:

$$\frac{\sqrt{N_{\text{ex}}^2 - N_{\text{ex}}}}{N_{\text{ex}}} = \sqrt{\frac{1 - \eta}{\eta N_{\text{tot}}}}. \quad (24)$$

This implies that the relative fluctuation of the number of emitters excited by an incident electron becomes smaller as N_{tot} or η increases. Therefore, if the total number N_{tot} of emitters in the excited region is sufficiently large or if the single-emitter-excitation probability η is near 1, we can assume that the excitation number is approximately the same single value, $\bar{N}_{\text{ex}} (= N)$, for each electron. This is the condition for model 1 to approximate model 2. In the Supplemental Material [47] (Refs. [15,44,48–51] are included therein), we numerically demonstrate this approximate relation between models 1 and 2.

We also note the difference between models for large excitation rate. Unlike the excitation term (5) of model 1, Eq. (21) of model 2 can excite emitters even for large $\tau_{\text{rad}}\gamma$. This suggests that model 2 is applicable even for $\tau_{\text{rad}}\gamma > 1$. In the Supplemental Material [47], we numerically demonstrate that this is the case. There, we find that $g^{(2)}(0)$ in model 2 approaches $1 - 1/N_{\text{tot}}$ for large $\tau_{\text{rad}}\gamma$. Therefore, if N_{tot} is sufficiently large, $g^{(2)}(0)$ is nearly equal to 1, which is consistent with the experimental results [15,31,35,36].

V. STATE OF RADIATION FIELD

In the previous section, our analysis on $g^{(2)}$ of the radiation field is based on the steady state $\hat{\rho}_{\text{ss}}$ of the emitters. Understanding the state of the radiation field itself is also an interesting problem. In this section, to qualitatively understand the radiation field state in CL, we give a heuristic argument on this problem under the assumption of $z_N \tau_{\text{rad}}\gamma \ll 1$ [note that $z_N = \sum_{m=1}^N (1/m) \approx \log N + \delta$ with an irrelevant constant δ ($0 < \delta < 1$)].

We first revisit the process of the radiation in CL. Even though an electron beam irradiates a sample continuously, each electron in the beam exists discretely. An incoming electron excites multiple (say, N) emitters and the emitters decay with radiating photons. The radiation generated in this process is considered to have a time profile of the intensity $I_{\text{rad}}(t)$ that is composed of a sequence of random pulses, as schematically depicted in Fig. 3. In each pulse, the excited emitters radiate photons within the duration of the emission process. The duration is random due to the spontaneous emission process of the emitters, and the mean duration is roughly equal to $\tau_{\text{rad}}(\log N + \delta)$ because $I_{\text{rad}} \approx N e^{-\tau/\tau_{\text{rad}}} > \epsilon$ (with some small positive constant ϵ) should be satisfied for τ within the duration. The instance at which a single pulse starts is also random, reflecting the randomness of the incoming electrons, and the

mean time between successive pulses is equal to $1/\gamma$, where γ is the excitation rate.

From the above argument, we can classify total time of photodetection into pulse-existing regions and zero-intensity regions, and we estimate the ratio q of the former regions to the total as $q \approx \tau_{\text{rad}}\gamma(\log N + \delta)$ ($\ll 1$). In the former regions, the radiation field is in a certain photonic state $\hat{\rho}_N^{\text{rad}}$ whose average photon number is around N . In the latter, it is in the vacuum state $\hat{\rho}_{\text{vac}}^{\text{rad}} = |\text{vac}\rangle\langle\text{vac}|$. Therefore, we can consider the steady state of the radiation field in CL as the average of $\hat{\rho}_N^{\text{rad}}$ and $\hat{\rho}_{\text{vac}}^{\text{rad}}$:

$$\hat{\rho}_{\text{avg}}^{\text{rad}} = q \hat{\rho}_N^{\text{rad}} + (1 - q) \hat{\rho}_{\text{vac}}^{\text{rad}}. \quad (25)$$

In this case, the zero-time delay correlation function of the radiation field $\hat{\rho}_{\text{avg}}^{\text{rad}}$ yields a $(1/q)$ multiple of that of $\hat{\rho}_N^{\text{rad}}$. In fact, for single-mode radiation, we have

$$g^{(2)}(0) = \frac{\text{Tr}[\hat{\rho}_{\text{avg}}^{\text{rad}} \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a}]}{\text{Tr}[\hat{\rho}_{\text{avg}}^{\text{rad}} \hat{a}^\dagger \hat{a}]^2} = \frac{1}{q} \frac{\text{Tr}[\hat{\rho}_N^{\text{rad}} \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a}]}{\text{Tr}[\hat{\rho}_N^{\text{rad}} \hat{a}^\dagger \hat{a}]^2}, \quad (26)$$

where \hat{a}^\dagger and \hat{a} are the creation and destruction operators of the mode, respectively. This result with $q \approx \tau_{\text{rad}}\gamma(\log N + \delta)$ reproduces the approximate proportionality of formula (16) to $1/\tau_{\text{rad}}\gamma$ and gives rise to the superbunching, $g^{(2)}(0) \gg 2$.

To be more concrete, we assume that the emitters' population is directly transferred to the single-mode photonic population as

$$\hat{\rho}_{\text{avg}}^{\text{rad}} = \sum_{n=0}^N P_{\text{ss}}(n) |n\rangle\langle n|, \quad (27)$$

where $|n\rangle$ is the n -photon state of the single mode (in particular, $|0\rangle = |\text{vac}\rangle$) and $P_{\text{ss}}(n)$ is the steady-state probability of the number of excited emitters [Eqs. (10) and (11)]. This state with Eq. (26) exactly reproduces formula (16) for $g^{(2)}(0)$. Moreover, we can write this state in the form of Eq. (25), where $q = 1 - P_{\text{ss}}(0) = z_N \tau_{\text{rad}}\gamma / (1 + z_N \tau_{\text{rad}}\gamma)$ and

$$\hat{\rho}_N^{\text{rad}} = \sum_{n=1}^N \frac{P_{\text{ss}}(n)}{1 - P_{\text{ss}}(0)} |n\rangle\langle n| = \sum_{n=1}^N \frac{1}{z_N n} |n\rangle\langle n|. \quad (28)$$

For $z_N \tau_{\text{rad}}\gamma \ll 1$, $q \approx z_N \tau_{\text{rad}}\gamma \approx \tau_{\text{rad}}\gamma(\log N + \delta)$ holds as expected.

To interpret the superbunching effect in different systems, similar arguments are given in Refs. [53,54] (see also Ref. [55]): a classical (incoherent) mixture of high- and low-intensity states with a large weight on the lower as in Eq. (25) leads to an enhancement of $g^{(2)}(0)$. We note that a quantum superposition state $\sqrt{q}|\text{rad}_N\rangle + \sqrt{1-q}|\text{vac}\rangle$ with some pure photonic state $|\text{rad}_N\rangle$ such as $|N\rangle$ also leads to a similar enhancement [54] and, only from $g^{(2)}(0)$, it is impossible to distinguish whether the radiation state is a classical or quantum mixture. In our model of CL, however, it should be a classical one because the steady state of the emitters is diagonal in the standard basis.

On the basis of this argument, we also discuss the difference between CL and PL photon statistics: the superbunching is observed in the CL whereas $g_{\text{PL}}^{(2)}(0) \simeq 1$ for the PL of the same sample [15]. While an electron instantly excites multiple emitters in CL, a continuous-wave laser for PL has a typical

excitation duration time, which reflects the coherence time. Thus, the interval of successive excitations in PL is much shorter than the radiation lifetime, such that the radiation in PL is continuous without pulsing (unlike Fig. 3 of CL). Therefore, the state of the PL radiation field can be considered simply as $\hat{\rho}_N^{\text{rad}}$ and not mixed with the vacuum state. Although the form of $\hat{\rho}_N^{\text{rad}}$ given by Eq. (28) cannot correctly express the state of PL because the present model is not valid under continuous conditions, the form of $\hat{\rho}_{\text{avg}}^{\text{rad}}$ given by Eq. (27) approaches Eq. (28) for $\tau_{\text{rad}}\gamma \gg 1$. This supports the above statement of $g_{\text{PL}}^{(2)}(0) \simeq 1$. If there is almost no difference between CL and PL in the material response related to the upper state excitation, once the state of PL is known, we can estimate the state of CL, providing the calculation of optical systems such as an interferometer with CL. This opens up application of CL itself (e.g., superbunched light source) and nanoscale analysis using the CL photon state property.

VI. DISCUSSION AND SUMMARY

In this study, we have constructed a QME model that captures essential aspects of CL: simultaneous excitation and individual decay of emitters. We have derived the exact formula for the zero-time delay correlation $g^{(2)}(0)$, which successfully describes the superbunching. We have also derived an approximate form for the finite-time delay correlation $g^{(2)}(\tau)$, which shows that the radiative lifetime τ_{rad} can be extracted from its τ dependence for small $N\tau_{\text{rad}}\gamma$.

In the present model, we have assumed that the properties (transition energy $\hbar\omega_e$, radiative lifetime τ_{rad} , and excitation strength) of all the emitters are the same and that there is no interaction among them. Also, they have randomly oriented dipole moments. It is expected that introducing inhomogeneity of emitters does not drastically alter the main conclusion of the model (for small $\tau_{\text{rad}}\gamma$) on the superbunching in CL. One interesting direction for future research is introducing interaction. If there is interaction, the QME is not simply reduced to the semiclassical master equation and some genuine quantum effects may emerge. The present model provides a simple basis for this direction.

We also note that, although we have not presented the spectrum of CL in this work, we can also investigate it in the model introducing the spectral resolution [56,57]. In calculating it, we can apply the QRT [44,45] to steady-state correlation functions.

In the model, the simultaneous excitation of multiple emitters by an incoming electron is phenomenologically introduced in Eq. (5). In the Supplemental Material [47] (Refs. [15,44,48–51] are included therein), we give a justification of this excitation term [Eq. (5)], where we derive another QME from a microscopic model with a stochastic interaction term emulating the process that an electron, randomly incident on the sample, randomly excites the emitters while traveling through the sample. We numerically demonstrate that our model [Eqs. (2)–(6)] can approximately describe the features of the microscopically derived QME in the Supplemental Material [47]. It is an important future work to derive this excitation effect from a first-principles analysis on the complicated elementary processes in CL starting from nonstochastic interactions.

Another implication of this phenomenological incorporation is that the superbunching is not specific to CL experiments; we can observe superbunching if there could be simultaneous excitation and individual radiative decay of multiple emitters and if their timescales are largely different. Indeed, giant photon bunching can be observed in other systems that have cooperative emission [20,58,59]. In these systems, we can interpret that excitations via dark states play the role of the simultaneous excitation of multiple emitters. In a similar manner, we can explain another example of giant bunching in a system composed of a quantum dot and a metal nanoparticle [21]: in this system, since the transition rate is suppressed by the Fano destructive interference, multiple photons can be efficiently excited through the dark state. This plays the role of (nearly) simultaneous multiple excitation.

We have also discussed a possible state of radiation field in the CL. Through a heuristic argument, we have proposed the state in Eq. (25) and confirmed that it is consistent with the QME analysis and well describes superbunching. This result implies that we may observe superbunching with mechanisms other than the simultaneous excitation of multiple emitters. Indeed, in Ref. [55], an enhancement of $g^{(2)}(0)$ is discussed in a train of pulses with a regular interval. Our argument, schematically depicted in Fig. 3, shows that the same enhancement is observed in random pulses and that the state of the radiation field in CL should be similar to that of the randomly modulated optical beam. Also, Ref. [53] discusses superbunching with states similar to Eq. (25) and shows that a bimodal microcavity laser with an emitter achieves such a state as its steady state. Indeed, superbunching is reported in bimodal microlaser systems [23–27].

In other words, a potent way to observe superbunching is generating a mixture of photonic states as in Eq. (25), and there are several systems and methods which can generate these types of states. The present study by the master equation reveals that the simultaneous excitation of multiple emitters in CL is one of them and gives quantum insight into CL photon statistics. Since the time-correlation measurement of CL has given new functionalities to nanoscale optical imaging, the obtained results imply a potential of CL to reveal and even utilize the quantum nature of light-matter interaction on the nanoscale.

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APPENDIX A: MASTER EQUATION FOR $P(n, t)$

In this Appendix, we derive the master equation for the number of excited emitters [Eqs. (7)–(9)]. For this purpose, we introduce some notation. We denote each of the standard basis states as $|s\rangle = \otimes_{j=1}^N |s_j\rangle_j$ with $s = \sum_{j=1}^N 2^{j-1} s_j$. Then, the standard basis is represented as $\{|s\rangle \mid s = 0, 1, 2, \dots, 2^N -$

1}. We write the diagonal elements of the system state $\hat{\rho}(t)$ as $\rho(s, t) = \langle s | \hat{\rho}(t) | s \rangle$. In addition, we define $|s^{j+}\rangle = \hat{\sigma}_j^+ |s\rangle$, which is also one of the standard basis states if $s_j = 0$.

From QME (2), we can show $\langle s | \{d\hat{\rho}(t)/dt\} | s \rangle = \langle s | \mathcal{L}\hat{\rho}(t) | s \rangle$ yields

$$\frac{\partial}{\partial t} \rho(0, t) = \frac{1}{\tau_{\text{rad}}} \sum_{j=1}^N \rho(0^{j+}, t) - \gamma \rho(0, t), \quad (\text{A1})$$

$$\frac{\partial}{\partial t} \rho(s, t) = \frac{1}{\tau_{\text{rad}}} \sum_{j=1}^N [\rho(s^{j+}, t) \delta_{s_j, 0} - \rho(s, t) \delta_{s_j, 1}], \quad (\text{A2})$$

$$\frac{\partial}{\partial t} \rho(2^N - 1, t) = -\frac{N}{\tau_{\text{rad}}} \rho(2^N - 1, t) + \gamma \rho(0, t), \quad (\text{A3})$$

where Eq. (A2) is for $1 \leq s \leq 2^N - 2$. We note that Eqs. (A1)–(A3) form a closed set of equations for the diagonal elements (Pauli master equation [44,60]).

To transform Eqs. (A1)–(A3) to the master equation for $P(n, t)$, we note the connection between $P(n, t)$ and $\rho(s, t)$:

$$P(n, t) = \sum_{s=0}^{2^N-1} \rho(s, t) \delta_{n_s, n}, \quad (\text{A4})$$

where

$$n_s = \sum_{j=1}^N s_j = \sum_{j=1}^N \delta_{s_j, 1} \quad (\text{A5})$$

is the number of excited emitters in the state $|s\rangle$. Differentiating Eq. (A4) with respect to t and using Eqs. (A1)–(A3), we obtain the master equation for $P(n, t)$ [Eqs. (7)–(9)].

APPENDIX B: A PERTURBATIVE ANALYSIS OF DECAY RATES

In this Appendix, we evaluate the decay rates of $\sum_{j=1}^N \langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$. To this end, it is sufficient to investigate each term $\langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$ in the sum. In the estimation, we use the QRT and a perturbative analysis under the assumption of $\tau_{\text{rad}} \gamma \ll 1$.

We first note that $\hat{n} = \sum_{n=0}^N n \hat{P}_n$, where \hat{P}_n is the projection operator onto the subspace of states with n excited emitters. This leads to $\langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}} = \sum_n n \langle \hat{\sigma}_j^+ \hat{P}_n(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$, so it is reasonable to investigate the decaying behavior of $\langle \hat{\sigma}_j^+ \hat{P}_n(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$.

According to the QRT, $\langle \hat{\sigma}_j^+ \hat{P}_n(\tau) \hat{\sigma}_j^- \rangle$ obeys the differential equation whose form is the same as that of $\langle \hat{P}_n(\tau) \rangle = P(n, \tau)$. The latter is the master equation for $P(n, t)$ [Eqs. (7)–(9)], which is expressed as $d\mathbf{P}(t)/dt = -\Gamma \mathbf{P}(t)$ with $\mathbf{P}(t) \equiv (P(0, t), P(1, t), \dots, P(N, t))^T$ (Γ stands for transpose) and an $(N+1) \times (N+1)$ matrix Γ :

$$\Gamma = \begin{pmatrix} \gamma & -1/\tau_{\text{rad}} & & & \\ & 1/\tau_{\text{rad}} & -2/\tau_{\text{rad}} & & \\ & & 2/\tau_{\text{rad}} & \ddots & \\ & & & \ddots & -N/\tau_{\text{rad}} \\ -\gamma & & & & N/\tau_{\text{rad}} \end{pmatrix}. \quad (\text{B1})$$

Therefore, by using the QRT, we obtain the differential equation $d\mathbf{Q}_j(\tau)/d\tau = -\Gamma \mathbf{Q}_j(\tau)$ for $\mathbf{Q}_j(\tau) \equiv (\langle \hat{\sigma}_j^+ \hat{P}_0(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}, \langle \hat{\sigma}_j^+ \hat{P}_1(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}, \dots, \langle \hat{\sigma}_j^+ \hat{P}_N(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}})^T$. Moreover, using the eigenvalues λ_n and its corresponding left and right eigenvectors, ℓ_n and \mathbf{r}_n , of the non-Hermitian matrix Γ , we can show

$$\mathbf{Q}_j(\tau) = \sum_{n=0}^N [\ell_n \cdot \mathbf{Q}_j(0)] \mathbf{r}_n e^{-\lambda_n \tau}. \quad (\text{B2})$$

This implies that $\langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$ exhibits a multiple exponential decay with the rates λ_n ($n = 1, 2, \dots, N$).

Note that, as shown in Eqs. (10) and (11), Γ has a zero eigenvalue $\lambda_0 = 0$ and the corresponding right eigenvector is $\mathbf{r}_0 = \mathbf{P}_{\text{ss}} \equiv (P_{\text{ss}}(0), P_{\text{ss}}(1), \dots, P_{\text{ss}}(N))^T$. And it is straightforward to show that the corresponding left eigenvector is $\ell_0 = (1, 1, \dots, 1)^T$. From this result, we can show that the asymptotic value of $\mathbf{Q}_j(\tau)$ is $\lim_{\tau \rightarrow \infty} \mathbf{Q}_j(\tau) = [\ell_0 \cdot \mathbf{Q}_j(0)] \mathbf{r}_0 = \langle \hat{\sigma}_j^+ \hat{\sigma}_j^- \rangle_{\text{ss}} \mathbf{P}_{\text{ss}}$, where we used $\sum_n \hat{P}_n = 1$. Therefore, we obtain $\lim_{\tau \rightarrow \infty} \sum_j \langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}} = \sum_j \sum_n n \langle \hat{\sigma}_j^+ \hat{\sigma}_j^- \rangle_{\text{ss}} P_{\text{ss}}(n) = \langle \hat{n} \rangle_{\text{ss}}^2$, which leads to $\lim_{\tau \rightarrow \infty} g^{(2)}(\tau) = 1$.

Now we perturbatively estimate the eigenvalues by assuming $\tau_{\text{rad}} \gamma \ll 1$. For this purpose, we decompose Γ as $\Gamma = \Gamma^0 + \gamma \Gamma^1$. The unperturbed part Γ^0 is the matrix where γ in Eq. (B1) is replaced with zero. The perturbation matrix Γ^1 has only two nonzero elements: $\Gamma_{0,0}^1 = 1$ and $\Gamma_{N,0}^1 = -1$.

Since Γ^0 is an upper triangular matrix, its eigenvalues (the zeroth-order eigenvalues λ_n^0) are the diagonal elements of Γ^0 , that is, $\lambda_n^0 = n/\tau_{\text{rad}}$ ($0 \leq n \leq N$). The corresponding (zeroth-order) left and right eigenvectors, ℓ_n^0 and \mathbf{r}_n^0 , are determined by

$$(\Gamma^0)^T \ell_n^0 = \lambda_n^0 \ell_n^0, \quad (\text{B3})$$

$$\Gamma^0 \mathbf{r}_n^0 = \lambda_n^0 \mathbf{r}_n^0, \quad (\text{B4})$$

with the normalization $\ell_n^0 \cdot \mathbf{r}_n^0 = 1$. After some algebraic calculation, we obtain

$$\ell_n^0 = \left(\underbrace{0, \dots, 0}_n, 1, \binom{n+1}{1}, \binom{n+2}{2}, \dots, \binom{N}{N-n} \right)^T, \quad (\text{B5})$$

$$\mathbf{r}_n^0 = \left((-1)^n \binom{n}{n}, (-1)^{n-1} \binom{n}{n-1}, \dots, (-1) \binom{n}{1}, 1, \underbrace{0, \dots, 0}_{N-n} \right)^T, \quad (\text{B6})$$

where $\binom{m}{k} = m!/[k!(m-k)!]$ is a binomial coefficient.

The perturbative analysis for a non-Hermitian matrix is almost the same as that for Hermitian cases in quantum mechanics [61]—the first-order correction to the eigenvalue is

$$\begin{aligned} \lambda_n^1 &= \gamma \ell_n^0 \cdot \Gamma^1 \mathbf{r}_n^0 \\ &= \begin{cases} 0 & (n=0) \\ \gamma (-1)^{n+1} \binom{N}{N-n} & (1 \leq n \leq N). \end{cases} \end{aligned} \quad (\text{B7})$$

Therefore, in the first order of $\tau_{\text{rad}} \gamma$, we obtain an approximate form of the eigenvalues of Γ (except for the zero eigenvalue

$\lambda_0 = 0$):

$$\lambda_n \simeq \frac{n}{\tau_{\text{rad}}} \left[1 + \frac{(-1)^{n+1} \tau_{\text{rad}} \gamma}{n} \binom{N}{N-n} \right] \quad (1 \leq n \leq N). \quad (\text{B8})$$

We thus estimate the decay rates $\{\lambda_n\}_{n=1}^N$ of $\langle \hat{\sigma}_j^+ \hat{n}(\tau) \hat{\sigma}_j^- \rangle_{\text{ss}}$. In particular, when $N\tau_{\text{rad}}\gamma \ll 1$, the lowest rate is $\lambda_1 \simeq (1/\tau_{\text{rad}})(1 + N\tau_{\text{rad}}\gamma)$ and the second lowest is $\lambda_2 \simeq (2/\tau_{\text{rad}})[1 - N(N-1)\tau_{\text{rad}}\gamma/4]$.

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