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## Quantum photonics model for nonclassical light generation using integrated nanoplasmonic cavity-emitter systems

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The implementation of nonclassical light sources is becoming increasingly important for various quantum applications. A particularly interesting approach is to integrate such functionalities on a single chip as this could pave the way towards fully scalable quantum photonic devices. Several approaches using dielectric systems have been investigated in the past. However, it is still not understood how on-chip nanoplasmonic antennas, interacting with a single quantum emitter, affect the quantum statistics of photons reflected or transmitted in the guided mode of a waveguide. Here we investigate a quantum photonic platform consisting of an evanescently coupled nanoplasmonic cavity-emitter system and discuss the requirements for nonclassical light generation. We develop an analytical model that incorporates quenching due to the nanoplasmonic cavity to predict the quantum statistics of the transmitted and reflected guided waveguide light under weak coherent pumping. The analytical predictions match numerical simulations based on a master equation approach. It is moreover shown that for resonant excitation the degree of antibunching in transmission is maximized for an optimal cavity modal volume  $V_c$  and cavity-emitter distance  $s$ . In reflection, perfectly antibunched light can only be obtained for specific  $(V_c, s)$  combinations. Finally, our model also applies to dielectric cavities and as such can guide future efforts in the design and development of on-chip nonclassical light sources using dielectric and nanoplasmonic cavity-emitter systems.

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### I. INTRODUCTION

It has been a long-standing goal in optical science to implement nonlinear effects at the few-photon level. In this regime, individual photons interact so strongly with one another that the propagation of light pulses containing few photons varies substantially with photon number. The strong dependence of light propagation on photon number, for example, allows the sorting or nondestructive counting of photons which could be used to implement various sources of nonclassical light fields [1,2]. One route to reach the quantum regime consists of coupling light to individual quantum emitters [3]. The interaction between a single photon and a single quantum emitter is, in general, however, relatively weak and hence makes the implementation of such quantum interconnects quite challenging [4]. A particularly interesting approach is to route photons on photonic integrated circuits and interface them with on-chip quantum emitters as this allows integration of many functionalities on a single chip and hence paves the way towards truly scalable quantum devices [5,6]. Several reports investigated the interaction between quantum emitters and dielectric (Refs. [7–16]) and plasmonic (Refs. [17–24]) cavities and waveguides. Moreover, many theoretical investigations were devoted to the interaction between quantum emitters and dielectric waveguides either without (Refs. [7,8]) or with (Refs. [4,9,10]) intermediate coupling to a dielectric cavity, as well as on emitters coupled to plasmonic cavities

(Refs. [21–24]) and waveguides (Refs. [17–19]). Recently, the quantum statistics of photons scattered by a plasmonic nanocavity strongly coupled to a mesoscopic emitter ensemble was investigated under coherent pumping of the system [25]. However, the latter study did not incorporate plasmonic quenching nor the effect of coupling the cavity-emitter system to a nearby dielectric waveguide. As such, an outstanding question concerns how on-chip nanoplasmonic antennas, interacting with a single quantum emitter, affect the quantum statistics of photons reflected or transmitted in the guided mode of the waveguide. In this paper we will present a general quantum photonic model of evanescently coupled cavity-emitter systems and investigate the requirements for nonclassical light generation using integrated nanoplasmonic cavities. Our model shows excellent correspondence with numerical simulations and as such allows accurate predictions of the photon statistics generated in the guided mode of a waveguide, coupled to a nanoplasmonic cavity-emitter system.

### II. MODEL

We start from a quantum photonic platform which is closely related to the one studied in Ref. [26] (see Fig. 1). It consists of a dielectric nanophotonic waveguide supporting a one-dimensional (1D) continuum of left and right traveling modes which are characterized by the operators  $l_k$  and  $r_k$ , respectively ( $l_k$  and  $r_k$  annihilate a left- or right- traveling photon with wave number  $k = \omega_k/c$ ). These modes interact evanescently with a plasmonic cavity-emitter system, consisting of a cavity with resonance frequency  $\omega_c$  (frequency of the fundamental

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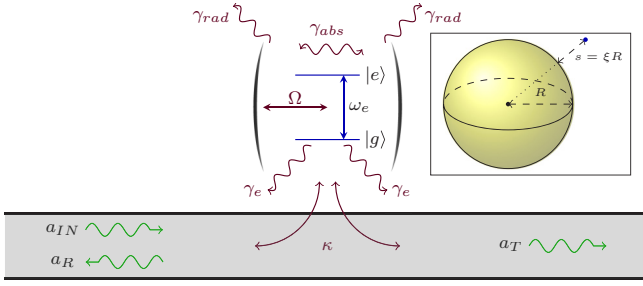


FIG. 1. Quantum photonic platform. The waveguide (gray) supports a 1D continuum of modes, carrying the excitation beam  $a_{IN}$  (at frequency  $\omega_L$ ) as well as the reflected ( $a_R$ ) and transmitted ( $a_T$ ) beam, and interacts evanescently with a cavity-emitter system. All coupling rates and frequencies are explained in the main text. The inset shows the case for a nanoplasmonic cavity-emitter system consisting of a spherical metallic nanoparticle with radius  $R$  and an emitter at a distance  $s = \xi R$  from the metal surface.

mode), and a two-level quantum emitter which has a frequency difference  $\omega_e$  between its ground  $|g\rangle$  and excited  $|e\rangle$  state ( $S_z = 1/2(|e\rangle\langle e| - |g\rangle\langle g|)$ ,  $S_+ = |e\rangle\langle g|$ ,  $S_- = |g\rangle\langle e|$ ). The intrinsic cavity linewidth  $\gamma_c$  is determined by the radiative decay rate to the nonguided modes ( $\gamma_{rad}$ ) and the absorption decay rate ( $\gamma_{abs}$ ), i.e.,  $\gamma_c = \omega_c/(2Q) = \gamma_{rad} + \gamma_{abs}$ , where  $Q$  is the unloaded quality factor. The overall linewidth  $\gamma_p = \gamma_c + \kappa$  also incorporates the coupling rate  $\kappa$  between the cavity mode (characterized by the operator  $p$ , annihilating a cavity excitation) and the waveguide mode. The coupling strength between the emitter and the cavity is given by  $\Omega$ . For dielectric cavities the emitter decay rate  $\gamma_e$  is approximated by the decay rate of a dipole emitter in a background dielectric with index  $n_d$ , i.e.,  $\gamma_e = \gamma_d = \gamma_0\sqrt{\epsilon_d} = \gamma_0 n_d$  with  $\gamma_0 = \frac{8\pi^2}{3\hbar\epsilon_0} \frac{|\mathbf{d}|^2}{\lambda^3}$ , where  $\mathbf{d}$  is the dipole moment vector of the emitter. For plasmonic cavities,  $\gamma_e$  contains an additional contribution due to nonradiative quenching by higher-order plasmon modes, as shown in Ref. [26], i.e.,  $\gamma_e = \gamma_d + \Omega^2 f_q$  with

$$f_q = \sum_{l=2}^{\infty} \left( \frac{(2l+1)(l+1)^2}{12l(1+\xi)^{2l-2}} \right) \left( \frac{\gamma_c}{(\omega_L - \omega_l)^2 + (\frac{\gamma_c}{2})^2} \right), \quad (1)$$

where  $\omega_l$  is the resonance frequency of the  $l$ th plasmon mode, assuming a spherical nanoplasmonic cavity with radius  $R$  and an emitter positioned at a distance  $s = \xi R$  from the metal surface, and  $\omega_L$  the frequency by which the system is driven. For this particular case, the cavity-emitter coupling strength  $\Omega = \sqrt{\frac{9\pi c^3 \gamma_0}{2\epsilon_d \omega_p^2 (1+\xi)^6 V_c}}$ , while the coupling rate between the cavity

and the waveguide is  $\kappa = \frac{2\chi^\kappa \omega_p^4 \epsilon_d \epsilon_{eff} V_c}{\pi^2 c^3 \epsilon_{wg}}$ , with  $\omega_p$  the plasma frequency of the metal,  $\chi^\kappa$  a factor containing the overlap between the electric fields of the waveguide and the cavity,  $\epsilon_{eff}$  the relative effective dielectric permittivity of the waveguide mode,  $\epsilon_{wg}$  the relative permittivity of the waveguide core, and  $V_c = \frac{\pi R^3}{\epsilon_d}$  the cavity modal volume [26].

As opposed to Ref. [26], where the decay of an initially excited emitter into the waveguide mode without any external driving was studied, we consider a system which is pumped by a continuous coherent probe field and study how the photon statistics of the classical probe field is affected by

the interference with the light emitted from the cavity-emitter system. Moreover, we discuss the differences between the transmitted and reflected guided light (which in the case of Ref. [26] were identical) and derive analytical formulas for the first- and second-order correlation functions in transmission and reflection. The Hamiltonian of this quantum photonic platform is given by

$$\begin{aligned} \mathcal{H} = & \hbar\omega_e S_z + \hbar\omega_c p^\dagger p + \hbar\Omega(pS_+ + p^\dagger S_-) \\ & + \hbar \int dk \omega_k l_k^\dagger l_k + \hbar \int dk \omega_k r_k^\dagger r_k + \mathcal{H}_{drive} \\ & + \hbar g_{wg} \int dk (l_k^\dagger p + l_k p^\dagger) + \hbar g_{wg} \int dk (r_k^\dagger p + r_k p^\dagger). \end{aligned}$$

It includes the free Hamiltonian of the emitter, cavity, and waveguide modes as well as the interaction between the emitter and the cavity and the interaction between the cavity and waveguide modes. The coupling constant between the cavity and the waveguide modes is  $g_{wg} = \sqrt{\frac{c\kappa}{4\pi}}$ . [8] Finally,  $\mathcal{H}_{drive}$  represents the coherent input driving field,

$$\mathcal{H}_{drive} = \hbar F p^\dagger e^{-i\omega_L t} + \hbar \bar{F} p e^{i\omega_L t} \quad (2)$$

oscillating at a frequency  $\omega_L$ . To model the overall dynamics, we effectively integrate out the waveguide modes. To this end we start from the Heisenberg equations for the  $r_k$  and  $l_k$  operators, which can be formally solved using an approach similar to Refs. [7–9]:

$$r_k(t) = r_k(t_0) e^{-i\omega_k(t-t_0)} - i g_{wg} \int_{t_0}^t p(u) e^{-i\omega_k(t-u)} du, \quad (3)$$

$$l_k(t) = l_k(t_0) e^{-i\omega_k(t-t_0)} - i g_{wg} \int_{t_0}^t p(u) e^{-i\omega_k(t-u)} du. \quad (4)$$

Substituting these formal solutions into the Heisenberg equation of the cavity operator, one eventually finds

$$\begin{aligned} \dot{p} = & -i\omega_c p(t) - i g_{wg} \left( \int r_k dk + \int l_k dk \right) - i\Omega S_- \\ = & -i\omega_c p(t) - i g_{wg} \left( \sqrt{2\pi} a_{input}(t) - \frac{4i\pi g_{wg}}{c} p(t) \right) - i\Omega S_- \\ = & -i\omega_c p(t) - \frac{4\pi g_{wg}^2}{c} p(t) - i g_{wg} \sqrt{2\pi} a_{input}(t) - i\Omega S_- \\ = & -i\omega_c p(t) - \kappa p(t) - i \sqrt{\frac{c\kappa}{2}} a_{input}(t) - i\Omega S_-, \end{aligned} \quad (5)$$

whereby

$$a_{input}(t) = \frac{1}{\sqrt{2\pi}} \int r_k(t_0) e^{-i\omega_k(t-t_0)} dk \quad (6)$$

is the right-propagating input field. There is no input field propagating to the left, hence  $\int l_k(t_0) e^{-i\omega_k(t-t_0)} dk = 0$ . Similar to the results obtained in Ref. [8], one can see from Eq. (5) that the infinite waveguide degrees of freedom can be effectively integrated out, such that the dynamics of the overall system is accurately described by incorporating an additional Lindblad term to the master equation, i.e.,  $\frac{\kappa}{2}(2pp^\dagger - p^\dagger pp - pp^\dagger p)$ , which describes the cavity decay into the guided modes. In a frame rotating at  $\omega_L$  the master equation then eventually

becomes

$$\begin{aligned} \frac{d\rho}{dt} = & -\frac{i}{\hbar}[\mathcal{H}_{\text{rot}}, \rho] + \frac{\gamma_p}{2}(2\rho p p^\dagger - p^\dagger p \rho - \rho p^\dagger p) \\ & + \frac{\gamma_e}{2}(2S_- \rho S_+ - S_+ S_- \rho - \rho S_+ S_-), \end{aligned} \quad (7)$$

where  $\mathcal{H}_{\text{rot}} = \hbar\delta_e S_z + \hbar\delta_c p^\dagger p + \hbar\Omega(pS_+ + p^\dagger S_-) + \hbar F p^\dagger + \hbar\bar{F} p$  with  $\delta_e = \omega_e - \omega_L$  and  $\delta_c = \omega_c - \omega_L$ . When incorporating a dephasing term  $\gamma^*(2S_z \rho S_z - S_z S_z \rho - \rho S_z S_z)/2$  in the master equation, no antibunching was observed for realistic room temperature values of  $\gamma^* \sim \mathcal{O}(\text{THz})$  (Ref. [26]), neither in transmission nor reflection. Hence, to observe the nonclassical effects described below, the dephasing rate needs to be negligible. Therefore we will not consider this dephasing term in our subsequent (numerical and analytical) calculations. The coherent driving field strength  $F$  can be related to the input field  $a_{\text{IN}}$  by considering that the coupling between the continuum of forward (right) propagating waveguide modes and the cavity is given by

$$\hbar g_{wg} \int dk r_k^\dagger p + \text{H.c.} \equiv \hbar\bar{F} p + \text{H.c.} \Rightarrow F = g_{wg} \int dk r_k(t_0) \quad (8)$$

and that the input field  $a_{\text{IN}}$  is just the initial field at  $t = t_0$ , i.e.,  $a_{\text{IN}} = \sqrt{2\pi}^{-1} \int dk r_k(t_0)$  [see Eq. (6)]. Hence  $a_{\text{IN}} = \frac{F}{\sqrt{2\pi}g_{wg}} = \sqrt{\frac{2}{c\kappa}} F$ . The cavity is evanescently coupled to the waveguide modes and the reflected (left-propagating)  $a_R$  and transmitted (right-propagating)  $a_T$  fields are respectively given by

$$\begin{aligned} a_R &= \frac{1}{\sqrt{2\pi}} \int l_k(t) dk = -i\sqrt{\frac{\kappa}{2c}} p, \\ a_T &= \frac{1}{\sqrt{2\pi}} \int r_k(t) dk = a_{\text{IN}} - i\sqrt{\frac{\kappa}{2c}} p. \end{aligned} \quad (9)$$

The power reflection and transmission coefficients are then respectively given by  $\mathcal{R} = |r|^2 = |\frac{a_R}{a_{\text{IN}}}|^2$  and  $\mathcal{T} = |\frac{a_T}{a_{\text{IN}}}|^2 = |1 + r|^2$ .

When solving the stationary Heisenberg equations in a frame rotating at the drive frequency  $\omega_L$  (Refs. [7,9]), the average value of the operator  $\langle \Psi | S_z | \Psi \rangle \triangleq s_z$  is approximately given by

$$s_z = -\frac{1}{2} \left( \frac{1}{1 + \frac{|a_{\text{IN}}|^2}{P_c}} \right) = -\frac{1}{2} \left( \frac{1}{1 + x} \right) \quad (11)$$

with

$$P_c = \frac{\gamma_e}{\Omega\sqrt{2c\kappa}\text{Im}(i\tau A) - \Omega^2\text{Re}(\tau)|A|^2}, \quad (12)$$

and  $\frac{1}{\tau} = i\delta_c + \kappa + \frac{\gamma_c}{2}$ ,  $\frac{1}{\beta} = i\delta_e + \frac{\gamma_c}{2}$ ,  $A = \frac{\beta\tau\Omega\sqrt{2c\kappa}}{1 + \Omega^2\beta\tau}$ . In the remainder of this paper we assume to work in the weak probe limit, i.e., when the input power is sufficiently small compared to the critical driving field power ( $\propto P_c$ ) such that  $|a_{\text{IN}}|^2 \ll P_c$  implying  $x \approx 0$ . This means that on average the emitter is in the ground state ( $s_z \approx -1/2$ ). In this limit, we can derive simple analytical approximations for the second-order correlation functions  $g_{\mathcal{T}/\mathcal{R}}^{(2)}$ , as will be shown further on. For all numerical

calculations, we assume a fixed coherent coupling strength  $F = 10^{-5}\omega_c$  to operate in the weak probe limit (in this paper  $\omega_c \approx 2960$  THz corresponding to a free space wavelength of 637 nm).

### III. DISCUSSION

Using the above model, we now investigate the properties of the transmitted and reflected guided light for a waveguide evanescently coupled to a spherical metallic nanoparticle (Fig. 2). We assume that the cavity is perfectly tuned with the emitter ( $\delta_c = \delta_e = \delta$ ). The coupling rate between the cavity and the waveguide is fixed to  $\kappa = 10\gamma_c$  [guaranteeing a coupling efficiency of  $\kappa/(\kappa + \gamma_c) \approx 91\%$ ]. Since  $\kappa \propto V_c$  is fixed, the cavity modal volume is fixed because all other parameters determining  $\kappa$  are fixed as well. Figures 2(a) and 2(b) and Figs. 2(c) and 2(d), respectively, show  $\mathcal{T}/g_{\mathcal{T}}^{(2)}(0)$  and  $\mathcal{R}/g_{\mathcal{R}}^{(2)}(0)$  as a function of the detuning  $\delta$  and emitter-cavity distance  $s$  (which basically tunes the cavity-emitter coupling strength as  $V_c$  is already fixed by fixing  $\kappa$ ). Please note that all quantities are plotted using a  $\log_{10}$  scale. These plots show that both for the transmitted and reflected case, antibunched light is only observed near  $\delta = 0$ . So no photon blockade effect appears for this particular system because for the given  $\kappa$  (which needs to exceed  $\gamma_c$ ),  $\Omega$  cannot be made large enough. Indeed, since  $\kappa \propto V_c \propto R^3$  needs to be larger than  $\gamma_c$  to observe significant antibunching in transmission, the radius of the nanoparticle (and by extension modal volume) needs to be large enough, which in turn makes  $\Omega \propto 1/\sqrt{V_c} \propto R^{-3/2}$  smaller.

One way to maintain a large  $\kappa$  without enlarging the modal volume would be to increase the cavity polarizability because, in general,  $\kappa \propto \alpha_p^2/V_c$ , with  $\alpha_p = \epsilon_0\alpha_0 V_c$  the cavity polarizability, yielding  $\kappa \propto \alpha_0^2 V_c$  (for a spherical nanocavity,  $\alpha_0 = 2\epsilon_d$ ) [26]. By placing two spherical nanoparticles close to each other, the fundamental mode is expected to have an increased dipole moment due to charges built up on both particles. Moreover, when the gap between these particles is sufficiently small, this will also result in an increased field enhancement with respect to a single spherical nanoparticle in the gap near the metal surface (yielding a larger  $\Omega$ ) [27]. To assess what would happen in case of increased cavity polarizability and field enhancement, we consider a hypothetical situation of a nanoplasmonic cavity with cavity polarizability ten times larger than the one for a spherical particle, while keeping  $\kappa = 10\gamma_c$  (implying a decrease in the modal volume by 100), and for which field enhancement effects further increase  $\Omega$  by a factor 100. The functional dependence of the quenching term  $\gamma_e = \gamma_d + \Omega^2 f_q$  is assumed to be the same as for the spherical particle. As shown in Figs. 2(e) and 2(f) an off-resonant anti-bunching in transmission is obtained for this case. However, the photon blockade effect is asymmetric with respect to  $\delta = 0$  due to the Lorentzian lineshape functions appearing in  $f_q$ . Photon blockade will at first only appear when the probe wavelength is redshifted (i.e.,  $\delta > 0$ ) compared to the resonance wavelength of the fundamental plasmonic mode. Since  $\{\omega_l - \omega_1 \geq 0, \forall l\}$ ,  $\gamma_e$  will first increase if  $\delta < 0$  due to the reduced denominator in the Lorentzian lineshape function of  $\gamma_e$ . Therefore a certain minimum  $\Omega$  is required to compensate for the increased quenching effect in the relevant  $\delta < 0$  region.

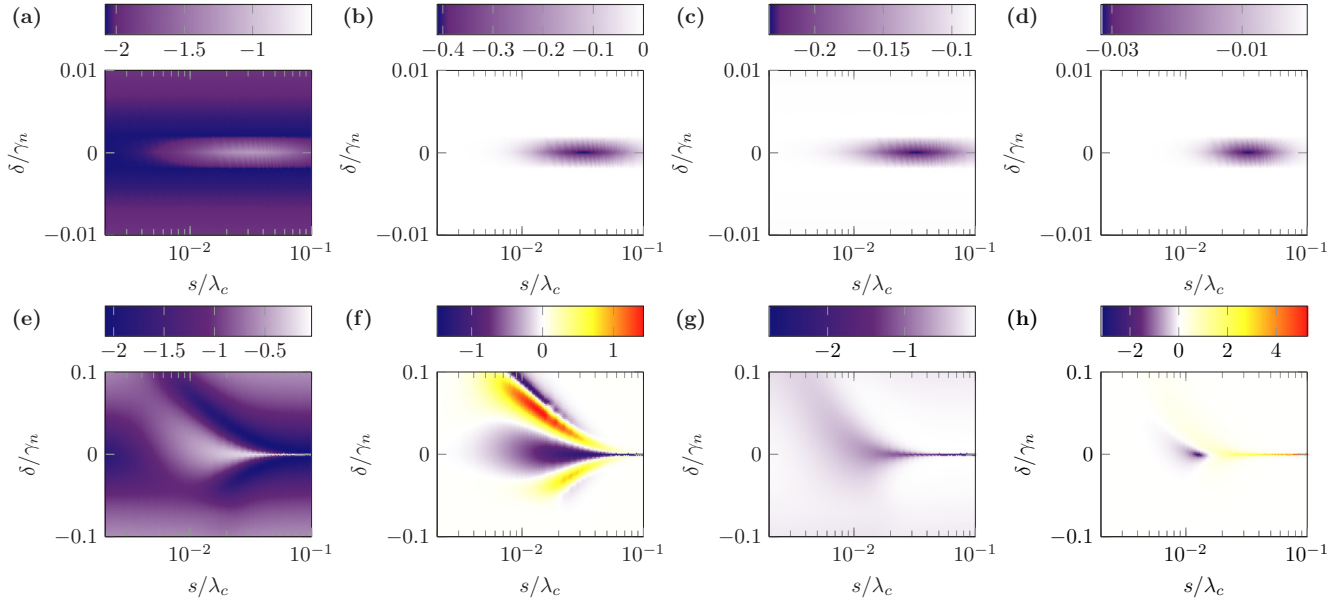


FIG. 2. Properties of the (a), (b), (e), (f) transmitted and (c), (d), (g), (h) reflected guided light for (a)–(d) an integrated spherical nanoparticle with  $Q = 15$  at  $\lambda_c = 637$  nm,  $\kappa = 10\gamma_c$ ,  $\gamma_D = 1$  GHz,  $\epsilon_{\text{eff}} = 2.56$ ,  $\epsilon_{wg} = 4$  (common values for a SiN waveguide),  $\chi^\kappa = 1$  and (e)–(h) a nanoplasmonic cavity with an increased polarizability and field enhancement (as explained in the main text). (a), (e)  $\mathcal{T}$  and (b), (f)  $g_{\mathcal{T}}^{(2)}(0)$  as a function of  $\delta_c = \delta_e = \delta$  ( $\gamma_n = 3\gamma_p$ ) and  $s$ . (c), (g)  $\mathcal{R}$  and (d), (h)  $g_{\mathcal{R}}^{(2)}(0)$  as a function of  $\delta$  and  $s$ . To incorporate quenching, we used 1000 higher-order modes. All quantities are plotted using a  $\log_{10}$  scale.

It is also clear that the transmission of the strongly off-resonant antibunched light is much smaller than the transmission of the resonant antibunched light. Other plasmonic cavities that potentially allow increased polarizability and field enhancement are, e.g., double rod or bowtie antennas [28,29]. Furthermore, quenching could be reduced by considering hybrid metal-dielectric cavities, as recently studied in Ref. [30]. For such intricate nanoplasmonic geometries, careful numerical simulations need to be performed to determine the set of parameters  $[\Omega, \kappa, \gamma_e(\Omega)]$ , which subsequently determine the nonclassical light effects.

In reflection, a negligible antibunching is observed for the spherical nanoplasmonic cavity and the given  $\kappa$ . Since the antibunching in this case solely stems from the light emitted by the quantum emitter and not from an interference between the excitation beam and the emitted light, the conditions are different. In reflection,  $\kappa$  may be smaller than  $\gamma_c$  to obtain strong resonant antibunching, as long as  $\Omega$  satisfies  $\Omega = \sqrt{\gamma_e(\gamma_e + \gamma_p)}/2$  (see below). This condition results in a quadratic equation for  $\Omega^2$  (because  $\gamma_e$  explicitly depends on  $\Omega^2$  through  $\gamma_e = \gamma_d + \Omega^2 f_q$ ), yielding two specific solutions for  $\Omega$  around which strong resonant antibunching is obtained (even if  $\kappa < \gamma_c$ ).

We will now investigate the case of resonant excitation in more detail. Since we assume to work in the weak probe limit, the reflection and transmission coefficients are given by:

$$\mathcal{R} = \left| \frac{\kappa}{\gamma_p(1+C)} \right|^2, \quad (13)$$

$$\mathcal{T} = \left| \frac{\gamma_c + \mathcal{C}\gamma_p}{\gamma_p + \mathcal{C}\gamma_p} \right|^2, \quad (14)$$

where  $\mathcal{C} = \frac{4\Omega^2}{\gamma_p\gamma_e}$  is the cooperativity of the system. In this limit, the Hilbert space used to evaluate the master equation can be truncated to states which have a maximum of two excitations (either emitter or cavity excitations), allowing us to derive analytical formulas for the second-order correlation function  $g^{(2)}(0)$  as a measure for the degree of antibunching. In Appendix B it is shown that in this limit the  $g_{\mathcal{T}/\mathcal{R}}^{(2)}(0)$  functions on resonance are, respectively, given by

$$g_{\mathcal{T}}^{(2)}(0) = \frac{(1+C)^2 \left( 1 + \frac{4\Omega^2[\gamma_c + 2(\gamma_e + \kappa)]}{\gamma_c\gamma_e(\gamma_e + \gamma_p)} + \frac{16\Omega^4}{\gamma_c^2\gamma_e(\gamma_e + \gamma_p)} \right)^2}{\left( 1 + \frac{\gamma_p}{\gamma_c}C \right)^4 \left( 1 + \frac{\gamma_e}{\gamma_e + \gamma_p}C \right)^2} \quad (15)$$

and

$$g_{\mathcal{R}}^{(2)}(0) = \frac{[4\Omega^2 + \gamma_e\gamma_p]^2 (\gamma_e(\gamma_e + \gamma_p) - 4\Omega^2)^2}{\gamma_e^4 [4\Omega^2 + \gamma_p(\gamma_e + \gamma_p)]^2}. \quad (16)$$

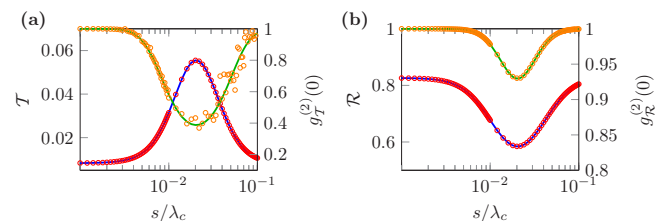


FIG. 3. (a) Numerically calculated (dots) and theoretically predicted (lines) values of  $\mathcal{T}$  (left axis, red dots and blue line) and  $g_{\mathcal{T}}^{(2)}(0)$  (right axis, orange dots and green lines) for  $\delta = 0$ . (b) Same as (a) but for the reflected field. The other parameter values are the same as for Fig. 2.



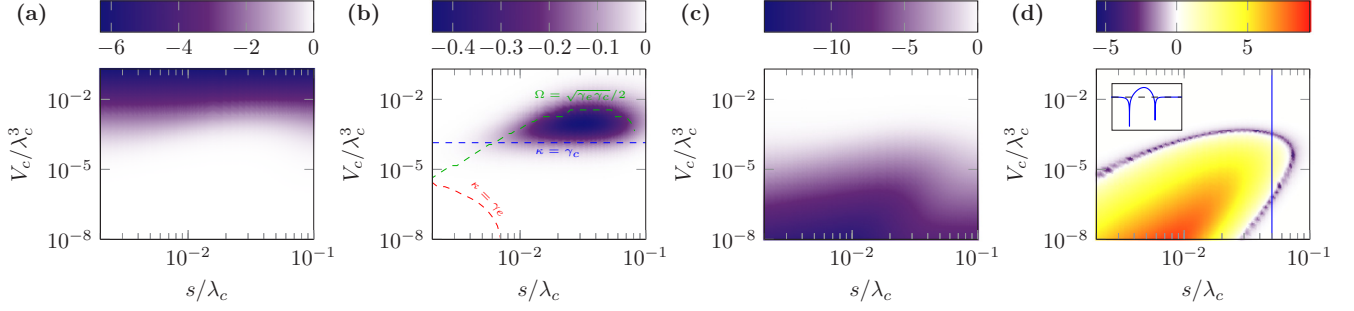


FIG. 4. Properties of the transmitted and reflected light for resonant excitation. (a)  $\mathcal{T}$  and (b)  $g_{\mathcal{T}}^{(2)}(0)$ . The blue dashed line marks  $\kappa = \gamma_c$ , the red dashed line  $\kappa = \gamma_e$  and the green dashed line  $\Omega = \sqrt{\gamma_e \gamma_c}/2$ . (c)  $\mathcal{R}$  and (d)  $g_{\mathcal{R}}^{(2)}(0)$ . The inset in (d) is a slice of  $g_{\mathcal{R}}^{(2)}(0)$  along a fixed  $s$ . All quantities are plotted using a  $\log_{10}$  scale.

Figures 3(a) and 3(b), respectively, show the properties of the transmitted and reflected light. The red and orange dots present the respective numerically calculated  $\mathcal{T}/\mathcal{R}$  and  $g_{\mathcal{T}}^{(2)}(0)/g_{\mathcal{R}}^{(2)}(0)$  while the blue and green solid lines give the respective analytical predictions of  $\mathcal{T}/\mathcal{R}$  and  $g_{\mathcal{T}}^{(2)}(0)/g_{\mathcal{R}}^{(2)}(0)$ . Details on the numerical calculation can be found in Appendix A. The correspondence between the numerical simulation and analytical formula is perfect for  $\mathcal{T}$ ,  $\mathcal{R}$ , and  $g_{\mathcal{R}}^{(2)}(0)$ , while some small deviation arises for  $g_{\mathcal{T}}^{(2)}(0)$ . In Appendix B we discuss a potential reason for this deviation, which we ascribe to finite accuracy in the numerical evaluation. Nevertheless, the oscillations remain small and indeed the predicted analytical value lies within the boundary of these oscillations. As such the analytical formulas derived here can be used to assess the degree of antibunching, in the weak probe limit, for any cavity-emitter system evanescently coupled to a waveguide (the formulas for off-resonant pumping are given in Appendix B as well). It should be noted that these formulas hold for any  $Q$  cavity, both dielectric and plasmonic.

Figure 4 shows the properties of the transmitted and reflected light as a function of  $V_c$  ( $\kappa$ ) and  $s$  ( $\Omega$  for fixed  $V_c$ ). Also here all quantities are plotted using a  $\log_{10}$  scale. It is clear that in transmission, a maximum degree of antibunching is obtained for an optimal cavity modal volume  $V_c$  and cavity-emitter distance  $s$ . Moreover, antibunching can, in general, only be obtained when  $\kappa > \max(\gamma_c, \gamma_e)$ . From the figure this is obvious for  $\kappa > \gamma_c$ . However, it also holds for  $\kappa > \gamma_e$ , e.g., for dielectric cavities with very high  $Q$  it is possible that  $\gamma_e > \gamma_c$ . For plasmonic cavities, the modal volume will generally be  $Q$  factor limited due to the very high  $\gamma_c$ . As such there is a minimum modal volume  $V_{\min}$  for the transmitted light to be antibunched, i.e.,

$$V_c > V_{\min} = \lambda_c^3 \left( \frac{\epsilon_{wg}}{32\pi Q \chi^{\kappa} \epsilon_d \epsilon_{\text{eff}}} \right). \quad (17)$$

Interestingly, this lower bound  $V_{\min}$  equals to the optimum modal volume required for single photon generation when the system is initially pumped to the excited state and the subsequent decay of the emitted light is observed [26]. On the other hand,  $\Omega$  should also be large enough to observe antibunching, as evidenced by the green dashed line in Fig. 4(b). When  $\Omega$  becomes significantly smaller than  $\sqrt{\gamma_e \gamma_c}/2$ , the light becomes coherent again. Thus there is also an upper bound for the cavity

modal volume  $V_{\max}$ , depending on the quenching factor  $f_q$ , i.e.,

$$V_c < V_{\max} = \lambda_c^3 \left( \frac{9Q(4 - \gamma_c f_q)}{8\pi^2 \epsilon_d (1 + \xi)^6} \right) \left( \frac{\gamma_0 \omega_c^2}{\gamma_d \omega_p^2} \right). \quad (18)$$

As a result, similar to the case of pure single photon generation [26], it is only possible to generate nonclassical light in transmission if the modal volume stays within the given bounds. As already highlighted in Ref. [26], it is hence not always beneficial to aim for a minimal cavity modal volume for nonclassical light generation. Despite the fact that the degree of antibunching can be optimized, it is nevertheless impossible to achieve perfectly antibunched light in transmission. Bunching on resonance is also not observed in transmission for the investigated platform.

However, in reflection, the situation is different and perfectly antibunched light can be achieved for  $\Omega_{\mathcal{R}}^{\text{opt}} = \sqrt{\gamma_e(\gamma_e + \gamma_p)}/2$ . The reflected light only exhibits nonclassical behavior in a very narrow region around  $\Omega_{\mathcal{R}}^{\text{opt}}$ , requiring a precise alignment of the quantum emitter with the cavity [Fig. 4(d)]. Within the boundary set by  $\Omega_{\mathcal{R}}^{\text{opt}} = \sqrt{\gamma_e(\gamma_e + \gamma_p)}/2$ , strong bunching is observed for the smaller modal volumes and cavity-emitter distances. Outside this boundary, the light is perfectly coherent. The overall reflection of nonclassical light is, however, relatively low [Fig. 4(c)]. Evaluating  $\mathcal{R}$  at  $\Omega_{\mathcal{R}}^{\text{opt}}$  yields  $\mathcal{R}_{\text{opt}} = \left( \frac{\kappa}{\gamma_e + 2\gamma_c + 2\kappa} \right)^2$ . For  $\kappa > \max(\gamma_e, \gamma_c)$  the maximum reflection of a nonclassical light state is achieved and equals 25%.

#### IV. CONCLUSION

We present a general quantum photonic model of evanescently coupled cavity-emitter systems and investigated the requirements for nonclassical light generation using integrated nanoplasmonic cavities. We consider a spherical metallic nanoparticle as a model system for the nanoplasmonic cavity because analytical formulas can be derived for this specific case. However, the model is generally applicable to any plasmonic (and dielectric) cavity, where the system parameters then need to be obtained from electromagnetic simulations. We derive analytical formulas for the first- and second-order correlation functions in the weak probe limit, both for

transmission and reflection, which showed good correspondence with numerical simulations using a quantum master equation approach. Moreover, we show that nonclassical light generation for the considered nanoplasmonic cavity-emitter system is only possible using resonant excitation and that photon blockade effects only appear for nanoplasmonic cavities with increased polarizability and field enhancement compared to a spherical metallic nanoparticle. This result stems from the intrinsic dependence of the effective emitter decay rate on the cavity-emitter coupling strength due to quenching effects near the plasmonic nanoparticle. Investigation of resonant excitation in more detail showed that in transmission the degree of antibunching is maximized for an optimal cavity modal volume  $V_c$  and cavity-emitter distance  $s$ . In reflection, perfectly antibunched light can be obtained for specific  $(V_c, s)$  combinations. The presented model allows accurate predictions of the photon statistics generated in the guided mode of a waveguide by an integrated nanoplasmonic cavity-emitter system, driven by a weak coherent probe beam. These results inform future efforts in the design and development of on-chip nonclassical light sources.

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#### APPENDIX A: NUMERICAL EVALUATION

The first- and second-order correlation functions can be evaluated using the steady-state ( $t \rightarrow \infty$ ) density matrix  $\rho^{ss}$  which is determined by  $d\rho/dt = 0$ . We can determine the steady state solution by evaluating the master equation in a predefined basis

$$\{|g, 0\rangle, |g, 1\rangle, |g, 2\rangle, \dots, |g, N_{\text{ex}}\rangle, \dots, |e, 0\rangle, |e, 1\rangle, |e, 2\rangle, \dots, |e, N_{\text{ex}} - 1\rangle\}, \quad (\text{A1})$$

which consists of the ground ( $g$ ) and excited ( $e$ ) states of the emitter and a certain number of cavity excitations such that the maximum number of excitations in the system is  $N_{\text{ex}}$ . If the emitter is in the ground state this means that the cavity can have  $N_{\text{ex}}$  excitations, while the cavity can only have up to  $N_{\text{ex}} - 1$  excitations if the emitter is in the excited state. So for  $N_{\text{ex}}$  excitations, the total number of basis states is  $N_b = 2N_{\text{ex}} + 1$ . These states will be numbered by  $\{|b\rangle, b = 1, \dots, N_b\}$ , such

that

$$\{|1\rangle = |g, 0\rangle, \dots, |N_{\text{ex}} + 1\rangle = |g, N_{\text{ex}}\rangle, \dots, |2N_{\text{ex}} + 2\rangle = |e, 0\rangle, \dots, |2N_{\text{ex}} + 1\rangle = |e, N_{\text{ex}} - 1\rangle\}. \quad (\text{A2})$$

To solve the system, we recast the elements of the density matrix  $\rho_{\alpha\beta}^{ss}, \alpha, \beta = 1, \dots, N_b$  into a column vector of length  $N_b^2$ , i.e.,  $V_j^\rho, \{j = 1, \dots, N_b^2\}$  and build up the Liouvillian superoperator  $\mathcal{L}^\rho$ , which is the  $(N_b^2 \times N_b^2)$  matrix representing all equations that determine  $\rho_{\alpha\beta}^{ss}$ , i.e.,

$$\sum_{j=1}^{N_b^2} \mathcal{L}_{ij}^\rho V_j^\rho = 0, \quad \forall i = 1, \dots, N_b^2. \quad (\text{A3})$$

The density matrix elements  $\rho^{ss}$  are now determined by the eigenvector corresponding to the zero eigenvalue of  $\mathcal{L}^\rho$  [31]. For all numerical evaluations we use a maximum number of excitations  $N_{\text{ex}} = 5$ , so the total Hilbert space then consists of 11 basis states.

#### 1. First-order correlation function

The transmission ( $\mathcal{T}$ ) and reflection ( $\mathcal{R}$ ) coefficients are determined by the first-order correlation function  $g_{\mathcal{R}, \mathcal{T}}^{(1)}$

$$g_{\mathcal{P}}^{(1)} = \sum_{m,n=1}^{N_b^2} \rho_{nm}^{ss} \langle m | a_{\mathcal{P}}^\dagger a_{\mathcal{P}} | n \rangle, \quad \mathcal{P} = \{\mathcal{R}, \mathcal{T}\}, \quad (\text{A4})$$

with

$$a_{\mathcal{P}} = a_{\mathcal{P}}^i + \psi p, \quad a_{\mathcal{P}}^i = a_{IN} \delta_{\mathcal{P}\mathcal{T}}, \quad \psi = -i \sqrt{\frac{\kappa}{2c}}. \quad (\text{A5})$$

such that

$$\mathcal{T} = \frac{g_{\mathcal{T}}^{(1)}}{a_{IN}^2}, \quad \mathcal{R} = \frac{g_{\mathcal{R}}^{(1)}}{a_{IN}^2}. \quad (\text{A6})$$

The combination of the matrix elements  $\langle m | a_{\mathcal{P}}^\dagger a_{\mathcal{P}} | n \rangle$  and  $\rho^{ss}$  (calculated using the Liouvillian superoperator method as described before) fully determines the transmission and reflection coefficients (and of course should give the same outcome as the Heisenberg equation analysis).

#### 2. Second-order correlation function

The steady-state second-order correlation function  $g_{\mathcal{R}, \mathcal{T}}^{(2)}(\tau = 0)$  for the reflected and transmitted field is determined by

$$g_{\mathcal{P}}^{(2)}(0) = \frac{\sum_{m,n=1}^{N_b^2} \rho_{nm}^{ss} \langle m | a_{\mathcal{P}}^\dagger a_{\mathcal{P}}^\dagger a_{\mathcal{P}} a_{\mathcal{P}} | n \rangle}{(g_{\mathcal{P}}^{(1)})^2}, \quad \mathcal{P} = \{\mathcal{R}, \mathcal{T}\}. \quad (\text{A7})$$

#### APPENDIX B: ANALYTICAL APPROXIMATION

We derive an approximate analytical formula for  $g_{\mathcal{R}, \mathcal{T}}^{(2)}$  by limiting the Hilbert space to only five possible states, i.e., the maximum number of excitations in the system is  $N_{\text{ex}} = 2$  such that the possible states are  $\{|1\rangle = |g, 0\rangle, |2\rangle = |g, 1\rangle, |3\rangle = |g, 2\rangle, |4\rangle = |e, 0\rangle, |5\rangle = |e, 1\rangle\}$ . For notational simplicity we note  $\rho^{ss} = \rho$  further on. Using the earlier introduced notations, the numerator of

$g_{\mathcal{P}}^{(2)}(0)$  ( $\mathcal{P} = \{\mathcal{R}, \mathcal{T}\}$ ) is then

$$|a_{\mathcal{P}}^i|^4 + 4|a_{\mathcal{P}}^i|^2 \text{Re}[\overline{a_{\mathcal{P}}^i} \psi(\rho_{21} + \sqrt{2}\rho_{32} + \rho_{54})] + 2\sqrt{2} \text{Re}[(\overline{a_{\mathcal{P}}^i})^2 \psi^2 \rho_{31}] + 4|a_{\mathcal{P}}^i|^2 |\psi|^2 (\rho_{22} + 2\rho_{33} + \rho_{55}) + 4\sqrt{2} |\psi|^2 \text{Re}[\overline{a_{\mathcal{P}}^i} \psi \rho_{32}] + 2|\psi|^4 \rho_{33}. \quad (\text{B1})$$

Similarly, the denominator is given by the square of

$$g_{\mathcal{P}}^{(1)} = |a_{\mathcal{P}}^i|^2 + 2\text{Re}[\overline{a_{\mathcal{P}}^i} \psi(\rho_{21} + \sqrt{2}\rho_{32} + \rho_{54})] + |\psi|^2 (\rho_{22} + 2\rho_{33} + \rho_{55}). \quad (\text{B2})$$

In the weak probe limit we assume a sufficiently small coherent driving strength  $F$  such that the expansion coefficients  $c_i$  ( $i = 1, \dots, 5$ ) in the wave function  $|\Psi\rangle = \sum_{i=1}^5 c_i |i\rangle$  are  $c_1 \approx 1$ ,  $c_{2,4} = \mathcal{O}(F)$  and  $c_{3,5} = \mathcal{O}(F^2)$ . This means that the cavity-emitter system is mostly in the ground state  $\rho_{11} \approx 1$  and that the one-excitation (2,4) and two-excitation (3,5) states are appropriately described by terms up to order one or two in  $F$ , respectively. As such we only keep terms up to order  $F$  for  $\rho_{12}, \rho_{14}$ , to order  $F^2$  for  $\rho_{13}, \rho_{15}, \rho_{22}, \rho_{2,4}, \rho_{44}$ , to order  $F^3$  for  $\rho_{23}, \rho_{25}, \rho_{3,4}, \rho_{45}$  and to order  $F^4$  for  $\rho_{33}, \rho_{35}, \rho_{55}$  in the master equation. The transmission and reflection coefficients are then determined by  $\mathcal{P} = \frac{g_{\mathcal{P}}^{(1)}}{|a_{IN}|^2}$ . For the transmission this means

$$\mathcal{T} \approx 1 + \frac{2}{|a_{IN}|^2} \text{Re}[\overline{a_{\mathcal{T}}^i} \psi \rho_{21}] + \frac{|\psi|^2}{|a_{IN}|^2} \rho_{22} = 1 + \frac{2|\psi|^2 c^2}{F^2} \text{Re}\left(\frac{-iF}{c} \rho_{21}\right) + \frac{|\psi|^4 c^2}{F^2} \rho_{22} = 1 + \frac{\kappa}{F} \text{Im}(\rho_{21}) + \frac{\kappa^2}{4F^2} \rho_{22},$$

while the reflection coefficient is given by

$$\mathcal{R} = \frac{\kappa^2}{4F^2} \rho_{22}. \quad (\text{B3})$$

Introducing the density matrix elements as calculated by the simplified master equation yields the same reflection and transmission coefficient as the one obtained through the Heisenberg equation analysis, which serves as a sanity check of the applied approximations. In a similar way the  $g_{\mathcal{T}}^{(2)}$  function is determined by

$$g_{\mathcal{T}}^{(2)}(0) \approx \frac{1}{\mathcal{T}^2} \left( 1 + \frac{2\kappa}{F} \text{Im}(\rho_{21}) - \frac{\sqrt{2}\kappa^2}{2F^2} \text{Re}(\rho_{31}) + \frac{\kappa^2}{F^2} \rho_{22} + \frac{\sqrt{2}\kappa^3}{2F^3} \text{Im}(\rho_{32}) + \frac{\kappa^4}{8F^4} \rho_{33} \right) \quad (\text{B4})$$

and  $g_{\mathcal{R}}^{(2)}$  is given by

$$g_{\mathcal{R}}^{(2)}(0) \approx \frac{\kappa^4}{8F^4 \mathcal{R}^2} \rho_{33} = \frac{2\rho_{33}}{\rho_{22}^2}. \quad (\text{B5})$$

The calculation of the density matrix elements  $\rho_{mn}$  is straightforward but tedious and can be done with MATHEMATICA. For nonzero detuning and  $\delta_e = \delta_c = \delta$  the second-order correlation function in transmission is given by

$$g_{\mathcal{T}}^{(2)}(0) = \left\{ [(4\Omega^2 + \gamma_e \gamma_p)^2 + 4\delta^2(\gamma_e^2 + \gamma_p^2 - 8\Omega^2) + 16\delta^4] [256\Omega^8 + 128\Omega^6(\gamma_c^2 + 2\gamma_c(\gamma_e + \kappa) - 12\delta^2) + (\gamma_c^2 + 4\delta^2)^2(\gamma_e^2 + 4\delta^2)((\gamma_e + \gamma_p)^2 + 16\delta^2) + 8\Omega^2(\gamma_c^2 + 4\delta^2)(\gamma_e \gamma_c(\gamma_e + \gamma_p)(\gamma_c + 2(\gamma_e + \kappa)) + 4\delta^2(\gamma_c(3\gamma_e - 4\kappa) - (\gamma_e + \kappa)(3\gamma_e + 2\kappa)) - 96\delta^4) + 16\Omega^4(\gamma_c^2(\gamma_c^2 + 2(\gamma_e + \kappa)(3\gamma_e + 2\kappa) + \gamma_c(6\gamma_e + 4\kappa)) + 8\delta^2(\gamma_c^2 - 5\gamma_c \gamma_e + (\gamma_e + \kappa)(\gamma_e + 2\kappa)) + 208\delta^4)] \right\} / \left\{ [(4\Omega^2 + \gamma_e \gamma_c)^2 + 4\delta^2(\gamma_e^2 + \gamma_c^2 - 8\Omega^2) + 16\delta^4] \right. \\ \left. \times (16\Omega^4 + 8\Omega^2(\gamma_p(\gamma_e + \gamma_p) - 8\delta^2) + (\gamma_p^2 + 4\delta^2)((\gamma_e + \gamma_p)^2 + 16\delta^2)) \right\} \quad (\text{B6})$$

while in reflection we find

$$g_{\mathcal{R}}^{(2)}(0) = \frac{[(4\Omega^2 + \gamma_e \gamma_p)^2 + 4\delta^2(\gamma_e^2 + \gamma_p^2 - 8\Omega^2) + 16\delta^4] [(\gamma_e(\gamma_e + \gamma_p) - 4\Omega^2)^2 + 4\delta^2(16\Omega^2 + 5\gamma_e^2 + 2\gamma_e \gamma_p + \gamma_p^2) + 64\delta^4]}{(\gamma_e^2 + 4\delta^2)^2 [(4\Omega^2 + \gamma_p(\gamma_e + \gamma_p))^2 + 4\delta^2(\gamma_e^2 + 2\gamma_e \gamma_p + 5\gamma_p^2 - 16\Omega^2) + 64\delta^4]}. \quad (\text{B7})$$

#### Remark on the local oscillation in $g_{\mathcal{T}}^{(2)}(0)$

Due to the perfect correspondence for  $\mathcal{T}$ ,  $\mathcal{R}$ , and  $g_{\mathcal{R}}^{(2)}(0)$  we can safely say that the driving field used in the calculations indeed satisfies the weak probe limit. Considering the analytical expressions for these quantities, it is reasonable to assume that the problem arises in the evaluation of  $\frac{\sqrt{2}\kappa^2}{2F^2} [-\text{Re}(\rho_{31}) + \frac{\kappa}{F} \text{Im}(\rho_{32})]$  because all the other terms in the evaluation of  $g_{\mathcal{T}}^{(2)}(0)$  also appear in the evaluation of  $\mathcal{T}$ ,  $\mathcal{R}$ , and  $g_{\mathcal{R}}^{(2)}(0)$ . Usually the quantities  $\rho_{31}$  and  $\rho_{32}$  will both be extremely small as the system is mostly in the ground state (weak probe approximation). We therefore expect that numerical inaccuracies can easily induce local oscillations in  $g_{\mathcal{T}}^{(2)}(0)$  due to the different signs in front of  $\text{Re}(\rho_{31})$  and  $\text{Im}(\rho_{32})$  (combined with the fact that both terms are usually so small that their numerical values are likely close to the finite accuracy limit of MATLAB).



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