Entanglement and four-wave mixing effects in the dissipation-free nonlinear interaction of two photons at a single atom

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We investigate the nonlinear interaction between two photons in a single-input pulse at an atomic two-level nonlinearity. A one-dimensional model for the propagation of light to and from the atom is used to describe the precise spatiotemporal coherence of the two-photon state. It is shown that the interaction generates spatiotemporal entanglement in the output state similar to the entanglement observed in parametric down-conversion. A method of generating photon pairs from coherent pump light using this quantum-mechanical four-wave mixing process is proposed.

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I. INTRODUCTION

Optical nonlinearities sensitive to individual photons may provide interesting new possibilities of controlling and manipulating the quantum states of light [1–10]. Possible applications of such nonlinearities include quantum nondemolition measurements of photon number [11] and quantum logic circuits for photonic qubits [12]. Experimentally, sufficiently strong nonlinearities have been achieved in cavity quantum electrodynamics, where cavity confinement can enhance the coupling between a single two-level atom and the input field [2]. By optimizing the suppression of uncontrollable photon losses in such systems, it may be possible to realize a fully quantum coherent photon-photon interaction [13]. The analysis of such a quantum level nonlinearity then requires a quantum-mechanical treatment of the spatiotemporal coherence in the input and output fields. Specifically, spontaneous four-wave mixing effects may entangle the two input photons in their spatial coordinates. This entanglement appears to introduce noise in the single-photon coherence, even though the two photons are still in a quantum-mechanically pure state.

In order to investigate such effects, we apply a one-dimensional model of light field propagation to and from a single two-level atom [14,15]. If photon losses are avoided, it is then possible to determine the response functions for single-photon and for two-photon inputs. Using these response functions, we derive the output state for a resonant rectangular input. We discuss the implications of this result for coherent input fields and show that it is possible to create entangled photon pairs from coherent input light by using an interferometric strategy similar to the one recently applied in parametric down-conversion [16].

II. ONE-DIMENSIONAL MODEL OF LIGHT FIELD PROPAGATION

If the transversal beam profile is known, it is sufficient to describe the propagation of light to and from a system using only a single-spatial coordinate. In free space, the propagation velocity c is constant. The linear propagation process can then be described by a dispersion relation of $\omega = ck$, where k is a scalar [14]. If this approximation is applied to the interaction of electromagnetic field with a single two-level system, the transversal profile of the k-space eigenmodes is defined by the coupling characteristics of the two-level system to the three-dimensional field in free space. As has been discussed in Ref. [14], the single-spatial coordinate r corresponding to the wave vector k then represents the distance from the system at $r=0$, where negative values indicate propagation towards the system and positive values indicate propagation away from the system.

Figure 1 shows a schematic representation of the model. The Hamiltonian of this system can be written as

$$\hat{H}_{1D} = \hat{H}_{\mathrm{prop}} + \hat{H}_{\mathrm{abs}},$$

with

$$\hat{H}_{\mathrm{prop}} = \int dk \;\; \hbar c k \;\; \hat{b}_k^\dagger \hat{b}_k,$$

$$\hat{H}_{\mathrm{abs}} = \int dk \;\; \hbar \sqrt{\frac{c\Gamma}{\pi}} (\hat{b}_k^\dagger \hat{\sigma}_- + \hat{\sigma}_-^\dagger \hat{b}_k), \qquad (1)$$

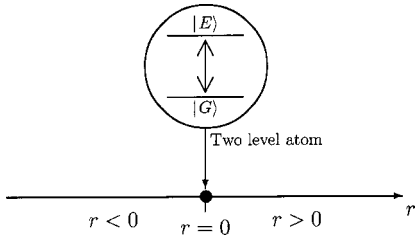

FIG. 1. Schematic representation of the one-dimensional model for light field propagation in the field-atom interaction. There is only one direction of propagation. $r<0$ represents light propagating towards the atom and $r>0$ represents light propagating away from the atom. The interaction takes place locally at $r=0$.

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where $\hat{b}_k$ is the photon annihilation operator in k space and $\sigma_- = |G\rangle\langle E|$ is the atomic annihilation operator describing coherence between the ground state $|G\rangle$ and the excited state $|E\rangle$. The coupling strength is expressed in terms of the dipole relaxation rate Γ. This rate defines the characteristic time scale of the coherent interaction between the light field and the two-level atom. For convenience, the resonant frequency of the atom has been set to zero. Note that this merely corresponds to a rotating frame of reference for the phase oscillations, so that all frequencies are expressed as frequency shifts relative to the resonant frequency $\omega_0 = ck_0$.

Experimentally, the model presented here could be realized using a one-sided microcavity [13,15]. Losses to transversal light field modes can then be minimized and almost all the light emitted by a two-level atom inside the cavity is emitted along the axis of the cavity. If this ideal condition cannot be met, the model described here could still be applied. However, it would be necessary to treat the losses as a transversal-mode mismatch between the input and output beams and the one-dimensional field actually interacting with the single atom.

III. LOCAL ABSORPTION AND EMISSION

It is now possible to formulate the Schrödinger equations for the single-photon case by defining the one-photon basis as $|k\rangle$ for one photon in a k eigenstate and $|E\rangle$ for the excited atom with no photon in free space. The quantum state $|\psi(t)\rangle$ is then described by the components $\psi(k;t) = \langle k|\psi(t)\rangle$ and $\psi(E;t) = \langle E|\psi(t)\rangle$. The temporal dynamics of these components is given by

$$\frac{d}{dt}\psi(k;t) = -ick\psi(k;t) - i\sqrt{\frac{c\Gamma}{\pi}}\psi(E;t),$$

$$\frac{d}{dt}\psi(E;t) = -i\sqrt{\frac{c\Gamma}{\pi}}\int dk \;\; \psi(k;t). \tag{2}$$

These equations of motion can now be transformed into real-space coordinates r by using the Fourier transform

$$\psi(r;t) = \frac{1}{\sqrt{2\pi}}\int dk \;\; \exp(ikr)\psi(k;t). \tag{3}$$

The equation for the propagating field then reads

$$\frac{d}{dt}\psi(r;t) = -c\frac{\partial}{\partial r}\psi(r;t) - i\sqrt{2c\Gamma}\,\delta(r)\psi(E;t). \tag{4}$$

As a result of the integration over k, this equation of motion now includes a δ function expressing the locality of emission. Since the time evolution should be continuous, this δ function implies a jump of $\psi(r;t)$ at $r=0$. By integrating Eq. (4), the discontinuity is found to be given by

$$\psi(r\to+0;t) - \psi(r\to-0;t) = -i\sqrt{\frac{2\Gamma}{c}}\psi(E;t). \tag{5}$$

Emission and absorption are therefore described by the instantaneous addition of an amplitude proportional to $\psi(E;t)$ to the single-photon wave function propagating from $r<0$ to $r>0$. At $r\neq 0$, the dynamics of $\psi(r;t)$ is simply described by linear propagation, $\psi(r;t) = \psi(r-ct;0)$.

In order to obtain the dynamics of $\psi(E;t)$, it is necessary to define the integral corresponding to $\psi(r=0;t)$. The proper result is obtained by taking the average of the incoming amplitude $\psi(r\to-0;t)$ and the outgoing amplitude $\psi(r\to+0;t)$. However, it is convenient to use the result of Eq. (5) to express the dynamics of $\psi(E;t)$ entirely in terms of the incoming amplitude $\psi(r\to-0;t)$. It then reads

$$\frac{d}{dt}\psi(E;t) = -\Gamma\psi(E;t) - i\sqrt{2c\Gamma}\,\psi(r\to-0;t). \tag{6}$$

The amplitude of the excited state $\psi(E;t)$ can therefore be obtained from an integration of the previous incoming field amplitudes $\psi(r\to-0;t)$. Since the dynamics of these amplitudes are given by linear propagation at a constant velocity c, they can be obtained from the initial single-photon wave function at $r<0$ using the linear propagation dynamics mentioned above.

With these results, it is possible to integrate the equations of motion from any initial time $t_{\rm in}$ to any final time $t_{\rm out}$. In particular, the output field within $0<r<c(t_{\rm out}-t_{\rm in})$ for $\psi(E;t_{\rm in})=0$ is given by

$$\begin{aligned}\psi(r;t_{\rm out}) &= \psi(r-c(t_{\rm out}-t_{\rm in});t_{\rm in}) - i\sqrt{\frac{2\Gamma}{c}}\psi(E;t_{\rm out}-r/c)\\ &= \psi(r-c(t_{\rm out}-t_{\rm in});t_{\rm in}) - 2\frac{\Gamma}{c}\int_{r-c(t_{\rm out}-t_{\rm in})}^{0} dr'\\ &\quad\times\exp\left(-\frac{\Gamma}{c}[r-r'-c(t_{\rm out}-t_{\rm in})]\right)\psi(r';t_{\rm in}).\end{aligned} \tag{7}$$

As the first line of Eq. (7) shows, the output wave function is a superposition of a component that propagated past the atom unchanged and a component emitted by the excited atom. Since the atom was initially in the ground state, the emission can be traced to absorptions of the incoming wave function, as represented by the integral in the last line of Eq. (7). The output wave function at $r>0$ can thus be represented as a linear function of the input wave function at $r<0$.

IV. MANY-PHOTON EFFECTS

The advantage of a local description of the field-atom interaction is that it is easily extended to multiple photons. No matter how high the photon density is, we can always define a region from $r=-\epsilon$ to $r=+\epsilon$ around the atom small enough to contain only one photon. In order to solve the field-atom interaction problem for many photons, it is therefore only necessary to consider what happens if a photon interacts with the excited atom.

For this purpose, it is useful to define the many-photon Hilbert space as a product space of independent particles.

The bosonic nature of photons must then be included in the symmetry of the initial state. For reasons of consistency, it is then also necessary to distinguish the origin of an excitation, effectively treating the excited state as a state of the photon. The two-photon wave function is then given by the amplitudes for two photons in free space, $\psi(r_1,r_2)$, the amplitudes for one photon in free space and one photon at the atom, $\psi(r_1,E)$ or $\psi(E,r_2)$, and the amplitude for a double excitation, $\psi(E,E)$. For a two-level atom, the latter must always be zero. In the Hamiltonian given by Eq. (1), this fact is expressed by the difference between the atomic annihilation operator $\hat{\sigma}_-$ and the annihilation operators of harmonic oscillators. Within the product space of independent particles, this difference is simply represented by setting the matrix elements between single excitation and double excitation to zero. The Schrödinger equation for the two-photon wave function then reads

$$\begin{aligned}
\frac{d}{dt}\psi(r_1,r_2;t) &= -c\frac{\partial}{\partial r_1}\psi(r_1,r_2;t) - i\sqrt{2c\Gamma}\,\delta(r_1)\psi(E,r_2;t) \\
&\quad - c\frac{\partial}{\partial r_2}\psi(r_1,r_2;t) \\
&\quad - i\sqrt{2c\Gamma}\,\delta(r_2)\psi(r_1,E;t), \\
\frac{d}{dt}\psi(E,r_2;t) &= -\Gamma\psi(E,r_2;t) - i\sqrt{2c\Gamma}\,\psi(r_1\to -0,r_2;t) \\
&\quad - c\frac{\partial}{\partial r_2}\psi(E,r_2;t) \quad (\cdots), \\
\frac{d}{dt}\psi(r_1,E;t) &= -c\frac{\partial}{\partial r_1}\psi(r_1,E;t) \quad (\cdots) \quad -\Gamma\psi(r_1,E;t) \\
&\quad - i\sqrt{2c\Gamma}\,\psi(r_1,r_2\to -0;t),
\end{aligned} \tag{8}$$

where $(\cdots)$ marks the missing two-photon absorption terms. This two-photon Schrödinger equation describes the nearly independent dynamics of two separate photons, except for the absence of absorption for one photon if the other photon has been absorbed by the atom. The integration of the two-photon Schrödinger equation can therefore be achieved by using the single-photon results and setting all contributions of double excitation to zero [15]. In the following, however, we will present an alternative solution of the dynamics based on the two-photon interaction represented by the missing double excitation terms in Eq. (8). This procedure has the advantage that it can be easily extended to three or more photons and may therefore provide a useful foundation for further investigations.

V. SINGLE-PHOTON AND TWO-PHOTON RESPONSE FUNCTIONS

Using the results for local emission and absorption, it is possible to evaluate the effects of the atom-field interaction on an arbitrary single-photon wave function. For this purpose, it is useful to define a time-independent characterization of the input and output wave functions. In the context of our model, this characterization is easy to obtain since the propagation before and after the interaction processes does not change the shape of the wave packet. For the single-photon cases, the input and output wave functions can therefore be given by

$$\begin{aligned}
\psi_{\rm in}(x) &= \lim_{t_{\rm in}\to-\infty}\psi(r=x+c\,t_{\rm in};t_{\rm in}), \\
\psi_{\rm out}(x) &= \lim_{t_{\rm out}\to+\infty}\psi(r=x+c\,t_{\rm out};t_{\rm out}).
\end{aligned} \tag{9}$$

According to Eq. (7), the output wave function can be obtained from the input wave function using a linear-response function $U_1(x;x')$ such that

$$\psi_{\rm out}(x) = \int_{-\infty}^{\infty} dx'\,U_1(x;x')\,\psi_{\rm in}(x'). \tag{10}$$

The single-photon response function reads

$$\begin{aligned}
&U_1(x;x') \\
&= \begin{cases} \delta(x'-x) - 2\dfrac{\Gamma}{c}\exp\left[-\dfrac{\Gamma}{c}(x'-x)\right] & \text{for } x\leqslant x' \\ 0 \quad \text{for } x>x'. \end{cases}
\end{aligned} \tag{11}$$

Note that the response function $U_1(x;x')$ is a representation of the unitary operation describing the time evolution of the field-atom interaction. It therefore preserves the norm of the wave function given by the integral over the absolute square.

Likewise, the field-atom interaction of a two-photon wave function can be described by a linear-response formalism. The input and output wave functions are then described by

$$\begin{aligned}
\psi_{\rm in}(x_1,x_2) &= \lim_{t_{\rm in}\to-\infty}\psi(r_1=x_1+c\,t_{\rm in},r_2=x_2+ct_{\rm in};t_{\rm in}), \\
\psi_{\rm out}(x_1,x_2) &= \lim_{t_{\rm out}\to+\infty}\psi(r_1=x_1+ct_{\rm out},r_2=x_2+c\,t_{\rm out};t_{\rm out}).
\end{aligned} \tag{12}$$

The unitary transform of the input state into the output state can also be described by linear-response function

$$\psi_{\rm out}(x_1,x_2) = \int_{-\infty}^{\infty} dx_1'dx_2'U_2(x_1,x_2;x_1',x_2')\,\psi_{\rm in}(x_1',x_2'). \tag{13}$$

If the two photons are always very far apart ($x_1-x_2 \gg \Gamma/c$), or if the atom is replaced with a harmonic oscillator, the propagation of the two photons must be independent of each other. In this case, the response function is equal to the product of two single-photon response functions,

$$U_{\mathrm{lin}}(x_1,x_2;x_1',x_2')=U_1(x_1;x_1')U_1(x_2;x_2'). \tag{14}$$

This response function corresponds to the linear part of the field-atom interaction. However, the absence of two-photon absorption in the dynamics causes a coupling between the photons that can be described by a nonlinear correction $\Delta U_{\mathrm{nonlin}}$ such that

$$U_2(x_1,x_2;x_1',x_2')=U_1(x_1;x_1')U_1(x_2;x_2') + \Delta U_{\mathrm{nonlin}}(x_1,x_2;x_1',x_2'). \tag{15}$$

According to the considerations in the preceding section, $\Delta U_{\mathrm{nonlin}}$ can be found by integrating the contributions from double excitations in U_{lin}. The result reads

$$\Delta U_{\mathrm{nonlin}}(x_1,x_2;x_1',x_2')=\begin{cases} -4\dfrac{\Gamma^2}{c^2}\exp\left[-\dfrac{\Gamma}{c}(x_1'-x_1)\right]\exp\left[-\dfrac{\Gamma}{c}(x_2'-x_2)\right] & \text{for } \max\{x_1,x_2\}<\min\{x_1',x_2'\} \\ 0 \quad \text{else,} \end{cases} \tag{16}$$

where the minimum $\min\{x_1',x_2'\}$ effectively defines the latest absorption time and the maximum $\max\{x_1,x_2\}$ defines the earliest emission. Thus the nonlinearity removes all components where the first emission occurs only after the second absorption [15].

It is now possible to derive the output wave function for any two-photon input wave function by integrating Eq. (13) using the expressions for U_1 and for $\Delta U_{\mathrm{nonlin}}$ given by Eqs. (11) and (15), respectively. If the input is a single-mode two-photon pulse, the input wave function can be written as a product state

$$\psi_{\mathrm{in}}(x_1,x_2)=\phi_{\mathrm{in}}(x_1)\phi_{\mathrm{in}}(x_2), \tag{17}$$

where ϕ_{in} defines the shape of the input pulse. The quantum state of the output field can then be described by

$$\psi_{\mathrm{out}}(x_1,x_2)=\phi_{\mathrm{out}}(x_1)\phi_{\mathrm{out}}(x_2)+\Delta\psi_{\mathrm{nonlin}}(x_1,x_2), \tag{18}$$

where ϕ_{out} describes the linear single-photon response given by

$$\phi_{\mathrm{out}}(x)=\int_{-\infty}^{\infty}dx'\,U_1(x;x')\phi_{\mathrm{in}}(x'), \tag{19}$$

and the nonlinear contribution is directly obtained from

$$\Delta\psi_{\mathrm{nonlin}}(x_1,x_2)=\int_{-\infty}^{\infty}dx_1'dx_2'\Delta U_{\mathrm{nonlin}}(x_1,x_2;x_1',x_2') \times\phi_{\mathrm{in}}(x_1')\phi_{\mathrm{in}}(x_2'). \tag{20}$$

These equations describe the nonlinear response of the two-level atom at the quantum level. It is now possible to apply this response function to a variety of input states. In the following, we will focus on the case of a resonant rectangular wave packet.

VI. THE QUANTUM LEVEL NONLINEARITY AT RESONANCE

Since the absorption of a photon is the strongest at resonance, a resonant input should also produce the strongest nonlinear effect in the field-atom interaction. In order to investigate this resonant nonlinearity, we consider the response to a rectangular input wave packet given by

$$\phi_{\mathrm{in}}(x)=\begin{cases}\dfrac{1}{\sqrt{L}} & \text{for } 0<x<L \\ 0 \quad \text{else.}\end{cases} \tag{21}$$

The linear and nonlinear parts of the output wave function for this rectangular wave packet can be determined analytically. They read

$$\phi_{\mathrm{out}}(x) =\begin{cases} -\dfrac{2}{\sqrt{L}}\left[1-\exp\left(-\dfrac{\Gamma L}{c}\right)\right]\exp\left(\dfrac{\Gamma}{c}x\right) & \text{for } x<0 \\ -\dfrac{1}{\sqrt{L}}\left\{1-2\exp\left[-\dfrac{\Gamma}{c}(L-x)\right]\right\} & \text{for } 0<x<L \\ 0 \quad \text{else} \end{cases} \tag{22}$$

and

$$\Delta\psi_{\mathrm{nonlin}}(x_1,x_2)=\begin{cases} -\dfrac{4}{L}\left[1-\exp\left(-\dfrac{\Gamma L}{c}\right)\right]^2\exp\left[\dfrac{\Gamma}{c}(x_1+x_2-2\max\{0,x_1,x_2\})\right] & \text{for } x_i<L \\ 0 \quad \text{else.}\end{cases} \tag{23}$$

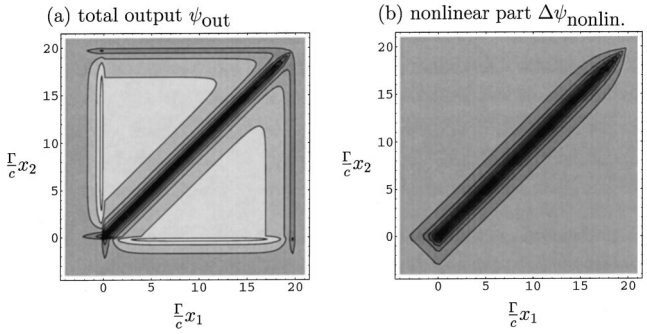

FIG. 2. Contour plots of (a) the output wave function $\psi_{\rm out}(x_1,x_2)$ and (b) the nonlinear component $\Delta\psi_{\rm nonlin}(x_1,x_2)$ of the output for a resonant rectangular input wave packet of length $L=20c/\Gamma$. The contour shading corresponds to amplitudes ranging from $-4/L$ for black to $+2/L$ for white. The dark gray shading at the edges of the graphs correspond to zero amplitude. The light gray shading of the triangular plateau regions in (a) correspond to an amplitude of $1/L$ equal to the input amplitude of the rectangular wave packet.

Figure 2 shows the output wave function $\psi_{\rm out}$ and the nonlinear component $\Delta\psi_{\rm nonlin}$ at an input pulse length of $L=20c/\Gamma$. The most remarkable feature of the nonlinear contribution is its localization around $x_1=x_2$. This is a direct consequence of the local interaction between the two photons.

A detailed discussion of the two-time correlation originating from this spatiotemporal locality of the interaction is given elsewhere [15]. In the present paper, we focus on the coherent properties of the two-photon wave function. For this purpose it is useful to simplify the results by assuming the limit of long pulses, $L\gg c/\Gamma$, and concentrating on the region within the pulse, $0<x_i<L$. In this limit, the photon-photon interaction becomes independent of the pulse shape effects caused by the sudden rise and fall of the rectangular pulse amplitude. The results should then apply to any pulse with an input amplitude varying slowly on a scale of c/Γ, where the pulse length parameter L defines the local photon density as $2/L$. The output amplitudes are then given by

$$\phi_{\rm out}(x_1)\phi_{\rm out}(x_2)=\frac{1}{L}, \tag{24}$$

$$\Delta\psi_{\rm nonlin}(x_1,x_2)=-\frac{4}{L}\exp\left(-\frac{\Gamma}{c}|x_1-x_2|\right), \tag{25}$$

$$\psi_{\rm out}(x_1,x_2)=\frac{1}{L}\left[1-4\exp\left(-\frac{\Gamma}{c}|x_1-x_2|\right)\right]. \tag{26}$$

In the long pulse limit, the linear part of the output wave function is nearly equal to the original input pulse. However, the nonlinear contribution reduces this overlap by scattering photons into other modes according to

$$\begin{aligned}\langle\psi_{\rm out}|\psi_{\rm in}\rangle&=1+\int dx_1dx_2\psi^*_{\rm out}(x_1,x_2)\Delta\psi_{\rm nonlin}(x_1,x_2)\\&\approx 1-\frac{4}{L}\int dx_-\exp\left(-\frac{\Gamma}{c}|x_-|\right)\\&=1-\frac{8c}{\Gamma L}.\end{aligned} \tag{27}$$

The probability that the two photons will be scattered out of the input mode is therefore approximately equal to

$$1-|\langle\psi_{\rm out}|\psi_{\rm in}\rangle|^2\approx\frac{16c}{\Gamma L}. \tag{28}$$

The long pulse limit requires that this fraction is never close to 1. However, the result can be used to define a scattering cross section for the two photons. If we think of the first photon as being in a random position within the pulse, the chance of finding the second photon within a distance $\leqslant\sigma$ should be equal to $2\sigma/L$. The interaction cross section σ for the two-photon nonlinearity can then be defined as $\sigma=8c/\Gamma$. Note that c/Γ is the coherence length of spontaneous emission from the atom. The nonlinear photon-photon interaction mediated by the two-level atom therefore appears to extend over a region eight times longer than this coherence length.

VII. ENTANGLEMENT AND FOUR-WAVE MIXING IN THE NONLINEAR COMPONENT

In the long pulse limit, the input mode is very nearly a plane wave resonant with the two-level atom ($k=0$). It is therefore possible to describe the scattering effect as a four-wave mixing effect changing the photon frequencies from $k_0=0$ to $+k$ and $-k$, respectively. The k-space representation of the output wave packet can be obtained by using the local Fourier transform in the spatial region from $x_i=0$ to $x_i=L$ given by

$$\begin{aligned}\psi_{\rm out}(k_1,k_2)&=\frac{1}{L}\int_0^L dx_1dx_2\exp(-ik_1x_1)\\&\quad\times\exp(-ik_2x_2)\psi_{\rm out}(x_1,x_2)\\&\approx\delta_{k_1,0}\delta_{k_2,0}-\frac{8\Gamma c}{L(\Gamma^2+c^2k_1^2)}\delta_{k_1,-k_2},\end{aligned} \tag{29}$$

where k_i can have values equal to integer multiples of $2\pi/L$. Note that this discretization of k_i is necessary to preserve the correct normalization of the quantum state. The phase-matching conditions of four-wave mixing is expressed in Eq. (29) as a Kronecker delta $\delta_{k_1,-k_2}$ ensuring that the sum of k_1 and k_2 is indeed zero. As a result of this strong correlation between k_1 and k_2, the k-space representation of the two-photon output is the Schmidt decomposition of the entangled state [17],

$$|\psi_{\rm out}\rangle=|k_1=0;k_2=0\rangle-\sum_k\frac{8\Gamma c}{L(\Gamma^2+c^2k^2)}|k;-k\rangle. \tag{30}$$

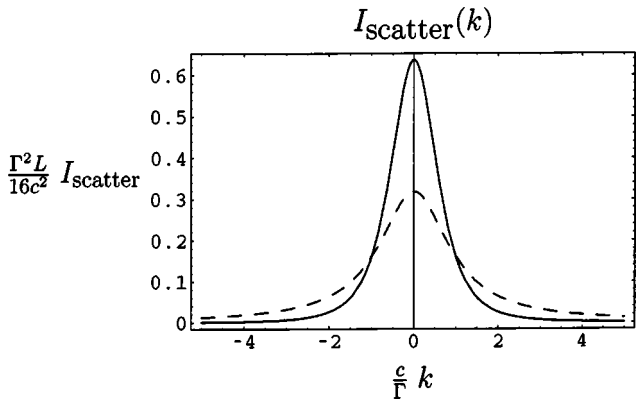

FIG. 3. Frequency spectrum I_{scatter} of the photons in the nonlinear component $\Delta\psi_{\mathrm{nonlin}}$. The intensity I and the frequency k have been scaled in such a way that the area of the spectral line in the graph is equal to one. The dashed line shows the Lorentzian line of spontaneous emission from the two-level atom derived from the same model [14]. Note that the area of this line is also one. The comparison shows that the spectrum of photons scatters by spontaneous four-wave mixing at the single atom is narrower than the spectrum of spontaneous emission.

According to this representation of the two-photon state, the single-photon density matrix can be written as a mixture of k eigenstates with

$$\hat{\rho}=\left(1-\frac{16c}{\Gamma L}\right)|k=0\rangle\langle k=0|+\sum_k\left(\frac{8\Gamma c}{L(\Gamma^2+c^2k^2)}\right)^2|k\rangle\langle k|. \tag{31}$$

This density matrix defines the single-photon coherence of the output. In particular, the frequency spectrum of the scattered light is given by a squared Lorentzian,

$$I_{\mathrm{scatter}}(k)=\frac{1}{\Delta k}\langle k|\hat{\rho}|k\rangle=\frac{16c}{\Gamma L}\,\frac{2c\Gamma^3}{\pi(\Gamma^2+c^2k^2)^2}. \tag{32}$$

Note that the resolution factor $\Delta k=2\pi/L$ is required to adjust the normalization of the continuous spectrum $I_{\mathrm{scatter}}(k)$ to the discrete distribution given by $\hat{\rho}$. Figure 3 shows this scattering spectrum in comparison with the spontaneous emission spectrum of the two-level atom. It should be noted that the squared Lorentzian of the scattering spectrum is narrower than the Lorentzian of spontaneous emission. This spectral feature clearly distinguishes the two-photon scattering process from an incoherent sequence of absorption and reemission and may serve as an indication of spontaneous four-wave mixing in experiments where low detection efficiencies prevent an evaluation of two-photon coincidences.

As this analysis shows, the resonant nonlinear interaction of the two photons at the atom causes correlated changes in the frequencies of the photons. Since the output state is completely quantum coherent, the noise in the single-photon density matrix actually indicates entanglement between the scattered photons. This situation is quite similar to the creation of photon pairs by spontaneous parametric down-conversion. In fact, it may also be possible to create entangled photon pairs from the spontaneous four-wave mixing effect at a single-atom nonlinearity by isolating the nonlinear part of the two-photon response to a coherent input field through destructive interference with an appropriate reference pulse. This method will be discussed in the following section.

VIII. GENERATION OF PHOTON PAIRS USING COHERENT INPUT LIGHT

Spontaneous four-wave mixing can only occur if two photons interact. Moreover, phase matching requires that a photon scattered to $+k$ must always be accompanied by a photon scattered to $-k$. It is therefore possible to use the nonlinear photon-photon interaction to generate correlated photon pairs from a coherent input pulse by selecting the corresponding output ports in a spectrometer. However, even better results for photon pair creation may be achieved if the linear component is removed by interference with another coherent light field using a method similar to the one applied to parametric down-conversion in Ref. [16].

For any pulse shape defined by the wave function ϕ, it is possible to define a weak coherent state $|\alpha\rangle$ with a low average photon number $|\alpha|^2\ll 1$. This coherent state can then be expanded into components with zero, one, and two photons. Using $|\mathrm{vac}\rangle$ for the vacuum state, $|\phi\rangle$ for the single-photon pulse, and $|\phi;\phi\rangle$ for the two-photon pulse, this expansion reads

$$|\alpha\rangle\approx|\mathrm{vac}\rangle+\alpha|\phi\rangle+\frac{\alpha^2}{\sqrt{2}}|\phi;\phi\rangle+\cdots. \tag{33}$$

The unitary operator $\hat{U}$ describing the response of the two-level system can now be applied separately to the vacuum, to the single-photon state, and to the two-photon state. The vacuum state is not changed by the interaction at all ($\hat{U}|\mathrm{vac}\rangle=|\mathrm{vac}\rangle$). In the resonant long pulse limit, the single-photon component changes its phase by π, but remains nearly unchanged otherwise. However, the two-photon component is changed by the addition of $|\Delta\psi_{\mathrm{nonlin}}\rangle$. The expansion of the output state therefore reads

$$\begin{aligned}|\psi_{\mathrm{out}}\rangle&\approx|\mathrm{vac}\rangle-\alpha|\phi\rangle+\frac{\alpha^2}{\sqrt{2}}|\phi;\phi\rangle+\frac{\alpha^2}{\sqrt{2}}|\Delta\psi_{\mathrm{nonlin}}\rangle\\&\approx|-\alpha\rangle+\frac{\alpha^2}{\sqrt{2}}|\Delta\psi_{\mathrm{nonlin}}\rangle.\end{aligned} \tag{34}$$

The linear component can therefore be represented by the weak coherent state $|-\alpha\rangle$ with the same coherence properties as the original pulse. This coherent pulse can be removed by destructive interference with a much stronger reference pulse of the same shape at a high reflectivity beam splitter. Note that the high reflectivity of the beam splitter is necessary to avoid quantum noise effects in the interaction that would appear as photon losses in the final output. If these

conditions are met, the destructive interference may be represented by the displacement operator $\hat{D}(\alpha)=\exp(\alpha\hat{a}^{\dagger}-\alpha^{*}\hat{a})$. For $|\alpha|\ll 1$, this operator is only slightly different from $\hat{1}$, but it does have the fundamental property that $\hat{D}(\alpha)|-\alpha\rangle=|\mathrm{vac}\rangle$. The final output therefore reads

$$\hat{D}(\alpha)|\psi_{\mathrm{out}}\rangle\approx\hat{D}(\alpha)|-\alpha\rangle+\hat{D}(\alpha)\frac{\alpha^{2}}{\sqrt{2}}|\Delta\psi_{\mathrm{nonlin}}\rangle$$

$$\approx|\mathrm{vac}\rangle+\frac{\alpha^{2}}{\sqrt{2}}|\Delta\psi_{\mathrm{nonlin}}\rangle. \tag{35}$$

This output wave function now contains only a zero and a two-photon component. The one-photon component has been eliminated by the interference effects at the high reflectivity beam splitter. It is therefore possible to generate entangled photon pairs with a two-photon wave function described by $\Delta\psi_{\mathrm{nonlin}}$ using a coherently driven dissipation-free two-level atom and an interferometric setup. The average number of photon pairs created in each pulse is then given by

$$\frac{|\alpha|^{4}}{2}\langle\Delta\psi_{\mathrm{nonlin}}|\Delta\psi_{\mathrm{nonlin}}\rangle=\frac{8c}{\Gamma L}|\alpha|^{4}. \tag{36}$$

In the long pulse limit, it is possible to approximate continuous input light as a sequence of rectangular pulses of length $L\gg c/\Gamma$. The intensity of the pump light is then given by $I_{\mathrm{in}}=c|\alpha|^{2}/L$ and the rate of pair creation R_{pair} is given by the average number of pairs per pulse divided by the pulse duration L/c. The result of this estimate reads

$$R_{\mathrm{pair}}=\frac{8}{\Gamma}I_{\mathrm{in}}^{2}, \tag{37}$$

where higher-order many-photon effects are negligible if $I_{\mathrm{in}}\ll\Gamma$. The pair-creation rate is therefore also limited to $R_{\mathrm{pair}}\ll\Gamma$. However, Γ is usually in the range of nanoseconds, so considerable pair rates should be possible.

According to Eq. (30), the quantum state of the emitted photon pair can be written as

$$|\Delta\psi_{\mathrm{nonlin}}\rangle=-\sum_{k}\frac{8\Gamma c}{L(\Gamma^{2}+c^{2}k^{2})}|k;-k\rangle, \tag{38}$$

where k represents the discretized k space with $\Delta k=2\pi/L$. In real-space representation, the same entanglement is expressed by the coefficients $\Delta\psi_{\mathrm{nonlin}}(x_1,x_2)$ given by Eq. (25). These representations show the same time-frequency correlations as a phase-matched parametric down-conversion, that is, $x_1\approx x_2$ and $k_1=-k_2$. It may therefore be possible to use photon pairs created by four-wave mixing in applications similar to those of down-converted photons.

IX. CONCLUSIONS

We have described the spatiotemporal dynamics of a one-dimensional light field interacting with a single two-level atom for input states with up to two-photons. In the case of a resonant two-photon input, the interaction at the atom results in spontaneous four-wave mixing effects, scattering the photons to higher and lower frequencies. Since this scattering effect is fully quantum coherent, the resulting output state is entangled in frequency and time.

For a coherent-state input, it is possible to remove the linear single-photon and two-photon components by destructive interference with a reference pulse. The remaining output then consists of the vacuum state and a small contribution from the nonlinear two-photon component. This output is very similar to the output from spontaneous parametric down-conversion. It may therefore be possible to realize a source of entangled photon pairs using the spontaneous four-wave mixing effects at a single two-level atom.

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