Analytic few-photon scattering in waveguide QED

David L. Hurst* and Pieter Kok†
Department of Physics and Astronomy, University of Sheffield, Hounsfield Road, Sheffield, S3 7RH, United Kingdom

(Received 22 May 2017; revised manuscript received 9 February 2018; published 20 April 2018)

We develop an approach to light-matter coupling in waveguide QED based upon scattering amplitudes evaluated via Dyson series. For optical states containing more than single photons, terms in this series become increasingly complex, and we provide a diagrammatic recipe for their evaluation, which is capable of yielding analytic results. Our method fully specifies a combined emitter-optical state that permits investigation of light-matter entanglement generation protocols. We use our expressions to study two-photon scattering from a Λ-system and find that the pole structure of the transition amplitude is dramatically altered as the two ground states are tuned from degeneracy.

DOI: 10.1103/PhysRevA.97.043850

I. INTRODUCTION

Proposals for devices such as a measurement-based quantum computer [1] or a quantum internet [2] require large entangled states with many stationary qubit nodes. Optical photons, with their long coherence times and large velocities, form the ideal carriers of quantum information between these nodes [3], and this means that understanding the light-matter interaction is necessary for the purposes of practical device design [4]. A possible route towards engineering this light-matter interaction involves coupling quantum emitters to the modes of a nanophotonic waveguide. Recently, there has been a great deal of theoretical interest [5,6] and experimental progress in this field [7–10].

Photon scattering from a waveguide-embedded emitter is a well-studied problem, with recent developments including the single- and multiphoton scattering matrix [11,12] and generalizations of the input-output formalism and master equation [13] to waveguide systems. There has also been a substantial body of work focused on applying techniques from relativistic quantum field theory to the problem, notably the Lehmann-Symanzik-Zimmermann (LSZ) reduction formula [14], cluster decomposition principle [15], and diagrammatic evaluation of Green's functions [16]. Interest in this simple system remains high today [17,18], with many authors noting also the possibility of engineering strong on-chip photon-photon interaction [19].

Schemes for engineering entanglement between matter qubits [20,21] require the stationary qubit state conditional on that of the optical field. This is not a universal feature of previously reported techniques, although it is rendered possible by some very recent works [22,23]. We develop a formalism that fully specifies the combined emitter-optical state following photon scattering from a waveguide-embedded emitter. It is interesting to note that despite the apparent simplicity of the system, many of the previous approaches involve extremely advanced mathematical techniques and tend not to encourage an intuitive understanding of the global dynamics. This is something that the method we develop here avoids, with terms in each expression corresponding very naturally to physical processes. We can use this to visualize the processes not allowed by our initial choice of system Hamiltonian.

In this paper, we consider multiphoton scattering from a waveguide-embedded emitter and derive a method to determine analytic expressions for the transition amplitude between arbitrary combined emitter-optical input and output states. In Sec. II we describe the system under analysis and outline a general procedure for specifying the global dynamics. In Sec. III we perform this procedure explicitly for the case of a single two-level-system (TLS) with one and two-photon optical inputs. In Sec. IV we demonstrate how to extend the developed diagrammatic approach for the scenario where the TLS is replaced with a Λ-system. This allows us, in Sec. V, to study the pole structure of the transition amplitude for both cases. Sec. VI is reserved for a summary, conclusions, and some suggestions for future work.

II. WAVEGUIDE QED SYSTEM

The system analyzed in this work is shown schematically in Fig. 1 and consists of some general local system chirally coupled to the right-propagating modes a_ω of an optical waveguide [8–10]. The system is in general complex and composed of multiple subsystems; therefore the coupling is characterized by the set of numbers $\{\gamma_i\}$. At some time $t_i \to -\infty$ the system state is given by $|\phi_{\rm in};\psi_{\rm in}\rangle$, where $\psi_{\rm in}$ represents the optical wave function and $|\phi_{\rm in}\rangle$ is the state in which the emitter is prepared. A scattering event then occurs, and the global system dynamics are in general complicated to describe until a time $t_f \to +\infty$, when the emitter has relaxed to some ground or metastable level and the optical state $|\psi_{\rm out}\rangle$ is coupled out of the waveguide. Working in the interaction picture, the input and output states are eigenstates of the free Hamiltonian (H_0) that describes the dynamics of an uncoupled waveguide-emitter system [24]. This allows us to construct input and output optical states from the usual creation and annihilation operators for photons. The transition amplitude $\mathcal{A} \equiv \langle\phi_{\rm out};\psi_{\rm out}|\,\mathcal{U}\,|\phi_{\rm in};\psi_{\rm in}\rangle$ gives the overlap between an output state $|\phi_{\rm out};\psi_{\rm out}\rangle$ and

*dhurst1@sheffield.ac.uk
†p.kok@sheffield.ac.uk

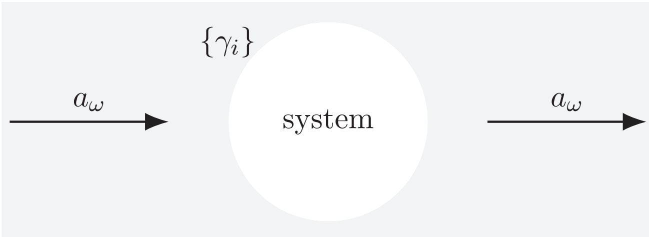

FIG. 1. Some unspecified local system interacting with the continuum of optical bosonic modes a_ω. The system may be composed of several subsystems, and thus the coupling is characterized in general by the set of rates $\{\gamma_i\}$.

an input state evolved from $t \to -\infty$ to $t \to +\infty$ by the operator $\mathcal{U}$. When $\mathcal{U}$ is the time evolution operator evaluated in the interaction picture, which in the long time limit is equivalent to the scattering matrix of the system, the transition amplitude $\mathcal{A}$ completely specifies the global system dynamics. The expansion of $\mathcal{U}$ is known as the Dyson series [25] and takes as an argument the global system interaction Hamiltonian $H_{\rm I}(t)$. This operator is defined by $H_{\rm I}(t) \equiv e^{iH_0 t} H_{\rm int} e^{-iH_0 t}$, where H_0 is the free Hamiltonian of the system, $H_{\rm int}$ the interaction Hamiltonian in the Schrödinger picture, and $\hbar = 1$. We expand the transition amplitude $\mathcal{A} \equiv \mathcal{A}^{(0)} + \mathcal{A}^{(1)} + \mathcal{A}^{(2)} + \cdots$ in terms of the Dyson series representation of $\mathcal{U}$, so that $\mathcal{A}^{(n)} \equiv \langle \phi_{\rm out}; \psi_{\rm out}| \mathcal{U}^{(n)} |\phi_{\rm in}; \psi_{\rm in}\rangle$. Using

$$\mathcal{U}^{(n)} = (-i)^n \int dt_1 \int^{t_1} dt_2 \cdots \int^{t_{n-1}} dt_n H_{\rm I}(t_1) H_{\rm I}(t_2) \cdots H_{\rm I}(t_n), \tag{1}$$

we determine that the nth-order term in the transition amplitude contains n copies of the interaction Hamiltonian. This will be an important observation in Secs. III and IV, where the interaction Hamiltonian is of Jaynes-Cummings form and conserves excitation number [26]. Here and throughout this article we adopt the convention that unspecified upper and lower integration limits correspond to ∞ and $-\infty$, respectively.

III. THE TWO-LEVEL SYSTEM

In this section we explicitly calculate the transition amplitude $\mathcal{A}$ for the scenario where the local system is a single TLS with states $\{|g\rangle, |e\rangle\}$ that are separated by the transition frequency Ω and coupled to bosonic modes of all frequencies equally at a rate γ—see Fig. 2. In Appendix A we show that the interaction Hamiltonian for this system is given by ($\hbar = 1$)

$$H_{\rm I}(t) = \gamma \int d\epsilon \, (e^{-i\Delta_\epsilon t} \sigma_+ a_\epsilon + e^{i\Delta_\epsilon t} \sigma_- a_\epsilon^\dagger), \tag{2}$$

where the waveguide's central frequency (around which we linearize the dispersion relation) is denoted by ω_0, and we define the detuning $\Delta_\epsilon \equiv \omega_0 + \epsilon - \Omega$. We assume that the TLS is prepared in the ground state $|g\rangle$ and, as $t_f \to \infty$, it is also true that $|\phi_{\rm out}\rangle = |g\rangle$.

The form of Hamiltonian (2) and our assumption of an initially and finally relaxed emitter means that the only nonzero contributions to $\mathcal{A}^{(n)}$ are those where n is even and the Pauli matrices are ordered as $\sigma_- \sigma_+ \cdots \sigma_- \sigma_+$. The general expression

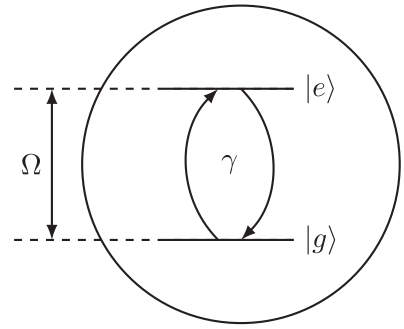

FIG. 2. A two-level system. The excited state $|e\rangle$ is separated from the ground level $|g\rangle$ by the energy gap Ω, and the two states are coupled with strength γ.

for the nth-order term in the transition amplitude is then

$$\mathcal{A}^{(n)} = (-i\gamma)^n \int d\tilde{t}^{(n)} \int d\bar{\epsilon}^{(n)} \, e^{i(\Delta_{\epsilon_1} t_1 - \Delta_{\epsilon_2} t_2 + \cdots - \Delta_{\epsilon_n} t_n)} \times \langle \psi_{\rm out}| a_{\epsilon_1}^\dagger a_{\epsilon_2} a_{\epsilon_3}^\dagger \cdots a_{\epsilon_n} |\psi_{\rm in}\rangle, \tag{3}$$

where $\int d\tilde{t}^{(n)} \equiv \int dt_1 \int^{t_1} dt_2 \cdots \int^{t_{n-1}} dt_n$ and $\int d\bar{\epsilon}^{(n)} \equiv \int d\epsilon_1 \int d\epsilon_2 \cdots \int d\epsilon_n$.

A. Single-photon scattering

We now demonstrate how to calculate the transition amplitude in Eq. (3) for the situation where a single incident photon with energy $\omega_0 + i$ scatters to an output photon of energy $\omega_0 + f$. It is simply a matter of applying the bosonic commutation relation to determine $\mathcal{A}^{(0)} = \delta(f - i)$. Consider now the nth-order term given by

$$\mathcal{A}^{(n)} = (-i\gamma)^n \int d\tilde{t}^{(n)} \int d\bar{\epsilon}^{(n)} \, e^{i\Delta_{\epsilon_1} t_1 - i\Delta_{\epsilon_2} t_2 \cdots - i\Delta_{\epsilon_n} t_n} \times \langle 0| a_f a_{\epsilon_1}^\dagger a_{\epsilon_2} \cdots a_{\epsilon_n} a_i^\dagger |0\rangle; \tag{4}$$

we can use the vacuum expectation value in Eq. (4) to eliminate the first, final, and half of the remaining frequency integrals:

$$\mathcal{A}^{(n)} = (-i\gamma)^n \int d\tilde{t}^{(n)} \int d\epsilon_3 d\epsilon_5 \cdots d\epsilon_{n-1} \times e^{i(\Delta_f t_1 - \Delta_{\epsilon_3} t_2 + \Delta_{\epsilon_3} t_3 + \cdots \Delta_i t_n)}. \tag{5}$$

The integrand in (5) can be further decomposed into its constituent Dirac delta functions, and we have then

$$\mathcal{A}^{(n)} = (-i\gamma)^n (2\pi)^{(\frac{n}{2}-1)} \int dt_1 \, e^{i\Delta_f t_1} \int^{t_1} dt_2 \times \int^{t_2} dt_3 \, \delta(t_3 - t_2) \cdots \int^{t_{n-1}} dt_n \, e^{-i\Delta_i t_n}. \tag{6}$$

Successively performing time integrals using the technique found in, e.g., Ref. [27] and reproduced here in Appendix B we arrive upon

$$\mathcal{A}^{(n)} = 2(-i\gamma)^n \delta(f - i) [\pi g(\Delta_i)]^{\frac{n}{2}}, \tag{7}$$

where we defined $g(\Delta) \equiv [\pi\delta(\Delta) + i\Delta^{-1}]$ for brevity. Summing over even n and using the binomial theorem, we

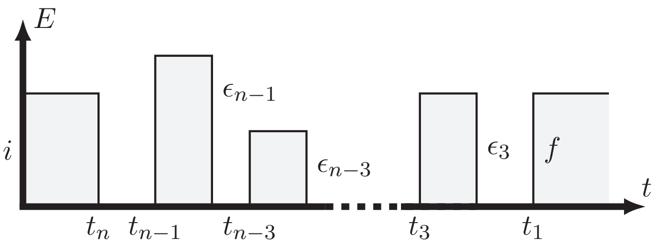

FIG. 3. Diagram for the nth-order single-photon scattering process. An incident photon of energy $\omega_0 + i$ is scattered to one of frequency $\omega_0 + f$. This occurs via the emission and absorption of $\frac{n}{2} - 1$ "internal" photons.

find

$$\mathcal{A} = \frac{1 - \gamma^2 \pi g(\Delta_i)}{1 + \gamma^2 \pi g(\Delta_i)}\, \delta(f - i) \equiv t(i)\, \delta(f - i). \tag{8}$$

Equation (8) is valid under the condition $|\pi\gamma^2 g(\Delta_i)| < 1$, which is required for application of the binomial theorem. However, in Appendix C we further use a Borel summation technique [28] to demonstrate the validity of the result for arbitrary values of $|\pi\gamma^2 g(\Delta_i)|$. Equation (8) is the first key result of this work and demonstrates that our method yields analytic expressions for the single-photon transition amplitude. In Appendix D we demonstrate its equivalence to the result of Fan *et al.* [11].

Note that it is quite easy to understand the nature of the physical process described by Eq. (5), and we have sketched it explicitly in Fig. 3. We see that the atom absorbs the original incident photon and, before emitting the outgoing photon, emits and absorbs $\frac{n}{2} - 1$ photons of frequencies $\{\epsilon_{n-1}, \epsilon_{n-3}, \ldots, \epsilon_3\}$. The energies of these 'internal' photons are uncertain, and we integrate over a continuum of possible values for each, which has the effect of reducing their duration to zero—a "pointlike" interaction.

B. Two-photon scattering

In this section we elaborate further on the diagrammatic method alluded to in Sec. III A in order to evaluate the two-photon transition amplitude. For input photons with energies $\omega_0 + i_0$ and $\omega_0 + i_1$, the nth-order term in the transition amplitude is

$$\mathcal{A}^{(n)} = (-i\gamma)^n \int d\tilde{t}^{(n)} \int d\bar{\epsilon}^{(n)}\, e^{i(\Delta_{\epsilon_1} t_1 - \Delta_{\epsilon_2} t_2 \cdots - \Delta_{\epsilon_n} t_n)} \times \langle 0|\, a_{f_0} a_{f_1} a^\dagger_{\epsilon_1} a_{\epsilon_2} \cdots a_{\epsilon_n} a^\dagger_{i_1} a^\dagger_{i_0}\, |0\rangle\,, \tag{9}$$

where f_0 and f_1 label the scattered photon frequencies. Evaluation of the vacuum expectation value in the integrand of Eq. (9) produces $2^{\frac{n}{2}+1}$ terms [29], and it is not feasible to mechanically calculate these. We instead use the physical interpretation of each term to provide further guidance.

For example, consider one of the 16 terms contributing to $\mathcal{A}^{(6)}$:

$$\mathcal{A}^{(6)}_{(1)} = -\gamma^6 \int d\tilde{t}^{(6)} \int d\omega \times e^{i(\Delta_{f_1} t_1 - \Delta_\omega (t_2 - t_3) - \Delta_{i_1} t_4 + \Delta_{f_0} t_5 - \Delta_{i_0} t_6)}, \tag{10}$$

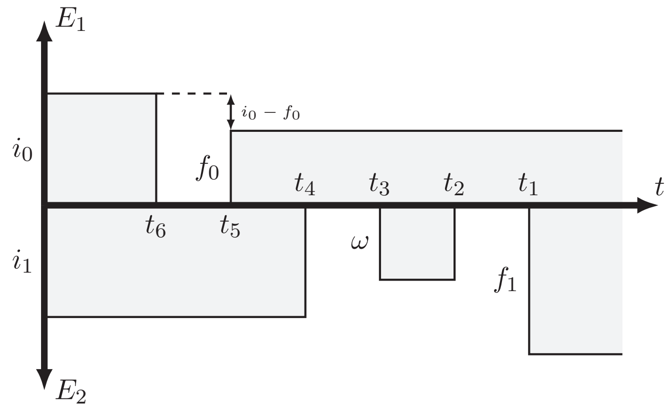

FIG. 4. Diagram for one of the possible $n = 6$ processes. Incident photons of energy $\omega_0 + i_0$ and $\omega_0 + i_1$ are scattered to energies $\omega_0 + f_0$ and $\omega_0 + f_1$. An internal photon "loop" of energy ω occurs, and ω is integrated over.

which, using exactly the same integration techniques as for the single-photon case, reduces to

$$\mathcal{A}^{(6)}_{(1)} = -2\pi^2\gamma^6 \delta(f_0 + f_1 - i_0 - i_1) \times g\big(\Delta_{i_0}\big) g\big(\Delta_{i_0} - \Delta_{f_0}\big) g\big(\Delta_{i_0} + \Delta_{i_1} - \Delta_{f_0}\big)^2. \tag{11}$$

By reassociating bosonic mode operators to their phases in the integrand of Eq. (10) we deduce that this term describes absorption by the atom of a photon with energy $\omega_0 + i_0$, prior to emission of a final $\omega_0 + f_0$ photon. Subsequently, the second incident photon is absorbed and emitted twice via an intermediate step of energy $\omega_0 + \omega$. Figure 4 gives a pictorial representation of the process, with time evolving from left to right and energies of the two populated modes relative to ω_0 given by the distance from the horizontal axis.

We can derive amplitudes in general from diagrams such as Fig. 4. By drawing the diagrams corresponding to the possible emission and absorption processes we can calculate the total transition amplitude. With each emission and absorption event in a diagram we associate a number Δ representing the difference between the total amount of absorbed radiation by the atom and the ground-excited energy gap. In Fig. 4, the atom absorbs a photon of frequency $\omega_0 + i_0$ (yielding Δ_{i_0}) and emits a photon with energy $\omega_0 + f_0$ yielding $\Delta_{i_0} - \Delta_{f_0}$ corresponding to the residual energy between the two photons. Absorbing the second incident photon produces the factor $\Delta_{i_0} + \Delta_{i_1} - \Delta_{f_0}$. These terms appear as arguments of the frequency dependent function $g(x)$ in Eq. (11), which describes the amplitude of the process depicted in Fig. 4. The "loop" indicated by ω in Fig. 4 increases the power of $g(\Delta_{i_0} + \Delta_{i_1} - \Delta_{f_0})$ by one. Finally, we impose energy conservation via $\delta(f_0 + f_1 - i_0 - i_1)$.

Suppose that for a given n we have drawn all diagrams corresponding to $\frac{n}{2}$ light-matter interaction events. Four of these diagrams (the permutations over initial and final photon frequencies) will always have one photon interacting with the emitter $\frac{n}{2}$ times, with the second photon passing through unperturbed (i.e., nonfrequency-mixing terms). These diagrams contribute amplitudes equivalent to the single-photon case. Another class of diagrams we immediately discard is

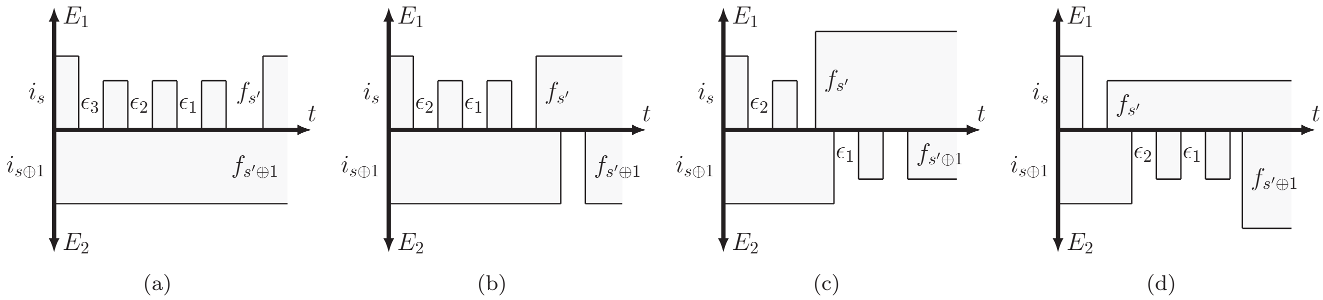

FIG. 5. The four nonzero types of diagram for the $\mathcal{A}^{(8)}$ term in the expansion of the two-photon amplitude. Panel (a) represents the nonfrequency-mixing term.

that in which an "internal" photon (such as ω in Fig. 4) is emitted at time t_m and *not* reabsorbed at $t = t_{m-1}$, since the interval $[t_m, t_{m-1}] \to 0$. We rigorously demonstrate this in Appendix E. The remaining diagrams are similar in structure to Fig. 4, with initial absorption and final emission separated by a number of internal photon loops. The structure of the integrals corresponding to these diagrams is the same as in Eq. (10) with additional frequency and time integrals corresponding to these internal loops.

The procedure for converting diagrams into $\mathcal{A}^{(n)}$ is as follows:

(i) Draw all possible diagrams with $\frac{n}{2}$ total interactions

(ii) Identify the single-photon (nonfrequency-mixing) terms

(iii) Discard the terms in which internal photons are emitted and not immediately reabsorbed

(iv) The remaining terms get the constant prefactor $\frac{2}{\pi}(i\sqrt{\pi}\gamma)^n$

(v) Each absorption event gets a factor $g(\Delta)$, where Δ corresponds to the total absorbed radiation, and each emission event gets $g(\Delta_{\text{res}})$, where Δ_{res} is the amount of absorbed radiation not reemitted

(vi) For each loop, multiply by an additional factor of $g(\Delta)$ with the same Δ as at the previous absorption

(vii) At the final emission, multiply by $\delta(f_0 + f_1 - i_0 - i_1)$.

The four species of diagram for the $n = 8$ case are shown in Fig. 5, and in Appendix F we explicitly perform this procedure to demonstrate equivalence between the diagrammatic and integral methods.

One interesting observation here is that for $n \geqslant 6$ the particular form of Eq. (2) causes vanishing of the terms with internal photon emission not immediately followed by reabsorption [step (iii) of the above outlined rules]. This behavior is due to the Hamiltonian's instantaneous coupling between the emitter and continuum of waveguide modes (without cutoff) at a constant rate. It is interesting to note that this oft-employed model makes this prediction and still agrees well with experimental data. General Hamiltonians with discretized waveguide modes would not necessarily lead to these terms vanishing. We show one of these disallowed diagrams in Fig. 6.

Let the frequency-mixing term in $\mathcal{A}^{(n)}$ for the two-photon case be given by $\delta(f_0 + f_1 - i_0 - i_1)\mathcal{M}^{(n)}$. From the above procedure we deduce that the total photon frequency-mixing term in the two-photon transition amplitude is given by $\mathcal{M} = \sum_{n=2}^{\infty} \mathcal{M}^{(n)}$ where

$$\mathcal{M}^{(n)} = \sum_{s=0,1} \sum_{s'=0,1} g(\Delta_{i_s}) g(\Delta_{i_s} - \Delta_{f_{s'}}) g(\Delta_{f_{s'\oplus1}}) \times \frac{2}{\pi}(-\pi\gamma^2)^n \sum_{k=0}^{n-2} g(\Delta_{i_s})^k g(\Delta_{f_{s'\oplus1}})^{n-2-k}. \quad (12)$$

The sum over k can be evaluated [30], and we sum over all n to find $\mathcal{M}$. Adding this to the nonfrequency-mixing component yields a final expression for the two-photon transition amplitude:

$$\begin{aligned}\mathcal{A} = {} & [t(i_0) + t(i_1) - 1] \\ & \times [\delta(f_0 - i_0)\delta(f_1 - i_1) + \delta(f_0 - i_1)\delta(f_1 - i_0)] \\ & + 2\pi\gamma^4 \delta(f_0 + f_1 - i_0 - i_1) \sum_{s=0,1} \sum_{s'=0,1} \\ & \times \frac{g(\Delta_{i_s}) g(\Delta_{i_s} - \Delta_{f_{s'}}) g(\Delta_{f_{s'\oplus1}})}{[1 + \pi\gamma^2 g(\Delta_{i_s})][1 + \pi\gamma^2 g(\Delta_{f_{s'\oplus1}})]},\end{aligned} \quad (13)$$

where $t(i)$ is defined in Eq. (8). Equation (13) is the second main result of this work and demonstrates our formalism's power to produce nonperturbative amplitudes for multiphoton processes. We demonstrate its equivalence the expression found by Fan *et al.* in Appendix D.

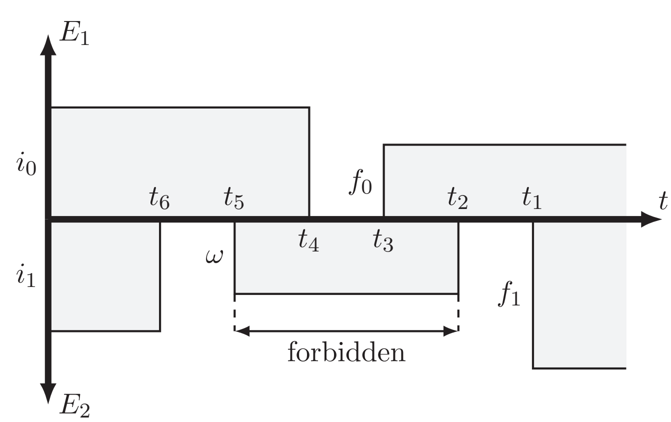

FIG. 6. Diagram for one of the impossible $n = 6$ processes. Incident photons of energy $\omega_0 + i_0$ and $\omega_0 + i_1$ are scattered to energies $\omega_0 + f_0$ and $\omega_0 + f_1$. An internal photon "loop" of energy ω occurs, and ω is integrated over.

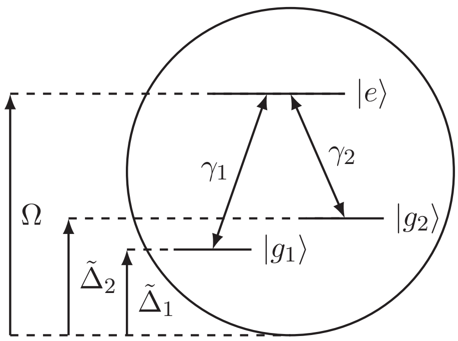

FIG. 7. A so-called Λ-system. Two ground levels $|g_1\rangle$ and $|g_2\rangle$ are coupled with amplitudes γ_1 and γ_2, respectively, to an excited state $|e\rangle$. We define a zero energy separated from $|e\rangle$ by Ω and denote the gap between $|g_i\rangle$ and zero by $\tilde{\Delta}_i$, where $i = 1,2$. In the following we assume $\Omega > \tilde{\Delta}_2 > \tilde{\Delta}_1$.

IV. Λ-SYSTEM

In many cases the perfect TLS is hard to realize, or some additional control is required over the system. This means that the emitter used in many light-matter interaction experiments has a more complex internal structure, e.g., in Refs. [31–33]. This motivates the extension of our method to a second species of local system. Consider the Λ-system shown schematically in Fig. 7. Neglecting polarization, the interaction Hamiltonian describing the dynamics of this system is readily derived [34] and given by

$$H_{\rm I}(t) = \sum_{\lambda=1}^{2} \gamma_\lambda \int d\epsilon \; e^{it\Delta_{\epsilon,\lambda}} a_\epsilon^\dagger \, |g_\lambda\rangle\langle e| + e^{-it\Delta_{\epsilon,\lambda}} a_\epsilon \, |e\rangle\langle g_\lambda| \,, \tag{14}$$

where we have defined the detuning $\Delta_{\epsilon,\lambda} = \omega_0 + \epsilon - \Omega + \tilde{\Delta}_\lambda$, again linearizing the waveguide dispersion relation about ω_0. In general, prior to and following a photon-scattering event, a Λ-system will be in some state described by $|\phi\rangle = \alpha\,|g_1\rangle + \beta\,|g_2\rangle$, as radiative transitions to each of the ground states are allowed but the $|g_1\rangle \leftrightarrow |g_2\rangle$ transitions are forbidden. In order to fully specify the dynamics, then, we need to evaluate matrix elements of the form

$$\mathcal{A}_{\mu\nu} = \langle\psi_{\rm out}; g_\mu|\,\mathcal{U}\,|\psi_{\rm in}; g_\nu\rangle \,, \tag{15}$$

where $\mu, \nu = 1,2$. Inserting Hamiltonian (14) into this expression for the transition amplitude then yields

$$\mathcal{A}_{\mu\nu}^{(n)} = (-i)^n \sum_{\{\lambda_1,\lambda_2\cdots\lambda_n\}=1}^{2} \gamma_{\lambda_1}\gamma_{\lambda_2}\cdots\gamma_{\lambda_n} \int d\tilde{t}^{(n)} \int d\bar{\epsilon}^{(n)} \times \langle\psi_{\rm out}; g_\mu| e^{it_1\Delta_{\epsilon_1,\lambda_1}} a_{\epsilon_1}^\dagger \,|g_{\lambda_1}\rangle\langle e| e^{-it_2\Delta_{\epsilon_2,\lambda_2}} a_{\epsilon_2}\,|e\rangle\,\langle g_{\lambda_2}| \cdots e^{-it_n\Delta_{\epsilon_n,\lambda_n}} a_{\epsilon_n}|e\rangle\langle g_{\lambda_n}|\,|\psi_{\rm in}; g_\nu\rangle \,, \tag{16}$$

where, at each time step, we inserted only the two terms from the Hamiltonian which either raise a ground state or lower an excited one—the two terms corresponding to the opposite behavior necessarily vanishing. This means that, again, Eq. (16) is nonzero only when n is even. The final simplification Eq. (16) permits, before requiring knowledge about the input and output optical states, utilizing the orthogonality of atomic states to eliminate $\frac{n}{2}+1$ of the sums over λ by replacing inner products between ground states with Kronecker delta functions, e.g., $\langle g_{\lambda_2}|g_{\lambda_3}\rangle = \delta_{\lambda_2\lambda_3}$.

A. Single-photon scattering

We can evaluate the amplitude of Eq. (16) for the case of single-photon scattering. We denote the input and output optical states by $|\psi_{\rm in}\rangle = |i\rangle$ and $|\psi_{\rm out}\rangle = |f\rangle$, respectively. It is simple to deduce that

$$\begin{aligned}\langle\psi_{\rm out}|\, a_{\epsilon_1}^\dagger a_{\epsilon_2}\cdots a_{\epsilon_n}\,|\psi_{\rm in}\rangle &= \langle 0|\, a_f a_{\epsilon_1}^\dagger a_{\epsilon_2}\cdots a_{\epsilon_n} a_i^\dagger\,|0\rangle \\ &= \delta(f-\epsilon_1)\delta(\epsilon_2-\epsilon_3)\cdots\delta(\epsilon_{n-2}-\epsilon_{n-1})\delta(\epsilon_n - i),\end{aligned} \tag{17}$$

and we can therefore eliminate $\frac{n}{2}+1$ of the integrals over ϵ in Eq. (16), leaving

$$\mathcal{A}_{\mu\nu}^{(n)} = (-i)^n \gamma_\mu\gamma_\nu \sum_{\{\lambda_2,\lambda_4\cdots\lambda_{n-2}\}=1}^{2} \gamma_{\lambda_2}^2\gamma_{\lambda_4}^2\cdots\gamma_{\lambda_{n-2}}^2 \int d\tilde{t}^{(n)} \times \int d\epsilon_2 \int d\epsilon_4 \cdots \int d\epsilon_{n-2} e^{it_1\Delta_{f,\mu}} e^{-it_2\Delta_{\epsilon_2,\lambda_2}} e^{it_3\Delta_{\epsilon_2,\lambda_2}} \cdots e^{it_{n-1}\Delta_{\epsilon_{n-2},\lambda_{n-2}}} e^{-it_n\Delta_{i,\nu}}. \tag{18}$$

Successively evaluating the frequency integrals in Eq. (18) in the same manner as for the TLS case, we find

$$\mathcal{A}_{\mu\nu}^{(n)} = 2(-i)^n\left[\pi\left(\gamma_1^2+\gamma_2^2\right)g(\Delta_{i,\nu})\right]^{\frac{n}{2}} \frac{\gamma_\mu\gamma_\nu}{\gamma_1^2+\gamma_2^2}\delta(\Delta_{f,\mu}-\Delta_{i,\nu}) \tag{19}$$

and again apply the binomial theorem and/or Borel summation to determine

$$\begin{aligned}\mathcal{A}_{\mu\nu} &= \delta(\Delta_{f,\mu}-\Delta_{i,\nu})\left[\delta_{\mu\nu} - \frac{2i\pi\gamma_\mu\gamma_\nu}{\Delta_{i,\nu}+i\pi\left(\gamma_1^2+\gamma_2^2\right)}\right] \\ &\equiv \delta(\Delta_{f,\mu}-\Delta_{i,\nu})[\delta_{\mu\nu}+s_{\mu\nu}(\Delta_{i\nu})].\end{aligned} \tag{20}$$

The predictions of Eq. (20) can be arrived upon via a variety of other methods, e.g., Refs. [35–38]. Specifically we see that Eqs. (23) of Ref. [37] are recovered under the transformation $\pi\gamma_i^2 \to \Gamma_i$. For a Λ-system with identical lifetimes into both ground states, i.e., $\gamma_1 = \gamma_2$, the prediction that a single resonant photon, incident upon an emitter prepared in the state $|g_1\rangle$, deterministically transfers the population to the state $|g_2\rangle$ is reproduced.

B. Two-photon scattering

We now argue that is possible to extend the diagrammatic approach used to compute the two-photon transition amplitude for the TLS to the Λ-system. In order to do this we need to demonstrate that the rules enumerated in Sec. III B continue to apply—with slight modifications specified by the added internal structure of the emitter. The first task therefore is to show that we can continue to discard terms in which internal photons are emitted and not immediately reabsorbed. These diagrams correspond to terms in the transition amplitude where an integral over the continuum of modes leads to a delta function connecting nonadjacent times. It is easy to determine that this continues to be the case by inspection of Eq. (16). We see that the structure of the time integral is not modified, and so any delta function in the integrand of the form $\delta(t_i - t_j)$, where $|i-j| > 1$, will continue to integrate to zero by the logic of Appendix E.

The nonfrequency-mixing diagrams for the Λ-system are again simple to analyze but yield a subtly different term to that found in the TLS case. This is expected [34] and related to the breaking of photon exchange symmetry, introduced by the nonunique ground states of the atomic system. Nonfrequency-mixing diagrams correspond to the four terms in the transition amplitude where, when the vacuum expectation value in Eq. (16) is evaluated, one of the creation operators for an initial photon state is commuted through one of the operators for a final state photon. This means that the structure of delta functions in the integrand of such terms is

$$\delta(f'-i')\delta(f-\epsilon_1)\delta(\epsilon_2-\epsilon_3)\delta(\epsilon_4-\epsilon_5)\cdots\delta(\epsilon_n-i), \quad (21)$$

where f,f',i and i' label the frequencies of final and initial state photons respectively. We can therefore construct the frequency-preserving portion of the transition amplitude:

$$\mathcal{N}_{\mu\nu} = \delta_{\mu\nu}[\delta(f_0-i_0)\delta(f_1-i_1)+\delta(f_0-i_1)\delta(f_1-i_0)] + \sum_{s=0,1}\sum_{s'=0,1}\delta(f_{s'\oplus 1}-i_{s\oplus 1})\delta\big(\Delta_{f_{s'}\mu}-\Delta_{i_s\nu}\big)s_{\mu\nu}\big(\Delta_{i_s\nu}\big). \quad (22)$$

Having evaluated the nonfrequency-mixing terms and those which do not contribute to the transition amplitude, the only species of terms remaining correspond to frequency-mixing processes. Applying the constraint that internal photons must be immediately reabsorbed following emission, we find that the structure of the vacuum expectation value in the integrand of frequency-mixing terms is

$$\delta(\epsilon_n-i)\delta(\epsilon_{n-1}-\epsilon_{n-2})\cdots\delta(f-\epsilon_{m+1})\delta(i'-\epsilon_m) \times\delta(\epsilon_{m-1}-\epsilon_{m-2})\cdots\delta(f'-\epsilon_1), \quad (23)$$

where m labels some point along the time evolution where one photon ceases its interaction with the emitter and the second one is absorbed. This completes our argument, as we see that again in order to calculate the nth-order term in the transition amplitude we have to sum all terms with $\frac{n}{2}$ total interactions, varying the number of times each of the initial photons interacts. Performing this procedure we calculate the nth-order frequency-mixing term

$$\mathcal{M}^{(n)}_{\mu\nu} = \frac{2}{\pi}(-\pi)^n\gamma_\mu\gamma_\nu(\gamma_1^2+\gamma_2^2)^{n-2}\sum_{s=0,1}\sum_{s'=0,1}\sum_{\lambda=1,2}\gamma_\lambda^2 \times g(\Delta_{i_s\nu})g(\Delta_{i_s\nu}-\Delta_{f_{s'}\lambda})g(\Delta_{f_{s'\oplus 1}\mu}) \times\sum_{k=0}^{n-2}g(\Delta_{i_s\nu})^{n-2-m}g(\Delta_{f_{s'\oplus 1}\mu})^m \times\delta(\Delta_{f_{s'\oplus 1}\mu}+\Delta_{f_{s'}}-\Delta_{i_{s\oplus 1}}-\Delta_{i_s\nu}), \quad (24)$$

which we see is similar in structure to Eq. (12) with an additional sum over the two possible mechanisms by which the two incident photons could now couple. After summing expression Eq. (24) over all n, adding this to the frequency-preserving term and algebraic rearrangement we find the total transition amplitude:

$$\mathcal{A}_{\mu\nu} = \mathcal{N}_{\mu\nu} + \frac{1}{2\gamma_\mu\gamma_\nu}\sum_{s,s',\lambda}\gamma_\lambda^2 s_{\mu\nu}\big(\Delta_{i_s\nu}\big)s_{\mu\nu}\big(\Delta_{f_{s'\oplus 1}\mu}\big) \times\delta\big(\Delta_{i_{s\oplus 1}\lambda}-\Delta_{f_{s'\oplus 1}\mu}\big)\delta\big(\Delta_{i_s\nu}-\Delta_{f_{s'}\lambda}\big) + \frac{i}{2\pi\gamma_\mu\gamma_\nu}\sum_{s,s',\lambda}\gamma_\lambda^2\frac{1}{\Delta_{i_s\nu}-\Delta_{f_{s'}\lambda}}s_{\mu\nu}\big(\Delta_{i_s\nu}\big) \times s_{\mu\nu}\big(\Delta_{f_{s'\oplus 1}\mu}\big)\delta\big(\Delta_{i_s\nu}-\Delta_{f_{s'\oplus 1}\mu}+\Delta_{i_{s\oplus 1}}-\Delta_{f_{s'}}\big). \quad (25)$$

The transition amplitude of Eq. (25) exactly specifies the combined emitter-optical state following the scattering of two initial photons with frequencies i_0 and i_1 on the Λ-system depicted in Fig. 7. We can use this to investigate the properties of light-matter scattering experiments, and we do this in the following section.

Fewer reported techniques exist that capture the physics of Eq. (25), compared with the single-photon case. However, methods derived from those of relativistic quantum field theory do exist as in, e.g., Ref. [39]. Here Pletyukhov and Gritsev derive an expression for the "T matrix," $T^{(2)}(\omega)$ when two photons scatter from a Λ-system. In Appendix G we demonstrate that Eq. (25) of this paper is equivalent to Eq. (46) of Ref. [39].

V. POLE STRUCTURE OF THE AMPLITUDE

Consider a two-photon scattering experiment. It is known that the properties of the scattered state are determined in large part by the pole structure of the transition amplitude [40]. In particular, poles in the complex plane of the scattered photon energy correspond to bound states of the system [41]. We might naively imagine that the pole structure of the amplitude is broadly similar whether the two photons scatter from a TLS or a Λ-system, with the added internal structure of the emitter only slightly shifting their location, for example. We can, however, demonstrate that this is not the case and that the addition of a second emitter ground state introduces a great deal of richness to the system. In Fig. 8 we consider the frequency-mixing portion of the transition amplitude of Eq. (25) and plot poles in the complex plane of f, which gives the energy of one of the scattered photons. Note that given f, the energy of the second photon is completely specified by the single energy-conserving delta function. In both Figs. 8(a) and 8(b) we drive the system with two single-frequency photons, one detuned negatively from the transition energy Ω by $\delta = 1\times 10^{14}$ rad/s and one positively by the same amount.

In Fig. 8(a) we plot the location of the poles for three different systems. Blue circles illustrate the locations of the poles for a simple TLS, with central frequency $\Omega = 2\times 10^{15}$ rad/s and coupling $\gamma = 2\times 10^4$ (rad/s)$^{\frac{1}{2}}$. This coupling strength would correspond to a lifetime of 1 ns, which is a reasonable estimate for a TLS formed by, e.g., a semiconductor quantum dot. We note the two poles at $f = \Omega \pm i\pi\gamma^2$, this is the result found by many previous authors and corresponds to the formation of a frequency-entangled pair of photons. It is interesting to ask under which circumstances the photons scattered from a Λ-system appear indistinguishable from those

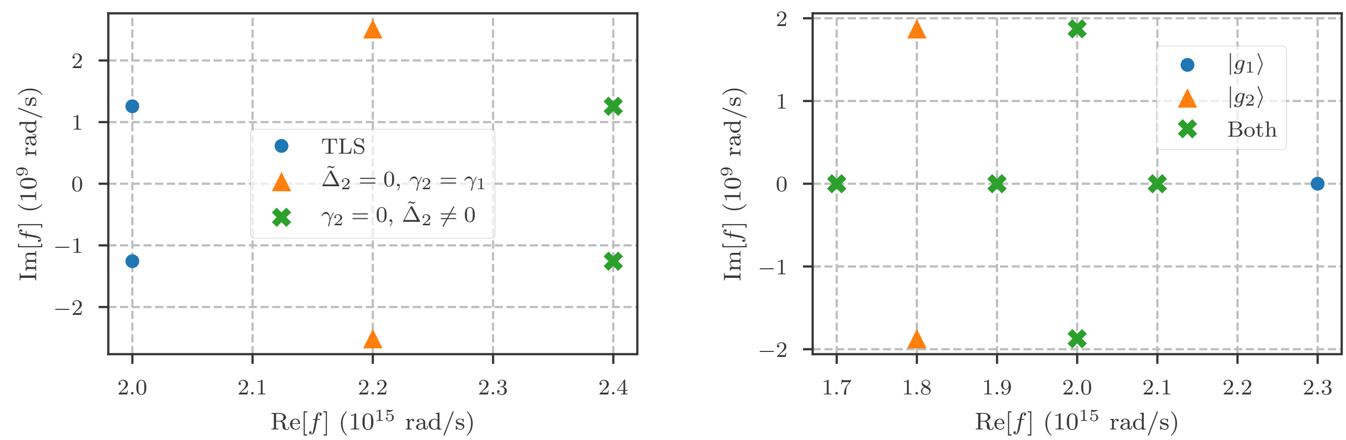

FIG. 8. Location of poles in the photon mixing component of the total transition amplitude on the complex energy plane of f—the energy of one of the scattered photons. The coupling $\gamma_1 = 2 \times 10^4$ (rad/s)$^{\frac{1}{2}}$ corresponds to a lifetime of approximately 1 ns. In both cases we drive the system with two single-frequency photons, one positively detuned from Ω by $\delta = 1 \times 10^{14}$ rad/s and one negatively detuned by the same amount. (a) TLS and special Λ-system configurations. Central angular frequencies are $\Omega = 2 \times 10^{15}$, 2.2×10^{15}, and 2.4×10^{15} rads^{-1} for systems represented by circles, triangles, and crosses respectively. The coupling $\gamma_1 = 2 \times 10^4$ (rad/s)$^{\frac{1}{2}}$. (b) Λ-system prepared initially in the state $|g_1\rangle$, with $\Omega = 2 \times 10^{15}$ rad/s, $\tilde{\Delta}_1 = 0$, and $\tilde{\Delta}_2 = \Omega/10$. We further set $\gamma_1 = 2 \times 10^4$ (rad/s)$^{\frac{1}{2}}$ and $\gamma_2 = \gamma_1/\sqrt{2}$. Some poles correspond specifically to the emitter scattering to a given state, and others are present in both cases.

scattered by a TLS. Obviously we would expect that when $\gamma_2 = 0$, for arbitrary $\tilde{\Delta}_2$, the system should behave as the TLS—photons have no access to the state $|g_2\rangle$. As a validity check of Eq. (25) we plot the poles of such as a system (with $\Omega = 2.4 \times 10^{15}$ rad/s now) using green crosses and find that this is indeed the predicted behavior. A more surprising result is indicated by the orange triangles of Fig. 8(a). Here we set $\gamma_2 = \gamma_1$, with γ_1 the same as for the TLS. We find that, when $\tilde{\Delta}_2 = 0$, the pole structure of the Λ-system is again the same as that of the TLS—though the poles are now located at $f = \Omega \pm i\pi(\gamma_1^2 + \gamma_2^2)$. This is due to the degenerate ground states appearing indistinguishable to incoming photons, and thus their only effect is a strengthening of the light-matter interaction, evidenced by shifting of the poles away from the real axis.

It is not generally true that the dynamics of scattering from a Λ-system are well approximated by the TLS. In Fig. 8(b) we consider a more general Λ-system with $\Omega = 2 \times 10^{15}$ rad/s, $\tilde{\Delta}_1 = 0$ and $\tilde{\Delta}_2 = \Omega/10$. We further assume the system is prepared initially in the lower ground state $|g_1\rangle$ and set the couplings asymmetrically so that again $\gamma_1 = 2 \times 10^4$ (rad/s)$^{\frac{1}{2}}$ but $\gamma_2 = \gamma_1/\sqrt{2}$. Now, it is important to note that the frequency-mixing component of Eq. (25) corresponds to two distinct processes. In one the emitter returns to the state $|g_1\rangle$ following the scattering, while in the other it scatters to $|g_2\rangle$. We plot both species of poles in Fig. 8(b), using blue circles and orange triangles, respectively, and also use green crosses to denote the location of poles common to both parts of the transition amplitude.

The most striking feature of Fig. 8(b) compared to Fig. 8(a) is the emergence of poles on the Im$[f] = 0$ axis of the complex plane. This means that there are now singularities in the transition amplitude corresponding to physical scattered photon energies—resonances. These occur at the frequencies of the photons input to the system, plus the input frequencies minus the energy gap $\tilde{\Delta}_2$. For the emitter state-preserving portion of the amplitude there is an additional resonance at $\Omega + \tilde{\Delta}_2 + \delta$, stemming from a process where one of the photons scatters the system to $|g_2\rangle$, with the second photon then picking up this excess energy. The final point to note is the emergence of a second pair of imaginary poles at $\Omega - \tilde{\Delta}_2 \pm i\pi(\gamma_1^2 + \gamma_2^2)$ in the portion of the transition amplitude in which the emitter is scattered to the state $|g_2\rangle$. This has a simple physical interpretation; if we were to postselect onto this state, the bound state of entangled photons that formed would have its central frequency shifted so as to conserve overall energy.

VI. CONCLUSIONS

We have developed an intuitive, diagrammatic approach to the problem of light-matter coupling in waveguide QED. In contrast to previously reported techniques, our method allows visualization of the photon-atom dynamics. We have demonstrated analytical results for both single- and two-photon input optical states for both the TLS and Λ-systems. The diagrammatic approach is straightforward to extend to higher photon number input states (though increasingly computationally expensive) and potentially more realistic Hamiltonians, and analytic results are expected to follow. Several open questions emerge from this work. For instance, how does the choice of Hamiltonian in Eq. (2) impact the transition amplitude? In particular, a waveguide will have a range of supported frequency modes defined largely by its dimensions. In theory this leads to observable consequences [42], and this would seem to suggest that some of the processes associated with forbidden diagrams actually contribute in physical systems. The limit on our method is ultimately a computational one, with an N-photon event requiring N permutations over both initial and final frequencies.

ACKNOWLEDGMENTS

D.L.H. acknowledges useful suggestions from M. E. Pearce and G. Ferenczi and is supported by an EPSRC studentship.

APPENDIX A: THE HAMILTONIAN

In this appendix we derive the interaction Hamiltonian (2) that describes a TLS coupled to an optical waveguide. We begin by dividing the total Hamiltonian into free and interacting parts, H_0 and H_{int}, respectively. The dynamics of an isolated emitter and bare waveguide are described by H_0, while the coupling between them—which we assume is of dipole form—is specified by H_{int}. We take a limit where the waveguide supports a continuum of optical modes with wave number k and apply the rotating-wave approximation. This leads to

$$\begin{aligned} H_0 &= \frac{1}{2}\Omega\sigma_z + \int_0 dk\ \omega(k)\tilde{a}_k^\dagger\tilde{a}_k, \\ H_{\text{int}} &= \tilde{\gamma}\int_0 dk\ (\sigma_+\tilde{a}_k + \tilde{a}_k^\dagger\sigma_-), \end{aligned} \tag{A1}$$

where $\omega(k)$ gives the waveguide dispersion relation and the operator $\tilde{a}_k$ destroys a photon of wave number k while obeying $[\tilde{a}_k,\tilde{a}_{k'}^\dagger] = \delta(k-k')$. We have assumed the fixed coupling rate $\tilde{\gamma}$ between optical modes of wave number k and atomic transition and adopted the convention that unspecified lower and upper integration limits imply negative and positive infinity, respectively.

It is shown by, e.g, Maier [43] that the dispersion relation for waveguide confined optical modes is surface-plasmonic. We linearize this about some central wave number k_0 so that $\tilde{\omega}(k) \approx \omega_0 + v_g(k-k_0)$, where v_g represents the photon group velocity. This means that

$$\begin{aligned} H_0 &= \frac{1}{2}\Omega\sigma_z + \int dk\ \omega_0\tilde{a}_k^\dagger\tilde{a}_k + v_g(k-k_0)\tilde{a}_k^\dagger\tilde{a}_k, \\ H_{\text{int}} &= \tilde{\gamma}\int dk\ (\sigma_+\tilde{a}_k + \tilde{a}_k^\dagger\sigma_-), \end{aligned} \tag{A2}$$

where we have also extended the limits of integration to cover the entirety of wave number space—an appropriate approximation when the band of populated modes is narrow. We next introduce the variable: $\epsilon \equiv v_g(k-k_0)$, which we use to rewrite the Hamiltonian

$$\begin{aligned} H_0 &= \frac{1}{2}\Omega\sigma_z + \int d\epsilon\ (\omega_0+\epsilon)a_\epsilon^\dagger a_\epsilon, \\ H_{\text{int}} &= \gamma\int d\epsilon\ (\sigma_+ a_\epsilon + \sigma_- a_\epsilon^\dagger), \end{aligned} \tag{A3}$$

where we have defined $\gamma \equiv v_g^{-\frac{1}{2}}\tilde{\gamma}$ and $a_\epsilon = v_g^{-\frac{1}{2}}\tilde{a}_{k_0+v_g^{-1}\epsilon}$. It can be easily shown that the commutation relation $[a_\epsilon,a_{\epsilon'}^\dagger] = \delta(\epsilon-\epsilon')$ is preserved.

At this point we can simply use the definition of the interaction Hamiltonian [24] and equation (A3) to deduce that

$$H_{\text{I}}(t) = \gamma\int d\epsilon\ (e^{-i\Delta_\epsilon t}\sigma_+ a_\epsilon + e^{i\Delta_\epsilon t}\sigma_- a_\epsilon^\dagger), \tag{A4}$$

which is the desired result, with the detuning defined by $\Delta_\epsilon \equiv \omega_0 + \epsilon - \Omega$. Eq. (A4) has the expected structure of an interaction Hamiltonian, with phases on the operators given by the energy mismatch between photons and emitter. The last point to note is the slight difference in structure between Hamiltonian (A4) and the version used by other authors (e.g., Ref. [11]). The discrepancies can be ascribed simply to our not working in a frame rotating at the waveguide's central frequency and our inclusion of the free emitter Hamiltonian in H_0 as opposed to H_{int}.

APPENDIX B: INTEGRATION TECHNIQUE

For completeness we describe here the integration technique used to evaluate the explicit integral expressions for the single- and two-photon transition amplitudes. This is a relatively well-known result and can be found in, e.g., the appendix of Ref. [27]. We define the integral $\mathcal{I}$ and begin by changing variables so as to shift the limits of integration

$$\mathcal{I} \equiv \int_{-\infty}^{t_1} dt_2\ e^{-i\Delta_i t_2} = \int_0^\infty dt_2\ e^{-i\Delta_i(t_1-t_2)}.$$

This can be decomposed and multiplied by unity to give

$$\mathcal{I} = e^{-i\Delta_i t_1}\lim_{\alpha\to 0}\int_0^\infty dt_2\ e^{-\alpha t_2}[\cos(\Delta_i t_2) + i\sin(\Delta_i t_2)], \tag{B1}$$

and we then make use of standard results [44], for example, noting

$$\delta(x) = \frac{1}{\pi}\lim_{a\to 0}\frac{a}{a^2+x^2} \tag{B2}$$

to find that

$$\begin{aligned} \mathcal{I} &= e^{-i\Delta_i t_1}\lim_{\alpha\to 0}\left(\frac{\alpha}{\alpha^2+\Delta_i^2} + i\frac{\Delta_i}{\alpha^2+\Delta_i^2}\right) \\ &= e^{-i\Delta_i t_1}\left[\pi\delta(\Delta_i) + \frac{i}{\Delta_i}\right], \end{aligned} \tag{B3}$$

which is the desired formula.

APPENDIX C: BOREL SUMMATION

In order to find the single-photon transition amplitude it is necessary to evaluate the sum

$$\sigma = \sum_{n=1}^{\infty}[-\gamma^2\pi g(\Delta_i)]^n, \tag{C1}$$

which is rendered possible for the case of $|\pi\gamma^2 g(\Delta_i)| < 1$ via the binomial theorem. Terms in the series are divergent when this condition is not satisfied, and we therefore need to take a more nuanced approach to assign a value to the sum outside of this regime. In fact, such divergent series are a common occurrence in quantum electrodynamics [45], and there are a range of methods used to extract meaning from them. The tool we utilize here is the *Borel summation*—a technique applied in a diverse range of fields [46] to analyze series with the nth term divergent up to a factor of $n!$.

We first demand that $\Delta_i \neq 0$ and find

$$\sigma = \sum_{n=0}^{\infty}\left(-\frac{i\pi\gamma^2}{\Delta_i}\right)^n. \tag{C2}$$

The Borel transformation of Eq. (C2) is defined by [47]

$$\phi(z) \equiv \sum_{n=0}^{\infty}\frac{1}{n!}(-i\pi z)^n = e^{-i\pi z} \tag{C3}$$

and the Borel sum by

$$\mathcal{B}\left(\frac{\gamma^2}{\Delta_i}\right) \equiv \int_0^\infty dt\, e^{-t}\phi\left(\frac{\gamma^2}{\Delta_i}t\right) = \frac{1}{1+i\frac{\pi\gamma^2}{\Delta_i}} \tag{C4}$$

under the condition now that $\mathrm{Im}[\frac{\pi\gamma^2}{\Delta_i}] < 1$. However, it is possible to derive the Heisenberg-Langevin equations associated with the Hamiltonian of Eq. (2), as in, e.g., Ref. [48], and in doing so we find that the emitter lifetime is directly proportional to γ^{-2}. This means that $\frac{\pi\gamma^2}{\Delta_i}$ is entirely real, and thus the condition is always satisfied. The Borel-summed result is then

$$\mathcal{A} = \delta(f-i)\frac{\Delta_i - i\pi\gamma^2}{\Delta_i + i\pi\gamma^2}, \tag{C5}$$

valid for all coupling strengths.

APPENDIX D: EQUIVALENCE TO FAN *et al.* RESULT

In this section we demonstrate the equivalence between our results for the one and two-photon transition amplitudes for a TLS and those found by Fan *et al.* [11]. As our transition amplitudes are evaluated in the limit $t \to \infty$ and the single final atomic state $|g\rangle$ is assumed, then the scattering matrix is in fact the quantity given by these amplitudes. For the single-photon case we find that

$$\mathcal{A} = \frac{1-\pi\gamma^2 g(\Delta_i)}{1+\pi\gamma^2 g(\Delta_i)}\delta(f-i). \tag{D1}$$

We can substitute our definition of $g(\Delta)$ into Eq. (D1) to determine

$$\mathcal{A} = \frac{\Delta_i - i\pi\gamma^2 - \pi^2\gamma^2\Delta_i\delta(\Delta_i)}{\Delta_i + i\pi\gamma^2 + \pi^2\gamma^2\Delta_i\delta(\Delta_i)}\delta(f-i), \tag{D2}$$

which is naturally equal to that found by Fan *et al.*:

$$\mathcal{A} = \frac{\Delta_i - i\pi\gamma^2}{\Delta_i + i\pi\gamma^2}\delta(f-i). \tag{D3}$$

The two-photon result requires a little more effort, our result is that

$$\begin{aligned}\mathcal{A} = {} & [t(i_0)+t(i_1)-1][\delta(f_0-i_0)\delta(f_1-i_1) \\ & +\delta(f_0-i_1)\delta(f_1-i_0)] \\ & +2\pi\gamma^4\delta(f_0+f_1-i_0-i_1) \\ & \times\sum_{s=0,1}\sum_{s'=0,1}\frac{g(\Delta_{i_s})g(\Delta_{i_s}-\Delta_{f_{s'}})g(\Delta_{f_{s'\oplus 1}})}{[1+\pi\gamma^2 g(\Delta_{i_s})][1+\pi\gamma^2 g(\Delta_{f_{s'\oplus 1}})]}.\end{aligned} \tag{D4}$$

Now, if we expand out the factor $g(\Delta_{i_s}-\Delta_{f_{s'}})$ so that

$$\begin{aligned}&\frac{g(\Delta_{i_s})g(\Delta_{i_s}-\Delta_{f_{s'}})g(\Delta_{f_{s'\oplus 1}})}{[1+\pi\gamma^2 g(\Delta_{i_s})][1+\pi\gamma^2 g(\Delta_{f_{s'\oplus 1}})]} \\ &= \frac{\pi g(\Delta_{i_s})\delta(\Delta_{i_s}-\Delta_{f_{s'}})g(\Delta_{f_{s'\oplus 1}})}{[1+\pi\gamma^2 g(\Delta_{i_s})][1+\pi\gamma^2 g(\Delta_{f_{s'\oplus 1}})]} \\ &\quad + \frac{i g(\Delta_{i_s})g(\Delta_{f_{s'\oplus 1}})}{(\Delta_{i_s}-\Delta_{f_{s'}})[1+\pi\gamma^2 g(\Delta_{i_s})][1+\pi\gamma^2 g(\Delta_{f_{s'\oplus 1}})]}.\end{aligned} \tag{D5}$$

It is then true that

$$\begin{aligned}\mathcal{A} = {} & [t(i_0)+t(i_1)-1][\delta(f_0-i_0)\delta(f_1-i_1)+\delta(f_0-i_1)\delta(f_1-i_0)] \\ & +4\pi^2\gamma^4[\delta(f_0-i_0)\delta(f_1-i_1)+\delta(f_0-i_1)\delta(f_1-i_0)]\frac{g(\Delta_{i_0})g(\Delta_{i_1})}{[1+\pi\gamma^2 g(\Delta_{i_0})][1+\pi\gamma^2 g(\Delta_{i_1})]} \\ & +2\pi i\gamma^4\delta(f_0+f_1-i_0-i_1)\left\{\frac{g(\Delta_{i_0})g(\Delta_{f_0})}{(\Delta_{i_0}-\Delta_{f_1})[1+\pi\gamma^2 g(\Delta_{i_0})][1+\pi\gamma^2 g(\Delta_{f_0})]}\right. \\ & +\frac{g(\Delta_{i_0})g(\Delta_{f_1})}{(\Delta_{i_0}-\Delta_{f_0})[1+\pi\gamma^2 g(\Delta_{i_0})][1+\pi\gamma^2 g(\Delta_{f_1})]}+\frac{g(\Delta_{i_0})g(\Delta_{f_1})}{(\Delta_{i_1}-\Delta_{f_0})[1+\pi\gamma^2 g(\Delta_{i_1})][1+\pi\gamma^2 g(\Delta_{f_1})]} \\ & \left.+\frac{g(\Delta_{i_1})g(\Delta_{f_0})}{(\Delta_{i_1}-\Delta_{f_1})[1+\pi\gamma^2 g(\Delta_{i_1})][1+\pi\gamma^2 g(\Delta_{f_0})]}\right\}.\end{aligned} \tag{D6}$$

We can rearrange the frequency-conserving terms and again substitute the definition of $g(\Delta)$ into the frequency-mixing term to determine

$$\begin{aligned}\mathcal{A} = {} & t(i_0)t(i_1)[\delta(f_0-i_0)\delta(f_1-i_1)+\delta(f_0-i_1)\delta(f_1-i_0)] \\ & +\frac{2\pi i\gamma^4\delta(f_0+f_1-i_0-i_1)}{[\Delta_{f_0}+i\pi\gamma^2][\Delta_{f_1}+i\pi\gamma^2]}\left\{\frac{\Delta_{f_1}+i\pi\gamma^2}{(\Delta_{f_1}-\Delta_{i_0})[\Delta_{i_0}+i\pi\gamma^2]}+\frac{\Delta_{f_0}+i\pi\gamma^2}{(\Delta_{f_0}-\Delta_{i_0})[\Delta_{i_0}+i\pi\gamma^2]}\right. \\ & \left.+\frac{\Delta_{f_0}+i\pi\gamma^2}{(\Delta_{f_0}-\Delta_{i_1})[\Delta_{i_1}+i\pi\gamma^2]}+\frac{\Delta_{f_1}+i\pi\gamma^2}{(\Delta_{f_1}-\Delta_{i_1})[\Delta_{i_1}+i\pi\gamma^2]}\right\}.\end{aligned} \tag{D7}$$

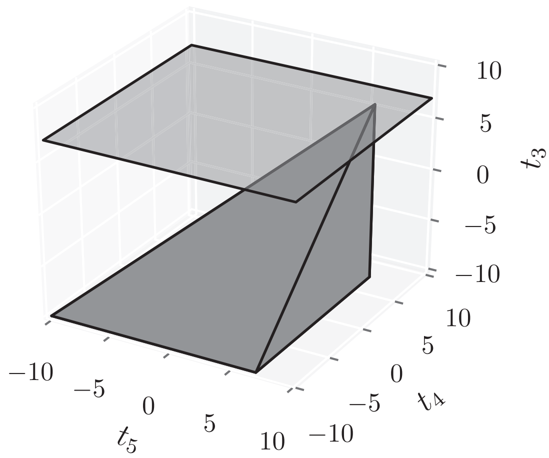

FIG. 9. Integration region defined by the enclosed volume. We see that it intersects the surface defined by $t_5 = t_2$ at only a single point.

Straightforward algebraic manipulation of Eq. (D7) then leads us to

$$\begin{aligned}\mathcal{A} &= t(i_0)t(i_1)[\delta(f_0-i_0)\delta(f_1-i_1)+\delta(f_0-i_1)\delta(f_1-i_0)]\\ &\quad+\frac{4\pi i\gamma^4\delta(f_0+f_1-i_0-i_1)}{[\Delta_{f_0}+i\pi\gamma^2][\Delta_{f_1}+i\pi\gamma^2]}\\ &\quad\times\left(\frac{1}{\Delta_{i_0}+i\pi\gamma^2}+\frac{1}{\Delta_{i_1}+i\pi\gamma^2}\right)\end{aligned} \tag{D8}$$

which is the result by Fan *et al.*

APPENDIX E: VANISHING TERMS IN HIGHER ORDER TRANSITION AMPLITUDES

In this appendix we show mathematically why diagrams with internal photon loops spanning multiple time integrals, as described in Sec. III B and shown in Fig. 6, should be discarded. If we methodically calculate $\mathcal{A}^{(6)}$ we arrive upon many terms, for example,

$$-\gamma^6\int d\tilde{t}^{(6)}\int d\omega\, e^{i(\Delta_{f_1}t_1-\Delta_\omega(t_2-t_5)+\Delta_{f_0}t_3-\Delta_{i_0}t_4-\Delta_{i_1}t_6)}. \tag{E1}$$

Evaluation of the frequency integral in this expression yields the Dirac delta function $\delta(t_5-t_2)$, and so we are evaluating an integral of the form

$$\int^{t_2}dt_3\int^{t_3}dt_4\int^{t_4}dt_5\; h(t_5,t_4,t_3)\delta(t_5-t_2), \tag{E2}$$

where $h(t_5,t_4,t_3)$ is some exponential function. The integral here is over a volume in time-space, bounded by the surfaces $t_5=t_4$ and $t_4=t_3$. The delta function has the effect of converting this volume integral into one over a surface, where the surface is defined by projection of the original volume onto $t_5=t_2$. A representation of this is depicted in Fig. 9, and we see that the resulting surface is given by a point. This term therefore does not contribute to the transition amplitude.

APPENDIX F: INTEGRAL AND DIAGRAMMATIC EVALUATION OF $\mathcal{A}^{(8)}$ FOR THE TWO-PHOTON CASE

1. Direct Integration Approach

In this appendix we demonstrate that for $n=8$ the diagrammatic and integral approaches to evaluation of the nth-order transition amplitude agree. By definition we have that

$$\begin{aligned}\mathcal{A}^{(8)} &= \gamma^8\int d\tilde{t}^{(8)}\int d\bar{\epsilon}^{(8)}\\ &\quad\times e^{i(\Delta_{\epsilon_1}t_1-\Delta_{\epsilon_2}t_2+\Delta_{\epsilon_3}t_3-\Delta_{\epsilon_4}t_4+\Delta_{\epsilon_5}t_5-\Delta_{\epsilon_6}t_6+\Delta_{\epsilon_7}t_7-\Delta_{\epsilon_8}t_8)}\\ &\quad\times\langle 0|a_{f_0}a_{f_1}a^\dagger_{\epsilon_1}a_{\epsilon_2}a^\dagger_{\epsilon_3}a_{\epsilon_4}a^\dagger_{\epsilon_5}a_{\epsilon_6}a^\dagger_{\epsilon_7}a_{\epsilon_8}a^\dagger_{i_1}a^\dagger_{i_0}|0\rangle.\end{aligned} \tag{F1}$$

The vacuum-expectation value in this expression can be directly evaluated, and we find expressions for a total of 32 terms:

$$\begin{aligned}\mathcal{A}^{(8)} &= \gamma^8\int d\tilde{t}^{(8)}\int d\epsilon_1\int d\epsilon_2\\ &\quad\times\big[e^{i(\Delta_{f_0}t_1-\Delta_{\epsilon_1}t_2+\Delta_{f_1}t_3-\Delta_{\epsilon_2}t_4+\Delta_{\epsilon_1}t_5-\Delta_{i_1}t_6+\Delta_{\epsilon_2}t_7-\Delta_{i_0}t_8)}\\ &\quad+e^{i(\Delta_{f_1}t_1-\Delta_{\epsilon_1}t_2+\Delta_{f_0}t_3-\Delta_{\epsilon_2}t_4+\Delta_{\epsilon_1}t_5-\Delta_{i_1}t_6+\Delta_{\epsilon_2}t_7-\Delta_{i_0}t_8)}\\ &\quad+\cdots\\ &\quad+e^{i(\Delta_{f_1}t_1-\Delta_{i_0}t_2+\Delta_{f_0}t_3-\Delta_{\epsilon_2}t_4+\Delta_{\epsilon_2}t_5-\Delta_{\epsilon_1}t_6+\Delta_{\epsilon_1}t_7-\Delta_{i_1}t_8)}\big],\end{aligned} \tag{F2}$$

where we have used the delta functions from the decomposed vacuum-expectation value to eliminate six of the eight frequency integrals. We can then use the definition of the Dirac delta function to transform the remaining frequency integrals and integrands into delta functions in time. Using the method outlined in Appendix E, we can then eliminate any term with a delta function connecting nonadjacent times [e.g., $\delta(t_7-t_4)$, $\delta(t_4-t_1)$, etc.], and 16 terms remain. There are, however, only four "categories" of terms, with each category containing four terms that are permutations over initial and final photon energies. We find that

$$\begin{aligned}\mathcal{A}^{(8)} &= (2\pi)^2\gamma^8\sum_{s=0,1}\sum_{s'=0,1}\int d\tilde{t}^{(8)}\big[2\pi\delta(f_{s'}-i_s)\delta(t_7-t_6)\delta(t_5-t_4)\delta(t_3-t_2)\,e^{i(\Delta_{f_{s'\oplus1}}t_1-\Delta_{i_{s\oplus1}}t_8)}\\ &\quad+\delta(t_7-t_6)\delta(t_5-t_4)e^{i(\Delta_{f_{s'}}t_1-\Delta_{i_{s\oplus1}}t_2+\Delta_{f_{s'\oplus1}}t_3-\Delta_{i_s}t_8)}+\delta(t_3-t_2)\delta(t_7-t_6)e^{i(\Delta_{f_{s'}}t_1-\Delta_{i_{s\oplus1}}t_4+\Delta_{f_{s'\oplus1}}t_5-\Delta_{i_s}t_8)}\\ &\quad+\delta(t_5-t_4)\delta(t_3-t_2)e^{i(\Delta_{f_{s'}}t_1-\Delta_{i_{s\oplus1}}t_6+\Delta_{f_{s'\oplus1}}t_7-\Delta_{i_s}t_8)}\big].\end{aligned} \tag{F3}$$

The integrals in Eq. (F3) can be evaluated directly, as in the main text for $n = 6$, and we find

$$\mathcal{A}^{(8)} = 2\pi^3\gamma^8 \sum_{s=0,1}\sum_{s'=0,1} \big[\pi g^4(\Delta_{i_s})\delta(f_{s'} - i_s)\delta(f_{s'\oplus 1} - i_{s\oplus 1}) + g^3(\Delta_{i_s})g(\Delta_{i_s} - \Delta_{f_{s'}})g(\Delta_{f_{s'\oplus 1}})\delta(f_0 + f_1 - i_0 - i_1)$$
$$+ g^2(\Delta_{i_s})g(\Delta_{i_s} - \Delta_{f_{s'}})g^2(\Delta_{f_{s'\oplus 1}})\delta(f_0 + f_1 - i_0 - i_1) + g(\Delta_{i_s})g(\Delta_{i_s} - \Delta_{f_{s'}})g^3(\Delta_{f_{s'\oplus 1}})\delta(f_0 + f_1 - i_0 - i_1)\big], \quad \text{(F4)}$$

which is the final result for the $n = 8$ term in the two-photon transition amplitude.

2. Diagrammatic Method

The diagrams corresponding to the four species of term in Eq. (F3) are shown in Fig. 5. In Fig. 5(a) a single photon is absorbed and emitted by the atom four times, in Fig. 5(b) a photon is absorbed and emitted three times, before a second photon is absorbed and emitted once. Figure 5(c) shows both photons being absorbed and emitted twice and the Fig. 5(d) shows a single absorption or emission for the first photon, followed by three for the second. Diagram 6(a) represents the nonfrequency-mixing component of the $n = 8$ term and therefore contributes a factor of $2\pi^4\gamma^8 g^4(\Delta_{i_s})\delta(f_{s'} - i_s)\delta(f_{s'\oplus 1} - i_{s\oplus 1})$ to the amplitude—this being the single-photon result multiplied by an additional delta function to impose conservation of energy for the second photon.

The three frequency-mixing diagrams require application of the rules supplied in the main text. For example, consider the diagram shown in Fig. 5(c). We first associate the prefactor $2\pi^3\gamma^8$ to this diagram's term, substituting $n = 8$ into the expression $\frac{2}{\pi}(\sqrt{\pi}\gamma)^n$ for the nth-order case. The first absorption event then yields a factor of $g(\Delta_{i_s})$ as per the rules, and we gain an additional factor of this term from the internal emission and absorption of the ϵ_1 photon. Emission of the photon with frequency $f_{s'}$ then yields the factor $g(\Delta_{i_s} - \Delta_{f_{s'}})$ before the next incident photon is absorbed, producing $g(\Delta_{i_{s\oplus 1}} + \Delta_{i_s} - \Delta_{f_{s'}})$. One additional copy of this factor is required, because of the second internal photon emission or absorption process but its argument can be simplified, as the final emission event yields the factor $\delta(f_0 + f_1 - i_0 - i_1)$, meaning that $\Delta_{i_{s\oplus 1}} + \Delta_{i_s} - \Delta_{f_{s'}} = \Delta_{f_{s'\oplus 1}}$. Multiplying individual factors together yields the expression $2\pi^3\gamma^8 g^2(\Delta_{i_s})g(\Delta_{i_s} - \Delta_{f_{s'}})g^2(\Delta_{f_{s'\oplus 1}})\delta(f_0 + f_1 - i_0 - i_1)$, exactly as found in Eq. (F4).

APPENDIX G: EQUIVALENCE OF $\mathcal{A}_{\mu\nu}$ AND $T^{(2)}(\omega)$

In Ref. [39], Pletyukhov and Gritsev derive the following expression [their Eq. (46)] for the T-matrix when two photons scatter from a Λ-system:

$$T^{(2)}(\omega) = \big[g_{31}^2 P_1 + g_{32}^2 P_2 + g_{31}g_{32}(|2\rangle\langle 1| + |1\rangle\langle 2|)\big] \times a_{\nu_1}^\dagger G_3 a_{\nu_2}\big(g_{31}^2 G_1 + g_{32}^2 G_2\big)a_{\nu_3}^\dagger G_3 a_{\nu_4}. \quad \text{(G1)}$$

Here g_{31} and g_{32} represent the ground-excited state couplings, equivalent to our γ_1 and γ_2, respectively. The states $|1\rangle$ and $|2\rangle$ are the atomic ground levels, which we labeled $|g_1\rangle$ and $|g_2\rangle$ and the operators P_1 and P_2 project onto these states. Bosonic operators are given by a_ν and propagators by G_1, G_2, and G_3.

The first step in demonstrating the equivalence between Eq. (G1) and our Eq. (25) is to apply the "intertwining property" [Eq. (15) in Ref. [39]] and rearrange bosonic operators and propagators. This has the effect of adding terms to the propagator's denominator, and we note this in the propagator argument. Note that integration over the internal photon frequencies ν is implied, and we arrive upon

$$T^{(2)}(\omega) = \big[g_{31}^2 P_1 + g_{32}^2 P_2 + g_{31}g_{32}(|2\rangle\langle 1| + |1\rangle\langle 2|)\big] \times G_3(\nu_1)\big[g_{31}^2 G_1(\nu_1 - \nu_2) + g_{32}^2 G_2(\nu_1 - \nu_2)\big] \times G_3(\nu_1 + \nu_3 - \nu_2)a_{\nu_1}^\dagger a_{\nu_2} a_{\nu_3}^\dagger a_{\nu_4}. \quad \text{(G2)}$$

Applying Wick's Theorem to the string of bosonic operators on the right-hand side of Eq. (G2), we see that there are two distinct species of term in the resulting T-matrix elements. The first is where the "internal" photon annihilation operators are contracted only with "external" photon creation operators, which we label with frequencies k_0 and k_1. In this case energies of incoming and outgoing particles are not individually preserved. In the second species of term, the operator $a_{\nu_3}^\dagger$ is contracted with a_{ν_2}. This means that an additional delta function arises from the overlap between incoming and outgoing states.

Using the definition of the T matrix in the two-photon sector, we determine that the requirement for equivalence between our result and that of Pletyukhov and Gritsev is

$$-2\pi i T_{p_0,p_1,k_0,k_1}^{(2)\mu\nu}\delta(E^{\text{out}} - E^{\text{in}}) \stackrel{?}{=} \mathcal{A}_{\mu\nu} - \mathcal{N}_{\mu\nu}, \quad \text{(G3)}$$

where $T_{p_0,p_1,k_0,k_1}^{(2)\mu\nu} \equiv \langle p_0,p_1;\mu|T^{(2)}|k_0,k_1;\nu\rangle$ and $T^{(2)}$ is defined as the on-shell evaluation of $T^{(2)}(\omega)$. We see immediately that the bound-state contributions to the left- and right-hand sides of Eq. (G3) are indeed equivalent. Consider

$$\begin{aligned}
&-2\pi i g_{3\mu}g_{3\nu}g_{31}^2 G_3^{\text{os}}G_1^{\text{os}}G_3^{\text{os}}\delta_{\nu_2 k_1}\delta_{\nu_4 k_0}\delta_{\nu_1 p_1}\delta_{\nu_3 p_0}\\
&\quad = \frac{1}{g_{3\mu}g_{3\nu}}\frac{-2\pi i g_{31}^2}{k_0 + \varepsilon_\nu - p_0 - \varepsilon_1}\frac{g_{3\mu}g_{3\nu}}{p_1 + \varepsilon_\mu - \varepsilon_3 + i\pi g^2}\\
&\qquad \times \frac{g_{3\mu}g_{3\nu}}{k_0 + \varepsilon_\nu - \varepsilon_3 + i\pi g^2}\\
&\quad = \frac{i}{2\pi\gamma_\mu\gamma_\nu}\frac{1}{k_0 +,\varepsilon_\nu - p_0 - \varepsilon_1}\frac{2\pi i\gamma_\mu\gamma_\nu}{\Delta_{p_1\mu} + i\pi\gamma^2}\frac{2\pi i\gamma_\mu\gamma_\nu}{\Delta_{k_0\nu} + i\pi\gamma^2},
\end{aligned} \quad \text{(G4)}$$

where G^{os} indicates that the propagator is to be evaluated on-shell, we made the transformations $g_{3i} \to \gamma_i$, $\varepsilon_3 \to \Omega$ and defined $\gamma^2 \equiv \gamma_1^2 + \gamma_2^2$. We see that Eq. (G4) is nothing more than one of the eight components of the bound state amplitude in Eq. (25). As we must sum over all possible permutations of initial and final photon states (and both of the propagators G_1 and G_2), the proof is complete.

It then only remains to treat the component of Eq. (G3) where individual photon energies are preserved separately.

Consider

$$-2\pi^2 g_{3\mu} g_{3\nu} g_{31}^2 G_3^{\mathrm{os}} G_3^{\mathrm{os}} \delta_{\nu_2\nu_3} \delta_{\nu_4 k_0} \delta_{\nu_1 p_0} \delta_{p_1+\mu,k_1+\varepsilon_1} = \frac{\gamma_1^2}{2\gamma_\mu\gamma_\nu} \frac{2\pi i \gamma_\mu\gamma_\nu}{\Delta_{k_0\nu} + i\pi g^2} \frac{2\pi i \gamma_\mu\gamma_\nu}{\Delta_{p_0\mu} + i\pi g^2} \delta_{p_1+\mu,k_1+\varepsilon_1}, \quad \text{(G5)}$$

where we have simply made the same transformations as for the bound-state case and also applied Eq. (16) of Ref. [39]. Again, Eq. (G5) is one of the eight components of the nonbound state portion of Eq. (25) and, owing to the sum over internal propagators, initial and final photon configurations, our main result is recovered.

[1] R. Raussendorf and H. J. Briegel, Phys. Rev. Lett. **86**, 5188 (2001).
[2] H. J. Kimble, Nature (London) **453**, 1023 (2008).
[3] A. Reiserer and G. Rempe, Rev. Mod. Phys. **87**, 1379 (2015).
[4] S. Mahmoodian, P. Lodahl, and A. S. Sørensen, Phys. Rev. Lett. **117**, 240501 (2016).
[5] P. Facchi, M. S. Kim, S. Pascazio, F. V. Pepe, D. Pomarico, and T. Tufarelli, Phys. Rev. A **94**, 043839 (2016).
[6] P. Facchi, S. Pascazio, F. V. Pepe, and K. Yuasa, J. Phys. Commun. **2**, 035006 (2018).
[7] A. Sipahigil, R. E. Evans, D. D. Sukachev, M. J. Burek, J. Borregaard, M. K. Bhaskar, C. T. Nguyen, J. L. Pacheco, H. A. Atikian, C. Meuwly, R. M. Camacho, F. Jelezko, E. Bielejec, H. Park, M. Lončar, and M. D. Lukin, Science **354**, 847 (2016).
[8] P. Lodahl, S. Mahmoodian, S. Stobbe, A. Rauschenbeutel, P. Schneeweiss, J. Volz, H. Pichler, and P. Zoller, Nature (London) **541**, 473 (2017).
[9] I. Söllner, S. Mahmoodian, S. L. Hansen, L. Midolo, A. Javadi, G. Kiršanskė, T. Pregnolato, H. El-Ella, E. H. Lee, J. D. Song, S. Stobbe, and P. Lodahl, Nat. Nano **10**, 775 (2015).
[10] R. Coles, D. Price, J. Dixon, B. Royall, E. Clarke, P. Kok, M. Skolnick, A. Fox, and M. Makhonin, Nat. Commun. **7**, 11183 EP (2016).
[11] S. Fan, Ş. E. Kocabaş, and J.-T. Shen, Phys. Rev. A **82**, 063821 (2010).
[12] S. Xu and S. Fan, Phys. Rev. A **91**, 043845 (2015).
[13] T. Caneva, M. Manzoni, T. Shi, J. Douglas, J. Cirac, and D. Chang, New J. Phys. **17**, 113001 (2015).
[14] T. Shi and C. P. Sun, Phys. Rev. B **79**, 205111 (2009).
[15] S. Xu, E. Rephaeli, and S. Fan, Phys. Rev. Lett. **111**, 223602 (2013).
[16] T. F. See, C. Noh, and D. G. Angelakis, Phys. Rev. A **95**, 053845 (2017).
[17] W. Konyk and J. Gea-Banacloche, Phys. Rev. A **93**, 063807 (2016).
[18] T. Shi, Y. Chang, and J. J. García-Ripoll, Phys. Rev. Lett. **120**, 153602 (2018).
[19] A. Nysteen, P. T. Kristensen, D. P. S. McCutcheon, P. Kaer, and J. Mørk, New J. Phys. **17**, 023030 (2015).
[20] S. D. Barrett and P. Kok, Phys. Rev. A **71**, 060310 (2005).
[21] G. Z. Cohen and L. J. Sham, Phys. Rev. B **88**, 245306 (2013).
[22] T. Shi, D. E. Chang, and J. I. Cirac, Phys. Rev. A **92**, 053834 (2015).
[23] A. Roulet and V. Scarani, New J. Phys. **18**, 093035 (2016).
[24] M. Schwartz, *Quantum Field Theory and the Standard Model* (Cambridge University Press, Cambridge, 2014).
[25] N. Quesada and J. E. Sipe, Phys. Rev. A **90**, 063840 (2014).
[26] E. T. Jaynes and F. W. Cummings, Proc. IEEE **51**, 89 (1963).
[27] A. Brańczyk, Ph.D. thesis, University of Queensland, 2010.
[28] I. M. Suslov, J. Exp. Theor. Phys. **100**, 1188 (2005).
[29] G. C. Wick, Phys. Rev. **80**, 268 (1950).
[30] M. Spivak, *Calculus*, Addison-Wesley World Student Series (Addison-Wesley, New York, 1980).
[31] I. Shomroni, S. Rosenblum, Y. Lovsky, O. Bechler, G. Guendelman, and B. Dayan, Science **345**, 903 (2014).
[32] A. Reiserer, S. Ritter, and G. Rempe, Science **342**, 1349 (2013).
[33] A. M. C. Dawes, L. Illing, S. M. Clark, and D. J. Gauthier, Science **308**, 672 (2005).
[34] S. Xu and S. Fan, Phys. Rev. A **95**, 063809 (2017).
[35] T. S. Tsoi and C. K. Law, Phys. Rev. A **80**, 033823 (2009).
[36] D. Witthaut and A. S. Sørensen, New J. Phys. **12**, 043052 (2010).
[37] M. Bradford and J.-T. Shen, Phys. Rev. A **85**, 043814 (2012).
[38] K. Koshino, K. Inomata, T. Yamamoto, and Y. Nakamura, New J. Phys. **15**, 115010 (2013).
[39] M. Pletyukhov and V. Gritsev, New J. Phys. **14**, 095028 (2012).
[40] S. Weinberg, *The Quantum Theory of Fields*, Vol. 1 (Cambridge University Press, Cambridge, 1995).
[41] J. Taylor, *Scattering Theory: The Quantum Theory of Nonrelativistic Collisions*, Dover Books on Engineering (Dover Publications, New York, 2012).
[42] J.-F. Huang, T. Shi, C. P. Sun, and F. Nori, Phys. Rev. A **88**, 013836 (2013).
[43] S. Maier, *Plasmonics: Fundamentals and Applications* (Springer, Boston, 2007), pp. 21–37.
[44] G. Arfken and H. Weber, *Mathematical Methods for Physicists, International Student Edition* (Elsevier Science, London, 2005).
[45] F. J. Dyson, Phys. Rev. **85**, 631 (1952).
[46] H. Kleinert, *Path Integrals in Quantum Mechanics, Statistics, Polymer Physics, and Financial Markets* (World Scientific, Singapore, 2009).
[47] C. Bender and S. Orszag, *Advanced Mathematical Methods for Scientists and Engineers I: Asymptotic Methods and Perturbation Theory* (Springer, New York, 1978).
[48] A. Auffèves-Garnier, C. Simon, J.-M. Gérard, and J.-P. Poizat, Phys. Rev. A **75**, 053823 (2007).