Few-photon SUPER: Quantum emitter inversion via two off-resonant photon modes

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With the realization of controlled quantum systems, exploring excitations beyond the resonant case opens new possibilities. We investigate an extended Jaynes-Cummings model in which two nondegenerate photon modes are coupled off resonantly to a quantum emitter. This allows us to identify few-photon scattering mechanisms that lead to a full inversion of the emitter while transferring off-resonant photons from one mode to another. This behavior is connected to recent measurements of a two-level emitter scattering two off-resonant photons simultaneously. Furthermore, our results can be understood as a quantized analog of the recently developed off-resonant quantum control scheme known as swing-up of quantum emitter (SUPER). Our intuitive formalism gives deeper insight into the interaction of a two-level emitter with off-resonant light modes with the prospect of novel photonic applications.

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I. INTRODUCTION

The two-level emitter and its interaction with light lies at the heart of quantum optics. The most prominent case is when the frequency of the light field is in resonance with the transition frequency of the two-level system, leading to phenomena such as vacuum Rabi oscillations [1–3] and the Mollow triplet [4]. Adding a chirp to the exciting laser pulse leads to adiabatic rapid passage [5,6]. From textbooks we learn that for a two-level emitter that is excited with a single-frequency field, a full inversion can be achieved only in resonance, while a detuned single-frequency pulse cannot lead to a full inversion.

This changes drastically when, instead of a single off-resonant pulse, multicolor pulses that are all off resonant are used [7–15]. Alternatively, a frequency modulation of an off-resonant pulse has similar effects [9,16–18]. An intuitive explanation of the inversion process can be given by the Swing-UP of quantum EmitteR (SUPER) mechanism [9]. The SUPER mechanism has also been demonstrated experimentally [12–15]. In the most widely discussed case, a pair of two red-detuned pulses, i.e., both pulses have an energy below the transition frequency, are employed to excite a two-level system. This prompts the question of energy conversion: In a single-photon picture, neither of the pulses will have enough energy to excite the two-level system, and also their sum or difference does not match the resonance condition.

In this paper, we clarify this question by considering an intuitive model of an extended Jaynes-Cummings model with two photon modes. We find distinct resonance conditions which lead to an inversion of the two-level emitter coupled to two off-resonant, nondegenerate modes. At these resonances, a multiphoton scattering process takes place.

Understanding off-resonant excitation schemes is not only essential for the generation of nonclassical single-photon states for quantum communication [19,20] but also interesting for understanding the fundamentals of the interaction of a two-level emitter with photons [21,22]. The interaction of a single two-level emitter with few photons, either resonant or off resonant, results in a multiphoton scattering, visible in the correlation function [23,24]. The $g^{(2)}$-correlation function is a standard measure for photon-photon correlations that occur in resonance fluorescence [25]. References [22,24] showed that a correlated photon scattering takes place. In the far-off-resonant case, this will lead to the excitation of the two-level emitter in a three-photon process, where two drive photons disappear, while one photon appears at even larger detuning. Here, we connect these multiphoton scatterings to the SUPER scheme.

Following this introduction, we will give a brief summary of the excitation of a two-level emitter either using a single monochromatic pulse, resulting in Rabi oscillations, or using two-color excitation via the SUPER scheme in the semiclassical picture in Sec. II. Next, we introduce the two-mode Jaynes-Cummings model in Sec. III. Within the two-mode Jaynes-Cummings model we discuss the dynamics of the participating states and the maximally achievable occupation of the two-level emitter to identify a quantum analog of SUPER in Sec. IV, before discussing and summarizing our findings in Sec. V.

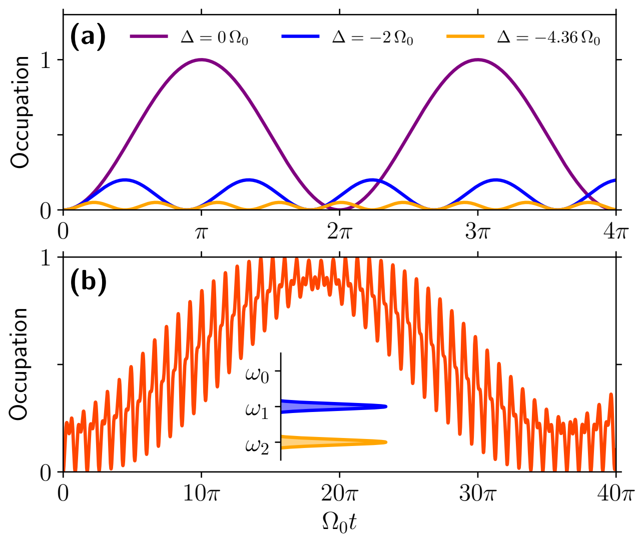

FIG. 1. Rabi and SUPER scheme in the semiclassical picture: (a) Rabi oscillations in the occupation of the excited states as a function of time for the resonant case (purple) and the off-resonant cases with detunings Δ_1 (blue) and Δ_2 (yellow). (b) Swing-up dynamics of the occupation induced by using two fields with constant amplitude Ω_0 and different detunings $\Delta_1 = -2\,\Omega_0$ and $\Delta_2 = -4.26\,\Omega_0$. The inset indicates the central frequencies of the two exciting lasers.

II. RABI AND SUPER IN THE SEMICLASSICAL PICTURE

We consider a two-level emitter consisting of the ground state $|g\rangle$ and the excited state $|x\rangle$ with energy difference $E_x - E_g = \hbar\omega_0$ coupled to the electric field in dipole approximation, with resonant or close-to-resonant excitation, such that the rotating wave approximation is applicable.

In the context of optical excitation schemes of quantum emitters, usually, a semiclassical picture via the standard Hamiltonian is employed, reading

$$H = \hbar\omega_0\,|x\rangle\,\langle x| - \frac{\hbar}{2}[\Omega_{\text{laser}}(t)\,|x\rangle\,\langle g| + \text{H.c.}]. \quad (1)$$

In the single-field case, the coupling to the light field is given by

$$\Omega_{\text{laser}}^{\text{single}}(t) = \Omega(t)e^{-i\omega_L t}. \quad (2)$$

The electric field E is contained in the Rabi frequency $\Omega = 2dE/\hbar$ via its coupling to the dipole moment d. Within the rotating wave approximation we use a notation splitting the field into the complex-valued part oscillating with frequency ω_L and the real-valued envelope $E(t) \sim \Omega(t)$. The detuning is then defined as the difference of the laser frequency and the transition frequency of the two-level system, $\Delta = \omega_L - \omega_0$. A cw excitation switched on at $t = 0$ leads to Rabi oscillations in the occupation of the excited state, as displayed in Fig. 1(a).

A two-color light field contains two frequencies ω_i, such that the coupling to the laser reads

$$\Omega_{\text{laser}}^{\text{two-color}}(t) = \Omega_1(t)e^{-i\omega_1 t} + \Omega_2(t)e^{-i\omega_2 t}. \quad (3)$$

Besides the resonant case, the two-level system can be excited with an off-resonant pulse pair, i.e., $\omega_1, \omega_2 \neq \omega_0$ or, in terms of detunings, $\Delta_1, \Delta_2 \neq 0$. In Ref. [9] we numerically found that an excitation with a Gaussian pulse pair leads to a complete inversion if the detunings fulfill the resonance condition

$$\Delta_2 = \Delta_1 + \sqrt{\Delta_1^2 + \left(\Omega_1^{\max}\right)^2}, \quad (4)$$

with $\Omega_1^{\max}$ being the maximal pulse strength. Employing a semiclassical dressed-state picture gives further insight into the resonance condition [26].

For the case of two off-resonant fields with the same constant amplitude Ω_0 switched on at $t = 0$, shown in Fig. 1(b), we find complete inversion for the condition

$$\sqrt{\Omega_0^2 + \Delta_2^2} = 2\sqrt{\Omega_0^2 + \Delta_1^2} \quad \text{or} \quad \Omega_{R,2} = 2\Omega_{R,1}, \quad (5)$$

with $\Omega_{R,i}$ being the respective Rabi frequency. The corresponding occupation dynamics shows a behavior that is typical for SUPER [9]. Note that here, both detunings are negative, i.e., $\omega_{1,2} < \omega_0$. The same dynamics occur for both detunings being positive, $\omega_{1,2} > \omega_0$. We remark that the two resonant conditions (4) and (5) are for two different excitation conditions, namely, pulsed excitation, where the instantaneous Rabi frequency changes along the pulse, and two constant fields switched on instantaneously with the same amplitude, respectively.

III. TWO-MODE JAYNES-CUMMINGS MODEL

To describe the two-color excitation in a fully quantum mechanical picture, we describe the light field in terms of photon creation ($a_i^\dagger$) and annihilation (a_i) operators, considering a two-mode Jaynes-Cummings Hamiltonian. In this system, the two-level system is simultaneously coupled to two photon modes ω_i. We focus on the case of both modes being off resonant with the two-level system,

$$\omega_0 \neq \omega_1 \neq \omega_2 \neq \omega_0. \quad (6)$$

Such a configuration was realized using atoms and two orthogonal cavities [27]. There, the interaction between the two-level system and the two modes was controlled by stepwise changing of the detuning [27,28].

Another realization of the two-mode Jaynes-Cummings model could be via superconducting circuits [29–31], where the frequency of the modes can be adjusted by changing the capacitance or inductance of the resonators.

The corresponding Hamiltonian reads

$$\begin{aligned} H &= \hbar\omega_0|x\rangle\langle x| + \sum_{i=1}^{2}\hbar[\omega_i a_i^\dagger a_i + \Lambda_i(|g\rangle\langle x|a_i^\dagger + |x\rangle\langle g|a_i)] \\ &= \hbar\omega_0\sigma_+\sigma_- + \sum_{i=1}^{2}\hbar[\omega_i a_i^\dagger a_i + \Lambda_i(\sigma_- a_i^\dagger + \sigma_+ a_i)]. \end{aligned} \quad (7)$$

The coupling strength between the two-level emitter and mode i is given by the parameter Λ_i. These coupling strengths translate into the Rabi frequencies, which depend on the total number of excitations in the coupled emitter-cavity system. While in the first row we use the notation commonly used by the quantum dot community, in the second row we have rewritten the equation using Pauli matrices, as commonly used by the atomic physics community.

We emphasize that our focus is different to the case of two resonant modes, i.e., $\omega_1 = \omega_2 = \omega_0$ or on two off-resonant but degenerate modes with $\omega_1 = \omega_2 \neq \omega_0$. These cases were studied previously [31–36]. For quantum dots, the case of two resonant modes with different polarizations has been realized [37]. We note that our model is distinct from the two-photon Jaynes-Cummings model with bimodal coupling, where the deexcitation leads to the direct emission of two photons ($\sim\sigma_- a_1^\dagger a_2^\dagger$) [38,39], which has been analyzed in the context of squeezing [40].

We compose the basis $|\nu, n_1, n_2\rangle$ as product states of the electronic states $|\nu\rangle \in \{|g\rangle, |x\rangle\}$ and the Fock states $|n_i\rangle$ in mode i. The interaction between the two-level system and the photon modes is allowed by either absorption of a photon coupling the states, meaning

$$|g, n_1, n_2\rangle \leftrightarrow |x, n_1 - 1, n_2\rangle, \tag{8a}$$

$$|g, n_1, n_2\rangle \leftrightarrow |x, n_1, n_2 - 1\rangle, \tag{8b}$$

or emission of a photon if one reads the equations from right to left. We emphasize that it does not allow for a direct photon exchange between the two modes, but all interactions are mediated via the two-level system.

IV. DYNAMICS AND RESONANCE CONDITIONS

To study the conditions under which few-photon scattering might lead to an excitation of the emitter, we analyze the dynamics of the two-level system coupled by a given number of initially present photons and search for the maximal occupation of the excited state. To this end, we set up the equations of motion with the initial state of the two-level system being in the ground state and a given number of photons as $|g, n_1^{(0)}, n_2^{(0)}\rangle$. Defining the operator for the total number of excitations as $\hat{N} = \sum_j a_j^\dagger a_j + |x\rangle\langle x|$, it is easily seen that $N^{\rm tot} = \langle \hat{N}\rangle$ is conserved in the Jaynes-Cummings model. Our initial conditions thus restrict the dynamics to eigenstates of $\hat{N}$ with total excitation number $N^{\rm tot} = n_1^{(0)} + n_2^{(0)}$, which is a manifold with finite dimension.

Diagonalization of the two-mode Jaynes-Cummings Hamiltonian for given initial conditions provides a simple solution of the time-dependent Schrödinger equation via the time-evolution operator. From the dynamics, we obtain the maximally achievable occupation of the excited state as displayed in the results. Additionally, we checked our results by direct numerical integration via the free software package QUTIP [41].

For a single photon in either of the two modes, i.e., $|i\rangle = |g, 1, 0\rangle$ or $|i\rangle = |g, 0, 1\rangle$, according to the Rabi condition for a two-level system, the occupation of the excited state remains small for $\Delta/\Lambda \gg 1$. Here, we are interested in large occupations for detunings in this parameter regime, where for the initial condition of a single photon no significant excited-state occupation is reached.

A. Two-photon scattering

The minimal number of photons required to achieve non-negligible excitation of the two-level emitter is two photons in the mode with the smaller absolute value of detuning. This corresponds to the initial state

$$|i\rangle = |g, 2, 0\rangle. \tag{9}$$

We scan the maximally achievable occupation of the excited state in the two-level system as a function of the two detunings Δ_1 and Δ_2, as shown in Fig. 2(a), while setting $\Lambda = \Lambda_1 = \Lambda_2$. A cut for $\Delta_2 = 10\Lambda$ is shown in Fig. 2(b).

If the emitter is coupled to only a single mode, the maximally achievable occupation of the excited state will show a Lorentzian-shaped decrease for increasing detuning Δ_1. In the color map, this is seen when going from the bottom to top, where the color fades from yellow (1) to blue (0).

Most strikingly, a sharp maximum of the excited-state occupation is found on top of the Lorentzian decrease. This sharp maximum reaches almost unity. In the color map, this is observed as a clear line with a slope of about $\Delta_1 \approx \frac{1}{2}\Delta_2$. This line matches the resonance condition where the SUPER mechanism enables the inversion of the two-level system. In the cut in Fig. 2(b) we see this sharp resonance appearing at $\Delta_1/\Lambda = 4.62$.

To understand what is happening at the resonance line, we look at the exemplary dynamics of different quantities in Figs. 2(c)–2(e). Figure 2(c) shows the dynamics of the occupation of the excited state $\langle |x\rangle\langle x|\rangle$ alongside the dynamics of the photon mode occupation $n_i = \langle a_i^\dagger a_i\rangle$ in Fig. 2(d). The excited-state occupation exhibits a fast oscillation on top of a slower oscillation, and eventually, the system reaches inversion. This behavior reminds us of the semiclassical SUPER case [compare with Fig. 1(b)]. Surprisingly, the excitation is achieved just by coupling the system to the second mode, although the latter holds no initial photon.

From the dynamics of the photon numbers it becomes clear that the two photons in mode 1 both vanish, i.e., $|n_1 = 2\rangle \rightarrow |n_1 = 0\rangle$. Simultaneously with the excited-state occupation, photon mode 2 becomes occupied and holds a single photon with $|n_2 = 1\rangle$ at its maximum. We also see that the fast oscillation mostly takes place only in the photon mode $|n_1\rangle$.

According to the coupling rules in the Jaynes-Cummings model, the transition from the initial state $|i\rangle = |g, 2, 0\rangle$ to the final state $|f\rangle = |x, 0, 1\rangle$ does not take place directly, but via

$$|i\rangle = |g, 2, 0\rangle \leftrightarrow |x, 1, 0\rangle \leftrightarrow |g, 1, 1\rangle \leftrightarrow |x, 0, 1\rangle = |f\rangle. \tag{10}$$

For the conditions required to achieve maximal excited-state occupation, the initial state $|i\rangle$ and final state $|f\rangle$ are almost degenerate, leading to a resonant transfer, whereas the other states are mostly unoccupied during the dynamics. This is confirmed in Fig. 2(e), where the dynamics of the occupation of these states is shown. Indeed, the occupations of the intermediate states remains small. On top of the overall dynamics, a fast oscillation is observed as a signature of the SUPER mechanism.

In the SUPER scheme in the semiclassical picture [9,26] the resonance condition [see Eq. (4)] was derived as

$$\Delta_2 = \Delta_1 + \sqrt{\Delta_1^2 + \left(\Omega_1^{\rm max}\right)^2} \xrightarrow{\Delta_1 \gg \Lambda_i} 2\Delta_1. \tag{11}$$

A similar resonance condition has been predicted for the two-photon scattering in resonance fluorescence with $\Delta_2 = 2\Delta_1$ in the limit of large detunings [22,23].

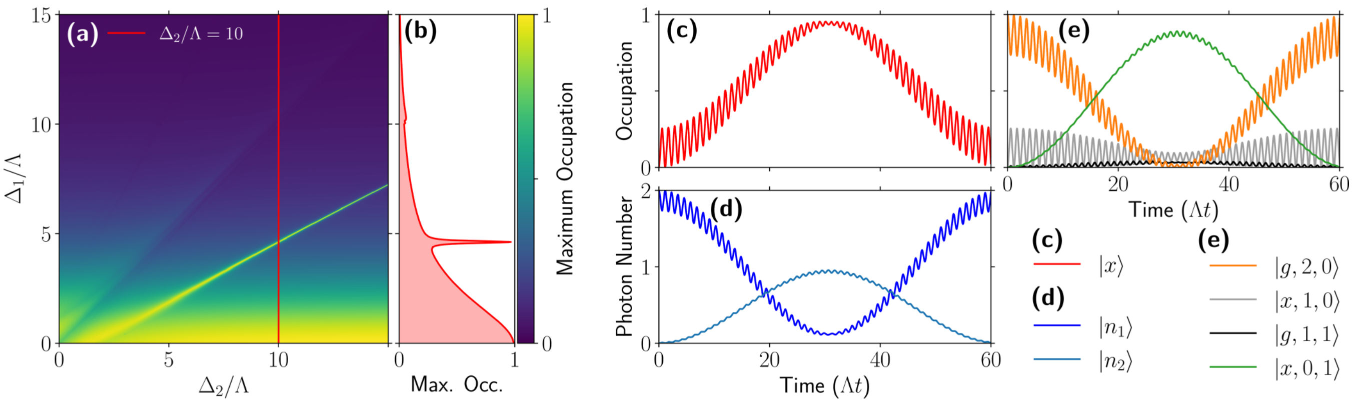

FIG. 2. Dynamics of a two-level system in the two-mode Jaynes Cummings model: (a) Color map of the maximum occupation as a function of the detunings Δ_1 and Δ_2. The initial state is $|g, 2, 0\rangle$. (b) A cut for $\Delta_2 = 10\Lambda$, as indicated by the red line. Dynamics of the (c) excited-state occupation and (d) occupation in the two photonic modes. (e) Dynamics of the occupations of selected states participating in the process.

An adiabatic elimination can be performed for the equations of motion of the two-mode Jaynes-Cummings model in order to derive an approximate resonance condition for large detunings,

$$\Delta_2 \overset{\Delta_2 \gg \Lambda}{\approx} 2\Delta_1 + \frac{4\Lambda^2}{\Delta_1}. \tag{12}$$

The details of the calculation are given in Appendix A. This resonance condition agrees well with our results in Fig. 2(a). In addition, similar results are found in both the semiclassical and quantum pictures. This corroborates our assumption that the Jaynes-Cummings model with two off resonantly coupled modes is indeed the adequate quantum mechanical analog of the semiclassical SUPER scheme.

It is interesting that these findings are connected to the idea of two-photon scattering, which was recently observed in experiments [21,22] in resonance fluorescence.

We note that in the area close to the diagonal line, where both modes are degenerate with $\Delta_1 = \Delta_2$, there is a bump observable in the cut as well. The degenerate case has been discussed in the literature [33–36], as it shows different physics compared to the coupling to a single mode. The bump appears in the vicinity of the diagonal line including several small features, which we attribute to multiphoton processes. Since here we are interested in the off-resonant case where both modes are distinct from each other and detuned to the two-level quantum emitter, we leave the detailed discussion of these processes to future research.

B. *N*-photon scattering

Next, we consider the case of an initial state with five photons in one mode,

$$|i\rangle = |g, 5, 0\rangle. \tag{13}$$

The maximally achievable occupation of the excited state as a function of both detunings Δ_i is shown in Fig. 3. On top of the single-mode Rabi background, we now find four distinct lines of maximal excitation probability. For example, for a cut at $\Delta_2/\Lambda = 7$ we find resonances at $\Delta_1/\Lambda \approx 2.400$, 4.039, 4.839, and 5.298. Furthermore, we observe that the width of the resonance peaks decreases with increasing detuning.

Inspecting the lines, we find that they belong to multiphoton scattering processes effectively connecting the initial and final states,

$$|g, 5, 0\rangle \leftrightarrow |x, 5 - N, N - 1\rangle. \tag{14}$$

The line with the lowest slope belongs to the two-photon scattering processes $|g, 5, 0\rangle \leftrightarrow |x, 3, 1\rangle$ discussed previously. The next line then features a three-photon scattering process with $|g, 5, 0\rangle \leftrightarrow |x, 2, 2\rangle$. In this case, the five photons in the initial mode are distributed such that one photon excites the two-level system, two photons are scattered into the mode with higher detuning, and two photons remain in the initial mode with lower detuning. In the curve with highest slope, we have a five-photon scattering process $|g, 5, 0\rangle \leftrightarrow |x, 0, 4\rangle$. While one photon excites the two-level system, the other four photons are scattered into the higher detuned mode.

The time required to achieve inversion increases for the higher-order processes as more photons N are scattered from one mode to the other. This translates to a decreased effective

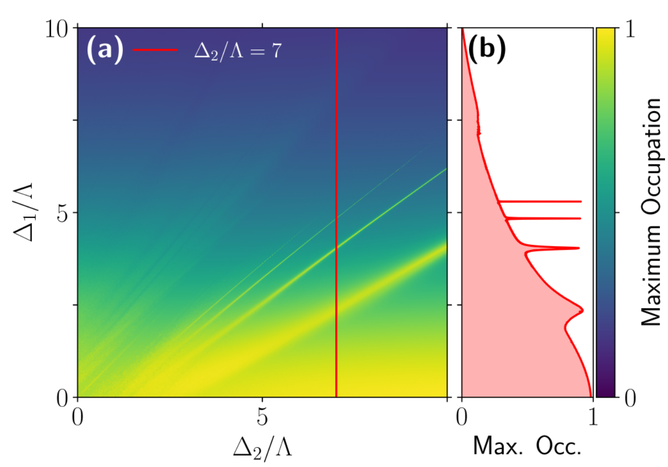

FIG. 3. Resonances for the multiphoton scattering: (a) Color map of the maximum occupation of the excited state as a function of detunings Δ_1 and Δ_2 with (b) a cut at $\Delta_2/\Lambda = 7$. The initial state is $|g, 5, 0\rangle$.

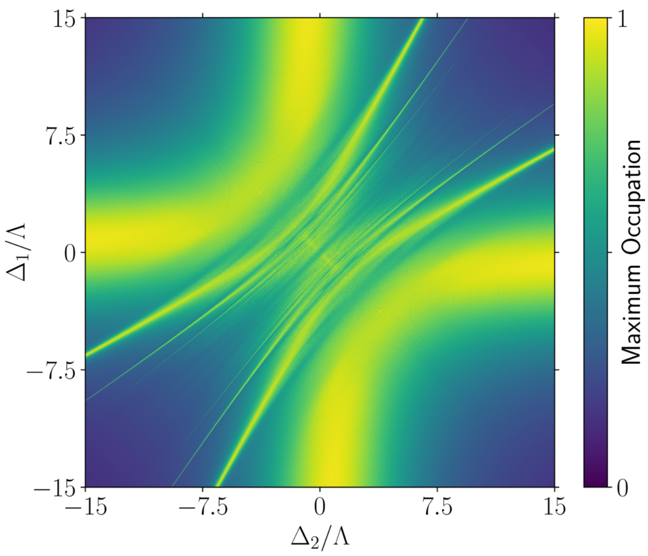

FIG. 4. Resonances for symmetric N-photon scattering. Color map of the maximum occupation of the excited state as a function of detunings Δ_1 and Δ_2 for the initial state $|i\rangle = |g, 5, 5\rangle$.

Rabi frequency and is also reflected in the linewidths that become narrower for higher N. We assume that in an experiment that also includes dissipation, these processes are much less likely to occur and most likely cannot be stimulated.

C. Scattering from both modes

In this section, we discuss the full scan of the maximally achievable occupation of the two-level emitter that is performed for the initial state

$$|i\rangle = |g, 5, 5\rangle. \tag{15}$$

In addition to both modes having negative detuning, we scan the region of positive detuning as well as the regions with one mode that is positive and one mode that is negative. The results are shown in Fig. 4.

Several symmetries are apparent, as expected from our model: If we switch the sign of both detunings, the picture is symmetric. At the origin $\Delta_1 = \Delta_2 = 0$, we do not achieve inversion, as expected from the single-mode case. This is also found in Figs. 2 and 3. Here, the photon modes can be rewritten into two new modes that are coupled and uncoupled to the two-level system, respectively. Then, full inversion will be reached only if the coupled mode is initially in a pure Fock state with one or more photons. However, in our case the two coupled and uncoupled modes are initially entangled, resulting in a lower maximal occupation depending on the number of initially available photons. We provide a detailed discussion of this in Appendix B.

We also see distinct resonance conditions for both detunings being either negative or positive. Although we now also get resonances for $\Delta_1 > \Delta_2$, in all cases photons are scattered from the lower detuned mode to the higher detuned mode. Note that in the case with nonequal coupling constants $\Lambda_1 \neq \Lambda_2$ we get a similar behavior, as shown in Appendix C.

Let us now turn to the case with $\Delta_1 > 0$ and $\Delta_2 < 0$ ($\Delta_1 < 0$ and $\Delta_2 > 0$). This case was studied in the atomic community [7,8]. In the quantum dot community it was named dichromatic excitation [10,11] and studied for single-photon generation. It can be understood as a special case of the SUPER scheme [26].

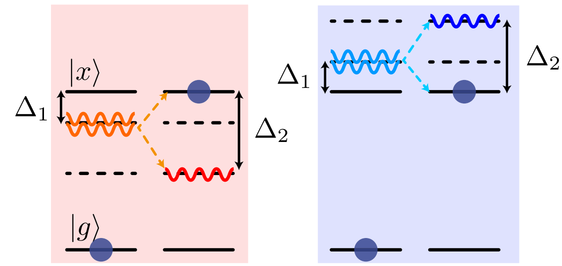

FIG. 5. Sketch of the photon-photon scattering: Illustration of a two-level system that interacts with two photon modes with detunings $\Delta_{1,2}$, either red or blue detuned to the transition of the two-level system. Initially, one mode holds two photons, which results in an occupation of the two-level system and a photon in the other mode.

For the detunings with opposite signs, we find a broad area where the excited-state occupation reaches close-to-unity values. From the dichromatic SUPER case [26], the predicted resonance condition will be

$$\Delta_2 = |\Delta_1| - \Omega^{\text{Rabi}}, \tag{16}$$

with Δ_1 belonging to the initially occupied mode and Ω^{Rabi} being the generalized Rabi frequency in the classical picture. Accordingly, the second detuning Δ_2 is rather small, in agreement with the broad area lying close to zero detuning.

V. DISCUSSION AND CONCLUSIONS

In summary, we found a scattering mechanism in the two-mode Jaynes Cummings model, where the electronic system is promoted from its ground state to its excited state while off-resonant photons are transferred from one mode to another according to

$$|g, n_1, n_2\rangle \leftrightarrow |x, n_1 - N, n_2 + (N - 1)\rangle. \tag{17}$$

We found that N, with $N \geqslant 2$, photons scatter in such a way that one photon excites the two-level emitter while the other $N - 1$ photons scatter into the second off-resonant mode. This second off-resonant mode must have at least twice the detuning of the lower detuned mode.

Our findings relate to different results in quantum optics: They are in line with the observations of multiphoton scattering processes in resonance fluorescence, predicted theoretically [23] and recently observed experimentally [22]. Going beyond the two-photon scattering, we furthermore discussed that scattering processes including three or more photons are also, in principle, possible but harder to realize in experiment.

The dynamics and resonance conditions are also in line with the SUPER scheme, which was found for laser pulses described by classical light fields [9,12]. In the SUPER scheme, full inversion became possible only when two laser pulses with different detunings were applied to the two-level system. This corresponds to stimulating the two-photon process described in the present paper. Note that in SUPER the excitation is usually performed with finite laser pulses, while in Fig. 1 we considered the case of an excitation with constant amplitude that is switched on instantaneously. For smooth pulses, adiabatic undressing effects [42] may push the occupation to its maximum.

We depict the two-photon scattering process in Fig. 5. In the red-detuned case, the scattering leads to the creation of photons with lower energy, while the photon scattering for blue-detuned photons leads to a rise in photon energy. The process also reminds one of Auger scattering, which in the solid-state community is known to occur only between electrons, but recently, in quantum dots the radiative Auger process, in which two electrons scatter, accompanied by the emission of a photon, was also demonstrated [43,44]. Our analysis provides a deeper understanding of the SUPER scheme at the fully quantized level. The results show that a SUPER-type inversion of a quantum emitter not only is possible at high laser intensities where the semiclassical description applies but can, in principle, already be achieved with as few as two off-resonant photons provided a second off-resonant mode is available [45].

The quantum emitter inversion enables photon scattering between off-resonant modes that might lead to new applications in quantum photonics like frequency conversion or photon addition and subtraction. In order to describe these processes in a realistic system, one needs to account for dissipative effects. To make the effects observable the coupling strengths of both modes have to exceed the dissipation rates. For example, in a quantum dot system, the electron-phonon interaction is a prominent source of dissipation [46] and also affects the SUPER scheme [47] even for two red-detuned lasers. The dissipative effect will lead to asymmetry between positive and negative detunings. Additionally, one should account for cavity losses. Therefore, we assume that in an experiment photon scattering with $N \geqslant 3$ will be extremely difficult to observe. In the case of superconducting circuits, the coupling between the bosonic modes can also become important, introducing new dephasing channels [29]. To simulate experiments on such systems, dissipative effects should be included, which we leave for future work. We are confident that with advancements in solid-state cavities [48,49] the presented effects will be observable in solid-state quantum emitters.

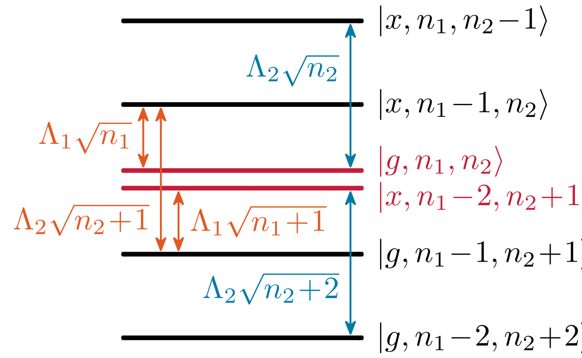

FIG. 6. Scheme of the most relevant states: Display of the most relevant states for coupling between the initial state $|i\rangle = |g, n_1, n_2\rangle$ and $|f\rangle = |x, n_1 - 2, n_2 + 1\rangle$.

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APPENDIX A: REDUCTION TO A TWO-LEVEL SYSTEM

As discussed in the main text, at the discussed resonance conditions the dynamics usually effectively takes place between only two states, namely,

$$|i\rangle = |g, n_1, n_2\rangle \leftrightarrow |f\rangle = |x, n_1 - N, n_2 + (N - 1)\rangle. \quad \text{(A1)}$$

For the condition that

$$\Lambda_i\sqrt{n_i} \ll |\Delta_1 - \Delta_2|, \quad \text{(A2)}$$

the system can be reduced to the most relevant states $|1\rangle = |x, n_1, n_2 - 1\rangle$, $|2\rangle = |x, n_1 - 1, n_2\rangle$, $|3\rangle = |g, n_1, n_2\rangle$, $|4\rangle = |x, n_1 - 2, n_2 + 1\rangle$, $|5\rangle = |g, n_1 - 1, n_2 + 1\rangle$, and $|6\rangle = |g, n_1 - 2, n_2 + 2\rangle$, as indicated in Fig. 6. We go into a rotating frame and shift the energy to be zero at the initial state. In this case the Hamiltonian reduces to

$$H = \hbar \begin{pmatrix} \Delta_2 & 0 & \Lambda_2\sqrt{n_2} & 0 & 0 & 0 \\ 0 & \Delta_1 & \Lambda_1\sqrt{n_1} & 0 & \Lambda_2\sqrt{n_2+1} & 0 \\ \Lambda_2\sqrt{n_2} & \Lambda_1\sqrt{n_1} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 2\Delta_1 - \Delta_2 & \Lambda_1\sqrt{n_1-1} & \Lambda_2\sqrt{n_2+2} \\ 0 & \Lambda_2\sqrt{n_2+1} & 0 & \Lambda_1\sqrt{n_1-1} & \Delta_1 - \Delta_2 & 0 \\ 0 & 0 & 0 & \Lambda_2\sqrt{n_2+2} & 0 & 2\Delta_1 - 2\Delta_2 \end{pmatrix}.$$

With these states, the equations of motion for the coefficients of the state $|\psi\rangle = \sum_{i=1}^{6} C_i|i\rangle$ can be set up. From this, an adiabatic elimination can be performed to highlight the coupling between states $|3\rangle = |i\rangle$ and $|4\rangle = |f\rangle$. In adiabatic elimination, the time derivatives of the coefficients C_1, C_2, C_5, and C_6 are set to zero. The resulting equations can be further approximated on the same level as Eq. (A2) and yield expressions for C_1, C_2, C_5, and C_6 as functions of C_3 and C_4, such that closed equations of motion can be derived for the latter. This results in

$$i\frac{d}{dt}C_3(t) = \left(-\frac{\Lambda_2^2 n_2}{\Delta_2} - \frac{\Lambda_1^2 n_1}{\Delta_1}\right)C_3(t) + \frac{\Lambda_1^2\Lambda_2\sqrt{n_1-1}\sqrt{n_1}\sqrt{n_2+1}}{\Delta_1(\Delta_1-\Delta_2)}C_4(t), \tag{A3a}$$

$$i\frac{d}{dt}C_4(t) = \frac{\Lambda_1^2\Lambda_2\sqrt{n_1-1}\sqrt{n_1}\sqrt{n_2+1}}{\Delta_1(\Delta_1-\Delta_2)}C_3(t) + \left[2\Delta_1 - \Delta_2 - \frac{\Lambda_1^2(n_1-1)}{(\Delta_1-\Delta_2)} - \frac{\Lambda_2^2(n_2+2)}{2(\Delta_1-\Delta_2)}\right]C_4(t). \tag{A3b}$$

Now, we have reduced the system to an effective two-level system described by the effective Hamiltonian

$$H^{\mathrm{eff}} = \hbar\begin{pmatrix} E_1 & \frac{\Omega_R^{\mathrm{eff}}}{2} \\ \frac{\Omega_R^{\mathrm{eff}}}{2} & E_2 \end{pmatrix}, \tag{A4}$$

introducing the effective Rabi frequency

$$\Omega_R^{\mathrm{eff}} = \frac{2\Lambda_1^2\Lambda_2\sqrt{n_1-1}\sqrt{n_1}\sqrt{n_2+1}}{\Delta_1(\Delta_1-\Delta_2)} \tag{A5}$$

and the effective energies of the two states including Stark shifts

$$E_1 = -\frac{\Lambda_2^2 n_2}{\Delta_2} - \frac{\Lambda_1^2 n_1}{\Delta_1}, \tag{A6a}$$

$$E_2 = 2\Delta_1 - \Delta_2 + \frac{\Lambda_1^2(n_1-1)}{(\Delta_2-\Delta_1)} + \frac{\Lambda_2^2(n_2+2)}{2(\Delta_2-\Delta_1)}. \tag{A6b}$$

The resonance condition is given for $E_2 - E_1 = 0$, which, assuming $2\Delta_1 - \Delta_2 \approx 0$, is approximated by

$$\Delta_2 = 2\Delta_1 + \frac{\Lambda_1^2}{\Delta_1}(2n_1-1) + \frac{\Lambda_2^2}{\Delta_1}(n_2+1). \tag{A7}$$

In the case discussed in Sec. IV A we have $n_1 = 2$ and $n_2 = 0$ as well as $\Lambda_1 = \Lambda_2 = \Lambda$. Under these circumstances, the resonance condition reads

$$\Delta_2 = 2\Delta_1 + \frac{4\Lambda^2}{\Delta_1}. \tag{A8}$$

We recall that this describes the behavior for detuning differences that are large compared to the coupling strength as given in Eq. (A2).

The same procedure of adiabatic elimination can also be performed for cases with $N \geqslant 3$. However, because these cases are much less likely to be observed, we present only the numerical results obtained in the main text.

APPENDIX B: OCCUPATION AT $\Delta_1 = \Delta_2 = 0$

A special case is given at the origin of the color plots (Figs. 2–4) where both detunings vanish:

$$\Delta_1 = \Delta_2 = 0. \tag{B1}$$

While this case might seem the same as the single-mode Rabi oscillation behavior and an occupation reaching unity is expected, a significantly smaller occupation is found. In the following we elaborate on this counterintuitive finding.

Let us consider a single photon that initially occupies one of the modes, e.g., $|i\rangle = |g, 1, 0\rangle$. Then, the occupation of the excited state indeed reaches only 0.5. This can be understood by transforming the coupling into a symmetric mode $a_+ = \frac{1}{\sqrt{2}}(a_1 + a_2)$ that couples to the emitter and an antisymmetric mode $a_+ = \frac{1}{\sqrt{2}}(a_1 - a_2)$ that does not couple to the emitter. The initial wave function for a single photon transforms as

$$|i\rangle = |g, 1, 0\rangle \;\rightarrow\; \tfrac{1}{\sqrt{2}}|g\rangle|1\rangle_+|0\rangle_- + \tfrac{1}{\sqrt{2}}|g\rangle|0\rangle_+|1\rangle_-. \tag{B2}$$

From this expression, it is apparent that the interaction with the coupled mode leads to a total maximal population of 0.5. With similar arguments it can be shown that for $|i\rangle = |g, 1, 1\rangle$ the maximal population is also 0.5.

In the next example of two photons in one mode, e.g., $|i\rangle = |g, 2, 0\rangle$, the wave function in the symmetric and antisymmetric mode picture becomes

$$|i\rangle = |g, 2, 0\rangle \;\rightarrow\; \tfrac{1}{2}|g\rangle|2\rangle_+|0\rangle_- + \tfrac{1}{\sqrt{2}}|g\rangle|1\rangle_+|1\rangle_- + \tfrac{1}{2}|g\rangle|0\rangle_+|2\rangle_-, \tag{B3}$$

yielding a maximal occupation of the excited state close to 0.75 by incoherently adding the maximum occupation resulting from the first two terms. It is straightforward to extend these considerations to more photons, showing that the maximally achievable occupation depends on the number of initial photons in the coupled mode. Full inversion will be reached only if the coupled mode is initially in a pure Fock state with one or more photons. This condition is not met in Figs. 2–4,

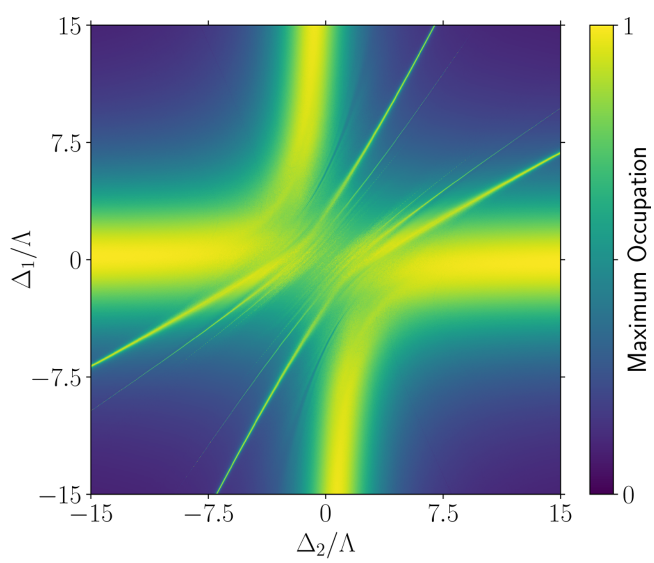

FIG. 7. Resonances for symmetric N-photon scattering. Color map of the maximum occupation as a function of detunings Δ_1 and Δ_2 for the initial state $|i\rangle = |g, 5, 5\rangle$ and unsymmetric coupling constants. We set $\Lambda = \Lambda_1$ and $\Lambda_2 = \frac{1}{2}\Lambda$.

where the coupled mode and the uncoupled mode are initially entangled. Hence, in Figs. 2–4, a maximally achievable occupation below 1 is found at $\Delta_1 = \Delta_2 = 0$.

APPENDIX C: ASYMMETRIC COUPLING CONSTANTS

In the main text we always set the coupling constants of the two modes as equal. This might lead to the question of whether this is a special situation. Thus, we analyze the case of unequal coupling constants in Fig. 7, where we set the coupling constants to $\Lambda_1 = \frac{1}{2}\Lambda_2$. The initial state is set to $|g, 5, 5\rangle$. As seen from the scan, this introduces an asymmetry with respect to the antidiagonal; e.g., the two-photon scattering $|g, 5, 5\rangle \leftrightarrow |x, 3, 6\rangle$ is more pronounced than $|g, 5, 5\rangle \leftrightarrow |x, 6, 3\rangle$. Nonetheless, the overall behavior is very similar, underlining that this is not exceptional behavior just found for equal coupling constants.

[1] I. I. Rabi, Space quantization in a gyrating magnetic field, Phys. Rev. **51**, 652 (1937).

[2] J. P. Reithmaier, G. Sęk, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel, Strong coupling in a single quantum dot–semiconductor microcavity system, Nature (London) **432**, 197 (2004).

[3] G. Khitrova, H. Gibbs, M. Kira, S. W. Koch, and A. Scherer, Vacuum Rabi splitting in semiconductors, Nat. Phys. **2**, 81 (2006).

[4] B. R. Mollow, Power spectrum of light scattered by two-level systems, Phys. Rev. **188**, 1969 (1969).

[5] L. Landau, On the theory of energy transmission in collisions. I, Phys. Z. Sowjetunion **1**, 88 (1932).

[6] C. Zener, Non-adiabatic crossing of energy levels, Proc. R. Soc. Lond. Ser. A **137**, 696 (1932).

[7] S. Guérin, L. P. Yatsenko, and H. R. Jauslin, Dynamical resonances and the topology of the multiphoton adiabatic passage, Phys. Rev. A **63**, 031403(R) (2001).

[8] Á. P. Conde, L. P. Yatsenko, J. Klein, M. Oberst, and T. Halfmann, Experimental demonstration of population inversion driven by retroreflection-induced bichromatic adiabatic passage, Phys. Rev. A **72**, 053808 (2005).

[9] T. K. Bracht, M. Cosacchi, T. Seidelmann, M. Cygorek, A. Vagov, V. M. Axt, T. Heindel, and D. E. Reiter, Swing-up of quantum emitter population using detuned pulses, PRX Quantum **2**, 040354 (2021).

[10] Z. X. Koong, E. Scerri, M. Rambach, M. Cygorek, M. Brotons-Gisbert, R. Picard, Y. Ma, S. I. Park, J. D. Song, E. M. Gauger, and B. D. Gerardot, Coherent dynamics in quantum emitters under dichromatic excitation, Phys. Rev. Lett. **126**, 047403 (2021).

[11] Y.-M. He, H. Wang, C. Wang, M.-C. Chen, X. Ding, J. Qin, Z.-C. Duan, S. Chen, J.-P. Li, R.-Z. Liu *et al.*, Coherently driving a single quantum two-level system with dichromatic laser pulses, Nat. Phys. **15**, 941 (2019).

[12] Y. Karli, F. Kappe, V. Remesh, T. K. Bracht, J. Münzberg, S. Covre da Silva, T. Seidelmann, V. M. Axt, A. Rastelli, D. E. Reiter *et al.*, Super scheme in action: Experimental demonstration of red-detuned excitation of a quantum emitter, Nano Lett. **22**, 6567 (2022).

[13] K. Boos, F. Sbresny, S. K. Kim, M. Kremser, H. Riedl, F. W. Bopp, W. Rauhaus, B. Scaparra, K. D. Jöns, J. J. Finley, K. Müller, and L. Hanschke, Coherent dynamics of the swing-up excitation technique, Adv. Quantum Technol. **7**, 2300359 (2024).

[14] R. Joos, S. Bauer, C. Rupp, S. Kolatschek, W. Fischer, C. Nawrath, P. Vijayan, R Sittig, M. Jetter, S. L. Portalupi, and P. Michler, Coherently and Incoherently Pumped Telecom C-Band Single-Photon Source with High Brightness and Indistinguishability, Nano. Lett. **24**, 8626 (2024).

[15] C. G. Torun, M. Gökçe, T. K. Bracht, M. I. Monsalve, S. Benbouabdellah, Ö. O. Nacitarhan, M. E. Stucki, M. L. Markham, G. Pieplow, T. Pregnolato *et al.*, Super and subpicosecond coherent control of an optical qubit in a tin-vacancy color center in diamond, arXiv:2312.05246.

[16] N. V. Vitanov, L. P. Yatsenko, and K. Bergmann, Population transfer by an amplitude-modulated pulse, Phys. Rev. A **68**, 043401 (2003).

[17] D. Gagnon, F. Fillion-Gourdeau, J. Dumont, C. Lefebvre, and S. MacLean, Suppression of multiphoton resonances in driven quantum systems via pulse shape optimization, Phys. Rev. Lett. **119**, 053203 (2017).

[18] Z.-C. Shi, Y.-H. Chen, W. Qin, Y. Xia, X. X. Yi, S.-B. Zheng, and F. Nori, Two-level systems with periodic *n*-step driving fields: Exact dynamics and quantum state manipulations, Phys. Rev. A **104**, 053101 (2021).

[19] P. Senellart, G. Solomon, and A. White, High-performance semiconductor quantum-dot single-photon sources, Nat. Nanotechnol. **12**, 1026 (2017).

[20] D. A. Vajner, L. Rickert, T. Gao, K. Kaymazlar, and T. Heindel, Quantum communication using semiconductor quantum dots, Adv. Quantum Technol. **5**, 2100116 (2022).

[21] H. Le Jeannic, A. Tiranov, J. Carolan, T. Ramos, Y. Wang, M. H. Appel, S. Scholz, A. D. Wieck, A. Ludwig, N. Rotenberg *et al.*, Dynamical photon–photon interaction mediated by a quantum emitter, Nat. Phys. **18**, 1191 (2022).

[22] L. Masters, X.-X. Hu, M. Cordier, G. Maron, L. Pache, A. Rauschenbeutel, M. Schemmer, and J. Volz, On the simultaneous scattering of two photons by a single two-level atom, Nat. Photon. **17**, 972 (2023).

[23] J. Dalibard and S. Reynaud, Correlation signals in resonance fluorescence: Interpretation via photon scattering amplitudes, J. Phys. France **44**, 1337 (1983).

[24] A. Aspect, G. Roger, S. Reynaud, J. Dalibard, and C. Cohen-Tannoudji, Time correlations between the two sidebands of the resonance fluorescence triplet, Phys. Rev. Lett. **45**, 617 (1980).

[25] H. Kimble and L. Mandel, Resonance fluorescence with excitation of finite bandwidth, Phys. Rev. A **15**, 689 (1977).

[26] T. K. Bracht, T. Seidelmann, Y. Karli, F. Kappe, V. Remesh, G. Weihs, V. M. Axt, and D. E. Reiter, Dressed-state analysis of two-color excitation schemes, Phys. Rev. B **107**, 035425 (2023).

[27] A. Rauschenbeutel, P. Bertet, S. Osnaghi, G. Nogues, M. Brune, J. M. Raimond, and S. Haroche, Controlled entanglement of two field modes in a cavity quantum electrodynamics experiment, Phys. Rev. A **64**, 050301(R) (2001).

[28] D. Gonţa, T. Radtke, and S. Fritzsche, Generation of two-dimensional cluster states by using high-finesse bimodal cavities, Phys. Rev. A **79**, 062319 (2009).
[29] M. Mariantoni, F. Deppe, A. Marx, R. Gross, F. K. Wilhelm, and E. Solano, Two-resonator circuit quantum electrodynamics: A superconducting quantum switch, Phys. Rev. B **78**, 104508 (2008).
[30] M. Neeley, M. Ansmann, R. C. Bialczak, M. Hofheinz, E. Lucero, A. D. O'Connell, D. Sank, H. Wang, J. Wenner, A. N. Cleland *et al.*, Emulation of a quantum spin with a superconducting phase qudit, Science **325**, 722 (2009).
[31] P. Rosario, A. C. Santos, C. J. Villas-Boas, and R. Bachelard, Collateral coupling between superconducting resonators: Fast high-fidelity generation of qudit-qudit entanglement, Phys. Rev. Appl. **20**, 034036 (2023).
[32] J. Larson and T. Mavrogordatos, *The Jaynes–Cummings Model and Its Descendants* (IOP Publishing, Bristol, UK, 2021), pp. 2053–2563.
[33] G. J. Papadopoulos, Energetics of radiation and a two-level atom in an ideal resonant cavity, Phys. Rev. A **37**, 2482 (1988).
[34] G. Benivegna and A. Messina, New quantum effects in the dynamics of a two-mode field coupled to a two-level atom, J. Mod. Opt. **41**, 907 (1994).
[35] G. Benivegna, Quantum effects in the transient dynamics of a many mode Jaynes-Cummings model, J. Mod. Opt. **43**, 1589 (1996).
[36] Y. Xie, Photon conversion among multichromatic waves in two-level atom systems, J. Mod. Opt. **42**, 2239 (1995).
[37] F. Meier and D. D. Awschalom, Spin-photon dynamics of quantum dots in two-mode cavities, Phys. Rev. B **70**, 205329 (2004).
[38] S.-C. Gou, Characteristic oscillations of phase properties for pair coherent states in the two-mode Jaynes-Cummings-model dynamics, Phys. Rev. A **48**, 3233 (1993).
[39] S.-C. Gou, Quantum behavior of a two-level atom interacting with two modes of light in a cavity, Phys. Rev. A **40**, 5116 (1989).
[40] L. Duan, S. He, D. Braak, and Q.-H. Chen, Solution of the two-mode quantum Rabi model using extended squeezed states, Europhys. Lett. **112**, 34003 (2015).
[41] J. R. Johansson, P. D. Nation, and F. Nori, Qutip: An open-source python framework for the dynamics of open quantum systems, Comput. Phys. Commun. **183**, 1760 (2012).
[42] A. M. Barth, A. Vagov, and V. M. Axt, Path-integral description of combined Hamiltonian and non-Hamiltonian dynamics in quantum dissipative systems, Phys. Rev. B **94**, 125439 (2016).
[43] M. C. Löbl, C. Spinnler, A. Javadi, L. Zhai, G. N. Nguyen, J. Ritzmann, L. Midolo, P. Lodahl, A. D. Wieck, A. Ludwig *et al.*, Radiative Auger process in the single-photon limit, Nat. Nanotechnol. **15**, 558 (2020).
[44] J.-Y. Yan, C. Chen, X.-D. Zhang, Y.-T. Wang, H.-G. Babin, A. D. Wieck, A. Ludwig, Y. Meng, X. Hu, H. Duan *et al.*, Coherent control of a high-orbital hole in a semiconductor quantum dot, Nat. Nanotechnol. **18**, 1139 (2023).
[45] Recently, a similar work was published [50].
[46] D. E. Reiter, T. Kuhn, and V. M. Axt, Distinctive characteristics of carrier-phonon interactions in optically driven semiconductor quantum dots, Adv. Phys.: X **4**, 1655478 (2019).
[47] T. K. Bracht, T. Seidelmann, T. Kuhn, V. M. Axt, and D. E. Reiter, Phonon wave packet emission during state preparation of a semiconductor quantum dot using different schemes, Phys. Status Solidi B **259**, 2100649 (2022).
[48] C. Schneider, P. Gold, S. Reitzenstein, S. Hoefling, and M. Kamp, Quantum dot micropillar cavities with quality factors exceeding 250,000, Appl. Phys. B **122**, 19 (2016).
[49] P. Androvitsaneas, R. N. Clark, M. Jordan, M. Alvarez Perez, T. Peach, S. Thomas, S. Shabbir, A. D. Sobiesierski, A. Trapalis, I. A. Farrer *et al.*, Direct-write projection lithography of quantum dot micropillar single photon sources, Appl. Phys. Lett. **123**, 094001 (2023).
[50] L. Vannucci and N. Gregersen, Super excitation of quantum emitters is a multi-photon process, Opt. Express **32**, 35381 (2024).