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 \( \omega = 
k \), 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 eigen-modes 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

\[
\hat{H}_{1D} = \hat{H}_{\text{prop}} + \hat{H}_{\text{abs}},
\]

with

\[
\hat{H}_{\text{prop}} = \int dk \ h c k \ \hat{b}^\dagger_k \hat{b}_k,
\]

\[
\hat{H}_{\text{abs}} = \int dk \ h \sqrt{\frac{\kappa}{\pi}} (\hat{b}^\dagger_k \hat{\sigma}_+ + \hat{\sigma}^\dagger_k \hat{b}_k),
\]

(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 \( r = 0 \).
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 \( \Gamma \). 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 frequency of the atom has been set to zero. Note that this can then be described by the components

\[
\rho_{\alpha} = c \kappa_0,
\]

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

\[
\frac{d}{dt} \psi(k;t) = -i c \kappa \psi(k;t) - i \frac{c \Gamma}{\pi} \psi(E;t),
\]

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

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

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

The equation for the propagating field then reads

\[
\frac{d}{dt} \psi(r;t) = -c \frac{\partial}{\partial r} \psi(r;t) - i \frac{2 \Gamma}{c} \delta(r) \psi(E;t).
\] (4)

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

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

\[
\frac{d}{dt} \psi(E;t) = -\Gamma \psi(E;t) - i \frac{\sqrt{2 \Gamma}}{c} \psi(r \to -0; t).
\] (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 \( t_{\text{in}} \) to any final time \( t_{\text{out}} \). In particular, the output field within \( 0 < r < c(t_{\text{out}} - t_{\text{in}}) \) for \( \psi(E;t_{\text{in}}) = 0 \) is given by

\[
\psi(r; t_{\text{out}}) = \psi(r-c(t_{\text{out}} - t_{\text{in}}); t_{\text{in}}) - i \frac{\sqrt{2 \Gamma}}{c} \psi(E; t_{\text{out}} - r/c)
\]

\[
= \psi(r-c(t_{\text{out}} - t_{\text{in}}); t_{\text{in}}) - \frac{\Gamma}{c} \int_{r-c(t_{\text{out}} - t_{\text{in}})}^{0} dr' \exp \left( -\frac{\Gamma}{c} (r-r' - c(t_{\text{out}}-t_{\text{in}})) \right) \psi(r'; t_{\text{in}}).
\] (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, \( \psi(\mathbf{r}_1, \mathbf{r}_2) \), the amplitudes for one photon in free space and one photon at the atom, \( \psi(\mathbf{r}_1, \mathbf{E}) \) or \( \psi(\mathbf{E}, \mathbf{r}_2) \), 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 \( \hat{a}_\sigma \) 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 terms in Eq. (2), using the single-photon results and setting all contributions of double excitation to zero \( \psi(E, E) = 0 \). For a two-level atom, 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

\[
\psi_{\text{in}}(x) = \lim_{t_{\text{in}} \to -\infty} \psi(x = x_0 + c t_{\text{in}}; t_{\text{in}}),
\]

\[
\psi_{\text{out}}(x) = \lim_{t_{\text{out}} \to +\infty} \psi(x = x_0 + c t_{\text{out}}; t_{\text{out}}).
\]

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

\[
\psi_{\text{out}}(x) = \int_{-\infty}^{\infty} dx' U_1(x; x') \psi_{\text{in}}(x').
\]

The single-photon response function reads

\[
U_1(x; x') = \begin{cases} 
\delta(x' - x) - \frac{\Gamma}{c} \exp \left[ - \frac{\Gamma}{c} (x' - x) \right] & \text{for } x \leq x' \\
0 & \text{for } x > x'.
\end{cases}
\]

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

\[
\psi_{\text{in}}(x_1, x_2) = \lim_{t_{\text{in}} \to -\infty} \psi(x_1 = x_1 + c t_{\text{in}}, x_2 = x_2 + c t_{\text{in}}; t_{\text{in}}),
\]

\[
\psi_{\text{out}}(x_1, x_2) = \lim_{t_{\text{out}} \to +\infty} \psi(x_1 = x_1 + c t_{\text{out}}, x_2 = x_2 + c t_{\text{out}}; t_{\text{out}}).
\]

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

\[
\psi_{\text{out}}(x_1, x_2) = \int_{-\infty}^{\infty} dx'_1 dx'_2 U_2(x_1, x_2; x'_1, x'_2) \psi_{\text{in}}(x'_1, x'_2).
\]

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,
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 $\Delta U_{\text{nonlin}}$ such that

$$U_{\text{lin}}(x_1, x_2; x'_1, x'_2) = U_1(x_1; x'_1)U_1(x_2; x'_2).$$

(14)

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

$$U_2(x_1, x_2; x'_1, x'_2) = U_1(x_1; x'_1)U_1(x_2; x'_2) + \Delta U_{\text{nonlin}}(x_1, x_2; x'_1, x'_2).$$

(15)

$$\Delta U_{\text{nonlin}}(x_1, x_2; x'_1, x'_2) = \left\{ \begin{array}{ll}
- \frac{4\Gamma^2}{c^2} \exp \left[ - \frac{\Gamma}{c} (x'_1 - x_1) \right] & \exp \left[ - \frac{\Gamma}{c} (x'_2 - x_2) \right] \quad \text{for} \ max \{ x_1, x_2 \} < \min \{ x'_1, x'_2 \} \\
0 & \text{else},
\end{array} \right.$$

(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 $\Delta U_{\text{nonlin}}$ 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

$$\psi_{\text{in}}(x_1, x_2) = \phi_{\text{in}}(x_1) \phi_{\text{in}}(x_2),$$

(17)

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

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

(18)

where $\phi_{\text{out}}$ describes the linear single-photon response given by

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

(19)

and the nonlinear contribution is directly obtained from

$$\Delta \psi_{\text{nonlin}}(x_1, x_2) = \int_{-\infty}^{\infty} dx'_1 dx'_2 \Delta U_{\text{nonlin}}(x_1, x_2; x'_1, x'_2) \times \phi_{\text{in}}(x'_1) \phi_{\text{in}}(x'_2).$$

(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

$$\phi_{\text{in}}(x) = \begin{cases} 
\frac{1}{\sqrt{L}} & \text{for } 0 < x < L \\
0 & \text{else}
\end{cases}$$

(21)

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

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

(22)

and

$$\Delta \psi_{\text{nonlin}}(x_1, x_2) = \begin{cases} 
\frac{4}{L} \left[ 1 - \exp \left( - \frac{\Gamma L}{c} \right) \right]^2 \exp \left( \frac{\Gamma}{c} (x_1 + x_2 - 2 \max \{ 0, x_1, x_2 \}) \right) & \text{for } x, L
\end{cases}$$

(23)
Entanglement and Four-Wave Mixing Effects

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

\[
\psi_{\text{out}}(k_1, k_2) = \frac{1}{L} \int_0^L dx_1 dx_2 \exp(-ik_1 x_1) \times \exp(-ik_2 x_2) \psi_{\text{out}}(x_1, x_2),
\]

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 \( |\psi_{\text{out}}\rangle \),

\[
|\psi_{\text{out}}\rangle = |k_1 = 0; k_2 = 0\rangle - \sum_k \frac{8\Gamma c}{L(\Gamma^2 + c^2 k^2)} |k; -k\rangle.
\]
The output. In particular, the frequency spectrum of the scattered photons indicates entanglement between the scattered light and the single-photon coherence of the output. This frequency spectrum actually indicates entanglement between the scattered photons. This situation is quite similar to the creation of photon pairs by spontaneous parametric down-conversion. Moreover, phase matching requires 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 \( \phi \), 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 \( |\text{vac}\rangle \) for the vacuum state, \( |\phi\rangle \) for the single-photon pulse, and \( |\phi;\phi\rangle \) for the two-photon pulse, this expansion reads

\[
|\alpha\rangle \approx |\text{vac}\rangle + \alpha |\phi\rangle + \frac{\alpha^2}{\sqrt{2}} |\phi;\phi\rangle + \cdots.
\]

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 (\( \hat{U}|\text{vac}\rangle = |\text{vac}\rangle \)). In the resonant long pulse limit, the single-photon component changes its phase by \( \pi \), but remains nearly unchanged otherwise. However, the two-photon component is changed by the addition of \( |\Delta \psi_{\text{nonlin}}\rangle \). The expansion of the output state therefore reads

\[
|\psi_{\text{out}}\rangle \approx |\text{vac}\rangle - \alpha |\phi\rangle + \alpha^2 |\phi;\phi\rangle + \frac{\alpha^2}{\sqrt{2}} |\Delta \psi_{\text{nonlin}}\rangle.
\]

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
In the long pulse limit, it is possible to approximate continuation $\hat{D}(\alpha) = \exp(\alpha \hat{a}^\dagger - \alpha^* \hat{a})$. For $|\alpha| \ll 1$, this operator is only slightly different from $\hat{1}$, but it does have the fundamental property that $\hat{D}(\alpha)|-\alpha\rangle = |\text{vac}\rangle$. The final output therefore reads

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

$$= |\text{vac}\rangle + \frac{\alpha^2}{\sqrt{2}} |\Delta \psi_{\text{nonlin}}\rangle. \quad (35)$$

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 $\Delta \psi_{\text{nonlin}}$ 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

$$|\alpha|^4 \langle \Delta \psi_{\text{nonlin}} | \Delta \psi_{\text{nonlin}} \rangle = \frac{8c}{\Gamma L} |\alpha|^4. \quad (36)$$

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 $I_{\text{in}} = c |\alpha|^2/L$, and the rate of pair creation $R_{\text{pair}}$ is given by the average number of pairs per pulse divided by the pulse duration $L/c$. The result of this estimate reads

$$R_{\text{pair}} = \frac{8}{\Gamma} I_{\text{in}}^2. \quad (37)$$

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

**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.