Pulse-shape effects on photon-photon interactions in nonlinear optical quantum gates

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Ideally, strong nonlinearities could be used to implement quantum gates for photonic qubits by well-controlled two-photon interactions. However, the dependence of the nonlinear interaction on frequency and time makes it difficult to preserve a coherent pulse shape that could justify a single-mode model for the time-frequency degree of freedom of the photons. In this paper, we analyze the problem of temporal multimode effects by considering the pulse shape of the average output field obtained from a coherent input pulse. It is shown that a significant part of the two-photon state transformation can be derived from this semiclassical description of the optical nonlinearity. The effect of a nonlinear system on a two-photon state can then be determined from the density-matrix dynamics of the coherently driven system using input-output theory. As an example, the resonant nonlinearity of a single two-level atom is characterized. The results indicate that the most efficient nonlinear effect may not be the widely studied single-mode phase shift, but the transfer of one of the photons to an orthogonal mode distinguished by its temporal and spectral properties.

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I. INTRODUCTION

Photonic qubits based on the polarization or the transverse-mode structure of single photons are an attractive candidate for the implementation of quantum information protocols because it is relatively easy to establish and to maintain single-qubit coherences by conventional linear optics. However, the realization of well-controlled two-photon interactions remains a challenging problem on the road to larger networks and more efficient operations. Early on, it has been suggested that nonlinear materials might provide the interaction necessary to couple pairs of photons [1], and sufficiently strong nonlinear effects were demonstrated experimentally using single atoms in a high-finesse cavity [2]. Recent advances in solid-state cavity designs seem to be putting the prospect of integrated devices implementing optical nonlinear quantum gates within the reach of present technological capabilities [3–7]. However, some quite fundamental problems still need to be addressed before the proper functions of a quantum gate can be realized. In particular, it is necessary to preserve single-mode coherence, not only in polarization and in transverse-mode structure, but also in the time-frequency domain. The latter problem is fundamentally linked to the dynamics of optical nonlinearities in time and space [8–17] and has recently been identified as a critical problem in the realization of quantum gates [18–20]. Initial attempts at optimizing the pulse shape and the pulse durations focused on the possibility of obtaining a large phase flip, represented by a negative two-photon amplitude in a single intended target mode [11,21]. In that context, Koshino and Ishihara pointed out that the two-photon amplitude can be evaluated from the semiclassical response of the system [11]. This result suggests that a more detailed analysis of the relation between semiclassical field expectation values and the transformation of two-photon wave functions may be possible. Such an analysis could provide both a more thorough foundation for the evaluation of experimental results like the ones recently reported in [22,23] and a more detailed characterization and classification of the spectral and the temporal features observed in highly nonlinear devices. In the following, we therefore present a systematic analysis of the relation between the average field response obtained from a coherent input and the transformation of the two-photon component of the quantum-mechanical wave function.

In Sec. II, a quantum-mechanical formulation of the semiclassical nonlinear response is developed using field operators. In Sec. III, the nonlinear interaction is expanded in terms of its effects on photon number states of suitably defined modes. It is then possible to express the semiclassical output in terms of the wave functions of these modes. In Sec. IV, it is shown how the matrix elements of the photon number expansion can be derived from the overlap integrals of the semiclassical result. According to the results of Secs. II–IV, the nonlinear transformation has two distinct effects: a change in phase and amplitude of the two-photon component of the linear output pulse and a change in pulse shape represented by the transfer of a single photon to an orthogonal mode. In Sec. VI, the magnitude of these effects is investigated for the case of a resonant two-level system. It is shown that the conditional transfer of one photon to an orthogonal mode is much stronger than the nonlinear phase shift, suggesting that it may be more efficient to use this effect in optical quantum gates. In Sec. VII, the pulse shapes of the modes involved in the nonlinear photon transfer are presented and the dynamics of the effect is discussed. The conclusions are summarized in Sec. VIII.

II. SEMICLASSICAL CHARACTERIZATION OF A NONLINEAR QUANTUM SYSTEM

In order to analyze the quantum level effects of an optical nonlinearity, we start by considering the effects of a quantum

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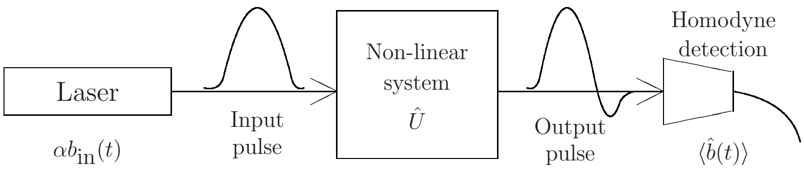

FIG. 1. Illustration of the semiclassical characterization of an optically nonlinear system using coherent input light and homodyne detection of the output pulse shape.

system on the pulse shape of a coherent input pulse. Such an input pulse can be described by a classical time-dependent field amplitude $\alpha b_{\rm in}(t)$, where $b_{\rm in}(t)$ is the wave function of a single-mode light field normalized to one photon. Hence, the input quantum state is effectively a single-mode coherent state $|\alpha\rangle$. This state interacts with the nonlinear quantum system, temporarily exciting it from its initial ground state. After this interaction, the system returns to the ground state, re-emitting any photons it might have absorbed in the process of the interaction.

If decoherence effects can be neglected, the total effect of the interaction on the quantum state of the input pulse can then be written as a unitary transformation $\hat{U}$ acting only on the state of the light field. In general, this unitary transformation acts on the full continuum of modes in free space. A complete characterization of the output state would therefore require multiphoton coherences between all frequencies or times observed in the output [9,16,18]. However, we now simplify this analysis by considering only the expectation values of the output fields $\langle \hat{b}(t) \rangle$. Experimentally, this would correspond to the averages of a homodyne measurement, as used in the pioneering work of Turchette *et al.* [2]. Figure 1 shows a schematic illustration of this semiclassical characterization of a quantum level nonlinearity. Theoretically, such averages are easily obtained from the density-matrix dynamics of the system using input-output theory, as will be explained in more detail in Sec. V.

In terms of the unitary operation $\hat{U}$ describing the effects of the nonlinear system on the quantum state of the input light, the average output field is given by the expectation value of the field operator $\hat{b}(t)$ in the time domain,

$$b_{\rm out}(t) = \langle \hat{b}(t) \rangle = \langle \alpha | \hat{U}^\dagger \hat{b}(t) \hat{U} | \alpha \rangle . \tag{1}$$

We can then proceed to analyze the specific form of the output pulse $b_{\rm out}(t)$ by expanding the output in terms of the input amplitude α. The nonlinear effect arising from photon-photon interactions is given by the third-order term in this expansion. For sufficiently small amplitudes α, the semiclassical effect of photon-photon interactions on the transformation of an input pulse $\alpha b_{\rm in}(t)$ can therefore be expressed in terms of the third-order nonlinear response,

$$b_{\rm out}(t) = \alpha b^{(1)}(t) + \alpha |\alpha|^2 b^{(3)}(t), \tag{2}$$

where $b^{(1)}(t)$ is the pulse shape of the linear output and $b^{(3)}(t)$ is the pulse shape of the third-order nonlinearity.

If there is neither decoherence nor photon loss in the system, $b^{(1)}(t)$ describes a normalized single output mode that characterizes the transformation of the one-photon wave function from $\psi_{\rm in}(t) = b_{\rm in}(t)$ to $\psi_1(t) = b^{(1)}(t)$. On the other hand, $b^{(3)}(t)$ is neither normalized nor does it represent a wave function orthogonal to $\psi_1(t)$. However, it is possible to interpret the pulse shape $b^{(3)}(t)$ as a linear superposition of a component proportional to $\psi_1(t)$ and another component proportional to a normalized wave function $\psi_2(t)$ that is orthogonal to $\psi_1(t)$. Since the wave functions $\psi_1(t)$ and $\psi_2(t)$ are orthogonal, they represent two distinct modes in the time-frequency continuum. It is therefore possible to quantize the light field by assigning separate annihilation operators $\hat{a}_1$ and $\hat{a}_2$ to these orthogonal modes. The output pulse shape $b_{\rm out}(t)$ can then be expressed in terms of the two orthonormal wave functions $\psi_1(t)$ and $\psi_2(t)$ and the expectation values $\langle \hat{a}_1 \rangle$ and $\langle \hat{a}_2 \rangle$ of their complex amplitudes,

$$b_{\rm out}(t) = \langle \hat{a}_1 \rangle \psi_1(t) + \langle \hat{a}_2 \rangle \psi_2(t). \tag{3}$$

The expectation values can be determined by analyzing the pulse-shape functions $b^{(1)}(t)$ and $b^{(3)}(t)$ in Eq. (2). Thus, the semiclassical representation of the nonlinearity in terms of field expectation values can be interpreted within a fully quantum-mechanical model focusing on the two modes defined by the pulse shapes observed in the output field.

III. QUANTUM MECHANICS OF THE PHOTON-PHOTON INTERACTION

In the previous section, we have shown that the third-order nonlinear response to a coherent input pulse can be described in terms of two quantized modes defined by the linear and the nonlinear parts of the average output field $b_{\rm out}(t)$. We can now use this two-mode representation to formulate the matrix elements of the unitary transformation $\hat{U}$ that describes the transitions between the photon number states of these modes. If photon losses can be neglected, the unitary transformation preserves the total photon number and we can look at each subspace of fixed total photon number separately. In particular, the vacuum state will not be changed by the interaction with the system, so the effect of $\hat{U}$ on the zero-photon subspace is simply given by

$$\hat{U} | \text{vac} \rangle = | \text{vac} \rangle . \tag{4}$$

For low-intensity fields ($\alpha \ll 1$), the expectation values $\langle \hat{b}(t) \rangle$ of the field amplitudes are given by the coherence between the single-photon wave function at t and the vacuum. Therefore, the linear part of the semiclassical pulse-shape transformation is equivalent to the transformation of the single-photon wave function. Specifically, the single-photon input wave function $\psi_{\rm in}(t) = b_{\rm in}(t)$ is transformed into the linear output mode wave function $\psi_1(t) = b^{(1)}(t)$. In terms of the annihilation operators of the input mode $\hat{a}_{\rm in}$ and the linear output mode $\hat{a}_1$, the effect of the unitary transformation $\hat{U}$ in the single-photon subspace can therefore be expressed as

$$\hat{U}(\hat{a}_{\rm in}^\dagger | \text{vac} \rangle) = \hat{a}_1^\dagger | \text{vac} \rangle . \tag{5}$$

The transformation in the two-photon subspace is more complicated, because the photon-photon interaction generally entangles the wave functions of the two photons [10]. However, we know from Eq. (2) that a significant part of the two-photon output wave function can be described in terms of the modes $\hat{a}_1$ and $\hat{a}_2$. We can therefore expand the effect of $\hat{U}$ in the two-photon subspace in terms of these output modes. Specifically, the contributions to the expectation values of the annihilation operators in Eq. (3) originate from an inner product of a one-photon wave function generated by annihilating a photon from the output two-photon wave function and the output one-photon state. As a result, the components of the two-photon output contributing to the averages in Eq. (3) must have at least one photon in the linear output mode $\hat{a}_1$. The components of the two-photon output that contribute to $b_{\text{out}}(t)$ are therefore (i) a state where both photons are in mode $\hat{a}_1$ and (ii) a state where one photon is in mode $\hat{a}_1$ and one photon is in mode $\hat{a}_2$. In addition, there is a third component $|\text{rest}\rangle$ that does not contribute to $b_{\text{out}}(t)$ because it does not have any photons in the linear output mode $\hat{a}_1$. Thus, the expansion of the two-photon output state can be written as

$$\hat{U}\left(\frac{1}{\sqrt{2}}\hat{a}_{\text{in}}^{\dagger}\hat{a}_{\text{in}}^{\dagger}|\text{vac}\rangle\right) = \frac{C_{11}}{\sqrt{2}}\hat{a}_1^{\dagger}\hat{a}_1^{\dagger}|\text{vac}\rangle + C_{12}\hat{a}_1^{\dagger}\hat{a}_2^{\dagger}|\text{vac}\rangle + C_r|\text{rest}\rangle, \tag{6}$$

where C_{11}, C_{12}, and C_r are the two-photon amplitudes characterizing the photon-photon interaction described by $\hat{U}$. C_{11} describes the amplitude of obtaining both photons in the same mode as the single-photon output, C_{12} describes a nonlinear transfer of one photon to a well-defined orthogonal mode, and C_r describes the amplitude of processes where both photons are transferred to other modes with shapes that cannot be identified using only the semiclassical output average $b_{\text{out}}(t)$.

We can now solve Eq. (1) by applying the unitary transformation $\hat{U}$ to the single-mode coherent input state $|\alpha\rangle$. In order to describe the third-order nonlinearity, it is convenient to expand the coherent state, neglecting all terms that only contribute terms of fourth or higher order in α to the final field expectation value in Eq. (1). The coherent state can then be approximated by

$$|\alpha\rangle \approx \left(1 - \frac{|\alpha|^2}{2}\right)|\text{vac}\rangle + \alpha\left(1 - \frac{|\alpha|^2}{2}\right)\hat{a}_{\text{in}}^{\dagger}|\text{vac}\rangle + \frac{\alpha^2}{2}\hat{a}_{\text{in}}^{\dagger}\hat{a}_{\text{in}}^{\dagger}|\text{vac}\rangle. \tag{7}$$

The application of $\hat{U}$ to this input state results in an output state of

$$\hat{U}|\alpha\rangle = \left(1 - \frac{|\alpha|^2}{2}\right)|\text{vac}\rangle + \alpha\left(1 - \frac{|\alpha|^2}{2}\right)\hat{a}_1^{\dagger}|\text{vac}\rangle + \frac{\alpha^2}{\sqrt{2}}\left(\frac{C_{11}}{\sqrt{2}}\hat{a}_1^{\dagger}\hat{a}_1^{\dagger}|\text{vac}\rangle + C_{12}\hat{a}_1^{\dagger}\hat{a}_2^{\dagger}|\text{vac}\rangle + C_r|\text{rest}\rangle\right). \tag{8}$$

From this output state, we can obtain the expectation values of $\hat{a}_1$ and $\hat{a}_2$,

$$\langle\hat{a}_1\rangle = \langle\psi_{\text{out}}|\hat{a}_1|\psi_{\text{out}}\rangle = \alpha + (C_{11} - 1)\alpha|\alpha|^2,$$

$$\langle\hat{a}_2\rangle = \langle\psi_{\text{out}}|\hat{a}_2|\psi_{\text{out}}\rangle = \frac{C_{12}}{\sqrt{2}}\alpha|\alpha|^2. \tag{9}$$

These expectation values establish the connection between the few-mode formulation of $b_{\text{out}}(t)$ in Eq. (3) and the expansion up to third order in α given by Eq. (2). Specifically, the expression for $b_{\text{out}}(t)$ obtained by inserting the results given by Eq. (9) into Eq. (3) reads

$$b_{\text{out}}(t) = \alpha\psi_1(t) + \alpha|\alpha|^2\left[(C_{11} - 1)\psi_1(t) + \frac{C_{12}}{\sqrt{2}}\psi_2(t)\right]. \tag{10}$$

Based on this relation, it is possible to obtain the two-photon amplitudes C_i that characterize the unitary transformation $\hat{U}$ by decomposing the nonlinear output pulse shape $b^{(3)}(t)$ of Eq. (2) into its $\psi_1(t)$ and $\psi_2(t)$ components.

IV. DERIVATION OF TWO-PHOTON AMPLITUDES C_i FROM SEMICLASSICAL PULSE SHAPES

The two-photon amplitudes C_i characterize the essential properties of the nonlinear system for applications as an optical quantum gate. In particular, C_{11} describes any nonlinear phase shift, with $C_{11}=-1$ corresponding to the ideal controlled phase flip needed for the implementation of a quantum controlled NOT [8,11,21]. On the other hand, C_{12} describes a well-controlled transfer of one photon to a new mode. Since the output is fully quantum coherent, this process may also be a suitable candidate for quantum information processing. Finally, the coefficient C_r represents a component of unknown coherence that may be interpreted as a quantitative representation of the dispersion problem discussed in [18].

Equation (10) shows how the two-photon amplitudes C_{11}, C_{12}, and C_r can be determined from the semiclassical description of the nonlinearity in terms of $b^{(1)}(t)$ and $b^{(3)}(t)$. Specifically, the amplitude of the $\psi_1(t)$ component in $b^{(3)}(t)$ is equal to $C_{11}-1$ and the amplitude of the $\psi_2(t)$ component is equal to $C_{12}/\sqrt{2}$. Since $\psi_1(t)$ is equal to the linear component $b^{(1)}(t)$ of the semiclassical output, the $\psi_1(t)$ amplitude can be determined from the overlap integral of $b^{(3)}(t)$ and $b^{(1)}(t)$. The two-photon amplitude C_{11} is therefore given by

$$C_{11} = 1 + \int b^{(1)}(t)^* b^{(3)}(t)dt. \tag{11}$$

As mentioned above, this parameter describes nonlinear phase shifts that do not change the pulse shape of the photon wave packets. This kind of single-mode phase shift has been the focus of most of the previous work on nonlinear optical quantum gates. In fact, C_{11} is equivalent to the parameter previously introduced by Koshino and Ishihara to evaluate the performance of a quantum nonlinearity based on a semiclassical result [11]. Our analysis completes this approach by taking into account the details of the pulse shape $b^{(3)}(t)$ that describes the semiclassical effects of the third-order nonlinearity. In terms of the quantum-mechanical description, this results in the introduction of the additional output mode $\psi_2(t)$ and the associated two-photon amplitude C_{12} describing the transfer of one photon to this new mode. The magnitude of this amplitude can be determined by taking the total intensity of $b^{(3)}(t)$ and subtracting the intensity accounted for by C_{11},

$$|C_{12}|^2 = 2\left[\int |b^{(3)}(t)|^2 dt - |C_{11}-1|^2\right] = 2\left[\int |b^{(3)}(t)|^2 dt - \left|\int b^{(1)}(t)^* b^{(3)}(t) dt\right|^2\right]. \quad (12)$$

The phase of C_{12} depends on the definition of $\psi_2(t)$. It can therefore always be set to zero. Since the wave function $\psi_2(t)$ of the target mode can also be obtained from the pulse shape $b^{(3)}(t)$, the amplitude C_{12} also describes a fully coherent process that may be used to implement well-controlled nonlinear operations on optical quantum states.

All effects that cannot be described by the two-photon amplitudes C_{11} and C_{12} are summarized by the amplitude C_r. Since the two-photon output wave function given by Eq. (6) is normalized, C_r can be obtained from

$$|C_r|^2 = 1 - |C_{11}|^2 - |C_{12}|^2. \quad (13)$$

Because this amplitude is associated with a two-photon wave function that cannot be described within the two-mode expansion of Sec. II, it has to be regarded as a source of decoherence in any straightforward implementation of a nonlinear quantum gate. It thus provides a quantitative measure of the dispersion problems raised in [18], and it is an interesting question to what extent the amplitude C_r can be minimized while retaining the desired nonlinear effects described by C_{11} and C_{12}.

Before moving on to specific examples of quantum level nonlinearities, it may be interesting to consider the natural limits imposed on semiclassical third-order nonlinearities by the amplitudes C_{11} and C_{12} of the two-photon output component. While classical physics imposes no such limits, quantization makes it impossible to have nonlinear effects for light field intensities far below the single-photon level. In the present theory, this limit is expressed quantitatively as restrictions on the magnitude of $b^{(3)}(t)$ corresponding to the requirement that $|C_{11}|^2+|C_{12}|^2 \leq 1$. Considering only C_{11}, the overlap between $b^{(3)}(t)$ and the normalized linear output $b^{(1)}(t)$ is limited by

$$\left|\int b^{(1)}(t)^* b^{(3)}(t) dt + 1\right| \leq 1. \quad (14)$$

Figure 2 shows this limit of the overlap integral between the nonlinear and the linear output wave functions as a circle in the complex plane. Note that the real part of the overlap is always negative, indicating that all third-order nonlinearities reduce the output amplitude. Moreover, phase shifts of $|\phi| \leq \pi/2$ are associated with a minimal amplitude reduction of $(1-\cos\phi)$. Likewise, a change in pulse shape described by the photon interaction amplitude C_{12} imposes a minimum on the negative value of the overlap between $b^{(1)}(t)$ and $b^{(3)}(t)$.

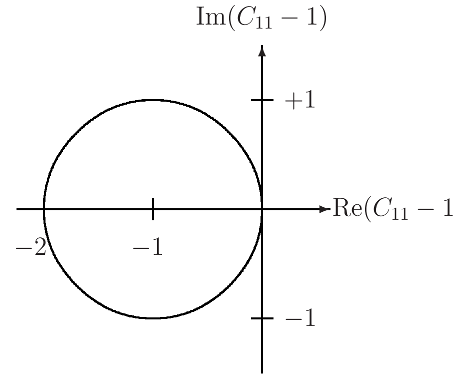

FIG. 2. Illustration of the quantum limit of the integral $\int b^{(1)}(t)^* b^{(3)}(t) dt = C_{11}-1$ describing the overlap between the linear and the nonlinear parts of the semiclassical output pulse.

As C_{12} increases, the radius of the circle in Fig. 2 is reduced to $\sqrt{1-|C_{12}|^2}$, resulting in a minimal amplitude reduction of

$$\mathrm{Re}\left[\int b^{(1)}(t)^* b^{(3)}(t) dt\right] \leq -(1-\sqrt{1-|C_{12}|^2}). \quad (15)$$

Thus, any nonlinearity includes a reduction in the coherent output amplitude in the linear mode $\psi_1(t)=b^{(1)}(t)$. In particular, the maximal nonlinear change in pulse shape $(C_{12}=1)$ requires an overlap of -1 between $b^{(1)}(t)$ and $b^{(3)}(t)$.

V. DYNAMICS OF THE NONLINEAR SYSTEM

As shown in the previous section, the two-photon amplitudes C_i can be determined from the output pulse shapes $b^{(1)}(t)$ and $b^{(3)}(t)$ obtained from a semiclassical analysis of the nonlinear optical response. It is therefore possible to determine a significant part of the transformation acting on a two-photon wave packet by solving the nonlinear dynamics of the system in response to a specific input pulse shape. In general, the dynamics of absorption and emission in an optical system excited by a coherent light field pulse can be represented by the dynamics of the density matrix, where the excitation is driven by a term linear in the coherent field amplitude α. Initially, the system is in its ground state $\rho^{(0)}$. Weak excitations are described by a linear response $\rho^{(1)}$, such that the density-matrix dynamics are approximately given by $\rho(t)=\rho^{(0)}+\alpha\rho^{(1)}$. The field $b_{\mathrm{out}}(t)=\alpha b^{(1)}(t)$ emitted by the system is then determined by the dipole or the field expectation values of $\rho^{(1)}$ according to input-output theory [24]. To describe the nonlinear response, the density-matrix dynamics can be expanded to include higher orders of α. Specifically, the excitation of the density matrix caused by an input pulse of intensity $|\alpha|^2$ can be described by a term $|\alpha|^2\rho^{(2)}$. However, the dipole or the field expectations of this term are zero in all symmetric systems since this contribution to ρ is invariant under a change of sign in α. Hence the lowest-order nonlinearity is obtained from the third-order term $\alpha|\alpha|^2\rho^{(3)}$ that describes the effects of the excitations on the response of the system. As the example given in the following will show, this kind of expansion can be done sequentially, resulting in a fairly simple and straightforward integration of a series of linear-response equations.

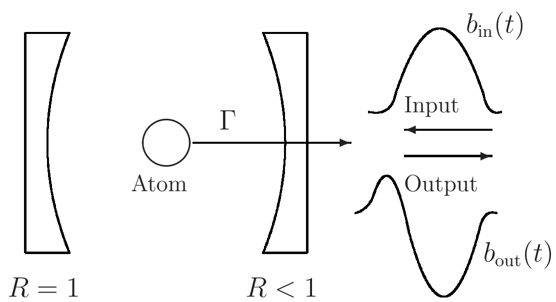

FIG. 3. Schematic representation of an atom-cavity system. If the reflectivity R of the back mirror is close to 1, the cavity mediated emission determines the coupling Γ with the field reflected by the front mirror.

One of the most important systems considered for the realization of nonlinear optical quantum gates is a single atom in a cavity. As Turchette *et al.* demonstrated experimentally [2], such a system exhibits a strong nonlinear phase shift in the weak-coupling regime, where the cavity dynamics can be adiabatically eliminated and the system response is described by the Bloch equations of a two-level atom. The problem of evaluating the strength of the nonlinear response in this system has recently attracted a lot of interest, largely motivated by the improved experimental possibilities due to advances in cavity design [3–7,12,13,17]. Our theory permits us to analyze the temporal and the spectral properties of this nonlinear response in terms of realistic pulse shapes without the complications introduced by a full quantum analysis of the entangled two-photon wave function. It should be noted that we achieve this without any approximations, simply by omitting the parts of the two-photon wave function that do not contribute to the average field $b_{\rm out}(t)$. In fact, the third-order solutions of the semiclassical Bloch equations discussed in the following can also be obtained from the two-photon wave functions determined in [9] if one of the two photons is projected onto the single-photon wave function. Thus the fully quantum-mechanical analysis of the two-photon response gives the same results for $b_{\rm out}(t)$ as the semiclassical analysis. It only provides additional information about the elusive component $|\text{rest}\rangle$, which usually requires an infinite number of additional modes for its precise characterization due to the spectral and the temporal entanglements of the two-photon wave function [10]. For the coefficient C_{11}, the exact correspondence of the two-photon response with the semiclassical result was first pointed out by Koshino and Ishihara in [11], where they suggested the application of a coherent amplitude of $\alpha=1/\sqrt{2}$ to obtain the maximal semiclassical response corresponding to C_{11}. In addition to this evaluation of C_{11}, the more detailed analysis developed here allows us to identify the photon transfer amplitude C_{12} that describes the nonlinear change in pulse shape in terms of a conditional transition between the modes in the two-photon output. As will be shown below, this effect is actually much stronger than the anticipated phase shift and may therefore play a significant role in possible realizations of nonlinear optical quantum gates. Finally, we can also determine the precise pulse shapes $\psi_1(t)$ and $\psi_2(t)$ that characterize the optical modes involved in the output of the quantum operation.

Figure 3 shows a schematic illustration of a single atom-cavity system and its input-output characteristics. An input pulse $b_{\rm in}(t)$ is incident on the front mirror of the cavity. The cavity field adiabatically couples the input pulse to the atom with a coupling strength corresponding to a cavity-enhanced spontaneous emission rate of 2Γ. The atom is excited and re-emits the absorbed energy by dipole emission through the cavity. Interference between this dipole emission and the reflected input pulse then results in the output pulse $b_{\rm out}(t)$. In the following, we assume that the losses of the system are negligible, so that all of the photons emitted by the atom will be found in $b_{\rm out}(t)$. The dipole response of the atom can be described by the well-known optical Bloch equations [8,24]

$$\frac{d}{dt}\langle\hat{\sigma}_-\rangle(t) = -\Gamma\langle\hat{\sigma}_-\rangle(t) - i2\sqrt{2\Gamma}\alpha b_{\rm in}(t)\langle\hat{\sigma}_z\rangle(t), \quad (16)$$

$$\frac{d}{dt}\langle\hat{\sigma}_z\rangle(t) = -2\Gamma\left[\langle\hat{\sigma}_z\rangle(t)+\tfrac{1}{2}\right] + i\sqrt{2\Gamma}[\alpha b_{\rm in}(t)\langle\hat{\sigma}_-\rangle^*(t) - \alpha^* b_{\rm in}^*(t)\langle\hat{\sigma}_-\rangle(t)], \quad (17)$$

where $\hat{\sigma}_-$ is the operator describing the atomic dipole and $\hat{\sigma}_z$ is the operator describing the excitation of the atom. In general, the output field can be determined from the input field and the corresponding dipole response $\langle\hat{\sigma}_-\rangle$ of the atom according to input-output theory [8,24]. For a lossless system, the corresponding relation reads

$$b_{\rm out}(t) = \alpha b_{\rm in}(t) + i\sqrt{2\Gamma}\langle\sigma_-\rangle(t). \quad (18)$$

In general, the dipole response $\langle\sigma_-\rangle(t)$ is a nonlinear function of the input amplitude α. In order to determine the linear and third-order response in α, we can now apply the procedure described at the beginning of this section to the Bloch equations (16) and (17).

Initially, the atom is in the ground state, so the zero-order density matrix is described by $\langle\hat{\sigma}_z\rangle^{(0)}(t)=-\frac{1}{2}$. The linear response of the atom then determines the first-order dipole term $\alpha\langle\hat{\sigma}_-\rangle^{(1)}$ according to the linear relaxation dynamics given by, using $\langle\hat{\sigma}_z\rangle^{(0)}(t)=-\frac{1}{2}$ in Eq. (16),

$$\frac{d}{dt}\langle\hat{\sigma}_-\rangle^{(1)}(t) = -\Gamma\langle\hat{\sigma}_-\rangle^{(1)}(t) + i\sqrt{2\Gamma}b_{\rm in}(t). \quad (19)$$

This equation can be solved for any input pulse shape by simply integrating the linear response. In principle, the second order of the density matrix is obtained by using the first-order result $\langle\hat{\sigma}_-\rangle^{(1)}$ as part of the excitation term in the relaxation dynamics described by Eq. (17). However, a comparison of Eqs. (16) and (17) shows that this equation is always solved by the absolute square of the first-order dipole term, $\langle\hat{\sigma}_z^{(2)}\rangle=|\langle\hat{\sigma}_-\rangle^{(1)}|^2$. We can then determine $\langle\hat{\sigma}_-\rangle^{(3)}(t)$ from Eq. (17) by using $\langle\hat{\sigma}_z\rangle^{(2)}(t)$ instead of $\langle\hat{\sigma}_z\rangle^{(0)}(t)$,

$$\frac{d}{dt}\langle\hat{\sigma}_-\rangle^{(3)}(t) = -\Gamma\langle\hat{\sigma}_-\rangle^{(3)}(t) - i2\sqrt{2\Gamma}b_{\rm in}(t)|\langle\hat{\sigma}_-\rangle^{(1)}(t)|^2. \quad (20)$$

This equation has the same form as Eq. (19) and can therefore be solved by the same kind of integration, corresponding to the linear response of the dipole to a modified input pulse of $-2b_{\text{in}}|\langle\hat{\sigma}_-\rangle^{(1)}|^2$. Specifically, the third-order response describes the reduction in the linear response by the saturation of the gradually excited atoms, as indicated by the negative sign of the modified input pulse.

Having obtained the linear and third-order responses of the atomic dipole, we can now express the output field up to third order in α using input-output theory as given by Eq. (18). The result reads

$$b_{\text{out}}(t) = \alpha[b_{\text{in}}(t) + i\sqrt{2\Gamma}\langle\hat{\sigma}_-\rangle^{(1)}(t)] + \alpha|\alpha|^2[i\sqrt{2\Gamma}\langle\hat{\sigma}_-\rangle^{(3)}(t)]. \tag{21}$$

Comparison with Eq. (2) shows how the solutions for the expectation values of the atomic dipole define the linear and the nonlinear output pulse shapes $b^{(1)}(t)$ and $b^{(3)}(t)$. We can thus obtain the semiclassical characterization of the optical nonlinearity necessary for the determination of the two-photon amplitudes C_i and the wave functions $\psi_1(t)$ and $\psi_2(t)$ describing the modes used for the quantization by solving the density-matrix dynamics of the system up to third order in the coherent excitation amplitude α.

VI. PULSE DURATION DEPENDENCE OF TWO-PHOTON AMPLITUDES

In general, the two-photon amplitudes C_i depend on the specific shape of the input pulse defined by $b_{\text{in}}(t)$. In the following, we focus on resonant pulses since such pulses seem to be the most promising candidates for strong nonlinear effects [8]. For a fixed pulse shape, the nonlinearity then depends only on the ratio between pulse duration T and the relaxation time $1/\Gamma$ of the atomic dipole. By applying our method of analysis, we can determine this dependence of nonlinear effects on the scaled pulse duration ΓT.

TABLE I. Definition of input pulse shapes $b_{\text{in}}(t)$.

Type of pulse	Wave function for pulse duration T
Rectangular pulse	$b_{\text{in}}(t)=\begin{cases}1/\sqrt{T}, & \text{for } -T<t<0\\ 0, & \text{else}\end{cases}$
Rising exponential pulse	$b_{\text{in}}(t)=\begin{cases}\sqrt{2/T}\exp(t/T), & \text{for } t<0\\ 0, & \text{for } t>0\end{cases}$
Symmetric exponential pulse	$b_{\text{in}}(t)=\sqrt{2/T}\exp(-2\lvert t\rvert/T)$
Gaussian pulse	$b_{\text{in}}(t)=\sqrt{2/(\sqrt{\pi}T)}\exp(-2t^2/T^2)$

To cover a sufficiently wide range of possible pulse shapes while keeping the calculations relatively simple and efficient, we have chosen the four pulse shapes given in Table I. The most notable difference between the pulse shapes is that the change in the field is not continuous for the rectangular and the rising exponential pulse, while it is continuous for the symmetric exponential and the Gaussian pulses. Moreover, only the rising exponential pulse is not symmetric around its peak. It should thus be possible to get insights into the effects of discontinuities and symmetry on the nonlinear transformation of the pulses.

For any given pulse shape and pulse duration, the nonlinear output wave functions can be determined by solving Eqs. (19)–(21). Using these results, it is then possible to determine the two-photon amplitudes C_{11}, C_{12}, and C_r from Eqs.

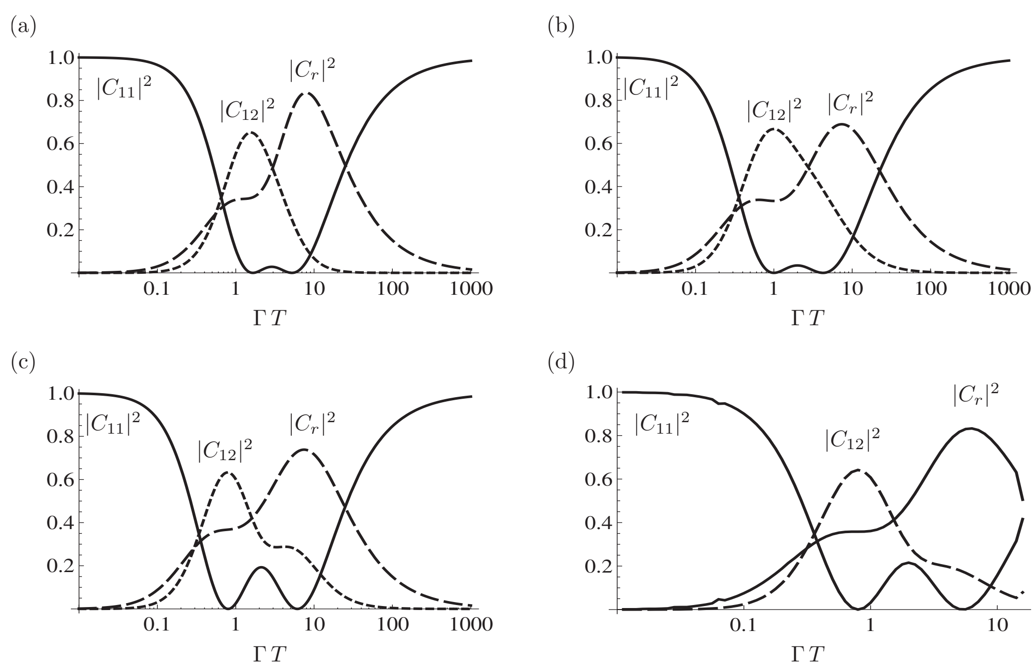

FIG. 4. Dependence of the squared two-photon amplitudes $|C_i|^2$ on pulse duration ΓT for (a) rectangular input pulses, (b) rising exponential input pulses, (c) symmetric exponential input pulses, and (d) Gaussian input pulses. Note that the pulse duration is given on a logarithmic scale.

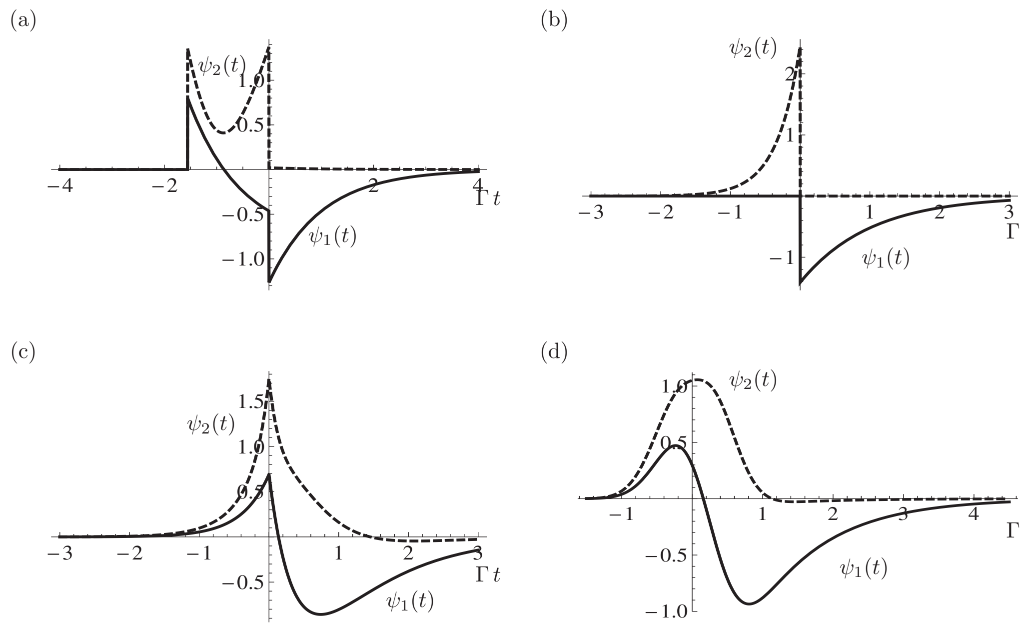

FIG. 5. Output pulse shapes $\psi_1(t)$ and $\psi_2(t)$ describing the linear output wave function and the target pulse of the conditional single photon transfer associated with C_{12}. (a) shows the pulse shapes for a rectangular input pulse, (b) for a rising exponential input pulse, (c) for a symmetric exponential input pulses, and (d) for a Gaussian input pulse.

(11)–(13). The dependence of the results on pulse duration T for each of the four input pulse shapes is shown in Fig. 4.

All four pulse shapes show a very similar pulse duration dependence, indicating that the two-photon amplitudes C_i do not depend much on the specific pulse shape. Significant nonlinear effects can typically be observed for pulse durations between about $0.1/\Gamma$ and $100/\Gamma$. For shorter pulses ($\Gamma T < 0.1$), the bandwidth is too broad for resonant absorption; and for longer pulses ($\Gamma T > 100$), the photon density is too low for efficient nonlinear interactions. Between these limits, the amplitude C_{11} drops from its linear value of 1 to negative values representing the nonlinear phase flip originally proposed for use in nonlinear optical quantum gates [2,8,11]. However, even the maximal values of the negative amplitude C_{11} represent only small fractions of the two-photon output wave function. For the symmetric exponential input pulse [Fig. 4(c)] and the Gaussian input pulse [Fig. 4(d)], the maximum is at about 0.2, while it is below 0.05 for the discontinuous pulse shapes of the rectangular input pulse [Fig. 4(a)] and the rising exponential pulse [Fig. 4(b)]. Thus, the nonlinear phase flip is both limited in magnitude and sensitive to discontinuities in the input pulse shape.

On the other hand, the squared amplitude $|C_{12}|^2$ describing the probability of a nonlinear transfer of exactly one photon to an orthogonal output mode $\psi_2(t)$ has a peak value of about 2/3 for all four pulse shapes. This means that the conditional photon transfer is more efficient and less sensitive to pulse-shape effects such as discontinuities. It may therefore be useful to consider this effect as an alternative option for the realization of nonlinear optical quantum gates.

VII. ANALYSIS OF OUTPUT PULSE SHAPES AT MAXIMAL PHOTON TRANSFER PROBABILITY

Since the conditional photon transfer described by C_{12} is the strongest nonlinear effect modifying the two-photon output state, it may be useful to take a closer look at the coherent pulse shapes $\psi_1(t)$ and $\psi_2(t)$ that determine about two thirds of the two-photon output state. These pulse shapes can be obtained from the semiclassical results for $b^{(1)}(t)$ and $b^{(3)}(t)$ using the amplitudes C_{11} and C_{12} and Eqs. (2) and (10). The maximal values of C_{12} are (a) $|C_{12}|^2=0.66$ at $\Gamma T=1.56$ for rectangular input pulses, (b) $|C_{12}|^2=2/3$ at $\Gamma T=1$ for rising exponential input pulses, (c) $|C_{12}|^2=0.67$ at $\Gamma T=0.78$ for symmetric input pulses, and (d) $|C_{12}|^2=0.64$ at $\Gamma T=2$ for Gaussian input pulses. For these pulse durations, the values of C_{11} are all vanishingly small, so the probability of finding both photons in the linear output mode is close to zero.

Figure 5 shows the output pulse shapes at the maximal mode transfer amplitudes given above. Interestingly, the shapes of the nonlinear target modes $\psi_2(t)$ are somewhat similar in shape to the original input modes, while the wave functions $\psi_1(t)$ are all delayed and change their sign to negative amplitudes after an initial positive peak [except for the rising exponential input in Fig. 5(b), where the amplitude is exactly zero until $t=0$]. This observation suggests a simple intuitive explanation for the nonlinear mode transfer effect described by C_{12}. If the pulse duration is perfectly matched to the absorption time, the atom is excited by one of the two photons for the entire pulse duration. Therefore, the second photon cannot be absorbed and passes the atom without the changes to its wave function otherwise induced by the linear dipole dynamics. If the transmitted mode $\psi_2(t)$ is orthogonal to the linear-response mode $\psi_1(t)$, then the two-photon output wave function will have exactly one photon in each of the two modes. A particularly interesting case may be that of the rising exponential input pulse shown in Fig. 5(b). Here, the input and the output pulses are clearly separated in time. It may therefore be possible to distinguish the two modes by a sufficiently fast time-dependent gate. Specifically, any photon detected before $t=0$ indicates that a second photon was absorbed by the atom since the linear output is exactly zero for $t<0$. This effect could be used for a highly efficient elimination of multiphoton components in single-mode quantum states.

VIII. CONCLUSIONS

We have presented an analysis of the relation between the average field amplitudes obtained from the density-matrix dynamics of coherently driven systems and the transformation of two-photon quantum states by a nonlinear system. The results show how the spectral and the temporal features of nonlinear field transformations affect the performances of nonlinear quantum gates operating on few-photon states. It is thus possible to predict whether a given nonlinear system can be used to implement a nonlinear optical quantum gate operating on superpositions of quantum states with zero, one, and two photons based on quantitative data of the intensity dependence of the coherent output field. In general, such data can be determined either experimentally, by using coherent light inputs and homodyne detection or, theoretically, by solving the density-matrix dynamics.

The application to a resonant single-atom nonlinearity shows that the most promising nonlinearity may not be the widely investigated two-photon phase shift, but a nonlinear photon transfer process to a two-photon output wave function where exactly one of the two photons is in a mode orthogonal to the single-photon output mode. The investigation of the specific pulse shape shows that the effect may be understood in terms of the saturation of the atom when the linear output wave function $\psi_1(t)$ is approximately orthogonal to the transmitted wave function $\psi_2(t)$. If the two wave functions can be separated by appropriate time-dependent gates, it may be possible to implement optical quantum gates based on this fundamental property of single-atom nonlinearities.

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