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Generalized cluster decomposition principle illustrated in waveguide quantum electrodynamics

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We show that the form of the cluster decomposition principle, commonly used in quantum field theory, needs to be significantly generalized. As an illustration, we consider the general structure of two-photon S matrix for a waveguide coupled to a local quantum system that supports multiple ground states. The presence of the multiple ground states results in a non-commutative aspect of the system with respect to the exchange of the orders of photons. Consequently, the two-photon S matrix significantly differs from the standard form in the quantum field theory.

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The cluster decomposition principle is one of the fundamental principles in quantum field theory [1]. In its general form, the cluster decomposition principle states that in a certain limit, a correlation function involving many quantum operators can be decomposed into products of correlation function involving smaller number of operators. This principle, and the possibility that it might fail, have played a significant role in theoretical physics. On one hand, this principle has been widely used to constrain the form of scattering matrix in field theory [2–4]. Moreover, it has been used to argue for the correct form of quantum vacuum in two-dimensional quantum electrodynamics (QED) and quantum chromodynamics (QCD) [5–7]. On the other hand, it has been shown that the possible failure of the cluster decomposition principle is sufficient to ensure confinement in QCD [8–13]. This observation has motivated significant efforts seeking to prove the failure of this principle in QCD. All these previous works on the cluster decomposition principle, however, are purely theoretical. And there have not been a single example that transparently illustrates the condition at which the standard form of cluster decomposition principle might fail, in a system that is experimentally accessible.

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In this letter, we introduce a generalized form of cluster decomposition principle, by considering the scattering matrix (S matrix) in two photon scattering in waveguide QED. Specialized to the S matrix, the cluster decomposition principle results in the standard form of the two-particle S-matrix [2, 14–16] as $S = S^0 + iT$, where S^0, the non-interacting part of the S matrix, is of the form

$$S^0_{p_1 p_2 k_1 k_2} = t_{k_1} t_{k_2} \left[\delta(p_1 - k_1)\delta(p_2 - k_2) + \delta(p_1 - k_2)\delta(p_2 - k_1)\right] \tag{1}$$

and contains the product of two δ functions. The T matrix, which describes the interaction, is of the form

$$T_{p_1 p_2 k_1 k_2} = \mathcal{M}_{p_1 p_2 k_1 k_2} \delta(p_1 + p_2 - k_1 - k_2) \tag{2}$$

and contains a single δ function. Here, $k_{1,2}$ and $p_{1,2}$ are the momenta of the incident and outgoing particles, respectively. t_k is the individual particle transmission amplitude and $\mathcal{M}_{p_1 p_2 k_1 k_2}$ characterizes the strength of the interactions between two particles. Recently, this form is also shown to apply in waveguide QED systems, where a few waveguide photons interact with a local quantum system [17–34].

In this letter, we show that there in fact exists a class of waveguide QED systems, in which the two-photon S matrix does not have the form of (1). The key attribute of these systems is that the local quantum system has multiple ground states. We show that this attribute results in a non-commutative aspect of the system with respect to the exchange of the orders of photons, which strongly constrains the form of the S matrix. This is in contrast to a large number of systems previously considered that have S matrix of the form shown in (1). In these systems the local quantum system has a unique ground state and hence does not have such non-commutative property.

The results here point to a much richer set of analytic properties in the structure of S matrix than previously anticipated. Also, examples of local quantum system with multiple ground states include three-level Λ-type atomic systems, which support two ground states in the electronic levels, as well as optomechanical cavities where the lowest lying photon-state manifolds contain multiple phonon sidebands. The three-level Λ-type systems play an essential role in constructing quantum memory and quantum gates for photons [35–38], whereas reaching the photon-blockade regime with optomechanical cavities has been a long-standing experimental objective in quantum optomechanics [39, 40]. Exploring the nature of photon-photon interaction in these systems in the context of waveguide QED is therefore of significance in a number of directions that are of importance for quantum optics. While there have been several calculations on the two-photon scattering properties of these systems [41–44], there have not been any discussions on the general analytic structure of the two-photon S matrix in this class of systems.

We start by considering the simplest example of a single-mode waveguide coupled to a three-level Λ-type atom as shown in Fig.1 (a). The Hamiltonian is described as

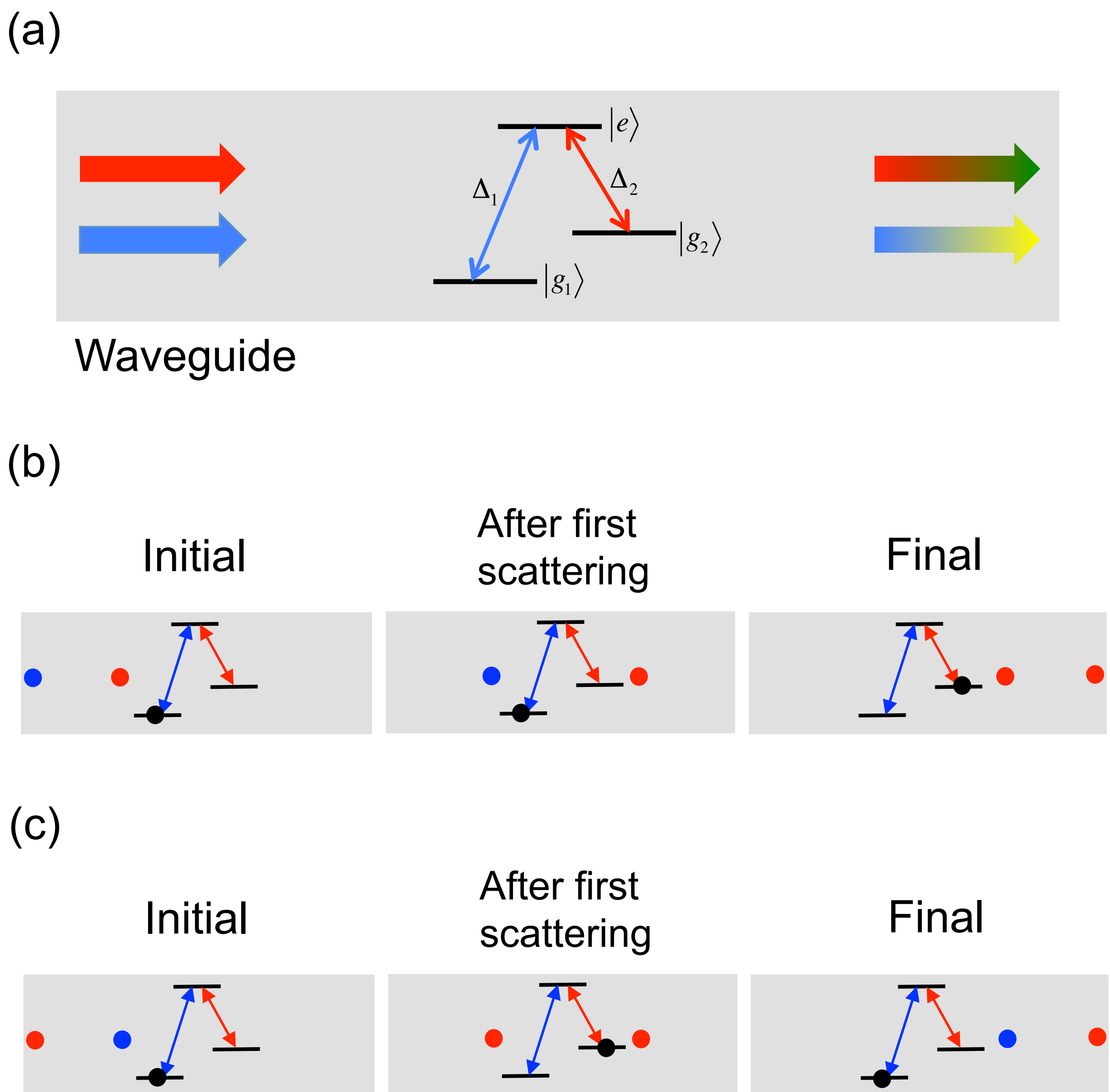

FIG. 1: (a) The system we consider: a photonic waveguide coupled to a three-level Λ-type atom. (b) A sequential scattering event where two photons incident from the left scatter against a three-level atom with $\gamma_1 = \gamma_2 \ll \Delta_1, \Delta_2$. The photons are represented by red or blue colors with different colors representing different frequencies of photons. We send in the red photon followed by the blue photon, in which the initial, intermediate and final states are shown in the subplots. (c) Another sequential scattering event that is the same as (b) except for the reverse photon ordering.

$$H = \int dk\, k\, c_k^\dagger c_k + \sum_{\lambda=1}^{2} \widetilde{\Delta}_\lambda |g_\lambda\rangle\langle g_\lambda| + \Omega|e\rangle\langle e| + \sum_{\lambda=1}^{2} \sqrt{\frac{\gamma_\lambda}{2\pi}} \int dk \left(c_k^\dagger \, |g_\lambda\rangle\langle e| + |e\rangle\langle g_\lambda| c_k \right) , \tag{3}$$

where c_k ($c_k^\dagger$) is the annihilation (creation) operator of the photon state in the waveguide. These operators satisfy the standard commutation relation $[c_k, c^\dagger_{k'}] = \delta(k-k')$. We linearize the dispersion relation and set the group velocity $v_g = 1$ so that the single photon's frequency is equal to its momentum. In our calculation, the relevant bandwidth is set by the atom-waveguide coupling strength, which is typically on the order of MHz-GHz scale. Over such frequency scale, the group velocity of an optical waveguide typically does not change significantly, and hence the linearization procedure is valid [19]. Here for simplicity we consider a waveguide consisting of only a single mode in the sense of Ref.[18]. The argument here, however, can be straightforwardly generalized to waveguides supporting multiple modes. $\widetilde{\Delta}_1$, $\widetilde{\Delta}_2$ and Ω are the respective energy of the ground states $|g_1\rangle$, $|g_2\rangle$ and the excite state $|e\rangle$ of the atom satisfying $\widetilde{\Delta}_1 < \widetilde{\Delta}_2 < \Omega$. We define $\Delta_\mu \equiv \Omega - \widetilde{\Delta}_\mu$ for $\mu = 1, 2$. The waveguide photons couple to both $|g_1\rangle - |e\rangle$ and $|g_2\rangle - |e\rangle$ transitions of the atom with respective coupling constants $\sqrt{\gamma_1/2\pi}$ and $\sqrt{\gamma_2/2\pi}$. In general we assume that $\gamma_1, \gamma_2 \ll \Delta_1, \Delta_2$. The single-photon S matrix for this system is

$$[\mathbf{S}_{pk}]_{\mu\nu} \equiv \langle p, g_\mu|S|k, g_\nu\rangle = t_{\mu\nu}(k)\,\delta(p - \Delta_\mu - k + \Delta_\nu)\,, \tag{4}$$

where μ, ν take values of $1, 2$ and

$$t_{\mu\nu}(k) = \delta_{\mu\nu} - i\frac{\sqrt{\gamma_\mu\gamma_\nu}}{k - \Delta_\nu + i\left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2}\right)} \tag{5}$$

is the transmission amplitude of the waveguide photon $|k\rangle$ when the initial and final states of the atom are $|g_\nu\rangle$ and $|g_\mu\rangle$, respectively [46–49].

We proceed to provide an intuitive argument about the structure of the two-photon S matrix. As an example, we consider a specific three-level system where $\gamma_1 = \gamma_2$. For notation simplicity, we refer photons with energy Δ_1 and Δ_2 as "blue" and "red" photons, respectively. From (30) and (5), if the atom is initially in the ground state $|g_1\rangle$, an incident blue photon will be on resonance to the atomic transition. Therefore, upon scattering against the atom, it will be converted to a red photon while the atomic state is changed to $|g_2\rangle$, whereas an incident red photon in the same situation will pass through the atom unchanged without affecting the atomic state, since it is off resonance from the atomic transition. A complementary behavior occurs when the atom is initially in the ground state $|g_2\rangle$, as can be deduced from (30) and (5).

To illustrate the structure of the non-interacting part of the S matrix, we now construct a thought experiment as shown in Fig.1 (b) and (c) by considering the outcome of two different sequential scattering events where two photons are sent toward the atom with a sufficiently large time delay between the two photons. In both events, we assume that the atom is initially in the ground state $|g_1\rangle$. In the first event (Fig.1 (b)), we send in the red photon first, it passes by the atom without interaction. The blue photon then comes in and scatters against the atom. The scattering changes the atomic state from $|g_1\rangle$ to $|g_2\rangle$, with the photon converted to red. Therefore, at the end of the two-photon scattering event, we end up with two red photons and the atom in the state $|g_2\rangle$. In the second event (Fig.1 (c)), we send in the blue photon first and then the red photon. With a similar analysis as discussed above, we can show that we will end up with the red photon first and then the blue photon, with the atomic state remaining in $|g_1\rangle$. In this system, the outcome of a two-photon scattering event depends on the order of the photons being sent in. We note that each of two different incident states above can be described by a symmetrized two-photon wavefunction. The two states are mapped to each other, not by an exchange symmetry operator, but rather by an operator $\hat{R}$ that exchanges the order of the photons. The observation above then indicates that $\left[\hat{R}, S\right] \neq 0$. Such a non-commutivity with respect to photon-order exchange operator arises from the existence of multiple ground states in the local quantum system. For local quantum system with a unique ground state, one can easily show with a similar thought experiment [34] that the outcome of the two-photon sequential scattering does not depend on the orders of the photons sent in.

The non-commutivity between the two-photon S matrix and photon-order exchange operator points to interesting aspects of the structure of two-photon S matrix. The two-photon S-matrix is typically computed with respect to a two-photon symmetrized plane wave:

$$\psi_{\text{in}}(x_1, x_2) \equiv \frac{1}{2\sqrt{2}\pi}\left(e^{ik_1x_1}e^{ik_2x_2} + e^{ik_1x_2}e^{ik_2x_1}\right). \tag{6}$$

To apply the argument above, we decompose $\psi_{\text{in}}(x_1, x_2) = \psi_{\text{in}}^{(1)}(x_1, x_2) + \psi_{\text{in}}^{(2)}(x_1, x_2)$, where

$$\psi_{\text{in}}^{(1)}(x_1, x_2) = \frac{1}{2\sqrt{2}\pi}\left[e^{ik_1x_1}e^{ik_2x_2}\,\theta(x_1 - x_2) + e^{ik_1x_2}e^{ik_2x_1}\,\theta(x_2 - x_1)\right], \tag{7}$$

$$\psi_{\text{in}}^{(2)}(x_1, x_2) = \frac{1}{2\sqrt{2}\pi}\left[e^{ik_1x_1}e^{ik_2x_2}\,\theta(x_2 - x_1) + e^{ik_1x_2}e^{ik_2x_1}\,\theta(x_1 - x_2)\right]. \tag{8}$$

With the θ functions in (7) and (8), $\psi_{\text{in}}^{(1)}(x_1, x_2)$ can be viewed as the plane wave limit of two sequential single-photon pulses with the center frequencies of the leading and the trailing pulses centering at k_1 and k_2, respectively, while $\psi_{\text{in}}^{(2)}(x_1, x_2)$ is the limit of the same two pulses but with the order of the center frequency reversed. We now consider all the scattering pathways in which the atom changes from state $|g_\nu\rangle$ to $|g_\mu\rangle$ through the two-photon sequential scattering process. For $\psi_{\text{in}}^{(1)}(x_1, x_2)$, the photon with frequency k_1 arrives first. As one of the many possible scattering pathways, upon scattering of this photon, the atom is driven from the state $|g_\nu\rangle$ to a ground state $|g_\lambda\rangle$, whereas the wavefunction of the outgoing photon takes the form of $\phi_{k_1\lambda\nu}(x_1) \equiv t_{\lambda\nu}(k_1)e^{i(k_1-\Delta_\nu+\Delta_\lambda)x_1}/\sqrt{2\pi}$. Then the photon with frequency k_2 arrives. It drives the atom from the state $|g_\lambda\rangle$ to the state $|g_\mu\rangle$, and as a result is converted to an outgoing photon with the wavefunction $\phi_{k_2,\mu\lambda}(x_2) \equiv t_{\mu\lambda}(k_2)e^{i(k_2-\Delta_\lambda+\Delta_\mu)x_2}/\sqrt{2\pi}$. Summing over all the pathways as labelled by λ, the final state associated with $\psi_{\text{in}}^{(1)}(x_1, x_2)$ is then $\psi_{\text{out}}^{(1)}(x_1, x_2) = \frac{1}{\sqrt{2}}\sum_\lambda \phi_{k_2,\mu\lambda}(x_2)\phi_{k_1,\lambda\nu}(x_1)\theta(x_1 - x_2) + [x_1 \longleftrightarrow x_2]$.

Consider both $\psi_{\rm in}^{(1)}(x_1,x_2)$ and $\psi_{\rm in}^{(2)}(x_1,x_2)$, the sequential scattering process then leads to the final state

$$\begin{aligned}\psi_{\rm out}(x_1,x_2) &= \psi_{\rm out}^{(1)}(x_1,x_2)+\psi_{\rm out}^{(2)}(x_1,x_2)\\ &= \frac{1}{2\sqrt{2}\pi}\sum_{\lambda=1}^{2} t_{\mu\lambda}(k_2)t_{\lambda\nu}(k_1)e^{i(k_2-\Delta_\lambda+\Delta_\mu)x_2}e^{i(k_1-\Delta_\nu+\Delta_\lambda)x_1}\theta(x_1-x_2)+[x_1\longleftrightarrow x_2, k_1\longleftrightarrow k_2]\ . \end{aligned} \quad (9)$$

We note that the θ functions in (9) don't compensate each other, as a direct result of the non-commutivity in the sequential scattering process. From (9), by Fourier transformation, we obtain the non-interacting part of the two-photon S matrix as

$$\begin{aligned}\left[\mathbf{S}^0_{p_1p_2k_1k_2}\right]_{\mu\nu} &\equiv \langle p_1,p_2,g_\mu|S^0|k_1,k_2,g_\nu\rangle = \frac{1}{2\sqrt{2}\pi}\int dx_1dx_2\left(e^{-ip_1x_1}e^{-ip_2x_2}+e^{-ip_1x_2}e^{-ip_1x_2}\right)\psi_{\rm out}(x_1,x_2)\\ &= \sum_{P,Q}\sum_{\lambda=1}^{2}\frac{i}{2\pi}\frac{t_{\mu\lambda}(k_{P(2)})t_{\lambda\nu}(k_{P(1)})}{p_{Q(2)}-\Delta_\mu-k_{P(2)}+\Delta_\lambda+i0^+}\delta(p_1+p_2-\Delta_\mu-k_1-k_2+\Delta_\nu)\,, \end{aligned} \quad (10)$$

where P and Q are permutation operators that act on indices $1,2$. In (10), the denominator arises from the arguments above regarding sequential scattering. When $\Delta_\mu+\Delta_\nu=2\Delta_\lambda$, one recovers the familiar form of S^0 that contains two δ functions. Here however, the S^0 contains only a single δ function. Therefore, in the sequential scattering process, the single photon energy is not conserved, if the incident wave is the symmetrized plane wave as shown in (6).

The three-level Λ-type atom we consider here exhibits the effect of electromagnetically induced transparency (EIT) when a classical field is used to couple the metastable level $|g_2\rangle$ and the excited state $|e\rangle$ [50]. In the presence of the classical field, the system is described by a Hamiltonian with a unique ground state and hence the general structure of two-photon S matrix (10) is not present [41]. Therefore, the form of the two-photon S matrix in fact can be controlled by the classical field in the Λ-type atom relevant for EIT physics. Also, experimentally, the form of the S matrix that we predict here can be probed by injecting into the system a weak coherent state and measuring the correlation function $G^{(2)}(\tau)$ as a function of incident photon energy E in the regime when the time delay τ is larger than the atom lifetime. We provide a more detailed discussion in the Appendix.

The heuristic arguments above that lead to (10) can be applied to other systems supporting multiple ground states, including optomechanical cavities [44] which also contains multiple ground states due to the phonon side bands. Here, by multiple ground states, we include the cases where the ground state manifolds contain metastable states, as long as the lifetime of these states significantly exceed the relevant interaction or scattering time-scales [45]. Consequently, instead of validating the heuristic arguments in the above specific system with the Hamiltonian (23), we prove the form (10) by computing the two-photon S matrix explicitly for a general waveguide QED system consisting of a single mode waveguide coupled to a local system

$$H = \int dk\, k\, c_k^\dagger c_k + \sqrt{\frac{\gamma}{2\pi}} \int dk \left(c_k^\dagger a + a^\dagger c_k \right) + H_{\rm sys}[a, b]\,, \tag{11}$$

where $H_{\rm sys}[a,b]$ is the Hamiltonian of the local system. a is one of the local system's operators that couples to the waveguide and b denotes its other degrees of freedom. For example, the local system can be a cavity like the James-Cummings model or the optomechanical cavity, where a is the bosonic annihilation operator of the cavity mode and b is related to the atom in the James-Cummings model or phonons in an optomechanical cavity. The local system can also be the multi-level atom such as the three-level atom in (23) with $a \equiv \sum_{\lambda=1}^{2} \sqrt{\frac{\gamma_\lambda}{\gamma}} |g_\lambda\rangle\langle e|$ as proved in the Appendix. In all these cases, one can integrate out the waveguide photons to obtain an effective Hamiltonian of the local system [30, 52, 53]

$$H_{\rm eff}[a, b] = H_{\rm sys}[a, b] - i\frac{\gamma}{2} a^\dagger a\,. \tag{12}$$

We also assume that there exits some total excitation operator of the form $\hat{N} = a^\dagger a + \hat{O}(b)$ such that $\hat{O} \geq 0$ and $\left[\hat{N}, H_{\rm eff}\right] = 0$ [55]. With such $\hat{N}$, $H_{\rm eff}[a,b]$ can be block diagonalized as

$$H_{\rm eff}\,|\lambda\rangle_N = \mathcal{E}_N^\lambda\,|\lambda\rangle_N\,, \quad {}_N\langle\bar{\lambda}|\,H_{\rm eff} = {}_N\langle\bar{\lambda}|\,\mathcal{E}_N^\lambda. \tag{13}$$

Because $H_{\rm eff}$ in (12) is non-Hermitian, its eigenvalues $\mathcal{E}_N^\lambda$ are in general complex, except for a set of ground states $|g_\lambda\rangle$ which has zero excitation and hence real eigenvalue E_0^λ. Using the input-output formalism [19, 51, 52, 54], we can compute the general single photon S matrix as

$$\left[\mathbf{S}_{pk}\right]_{\mu\nu} \equiv \langle p, g_\mu|S|k, g_\nu\rangle = t_{\mu\nu}(k)\,\delta(p + E_0^\mu - k - E_0^\nu)\,, \tag{14}$$

with

$$t_{\mu\nu}(k) = \delta_{\mu\nu} + \sum_\rho s_\nu^\rho(k)\langle g_\mu|a|\rho\rangle_1\,{}_1\langle\bar{\rho}|a^\dagger|g_\nu\rangle\,, \quad s_\nu^\rho(k) \equiv -i\frac{\gamma}{k + E_0^\nu - \mathcal{E}_1^\rho}\,. \tag{15}$$

where we insert the biorthogonal basis as defined in (13) to compute the cavity's Green function [30]. Similarly, we can compute the two-photon S matrix $\left[\mathbf{S}_{p_1p_2k_1k_2}\right]_{\mu\nu} \equiv \langle p_1, p_2, g_\mu|S|k_1, k_2, g_\nu\rangle$ as

$$\mathbf{S}_{p_1p_2k_1k_2} = \mathbf{S}^0_{p_1p_2k_1k_2} + i\,\mathbf{T}_{p_1p_2k_1k_2}\,, \tag{16}$$

where

$$\left[\mathbf{S}^0_{p_1p_2k_1k_2}\right]_{\mu\nu} \;=\; \sum_{P,Q}\sum_\lambda \frac{i}{2\pi}\,\frac{t_{\mu\lambda}(k_{P(2)})t_{\lambda\nu}(k_{P(1)})}{p_{Q(2)} + E_0^\mu - k_{P(2)} - E_0^\lambda + i0^+}\delta(p_1 + p_2 + E_0^\mu - k_1 - k_2 - E_0^\nu)\,, \tag{17}$$

$$i\left[\mathbf{T}_{p_1p_2k_1k_2}\right]_{\mu\nu} \;=\; i\left[\mathcal{M}_{p_1p_2k_1k_2}\right]_{\mu\nu}\delta(p_1+p_2+E_0^{\mu}-k_1-k_2-E_0^{\nu})\,, \tag{18}$$

$$\begin{aligned} i\left[\mathcal{M}_{p_1p_2k_1k_2}\right]_{\mu\nu} \;=\;& \frac{1}{2\pi\gamma}\sum_{\lambda\rho\sigma}\left[s_\mu^\rho(p_1)+s_\mu^\rho(p_2)\right]\left[s_\lambda^\rho(k_1)s_\nu^\sigma(k_2)+s_\lambda^\rho(k_2)s_\nu^\sigma(k_1)\right]\langle g_\mu|a|\rho\rangle_{1\,1}\langle\bar\rho|a^\dagger|g_\lambda\rangle\langle g_\lambda|a|\sigma\rangle_{1\,1}\langle\bar\sigma|a^\dagger|g_\nu\rangle \\ &+\frac{i}{2\pi}\sum_{\lambda\rho\sigma}\left[s_\mu^\rho(p_1)+s_\mu^\rho(p_2)\right]\left[s_\nu^\sigma(k_1)+s_\nu^\sigma(k_2)\right]\frac{\langle g_\mu|a|\rho\rangle_{1\,1}\langle\bar\rho|a|\lambda\rangle_{2\,2}\langle\bar\lambda|a^\dagger|\sigma\rangle_{1\,1}\langle\bar\sigma|a^\dagger|g_\nu\rangle}{k_1+k_2+E_0^\nu-\mathcal{E}_2^\lambda}\,. \end{aligned} \tag{19}$$

In the above decomposition, the T matrix (18), which describes the effect of photon-photon interaction, only contains single and two excitation poles as well as a single δ function related to the energy conservation, as required by the cluster decomposition principle [34]. The non-interacting part of S matrix (17) has the same structure as in (10), which becomes the usual direct product of two single-photon S matrix only in the cases of a single ground state or multiple degenerate ground states. In general, however, S^0 is not a direct product of the single photon S matrix.

With the two-photon S matrix (16)-(19), we now confirm the heuristic argument presented in Fig.1 by an explicit calculation. We consider the scattering event of two sequential single photon pulses spatially well separated from each other. By the identical-particle postulate the two-photon in-state has the form

$$|\,\bar k_1,\bar k_2,L,g_\nu\rangle \equiv \frac{1}{\sqrt{2}}\left[|\bar k_2\rangle\otimes e^{-i\hat pL}|\bar k_1\rangle+|\bar k_1\rangle\otimes e^{-i\hat pL}|\bar k_2\rangle\right]\otimes|g_\nu\rangle\,, \tag{20}$$

where $|\bar k\rangle=\int dk\, f_{\bar k}(k)\,|k\rangle$ describe a single photon pulse with mean momentum $\bar k$ [56]. $\hat p$ is the momentum operator and L is the spatial separation between two pulses. When L is large enough, there should be no photon-photon interaction. Indeed, one can check explicitly that (18)-(19) satisfies the requirement [34]

$$\lim_{L\to\infty} T\,|\bar k_1,\bar k_2,L,g_\nu\rangle = 0\,. \tag{21}$$

As a result, the out-state all comes from the non-interacting part of S matrix (16), that is,

$$\begin{aligned} |\mathrm{out}\rangle \;&=\; \lim_{L\to\infty} S^0\,|\bar k_1,\bar k_2,L,g_\nu\rangle \\ &=\; \lim_{L\to\infty}\frac{1}{4}\sum_\mu\int dp_1dp_2|p_1,p_2,g_\mu\rangle\int dk_1dk_2\left[\mathbf{S}^0_{p_1p_2k_1k_2}\right]_{\mu\nu}\langle k_1,k_2,g_\nu|\,\bar k_1,\bar k_2,L,g_\nu\rangle\,, \\ &=\; \frac{1}{\sqrt{2}}\sum_{\mu,\lambda}\left[|\bar k_2\rangle_{\mu\lambda}\otimes e^{-i\hat pL}|\bar k_1\rangle_{\lambda\nu}+|\bar k_1\rangle_{\mu\lambda}\otimes e^{-i\hat pL}|\bar k_2\rangle_{\lambda\nu}\right]\otimes|g_\mu\rangle\,, \end{aligned} \tag{22}$$

where $|\bar k\rangle_{\lambda\nu}\equiv\int dk\,t_{\lambda\nu}(k)f_{\bar k+E_0^\nu-E_0^\lambda}(k)|k\rangle$ describes the outgoing single photon pulse with mean momentum $\bar k+E_0^\nu-E_0^\lambda$ after scattering. By comparing the initial state (20) and the final state (22), one can see that our main result (10) indeed preserves the sequential ordering as represented by the translation operator $e^{-i\hat pL}$, and thus produces the correct result of sequential scattering that agrees with previous thought experiment.

In summary, we generalize the cluster decomposition principle and present the general structure of two-photon S matrix for a waveguide coupled to a local quantum system with multiple ground states. Such two-photon S matrix has an analytic structure that differs significantly from the standard connectedness decomposition of the two-particle S matrix in quantum field theory. We show that such a structure arises from a non-commutivity between the two-photon S matrix and an operator that exchanges photon orders. Our results here points to significant additional richness in the analytic structure of S matrix as compared to commonly anticipated. The results also provide a complete description of photon-photon interaction in several waveguide QED systems, including systems with quantum emitters with multiple ground states and systems with optomechanical cavities, that are of importance for on-chip manipulation of photon-photon interactions. Finally, the heuristic arguments we use to argue the generalized form of the cluster decomposition principle are quite general and can be applied to other three-body scattering scenarios where at least one body has an internal structure. We thus anticipate that similar S matrix structure exists in inelastic three-body scattering in other areas of physics as well.

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I. APPENDIX

We compute that two-photon S matrix associated with the specific Hamiltonian (3) explicitly and then show that the result is the same as that reduced from the general results (11)-(19).

We start by the specific Hamiltonian (3)

$$H = \int dk\, k\, c_k^\dagger\, c_k - \sum_{\lambda=1}^{2} \Delta_\lambda |g_\lambda\rangle\langle g_\lambda| + \sum_{\lambda=1}^{2} \sqrt{\frac{\gamma_\lambda}{2\pi}} \int dk \left(c_k^\dagger\, |g_\lambda\rangle\langle e| + |e\rangle\langle g_\lambda| c_k \right) . \tag{23}$$

Let $b_\lambda \equiv |g_\lambda\rangle\langle e|$ and $D_\lambda \equiv \sqrt{\gamma_1}|g_\lambda\rangle\langle g_1| + \sqrt{\gamma_2}|g_\lambda\rangle\langle g_2| - \sqrt{\gamma_\lambda}|e\rangle\langle e|$ for $\lambda = 1, 2$, the Heisenberg equations of motion are

$$\begin{aligned} \frac{d}{dt} c_k &= -i\, k\, c_k - i \sum_{\lambda=1}^{2} \sqrt{\frac{\gamma_\lambda}{2\pi}}\, b_\lambda\, , \\ \frac{d}{dt} b_\lambda &= -i\, \Delta_\lambda\, b_\lambda - i\, D_\lambda \int \frac{dk}{\sqrt{2\pi}} c_k\, . \end{aligned}$$

We then define the input and output operator as

$$\begin{aligned} c_{\text{in}}(t) &\equiv \int \frac{dk}{\sqrt{2\pi}}\, c_k(t_0)\, e^{-ik(t-t_0)}\, , \qquad t_0 \to -\infty \\ c_{\text{out}}(t) &\equiv \int \frac{dk}{\sqrt{2\pi}}\, c_k(t_1)\, e^{-ik(t-t_1)}\, , \qquad t_1 \to +\infty \end{aligned}$$

and derive the following input-output formalism from the above Heisenberg equations of motion

$$c_{\text{out}}(t) = c_{\text{in}}(t) - i\, \sqrt{\gamma_1}\, b_1(t) - i\, \sqrt{\gamma_2}\, b_2(t)\, , \tag{24}$$

$$\frac{d}{dt} b_\lambda = -i \left[\Delta_\lambda - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] b_\lambda - i\, D_\lambda\, c_{\text{in}} \tag{25}$$

$$= -i\left[\Delta_\lambda + i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)\right] b_\lambda - i\,D_\lambda\,c_{\rm out}\,. \tag{26}$$

A key property with respect to the (25)-(26) the quantum causality condition which states that

$$[b_\lambda(t), c_{\rm in}(t')] = \left[b_\lambda^\dagger(t), c_{\rm in}(t')\right] = \left[b_\lambda(t), c_{\rm in}^\dagger(t')\right] = \left[b_\lambda^\dagger(t), c_{\rm in}^\dagger(t')\right] = 0\,, \quad \text{for } t < t'\,, \tag{27}$$

$$[b_\lambda(t), c_{\rm out}(t')] = \left[b_\lambda^\dagger(t), c_{\rm out}(t')\right] = \left[b_\lambda(t), c_{\rm out}^\dagger(t')\right] = \left[b_\lambda^\dagger(t), c_{\rm out}^\dagger(t')\right] = 0\,, \quad \text{for } t > t'\,. \tag{28}$$

The N-photon S matrix can be related to the input and output operators as

$$[\mathbf{S}_{p_1\cdots p_N;k_1\cdots k_N}]_{\mu\nu} = \left(\prod_{i=1}^N \int \frac{dt_i'}{\sqrt{2\pi}} e^{ip_i t_i'} \prod_{j=1}^N \int \frac{dt_j}{\sqrt{2\pi}} e^{-ik_j t_j}\right) \langle 0, g_\mu| \prod_{i=1}^N c_{\rm out}(t_i') \prod_{j=1}^N c_{\rm in}^\dagger(t_j)|0, g_\nu\rangle\,, \tag{29}$$

and all we need to compute is the N-photon S matrix in the time domain

$$\mathbf{S}_{\mu\nu}\left(t_1'\cdots t_N'; t_1\cdots t_N\right) \equiv \langle 0, g_\mu| \prod_{i=1}^N c_{\rm out}(t_i') \prod_{j=1}^N c_{\rm in}^\dagger(t_j)|0, g_\nu\rangle\,.$$

Let $A \equiv \sqrt{\gamma_1}\,b_1 + \sqrt{\gamma_2}\,b_2$. Using the input-output relation (24) and the quantum causality condition (27)-(28), we can reduce the computation of the S matrix to the computation of the Green function consisting of only the operator A. For the single and two-photon S matrices, we have

$$\begin{aligned}
\mathbf{S}_{\mu\nu}\left(t';t\right) &= \delta(t'-t)\delta_{\mu\nu} - \langle 0, g_\mu|\mathcal{T}A(t')A^\dagger(t)|0, g_\nu\rangle\,,\\
\mathbf{S}_{\mu\nu}\left(t_1't_2';t_1t_2\right) &= \delta(t_1'-t_1)\delta(t_2'-t_2)\delta_{\mu\nu} + \delta(t_1'-t_2)\delta(t_2'-t_1)\delta_{\mu\nu}\\
&\quad -\langle 0, g_\mu|\mathcal{T}A(t_1')A^\dagger(t_1)|0, g_\nu\rangle\delta(t_2'-t_2) - \langle 0, g_\mu|\mathcal{T}A(t_2')A^\dagger(t_2)|0, g_\nu\rangle\delta(t_1'-t_1)\\
&\quad -\langle 0, g_\mu|\mathcal{T}A(t_1')A^\dagger(t_2)|0, g_\nu\rangle\delta(t_2'-t_1) - \langle 0|\mathcal{T}A(t_2')A^\dagger(t_1)|0, g_\nu\rangle\delta(t_1'-t_2)\\
&\quad +\langle 0, g_\mu|\mathcal{T}A(t_1')A(t_2')A^\dagger(t_1)A^\dagger(t_2)|0, g_\nu\rangle\,.
\end{aligned}$$

All the left is to compute the Green functions $\langle 0, g_\mu|\mathcal{T}A(t')A^\dagger(t)|0, g_\nu\rangle$ and $\langle 0, g_\mu|\mathcal{T}A(t_1')A(t_2')A^\dagger(t_1)A^\dagger(t_2)|0, g_\nu\rangle$.

Finally, the Green functions can be computed using the effective Hamiltonian of the atom

$$H_{\rm eff} = -\sum_{\lambda=1}^2 \Delta_\lambda |g_\lambda\rangle\langle g_\lambda| - i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)|e\rangle\langle e| \tag{30}$$

without involving the waveguide photon's degrees of freedom. That is, if we define

$$\widetilde{A}(t) \equiv e^{iH_{\rm eff}t}\,A\,e^{-iH_{\rm eff}t}\,, \quad \widetilde{A^\dagger}(t) \equiv e^{iH_{\rm eff}t}\,A^\dagger\,e^{-iH_{\rm eff}t}\,, \tag{31}$$

then we have

$$\langle 0, g_\mu|\mathcal{T}A(t_1')\cdots A(t_M')A^\dagger(t_1)\cdots A^\dagger(t_M)|0, g_\nu\rangle = \langle 0, g_\mu|\mathcal{T}\widetilde{A}(t_1')\cdots\widetilde{A}(t_M')\widetilde{A^\dagger}(t_1)\cdots\widetilde{A^\dagger}(t_M)|0, g_\nu\rangle\,. \tag{32}$$

Note that $A \equiv \sum_{\lambda=1}^{2} \sqrt{\gamma_\lambda}\, b_\lambda$. More generally, we have

$$\langle 0, g_\mu | \mathcal{T} b_{\rho_1}(t'_1) \cdots b_{\rho_M}(t'_M) b^\dagger_{\lambda_1}(t_1) \cdots b^\dagger_{\lambda_M}(t_M) | 0, g_\nu \rangle = \langle 0, g_\mu | \mathcal{T} \widetilde{b}_{\rho_1}(t'_1) \cdots \widetilde{b}_{\rho_M}(t'_M) \widetilde{b^\dagger}_{\lambda_1}(t_1) \cdots \widetilde{b^\dagger}_{\lambda_M}(t_M) | 0, g_\nu \rangle \quad (33)$$

with $\widetilde{b}_\lambda(t) \equiv e^{iH_{\rm eff} t}\, b_\lambda\, e^{-iH_{\rm eff} t}$, $\widetilde{b^\dagger}_\lambda(t) \equiv e^{iH_{\rm eff} t}\, b^\dagger_\lambda\, e^{-iH_{\rm eff} t}$.

The proof of (33) is as follows. Consider the derivatives with respect to some time labels t'_i and t_j, the left side of (33) satisfies

$$\begin{aligned}
\frac{\partial}{\partial t'_i}\mathrm{LHS} &= \frac{\partial}{\partial t'_i}\langle 0, g_\mu | \cdots b_{\rho_i}(t'_i) \cdots | 0, g_\nu \rangle = \langle 0, g_\mu | \cdots \frac{d b_{\rho_i}(t'_i)}{d t'_i} \cdots | 0, g_\nu \rangle \\
&= -i \left[\Delta_{\rho_i} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \langle 0, g_\mu | \cdots b_{\rho_i}(t'_i) \cdots | 0, g_\nu \rangle - i \, \langle 0, g_\mu | \cdots D_{\rho_i}(t'_i)\, c_{\rm in}(t'_i) \cdots | 0, g_\nu \rangle \\
&= -i \left[\Delta_{\rho_i} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \mathrm{LHS}\,,
\end{aligned}$$

where in the first line we reorder the order of operators as required by the time-ordering operator. The symbols "$\cdots$" before and after $b_{\rho_i}(t'_i)$ represent all the operators with time labels smaller and larger than t'_i, respectively. In the second line, we plug in the input-output formalism (25). Finally, using the quantum causality condition (27), we have $\langle 0, g_\mu | \cdots D_{\rho_i}(t'_i)\, c_{\rm in}(t'_i) \cdots | 0, g_\nu \rangle = \langle 0, g_\mu | \cdots D_{\rho_i}(t'_i) \cdots c_{\rm in}(t'_i) | 0, g_\nu \rangle = 0$. Similarly,

$$\begin{aligned}
\frac{\partial}{\partial t_j}\mathrm{LHS} &= \frac{\partial}{\partial t_j}\langle 0, g_\mu | \cdots b^\dagger_{\lambda_j}(t_j) \cdots | 0, g_\nu \rangle = \langle 0, g_\mu | \cdots \frac{d b^\dagger_{\lambda_j}(t_j)}{d t_j} \cdots | 0, g_\nu \rangle \\
&= i \left[\Delta_{\lambda_j} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \langle 0, g_\mu | \cdots b^\dagger_{\lambda_j}(t_j) \cdots | 0, g_\nu \rangle + i \, \langle 0, g_\mu | \cdots c^\dagger_{\rm out}(t_j) D^\dagger_{\lambda_j}(t_j) \cdots | 0, g_\nu \rangle \\
&= i \left[\Delta_{\lambda_j} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \mathrm{LHS}\,,
\end{aligned}$$

where we use the the input-output formalism (26) in the second line and use the quantum causality condition (28) so that $\langle 0, g_\mu | \cdots c^\dagger_{\rm out}(t_j) D^\dagger_{\lambda_j}(t_j) \cdots | 0, g_\nu \rangle = \langle 0, g_\mu | c^\dagger_{\rm out}(t_j) \cdots D^\dagger_{\lambda_j}(t_j) \cdots | 0, g_\nu \rangle = 0$. On the other hand, the right side of (33) satisfies

$$\begin{aligned}
\frac{\partial}{\partial t'_i}\mathrm{RHS} &= \frac{\partial}{\partial t'_i}\langle 0, g_\mu | \cdots \widetilde{b}_{\rho_i}(t'_i) \cdots | 0, g_\nu \rangle = \langle 0, g_\mu | \cdots \frac{d\, \widetilde{b}_{\rho_i}(t'_i)}{d t'_i} \cdots | 0, g_\nu \rangle \\
&= i \langle 0, g_\mu | \cdots e^{iH_{\rm eff} t'_i} \left[H_{\rm eff}, b_{\rho_i} \right] e^{-iH_{\rm eff} t'_i} \cdots | 0, g_\nu \rangle \\
&= -i \left[\Delta_{\rho_i} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \langle 0, g_\mu | \cdots \widetilde{b}_{\rho_i}(t'_i) \cdots | 0, g_\nu \rangle \\
&= -i \left[\Delta_{\rho_i} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \mathrm{RHS}\,, \\
\frac{\partial}{\partial t_j}\mathrm{RHS} &= \frac{\partial}{\partial t_j}\langle 0, g_\mu | \cdots \widetilde{b^\dagger}_{\lambda_j}(t_j) \cdots | 0, g_\nu \rangle = \langle 0, g_\mu | \cdots \frac{d\, \widetilde{b^\dagger}_{\lambda_j}(t_j)}{d t_j} \cdots | 0, g_\nu \rangle \\
&= i \langle 0, g_\mu | \cdots e^{iH_{\rm eff} t_j} \left[H_{\rm eff}, b^\dagger_{\lambda_j} \right] e^{-iH_{\rm eff} t_j} \cdots | 0, g_\nu \rangle \\
&= i \left[\Delta_{\lambda_j} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \langle 0, g_\mu | \cdots \widetilde{b^\dagger}_{\lambda_j}(t_j) \cdots | 0, g_\nu \rangle \\
&= i \left[\Delta_{\lambda_j} - i \left(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} \right) \right] \mathrm{RHS}\,,
\end{aligned}$$

where we use the definition of $\widetilde{b}_{\rho_i}(t'_i)$, $\widetilde{b^\dagger}_{\lambda_j}(t_j)$ as well as the effective Hamiltonian (30) and then compute their commutators explicitly. Therefore, both the left and right side of (33) satisfy the same set of partial differential equations with respect to t'_i and t_j for any $1 \le i, j \le M$. As a result, (33) holds and so does (32).

With all the above preparations, we are already to compute the single and two-photon S matrices. We start by computing the two- and four-point Green functions as

$$\begin{aligned} G_{\mu\nu}(t';t) &\equiv \langle 0, g_\mu|\mathcal{T}A(t')A^\dagger(t)|0, g_\nu\rangle = \langle 0, g_\mu|\mathcal{T}\widetilde{A}(t')\widetilde{A^\dagger}(t)|0, g_\nu\rangle \\ &= \sum_{\rho,\lambda=1}^{2} \sqrt{\gamma_\rho\gamma_\lambda}\,\langle 0, g_\mu|\widetilde{b}_\rho(t')\widetilde{b^\dagger}_\lambda(t)|0, g_\nu\rangle\theta(t'-t) \\ &= \sqrt{\gamma_\mu\gamma_\nu}\,\langle 0, g_\mu|e^{iH_{\text{eff}}t'}\widetilde{b}_\mu e^{-iH_{\text{eff}}(t'-t)}\widetilde{b^\dagger}_\nu e^{-iH_{\text{eff}}t}|0, g_\nu\rangle\theta(t'-t) \\ &= \sqrt{\gamma_\mu\gamma_\nu}e^{-i\Delta_\mu t'+i\Delta_\nu t}e^{-(\frac{\gamma_1}{2}+\frac{\gamma_2}{2})(t'-t)}\theta(t'-t)\,, \end{aligned}$$

and

$$\begin{aligned} &\langle 0, g_\mu|\mathcal{T}A(t'_1)A(t'_2)A^\dagger(t_1)A^\dagger(t_2)|0, g_\nu\rangle = \langle 0, g_\mu|\mathcal{T}\widetilde{A}(t'_1)\widetilde{A}(t'_2)\widetilde{A^\dagger}(t_1)\widetilde{A^\dagger}(t_2)|0, g_\nu\rangle \\ &= \sqrt{\gamma_\mu\gamma_\nu}\sum_{P,Q}\sum_{\lambda=1}^{2}\gamma_\lambda\langle 0, g_\mu|\widetilde{b}_\mu\left(t'_{P(1)}\right)\widetilde{b^\dagger}_\lambda\left(t_{Q(1)}\right)\widetilde{b}_\lambda\left(t'_{P(2)}\right)\widetilde{b^\dagger}_\nu\left(t_{Q(2)}\right)|0, g_\nu\rangle\theta\left(t'_{P(1)}-t_{Q(1)}\right)\theta\left(t_{Q(1)}-t'_{P(2)}\right)\theta\left(t'_{P(2)}-t_{Q(2)}\right) \\ &= \sum_{P,Q}\sum_{\lambda=1}^{2} G_{\mu\lambda}(t'_{P(1)};t_{Q(1)})G_{\lambda\nu}(t'_{P(2)};t_{Q(2)})\theta\left(t_{Q(1)}-t'_{P(2)}\right)\,, \end{aligned}$$

where P, Q are permutations over indices $1, 2$. By Fourier transformation, we have

$$[\mathbf{G}_{pk}]_{\mu\nu} \equiv \int \frac{dt'}{\sqrt{2\pi}}e^{ipt'}\frac{dt}{\sqrt{2\pi}}e^{-ikt}\,G_{\mu\nu}(t';t) = \frac{i\sqrt{\gamma_\mu\gamma_\nu}}{k-\Delta_\nu+i(\frac{\gamma_1}{2}+\frac{\gamma_2}{2})}\delta(p-\Delta_\mu-k+\Delta_\nu)\,.$$

As a result, the single photon S matrix is

$$[\mathbf{S}_{pk}]_{\mu\nu} = \delta(p-k)\delta_{\mu\nu} - [\mathbf{G}_{pk}]_{\mu\nu} = t_{\mu\nu}(k)\,\delta(p-\Delta_\mu-k+\Delta_\nu)\,,$$

where $t_{\mu\nu}(k)$ is exactly the same as (5). For two-photon S matrix,

$$\begin{aligned} [\mathbf{S}_{p_1p_2k_1k_2}]_{\mu\nu} &= \frac{\delta_{\mu\nu}}{2}\sum_{P,Q}\delta\left(p_{Q(1)}-k_{P(1)}\right)\delta\left(p_{Q(2)}-k_{P(2)}\right) - \sum_{P,Q}\left[\mathbf{G}_{p_{Q(1)}k_{P(1)}}\right]_{\mu\nu}\delta\left(p_{Q(2)}-k_{P(2)}\right) \\ &+\sum_{P,Q}\sum_{\lambda=1}^{2}\int\frac{dq}{\sqrt{2\pi}}\frac{dl}{\sqrt{2\pi}}\frac{i\left[\mathbf{G}_{p_{Q(2)}q}\right]_{\mu\lambda}\left[\mathbf{G}_{lk_{P(1)}}\right]_{\mu\lambda}}{l-p_{Q(1)}+i0^+}\delta\left(p_{Q(1)}+q-l-k_{P(2)}\right)\,. \end{aligned}$$

Submitting the expression of $[\mathbf{G}_{pk}]_{\mu\nu}$ leads to the final result:

$$[\mathbf{S}_{p_1p_2k_1k_2}]_{\mu\nu} = \left[\mathbf{S}^0_{p_1p_2k_1k_2}\right]_{\mu\nu} + i\,[\mathcal{M}_{p_1p_2k_1k_2}]_{\mu\nu}\,\delta(p_1+p_2-\Delta_\mu-k_1-k_2+\Delta_\nu)\,, \tag{34}$$

where

$$\left[\mathbf{S}^0_{p_1p_2k_1k_2}\right]_{\mu\nu} = \sum_{P,Q}\sum_{\lambda=1}^{2}\frac{i}{2\pi}\frac{t_{\mu\lambda}(k_{P(2)})t_{\lambda\nu}(k_{P(1)})}{p_{Q(2)}-\Delta_\mu-k_{P(2)}+\Delta_\lambda+i0^+}\delta(p_1+p_2-\Delta_\mu-k_1-k_2+\Delta_\nu)\,, \tag{35}$$

$$\begin{aligned} i\left[\mathcal{M}_{p_1p_2k_1k_2}\right]_{\mu\nu} &= \frac{i}{2\pi}\sqrt{\gamma_\mu\gamma_\nu}\left(\frac{1}{p_1-\Delta_\mu+i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)}+\frac{1}{p_2-\Delta_\mu+i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)}\right)\times \\ &\sum_{P}\sum_{\lambda=1}^{2}\gamma_\lambda\frac{1}{k_{P(1)}-\Delta_\lambda+i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)}\frac{1}{k_{P(2)}-\Delta_\nu+i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)}\,. \end{aligned} \tag{36}$$

(35) is exactly the same as that obtained by our heuristic arguments.

Now we check that our general result (11)-(19) can give the correct result of the two-photon S matrix for three-level atom as computed above. Applying the general Hamiltonian (11) to the specific Hamiltonian (3), we have

$$a=\sum_{\lambda=1}^{2}\sqrt{\frac{\gamma_\lambda}{\gamma}}|g_\lambda\rangle\langle e|\,. \tag{37}$$

The resulting effective Hamiltonian from (12) is

$$H_{\text{eff}}=\sum_{\lambda=1}^{2}\widetilde{\Delta}_\lambda|g_\lambda\rangle\langle g_\lambda|+\Omega|e\rangle\langle e|-i\frac{\gamma}{2}a^\dagger a \;=\; \sum_{\lambda=1}^{2}\widetilde{\Delta}_\lambda|g_\lambda\rangle\langle g_\lambda|+\left[\Omega-i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)\right]|e\rangle\langle e|\,, \tag{38}$$

which can be diagonalized as

$$H_{\text{eff}}\,|g_1\rangle=\widetilde{\Delta}_1|g_1\rangle\,,\qquad H_{\text{eff}}\,|g_2\rangle=\widetilde{\Delta}_2|g_2\rangle\,,\qquad H_{\text{eff}}\,|e\rangle=\left[\Omega-i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)\right]|e\rangle\,.$$

In other word, we have $E_0^\mu\equiv\widetilde{\Delta}_\mu$ associated with the ground state $|g_\mu\rangle$ for $\mu=1,2$ and $\mathcal{E}_1^\rho\equiv\Omega-i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)$ together with $|\rho\rangle_1\equiv|e\rangle$, ${}_1\langle\bar{\rho}|\equiv\langle e|$ for $\rho=1$. In this special case, the excitation number cannot be larger than one and the second line of (19) vanishes. Furthermore,

$$\langle g_\mu|a|\rho\rangle_1=\langle\bar{\rho}|a|g_\mu\rangle_1=\sqrt{\frac{\gamma_\mu}{\gamma}}\,,\qquad s^\rho_\mu(k)=-i\frac{\gamma}{k+\widetilde{\Delta}_\mu-\Omega+i\left(\frac{\gamma_1}{2}+\frac{\gamma_2}{2}\right)} \tag{39}$$

for $\mu=1,2$ and $\rho=1$. Submitting (39) into (17)-(19) , the general results (17)-(19) indeed reduce to the form of (34)-(36).

Finally, we consider the two-photon correlation function that describes the photon statistics of the outgoing two-photon state. Without loss of generality, we consider an incident two-photon plane-wave state $|k_1,k_2,g_\nu\rangle$ comprised of two photons with individual frequencies k_1 and k_2, as described by

$$|k_1,k_2,g_\nu\rangle=\int dx_1dx_2P_{k_1k_2}\left(x_1,x_2\right)\frac{1}{\sqrt{2}}c^\dagger(x_1)c^\dagger(x_2)|0,g_\nu\rangle\,,$$

where $P_{k_1k_2}\left(x_1,x_2\right)=\frac{1}{\sqrt{2}2\pi}\left[e^{ik_1x_1}e^{ik_2x_2}+e^{ik_1x_2}e^{ik_2x_1}\right]$ is a symmetrized two-photon plane wave. With the two-photon S matrix (34)-(36), the outgoing state with the atom state $|g_\mu\rangle$ can be computed as $|\text{out}\rangle_\mu=$

$\frac{1}{2}\int dp_1 dp_2 \left[\mathbf{S}_{p_1p_2k_1k_2}\right]_{\mu\nu} |p_1, p_2, g_\mu\rangle$ and the two-photon correlation function associated with this outgoing state is

$$G^{(2)}(\tau) = {}_\mu\langle \text{out}|\, c(y)c(y+\tau)c^\dagger(y+\tau)c^\dagger(y)\, |\text{out}\rangle_\mu \,. \tag{40}$$

For large τ, the interacting part of the S matrix has no contributions. Only the non-interacting part S^0 matters. That is, for large τ, we can compute (40) using

$$|\text{out}\rangle_\mu = \frac{1}{2}\int dp_1 dp_2 \left[\mathbf{S}^0_{p_1p_2k_1k_2}\right]_{\mu\nu} |p_1, p_2, g_\mu\rangle \,. \tag{41}$$

Therefore, experimentally we can validate our main result (35) by measuring the $G^{(2)}(\tau)$ when τ is very large, as shown in Fig.2. Our main result (35) differs from the commonly anticipated disconnected form

$$\left[\mathbf{S}^{\text{disconnected}}_{p_1p_2k_1k_2}\right]_{\mu\nu} = \frac{1}{2}\sum_{\lambda=1}^{2}\left\{\left[\mathbf{S}_{p_1k_1}\right]_{\mu\lambda}\left[\mathbf{S}_{p_2k_2}\right]_{\lambda\nu} + \left[\mathbf{S}_{p_2k_1}\right]_{\mu\lambda}\left[\mathbf{S}_{p_1k_2}\right]_{\lambda\nu} + \left[\mathbf{S}_{p_1k_2}\right]_{\mu\lambda}\left[\mathbf{S}_{p_2k_1}\right]_{\lambda\nu} + \left[\mathbf{S}_{p_2k_2}\right]_{\mu\lambda}\left[\mathbf{S}_{p_1k_1}\right]_{\lambda\nu}\right\}$$

when $\Delta_1 \neq \Delta_2$, leading to a qualitative difference on $G^{(2)}(\tau)$.

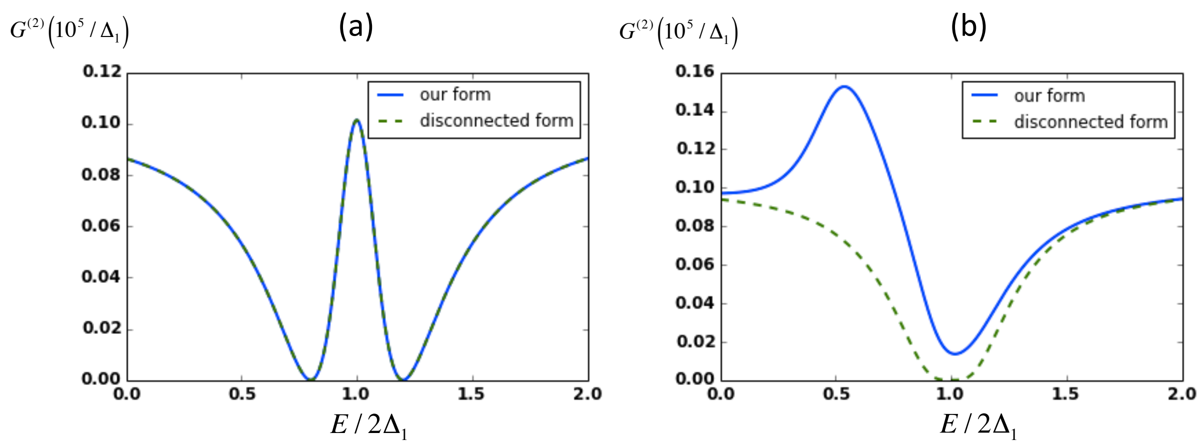

FIG. 2: The two-photon correlation function $G^{(2)}(\tau = 10^5/\Delta_1)$ as a function of the two photons' total energy E when $k_1 = k_2 = E/2$ and $\gamma_1 = \gamma_2 = \Delta_1/5$. In both cases, the initial and final states of the atom are the ground state $|g_1\rangle$. (a) $\Delta_2 = \Delta_1$. In this case, our result (35) is the same as the commonly anticipated disconnected form and thus the two-photon correlation functions are identical. (b) $\Delta_2 = \Delta_1/2$. In this case, our main result (35) leads to a significant 'bump' in the two-photon correlation function as compared to the disconnected form.

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[55] For example, $\hat{N} = a^\dagger a + \sigma_z/2$ in the Jaynes-Cummings model and $\hat{N} = a^\dagger a$ in the optomechanical cavity.

[56] For example, we could take the envelop of the pulse to be Lorentzian like $f_{\bar{k}}(k) = \frac{\alpha}{\pi(\alpha^2+(k-\bar{k})^2)}$.