# Scattering of Few Photons by a Ladder-type Quantum System 

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#### Abstract

We consider the inelastic scattering of few photons by a ladder-type quantum system residing in a semi-infinite waveguide. Two scenarios, namely 2 -photon scattering and 3-photon scattering, are investigated. The single photons are described by continuous temporal pulse functions, according to which the exact forms of the stationary output field states can be derived. Based on the exact analysis of the nonlinear dynamics, the spectral entanglement among the output photons mediated by the ladder-type quantum system is simulated. In the 2-photon scattering case, strong correlation between the output photons can be observed if each input photon is in resonance with a transition frequency of the system. Particularly, there exists a weak two-photon process for the non-resonance case, where two input photons can couple to the transition between the first and third levels of the system if a two-photon resonance condition is satisfied. In the 3-photon scattering case, the presence of an ancillary photon could significantly influence the correlation pattern. There exist two nonlinear terms, one is for 2-photon inelastic scattering and the other for 3-photon inelastic scattering, in the 3 -photon output state. As a result, both two-photon and three-photon processes can be observed in the pattern of spectral entanglement.


## I. INTRODUCTION

The scattering of few photons by a quantum system has received considerable attention recently, as the precise control of photons has fundamental interests in optical physics and potential applications in quantum information science [1]. Single photon transistors and switches could be realized by engineering photon-matter interactions [2-4]. A single atom can induce a phase shift on a photon [5]. When photons are used to encode quantum information, the photon-photon interaction mediated by a quantum system can be exploited to synthesize the controlled-phase gate for quantum computation [6, 7]. Moreover, engineered routing and scattering of single photons could provide a scalable way for implementing quantum computation [8].

The nonlinear dynamics of few-photon scattering has been studied both analytically and numerically [9-16]. The nonlinear interaction may induce correlations among the frequency-domain variables of the photons. This correlation is often referred to as spectral entanglement [ $7,17,18$ ] which can be observed using a frequencydomain representation of the output state. The spectral entanglement is the consequence of inelastic scattering of the photons to other modes of the field, i.e., the frequency of a monochromatic single photon is not conserved. Photon-photon interaction can be enhanced by confining photons in a one-dimensional waveguide [19, 20], with their interaction mediated by a finite-level single atom or artificial atom coupled to the waveguide [17-21]. The waveguide system in the context of quantum electrodynamics (QED) has become one of the most

[^0]promising platforms for studying the nonlinear behaviors of photon scattering, such as transmission and reflection properties [4, 10, 19, 20], non-markovian dynamics [12, 14], and fundamental problems in light-matter interaction [9, 11, 15, 22].

In this paper, we consider the scattering of two photons and three photons, respectively, by a ladder-type quantum system. In particular, the output channels of the input photons can be distinguished so that we can focus on the analysis of spectral entanglement. Without the reflection process, the photon number is conserved for each input-output channel. This setup is commonly seen in the construction of a quantum phase gate $[6,7]$, where the scattering process only induces a phase shift on the logical state of the flying photons via the cross-Kerr effect [6, 7, 16, 23, 24]. The cross-Kerr effect can further be enhanced by cascading a series of finitelevel systems [6, 7, 16], or by coupling the microwave photons directly to an artificial atom in a QED system [20]. Moreover, the 2-photon scattering of a three-level ladder-type system has been studied in [17] by solving the Schrödinger equation. In this paper, the photons are encoded using a continuum of field modes, which results in a continuous pulse function for each photon. For example, an atom can couple to a continuum of modes of a one-dimensional waveguide [17, 18]. For arbitrarily given pulse functions of the input photons, we derive the exact output states for the 2-photon scattering and 3photon scattering. Since cross-Kerr interaction between the photons dominates the scattering process, strongly correlated photons can be generated at the output. Our calculation makes use of the Heisenberg picture equations of motion for the coupling operators. The exact scattering matrix in the frequency domain is derived, which is found to be quite different from the scattering matrix of a


FIG. 1. A 4-level quantum system in interaction with few photons. When the input is in a 2 -photon state with Photon 1 and Photon 2, the system state $\left|3_{s}\right\rangle$ is not relevant and the model is reduced to 3 -level. The input photon $i$ is exclusively coupled to the transition $\left|i_{s}\right\rangle \leftrightarrow\left|(i-1)_{s}\right\rangle$. For example, Photon 1 can induce the transition between $\left|0_{s}\right\rangle$ and $\left|1_{s}\right\rangle$. The output photons can be distinguished from each other as well. The single input and output channel can be realized by placing the quantum scatter at the end of a semi-infinite waveguide. The incident photon is travelling to the right, and the output photon is reflected and travelling unidirectionally to the left.
$V$-type quantum system [6]. For the 3-photon scattering case, the existence of an ancillary photon could significantly influence the correlation pattern. Single-photon, two-photon and three-photon resonances can be observed both in the analytical form and in the simulation.

## II. MODEL OF A 4-LEVEL SYSTEM

The scattering model is depicted in Fig. 1. The states of a 4-level ladder-type system are denoted as $\left|0_{s}\right\rangle,\left|1_{s}\right\rangle,\left|2_{s}\right\rangle,\left|3_{s}\right\rangle$ respectively, with $\left|0_{s}\right\rangle$ being the ground and initial state. Invoking the rotating wave approximation, the interaction Hamiltonian between the system and the input photons is written as

$$
\begin{equation*}
H_{\mathrm{int}}=\sum_{i} \int_{-\infty}^{\infty}\left(l_{i} L_{i} b_{i}^{\dagger}\left(\omega_{i}\right)+l_{i}^{*} L_{i}^{\dagger} b_{i}\left(\omega_{i}\right)\right) d \omega_{i} \tag{1}
\end{equation*}
$$

where $\left\{b_{i}\left(\omega_{i}\right), i=1,2,3\right\}$ are frequency-domain field annihilation operators defined on three independent input channels. $\left\{l_{i}\right\}$ are the coupling constants and $\left\{L_{i}\right\}$ are transitions between two neighboring levels defined as

$$
\begin{equation*}
L_{1}=\left|0_{s}\right\rangle\left\langle 1_{s}\right|, \quad L_{2}=\left|1_{s}\right\rangle\left\langle 2_{s}\right|, \quad L_{3}=\left|2_{s}\right\rangle\left\langle 3_{s}\right| . \tag{2}
\end{equation*}
$$

The Hamiltonian of the system is

$$
\begin{equation*}
H=\lambda_{1}\left|1_{s}\right\rangle\left\langle 1_{s}\right|+\lambda_{2}\left|2_{s}\right\rangle\left\langle 2_{s}\right|+\lambda_{3}\left|3_{s}\right\rangle\left\langle 3_{s}\right|, \tag{3}
\end{equation*}
$$

with the energy differences denoted as $h_{i}=\lambda_{i}-\lambda_{i-1}$, $i=1,2,3$. We have let $\lambda_{0}=0$ without loss of generality.

The interaction Hamiltonian (1) can be used to model the scattering of one-dimensional waveguide photons. For waveguide photons, Eq. (1) indicates that the input
and output photons are travelling unidirectionally. Experimentally, this type of interaction can be engineered by placing the atom at the end of a semi-infinite waveguide such that the input photon will be bounced back after interacting with the atom. The interaction Hamiltonian (1) also indicates that different input photons drive different transitions between two neighboring levels, which can be achieved if the photons are encoded in different polarizations, or the transition frequencies differ significantly [17]. For example, the first input photon defined using the field operators $\left\{b_{1}^{\dagger}\left(\omega_{1}\right), b_{1}\left(\omega_{1}\right)\right\}$ can only drive the transition $\left|0_{s}\right\rangle \leftrightarrow\left|1_{s}\right\rangle$. The system Hamiltonian (3) can be realized as an artificial atom made by superconducting circuits [20]. The artificial atom can be tuned to possess anharmonic energy levels, which allow the controlling of the atomic transitions with different microwave photons.

The overall dynamics of the system plus the fields is governed by a unitary operator $U\left(t, t_{0}\right)$, where $t_{0}$ is the initial time of the interaction. The dynamical equation of $U\left(t, t_{0}\right), t \geq t_{0}$, is given by [25]

$$
\begin{equation*}
d U\left(t, t_{0}\right)=\left\{b^{\dagger}(t) L-L^{\dagger} b(t)-\left(\frac{1}{2} L^{\dagger} L+\mathrm{i} H\right)\right\} U\left(t, t_{0}\right) d t \tag{4}
\end{equation*}
$$

with $U\left(t_{0}, t_{0}\right)=I \otimes I$ being the identity operator of the composite system. Here, $b(t)=\left(b_{1}(t) b_{2}(t) b_{3}(t)\right)^{T}$ is the column vector of field annihilation operators. Since the input photons are coupled to the system in parallel [26], the coupling operator $L$ is

$$
L=\left(\begin{array}{c}
l_{1} L_{1}  \tag{5}\\
l_{2} L_{2} \\
l_{3} L_{3}
\end{array}\right)
$$

Note that Markovian approximation has been invoked in the derivation of $d U\left(t, t_{0}\right)$, as (4) indicates Markovian dynamics for the open quantum system.

The Heisenberg-picture evolution of a system operator $X$ can be calculated by $X(t)=U^{\dagger}\left(t, t_{0}\right)(I \otimes X) U\left(t, t_{0}\right)$, with $I$ being the identity operator on the fields. The dynamical equation of $X(t)$ is then given by [25-27]

$$
\begin{align*}
\dot{X}(t)= & \mathcal{L}^{\dagger}(X(t)) \\
& +b^{\dagger}(t)[X(t), L(t)]+\left[L^{\dagger}(t), X(t)\right] b(t),  \tag{6}\\
b_{\text {out }}(t)= & L(t)+b(t) \tag{7}
\end{align*}
$$

where

$$
\begin{align*}
& \mathcal{L}^{\dagger}(X(t)) \\
\triangleq & -\mathrm{i}[X(t), H(t)]+L^{\dagger}(t) X(t) L(t) \\
& -\frac{1}{2} L(t)^{\dagger} L(t) X(t)-\frac{1}{2} X(t) L^{\dagger}(t) L(t) \tag{8}
\end{align*}
$$

Moreover, the output $b_{\text {out }}(t)$ is connected to the input $b(t)$ via the following relation [27]

$$
\begin{equation*}
b_{\text {out }}(t)=U^{\dagger}\left(t, t_{0}\right) b(t) U\left(t, t_{0}\right) \tag{9}
\end{equation*}
$$

In this paper, we define the Fourier transform as

$$
\begin{equation*}
f(\omega)=\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} f(t) e^{-\mathrm{i} \omega t} d t \tag{10}
\end{equation*}
$$

For clarity, we use $\omega, v, p$ to denote the variables in the frequency domain, and $t, \tau$ to denote the variables in the time domain. $u(t)$ is the Heaviside step function.

## III. 2-PHOTON SCATTERING

## A. Exact dynamics of 2-photon scattering

The 2-photon input state is defined as

$$
\begin{equation*}
\left|1_{\xi_{1}} 1_{\xi_{2}}\right\rangle=\int_{-\infty}^{\infty} d t_{1} \xi_{1}\left(t_{1}\right) b_{1}^{\dagger}\left(t_{1}\right) \int_{-\infty}^{\infty} d t_{2} \xi_{2}\left(t_{2}\right) b_{2}^{\dagger}\left(t_{2}\right)|00\rangle \tag{11}
\end{equation*}
$$

where $\xi_{i}\left(t_{i}\right)$ is the temporal pulse function for the $i$ th photon which satisfies the condition $\int_{-\infty}^{\infty}\left|\xi_{i}\left(t_{i}\right)\right|^{2} d t_{i}=1$. $|00\rangle$ is the abbreviation for $|0\rangle \otimes|0\rangle$ which denotes the vacuum field state of both input channels. Equation (11) describes a product state, i.e., the two input photons are uncorrelated. Assume initially there is no excitation within the finite-level system, in other words, the system is initialized in the ground $\left|0_{s}\right\rangle$. The joint output state of the total system is expressed as the unitary evolution of the initial state:

$$
\begin{equation*}
|\Psi(\infty)\rangle=U(\infty,-\infty)\left|1_{\xi_{1}} 1_{\xi_{2}} 0_{s}\right\rangle \tag{12}
\end{equation*}
$$

Note that (12) gives the steady-state output by letting $t_{0} \rightarrow-\infty$ and $t \rightarrow \infty$. In the steady-state limit, the system has returned to the ground state $\left|0_{s}\right\rangle$ and the output is a 2 -photon field state [28, 29]. In order to obtain an analytical form of the output field state, we take the partial trace of $|\Psi(\infty)\rangle$ on the system subspace, and then project the resulting field state onto the 2-photon subspace. By this way we arrive at

$$
\begin{equation*}
\left|\Psi_{\text {field }}(\infty)\right\rangle=\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d \tau_{1} d \tau_{2} \xi_{12}\left(\tau_{1}, \tau_{2}\right) b_{1}^{\dagger}\left(\tau_{1}\right) b_{2}^{\dagger}\left(\tau_{2}\right)|00\rangle \tag{13}
\end{equation*}
$$

with

$$
\begin{align*}
& \xi_{12}\left(\tau_{1}, \tau_{2}\right) \\
= & \left\langle 000_{s}\right| b_{2}\left(\tau_{2}\right) b_{1}\left(\tau_{1}\right) \int_{-\infty}^{\infty} d t_{1} \int_{-\infty}^{\infty} d t_{2} \\
& \times U(\infty,-\infty) \xi_{1}\left(t_{1}\right) b_{1}^{\dagger}\left(t_{1}\right) \xi_{2}\left(t_{2}\right) b_{2}^{\dagger}\left(t_{2}\right)\left|000_{s}\right\rangle . \tag{14}
\end{align*}
$$

Here, $\xi_{12}\left(\tau_{1}, \tau_{2}\right)$ is the time-domain pulse function of the 2-photon output field state. The pulse function in (14)
can be further simplified, specifically,

$$
\begin{align*}
& \xi_{12}\left(\tau_{1}, \tau_{2}\right) \\
= & \int_{-\infty}^{\infty} d t_{1} \int_{-\infty}^{\infty} d t_{2} \xi_{1}\left(t_{1}\right) \xi_{2}\left(t_{2}\right) \\
& \times\left\langle 000_{s}\right| U(\infty,-\infty) U^{\dagger}(\infty,-\infty) b_{2}\left(\tau_{2}\right) U(\infty,-\infty) \\
& \times U^{\dagger}(\infty,-\infty) b_{1}\left(\tau_{1}\right) U(\infty,-\infty) b_{1}^{\dagger}\left(t_{1}\right) b_{2}^{\dagger}\left(t_{2}\right)\left|000_{s}\right\rangle \\
= & \int_{-\infty}^{\infty} d t_{1} \int_{-\infty}^{\infty} d t_{2} \xi_{1}\left(t_{1}\right) \xi_{2}\left(t_{2}\right) \\
& \times\left\langle 000_{s}\right| b_{\text {out }, 1}\left(\tau_{1}\right) b_{\text {out }, 2}\left(\tau_{2}\right) b_{1}^{\dagger}\left(t_{1}\right) b_{2}^{\dagger}\left(t_{2}\right)\left|000_{s}\right\rangle, \tag{15}
\end{align*}
$$

where we have made use of the input-output relation (9). More details on the derivation of (15) can be found in [28]. The stationary output field state can thus be expressed in terms of a time-domain scattering matrix $S\left(\tau_{1}, \tau_{2} ; t_{1}, t_{2}\right)=$ $\left\langle 000_{s}\right| b_{\text {out }, 1}\left(\tau_{1}\right) b_{\text {out }, 2}\left(\tau_{2}\right) b_{1}^{\dagger}\left(t_{1}\right) b_{2}^{\dagger}\left(t_{2}\right)\left|000_{s}\right\rangle$.

The frequency domain representation of the 2-photon input state (11) is
$\left|1_{\xi_{1}} 1_{\xi_{2}}\right\rangle=\int_{-\infty}^{\infty} d p_{1} \xi_{1}\left(p_{1}\right) b_{1}^{\dagger}\left(p_{1}\right) \int_{-\infty}^{\infty} d p_{2} \xi_{2}\left(p_{2}\right) b_{2}^{\dagger}\left(p_{2}\right)|00\rangle$,
with $\left\{\xi_{i}\left(p_{i}\right)\right\}$ being the frequency-domain pulse functions for the two photons. As a result, the scattering matrix becomes $S\left(\tau_{1}, \tau_{2} ; p_{1}, p_{2}\right)$ with the input taking the form of (16). We can further transform the time-domain variables $\tau_{1}, \tau_{2}$ of the scattering matrix $S\left(\tau_{1}, \tau_{2} ; p_{1}, p_{2}\right)$ to frequency-domain variables $\omega_{1}, \omega_{2}$, respectively, which leads to the following expression for the frequency-domain pulse function of the stationary output state

$$
\begin{align*}
& \xi_{12}\left(\omega_{1}, \omega_{2}\right) \\
= & \int_{-\infty}^{\infty} d p_{1} \int_{-\infty}^{\infty} d p_{2} \xi_{1}\left(p_{1}\right) \xi_{2}\left(p_{2}\right) S\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right),( \tag{17}
\end{align*}
$$

where the frequency-domain scattering matrix $S\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)$ has been calculated in Appendix A:

$$
\begin{align*}
& S\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)  \tag{18}\\
= & T_{1}\left(\omega_{1}\right) \delta\left(\omega_{1}+p_{1}\right) \delta\left(\omega_{2}+p_{2}\right) \\
& +\frac{\mathrm{i}\left|l_{1}\right|^{2}\left|l_{2}\right|^{2} \delta\left(\omega_{2}+\omega_{1}+p_{1}+p_{2}\right)}{2 \pi \Gamma_{1}\left(-p_{1},-\lambda_{1}\right) \Gamma_{1}\left(\omega_{1},-\lambda_{1}\right) \Gamma_{2}\left(\omega_{2}+\omega_{1},-\lambda_{2}\right)}
\end{align*}
$$

with $\Gamma_{1}\left(\omega_{1}, a\right)=\omega_{1}-a-\frac{\left|l_{1}\right|^{2}}{2} \mathrm{i}, \Gamma_{2}\left(\omega_{2}, b\right)=\omega_{2}-b-\frac{\left|l_{2}\right|^{2}}{2} \mathrm{i}$ and $T_{1}\left(\omega_{1}\right)=\left(-\frac{\left|l_{1}\right|^{2}}{2}+\mathrm{i} \omega_{1}+\mathrm{i} \lambda_{1}\right) /\left(\frac{\left|l_{1}\right|^{2}}{2}+\mathrm{i} \omega_{1}+\mathrm{i} \lambda_{1}\right)$. The energy conservation term $\delta\left(\omega_{2}+\omega_{1}+p_{1}+p_{2}\right)$ in the scattering matrix $S\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)$ induces the correlations between the frequency components of the two photons. Plugging this scattering matrix back to (17), $\xi_{12}\left(\omega_{1}, \omega_{2}\right)$ takes the form

$$
\begin{equation*}
\xi_{12}\left(\omega_{1}, \omega_{2}\right)=\int_{-\infty}^{\infty} d p_{2} \frac{\mathrm{i}\left|l_{1}\right|^{2}\left|l_{2}\right|^{2} \xi_{1}\left(-\omega_{2}-\omega_{1}-p_{2}\right) \xi_{2}\left(p_{2}\right)}{2 \pi \Gamma_{1}\left(\omega_{1}+\omega_{2}+p_{2},-\lambda_{1}\right) \Gamma_{1}\left(\omega_{1},-\lambda_{1}\right) \Gamma_{2}\left(\omega_{2}+\omega_{1},-\lambda_{2}\right)}+T_{1}\left(\omega_{1}\right) \xi_{1}\left(-\omega_{1}\right) \xi_{2}\left(-\omega_{2}\right) \tag{19}
\end{equation*}
$$



FIG. 2. The center frequency are defined as $\Omega_{1}=-1, \Omega_{2}=$ -0.9 and we let $\lambda_{2}=1.9, \lambda_{1}=-\Omega_{1}$ and $\left|l_{1}\right|^{2}=\kappa_{1}$. The probability amplitudes are plotted for different $\kappa_{1}, \kappa_{2}$ and coupling strength $\left|l_{2}\right|^{2}$. The top left figure corresponds to probability amplitude of the interaction-free linear term which is not influenced by $\left|l_{2}\right|^{2}$.

The second term on the RHS of the equality in (19) corresponds to a linear process: the two photons just pick up a single photon phase factor $T_{1}\left(\omega_{1}\right)$ without inducing any correlation. That is, the frequency of the input photon is conserved if it is a monochromatic wave. Note that the second photon passes the system without picking up a phase factor, which is in contrast to the other schemes ( $V$-type system [6], or interacting two-level atoms [16]) for inducing Kerr interaction. This particular phenomenon has been observed before for a laddertype system by calculating the output wavepackets in Schrödinger picture [17]. $T_{1}\left(\omega_{1}\right)$ is called a single photon phase factor because a single photon input $\left|1_{\xi_{1}}\right\rangle$ will pick up this phase factor after interaction with a two-level quantum system [10, 30, 31]. The first term on the RHS of the equality in (19) indicates a nonlinear scattering process involving $\omega_{1}+\omega_{2}$, which is responsible for the correlation of frequency-domain variables between the two output channels.

It is worth mentioning that the input-output relation (7) brings an extra minus sign to the frequency variables $\left(\omega_{i} \rightarrow-\omega_{i}\right)$ as compared to other input-output formalisms, e.g. [10].

## B. Spectral entanglement

For arbitrarily given pulse functions for the input photons, the output state can be exactly calculated by (19). Here we consider Lorentzian-type pulse functions as

$$
\begin{equation*}
\xi_{i}\left(p_{i}\right)=\frac{\mathrm{i} \sqrt{\kappa_{i}}}{\sqrt{2 \pi}\left(-\frac{\kappa_{i}}{2} \mathrm{i}+p_{i}+\Omega_{i}\right)}, \quad \kappa_{i}>0, i=1,2 \tag{20}
\end{equation*}
$$

which correspond to exponentially rising pulse function in the time domain. $\Omega_{i}$ is the center frequency for the $i$ th photon. $\kappa_{i}$ characterizes the width of the pulse. We assume $\kappa_{1}=\left|l_{1}\right|^{2}$ and $-\Omega_{1}=\lambda_{1}$ for the calculation of the output state. In this case, the first input photon alone can fully excite the system from $\left|0_{s}\right\rangle$ to $\left|1_{s}\right\rangle$ at $t=0$ [30, 31].

In order to demonstrate the spectral entanglement induced by the photon-photon nonlinear interaction, firstly we remove the linear deformation from the 2-photon output state in (19). The operation that removes the linear deformation is given by

$$
\begin{equation*}
T_{1}^{-1}\left(\omega_{1}\right)=-\frac{\Gamma_{1}\left(\omega_{1},-\lambda_{1}\right)}{\Gamma_{1}\left(-\omega_{1}, \lambda_{1}\right)} \tag{21}
\end{equation*}
$$

Note that it is experimentally feasible to implement this reverse operation using linear optics [6, 32]. In this section and Fig. 2, the notation $\xi_{12}\left(\omega_{1}, \omega_{2}\right)$ is redefined as $T_{1}^{-1}\left(\omega_{1}\right) \xi_{12}\left(\omega_{1}, \omega_{2}\right)$. Accordingly, the linear component of the pulse function is thus given by $\xi_{1}\left(-\omega_{1}\right) \xi_{2}\left(-\omega_{2}\right)$, which is just the product of the pulse functions of the two input photons. As shown in the top left figure of Fig. 2, there exists no correlation between the two input channels. The probability amplitude of $\xi_{1}\left(-\omega_{1}\right) \xi_{2}\left(-\omega_{2}\right)$ peaks at the single-photon resonance point $\omega_{1}=-1=$ $-\lambda_{1}=\Omega_{1}, \omega_{2}=-0.9=-\left(\lambda_{2}-\lambda_{1}\right)=\Omega_{2}$.

The nonlinear term in (19) interferes with the linear one and scatters the frequencies of the photons around the line $\omega_{1}+\omega_{2}=-1.9=\Omega_{1}+\Omega_{2}$ (top right figure of Fig. 2). Therefore, strong interference occurs when the two photons are resonant with the transition between $\left|2_{s}\right\rangle$ and $\left|0_{s}\right\rangle$ [33]. As we increase the coupling strength $\left|l_{2}\right|^{2}$ and $\kappa_{1}, \kappa_{2}$, the frequency components of the output state is distributed more and more uniformly around the line $\omega_{1}+\omega_{2}=-1.9$. In particular, we can see that the nonlinear process dominates over the linear process for the extreme case $\left|l_{2}\right|^{2}=0.5$ and $\kappa_{1}=\kappa_{2}=0.3$ (the bottom right figure of Fig. 2). That is, the probability amplitude of $\xi_{12}\left(\omega_{1}, \omega_{2}\right)$ vanishes at the single-photon resonance point $\omega_{1}=-1, \omega_{2}=-0.9$ where the linear term reaches its maximum. It is worth mentioning that similar pattern of spectral entanglement has been observed in $[17,18]$ based on different calculation methods.

Fig. 3 depicts the calculation results of the output state in the frequency domain with $\Omega_{1}+\Omega_{2} \neq-\lambda_{2}$. If one photon is not resonant with the transition frequency, i.e., $\Omega_{1} \neq-\lambda_{1}$ or $\Omega_{2} \neq-\left(\lambda_{2}-\lambda_{1}\right)$, only one peak can be observed for $\left|\xi_{12}\left(\omega_{1}, \omega_{2}\right)\right|^{2}$ with the other one reduced, which corresponds to a weakened twophoton process. For the off-resonance case, the pattern of photon-photon interference is determined by the difference $\Omega_{1}+\Omega_{2}-\left(-\lambda_{2}\right)$. The figures on the top left and bottom left are similar and they correspond to a negative difference $\Omega_{1}+\Omega_{2}-\left(-\lambda_{2}\right)<0$. The rest two figures correspond to a positive difference $\Omega_{1}+\Omega_{2}-\left(-\lambda_{2}\right)>0$.


FIG. 3. The parameters are set as $\left|l_{2}\right|^{2}=\kappa_{1}=\kappa_{2}=0.3$. $\lambda_{2}=1.9$ and $\left|l_{1}\right|^{2}=\kappa_{1}=0.3$. The figures are plotted for different center frequencies $\Omega_{1}$ and $\Omega_{2}$ and $\lambda_{1}=-\Omega_{1}$.

## IV. 3-PHOTON SCATTERING

## A. Exact dynamics of 3-photon scattering

In analogy to the development in Section III A, for a 3 -photon input state

$$
\begin{align*}
& \left|1_{\xi_{1}} 1_{\xi_{2}} 1_{\xi_{3}}\right\rangle \\
= & \int_{-\infty}^{\infty} d t_{1} \int_{-\infty}^{\infty} d t_{2} \int_{-\infty}^{\infty} d t_{3} \\
& \times \xi_{1}\left(t_{1}\right) b_{1}^{\dagger}\left(t_{1}\right) \xi_{2}\left(t_{2}\right) b_{2}^{\dagger}\left(t_{2}\right) \xi_{3}\left(t_{3}\right) b_{3}^{\dagger}\left(t_{3}\right)|000\rangle \tag{22}
\end{align*}
$$

the time-domain pulse function of the stationary 3photon output state can be expressed as

$$
\begin{align*}
& \xi_{123}\left(\tau_{1}, \tau_{2}, \tau_{3}\right) \\
= & \left\langle 0000_{s}\right| b_{1}\left(\tau_{1}\right) b_{2}\left(\tau_{2}\right) b_{3}\left(\tau_{3}\right) U(\infty,-\infty)\left|1_{\xi_{1}} 1_{\xi_{2}} 1_{\xi_{3}} 0_{s}\right\rangle \\
= & \left\langle 0000_{s}\right| b_{\text {out }, 1}\left(\tau_{1}\right) b_{\text {out }, 2}\left(\tau_{2}\right) b_{\text {out }, 3}\left(\tau_{3}\right)\left|1_{\xi_{1}} 1_{\xi_{2}} 1_{\xi_{3}} 0_{s}\right\rangle . \tag{23}
\end{align*}
$$

Similar to the 2-photon scattering case, the output state is expanded on the basis vectors $\left\{\langle 000| b_{1}\left(\tau_{1}\right) b_{2}\left(\tau_{2}\right) b_{3}\left(\tau_{3}\right)\right\}$. In order to get the frequencydomain expression of the pulse function of the 3 -photon output state, we first transform $\left\langle 0000_{s}\right| b_{\text {out }, 1}\left(\tau_{1}\right) b_{\text {out }, 2}\left(\tau_{2}\right)$ in (23) to the frequency domain with respect to the two variables $\tau_{1}, \tau_{2}$. By this we can express $\xi_{123}\left(\omega_{1}, \omega_{2}, \tau_{3}\right)$ in terms of the 2 -photon scattering matrix as

$$
\begin{align*}
& \xi_{123}\left(\omega_{1}, \omega_{2}, \tau_{3}\right) \\
= & \int_{-\infty}^{\infty} d p_{1} \int_{-\infty}^{\infty} d p_{2} S\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right) \\
& \times\left\langle 0000_{s}\right| b_{1}\left(p_{1}\right) b_{2}\left(p_{2}\right) b_{\mathrm{out}, 3}\left(\tau_{3}\right)\left|1_{\xi_{1}} 1_{\xi_{2}} 1_{\xi_{3}} 0_{s}\right\rangle \tag{24}
\end{align*}
$$

The key step is the calculation of the Fourier transform of the following term

$$
\begin{align*}
& \left\langle 0000_{s}\right| b_{1}\left(p_{1}\right) b_{2}\left(p_{2}\right) b_{\mathrm{out}, 3}\left(\tau_{3}\right)\left|1_{\xi_{1}} 1_{\xi_{2}} 1_{\xi_{3}} 0_{s}\right\rangle \\
= & \left\langle 0000_{s}\right| b_{1}\left(p_{1}\right) b_{2}\left(p_{2}\right)\left(l_{3} L_{3}\left(\tau_{3}\right)+b_{3}\left(\tau_{3}\right)\right)\left|1_{\xi_{1}} 1_{\xi_{2}} 1_{\xi_{3}} 0_{s}\right\rangle . \tag{25}
\end{align*}
$$

The Fourier representation of the term

$$
\begin{align*}
& f_{2}\left(p_{1}, p_{2}, \tau_{3}\right)  \tag{26}\\
= & \left\langle 0000_{s}\right| b_{1}\left(p_{1}\right) b_{2}\left(p_{2}\right) L_{3}\left(\tau_{3}\right) b_{1}^{\dagger}\left(v_{1}\right) b_{2}^{\dagger}\left(v_{2}\right) b_{3}^{\dagger}\left(v_{3}\right)\left|0000_{s}\right\rangle
\end{align*}
$$

with respect to the variable $\tau_{3}$ is given in Appendix B. Based on (24), the frequency-domain pulse function of the 3 -photon output field state is given by

$$
\begin{align*}
& \xi_{123}\left(\omega_{1}, \omega_{2}, \omega_{3}\right) \\
= & T_{1}\left(\omega_{1}\right) \xi_{1}\left(-\omega_{1}\right) \xi_{2}\left(-\omega_{2}\right) \xi_{3}\left(-\omega_{3}\right)+\int_{-\infty}^{\infty} d v_{1} \frac{\mathrm{i}\left|l_{1}\right|^{2}\left|l_{2}\right|^{2} \xi_{1}\left(v_{1}\right) \xi_{2}\left(-v_{1}-\omega_{1}-\omega_{2}\right) \xi_{3}\left(-\omega_{3}\right)}{2 \pi \Gamma_{1}\left(\lambda_{1}, v_{1}\right) \Gamma_{1}\left(\omega_{1},-\lambda_{1}\right) \Gamma_{2}\left(\omega_{1}+\omega_{2}, \lambda_{2}\right)} \\
& +\int_{-\infty}^{\infty} d v_{1} \int_{-\infty}^{\infty} d v_{2} \frac{\mathrm{i}\left|l_{1}\right|^{2}\left|l_{2}\right|^{2}\left|l_{3}\right|^{2} \xi_{1}\left(v_{1}\right) \xi_{2}\left(v_{2}\right) \xi_{3}\left(-v_{1}-v_{2}-\omega_{3}-\omega_{1}-\omega_{2}\right)}{4 \pi^{2} \Gamma_{1}\left(\omega_{1},-\lambda_{1}\right) \Gamma_{1}\left(-v_{1},-\lambda_{1}\right) \Gamma_{2}\left(-v_{1}-v_{2},-\lambda_{2}\right) \Gamma_{2}\left(\omega_{1}+\omega_{2},-\lambda_{2}\right) \Gamma_{3}\left(\omega_{3}+\omega_{1}+\omega_{2},-\lambda_{3}\right)} \\
= & T_{1}\left(\omega_{1}\right) \xi_{1}\left(-\omega_{1}\right) \xi_{2}\left(-\omega_{2}\right) \xi_{3}\left(-\omega_{3}\right)+\int_{-\infty}^{\infty} d v_{1} \frac{\mathrm{i}\left|l_{1}\right|^{2}\left|l_{2}\right|^{2} \xi_{1}\left(v_{1}\right) \xi_{2}\left(-v_{1}-\omega_{1}-\omega_{2}\right)}{2 \pi \Gamma_{1}\left(\lambda_{1}, v_{1}\right) \Gamma_{1}\left(\omega_{1},-\lambda_{1}\right) \Gamma_{2}\left(\omega_{1}+\omega_{2}, \lambda_{2}\right)} \xi_{3}\left(-\omega_{3}\right) \\
& +\int_{-\infty}^{\infty} d v_{1} \int_{-\infty}^{\infty} d v_{2} \frac{\mathrm{i}\left|l_{1}\right|^{2}\left|l_{2}\right|^{2}\left|l_{3}\right|^{2} \xi_{1}\left(v_{1}\right) \xi_{2}\left(v_{2}\right) \xi_{3}\left(-v_{1}-v_{2}-\omega_{3}-\omega_{1}-\omega_{2}\right)}{4 \pi^{2} \Gamma_{1}\left(\omega_{1},-\lambda_{1}\right) \Gamma_{1}\left(-v_{1},-\lambda_{1}\right) \Gamma_{2}\left(-v_{1}-v_{2},-\lambda_{2}\right) \Gamma_{2}\left(\omega_{1}+\omega_{2},-\lambda_{2}\right) \Gamma_{3}\left(\omega_{3}+\omega_{1}+\omega_{2},-\lambda_{3}\right)} . \tag{27}
\end{align*}
$$

Here we have used the notation $\Gamma_{3}(\omega, \lambda)=1 /(\omega-\lambda-$ $\frac{\left|l_{3}\right|^{2}}{2} \mathrm{i}$ ). The first two terms of (27) can be expressed as $\xi_{12}\left(\omega_{1}, \omega_{2}\right) \xi_{3}\left(-\omega_{3}\right)$, which corresponds to the case that only the photon-photon interaction between $\left|1_{\xi_{1}}\right\rangle$ and
$\left|1_{\xi_{2}}\right\rangle$ is in effect. The third photon just passes the system without any interaction. Similar to the 2 -photon scattering case, the third photon does not pick up a phase factor in this process. The last term in (27) describes a 3-


FIG. 4. $\left|l_{1}\right|^{2}=\left|l_{2}\right|^{2}=\left|l_{3}\right|^{3}=\kappa_{1}=\kappa_{2}=\kappa_{3}=$ 0.3. The energy differences are defined by $\lambda_{3}=2.7, \lambda_{2}=$ 1.9 and $\lambda_{1}=-\Omega_{1}=1$. The probability amplitudes $\left|T_{1}^{-1}\left(\omega_{1}\right) \xi_{123}\left(\omega_{1}, \omega_{2}, \omega_{3}\right)\right|^{2}$ are plotted with fixed $\omega_{3}$.
photon inelastic scattering process, where $\omega_{3}+\omega_{1}+\omega_{2}$ is responsible for the correlation among the frequency components of all the three photons. As a result, there exist two terms in (27) that are related to the nonlinear interaction among photons. The interference between the two nonlinear terms has significant impact on the photonphoton correlations.

## B. Spectral entanglement mediated by an ancillary photon

We define the pulse functions $\xi_{i}\left(v_{i}\right), i=1,2,3$ by (20). The parameters for the simulation are explained in Fig. 4. We fix $\omega_{3}$ such that the plot of $\left|T_{1}^{-1}\left(\omega_{1}\right) \xi_{123}\left(\omega_{1}, \omega_{2}, \omega_{3}\right)\right|^{2}$ can be obtained. In contrast to 2 -photon scattering, the correlation between the frequency variables $\omega_{1}, \omega_{2}$ of the first two photons is mediated by a third photon which is coupled to an additional level $\left|3_{s}\right\rangle$. For the resonant case (the top left figure of Fig. 4), the plot is similar to the 2 -photon scattering case. However, an interesting feature for this case is that there is non-vanishing probability around the single-photon resonance point $\omega_{1}=-1, \omega_{2}=-0.9$, in contrast to the 2 -photon scattering. The inelastic scattering occurs exactly along the line $\omega_{1}+\omega_{2}=-1.9$. When $\omega_{3} \neq \lambda_{3}-\lambda_{2}$ (the top right figure of Fig. 4), the two peaks for the probability amplitudes are unbalanced. In this case, the third photon is not resonant with $\left|2_{s}\right\rangle \leftrightarrow\left|3_{s}\right\rangle$ and so the three-photon process is weak. However, we can still observe the effect of two-photon resonance in the bottom left figure of Fig. 4, where the center frequencies satisfy $\Omega_{2}+\Omega_{3}=-1.7=-\left(\lambda_{3}-\lambda_{1}\right)$. Although the $\Omega_{3}$ in the
bottom right figure ( $\Omega_{2}+\Omega_{3} \neq-1.7$ ) is more close to the single photon transition frequency $-\left(\lambda_{3}-\lambda_{2}\right)=-0.8$, the two peaks of the correlation pattern is less symmetric as compared to the two-photon resonance case.

## V. CONCLUSION

We have investigated inelastic scattering of few photons by a Ladder-type four-level system. The whole system could be easily realized by waveguide QED systems. The exact forms of the stationary output field states have been derived, for the 2-photon scattering and 3-photon scattering cases. By means of exponentially rising functions as input pulse shapes, spectral entanglement of the output photons have been visualized. For the 2 -photon scattering scenario, if the two input photons are resonant with the finite-level system, strong nonlinear correlation between the two output photons can be observed; on the other hand, for the non-resonance case, there exists a weak two-photon process. For the 3 -photon scattering scenario, a non-vanishing probability amplitude around the single-photon resonance point can be observed even when the three input photons are all resonant with the four-level system. Moreover, due to the presence of two nonlinear terms, the peaks of the probability amplitudes are unbalanced.

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Appendix A: Calculation of $S\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)$ in (18)
In this appendix, we give an exact calculation of $S\left(\omega_{1}, \omega_{2}, p_{1}, p_{2}\right)$ in (18). Let $X=L_{1}$ in (6). We have

$$
\begin{equation*}
\left\langle 000_{s}\right| \dot{L}_{1}(t)=\left\langle 000_{s}\right|\left[-\left(\mathrm{i} \lambda_{1}+\frac{1}{2}\left|l_{1}\right|^{2}\right) L_{1}(t)-l_{1}^{*} b_{1}(t)\right] \tag{A1}
\end{equation*}
$$

The solution to the above ordinary differential equation is given by

$$
\begin{equation*}
\left\langle 000_{s}\right| L_{1}(\tau)=-\left\langle 000_{s}\right| \int_{-\infty}^{\tau} l_{1}^{*} e^{\left(-\mathrm{i} \lambda_{1}-\frac{1}{2}\left|l_{1}\right|^{2}\right)(\tau-r)} b_{1}(r) d r \tag{A2}
\end{equation*}
$$

Using (9) we have $S\left(\tau_{1}, \tau_{2} ; p_{1}, p_{2}\right)=\left\langle 000_{s}\right|\left(l_{1} L_{1}\left(\tau_{1}\right)+\right.$ $\left.b_{1}\left(\tau_{1}\right)\right)\left(l_{2} L_{2}\left(\tau_{2}\right)+b_{2}\left(\tau_{2}\right)\right) b_{1}^{\dagger}\left(p_{1}\right) b_{2}^{\dagger}\left(p_{2}\right)\left|000_{s}\right\rangle$. Since

$$
\begin{aligned}
& \left\langle 000_{s}\right|\left(l_{1} L_{1}\left(\tau_{1}\right)+b_{1}\left(\tau_{1}\right)\right) \\
= & \left\langle 000_{s}\right| \int_{-\infty}^{\infty}\left[-\left|l_{1}\right|^{2} e^{\left(-\mathrm{i} \lambda_{1}-\frac{1}{2}\left|l_{1}\right|^{2}\right)\left(\tau_{1}-r\right)} u\left(\tau_{1}-r\right)\right.
\end{aligned}
$$

$$
\begin{equation*}
\left.+\delta\left(\tau_{1}-r\right)\right] b_{1}(r) d r \tag{A3}
\end{equation*}
$$

the Fourier transform of the 1-photon state (A3) with respect to the variable $r$ is given by

$$
\begin{equation*}
\left\langle 000_{s}\right| \int_{-\infty}^{\infty} \Gamma\left(\omega, \tau_{1}\right) b_{1}(\omega) d \omega \tag{A4}
\end{equation*}
$$

with the pulse function being

$$
\begin{equation*}
\Gamma\left(\omega, \tau_{1}\right)=\frac{1}{\sqrt{2 \pi}} e^{-\mathrm{i} \omega \tau_{1}} \frac{-\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega+\mathrm{i} \lambda_{1}}{\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega+\mathrm{i} \lambda_{1}} \tag{A5}
\end{equation*}
$$

Transforming $\tau_{1}$ to $\omega_{1}$ yields

$$
\begin{align*}
& S\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right) \\
= & T_{1}\left(\omega_{1}\right)\left[l_{2} f_{1}\left(-\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)+\delta\left(\omega_{1}+p_{1}\right) \delta\left(\omega_{2}+p_{2}\right)\right] . \tag{A6}
\end{align*}
$$

Here we have defined

$$
T_{1}\left(\omega_{1}\right)=\frac{-\frac{\left|l_{1}\right|^{2}}{2}+\mathrm{i} \omega_{1}+\mathrm{i} \lambda_{1}}{\frac{\left|l_{1}\right|^{2}}{2}+\mathrm{i} \omega_{1}+\mathrm{i} \lambda_{1}}
$$

$$
\begin{align*}
& \frac{\partial}{\partial \tau_{2}} f_{1}\left(\omega_{1}, \tau_{2} ; p_{1}, p_{2}\right) \\
= & \left(-\mathrm{i} h_{2}-\frac{\left|l_{1}\right|^{2}+\left|l_{2}\right|^{2}}{2}\right) f\left(\omega_{1}, \tau_{2} ; p_{1}, p_{2}\right)-\frac{l_{2}^{*}\left|l_{1}\right|^{2}}{2 \pi \sqrt{2 \pi}} e^{-\mathrm{i} p_{2} \tau_{2}} \int_{-\infty}^{\infty} e^{\mathrm{i} \omega_{1} r_{1}} e^{\left(\mathrm{i} \lambda_{1}-\frac{1}{2}\left|l_{1}\right|^{2}\right)\left(\tau_{2}-r_{1}\right)} u\left(\tau_{2}-r_{1}\right) d r_{1} \\
& \times \int_{-\infty}^{\infty} e^{\left(-\mathrm{i} \lambda_{1}-\frac{1}{2}\left|l_{1}\right|^{2}\right)\left(\tau_{2}-r_{2}\right)} u\left(\tau_{2}-r_{2}\right) e^{-\mathrm{i} p_{1} r_{2}} d r_{2}-\frac{l_{1}}{\sqrt{2 \pi}} e^{\mathrm{i} \omega_{1} \tau_{2}}\left\langle 000_{s}\right| L_{1}\left(\tau_{2}\right) L_{2}\left(\tau_{2}\right) b_{1}^{\dagger}\left(p_{1}\right) b_{2}^{\dagger}\left(p_{2}\right)\left|000_{s}\right\rangle . \tag{A10}
\end{align*}
$$

Note that the inserted identity operator reduces to $\left|000_{s}\right\rangle\left\langle 000_{s}\right|$ since $\left\langle 000_{s}\right| b_{1}\left(\omega_{1}\right) L_{1}^{\dagger}\left(\tau_{2}\right)=$ $\left\langle 000_{s}\right| b_{1}\left(\omega_{1}\right) U^{\dagger}\left(\tau_{2},-\infty\right) L_{1}^{\dagger} U\left(\tau_{2},-\infty\right)$ contains no excitation. Using (A4) and (A7) we can rewrite the last term as

$$
\begin{equation*}
=\frac{\left|l_{1}\right|^{2}}{2 \pi} \int_{-\infty}^{\infty} \frac{e^{\mathrm{i}\left(\omega_{1}-\omega\right) \tau_{2}}}{\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega+\mathrm{i} \lambda_{1}} f_{1}\left(\omega, \tau_{2} ; p_{1}, p_{2}\right) d \omega \tag{A11}
\end{equation*}
$$

Then we transform $\tau_{2}$ to frequency domain and get

$$
\begin{equation*}
=\frac{\left|l_{1}\right|^{2}}{2 \pi} \int_{-\infty}^{\infty} \frac{1}{\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega+\mathrm{i} \lambda_{1}} f_{1}\left(\omega, \omega_{2}+\omega-\omega_{1} ; p_{1}, p_{2}\right) d \omega \tag{A12}
\end{equation*}
$$

As a result, the frequency-domain representation of (A9) is given by

$$
\mathrm{i} \omega_{2} f_{1}\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)
$$

and

$$
\begin{align*}
& f_{1}\left(-\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)  \tag{A7}\\
= & \left\langle 000_{s}\right| b_{1}\left(-\omega_{1}\right) L_{2}\left(\omega_{2}\right) b_{1}^{\dagger}\left(p_{1}\right) b_{2}^{\dagger}\left(p_{2}\right)\left|000_{s}\right\rangle . \tag{A8}
\end{align*}
$$

Eq. (A8) can be calculated by considering the term $f_{1}\left(\omega_{1}, \tau_{2} ; p_{1}, p_{2}\right)=\left\langle 000_{s}\right| b_{1}\left(\omega_{1}\right) L_{2}\left(\tau_{2}\right) b_{1}^{\dagger}\left(p_{1}\right) b_{2}^{\dagger}\left(p_{2}\right)\left|000_{s}\right\rangle$. A powerful approach to calculate this term is by taking the derivative with respect to the time-domain variable $\tau_{2}[10,34]$ as

$$
\begin{align*}
& \frac{\partial}{\partial \tau_{2}} f_{1}\left(\omega_{1}, \tau_{2} ; p_{1}, p_{2}\right) \\
= & \left(-\mathrm{i} h_{2}-\frac{\left|l_{1}\right|^{2}+\left|l_{2}\right|^{2}}{2}\right) f\left(\omega_{1}, \tau_{2} ; p_{1}, p_{2}\right) \\
- & \frac{l_{2}^{*}}{\sqrt{2 \pi}} e^{-\mathrm{i} p_{2} \tau_{2}}\left\langle 000_{s}\right| b_{1}\left(\omega_{1}\right) L_{1}^{\dagger}\left(\tau_{2}\right) L_{1}\left(\tau_{2}\right) b_{1}^{\dagger}\left(p_{1}\right)\left|000_{s}\right\rangle \\
- & \frac{l_{1}}{\sqrt{2 \pi}} e^{\mathrm{i} \omega_{1} \tau_{2}}\left\langle 000_{s}\right| L_{1}\left(\tau_{2}\right) L_{2}\left(\tau_{2}\right) b_{1}^{\dagger}\left(p_{1}\right) b_{2}^{\dagger}\left(p_{2}\right)\left|000_{s}\right\rangle \tag{A9}
\end{align*}
$$

where the derivative of $L_{2}\left(\tau_{2}\right)$ is obtained using (6). Also we have used the equalities $\left\langle 000_{s}\right| b_{1}\left(\omega_{1}\right) b_{1}^{\dagger}\left(\tau_{2}\right)=\frac{1}{\sqrt{2 \pi}}\left\langle 000_{s}\right| e^{\mathrm{i} \omega_{1} \tau_{2}} \quad$ and $\left\langle 000_{s}\right| b_{2}\left(p_{2}\right) b_{2}^{\dagger}\left(\tau_{2}\right)=\frac{1}{\sqrt{2 \pi}}\left\langle 000_{s}\right| e^{\mathrm{i} p_{2} \tau_{2}}$. Sandwiching the identity operator between $L_{1}^{\dagger}\left(\tau_{2}\right)$ and $L_{1}\left(\tau_{2}\right)$ on the third line of (A9) and making use of (A2), we can obtain

$$
\begin{align*}
& =\left(-\mathrm{i} h_{2}-\frac{\left|l_{1}\right|^{2}+\left|l_{2}\right|^{2}}{2}\right) f_{1}\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right) \\
& -\frac{l_{2}^{*}\left|l_{1}\right|^{2}}{2 \pi} \frac{\delta\left(\omega_{2}-\omega_{1}+p_{1}+p_{2}\right)}{\left(\mathrm{i} \omega_{1}-\mathrm{i} \lambda_{1}+\frac{\left|l_{1}\right|^{2}}{2}\right)\left(-\mathrm{i} p_{1}+\mathrm{i} \lambda_{1}+\frac{\left|l_{1}\right|^{2}}{2}\right)} \\
& +\frac{\left|l_{1}\right|^{2}}{2 \pi} \int_{-\infty}^{\infty} \frac{1}{\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega+\mathrm{i} \lambda_{1}} f_{1}\left(\omega, \omega_{2}+\omega-\omega_{1} ; p_{1}, p_{2}\right) d \omega \tag{A13}
\end{align*}
$$

Now we follow the procedure in [16, Appendix A.2] to solve the above equation. First, make the replacement $\omega_{2} \rightarrow \omega_{2}+\omega_{1}$ and define $g\left(\omega_{1}, \omega_{2}\right)=f_{1}\left(\omega_{1}, \omega_{2}+\right.$ $\left.\omega_{1} ; p_{1}, p_{2}\right)$. Then we can write the above equation as

$$
\left[\mathrm{i}\left(\omega_{2}+\omega_{1}+h_{2}\right)+\frac{\left|l_{1}\right|^{2}+\left|l_{2}\right|^{2}}{2}\right] g\left(\omega_{1}, \omega_{2}\right)
$$

$$
\begin{equation*}
=-\frac{l_{2}^{*}\left|l_{1}\right|^{2}}{2 \pi} \frac{\delta\left(\omega_{2}+p_{1}+p_{2}\right)}{\left(\mathrm{i} \omega_{1}-\mathrm{i} \lambda_{1}+\frac{\left|l_{1}\right|^{2}}{2}\right)\left(-\mathrm{i} p_{1}+\mathrm{i} \lambda_{1}+\frac{\left|l_{1}\right|^{2}}{2}\right)} \tag{A14}
\end{equation*}
$$

$$
+\frac{\left|l_{1}\right|^{2}}{2 \pi} \int_{-\infty}^{\infty} \frac{1}{\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega+\mathrm{i} \lambda_{1}} g\left(\omega, \omega_{2}\right) d \omega
$$

Next, define $G\left(\omega_{2}\right)=\int_{-\infty}^{\infty} \frac{1}{\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega+\mathrm{i} \lambda_{1}} g\left(\omega, \omega_{2}\right) d \omega$ and rewrite (A14) as

$$
\begin{align*}
G\left(\omega_{2}\right)= & \int_{-\infty}^{\infty} \frac{-l_{2}^{*}\left|l_{1}\right|^{2} \delta\left(\omega_{2}+p_{1}+p_{2}\right)}{2 \pi\left(\mathrm{i} \omega_{1}-\mathrm{i} \lambda_{1}+\frac{\left|l_{1}\right|^{2}}{2}\right)\left(-\mathrm{i} p_{1}+\mathrm{i} \lambda_{1}+\frac{\left|l_{1}\right|^{2}}{2}\right)\left(\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega_{1}+\mathrm{i} \lambda_{1}\right)\left(\mathrm{i}\left(\omega_{2}+\omega_{1}+h_{2}\right)+\frac{\left|l_{1}\right|^{2}+\left|l_{2}\right|^{2}}{2}\right)} d \omega_{1} \\
& +G\left(\omega_{2}\right) \int_{-\infty}^{\infty} \frac{\left|l_{1}\right|^{2}}{2 \pi\left(\frac{\left|l_{1}\right|^{2}}{2}-\mathrm{i} \omega_{1}+\mathrm{i} \lambda_{1}\right)\left(\mathrm{i}\left(\omega_{2}+\omega_{1}+h_{2}\right)+\frac{\left|l_{1}\right|^{2}+\left|l_{2}\right|^{2}}{2}\right)} d \omega_{1} \tag{A15}
\end{align*}
$$

The integrals can be calculated using the residue theorem, which yields

$$
\begin{equation*}
G\left(\omega_{2}\right)=\frac{l_{2}^{*} \delta\left(\omega_{2}+p_{1}+p_{2}\right)}{\left(\lambda_{1}-p_{1}-\frac{\left|l_{1}\right|^{2}}{2} \mathrm{i}\right)\left(\omega_{2}+\lambda_{2}-\frac{\left|l_{2}\right|^{2}}{2} \mathrm{i}\right)} \tag{A16}
\end{equation*}
$$

Plugging this back to (A14) gives

$$
\begin{equation*}
g\left(\omega_{1}, \omega_{2}\right)=-\frac{\left|l_{1}\right|^{2} l_{2}^{*} \mathrm{i}}{2 \pi} \frac{\delta\left(\omega_{2}+p_{1}+p_{2}\right)}{\Gamma_{1}\left(\lambda_{1}, p_{1}\right) \Gamma_{1}\left(\omega_{1}, \lambda_{1}\right) \Gamma_{2}\left(\omega_{2},-\lambda_{2}\right)}, \tag{A17}
\end{equation*}
$$

where $\Gamma_{1}\left(\omega_{1}, a\right)=\left(\omega_{1}-a-\frac{\left|l_{1}\right|^{2}}{2} \mathbf{i}\right), \Gamma_{2}\left(\omega_{2}, b\right)=\left(\omega_{2}-b-\right.$ $\left.\frac{\left|l_{2}\right|^{2}}{2} \mathrm{i}\right)$. Finally, we obtain

$$
\begin{aligned}
& f_{1}\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right) \\
= & -\frac{\left|l_{1}\right|^{2} l_{2}^{*} \mathrm{i}}{2 \pi} \frac{\delta\left(\omega_{2}-\omega_{1}+p_{1}+p_{2}\right)}{\Gamma_{1}\left(\lambda_{1}, p_{1}\right) \Gamma_{1}\left(\omega_{1}, \lambda_{1}\right) \Gamma_{2}\left(\omega_{2}-\omega_{1},-\lambda_{2}\right)}
\end{aligned}
$$

Substituting (A18) into (A6) yields (18).

## Appendix B: Calculation of the frequency-domain <br> counterpart $f_{2}\left(p_{1}, p_{2}, \omega_{3}\right)$ of $f_{2}\left(p_{1}, p_{2}, \tau_{3}\right)$ in (26)

Take the derivative of $f_{2}\left(p_{1}, p_{2}, \tau_{3}\right)$ with respect to $\tau_{3}$ yields

$$
\begin{align*}
& \frac{\partial}{\partial \tau_{3}} f_{2}\left(p_{1}, p_{2}, \tau_{3}\right) \\
= & \left(-\mathrm{i} h_{3}-\frac{\left|l_{2}\right|^{2}+\left|l_{3}\right|^{2}}{2}\right) f_{2}\left(p_{1}, p_{2}, \tau_{3}\right) \\
& -l_{3}^{*}\left\langle 0000_{s}\right| b_{1}\left(p_{1}\right) b_{2}\left(p_{2}\right)\left(\left|2_{s}\right\rangle\left\langle 2_{s}\right|\right)\left(\tau_{3}\right) b_{3}\left(\tau_{3}\right) \\
& \times b_{1}^{\dagger}\left(v_{1}\right) b_{2}^{\dagger}\left(v_{2}\right) b_{3}^{\dagger}\left(v_{3}\right)\left|0000_{s}\right\rangle \\
& -l_{2}\left\langle 0000_{s}\right| b_{1}\left(p_{1}\right) b_{2}\left(p_{2}\right) b_{2}^{\dagger}\left(\tau_{3}\right)\left(\left|1_{s}\right\rangle\left\langle 3_{s}\right|\right)\left(\tau_{3}\right) \\
& \times b_{1}^{\dagger}\left(v_{1}\right) b_{2}^{\dagger}\left(v_{2}\right) b_{3}^{\dagger}\left(v_{3}\right)\left|0000_{s}\right\rangle \tag{B1}
\end{align*}
$$

By simplifying the equation and transforming $\tau_{3}$ to the frequency domain, we arrive at

$$
\begin{aligned}
& \mathrm{i} \omega_{3} f_{2}\left(p_{1}, p_{2}, \omega_{3}\right) \\
= & \left(-\mathrm{i} h_{3}-\frac{\left|l_{2}\right|^{2}+\left|l_{3}\right|^{2}}{2}\right) f_{2}\left(p_{1}, p_{2}, \omega_{3}\right)
\end{aligned}
$$

$$
\begin{align*}
& -\frac{l_{3}^{*}}{2 \pi} \delta\left(p_{1}+p_{2}-v_{3}-v_{1}-v_{2}-\omega_{3}\right) \int d \omega \\
& \times F_{1}^{*}\left(\omega, \omega-v_{1}-v_{2}-\omega_{3}-v_{3}, p_{1}\right) F_{1}\left(\omega, \omega-v_{1}-v_{2}, v_{1}\right) \\
& -\frac{l_{2}}{2 \pi} \int_{-\infty}^{\infty} d \omega_{4} \int_{-\infty}^{\infty} d \omega_{5} F_{1}\left(p_{1}, p_{1}-\omega_{4}-\omega_{5}, \omega_{4}\right) \\
& \times f_{2}\left(\omega_{4}, \omega_{5}, \omega_{3}+\omega_{4}+\omega_{5}-p_{1}-p_{2}\right) \tag{B2}
\end{align*}
$$

In (B2), we denote $f_{1}\left(\omega_{1}, \omega_{2} ; p_{1}, p_{2}\right)$ by $F_{1}\left(\omega_{1}, \omega_{2}, p_{1}\right) \delta\left(\omega_{2}-\omega_{1}+p_{1}+p_{2}\right)$, where

$$
\begin{equation*}
F_{1}\left(\omega, \omega_{2}, p_{1}\right)=\frac{c}{\Gamma_{1}\left(\lambda_{1}, p_{1}\right) \Gamma_{1}\left(\omega, \lambda_{1}\right) \Gamma_{2}\left(\omega_{2}-\omega,-\lambda_{2}\right)} \tag{B3}
\end{equation*}
$$

with $c=-\frac{\left|l_{1}\right|^{2} l_{2}^{*} \mathrm{i}}{2 \pi}$. Making the replacement $\omega_{3} \rightarrow \omega_{3}+$ $p_{1}+p_{2}$ and define

$$
\begin{align*}
& \begin{array}{l}
G\left(\omega_{3}\right) \\
= \\
\\
\\
\times f_{2}\left(p_{1}, p_{2}, \omega_{3}+p_{1}+p_{2}\right)
\end{array}
\end{align*}
$$

Eq. (B2) can be expressed as

$$
\begin{align*}
& G\left(\omega_{3}\right) \\
= & \frac{l_{3}^{*}|c|^{2} \delta\left(v_{3}+v_{1}+v_{2}+\omega_{3}\right) \mathrm{i}}{\left|l_{1}\right|^{2} \Gamma_{1}\left(\lambda_{1}, v_{1}\right) \Gamma_{2}\left(-v_{1}-v_{2},-\lambda_{2}\right)} \\
& \times \int d p_{1} \int d p_{2} \frac{1}{\omega_{3}+h_{3}+p_{1}+p_{2}-\frac{\left|l_{2}\right|^{2}+\left|l_{3}\right|^{2}}{2} \mathrm{i}} \\
& \times \frac{1}{\Gamma_{1}\left(\lambda_{1}, p_{1}\right) \Gamma_{2}\left(-p_{1}-p_{2},-\lambda_{2}\right)} \\
\times & \frac{1}{\Gamma_{1}^{*}\left(\lambda_{1}, p_{1}\right) \Gamma_{2}^{*}\left(-v_{1}-v_{2}-\omega_{3}-p_{1}-p_{2}-v_{3},-\lambda_{2}\right)} \\
+ & \frac{l_{2} c \mathrm{i}}{2 \pi} G\left(\omega_{3}\right) \int d p_{1} \int d p_{2} \frac{1}{\Gamma_{1}\left(\lambda_{1}, p_{1}\right) \Gamma_{1}\left(p_{1}, \lambda_{1}\right)}  \tag{B5}\\
\times & \frac{1}{\Gamma_{2}\left(-p_{1}-p_{2},-\lambda_{2}\right)\left(\omega_{3}+h_{3}+p_{1}+p_{2}-\frac{\left|l_{2}\right|^{2}+\left|l_{3}\right|^{2}}{2} \mathrm{i}\right)}
\end{align*}
$$

As a result, $G\left(\omega_{3}\right)$ can be calculated using the residue theorem as

$$
\begin{align*}
G\left(\omega_{3}\right)= & \frac{l_{3}^{*}\left|l_{2}\right|^{2} \delta\left(v_{3}+v_{1}+v_{2}+\omega_{3}\right)}{\Gamma_{1}\left(\lambda_{1}, v_{1}\right) \Gamma_{2}\left(-v_{1}-v_{2},-\lambda_{2}\right)} \\
& \times \frac{1}{\left(v_{1}+v_{2}+\omega_{3}+v_{3}-\left|l_{2}\right|^{2} \mathrm{i}\right)\left(\omega_{3}+\lambda_{3}-\frac{\left|l_{3}\right|^{2}}{2} \mathrm{i}\right)} \tag{B6}
\end{align*}
$$

Since

$$
\begin{align*}
& {\left[\mathrm{i}\left(\omega_{3}+h_{3}+p_{1}+p_{2}\right)+\frac{\left|l_{2}\right|^{2}+\left|l_{3}\right|^{2}}{2}\right] } \\
& \times f_{2}\left(p_{1}, p_{2}, \omega_{3}+p_{1}+p_{2}\right) \\
= & -\frac{l_{3}^{*}|c|^{2} \delta\left(v_{3}+v_{1}+v_{2}+\omega_{3}\right)}{\left|l_{1}\right|^{2} \Gamma_{1}^{*}\left(\lambda_{1}, p_{1}\right) \Gamma_{1}\left(\lambda_{1}, v_{1}\right) \Gamma_{2}\left(-v_{1}-v_{2},-\lambda_{2}\right)} \\
& \times \frac{1}{\Gamma_{2}^{*}\left(-v_{1}-v_{2}-\omega_{3}-p_{1}-p_{2}-v_{3},-\lambda_{2}\right)} \\
& -\frac{l_{2} c}{2 \pi \Gamma_{1}\left(p_{1}, \lambda_{1}\right)} G\left(\omega_{3}\right) \tag{B7}
\end{align*}
$$

replacing $\omega_{3}$ with $\omega_{3}-p_{1}-p_{2}$ we finally obtain

$$
\begin{align*}
& l_{3} f_{2}\left(p_{1}, p_{2}, \omega_{3}\right) \\
= & F_{2}\left(\omega_{3}, p_{1}, p_{2}, v_{1}, v_{2}, v_{3}\right) \delta\left(v_{3}+v_{1}+v_{2}+\omega_{3}-p_{1}-p_{2}\right) \tag{B8}
\end{align*}
$$

with

$$
\begin{align*}
& F_{2}\left(\omega_{3}, p_{1}, p_{2}, v_{1}, v_{2}, v_{3}\right) \\
= & \frac{\left|l_{1}\right|^{2}\left|l_{2}\right|^{2}\left|l_{3}\right|^{2}}{4 \pi^{2}\left(\mathrm{i}\left(\omega_{3}+h_{3}\right)+\frac{\left|l_{2}\right|^{2}+\left|l_{3}\right|^{2}}{2}\right)} \\
& \times \frac{1}{\Gamma_{1}\left(p_{1}, \lambda_{1}\right) \Gamma_{1}\left(\lambda_{1}, v_{1}\right) \Gamma_{2}\left(-v_{1}-v_{2},-\lambda_{2}\right)} \\
& \times\left[\frac{1}{\Gamma_{2}^{*}\left(-v_{1}-v_{2}-\omega_{3}-v_{3}-\lambda_{2}\right)}\right. \\
& +\frac{\left|l_{2}\right|^{2} \mathrm{i}}{\left(v_{1}+v_{2}+\omega_{3}-p_{1}-p_{2}+v_{3}-\left|l_{2}\right|^{2} \mathrm{i}\right)} \\
& \left.\times \frac{1}{\left(\omega_{3}-p_{1}-p_{2}+\lambda_{3}-\frac{\left|l_{3}\right|^{2}}{2} \mathrm{i}\right)}\right] . \tag{B9}
\end{align*}
$$

Thus, $f_{2}\left(p_{1}, p_{2}, \omega_{3}\right)$, the frequency-domain counterpart of (26) follows from (B9). Then a lengthy calculation leads to the 3 -photon pulse function (27).
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