

**Impact of nonradiative line broadening on emission in photonic and plasmonic cavities**Pavel Ginzburg,<sup>1,2,\*</sup> Alexey V. Krasavin,<sup>1</sup> David Richards,<sup>1</sup> and Anatoly V. Zayats<sup>1</sup><sup>1</sup>*Department of Physics, King's College London, Strand, London WC2R 2LS, United Kingdom*<sup>2</sup>*St. Petersburg National Research University of Information Technologies, Mechanics & Optics, St. Petersburg 197101, Russia*

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A light-matter interaction modified by the material environment is one of the central topics in quantum electrodynamics. While a strong coupling between a single emitter and a cavity and the Markovian (exponential) relaxation regime are most straightforwardly covered by theory, real physical systems that include also various line broadening effects may possess a much more complicated behavior. Here we propose a theoretical framework to account for nonradiative interaction effects in emission in photonic and plasmonic cavities. The quantum electrodynamics model formulated via a stochastic Hamiltonian approach has been developed with nonradiative line broadening introduced via the Kubo oscillator model. The impact of competing radiative and nonradiative processes on the emitter dynamics has been studied, showing that nonradiative relaxations, having significant impact on processes in high- $Q$  photonic cavities, are much less influential in the plasmonic regime. The developed theoretical framework is not restricted to the emitter in a cavity example, but represents a general tool for multiple stochastic Hamiltonian evolution, important for various types of interactions where either classical or quantum noise is present.

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**I. INTRODUCTION**

The phenomenon of spontaneous emission of electromagnetic radiation is the most common and transparent manifestation of a quantum noise. While in free space these vacuum fluctuations are solely dependent on fundamental physical constants and the frequency of the radiation, they can be manipulated through the introduction of a material environment. In spite of the overall conservation of the total density of states [1], the electromagnetic modes can be rearranged in the frequency domain and give rise to a modified spontaneous emission rate, the so-called Purcell effect [2]. The Purcell factor is approximately proportional to the ratio of the quality factor of a cavity to its modal volume  $\frac{Q}{V}$  and can be influenced by the manipulation of these two quantities. Photonic cavities may deliver high Purcell enhancements due to their high quality factors [3], while plasmonic nanostructures do so because of small modal volumes [4,5]. These unique properties make plasmonic structures perfect candidates for the realization of optical antennas, with various applications [6–9]. Being based on interference effects, modal volumes of confined modes in photonic structures are bounded from below by the conventional diffraction limit. On the other hand, negative permittivity plasmonic nanostructures can confine light much below the diffraction limit [10–12], but due to inherent material losses have small quality factors (of the order of tens when defined by the ratio of the real and imaginary parts of material permittivity at the operation frequency [13]). It is worth noting that in the plasmonic case, the general expression for the Purcell factor is not exact due to radiation quenching and can be used only as a qualitative design guideline [14].

The study of modified light-matter interactions is one of the central topics of cavity quantum electrodynamics and is typically restricted to single-mode to single-emitter

coupling. Nevertheless, real physical systems contain the whole span of additional physical mechanisms influencing the interaction dynamics, such as nonradiative transitions and associated line broadening present in molecular and solid-state systems [15,16]. Hence, in order to understand the dynamic evolution of complex quantum systems in the structured material environment, nonradiative deexcitation dynamics should be taken into account.

In this paper we investigate the influence of nonradiative transitions on an emitter in a cavity, either photonic [Fig. 1(a)] or plasmonic [Fig. 1(b)]. The quantum electrodynamics model for light-matter interaction in a cavity [17] has been reformulated with the help of a stochastic Hamiltonian approach [18] and the nonradiative broadening introduced via the Kubo oscillator model [19]. The competition between radiative and nonradiative processes in the system's dynamics has been investigated in both weak- and strong-coupling regimes.

**II. MODEL CONSIDERATIONS**

The leakage of a photon from a cavity is generally described via the interaction with a thermal bath of modes having infinite degrees of freedom. The bath degrees of freedom could be traced out by adopting the density-matrix approach or they could be treated as stochastic noise operators. The equations of motion corresponding to the latter approach are derived from the quantum Heisenberg-Langevin evolution and are preferable over the density-matrix formalism if the time dependence of the operators is of particular interest [17]. The Kossakowski-Lindblad equation, extensively used for the nonunitary description of dissipative and decoherence influenced systems, is, however, restricted to the treatment of Markovian dynamics and cannot be used in the strong-coupling regime [18]. Following the stochastic formulation of the interaction, the nonradiative line broadening can be introduced via the so-called Kubo oscillator model. The general idea behind this approach is to modulate the natural frequency of an oscillator  $\omega_{21}$  by a stochastic term  $\omega_{\text{Kubo}}(t)$

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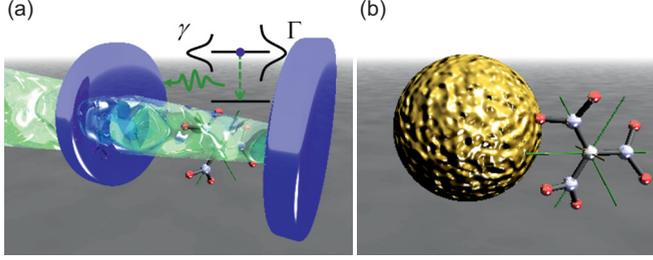


FIG. 1. (Color online) Schematic representations of an emitter in (a) photonic and (b) plasmonic cavities. The inset shows a two-level emitter with natural and nonradiative line broadenings.

describing the nonradiative broadening. The generic equation of motion can then be written as  $\dot{x}(t) = i[\omega_{21} + \omega_{\text{Kubo}}(t)]x(t)$  and will result in the frequency broadening of the emission line, governed by the statistics of the stochastic process in the model. This will be accompanied by dissipation according to the fluctuation-dissipation theorem. In the following, the stochastic approach for the description of a two-level system broadened by the Kubo Hamiltonian and situated in a single-mode leaky cavity will be developed and solved.

### III. MASTER EQUATIONS FOR AN EMITTER WITH NONRADIATIVE BROADENING IN A LEAKY CAVITY

The Hamiltonian of the system  $\hat{H}_{\text{tot}}$  relying on the above considerations and decomposed into several terms, i.e., the two-level system  $\hat{H}_{\text{TLS}}$ , photonic cavity  $\hat{H}_{\text{cavity}}$ , electromagnetic field outside the cavity  $\hat{H}_{\text{field}}$ , two-level-system–cavity photon coupling  $\hat{H}_{\text{TLS-cavity}}$ , cavity mode–outside field coupling  $\hat{H}_{\text{cavity-field}}$ , and Kubo oscillator as the nonradiative broadening description  $\hat{H}_{\text{Kubo}}$ , is given by

$$\begin{aligned}\hat{H}_{\text{TLS}} &= \hbar \frac{\omega_{21}}{2} \hat{\sigma}_0, \\ \hat{H}_{\text{cavity}} &= \hbar \omega_c \left( \hat{a}^\dagger \hat{a} + \frac{1}{2} \right), \\ \hat{H}_{\text{field}} &= \hbar \int_{-\infty}^{\infty} d\omega \omega \hat{b}^\dagger(\omega) \hat{b}(\omega), \\ \hat{H}_{\text{TLS-cavity}} &= \hbar \chi (\hat{a}^\dagger \hat{\sigma}_- + \hat{\sigma}_+ \hat{a}), \\ \hat{H}_{\text{cavity-field}} &= i \hbar \int_{-\infty}^{\infty} d\omega \nu(\omega) [\hat{b}^\dagger(\omega) \hat{a} - \hat{a}^\dagger \hat{b}(\omega)], \\ \hat{H}_{\text{Kubo}} &= \hbar \frac{\sqrt{\Gamma}}{2} n(t) \hat{\sigma}_0, \\ \hat{H}_{\text{tot}} &= \hat{H}_{\text{TLS}} + \hat{H}_{\text{cavity}} + \hat{H}_{\text{field}} \\ &\quad + \hat{H}_{\text{TLS-cavity}} + \hat{H}_{\text{cavity-field}} + \hat{H}_{\text{Kubo}},\end{aligned}\quad (1)$$

where  $\hat{a}^\dagger$  and  $\hat{a}$  are creation and annihilation photonic operators, respectively, with the corresponding Pauli matrices given by  $\hat{\sigma}_+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}$ ,  $\hat{\sigma}_- = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}$ , and  $\hat{\sigma}_0 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$ . In addition,  $\omega_{21}$  and  $\omega_c$  are the central frequencies of the two-level system and the cavity, respectively,  $\chi$  is the emitter-cavity coupling constant,  $\nu(\omega)$  describes the cavity mode leakage,  $\frac{\Gamma}{2}$  is the nonradiative linewidth, and  $n(t)$  is the classical noise and has

dimensions of  $[s^{-1/2}]$ , as follows from the definition of the commutation relation.

While the Kubo term  $\hat{H}_{\text{Kubo}}$  with  $\langle n(t)n(t') \rangle = \delta(t-t')$  is already written as the stochastic Hamiltonian with an uncorrelated noise model, the cavity leakage term can be recast as

$$\hat{H}_{\text{cavity-field}} = i \hbar \sqrt{\gamma} [\hat{b}^\dagger(t) \hat{a} - \hat{a}^\dagger \hat{b}(t)],\quad (2)$$

where the first Markov approximation of the frequency-independent coupling constant, namely,  $\nu(\omega) = \sqrt{\gamma/2\pi}$ , was employed [18]. Here  $b(t)$  and  $b^\dagger(t)$  are stochastic quantum operators, obeying the commutation relations  $[b(t), b^\dagger(t')] \sim \delta(t-t')$  and regarded as a quantum white noise with a Gaussian distribution. Consequently, the quantum Wiener process is defined as  $\hat{B}(t, t_0) = \int_{t_0}^t b(t') dt'$  and obeys the following relations:

$$\begin{aligned}d\hat{B}(t) d\hat{B}(t) &= d\hat{B}^\dagger(t) d\hat{B}^\dagger(t) = 0, \\ d\hat{B}(t) d\hat{B}^\dagger(t) &= (\bar{N}_{\text{th}} + 1) dt, \\ d\hat{B}^\dagger(t) d\hat{B}(t) &= \bar{N}_{\text{th}} dt,\end{aligned}\quad (3)$$

while the higher-order correlations are vanishing in the short-time limit. Here  $\bar{N}_{\text{th}}$  is the thermal photon occupation number that follows the Bose-Einstein statistics of a thermal bath  $\bar{N}_{\text{th}} = [\exp(\frac{\hbar\omega}{k_B T}) - 1]^{-1}$ . Hereafter we assume that the chromatic dispersion of the thermal bath can be neglected over the bandwidth of the cavity.

The system's propagator in the interaction picture satisfies the following Schrödinger equation:

$$i \hbar \frac{\partial \hat{U}(t, t_0)}{\partial t} = \hat{H}_{\text{int}} \hat{U}(t, t_0),\quad (4)$$

where  $\hat{H}_{\text{int}} = \hat{H}_{\text{TLS-cavity}} + \hat{H}_{\text{cavity-field}} + \hat{H}_{\text{Kubo}}$ . In order to derive the quantum stochastic differential equation (QSDE), the perturbation series should be kept to second order, as the multiplicative noise terms will give nonvanishing contributions. The perturbation series expansion leads to

$$\begin{aligned}\hat{U}(t, t_0) &= \exp\left(-\frac{i}{\hbar} \int_{t_0}^t \hat{H}_{\text{int}}(t') dt'\right) \\ &\approx 1 - \frac{i}{\hbar} \int_{t_0}^t \hat{H}_{\text{int}}(t') dt' - \frac{1}{\hbar^2} \int_{t_0}^t dt_1 \\ &\quad \times \int_{t_0}^{t_1} dt_2 \hat{H}_{\text{int}}(t_1) \hat{H}_{\text{int}}(t_2).\end{aligned}\quad (5)$$

The evaluation can be performed by means of the Itô formulation with proper time ordering of the integration. Using the definition of the quantum Wiener process, namely,  $\hat{b}(t) dt = d\hat{B}(t)$ , Eq. (5) can be rewritten as

$$\begin{aligned}\hat{U}(t + dt, t) &\approx 1 + \sqrt{\gamma} (d\hat{B}^\dagger \hat{a} - \hat{a}^\dagger d\hat{B}) \\ &\quad - \frac{1}{2} \gamma [\bar{N}_{\text{th}} \hat{a} \hat{a}^\dagger + (\bar{N}_{\text{th}} + 1) \hat{a}^\dagger \hat{a}] dt \\ &\quad - i \frac{\sqrt{\Gamma}}{2} \hat{\sigma}_0 dN - \frac{\Gamma}{8} dt,\end{aligned}\quad (6)$$

where  $dN$  is the classical Wiener process [20], corresponding to the Kubo oscillator noise. In the derivation of Eq. (6), the

following Itô integration identities were used:

$$I \int_{t_0}^t f(t') d\hat{B}(t') = I \int_{t_0}^t d\hat{B}(t') f(t'),$$

$$I \int_{t_0}^{t_1} \int_{t_0}^{t_2} d\hat{B}^\dagger(t') d\hat{B}(t'') f(t') g(t'') = \bar{N}_{\text{th}} \int_{t_0}^{\min(t_1, t_2)} f(t) g(t) dt,$$

where  $I$  indicates an Itô-type integration.

Now the QSDE governs the time evolution of any system's operator  $\hat{\Theta}$  in the interaction picture, given by

$$\hat{\Theta}(t + dt) = \hat{U}^\dagger(t + dt, t) \hat{\Theta}(t) \hat{U}(t + dt, t), \quad (7)$$

leading to

$$d\hat{\Theta} = \frac{\gamma}{2} \bar{N}_{\text{th}} [2\hat{\Theta} \hat{a}^\dagger - \hat{\Theta} \hat{a} \hat{a}^\dagger - \hat{a} \hat{a}^\dagger \hat{\Theta}] dt$$

$$+ \frac{\gamma}{2} (\bar{N}_{\text{th}} + 1) [2\hat{a}^\dagger \hat{\Theta} \hat{a} - \hat{\Theta} \hat{a}^\dagger \hat{a} - \hat{a}^\dagger \hat{a} \hat{\Theta}] dt$$

$$- \sqrt{\gamma} [d\hat{B}^\dagger \hat{a} - \hat{a}^\dagger d\hat{B}, \hat{\Theta}] - \frac{\Gamma}{4} \hat{\Theta}(t) dt$$

$$+ \frac{\Gamma}{4} \hat{\sigma}_0 \hat{\Theta}(t) \hat{\sigma}_0 dt + i \frac{\sqrt{\Gamma}}{2} [\hat{\sigma}_0, \hat{\Theta}(t)] dN. \quad (8)$$

The Schrödinger picture description can be connected to the above by adding the  $\frac{i}{\hbar} [\hat{H}_0, \hat{\Theta}] dt$  term, where  $\hat{H}_0 = \hat{H}_{\text{tot}} - \hat{H}_{\text{int}}$ . Equation (8) is regarded as a generalized master QSDE of the two-level system in a leaky cavity and nonradiative line broadening. In particular, restricting the consideration to the pure Kubo contributions [last three terms in Eq. (8)], the relation  $\hat{\sigma}_+(t) = \hat{\sigma}_+(0) \exp(i\sqrt{\Gamma} dN)$  can be derived, demonstrating the full correspondence with the classical Kubo oscillator case. The precise time propagator (8) enables investigation of full time-dependent dynamics not restricted to the Markovian approximation.

In general, the analytic description of the system of equations (1) by means of the operators' evolution, given by Eq. (8), involves an infinite number of coupled QSDEs and cannot be solved analytically. Nevertheless, neglecting the high (more than one) number of photons in the cavity [17], the problem can be reduced to the finite solvable set given by

$$d \begin{pmatrix} \langle \hat{\sigma}_0 \rangle \\ \langle \hat{a}^\dagger \hat{a} \rangle \\ \langle \hat{A}_1 \rangle \\ \langle \hat{A}_2 \rangle \\ \langle \hat{A}_3 \rangle \end{pmatrix} = \begin{pmatrix} 0 & 0 & -2\chi & 0 & 0 \\ 0 & -\gamma & \chi & 0 & 0 \\ \chi & 0 & -\frac{\gamma+\Gamma}{2} & 2\chi & \omega_c - \omega_{21} \\ \gamma \bar{N}_{\text{th}} & 0 & -\chi & -\gamma & 0 \\ 0 & 0 & \omega_{21} - \omega_c & 0 & -\frac{\gamma+\Gamma}{2} \end{pmatrix}$$

$$\times \begin{pmatrix} \langle \hat{\sigma}_0 \rangle \\ \langle \hat{a}^\dagger \hat{a} \rangle \\ \langle \hat{A}_1 \rangle \\ \langle \hat{A}_2 \rangle \\ \langle \hat{A}_3 \rangle \end{pmatrix} dt + \begin{pmatrix} 0 \\ \gamma \bar{N}_{\text{th}} \\ \chi \\ 0 \\ 0 \end{pmatrix}, \quad (9)$$

where the following auxiliary operators were defined:  $\hat{A}_1 = i(\hat{\sigma}_+ \hat{a} - \hat{a}^\dagger \hat{\sigma}_-)$ ,  $\hat{A}_2 = \hat{a}^\dagger \hat{\sigma}_0 \hat{a}$ , and  $\hat{A}_3 = \hat{\sigma}_+ \hat{a} + \hat{a}^\dagger \hat{\sigma}_-$ . In particular,  $\hat{A}_3$  should be taken into account if the central emission line of the two-level system is detuned from the cavity resonance, namely,  $\omega_{21} \neq \omega_c$ . The set of equations

(9) is formulated for statistically averaged operators, where nonanticipating noise terms were averaged out due to the Itô-type formulation of the stochastic process. The system of equations (9) can be solved analytically via  $(5 \times 5)$ -matrix diagonalization; however, numerical integration routines were used in the following in order to simplify cumbersome expressions.

#### IV. NUMERICAL RESULTS

We have studied several possible scenarios for different relative cavity and emission lifetimes, corresponding to weak- and strong-coupling regimes and low- $Q$  (plasmonic) and high- $Q$  (photonic) cavities. The two-level system's time-dependent population  $P_a = \frac{1+\langle \hat{\sigma}_0 \rangle}{2}$  and photon number state  $\langle \hat{a}^\dagger \hat{a} \rangle$  in the cavity dynamics have been investigated. The system's parameters were chosen to correspond to realistic characteristic values of similar optical arrangements. In particular, the radiative lifetime of an emitter in the free space  $\Gamma_0^{-1}$  was chosen to be 1 ns, the nonradiative damping time  $(\frac{\Gamma}{2})^{-1}$  is 1 ps, and the central emission frequency, resonant with the cavity ( $\omega_{21} = \omega_c$ ), corresponds to the vacuum wavelength 500 nm. The rest of the parameters were defined in the standard way to allow comparison between the model parameters ( $\omega_c$  and  $\gamma$ ) and the experimentally measured cavity parameters:  $Q = \frac{\omega_c}{\gamma}$ , the cavity quality factor, which depends on the leakage rate  $\gamma$ , and  $\Gamma_C = \Gamma_0 Q \frac{2\pi c^3}{V \omega_c^3}$ , the cavity decay rate, which depends on  $Q$  and the cavity modal volume  $V$  and is related to the coupling constant  $\chi = 0.5\sqrt{\gamma \Gamma_C}$ . While all the above quantities and relations were defined for the ideal (without nonradiative broadening) scenario [17], they will be used for the solution of the system of equations (9) in order to compare possible physical situations. The actual design of the cavity, such as material composition, size, and position of the emitter relative to the cavity, will obviously affect the quantum dynamics, but all these factors are taken into account in a model description via coupling constants in the Hamiltonians (1). The nonradiative and radiative times may become comparable in both plasmonic and photonic cavities, as the radiative rate is affected by the Purcell factor of a cavity. The differences in the quantum dynamics of the emitter in photonic and plasmonic cavities are described below.

##### A. Photonic cavity

The case of a photonic cavity is presented in Fig. 2. To understand the impact of the nonradiative broadening on the evolution of the emitter, the nonradiative scattering was first neglected [Figs. 2(a) and 2(b)] and then reintroduced [Figs. 2(c) and 2(d)]. For a small quality factor of the cavity, without nonradiative broadening, the exponential decay of the excited-state population is observed. Since the emitted photon leakage rate from the cavity is much faster than the Rabi oscillation frequency, the probability of photons to be reabsorbed by the emitter is negligible and they are radiated in the far field outside the cavity. Small  $Q$  and nonradiative broadening result in the exponential decay of the emitter as well. In this case, however, the inequality of rates  $\gamma \ll \Gamma$  governs the exponential decay due to the nonradiative energy relaxation. In both of the above cases, a pure exponential decay

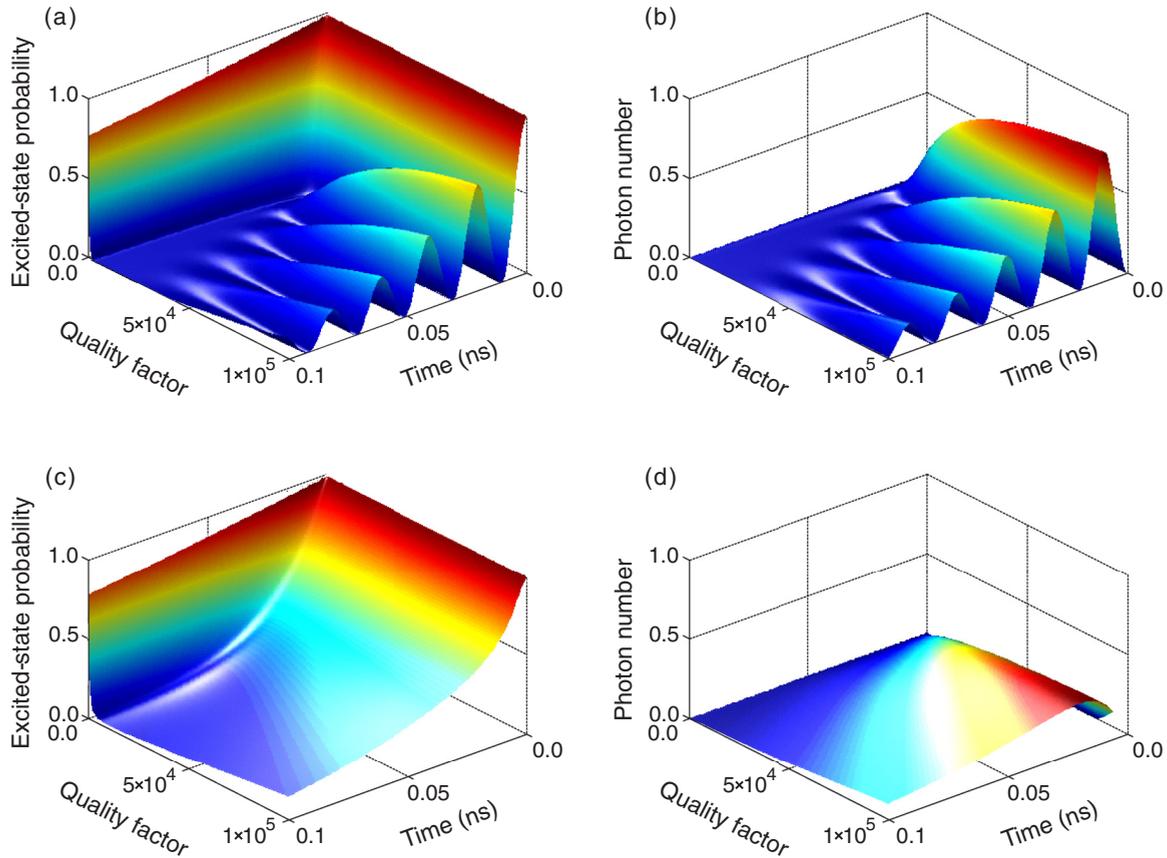


FIG. 2. (Color online) Dependence of the time evolution of an emitter in a photonic cavity on a cavity quality factor: (a) and (c) probability of the two-level system being in the excited state and (b) and (d) number state of the cavity photon. The emitter (a) and (b) without and (c) and (d) with nonradiative broadening has been considered.

is observed and is the direct result of vanishing environmental feedback, since the Purcell factor here is small. It is worth noting that the exponential lifetime decreases rapidly with the increase of the  $Q$  factor of the cavity.

However, the increase of the quality factor reduces the probability of the photon escaping from the cavity and, as a result, increases the probability of reabsorption. This manifests itself in the oscillatory behavior of the emitter's inversion [Fig. 2(a)], which represents multiple reabsorption-reemission events. While this intuitive description works in the absence of nonradiative processes, the presence of the nonradiative decay, according to the fluctuation-dissipation theorem, smears out the Rabi oscillations in the cavity. The coherence buildup between the emitter and the cavity photon observed in Fig. 2(a) is completely destroyed by the short-memory scattering process [Fig. 2(c)]. In other words, additional noise destroys the quantum entanglement between the emitter and the cavity photon. As a result, previously observable revivals in the dynamics are totally suppressed by a pure exponential decay [Fig. 2(c)]. Thus, in the high- $Q$  regime, the instantaneous Purcell factor is high, but does not reflect the emission rates because of the probable photon revivals (it should be noted that the Purcell factor defined as the ratio of the emission rates in vacuum and structured environments is strictly valid only for exponential decays).

The number state time evolution has also nontrivial dynamics in the high- $Q$  photonic cavity regime [Fig. 2(b)]

for some reasons as discussed above. Moreover, the number state of the cavity photon reaches much lower values when nonradiative processes are considered [Fig. 2(d)] because of the increased probability of dissipation. The overall quantum yield of nonradiatively broadened systems is worse than those with a natural linewidth.

## B. Plasmonic cavity

In the case of a plasmonic cavity, the radiation leakage rate is much faster since the quality factor (taken in the simulations to be 100 or 1000) is relatively low. Here the dissipative losses of the cavity's material components are ignored, but could be further introduced via additional degrees of freedom [21]. These low- $Q$  low- $V$  constraints impose the relation  $\gamma \gg \Gamma$  for any reasonable physical scenario of the emission. As a result, nonradiative processes do not influence the evolution of the system: The radiative processes become faster (due to small modal volumes), but emitted photons leave the cavity even faster. For example, in silver spherical nanoparticles with a radius bigger than 30 nm, radiation damping predominates internal material losses [13]. The dependence of the two-level system population in the cavity with  $Q = 100$  [Fig. 3(a)] and the cavity photon number state [Fig. 3(b)] on the cavity modal volume at constant quality factor is almost insensitive to the presence of the nonradiative damping. Here the Purcell factor is large owing to the small modal volume of the plasmonic cavity.

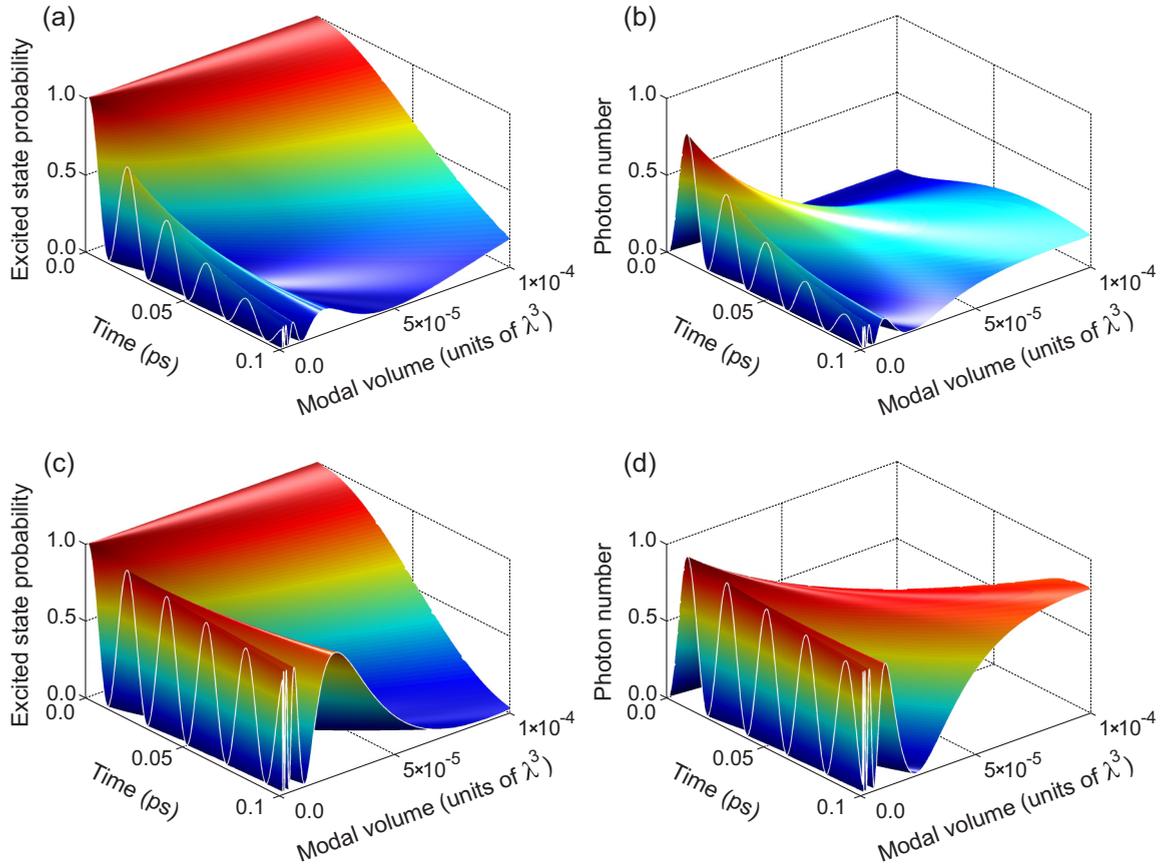


FIG. 3. (Color online) Dependence of the time evolution of an emitter in a plasmonic cavity on a cavity modal volume: (a) and (c) probability of the two-level system being in the excited state and (b) and (d) number state of the cavity plasmon for (a) and (b)  $Q = 100$  and (c) and (d)  $Q = 1000$ . The emitter without and with nonradiative broadening exhibits similar behavior.

In the large modal volume regime, the decay law is almost exponential; however, small deviations are observed owing to the relatively large quality factor of the cavity ( $Q = 100$ ). For smaller  $Q$ , the law has a pure exponential nature.

In order to observe a nontrivial time evolution of the emitter, namely, a strong-coupling regime, very small modal volumes of plasmonic cavities should be achieved. Nevertheless, the required values can be obtained with specifically designed structures [22] or with small nanoparticles. The strong-coupling regime in plasmonic nanostructures has already been observed experimentally [23–25]. As in the case of a photonic cavity, the Purcell factor has no direct physical meaning in the strong-coupling regime. The evolution of the plasmon population numbers, when small modal volumes are considered, also poses nontrivial (nonexponential) behavior. Similar effects can also emerge as the manifestation of the strong near-field feedback [21]. Recently, high quality plasmonic cavities were demonstrated, showing  $Q$  factors of more than 1000 [26]. While the qualitative behavior for both  $Q = 100$  and 1000 cavities is similar and the main features are preserved, such as the number of revivals, much longer characteristic radiative lifetimes and higher revival intensity for  $Q = 1000$  can be observed [cf. Figs. 3(a) and 3(b) and Figs. 3(c) and 3(d)]. Photonic cavities typically have higher  $Q$  factors than plasmonic ones but relatively large volume  $V$  and low damping, so that the main loss mechanism is the radiative

loss. In contrast, plasmonic cavities have a limited value of  $Q$  but small  $V$ , with a strong stochastic (Ohmic) loss channel. The influence of either a photonic or a plasmonic cavity is to make the radiative time comparable to the nonradiative one (i.e., 1 ps) and even faster (as in the case of small-volume plasmonic cavities), which leads to a competition between radiative and nonradiative transitions.

## V. CONCLUSION

The evolution of a quantum system with nonradiative broadening coupled to a leaky cavity has been investigated in different regimes. In particular, the regime of comparable radiative and nonradiative lifetimes, where commonly used perturbative approaches fail, was analytically treated via quantum stochastic differential equations. We have demonstrated that some nonradiative mechanisms, being of particular significance to radiative processes in high- $Q$  photonic cavities, are much less influential in the plasmonic regime. Optical cavities, based on various approaches, e.g., Fabry-Pérot, whispering gallery, and photonic crystal designs, could deliver  $Q$  factors up to  $10^{10}$  (whispering gallery) and modal volumes as small as  $(\lambda/n)^3$  (photonic crystal) and cover all the regimes investigated here. Plasmonic cavities in the localized plasmon resonance regime have  $Q$  factors not exceeding typically 100 (limited by the ratio between real and imaginary material

permittivity of metal components of resonators), while in the polaritonic regime (propagating plasmons) these  $Q$  factors may have higher values [26]. Modal volumes of these cavities could be as small as  $\lambda^3/1000$ , overlapping with the case studied in this paper. The proposed model allows one to describe the Purcell effects in various kinds of cavities in the presence of loss when a normal description via  $Q$  factors fails.

The developed theoretical framework is not limited to the presented examples and represents a general tool for multiple stochastic Hamiltonian evolutions. In particular, the process of quantum-dot blinking (the random transitions between bright

and dark states of an emitter, subject to continuous pumping) is the manifestation of the competition between the radiative and nonradiative processes [27] and could be treated with the help of the developed technique. Higher-order spontaneous processes, such as spontaneous two-photon emission [28], could be also analyzed, including plasmonic cavity-assisted enhancement [29,30].

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