Microscopic theory of cavity-enhanced single-photon emission from optical two-photon Raman processes

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We consider cavity-enhanced single-photon generation from stimulated two-photon Raman processes in three-level systems. We compare four fundamental system configurations, one Λ-, one V- and two ladder (Ξ-) configurations. These can be realized as subsystems of a single quantum dot or of quantum-dot molecules. For a new microscopic understanding of the Raman process, we analyze the Heisenberg equation of motion applying the cluster-expansion scheme. Within this formalism an exact and rigorous definition of a cavity-enhanced Raman photon via its corresponding Raman correlation is possible. This definition for example enables us to systematically investigate the on-demand potential of Raman-transition-based single-photon sources. The four system arrangements can be divided into two subclasses, A-type and V-type, which exhibit strongly different Raman-emission characteristics and Raman-emission probabilities. Moreover, our approach reveals whether the Raman path generates a single photon or just induces destructive quantum interference with other excitation paths. Based on our findings and as a first application, we gain a more detailed understanding of experimental data from the literature. Our analysis and results are also transferable to the case of atomic three-level-resonator systems, and can be extended to more complicated multi-level schemes.

I. INTRODUCTION

Light emission from laser-enhanced two-photon processes, also referred to as optical two-photon Raman processes, has been extensively studied in atomic three-level systems1–4, including quantum-network5 and single-photon storage applications6. In addition to that, coherent quantum-interference phenomena mediated by related two-photon excitations in generic three-level systems like electromagnetically induced transparency (EIT) have been thoroughly investigated7–10. Nowadays, optical Raman processes again attract attention in the context of cavity-enhanced single-photon generation from quantum-dot (QD) systems in different electronic configurations11–15. Moreover, an integrated nanophotonics platform16 and single microwave photon detectors17,18 have recently been realized operating on Raman processes. The setup to be considered is sketched in Fig. 1. Here, one photon is driven by an external control laser and the other photon is emitted spontaneously into a cavity mode. This mechanism allows for direct all-optical tunability of emission time, frequency, linewidth, spectral shape and polarization state of the emitted single photon15,19–22.

Despite all this previous work on the optical properties of the emitted light from cavity-enhanced optical Raman processes, thus far on-demand emission probability of a single photon has not been realized or studied in detail yet, neither from atoms4,23 nor from a QD19. For quantum-communication networks a minimum-extraction efficiency of 67% is needed19,21,25. Moreover, emission spectra measured in recent experiments raised an open question concerning the contradictory behaviour of peak-intensity and broadening for the resonant Raman process, as discussed in the Supplementary Information of Ref. 14. For that reason, a more detailed microscopic understanding of the Raman emission, revealing full information on the excitation processes and paths contributing to or diminishing the emission, is needed.

In this article, we present a comprehensive microscopic analysis of cavity-enhanced single-photon emission from two-photon Raman processes. Here, we focus on three-level systems in all possible configurations where the cavity enhances the emission of one photon of the respective two-photon Raman path. This yields one Λ-, one V- and two ladder (Ξ-) configurations (see Fig. 1). We assume that cavity and control laser have orthogonal polarizations. These three-level systems – and superpositions of them14,15 – appear as fundamental building blocks of Raman-emission-based single-photon sources realized by single QDs or QD molecules embedded in microresonators. The four system configurations can be divided into two subclasses where the cavity either couples to the first state 1 (V-type) or to the final state 3 (Λ-type). We will use the nomenclature \textit{T-type system} \((T=\Lambda\text{ or }V)\) whenever talking about the whole system \textit{class}, and the nomenclature \textit{configuration} whenever addressing a chosen \textit{representative} \((\Xi_1,\Xi_2,\Lambda\text{ or }V)\) of the respective class. We use the Heisenberg equation of motion together with the cluster-expansion method26 to study the single-photon emission dynamics in terms of fundamental physical processes present in the four QD-cavity systems.

This approach leads to new microscopic insight into the Raman process which, in general, cannot be extracted directly from the observation of optical spectra. As an example, Fig. 2 shows the time development of the cavity-
emission spectrum for a Λ-configuration with a resonant control pulse and a resonant cavity (see also inset of the figure). On the time scale of the coherent emission (dashed line), the spectrum exhibits a pronounced dip in the center of the emission peak which is the fingerprint of destructive quantum interference of different excitation paths.\(^{27}\) This effect is well known from three-band or three-level excitations of zero- and higher-dimensional semiconductor structures.\(^{28–31}\) In particular, EIT\(^{32,33}\) is a prominent example of quantum interference due to a two-photon excitation leading to a dip or even a spectral gap in the absorption spectrum.\(^{34–36}\) In our case, for larger measurement durations (solid line) the interference feature is not visible anymore. Here, immediately the question arises if the observed emission can be identified as a Raman photon or not. Another question would be which process causes the destructive interference feature observed in the spectrum. Since neither spectral nor polarization filtering will decompose the spectrum in the resonant case, these questions can only be answered by a microscopic analysis of the emission. In this regard and in contrast to commonly used Schrödinger-picture descriptions, the cluster-expansion description enables a new, rigorous and unambiguous definition of a cavity-enhanced single photon stemming from the Raman process.

Moreover, this approach goes beyond established effective-Hamiltonian descriptions\(^{5,38}\) which are limited to the far-off-resonant regime of the Raman process. Here, we will follow Ref. 39 and establish a decomposition of the full cavity-photon population \(N\) into contributions \(N_S\) of all emission sources \(S\) corresponding to different excitation (or emission) paths,

\[
N = \sum_S N_S. \tag{1}
\]

We will investigate under which conditions the decomposition of the full emission is dominated by the Raman source \(S = R\) and compute the corresponding emission probabilities \(P_S\). Based on our definitions, we will furthermore be able to distinguish whether the Raman process indeed generates a single Raman photon or causes destructive interference with the emission from other sources \(S\). We note that this approach can easily be extended to systems with more than three levels.
II. THREE-LEVEL SYSTEMS IN PHOTON-EMISSION CONFIGURATION

Let us first introduce the Hamiltonians of the four fundamental three-level configurations which have the form

\[
H(t) = H_0 + H_1(t)
\]

with the noninteracting part

\[
H_0 = \hbar \left( \sum_{i=1}^{3} \omega_i P_{i,i} + \omega_c b^\dagger b \right)
\]

and the light-matter interaction part \(H_1\). Here, we have defined the electronic operators \(P_{i,j} = |i\rangle\langle j|\) and the bosonic photon annihilation (creation) operators \(b^\dagger\). The energies of the electronic states and the cavity are given by \(\hbar \omega_i\) and \(\hbar \omega_c\). For the respective configuration (see Fig. 1 for the nomenclature) and in rotating-wave approximation (RWA), \(H_1\) is explicitly given by

\[
\begin{align*}
\Xi_1(t) &= \Omega^*(t) P_{2,1} + \Omega(t) P_{1,2} + g (P_{3,2} b^\dagger + P_{2,3} b), \quad (3) \\
\Lambda(t) &= \Omega(t) P_{2,1} + \Omega^*(t) P_{1,2} + g (P_{3,2} b^\dagger + P_{2,3} b), \quad (4) \\
\Xi_2(t) &= g (P_{2,1} b^\dagger + P_{1,2} b) + \Omega^*(t) P_{2,3} + \Omega(t) P_{2,3}, \quad (5) \\
V(t) &= g (P_{2,1} b^\dagger + P_{1,2} b) + \Omega(t) P_{3,2} + \Omega^*(t) P_{3,2}. \quad (6)
\end{align*}
\]

The constant \(g\) denotes the photon-material coupling and \(\Omega^*(t)\) is the Rabi energy for absorption of (or stimulation by) the coherent external light field at time \(t\). It is explicitly given by \(\Omega(t) = \Omega_{\text{env}}(t)e^{-i\omegaLt}\) where \(\Omega_{\text{env}}\) denotes the envelope part and \(\omega\) the excitation frequency. To be able to provide a more transparent discussion of relevant physical processes, in the present work we assume that laser field and cavity mode have orthogonal polarizations. The general discussion, however, can be extended to arbitrary selection rules. The Hamiltonians Eqs. (3) and (4) can be transformed into each other by interchanging \(\Omega\) and \(\Omega^*\). This is also valid for Eqs. (5) and (6). Hence, the four configurations can be divided into two general types, \(\Lambda\)-type and \(\Omega\)-type (see also Fig. 1).

III. EQUATION-OF-MOTION APPROACH

Here we only give a brief overview of the equation-of-motion approach combined with the cluster-expansion method which is meanwhile also established in textbooks\(^{26,39}\). To analyze the system dynamics, we evaluate the Heisenberg equation of motion

\[
\frac{i\hbar}{\partial t} \langle O \rangle = \{[O, H], \langle O \rangle\} + \sum_L \langle L \rangle L. \quad (7)
\]

describing the dynamics of the expectation value \(\langle O \rangle\) of an operator \(O\). We model losses and dephasing via the common Lindbladian

\[
\mathcal{L}_L(O) = \frac{1}{2} \left( [L^\dagger [O, L] + [L^\dagger, O] L \right).
\]

We include photon losses from the cavity with the Lindblad operator \(L = \sqrt{\kappa} b\) and polarization decay – also referred to as pure dephasing – via \(L = \sqrt{h\gamma_{i,j}}\sigma_{i,j}\) for \(i > j\). Here, we have introduced the inversion operator \(\sigma_{i,j} = P_{i,i} - P_{j,j}\). Assuming \(\gamma_{i,j} = \gamma_{j,i}\), all the dephasing contributions are the same for the four configurations. Radiative decay, i.e., recombination of the excited levels into the ground states, can be included for the configuration-specific optically allowed transitions \(P_{i,j}\) with \(E_j < E_i\) via \(L = \sqrt{h\gamma_{i,j}} P_{i,j}\). Explicitly, the decay term for \(O = b^\dagger\) is given by

\[
\sum_L \mathcal{L}_L(b^\dagger) = \frac{-i\hbar}{2} \frac{\kappa}{2} b^\dagger \quad (9)
\]

and the pure-dephasing term for \(O = P_{m,n,m}\) by

\[
\sum_L \mathcal{L}_L(P_{m,n}) \bigg|_{\text{pure}} = \frac{i\hbar}{2} \sum_{i,j} (2\delta_{m,i} \delta_{j,n} + 2\delta_{m,j} \delta_{i,n} - \delta_{i,m} - \delta_{j,m}) P_{m,n}. \quad (10)
\]

We note that the inclusion of radiative contributions to the decay is straightforward but complicates a direct comparison of the three-level systems of different type. A detailed discussion is given in Appendix B 2.

We use the cluster-expansion method\(^{26,39}\) to deal with the many-particle hierarchy problem resulting from Eq. (7), i.e., in normal order, we expand all one-, two-, three-operator and higher-order expectation values into their correlated clusters. These correlations are named singlets, doublets, triplets, quadruplets and so forth, corresponding to their respective interaction hierarchy, and are defined via a recursive factorization scheme. We indicate correlations of an operator quantity \(O\) with double brackets, \(\langle O \rangle\). The singlets are identical with their full expectation value, here \(\langle P_{i,j} \rangle = \langle P_{i,j} \rangle \) and \(\langle b^\dagger \rangle \) \(\langle b^\dagger \rangle \). For the doublets appearing in our calculations, we find

\[
\langle b^\dagger b \rangle = \langle b^\dagger \rangle \langle b \rangle + \langle b^\dagger \rangle, \quad \langle P_{i,j} b^\dagger b \rangle = \langle P_{i,j} \rangle \langle b^\dagger \rangle \langle b \rangle \quad (11)
\]

Note that under our excitation conditions (the laser polarization is assumed to be orthogonal to the cavity polarization) the photon singlets \(\langle b^\dagger \rangle \) are always zero. Analogously, we find for the triplets

\[
\langle P_{i,j} b^\dagger b^\dagger \rangle = \langle P_{i,j} \rangle \langle b^\dagger \rangle \langle b \rangle + \langle P_{i,j} \rangle \langle b^\dagger \rangle \quad (12)
\]

and for the quadruplets

\[
\langle P_{i,j} b^\dagger b^\dagger b \rangle = 2 \langle P_{i,j} \rangle \langle b^\dagger \rangle \langle b \rangle + \langle P_{i,j} \rangle \langle b^\dagger \rangle, \quad \langle P_{i,j} b^\dagger b^\dagger b^\dagger \rangle = 2 \langle P_{i,j} \rangle \langle b^\dagger \rangle \langle b \rangle \langle b \rangle + \langle P_{i,j} \rangle \langle b^\dagger \rangle \langle b \rangle \quad (13)
\]

In the following, we implement the singlet-doublet-triplet (SDT) approximation and neglect the quadruplets and all higher-order correlations.

To compute the full emission dynamics as a reference, we can directly solve the von-Neumann equation for the density operator \(\rho\),

\[
\frac{i\hbar}{\partial t} \rho = [H, \rho] + \sum_L \dot{\mathcal{L}}_L(\rho), \quad (14)
\]
where $\hat{L}(\rho)$ is the Lindbladian Eq. (8) in the Schrödinger picture,
\begin{equation}
\hat{L}(\rho) = \frac{i}{2} \left( 2L\rho L^\dagger - \rho L^\dagger L - L^\dagger L\rho \right).
\end{equation}

The expectation value of an operator $O$ is then defined via taking the trace with the density operator, $\langle O \rangle = \text{tr}(\rho O)$. This exact approach is used to verify the cluster-expansion approximation described above, see also Appendix B. In the following, we always assume that all the three-level systems are initialized in state 1 without any initial electronic polarizations and without any photons inside the cavity.

### IV. RESULTS

To thoroughly analyze the Raman-emission dynamics – which is strongly affected by light-induced energy shifts of the Stark type – we allow the control laser to be tunable around the bare Raman-resonance condition via an adjustable detuning $\Delta_L$ and define the excitation frequency $\omega_L$ as
\begin{equation}
\hbar \omega_L = |h(\omega_1 - \omega_3 - \omega_C)| + \Delta_L.
\end{equation}

We may further assume $|\omega_1 - \omega_3| < \omega_C$ for the V- and $\Lambda$-configurations. Moreover, we introduce the detuning of the cavity from the two-level subsystem as
\begin{equation}
\Delta_C = h(\omega_i - \omega_j - \omega_C)
\end{equation}

with $(i, j) = (1, 2)$ for the V-type and $(2, 3)$ for the $\Lambda$-type system, hence $\omega_i - \omega_j > 0$. Both definitions of these detunings are sketched in Fig. 3. In this context, we further assume typical energy scales of semiconductor-QD-based systems where energy differences of optically allowed transitions are on the order of $1-2 \text{ eV}$.

As detailed in the previous section, we derive the full sets of equations of motion for the two system classes within the SDT approximation, the equations are explicitly given in Appendix A. Whenever we use the $\pm$-notation the upper sign refers to the $\Lambda$- or V-configuration of the respective system class and the lower sign to the $\Xi$-configurations. To express the appearing energy differences by the above defined detunings $\Delta_L$ and $\Delta_C$, we go into the rotating frame of the light field. For a quantity $Q$ the envelope part $Q_{\text{env}}$ is given via $Q(t) = Q_{\text{env}}(t)e^{i\omega_L t}$.

To show that $\Lambda$- and V-type systems exhibit different Raman-emission behaviour, in the first instance we analyze and compare their equations of motion of the Raman-photon-assisted polarization
\begin{equation}
R \equiv \langle P_{3, 1} b^\dagger \rangle
\end{equation}

which is a doublet correlation. To discriminate between the photon-assisted polarization $R$ and the photon-assisted polarization of the respective cavity-two-level subsystem, $II \equiv \langle P_{3, 2} b^\dagger \rangle$ ($\Lambda$-type) and $\langle P_{2, 1} b^\dagger \rangle$ (V-type), we refer to $R$ as Raman correlation or Raman polarization and $II$ commonly as photon-assisted polarization. Moreover, for a quantity $Q$, we will indicate the system type $T$ by the respective superscript, $Q^T$ with $T = \Lambda$ or $V$.

#### A. Raman-correlation equation

The Raman correlation of $\Lambda$-type systems obeys the equation of motion (see Eq. (A3))
\begin{equation}
\frac{\partial}{\partial t} R_{\text{env}}^\Lambda = -i \left( \frac{\Delta_L}{\hbar} - i\frac{\kappa + \Gamma_{3, 1}}{2} \right) R_{\text{env}}^\Lambda - i\frac{\Omega_{\text{env}}}{\hbar} II^\Lambda
+ i\frac{g}{\hbar} P_{\text{env}}^\Lambda (1 + N^\Lambda) + i\frac{g}{\hbar} C_{\text{env}}^\Lambda
\end{equation}

and the Raman correlation of $V$-type systems obeys the equation (see Eq. (A13))
\begin{equation}
\frac{\partial}{\partial t} R_{\text{env}}^V = -i \left( \frac{\Delta_L}{\hbar} - i\frac{\kappa + \Gamma_{3, 1}}{2} \right) R_{\text{env}}^V + i\frac{\Omega_{\text{env}}}{\hbar} II^V
- i\frac{g}{\hbar} P_{\text{env}}^V N^V - i\frac{g}{\hbar} C_{\text{env}}^V.
\end{equation}

The Raman polarization is a coherent correlation as it only exists in the presence of the control-laser field. Here, we have introduced the dephasing constants $\Gamma_{i,j}$ for the polarizations – in general containing both radiative decay and pure dephasing – which are explicitly given by Eq. (10) modified by the respective radiative-loss constant (see Appendix B). $N^T$ denotes the cavity-photon population $\langle b^\dagger b \rangle$ of the $T$-type system and $n_i^T$ the electronic occupation of level $i$, $\langle P_{i, i} \rangle$. $P^T$ is the laser-induced electronic polarization with $P^\Lambda \equiv \langle P_{2, 1} \rangle$ and
In total, we find three fundamental source terms. In the first line of both equations, the laser field is explicitly entering and converts photon-assisted polarization $P$ into Raman correlation $R$. The first term in the second line describes how the laser-induced polarization $\Pi$ acts as a source of Raman correlation. Furthermore, there is a triplet source $C$ which is a correlation between the laser-induced polarization and the emitted photon-population. Equations (19) and (20) are formally exact within the SDT approximation because Raman correlation as a doublet correlation only couples to triplet correlations according to the corresponding hierarchy in the Heisenberg equation of motion.

Comparing the source terms of Eqs. (19) and (20), we identify structural and, due to the initial condition, further differences. In $V$-type systems, $P^V$ only enters through the feedback with the cavity field $N^V$ via $P^V_{env}N^V$. In $\Lambda$-type systems $P^\Lambda$ does not only enter via backaction of the cavity field but also appears as a separate source term via $P^\Lambda_{env}(1 + N^\Lambda)$. Another difference between the system classes is produced by the different dynamics of the $\Pi$-term. Since $V$-type systems spontaneously decay via the cavity once the initial state 1 is occupied, $\Pi^V$ contains a non-vanishing source term even when no control field is present. This spontaneous incoherent source term is immediately available for the conversion into Raman correlation applying an appropriate control pulse. For comparison, in $\Lambda$-type arrangements $P^\Lambda$ must be created first, before the build-up of Raman correlation can occur. These observations from the equation structure of the Raman correlation will become more clear later during the analysis of measurable quantities shown in Figs. 4–7.

Equations (19) and (20) can be solved analytically in the quasi-stationary limit for weak continuous-wave (cw) excitation (see Appendix B3). Many properties derived in this regime are – at least qualitatively – conserved also in the limit of non-stationary conditions, e.g., pulsed excitations or excitations causing occupation inversion. In particular, we can detect how Stark shifts affect the Raman-excitation conditions. Namely, for the case of optimum Raman-excitation conditions we can identify a common expression for the optimum-laser detuning in the cw limit ($\Omega_{env}(t) = \Omega_0$) for both system classes,

$$\Delta c_{\text{cw}} = \pm \frac{\Delta_c \Omega_0^2}{\Delta_c^2 + \left(\hbar \chi_{2,2,2} \frac{1}{2}\right)^2}, \quad (21)$$

for $\Lambda[V]$-type systems. Hence, the sign of the detuning $\Delta c$ decides if the control laser introduces a blue or red shift of the photon emission according to energy conservation of the two-photon process, in agreement with earlier investigations of a related situation. Furthermore, our above made observations concerning the fundamental differences between the source terms also become obvious in the analytic cw solution. Namely, we find that the $\Lambda$-type Raman correlation scales like

$$|R_{\text{cw}}^\Lambda|^2 \propto \frac{g^2 \Omega_0^2}{(\Delta_c \mp \Delta_L)^2 + \left(\hbar \chi_{2,2,2} \frac{1}{2}\right)^2}, \quad (22)$$

and the $V$-type Raman correlation like

$$|R_{\text{cw}}^V|^2 \propto \frac{g^2 \Omega_0^2}{\Delta_c^2 + \left(\hbar \chi_{2,2,2} \frac{1}{2}\right)^2}. \quad (23)$$

We clearly see that Eq. (22) contains an additional resonance condition for $\Delta_L$ besides Eq. (21) stemming from the specific $P^\Lambda$-coupling in Eq. (19). The condition $\Delta_c \mp \Delta_L = 0$ means that the control laser becomes resonant with the electronic transition between levels 1 and 2. In contrast to that, $R^V$ mainly scales monotonically with $\Omega_0$ according to Eq. (23). This difference will become clearly visible in the input-output characteristics of the Raman emission shown in Figs. 6 and 7.

So far we can summarize that $\Lambda$- and $V$-type systems show some significant differences mainly due to a qualitatively different dependence on the control-laser induced energy shifts. This has not been discussed to its full extent in previous studies where these shifts were neglected or their influence underestimated. In a next step, we analogously analyze the source terms of the cavity-photon population $N$. This will allow us to find an accurate and unambiguous definition of Raman population $N_R$ which will be needed for any further discussion of our systems as a Raman-based single-photon source.

### B. Definition of Raman population

The cluster-expansion method enables us to establish a reasonable decomposition of the total photon population $N$ created inside the cavity into different terms stemming from different microscopic physical processes present in our systems. For a $T$-type system, the total photon population is given via formal integration of Eqs. (A1) and (A11) by:

$$N^T(t) = \frac{2g}{\hbar} \text{Im} \int_0^t dt' e^{-\kappa(t-t')} \Pi^T(t'). \quad (24)$$

Here, the photon-assisted polarization $\Pi^T$ is analogously obtained from integration of Eq. (A2) yielding

$$\Pi^\Lambda(t) = \int_0^t dt' e^{i \left(\frac{\Delta_c}{\hbar} + \frac{i g \sigma_{3,2}}{\hbar} \right) (t-t')} \left[ -\frac{i \Omega_{env}(t')}{\hbar} R_{env}(t') + \frac{i g}{\hbar} n_{\Lambda}(t') \right]$$

$$+ \frac{i g}{\hbar} \sigma_{\Lambda}(t') N^\Lambda(t') - \frac{i g}{\hbar} \Sigma^\Lambda(t') \quad (25)$$
and integration of Eq. (A12) yielding
\[
\Pi^V(t) = \int_0^t dt' e^{-i \frac{\phi}{\hbar} (1 + e^{i\frac{\phi}{\hbar}})} \left[ \frac{i \omega_{\text{env}}(t')}{\hbar} R^V_{\text{env}}(t') + i g \sigma^{V}_{1,2}(t') R^V_{\text{env}}(t') \right].
\]

(26)

In Eqs. (25) and (26), the source term in the first line describes the Raman process (denoted as R). In the second line, the first term is a purely electronic source (denoted as E) related to electronic-occupation transfer into or from the upper level of the cavity-two-level subsystem. The second term describes the backaction of the electronic inversion \(\sigma^T_{i,j} = n^T_{i} - n^T_{j}\) with the created cavity population (denoted as \(B\)) and the third term, \(\Sigma^V = \{\sigma_{3,2}^V b^\dagger b\} \) and \(\Sigma^V = \{\sigma_{2,1}^V b^\dagger b\}\), is a triplet contribution (denoted as \(T\)) resulting from correlations between photon population and electronic inversion. Both equations are exact because – as mentioned in the previous section – the triplet order is the highest appearing order in the equation of motion of a doublet quantity.

According to the above defined source terms of type \(S\), we can eventually specify the photon-population decomposition, Eq. (1), yielding
\[
N^T = \sum_{S=R,E,B,T} N^T_S. \tag{27}
\]

Notably, the Raman population is explicitly given by
\[
N^V_{R[\Lambda]}(t) = \text{Im} \left\{ \left[ -\frac{2i g}{\hbar^2} \int_0^t dt' \int_0^{t'} dt'' e^{-\kappa(t'' - t')} \times e^{-i \frac{\phi}{\hbar} e^{i\frac{\phi}{\hbar}} (t'' - t')} \Omega_{\text{env}}(t'') R^V_{\text{env}}(t'') \right] \right\} \tag{28}
\]

where expressions in square brackets are for \(A\)-type systems. We note that this is an unambiguous and precise definition of Raman-photon population. It can be computed directly from the SDT equations or via the full solution of the von-Neumann equation, Eq. (14) (see Appendix B1). We note that \(N_{R}\) defined by Eq. (28) and other contributions \(N_S\) can become negative while \(N = \sum_S N_S\) as a physical population always remains positive. We will discuss and interpret this property below in Section IV C in the context of the corresponding photon-emission probability. By the introduction of a generalized emission probability, we will specify the condition for which the Raman process leads to the emission of a pure single Raman photon or just diminishes the photon emission stemming from other contributing processes.

To give a brief illustration of our decomposition, Eq. (27), and of the discussions above, we show typical examples of excitation dynamics for both system classes in Fig. 4. We present the SDT photon-emission dynamics for a Gaussian control pulse with a maximum Rabi energy of 0.7 meV [Figs. 4a–4c] and the dynamics of a rectangular pulse of the same pulse area \(\theta = 5\pi\) (maximum Rabi energy of 0.281 meV) [Figs. 4d–4f]. According to our RWA Hamiltonians, the pulse area is given by
\[
\theta = \frac{2}{\hbar} \int_0^\infty dt \Omega_{\text{env}}(t). \tag{29}
\]

At a first glance, our choice of the two control pulses might seem arbitrary, but they are carefully chosen to highlight that the Raman process exhibits a non-trivial dependence on pulse shape and peak intensity. Both pulse durations are chosen such that the induced Raman-photon populations are completely emitted inside the same time window (see the yellow lines in Fig. 4). This allows for the most reasonable comparison between both excitation scenarios. In particular, the rectangular pulse is designed to efficiently pick off the incoherent source (blue lines) of Raman correlation in the V-type case [Figs. 4c and 4f]. Remarkably, in the context of the Raman-emission probability (see discussion of Fig. 6 below), we will see that, in our example, the rectangular pulse – which has the same pulse area but a lower maximum-Rabi amplitude – in both system classes can actually lead to a Raman probability twice as high as for the Gaussian control pulse. We note that in the limit where cavity and laser are spectrally tuned into the vicinity of the electronic resonance – which will be necessary for achieving on-demand emission (see discussion of Fig. 5 later) – the Raman-emission properties are not exclusively determined by the bandwidth of the pulse. Our findings clearly indicate that they show a very complex dependency on the combination of pulse bandwidth, pulse shape, pulse intensity and cavity detuning together with the resulting excitation-induced energy shifts (see Eqs. (21)-(23) and Ref. 46). In our example, we choose the limit of strong cavity detuning where the emission should be dominated by the Raman process. In \(\Lambda\)-type systems [Figs. 4b and 4c], the full emission (shaded area) is completely stemming from the Raman emission (yellow line). Electronic-occupation induced photon population (blue line) hardly contributes. In V-type systems, additionally the initial occupation of level 1 instantaneously generates incoherent photon population (blue line) before the control pulse starts to induce Raman emission [Figs. 4c and 4f]. The sum of both contributions (black dashed line) in all four cases describes the full emission very well. In particular, this example shows that our decomposition definition indeed is physically reasonable. We note that the rectangular pulse is introduced here to generate a hybrid scenario between a pulsed and a cw scenario. We further note that – although the emission dynamics are non-stationary – our quasi-stationary cw approximations will apply and lead to a deeper understanding. In the following, we will investigate the emission characteristics more systematically in dependence of varying cavity detuning, control detuning and control power. In this regard, we will further demonstrate that the incoherent emission in V-type systems can actually be efficiently converted into Raman population.
For this purpose, we need a unique measure to characterize and compare the performance of our two system classes, given by the photon-emission probability as discussed in the following section.

**C. Generalized emission probability**

The total photon-emission probability up to time $t$ of our QD-cavity systems can be obtained from

$$P(t) = \kappa \int_0^t dt' N(t'),$$

within the Lindblad formalism. This quantity actually gives the total photon number emitted during the time interval $[0, t]$. Due to our initial condition and assuming that on the time scale of the photon life time $1/\kappa$ no loop $1 \rightarrow 3 \rightarrow 1$ among the electronic configurations has to be considered (e.g., via nonradiative spin relaxation between the two ground states 1 and 3 in a QD-$\Lambda$-configuration), a maximum of one photon can be emitted into the cavity mode during a single excitation cycle. Hence, we identify $P = 1$ with 100\% emission probability such that $0\% \leq P \leq 100\%$. The use of our decomposition, Eq. (27), in Eq. (30) yields

$$P(t) = \sum_S \kappa \int_0^t dt' N_S(t') \equiv \sum_S P_S(t)$$

(31)

where in particular the Raman-photon emission probability of type $T$,

$$P_R^T(t) = \kappa \int_0^t dt' N_R^T(t'),$$

(32)

is defined via Eq. (28). We have to emphasize that $P_S$ can become negative or greater than 100\% which is, as already mentioned above in the context of the population decomposition, the fingerprint of quantum interference. For that reason, $P_S$ has to be understood as a generalized emission probability. It can be identical with a physical probability in the case where $0\% \leq P_S \leq 100\%$ for all sources $S$. For a chosen cavity detuning and control amplitude, the optimum control detuning $\Delta_{L\text{opt}}$ is then defined as the control detuning $\Delta_L$ for which the Raman-emission probability reaches its maximum,

$$\Delta_{L\text{opt}} = \Delta_{L\text{max}(P_S)}.$$  

We can directly apply our emission-probability decomposition, Eq. (31), to our example shown in Fig. 4. For the Gauss-pulse dynamics we find $P_R^A(t = 30 \text{ ps}) = 2.85\%$ as the Raman-emission probability and $P_R^E(t = 30 \text{ ps}) = 0.04\%$ as the emission probability of the electronic contribution [Fig. 4b] and $P_{R\text{Tot}}(t = 30 \text{ ps}) = 2.82\%$ and $P_{R\text{Tot}}^V(t = 30 \text{ ps}) = 1.04\%$ [Fig. 4c]. For the rectangular-pulse dynamics we calculate $P_R^A(t = 30 \text{ ps}) = 2.2\%$ and $P_R^E(t = 30 \text{ ps}) = 0.04\%$ [Fig. 4d] and $P_{R\text{Tot}}^A(t = 30 \text{ ps}) = 2.25\%$ and $P_{R\text{Tot}}^V(t = 30 \text{ ps}) = 1.04\%$ [Fig. 4f]. That means, in all cases the emission probability of a Raman photon is only 2–3\% and far away from on-demand behaviour. We therefore have to systematically analyze how and under which conditions on-demand behaviour can be achieved or approached.

For a consistent fundamental comparison of $\Lambda$- and $V$-type systems based on simulations – where we want to create identical conditions for both system classes –, we neglect radiative decay of the electronic occupations which does not depend on the system class ($\Lambda$- or $V$-type) but on the actual configuration ($\Xi$, $\Lambda$ or $V$) of the respective system. Since radiative decay only modifies the electronic damping constant $\Gamma_{3,1}$ and does not change the structure of the Raman equations, Eqs. (19) and (20), this will hardly influence the physical observations from our numerical results presented in the following. In particular, neglecting radiative decay is valid for pulsed excitations where the duration of the control pulse is significantly shorter than the radiative lifetime of the occupations\cite{15, 20}. A detailed discussion of the influence of radiative decay is given in Appendix B2.

Figure 5 shows the emission probabilities for varying control detuning $\Delta_L$ and cavity detuning $\Delta_C$ of two
representative excitation scenarios for a Λ-type (upper block) in comparison with a V-type system (lower block), one for a weak rectangular pulse (θ = 0.1π) and one for a strong rectangular pulse (θ = 5π). The probabilities are calculated at time t = 30 ps, about 10 ps after the presence of the control pulse such that the Raman population has completely been emitted according to Fig. 4. We present the full emission probability \( P \) (first column) and the two most important components of the decomposition, the Raman-emission probability \( P_R \) (second column) and the electronic emission probability \( P_E \) (third column). In all panels we plot the analytic cw result for the light-induced energy shift, Eq. (21), as a guide to the eyes. The full emission is approximately represented by the sum of the Raman and electronic part. We observe that especially the weak-excitation scenario – where Stark shifts are hardly visible – of a Λ-type system [panels a-c] looks completely different than the one of a V-type system [panels g-i]. The main difference is that we find a small positive Raman probability (< 0.4%) in the Λ-type system [panel b] for the case of a resonant cavity, \( \Delta_C \approx 0 \), in combination with the bare Raman-resonance condition, \( \Delta_L \approx 0 \). In contrast to that, in the same region of the Raman map, the corresponding V-type system shows a purely negative Raman (generalized) emission probability, hence the Raman process does not generate any photon but causes destructive interference diminishing the emission stemming from the electronic source [panel i]. Additionally, we observe a further negative region in the Λ-type Raman map in Fig. 5b along the diagonal \( \Delta_L = -\Delta_C \) according to our observations from Eq. (22). As can be seen in Fig. 3, along this diagonal the control laser is always resonant with the electronic transition between states 1 and 2. Obviously, the V-type system [Fig. 5i] is not sensitive to this resonance, in agreement with Eq. (23). This is a direct consequence of the different electronic-polarization coupling observed in the Raman equations, Eqs. (19) and (20). The orientation of the Λ-type electronic emission and total emission also follow the diagonal. The orientation of the V-type electronic emission follows the cavity-resonance line \( \Delta_C = 0 \) because the fully occupied initial state can efficiently decay via the resonant cavity. In the case of strong excitation the Raman patterns of both system classes [panels e and k] become more similar, but their maxima are located differently. The analytic dependency shown by the white line especially describes the optimum line of the Λ-type Raman map very well. Obviously, Raman interference occurs in both classes in the region characterized by the extremal points of the white line, namely for

\[
|\Delta_C| < h \frac{\kappa + \Gamma_{3,2}[2,1]}{2}
\]

in a Λ[V]-type system. Hence, the critical detunings for interference are only determined by the cavity losses, \( \kappa \), and the total electronic-dephasing rate, \( \Gamma_{i,j} \), of the two-level-cavity subsystem. The corresponding control detunings of the extremal point are dependent on the pulse amplitude,

\[
|\Delta_L| = \frac{\theta^2}{h} \frac{\kappa + \Gamma_{3,2}[2,1]}{\kappa + \Gamma_{3,2}[2,1]}.
\]

At this point we want to emphasize that the maxima observed in the full emission [panels d and j] are not identical with the maxima of the Raman contribution. Notably, in the V-type system, even when Stark shifts are hardly visible [panel h], the light-induced effects are nevertheless contributing via the interference condition, Eq. (34). Interestingly, V-type Raman emission is quite insensitive against the choice of the control detuning over a range of almost 1 meV for the case of a near-resonant cavity [panels h and k]. Here the Raman emission exhibits a pronounced shoulder due to electronic-to-Raman emission conversion. Corresponding to this efficient conversion, the maximum stripe of the electronic contribution observed in panel i is much narrower in comparison with the stripe of the weak-excitation scenario [panel i]. This conversion effect is absent in Λ-type systems.
We do not show the emission maps for the corresponding Gauss-pulse scenario or for enhanced coupling strength $g$, because they qualitatively look very similar. We will discuss these scenarios again below in the context of input-output characteristics.

According to our observations from Figs. 5d-5f, we can now state that the interference dip observed in the spectrum, shown in Fig. 2, actually stems from the Raman process diminishing the emission from the electronic source. Hence, we observe destructive interference between the direct two-photon Raman path $1 \rightarrow \text{opt} \rightarrow 3$ via electronic-occupation transfer. Note that the interference feature can be masked in the spectrum if the dip is compensated by the emission from other channels, as can also be seen in Fig. 2 for longer measurement durations. This can especially happen in V-type systems where the resonant emission is strongly dominated by the electronic source [Figs. 5g-5l]. Only a detailed decomposition is able to discriminate the different contributing emission channels.

### D. Input-output characteristics

Although the Raman maps can have their absolute maxima in the limit of a near-resonant cavity, the efficiency of the Raman process compared with the total output is higher for cavities with intermediate detunings where $P_R = P$. For that reason we will now turn our attention to the input-output characteristics for the case where cavity and electronic resonance overlap only moderately. For our chosen system parameters, this is the case for about $|\Delta_C| = 0.8$ meV. We vary the control amplitude, i.e., vary the pulse area, and we calculate the corresponding maximum-Raman emission via the optimum-control detuning, Eq. (33). We present the input-output characteristics for pulse areas up to $\theta = 15\pi$ to highlight the physical differences between the two system classes in a better way. We note that the range up to $\theta \approx 10\pi$ should be experimentally relevant.\(^{45}\)

Figure 6 shows a typical example of the optimum-Raman characteristics induced by our Gauss [Figs. 6a and b] and our rectangular pulse [Figs. 6c and d]. The upper panels show the numerically and analytically calculated optimum-control detunings and the lower panels the corresponding total and Raman-emission probabilities, again taken 10 ps after the control pulse. We use our standard system parameters which represent an intermediate cavity quality as experimentally used in Ref. 14. Both scenarios have in common that the V-type system is a very pure Raman-photon source where the Raman contribution can be near identical with the total emission probability. As already discussed above, the V-type system exhibits a small background stemming from the incoherent electronic source which is also present without control pulse. This can be seen as a small offset at $\theta = 0$ (black solid lines in Figs. 6c and d). The big difference of the input-output characteristic lines is that the V-type system exhibits a quite monotonically increasing behaviour even in the regime of very strong pulses whereas for the Λ-type system we observe a saturation behaviour for the rectangular case or even a break-down for the Gauss pulse at $\theta \approx 6\pi$. We further observe that the rectangular pulse – representing a longer coupling duration in the chosen time window – strongly enhances the output compared with the Gauss-pulse scenario\(^{46}\), here by a factor of two (although the peak Rabi energy of the rectangular pulse is lower while both pulse areas are equal). It further induces a more efficient conversion of spontaneous background emission into Raman emission which can be observed if we compare the smaller difference between the solid lines in Fig. 6d with the difference observed in Fig. 6b. The optimum-control detunings of the rectangular pulse are nearly perfectly described by the cw approximation [panel b] and they are much smaller than the detunings for the case of the Gauss pulse [panel a]. Nevertheless, for the Gauss pulse, the sign and the order of magnitude of the V-type optimum shift are still well described by our cw expres-
FIG. 7. Optimum-emission probability for enhanced light-matter coupling. We present the results for the same situation as in Fig. 6 but with enhanced coupling strength between cavity photons and QD, \( g = \hbar \kappa = 186 \ \text{meV} \).

...for increasing pulse areas. In contrast to that the \( \Lambda \)-type input-output line always reveals intensity-induced damping. Figure 7d again demonstrates that incoherent photon emission can be transformed into Raman-photon population: while at \( \theta = 0 \) we find \( \mathcal{P}_V^V - \mathcal{P}_R^V = 40\% \) for the incoherent off-set, we end up with \( \mathcal{P}_V^V - \mathcal{P}_R^V = 5\% \) for \( \theta = 15\pi \).

Finally, we come back to the resonant Raman process where both control laser and cavity are resonant with the respective electronic transition, \( \Delta_C = \Delta_L = 0 \). Here, we focus only on \( \Lambda \)-type systems because as discussed before – \( V \)-type systems do not emit Raman photons in the sense of our definition in the resonant limit. Figure 8 shows the input-output characteristics of a \( \Lambda \)-type system for varying pulse area. We observe damped Rabi oscillations of the total emission \( \mathcal{P}_R^\Lambda \) (black dashed line). Obviously, these oscillations stem from the electronic source \( \mathcal{P}_E^\Lambda \) (shaded area) related to the path \( 1 \xrightarrow{\Omega(t)} 2 \xrightarrow{b^1} 3 \) which is the dominant positive contribution. The Raman signal \( \mathcal{P}_R^\Lambda \) (yellow dashed line) exhibits only a single Rabi oscillation for the \( \pi \)-pulse. It becomes negative beyond \( \theta = 2\pi \) and decreases quite monotonically for increasing pulse areas. Notably, the Raman correlation is responsible for the excitation-induced damping. We find that this is a general phenomenon of the resonant Raman process: efficient occupation transfer from the initial to the intermediate state and the simultaneous build-up of Raman correlation start to counteract for enhanced excitation intensities, because, in \( \Lambda \)-type systems, both processes share the laser-induced electronic polarization as the only initial source (see also Eqs. (A3) and (A6)). Interestingly, the \( \pi \)-pulse can nevertheless invert the system from the initial to the final state nearly deterministically – in our example, about 70% of the occupation is transferred (not shown) – which is commonly introduced as a spin-flip...
Spin-flip Raman transition in Λ-configurations\textsuperscript{19}. Obviously, this spin-flip Raman transition is physically different from the Raman-correlation-mediated process. Hence, we conclude that the measured damped Rabi oscillations in a Λ-system presented in the Supplemental Material of Ref. 19 – which are induced by resonant excitation – do not originate from the excitation channel $1 \rightarrow 3$ mediated by the Raman correlation but purely from the inversion-induced emission. In particular, according to Fig. 8, the damping – which in Ref. 19 is attributed to acoustic phonons and further non-Markovian dephasing reservoirs – can already be induced by the Raman correlation itself via destructive interference, even in the absence of phonons. Our findings also offer an alternative interpretation of the observed Raman-linewidth broadening for vanishing laser detuning: under certain conditions, going into the resonance limit, we do not observe the linewidth-broadening of the Raman process (see Appendix B3) anymore, but the linewidth of the electronic emission which commonly is broader than the Raman linewidth in the cw limit. With the same argument, we may also explain the measured unexpected Raman peak-intensity and broadening behaviour of the resonant Raman process which is discussed and left as an open question in the Supplementary Information of Ref. 14.

V. CONCLUSIONS

Using the microscopic cluster-expansion method, we discuss new aspects of cavity-enhanced single-photon generation from optical two-photon Raman processes. We find that either coupling the cavity with the initial or with the final state defines two different classes of three-level-cavity systems which can be characterized by their strongly different Raman-emission dynamics and emission probability. Physically, this difference mainly originates from the different coupling of the control laser with the two-photon Raman process: in contrast to Λ-type systems, V-type Raman correlation only weakly depends on the control-laser induced electronic polarization. Furthermore, V-type systems exhibit efficient conversion of incoherent emission into coherent Raman emission. We find that either coupling the cavity with the initial or with the final state defines two different classes of three-level-cavity systems which can be characterized by their strongly different Raman-emission dynamics and emission probability. Physically, this difference mainly originates from the different coupling of the control laser with the two-photon Raman process: in contrast to Λ-type systems, V-type Raman correlation only weakly depends on the control-laser induced electronic polarization. Furthermore, V-type systems exhibit efficient conversion of incoherent emission into coherent Raman emission. We further show that under resonant excitation conditions we may enter the limit of quantum interference where the Raman process does not generate a single Raman photon but acts as a source of destructive interference with other excitation paths. Hence, the so-called spin-flip Raman process in Λ-configurations – which allows for deterministic inversion from the initial to final state and which is driven under resonant excitation conditions – is physically different from the non-resonant Raman process – which additionally allows for all-optical control of the spectral properties of the emitted Raman photon via the control laser.

Based on our model study for a quantum-dot cavity system we predict that the non-resonant Raman protocol – mediated by the Raman correlation introduced in this article – is indeed able to deliver on-demand photons. This requires appropriately designed control pulses beyond the τ-pulse regime, where Stark shifts and interference effects have to be taken into account, together with suitably detuned high-quality resonators. The general approach and the fundamental properties reported here can also be transferred to atomic three-level-cavity systems. With the additional microscopic insight obtained in the present work, we are furthermore able to interpret and explain in more detail experimental data from the recent literature\textsuperscript{14,19}. The present work also lays the groundwork for the preparation of related experiments and for the optimization of Raman-process based devices for quantum-optical networking.

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Appendix A: Singlet-doublet-triplet equations

We present the SDT equations, computed from Eq. (7) for a light field of the form $Q(t) = Q\text{env}(t)e^{i\omega_L t}$ in the rotating frame. For any quantity $Q$, we define the envelope part via $Q = Q\text{env}e^{i\omega_L t}$ with the lower sign for Ξ-configurations and the upper sign for the respectively related Λ- or V-configuration. Furthermore, we introduce new dephasing constants $\Gamma_{i,j}$ for the polarizations which are explicitly given by Eq. (10). For a consistent and reasonable comparison between the system classes by numerical simulations, we do not consider radiative decay. The inclusion of radiative decay and its influence on the Raman process is discussed in the next section.

1. Λ-type systems

The equations of motion for Λ-type systems without radiative decay are

$$\frac{\partial}{\partial t} \langle b^\dagger b \rangle = -\kappa \langle b^\dagger b \rangle + \frac{2g}{\hbar} \text{Im} \langle P_{3,2} b^\dagger \rangle, \quad (A1)$$

$$\frac{\partial}{\partial t} \langle P_{3,2} b^\dagger \rangle = -i \left( \frac{\Delta_C}{\hbar} - i \frac{\kappa + \Gamma_{3,2}}{2} \right) \langle P_{3,2} b^\dagger \rangle - i\frac{\Omega_{\text{env}}}{\hbar} \langle P_{3,1} b^\dagger \rangle_{\text{env}} + i\frac{g}{\hbar} \langle P_{2,2} \rangle - i\frac{g}{\hbar} \langle \sigma_{3,2} b^\dagger b \rangle - i\frac{g}{\hbar} \langle \sigma_{3,2} b^\dagger b \rangle, \quad (A2)$$

$$\frac{\partial}{\partial t} \langle P_{3,1} b^\dagger \rangle_{\text{env}} = -i \left( \frac{\Delta_u}{\hbar} - i \frac{\kappa + \Gamma_{3,1}}{2} \right) \langle P_{3,1} b^\dagger \rangle_{\text{env}}$$
The equations of motion for V-type systems without radiative decay are

\[
\frac{\partial}{\partial t} \langle P_{2,1} \rangle_{\text{env}} = -i \left( \frac{-\Delta_C + \Delta_I}{\hbar} - \frac{i \Gamma_{2,1}}{2} \right) \langle P_{2,1} \rangle_{\text{env}} \\
+ i \frac{\Omega_{\text{env}}}{\hbar} \langle P_{2,1} b^\dagger \b^\dagger \rangle_{\text{env}}, \quad \text{(A3)}
\]

\[
\frac{\partial}{\partial t} \langle P_{3,2} \rangle_{\text{env}} = -i \left( \frac{-\Delta_C + \Delta_I}{\hbar} - \frac{i \Gamma_{3,2}}{2} \right) \langle P_{3,2} \rangle_{\text{env}} \\
- i \frac{\Omega_{\text{env}}}{\hbar} \langle \sigma_{3,2} \rangle - i \frac{g}{\hbar} \langle P_{3,1} b^\dagger \rangle_{\text{env}}, \quad \text{(A14)}
\]

\[
\frac{\partial}{\partial t} \langle P_{1,1} \rangle = - \frac{2 g}{\hbar} \text{Im} \langle \Omega_{\text{env}} \langle P_{2,1} b^\dagger \rangle_{\text{env}} \rangle, \quad \text{(A5)}
\]

\[
\langle P_{2,2} \rangle = 1 - \langle P_{1,1} \rangle - \langle P_{3,3} \rangle, \quad \text{(A6)}
\]

\[
\frac{\partial}{\partial t} \langle P_{3,3} \rangle = \frac{2 g}{\hbar} \text{Im} \langle P_{3,2} b^\dagger \rangle, \quad \text{(A7)}
\]

Equation (A3) describes the dynamics of the Raman correlation.

2. V-type systems

The equations of motion for V-type systems without radiative decay are

\[
\frac{\partial}{\partial t} \langle b^\dagger b \rangle = -\kappa \langle b^\dagger b \rangle + \frac{2 g}{\hbar} \text{Im} \langle P_{2,1} b^\dagger \rangle, \quad \text{(A11)}
\]

\[
\frac{\partial}{\partial t} \langle P_{2,1} b^\dagger \rangle_{\text{env}} = -i \left( \frac{-\Delta_C + \Delta_I}{\hbar} - \frac{i \Gamma_{2,1}}{2} \right) \langle P_{2,1} b^\dagger \rangle_{\text{env}} \\
+ i \frac{\Omega_{\text{env}}}{\hbar} \langle P_{2,1} b^\dagger \rangle_{\text{env}} + i \frac{g}{\hbar} \langle P_{1,1} \rangle, \quad \text{(A11)}
\]

\[
\frac{\partial}{\partial t} \langle P_{3,2} b^\dagger \rangle_{\text{env}} = -i \left( \frac{-\Delta_C + \Delta_I}{\hbar} - \frac{i \Gamma_{3,2}}{2} \right) \langle P_{3,2} b^\dagger \rangle_{\text{env}} \\
+ i \frac{\Omega_{\text{env}}}{\hbar} \langle P_{3,2} b^\dagger \rangle_{\text{env}} - i \frac{g}{\hbar} \langle P_{2,1} b^\dagger \b^\dagger \rangle_{\text{env}}, \quad \text{(A13)}
\]

Equation (A13) describes the dynamics of the Raman correlation.

Appendix B: Discussion of the SDT equations

In this section we will discuss the SDT equations presented in the previous section, also in comparison with the full solution of the von-Neumann equation Eq. (14). We will further analyze how the equations change if we include radiative decay of the electronic occupations.

1. Full vs. cluster-expansion solution

We compute the Raman maps using the full solution of the von-Neumann equation as input in our Raman-probability definition \( P_R \), Eq. (32). The results are exemplarily shown for the \( \Lambda \)-type system in Fig. 9a and for the V-type system in Fig. 9d for the 5π-pulse scenario from Fig. 5. For comparison, we plot the SDT solution [Figs. 9b and 9c]. To further understand the influence of the different source types on the Raman process, we additionally present the singlet-doublet (SD) result [Figs. 9c and 9f] where we neglect the triplet correlations and, in addition to it, the back coupling of the matter with the cavity-light field. On a first view, we can state that the shape of the Raman pattern remains nearly uninfluenced under the SDT and the SD approximation. Since
the Raman equation is of the triplet order, in particular the SDT result perfectly agrees with the full solution. In the SD approximation we only detect some minor differences in the quantum yield. Explicitly, the SD Raman equations without cavity backaction are

$$\frac{d}{dt} \langle P_{3,1} b^\dagger \rangle_{\text{env}} = -i \left( \frac{\Delta_L}{\hbar} - i \kappa + \frac{\Gamma_{3,1}}{2} \right) \langle P_{3,1} b^\dagger \rangle_{\text{env}}$$

$$- \frac{i}{\hbar} \Omega_{\text{env}} \langle P_{3,2} b^\dagger \rangle + \frac{i g}{\hbar} \langle P_{2,1} \rangle_{\text{env}}$$

(B1)

for Λ-type systems and

$$\frac{d}{dt} \langle P_{3,1} b^\dagger \rangle_{\text{env}} = -i \left( \frac{\Delta_L}{\hbar} - i \kappa + \frac{\Gamma_{3,1}}{2} \right) \langle P_{3,1} b^\dagger \rangle_{\text{env}}$$

$$+ \frac{i}{\hbar} \Omega_{\text{env}} \langle P_{2,1} b^\dagger \rangle$$

(B2)

for V-type systems. Hence, we can conclude that – in contrast to the Λ-type case – the V-type Raman process is quite insensitive against the control-laser induced electronic polarization between levels 2 and 3. Its fundamental source is the term describing the conversion of photon-assisted polarization of the two-level cavity sub-system into Raman correlation. This explains the pronounced shoulder around the near-resonant cavity close to the region of destructive interference [Figs. 9d and 9e]. Note that the analytic line misses this shoulder because, in the quasi-stationary regime, we assume a fixed initial occupation which cannot efficiently decay via the near-resonant cavity to be transformed into Raman correlation (see section B3). The shoulder feature is not visible in the Λ-type pattern [Figs. 9a and 9c], because the conversion of electronic emission into Raman correlation is not available in that region of the Raman map.

Nevertheless, if we want to describe the emission dynamics of all quantities accurately, we have to include the full set of SDT equations or use the von-Neumann result.

2. Radiative decay

Since the inclusion of radiative decay renders a reasonable and consistent comparison of the two different Raman-coupling mechanisms out, we discuss its influence in this subsection separately. Notably, in material systems with longlived occupations or in the case of short excitation durations, radiative decay can be neglected. Radiative decay can explicitly be included in the Ξ-configurations via the Lindblad operators \( L = \sqrt{hr_{2,1}} P_{2,1} \), \( \sqrt{hr_{3,2}} P_{3,2} \), in the Λ-configuration via \( L = \sqrt{hr_{1,2}} P_{1,2} \), \( \sqrt{hr_{3,2}} P_{3,2} \) and in the V-configuration via \( L = \sqrt{hr_{2,1}} P_{2,1} \), \( \sqrt{hr_{2,3}} P_{2,3} \). In particular this does not change the structure of the Raman equations, Eqs. (A3) and (A13), only our introduced total dephasing constant \( \Gamma_{3,1} \) will be modified in the Ξ-systems by \( r_{3,2} \) and in the V-system by \( r_{2,1} + r_{2,3} \). The Raman process in Λ-configurations is unaffected by radiative decay, here, in typical realizations with metastable ground states, the spin-dephasing time between the ground states can generate an additional contribution. The radiative-decay contributions for \( r_{2,1} \) are \( r_{3,2} + r_{2,1} \) (Ξ), \( r_{1,2} + r_{3,2} \) (Λ), \( r_{2,1} \) (V), and for \( r_{3,2} \) the contributions are \( r_{3,2} \) (Ξ), \( r_{1,2} + r_{3,2} \) (Λ), \( r_{2,3} \) (V). Only the occupation-singlet and -triplet equations change their structure, because the occupations explicitly couple to each other. Since the Raman process does not explicitly couple to the occupations, we do not discuss the modifications here. In V-type systems, of course, we need a robust initial occupation of level 1 to efficiently drive the Raman process. In total, we can state that radiative decay will lead to configuration- and device-specific changes of the quantum yield which have to be considered individually for any analysis of experiments. If necessary, further phonon-induced effects can be included.

3. Quasi-stationary solution

To gain first insight into the fundamental Raman-emission characteristics of the two system classes, we simplify the SDT equations for the limit of weak excitations and stationary electronic occupations \( \langle P_{1,1} \rangle = 1, \langle P_{2,2} \rangle = 0 \) such that we can neglect the triplet order and the backaction of the cavity field with the system. The essential properties found in this regime also survive in the non-stationary limit. Temporarily, let \( N \equiv \langle b^\dagger b \rangle \) (photon population), \( R \equiv \langle P_{3,1} b^\dagger \rangle_{\text{env}} \) (Raman correlation), \( \Pi \equiv \langle P_{3,2} b^\dagger \rangle \) or \( \langle P_{3,1} b^\dagger \rangle \) (photon-assisted polarization of Λ or V), and \( P \equiv \langle P_{2,1} \rangle_{\text{env}} \) or \( \langle P_{3,2} \rangle_{\text{env}} \) (Λ- or V-polarization induced by the control laser). For Λ-type
In the cw limit, $\Omega_{\text{env}}(t) = \Omega_0 = \text{const}$, we can easily solve both sets of equations and analyze the Raman-correlation equation. For $\Lambda$-type systems, we find

$$|R|^2 = \frac{g^2\Omega_0^2}{(\Delta_C + \Delta_L)^2 + (h\frac{\Gamma_{2,1}}{2})^2} \left[ \left( -\Delta_L + \frac{\Delta_C\Omega_0^2}{\Delta_C^2 + (h\frac{\kappa + \Gamma_{3,1}}{2})^2} \right)^2 - \frac{(\Delta_C + \Delta_L)g^2}{(\Delta_C + \Delta_L)^2 + (h\frac{\Gamma_{3,1}}{2})^2} \right]^{-1} \left[ \frac{1}{(\Delta_C + \Delta_L)^2 + (h\frac{\Gamma_{3,1}}{2})^2} + \frac{(h\frac{\Gamma_{2,1}}{2})^2}{(\Delta_C + \Delta_L)^2 + (h\frac{\kappa + \Gamma_{3,1}}{2})^2} \right]^{-1},$$

(B7)

and for $V$-type systems we obtain

$$|R|^2 = \frac{g^2\Omega_0^2}{\Delta_C^2 + (h\frac{\kappa + \Gamma_{2,1}}{2})^2} \left[ \left( -\Delta_L + \frac{\Delta_C\Omega_0^2}{\Delta_C^2 + (h\frac{\kappa + \Gamma_{2,1}}{2})^2} \right)^2 + \left( \frac{h\frac{\kappa + \Gamma_{2,1}}{2}}{(\Delta_C^2 + (h\frac{\kappa + \Gamma_{3,1}}{2})^2)^{1/2}} \right)^2 \right].$$

(B8)

If the $g^2$-shift is negligibly small in Eq. (B7) (e.g., if $g$ is small or $\Delta_C$ is significantly greater than $g$), we can identify a common expression for the optimum-laser detuning of both system classes,

$$\Delta_{\text{opt}} = \pm \frac{\Delta_C\Omega_0^2}{\Delta_C^2 + (h\frac{\kappa + \Gamma_{3,1}}{2})^2},$$

(B9)

with $(i, j) = (2, 1)$ for $V$- and $(i, j) = (3, 2)$ for $\Lambda$-type systems, and $-$ for $\Xi$- and $+$ for $\Lambda$- and $V$-configurations. We note that in any system configuration, the laser frequency – i.e. the length of the blue arrows in Fig. 3 – has to be tuned towards the intermediate state 2 by $|\Delta_{\text{opt}}|$ with respect to the bare resonance condition to achieve optimum Raman-photon emission. We also recognize a laser-induced additional damping of the Raman signal which should be minimized via the choice of $\Delta_C$ and $\Omega_0$ in comparison with the natural damping $(\kappa + \Gamma_{3,1})/2$. In particular, for a fixed control amplitude, we observe enhanced damping for vanishing cavity detuning.

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Note that, in this work, we restrict ourselves to optimizing constant (i.e., time-independent) control detunings \( \Delta_t \). In general, the Stark-type energy shifts induced by the control laser are explicitly dependent on the pulse amplitude and the pulse shape, and thus are time-dependent. Hence, self-consistent solutions are required. From our single-doublet equations we can derive an approximation for the time dependency of the optimum control-laser detuning in the limit of off-resonant cavities yielding

\[
\Delta_t^{\text{opt}}(t) = \text{Re} \left[ \int_0^t \int_0^{t'} \int_0^{t''} \Omega_{\text{env}}(t') \Omega_{\text{env}}(t'') e^{i \left( \frac{\Delta_t}{2} - \frac{\kappa t'^{2}}{2(1+\Gamma_0)} \right)} dt'' dt' dt \right],
\]

for A-type [V-type] systems. In comparison with the spectrum of the resulting chirped control pulse, in many cases an effective constant detuning can be found to produce the same Raman-photon output. This is, of course, especially the case for quasi-stationary excitations like rectangular pulses. A detailed investigation of the optimization potential with respect to appropriately chirped pulses will be the starting point of future work.