Nonperturbative quantum solutions to resonant four-wave mixing of two single-photon wave packets

Mattias Johnsson and Michael Fleischhauer
Fachbereich Physik, Universität Kaiserslautern, D-67663 Kaiserslautern, Germany

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We analyze both analytically and numerically the resonant four-wave mixing of two co-propagating single-photon wave packets. We present analytic expressions for the two-photon wave function, and show that quantum solutions exist which display a shape-preserving oscillatory exchange of excitations between the modes. Potential applications including quantum-information processing are discussed.

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I. INTRODUCTION

The cancellation of resonant linear absorption and refraction via electromagnetically induced transparency (EIT) [1,2] has led to a range of new possibilities in nonlinear optics. One important application is optical frequency mixing close to atomic resonances which allows the use of the enhanced nonlinear interaction without suffering from linear absorption and refraction. It has been predicted that EIT could lead to a new regime of nonlinear optics on the level of few light quanta [3–7]. Several schemes for resonant nonlinear processes have been proposed and analyzed, both theoretically and experimentally [6]. A particularly interesting system is the resonant four-wave mixing using atoms with a double-Λ configuration [8–11]. Efficient frequency conversion, generation of squeezing [12], as well as the possibility of mirrorless oscillations [13] with extremely low thresholds and narrow linewidth have been predicted [14] and, in part, experimentally observed [15].

Most theoretical and experimental studies of resonant nonlinear processes have been carried out for classical fields or assuming small quantum fluctuations. For one-dimensional setups where common comoving frames exist, full analytical solutions have been derived for the interaction of classical pulses in the adiabatic limit [16]. Quantum aspects of resonant nonlinear processes, such as the generation of squeezing, have been discussed almost exclusively within linearization approximations [17]. In view of the potential for an efficient nonlinear interaction on the level of few photons, however, a full quantum-theoretical analysis of these systems is necessary. In addition, in order to take into account finite-size effects which become increasingly important in the few-photon regime, and to analyze the potential for quantum-information processing, such a quantum analysis has to go beyond linearization approaches. Here, very little work has been done, the few exceptions being the integrable models of resonantly enhanced Kerr interaction [18] and photon blockade [3,19].

In a previous paper [7], we have shown numerically that if single-mode fields are considered, it is possible to use an atomic vapor in a four-wave mixing double-Λ configuration to obtain full conversion from two input fields into two generated fields within a few centimeters of interaction length, even if the input fields only consist of single-light quanta. The treatment was fully quantum, but being restricted to a single-mode analysis propagation effects of wave packets were not considered. In the present paper, we extend this study by undertaking a multimode analysis in one spatial dimension and for copropagating fields. In order to keep the problem tractable, we restrict ourselves to the special case of two single-photon wave packets as inputs. For this case, we obtain simple analytic solutions and compare them with numerical simulations. We show that quantum solutions exist, which display an oscillatory exchange of excitation between the two input and the two generated fields. Finally, we discuss briefly the possible applications of single-photon four-wave mixing to quantum-information processing and entanglement generation.

II. SYSTEM AND EFFECTIVE FIELD EQUATIONS

The situation we consider is resonantly enhanced four-wave mixing in the modified double-Λ system shown in Fig. 1. Note that a five-level atomic system is used instead of the original four-level system put forward in Refs. [8,13]. This is due to the fact that in the four-level system, the finite detuning Δ is associated with an ac Stark effect, which leads to intensity-dependent dynamical phase shifts of the fields. These phase shifts are of minor consequence in the case where the fields are counterpropagating [20], but for co-

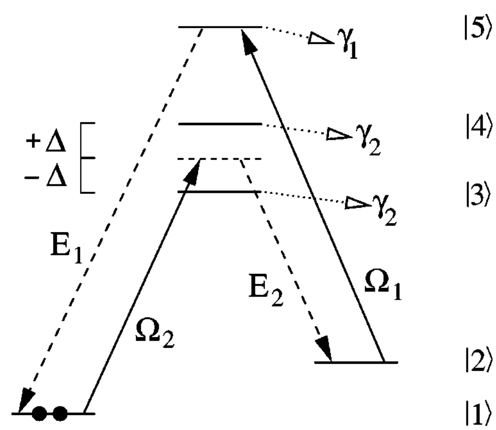

FIG. 1. Four-wave mixing in a modified double-Λ system with $\mathrm{sgn}(d_{42}/d_{41})] = -\mathrm{sgn}(d_{32}/d_{31})$, with d_{ij} being the dipole moment of the $|i\rangle - |j\rangle$ transition.

propagating fields these have a detrimental influence, leading to impaired phase matching and inefficient frequency conversion. As shown in Ref. [21], these phase shifts are eliminated in the five-level scheme when the relative sign of the dipole moments of the $|4\rangle \rightarrow |2\rangle, |1\rangle$ transitions is opposite to that of the $|3\rangle \rightarrow |2\rangle, |1\rangle$ transitions.

The two fields with carrier frequencies $\nu_{\Omega 1}$ and $\nu_{\Omega 2}$ and slowly varying amplitudes Ω_1 and Ω_2 are initially excited and form the pump fields. The other fields of carrier frequencies ν_{E1} and ν_{E2} and slowly varying amplitudes E_1 and E_2 are generated during the interaction process and are assumed to be initially zero. Ω_1 and E_1 are taken to be exactly on resonance, i.e., $\nu_{\Omega 1} = \omega_{52}$, $\nu_{E_1} = \omega_{51}$, while the other two fields are detuned by an amount Δ, i.e., $\nu_{\Omega 2} = \omega_{41} - \Delta = \omega_{31} + \Delta$ and $\nu_{E2} = \omega_{42} - \Delta = \omega_{32} + \Delta$. A finite detuning Δ, large compared to the Rabi frequencies, the Doppler broadening, and the decay rates from the excited states, is necessary to maximize the ratio of nonlinear gain to linear absorption. Decay from the two lower levels is considered to be negligible and all fields have the same propagation direction.

Because of energy conservation, there is an overall four-photon resonance, i.e., $\nu_{\Omega 1} + \nu_{\Omega 2} = \nu_{E1} + \nu_{E2}$. It can be shown that the contributions of the resonant transitions to the linear refractive index vanish if the fields are pairwise in the two-photon resonance. Phase matching will thus favor two-photon resonance, and we assume that this condition is fulfilled for the carrier frequencies of the four pulses, i.e., $\nu_{E1} - \nu_{\Omega 1} = \nu_{\Omega 2} - \nu_{E2} = \omega_{21}$.

Extending the analysis of Ref. [7] to a multimode description, the interaction can be described by the effective adiabatic Hamiltonian [22]

$$H_{\text{int}} = \frac{\hbar g c}{\Delta} \int dz \, \frac{\hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_2 + \hat{E}_1^\dagger \hat{E}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_2}{\hat{\Omega}_1^\dagger \hat{\Omega}_1 + \hat{E}_1^\dagger \hat{E}_1}. \tag{1}$$

The denominator commutes with the numerator and should of course be read as premultiplication or postmultiplication by $(\hat{\Omega}_1^\dagger \hat{\Omega}_1 + \hat{E}_1^\dagger \hat{E}_1)^{-1}$. Writing the Hamiltonian in the form above, however, highlights the resonant nature of the interaction. $\hat{\Omega}_1(z), \hat{\Omega}_1^\dagger(z)$, etc., denote dimensionless, slowly varying (both in time and space) positive- and negative-frequency components of the corresponding electric fields

$$\hat{E}_j(z) = \frac{1}{\sqrt{L}} \sum_k \hat{a}_{jk} e^{ikz} e^{i\nu_{Ej}(t-z/c)}, \tag{2}$$

$$\hat{\Omega}_j(z) = \frac{1}{\sqrt{L}} \sum_k \hat{b}_{jk} e^{ikz} e^{i\nu_{\Omega j}(t-z/c)}. \tag{3}$$

L is the quantization length and $k \geqslant -\nu/c$ is the wave-vector component in the z-direction relative to ν/c. In the derivation of Eq. (1), the rotating wave approximation was used in the atom-field interaction. This is only justified if all the fields change neither over times of the order of the oscillation period nor over distances of the order of the carrier wavelength. Thus, when discussing the local form of the approximate interaction operator, one should remember that a spatial coarse graining over distances larger than the wavelength is implied. The four fields are assumed to have either sufficiently different carrier frequencies or different polarizations. Thus, operators corresponding to different fields commute. The commutator between positive- and negative-frequency components can be approximated by a spatial δ function

$$[\hat{E}_i(z), \hat{E}_j^\dagger(z')] = \frac{\delta_{ij}}{L} \sum_k e^{ik(z-z')} \approx \delta_{ij} \delta(z-z'). \tag{4}$$

We furthermore assume in Eq. (1) that all the four transitions give rise to the same coupling $g = N d_i^2 \omega_i / (2\hbar c \epsilon_0) = 3N\lambda^2 \gamma / 8\pi$, where N is the atomic number density, λ the typical wavelength of the fields, and γ the typical radiative decay rate.

The structure of the denominator results from the saturation of the two-photon transition $|1\rangle$-$|2\rangle$, whose coherence lifetime is taken to be infinite. If a finite decay rate γ_0 of the $|1\rangle$-$|2\rangle$ coherence is taken into account, a term proportional to $\gamma\gamma_0$ has to be added in the denominator.

The nonpolynomial character of the interaction Hamiltonian causes the nonlinear coupling to behave unusually. As shown in Ref. [7], the interaction increases with decreasing pump field strength, making effective nonlinear frequency conversion possible even for single photons. In the derivation of the effective Hamiltonian in Ref. [7], adiabatic conditions were assumed. This limits the applicability of Eq. (1) in the multimode case to sufficiently long pulses. A discussion of nonadiabatic corrections and their effect on the propagation of the pulses is, however, outside of the scope of the present paper and will be discussed elsewhere.

As shown in the Appendix, the slowly varying amplitudes of the electric field obey

$$\partial_t \hat{E}_j(z,t) = -c \partial_z \hat{E}_j(z,t) + \frac{i}{\hbar} [\hat{H}_{\text{int}}, \hat{E}_j(z,t)] \tag{5}$$

and similarly for $\hat{\Omega}_j$. Thus, from Eq. (1) we arrive at the following equations of motion:

$$(\partial_t + c\,\partial_z)\hat{E}_1 = i\kappa c \hat{\Lambda} (\hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_1 \hat{E}_2 - \hat{\Omega}_1^\dagger \hat{E}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_1 \hat{\Omega}_2) \hat{\Lambda}, \tag{6}$$

$$(\partial_t + c\,\partial_z)\hat{\Omega}_1 = i\kappa c \hat{\Lambda} (\hat{E}_1^\dagger \hat{E}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_1 \hat{\Omega}_2 - \hat{E}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_1 \hat{E}_2) \hat{\Lambda},$$

$$(\partial_t + c\,\partial_z)\hat{E}_2 = -i\kappa c \hat{\Lambda} \hat{E}_1^\dagger \hat{\Omega}_1 \hat{\Omega}_2,$$

$$(\partial_t + c\,\partial_z)\hat{\Omega}_2 = -i\kappa c \hat{\Lambda} \hat{\Omega}_1^\dagger \hat{E}_1 \hat{E}_2,$$

where $\kappa = g/\Delta$ and $\hat{\Lambda} = (\hat{\Omega}_1^\dagger \hat{\Omega}_1 + \hat{E}_1^\dagger \hat{E}_1)^{-1}$.

These equations admit four independent constants of motion:

$$(\partial_t + c\,\partial_z)(\hat{\Omega}_1^\dagger \hat{\Omega}_1 + \hat{E}_1^\dagger \hat{E}_1) = 0, \tag{7}$$

$$(\partial_t + c\,\partial_z)(\hat{\Omega}_2^\dagger \hat{\Omega}_2 + \hat{E}_2^\dagger \hat{E}_2) = 0,$$

$$(\partial_t + c\partial_z)(\hat{\Omega}_1^\dagger \hat{\Omega}_1 - \hat{\Omega}_2^\dagger \hat{\Omega}_2) = 0,$$

$$(\partial_t + c\partial_z)(\hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_2 + \hat{\Omega}_1 \hat{\Omega}_2 \hat{E}_1^\dagger \hat{E}_2^\dagger) = 0,$$

which represent the quantum analogs of the Manley-Rowe relations and additionally the conservation of the relative phase between the fields.

If we assume that the input fields consist of two single-photon wave packets in $\hat{\Omega}_1$ and $\hat{\Omega}_2$, then it is clear, due to the constants of motion (7), that the state of the system can be represented at all times by

$$|\varphi(t)\rangle = \sum_{k,k'} \xi_{kk'}(t)|1_k 1_{k'} 00\rangle + \sum_{k,k'} \eta_{kk'}(t)|001_k 1_{k'}\rangle, \tag{8}$$

where $|n_k m_{k'} p_{k''} q_{k'''}\rangle$ denotes n photons in the kth mode of $\hat{\Omega}_1$, m photons in the k'th mode of $\hat{\Omega}_2$, and so on.

III. FIELD INTENSITIES

Since for the case of a single-photon input, the expectation values of all fields vanish at all times, all relevant information about the state of the system is given by the mean intensities of the fields

$$\langle \varphi(t)|\hat{\Omega}_j^\dagger(z)\hat{\Omega}_j(z)|\varphi(t)\rangle, \;\; \langle \varphi(t)|\hat{E}_j^\dagger(z)\hat{E}_j(z)|\varphi(t)\rangle, \tag{9}$$

and in expressions which we term as two-photon wave functions

$$\psi_\Omega(z,z',t) = \langle 0|\hat{\Omega}_1(z)\hat{\Omega}_2(z')|\varphi(t)\rangle = \sum_{k,k'=-\infty}^{\infty} e^{2\pi i k z/L} e^{2\pi i k' z'/L} \xi_{kk'}(t), \tag{10}$$

$$\psi_E(z,z',t) = \langle 0|\hat{E}_1(z)\hat{E}_2(z')|\varphi(t)\rangle = \sum_{k,k'=-\infty}^{\infty} e^{2\pi i k z/L} e^{2\pi i k' z'/L} \eta_{kk'}(t). \tag{11}$$

$\psi_\Omega(z,z',t)$ and $\psi_E(z,z',t)$ represent the amplitude of finding the $\hat{\Omega}_1$ ($\hat{E}_1$) photon at position z and simultaneously the $\hat{\Omega}_2$ ($\hat{E}_2$) photon at position z'. The ψ's are the two-dimensional Fourier transforms from the k-space representations $\xi_{kk'}$ and $\eta_{kk'}$ into a real-space representation.

It should be noted that although we call the ψ's "wave functions," they do not individually strictly meet the required criteria as they are not normalized to unity and, as will be seen later, can exhibit discontinuities. Because the individual $\psi_i(z,z',t)$ give the amplitude of finding photons at z and z' in field i, however, referring to ψ_i as wave functions is convenient.

We first discuss the dynamics of the mean intensities of the fields. Due to the constants of motion it is sufficient to calculate, say, $\langle \hat{\Omega}_1^\dagger \hat{\Omega}_1\rangle$:

$$\begin{aligned}(\partial_t + c\partial_z)\langle \varphi|\hat{\Omega}_1^\dagger \hat{\Omega}_1|\varphi\rangle &= \left\langle \varphi \left| \frac{i}{\hbar}[\hat{H}, \hat{\Omega}_1^\dagger \hat{\Omega}_1] \right| \varphi \right\rangle \\ &= i\kappa c\langle (\hat{\Omega}_1^\dagger \hat{\Omega}_1 \hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_2 \\ &\quad - \hat{\Omega}_1 \hat{\Omega}_2 \hat{E}_1^\dagger \hat{E}_2^\dagger \hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger)\hat{\Lambda}\rangle \\ &= i\kappa c\langle \hat{E}_1^\dagger \hat{E}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_2 - \hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_2\rangle,\end{aligned} \tag{12}$$

where we have dropped the common spatial coordinate z and used $\hat{\Lambda}|\varphi\rangle = |\varphi\rangle$, as well as the fact that three annihilation operators acting on $|\varphi\rangle$ give zero. Similarly, we find

$$\begin{aligned}&(\partial_t + c\partial_z)\langle \hat{E}_1^\dagger \hat{E}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_2 - \hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_2\rangle \\ &\quad = 2i\kappa c\langle \hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_2 - \hat{E}_1^\dagger \hat{E}_2^\dagger \hat{E}_1 \hat{E}_2\rangle,\end{aligned} \tag{13}$$

$$\begin{aligned}&(\partial_t + c\partial_z)\langle \hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_2 - \hat{E}_1^\dagger \hat{E}_2^\dagger \hat{E}_1 \hat{E}_2\rangle \\ &\quad = 2i\kappa c\langle \hat{E}_1^\dagger \hat{E}_2^\dagger \hat{\Omega}_1 \hat{\Omega}_2 - \hat{\Omega}_1^\dagger \hat{\Omega}_2^\dagger \hat{E}_1 \hat{E}_2\rangle.\end{aligned} \tag{14}$$

Consequently, the differential equation we must solve is

$$(\partial_t + c\partial_z)^3 \langle \hat{\Omega}_1^\dagger \hat{\Omega}_1\rangle = -4\kappa^2 c^2 (\partial_t + c\partial_z)\langle \hat{\Omega}_1^\dagger \hat{\Omega}_1\rangle. \tag{15}$$

We take the input to be two independent single-photon wave packets in $\hat{\Omega}_1$ and $\hat{\Omega}_2$ with the same spatial envelope $f_0(z)$ and vacuum in the other two fields, which corresponds to a separable initial state of the form

$$|\varphi\rangle_{\text{in}} = [\sum_k \xi_k^0 |1_k\rangle_{\Omega_1}] \otimes [\sum_k \xi_k^0 |1_k\rangle_{\Omega_2}] \otimes |0\rangle_{E1} \otimes |0\rangle_{E2}. \tag{16}$$

The ξ_k^0 are Fourier transforms of $f_0(z)$,

$$f_0(z) = \sum_k \xi_k^0 e^{2\pi i k z/L}. \tag{17}$$

Thus,

$$\langle \hat{\Omega}_1^\dagger(z,t)\hat{\Omega}_1(z,t)\rangle_{\text{in}} = \langle \hat{\Omega}_2^\dagger(z,t)\hat{\Omega}_2(z,t)\rangle_{\text{in}} = \psi_0(z-ct), \tag{18}$$

where $\psi_0(z) = f_0^2(z)$. With these initial conditions, one finds

$$\langle \hat{\Omega}_1^\dagger \hat{\Omega}_1\rangle = \langle \hat{\Omega}_2^\dagger \hat{\Omega}_2\rangle = \psi_0(z-ct)\cos^2(\kappa z), \tag{19}$$

i.e., a sinusoidal exchange of excitation between the two pump and the two generated fields. A complete conversion is achieved at $z = \pi/\kappa$. It is worthwhile noting that, as shown in Ref. [7], and contrary to the classical a dynamics, a complete conversion can only be achieved in the quantum case for initial Fock states with one or two photons in the two pump modes.

IV. DYNAMICS OF THE TWO-PHOTON WAVE FUNCTION

We proceed by calculating the two-photon wave functions. The calculation of $\psi_\Omega(z,z',t)$ can be split into two distinct cases, depending on whether $z=z'$ or $z\neq z'$. Let us first assume $z=z'$:

$$(\partial_t+c\partial_z)\langle 0|\hat{\Omega}_1\hat{\Omega}_2|\varphi\rangle=\left\langle 0\left|\frac{i}{\hbar}[\hat{H},\hat{\Omega}_1\hat{\Omega}_2]\right|\varphi\right\rangle$$
$$=-i\kappa c\langle 0|\hat{\Omega}_1\hat{\Omega}_2(\hat{\Omega}_1^\dagger\hat{\Omega}_2^\dagger\hat{E}_1\hat{E}_2+\hat{\Omega}_1\hat{\Omega}_2\hat{E}_1^\dagger\hat{E}_2^\dagger)\hat{\Lambda}|\varphi\rangle$$
$$=-i\kappa c\langle 0|\hat{E}_1\hat{E}_2|\varphi\rangle, \qquad (20)$$

where we have again used the results that $\hat{H}|0\rangle=0$, $\hat{\Lambda}|\varphi\rangle=|\varphi\rangle$, and that three annihilation operators acting on $|\varphi\rangle$ give zero. Operating on $\langle 0|\hat{E}_1\hat{E}_2|\psi\rangle$, we find

$$(\partial_t+c\partial_z)\langle 0|\hat{E}_1\hat{E}_2|\varphi\rangle=-i\kappa c\langle 0|\hat{\Omega}_1\hat{\Omega}_2|\varphi\rangle. \qquad (21)$$

Thus, to determine $\psi_\Omega(z,z,t)$, we need to solve the differential equation

$$(\partial_t+c\partial_z)^2\psi_\Omega(z,z,t)=-\kappa^2c^2\psi_\Omega(z,z,t). \qquad (22)$$

For an input consisting of two independent single-photon wave packets with the same spatial shape, as considered above, the initial two-photon wave function reads

$$\psi_\Omega(z,z',t)_{\rm in}=f(z-ct)f(z'-ct). \qquad (23)$$

We also have at the entrance of the medium

$$(\partial_t+c\partial_z)\psi_\Omega(z,z,t)|_{z=0}=-i\kappa c\langle 0|\hat{E}_1\hat{E}_2|\varphi\rangle|_{z=0}=0, \qquad (24)$$

so that solution to Eq. (22) is given by

$$\psi_\Omega(z,z,t)=\psi_0(z-ct)\cos(\kappa z), \qquad (25)$$

with $\psi_0(z)=f^2(z)$. Thus, the two-photon wave function at equal spatial points propagates through the medium modulated by a factor of $\cos(\kappa z)$. We see that after one full conversion cycle $z=\pi/\kappa$, the phase of the wave function has changed sign. This agrees with a numerical simulation of the quantum problem, the results of which are shown in Fig. 2.

We can use a similar procedure to find $\psi_E(z,z,t)=\langle 0|\hat{E}_1\hat{E}_2|\varphi\rangle$, the wave function of the generated fields. We obtain

$$\psi_E(z,z,t)=-i\psi_0(z-ct)\sin(\kappa z). \qquad (26)$$

The evolution of this field is shown in Fig. 3. Note that it is $\pi/2$ out of phase with the drive field wave functions, as expected.

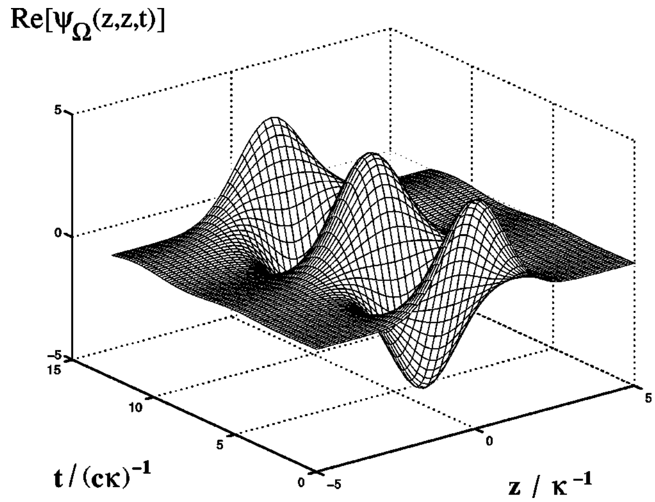

FIG. 2. 45-mode simulation of wave-function propagation in a comoving frame. Both initial wave packets are taken to be Gaussian.

For full knowledge of the state of the system, we also need to find $\psi(z,z',t)$, where $z\neq z'$. From Eq. (4) we see that operators at different points in the space commute, so that

$$(\partial_t+c\partial_z+c\partial_{z'})\langle 0|\hat{\Omega}_1(z)\hat{\Omega}_2(z')|\varphi\rangle$$
$$=\frac{i}{\hbar}\langle 0|[\hat{H},\hat{\Omega}_1(z)]\hat{\Omega}_2(z')+\hat{\Omega}_1(z)[\hat{H},\hat{\Omega}_2(z')]|\varphi\rangle. \qquad (27)$$

Expanding the Hamiltonian, normal ordering the expression and again noting the form of Eq. (8), we eventually arrive at

$$(\partial_t+c\partial_z+c\partial_{z'})\langle 0|\hat{\Omega}_1(z)\hat{\Omega}_2(z')|\varphi\rangle=0. \qquad (28)$$

Thus, the wave function of the system in the case where $z\neq z'$ is simply given by the corresponding input expression

$$\psi(z,z',t)=f(z-ct)f(z'-ct), \quad z\neq z'. \qquad (29)$$

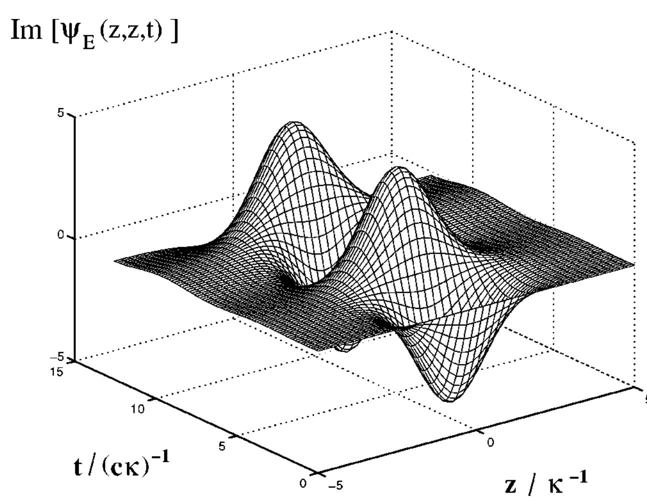

FIG. 3. Generated fields in the 45-mode simulation. Both initial wave packets are taken to be Gaussian.

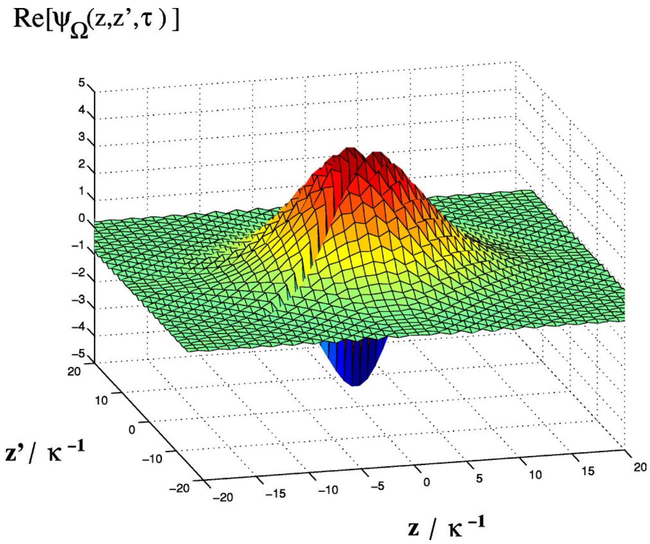

FIG. 4. Numerical 45-mode calculation of $\psi_\Omega(z,z',\tau)$, shown after one full cycle of energy transferal from pump to generated fields and back again. The sign flip for $z=z'$ is evident.

It is evident that the initial two-photon wave function at different coordinates propagates undisturbed throughout the medium, and that there is a discontinuous change in the behavior when moving away from the line $z=z'$. This is clearly seen in Fig. 4, which shows the two-dimensional wave function $\psi_\Omega(z,z',\tau)$ at time $\tau=\pi/\kappa$. This is the time required for one full conversion from the pump fields to the generated fields and back again. Essentially, we have at this time

$$\psi_\Omega(z,z',\tau)=-\psi_\Omega(z,z')_{\text{in}},\quad z=z', \tag{30}$$

$$\psi_\Omega(z,z',\tau)=\psi_\Omega(z,z')_{\text{in}},\quad z\neq z'. \tag{31}$$

This behavior can easily be understood. The two-photon wave function represents the joint probability amplitude that a measurement of the two photons will find them at positions z and z'. If they are separated, no nonlinear interaction can occur. Put in another way, since the (approximate) nonlinear interaction in Eq. (1) is local, the part of first wave packet at z can only interact with the part of the second wave packet at the same point z. Thus, only if $z=z'$, there is a conversion from the pump fields into the generated fields due to the local nonlinearity. When interpreting the discontinuity in the beaviour of the two-photon wave-function at $z=z'$, one should remember that in deriving the effective interaction Hamiltonian a spatial coarse graining was implied.

The simple sign change in the wave function for $z=z'$ hints at the possibility of using the system as a phase gate for quantum computation, as was mentioned in the single-mode case considered in Ref. [7]. One chooses the length of the nonlinear medium such that exactly one full conversion cycle can occur. If the entire wave function changes sign, we would have a system that behaves as a true phase gate: if only one of the two inputs is populated the pulse exits the medium unchanged, but if both are present a phase shift of π occurs.

However, due to the multimode nature of this problem, it is $\psi(z,z',t)$ for all z and z' which is a true reflection of the system rather than $\psi(z,z,t)$, and there is no sign change for $z\neq z'$. To see whether we can still use the system as an approximate phase gate, we consider the behavior of the state vector coefficients in Eq. (8). Returning briefly to a picture with $2N+1$ discrete modes, by using Eq. (A1) of the Appendix one can show that Eqs. (30) and (31) imply

$$\xi_{kk'}(\tau)=\xi_{kk'}(0)-\frac{2}{2N+1}\sum_{mn}\delta_{m+n,k+k'}\xi_{mn}(0). \tag{32}$$

This gives a good picture of how the different modes have mixed among themselves, and shows that in general a simple sign change of all the coefficients cannot exist. Only if we can enforce that at least temporarily only a single effective mode of the two pump fields is excited, e.g., by using a resonator setup is it possible to use the system as a phase gate.

We see, however, from Eq. (32) that after one full conversion cycle the initially factorized state (16) evolves into an entangled state between all modes of the fields $\hat{\Omega}_1$ and $\hat{\Omega}_2$. Thus, the nonlinear interaction generates entanglement.

On the other hand, if the initial state is a two-photon wave packet in an entangled state, such that the initial two-photon wave function has only a contribution for $z=z'$,

$$\psi_\Omega(z,z')|_{\text{in}}=\phi_0(z)\delta(z-z'), \tag{33}$$

only diagonal components of the two-photon wave function will ever be nonzero. According to Eqs. (25) and (26) they undergo sinusoidal oscillations

$$\psi_\Omega(z,t)=\phi_0(z-ct)\cos(\kappa z), \tag{34}$$

$$\psi_E(z,t)=-i\phi_0(z-ct)\sin(\kappa z). \tag{35}$$

The superposition of pump and generated fields

$$\Phi(z,t)\equiv\cos(\kappa z)\psi_\Omega(z,t)+i\sin(\kappa z)\psi_E(z,t)=\phi_0(z-ct) \tag{36}$$

propagates in a form-stable manner. The two-photon wave function $\Phi(z,t)$ of this quantum solution corresponds to a quasiparticle excitation

$$\hat{\Psi}^\dagger(z,t)\equiv\cos(\kappa z)\hat{\Omega}_1^\dagger(z,t)\hat{\Omega}_2^\dagger(z,t)-i\sin(\kappa z)\hat{E}_1^\dagger(z,t)\hat{E}_2^\dagger(z,t). \tag{37}$$

V. SUMMARY

In the present paper, we have presented a full quantum-theoretical treatment of resonant forward four-wave mixing for single-photon pulses. For this we have used an effective Hamiltonian derived from the interaction of the fields with an ensemble of atoms in a double-Λ configuration using an adiabatic approximation [7]. We were particularly interested in the few-photon regime, since here quantum effects dominate the dynamics and because of potential applications in quantum-information processing. Thus, we have restricted ourselves to the special case of an input consisting of two single-photon wave packets. For this case, we were able to analytically solve the propagation equations for the field intensities and two-photon wave functions which contain all relevant information about the quantum state.

We found that there is an oscillatory energy exchange between the two pump and generated fields with 100% conversion at periodic intervals of interaction. This result is characteristic for a few-photon Fock-state input; for a coherent input, complete conversion can only be achieved asymptotically for very large input power.

We have also shown that after even multiples of the conversion length the two-photon wave function $\psi(z,z',t)$ regains its initial form, while after odd multiples there is a sign flip for $z=z'$.

If the two input wave packets are not independent but are in a highly entangled state, the two-photon wave function can be made zero outside of the diagonal. It was shown that such a pair of input wave packets form a form-stable quantum solution, which is a superposition of pump and generated fields with oscillating coefficients.

The process of resonant four-wave mixing was shown to generate large entanglement between the modes forming the two single-photon wave packets. Furthermore, the nonlinear interaction strength is large enough to generate a controlled phase shift of a single photon by the presence of another one. Thus, if the number of relevant modes is at least temporarily restricted by some external means such as a resonator, the system could have interesting applications as a photonic phase gate.

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APPENDIX

To simplify the transition from a single-mode description to a multimode one, we will first consider one single-mode quantum field $\hat{a}$ and then generalize the result to our four-field system. Suppose that the single-mode Hamiltonian governing the evolution of $\hat{a}$ is given by $\hat{H}=\hbar\omega_0\hat{a}^\dagger\hat{a}+\hat{H}_{\mathrm{int}}(\hat{a},\hat{a}^\dagger)$. To go over to the multimode description, we consider an interaction region of length L, divided into $2N+1$ cells, and consider a discrete set of modes around the carrier frequency of the field, i.e., $k_n=k_0+2n\pi/L$, $-N\leqslant n\leqslant N$. We now define localized field operators (denoted by a tilde) via

$$\hat{\tilde{a}}_l=\sum_{k=-N}^{N}\hat{a}_k\exp\left[\frac{2\pi ikl}{2N+1}\right], \tag{A1}$$

$$\hat{a}_k=\frac{1}{2N+1}\sum_{l=-N}^{N}\hat{\tilde{a}}_l\exp\left[\frac{-2\pi ikl}{2N+1}\right], \tag{A2}$$

where the $\hat{a}_k$ are annihilation operators for mode k. $\hat{\tilde{a}}_l$ is related to the field strength in cell l.

These localized field operators have the commutation relations

$$[\hat{\tilde{a}}_l,\hat{\tilde{a}}_{l'}^\dagger]=(2N+1)\delta_{ll'}. \tag{A3}$$

The multimode Hamiltonian can now be written as

$$\hat{H}=\hbar\sum_k\omega_k\hat{a}_k^\dagger\hat{a}_k+\sum_l\hat{H}_{\mathrm{int}}(\hat{\tilde{a}}_l,\hat{\tilde{a}}_l^\dagger)=\frac{\hbar\omega_0}{2N+1}\sum_l\hat{\tilde{a}}_l^\dagger\hat{\tilde{a}}_l$$

$$-\hbar\sum_{ll'}\omega_{ll'}\hat{\tilde{a}}_l^\dagger\hat{\tilde{a}}_{l'}+\sum_l\hat{H}_{\mathrm{int}}(\hat{\tilde{a}}_l,\hat{\tilde{a}}_l^\dagger), \tag{A4}$$

where ω_0 is the carrier frequency and

$$\omega_{ll'}=\sum_{k=-N}^{N}\frac{2\pi kc}{(2N+1)^2L}\exp\left[\frac{2\pi ik(l-l')}{2N+1}\right]. \tag{A5}$$

Commutation relations yield the following Heisenberg equation of motion:

$$\frac{\partial}{\partial t}\hat{\tilde{a}}_l=-i\omega_0\hat{\tilde{a}}_l-i(2N+1)\sum_{l'}\omega_{ll'}\hat{\tilde{a}}_{l'}+\frac{i}{\hbar}[\hat{\tilde{H}}_{\mathrm{int}},\hat{\tilde{a}}]. \tag{A6}$$

Now, as we let $N\to\infty$ we find

$$lL/(2N+1)\to z, \tag{A7}$$

$$\hat{\tilde{a}}_l\to\hat{a}(z,t), \tag{A8}$$

$$-i(2N+1)\sum_{l'}\omega_{ll'}\hat{\tilde{a}}_{l'}\to-c\partial_z\hat{a}(z,t), \tag{A9}$$

$$[\hat{a}(z,t),\hat{a}^\dagger(z',t)]\to L\delta(z-z'). \tag{A10}$$

Introducing slowly time-varying amplitudes, we obtain

$$(\partial_t+c\partial_z)\hat{a}(z,t)=\frac{i}{\hbar}[\hat{H}_{\mathrm{int}}(t),\hat{a}(z,t)]. \tag{A11}$$

Thus, the multimode equations of motion look exactly like the single-mode equations of motion, with the exception that a $c\partial_z$ term has been added and the fields now have a spatial dependence.

Returning to the four-wave mixing situation described in this paper, we see that the interaction Hamiltonian given by Eq. (1) is of the form shown above, with the summation replaced by an integral in the continuum limit. The multimode annihilation and creation operators $\hat{E}(z)$ and $\hat{\Omega}(z)$ defined in Eqs. (2) and (3) are analogous to the localized field operators $\hat{a}(z,t)$ defined above, except for the factor of $\sqrt{L}$ inserted to ensure correct commutation relations, regardless of the quantization length. Thus, the equation of motion (5) follows from Eq. (A11) above.

[1] S.E. Harris, Phys. Today **50**(7), 36 (1997).

[2] E. Arimondo, Prog. Opt. **35**, 259 (1996); P.L. Knight, B. Stoicheff, and D.F. Walls, Philos. Trans. R. Soc. London, Ser. A **355**, 2215 (1997); J.P. Marangos, J. Mod. Opt. **45**, 471 (1998).

[3] S.E. Harris and Y. Yamamoto, Phys. Rev. Lett. **81**, 3611 (1998).

[4] M. Jain, Hui Xia, G.Y. Yin, A.J. Merriam, and S.E. Harris, Phys. Rev. Lett. **77**, 4326 (1996); S.E. Harris and L.V. Hau, *ibid.* **82**, 4611 (1999).

[5] H. Schmidt and A. Imamoglu, Opt. Lett. **21**, 1936 (1996); M.D. Lukin, and A. Imamoglu, Phys. Rev. Lett. **84**, 1419 (2000).

[6] M.D. Lukin, P.R. Hemmer, and M.O. Scully, Adv. At., Mol., Opt. Phys. **42**, 347 (2000).

[7] M.T. Johnsson, and M. Fleischhauer, Phys. Rev. A **66**, 043808 (2002).

[8] P.R. Hemmer, K.Z. Cheng, J. Kierstead, M.S. Shariar, and M.K. Kim, Opt. Lett. **19**, 296 (1994); B.S. Ham, M.S. Shariar, M.K. Kim, and P.R. Hemmer, *ibid.* **22**, 1849 (1997).

[9] S. Babin, U. Hinze, E. Tiemann, and B. Wellegehausen, Opt. Lett. **21**, 1186 (1996); S.A. Babin, E.V. Podivilov, D.A. Shapiro, U. Hinze, E. Tiemann, and B. Wellegehausen, Phys. Rev. A **59**, 1355 (1999); U. Hinze, L. Meyer, B.N. Chichkov, E. Tiemann, and B. Wellegehausen, Opt. Commun. **166**, 127 (1999).

[10] A.K. Popov and S.A. Myslivets, Kvant. Elektron. (Moscow) **24**, 1033 (1997).

[11] B. Lu, W.H. Burkett, and M. Xiao, Opt. Lett. **23**, 804 (1998).

[12] M.D. Lukin, A.B. Matsko, M. Fleischhauer, and M.O. Scully, Phys. Rev. Lett. **82**, 1847 (1999).

[13] M.D. Lukin, P.R. Hemmer, M. Löffler, and M.O. Scully, Phys. Rev. Lett. **81**, 2675 (1998); A.S. Zibrov, M.D. Lukin, and M.O. Scully, *ibid.* **83**, 4049 (1999).

[14] M. Fleischhauer, M.D. Lukin, A.B. Matsko, and M.O. Scully, Phys. Rev. Lett. **84**, 3558 (2000).

[15] A.S. Zibrov, M.D. Lukin, and M.O. Scully, Phys. Rev. Lett. **83**, 4049 (1999).

[16] E.A. Korsunsky and M. Fleischhauer, Phys. Rev. A **66**, 033808 (2002).

[17] For discussion of quantum noise in dark-state systems, see G.S. Agarwal, Phys. Rev. Lett. **71**, 1351 (1993); K.M. Gheri, D.F. Walls, and M.A. Marte, Phys. Rev. A **50**, 1871 (1994); K.M. Gheri and D.F. Walls, *ibid.* **46**, R6793 (1992); M. Jain, *ibid.* **50**, 1899 (1994); M. Fleischhauer, Phys. Rev. Lett. **72**, 989 (1994); M.D. Lukin, A.B. Matsko, M. Fleischhauer, and M.O. Scully, *ibid.* **82**, 1847 (1999).

[18] M.D. Lukin and A. Imamoğlu, Phys. Rev. Lett. **84**, 1419 (2000).

[19] A. Imamoğlu, H. Schmidt, G. Woods, and M. Deutsch, Phys. Rev. Lett. **79**, 1467 (1997); M. Dunstan, S. Rebic, S. Tan, S. Parkins, M. Collett, and D. F. Walls, in *Proceedings of Quantum Communication, Computing, and Measurement 2*, edited by P. Kumar, G. M. D'Ariano, and O. Hirota (Plenum Press, New York, 1999).

[20] M. Fleischhauer, in *Frontiers of Laser Physics and Quantum Optics*, edited by Z. Xu, S. Xie, S.-Y. Zhu, and M. O. Scully (Springer, Berlin, 2000), pp. 97–106 .

[21] M.T. Johnsson, E. Korsunsky, and M. Fleischhauer, Opt. Commun. **212**, 335 (2002).

[22] A.O. Melikyan and S.G. Saakyan, Zh. Eksp. Teor. Fiz. **76**, 1530 (1979) [Sov. Phys. JETP **49**, 776 (1979)]; A.R. Karapetyan and B.V. Kryzhanovskii, *ibid.* **99**, 1103 (1991) [*ibid.* **72**, 613 (1991)]; B. Kryzhanovsky and B. Glushko, Phys. Rev. A **45**, 4979 (1992).