Pulse shape effects on photon-photon interactions in non-linear optical quantum gates

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Ideally, strong non-linearities could be used to implement quantum gates for photonic qubits by well controlled two photon interactions. However, the dependence of the non-linear 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 multi-mode 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 semi-classical description of the optical non-linearity. The effect of a non-linear 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 non-linearity of a single two level atom is characterized. The results indicate that the most efficient non-linear 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 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 non-linear materials might provide the interaction necessary to couple pairs of photons [1], and sufficiently strong non-linear 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 non-linear quantum gates within the reach of present technological capabilities [3, 4, 5, 6, 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 non-linearities in time and space [8, 9, 10, 11, 12, 13, 14, 15, 16, 17], and has recently been identified as a critical problem in the realization of quantum gates [18, 19, 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 semi-classical response of the system [11]. This result suggests that a more detailed analysis of the relation between semi-classical field expectation values and the transformation of two photon wavefunctions 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 temporal features observed in highly non-linear 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 wavefunction.

In section II a quantum mechanical formulation of the semi-classical non-linear response is developed using field operators. In section III the non-linear interaction is expanded in terms of its effects on photon number states of suitably defined modes. It is then possible to express the semi-classical output in terms of the wavefunctions of these modes. In section IV it is shown how the matrix elements of the photon number expansion can be derived from the overlap integrals of the semi-classical result. According to the results of sections II to IV the non-linear 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 section V the magnitude of these effects is investigated for the case of a resonant two-level system. It is shown

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that the conditional transfer of one photon to an orthogonal mode is much stronger than the non-linear phase shift, suggesting that it may be more efficient to use this effect in optical quantum gates. In section VII, the pulse shapes of the modes involved in the non-linear photon transfer are presented and the dynamics of the effect is discussed. The conclusions are summarized in section VIII.

II. SEMI-CLASSICAL CHARACTERIZATION OF A NON-LINEAR QUANTUM SYSTEM

In order to analyze the quantum level effects of an optical non-linearity, we start by considering the effects of a quantum system on the pulse shape of a coherent input pulse. Such an input pulse can be described by a classical time dependent field amplitude $\alpha b_{\text{in}}(t)$, where $b_{\text{in}}(t)$ 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 non-linear 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.

![Diagram](Laser $\alpha b_{\text{in}}(t)$ Input pulse Non-linear system $\hat{U}$ Output pulse Homodyne detection $\langle \hat{b}(t) \rangle$)

FIG. 1: Illustration of the semi-classical characterization of an optically non-linear system using coherent input light and homodyne detection of the output pulse shape.

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 multi-photon 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]. Fig. 1 shows a schematic illustration of this semi-classical characterization of a quantum level non-linearity. 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 section V.

In terms of the unitary operation $\hat{U}$ describing the effects of the non-linear 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,

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

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

$$b_{\text{out}}(t) = \alpha b^{(1)}(t) + \alpha |\alpha|^2 b^{(3)}(t),$$ (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 non-linearity.

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 wavefunction from $\psi_{\text{in}}(t) = b_{\text{in}}(t)$ to $\psi_{1}(t) = b^{(1)}(t)$. On the other hand, $b^{(3)}(t)$ is neither normalized nor does it represent a wavefunction 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 wavefunction \( \psi_2(t) \) that is orthogonal to \( \psi_1(t) \). Since the wavefunctions \( \psi_1(t) \) 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 \( b_{\text{out}}(t) \) can then be expressed in terms of the two orthonormal wavefunctions \( \psi_1(t) \) and \( \psi_2(t) \) and the expectation values \( \langle \hat{a}_1 \rangle \) and \( \langle \hat{a}_2 \rangle \) of their complex amplitudes,

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

The expectation values can be determined by analyzing the pulse shape functions \( b^{(1)}(t) \) and \( b^{(3)}(t) \) in eq. (2). Thus, the semi-classical representation of the non-linearity 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 non-linear response to a coherent input pulse can be described in terms of two quantized modes defined by the linear and non-linear parts of the average output field \( b_{\text{out}}(t) \). We can now use this two mode representation to formulate the matrix elements of the unitary transformation \( \hat{U} \) that describe 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 wavefunction at \( t \) and the vacuum. Therefore, the linear part of the semi-classical pulse shape transformation is equivalent to the transformation of the single photon wavefunction. Specifically, the single photon input wavefunction \( \psi_{\text{in}}(t) = b_{\text{in}}(t) \) is transformed into the linear output mode wavefunction \( \psi_1(t) = b^{(1)}(t) \).

In terms of the annihilation operators of the input mode \( \hat{a}_{\text{in}} \) 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

\[
\hat{U} \left( \hat{a}_{\text{in}}^\dagger | \text{vac.} \rangle \right) = \hat{a}_1^\dagger | \text{vac.} \rangle. \tag{5}
\]

The transformation in the two photon subspace is more complicated, because the photon-photon interaction generally entangles the wavefunctions of the two photons. However, we know from eq. (2) that a significant part of the two photon output wavefunction 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 wavefunction generated by annihilating a photon from the output two photon wavefunction 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

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

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 non-linear 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 semi-classical 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 non-linearity, it is convenient to expand the coherent state, neglecting all terms that only contribute terms of fourth or higher order in \( \alpha \) 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 phase of $\psi_C$ to this new mode. The magnitude of this amplitude can be determined by taking the total intensity of a coherent process that may be used to implement well controlled non-linear operations on optical quantum states.

$\psi_{\text{additional output mode}}$ is equal to $C_b \exp(\alpha t)$ component of unknown coherence that may be interpreted as a quantitative representation of the dispersion problem.

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 non-linear 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 SEMI-CLASSICAL PULSE SHAPES**

The two photon amplitudes $C_i$ characterize the essential properties of the non-linear system for applications as an optical quantum gate. In particular, $C_{11}$ describes any non-linear 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 \[18\].

Eq.(10) shows how the two photon amplitudes $C_{11}$, $C_{12}$, and $C_r$ can be determined from the semi-classical description of the non-linearity 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 semi-classical 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 non-linear phase shifts that do not change the pulse shape of the photon wavepackets. This kind of single mode phase shift has been the focus of most of the previous work on non-linear 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 non-linearity based on a semi-classical result \[11\]. Our analysis completes this approach by taking into account the details of the pulse shape $b^{(3)}(t)$ that describes the semi-classical effects of the third-order non-linearity. 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 $C_{11}$,

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

$$= 2 \left( \int |b^{(3)}(t)|^2 dt - \left| \int b^{(1)}(t)^* b^{(3)}(t) dt \right|^2 \right). \tag{12}$$

The phase of $C_{12}$ depends on the definition of $\psi_2(t)$. It can therefore always be set to zero. Since the wavefunction $\psi_2(t)$ 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 non-linear 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 wavefunction given by eq. (6) is normalized, $C_r$ can be obtained from

$$|C_r|^2 = 1 - |C_{11}|^2 - |C_{12}|^2$$

(13)

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

$$\int b^{(1)}(t)^* b^{(3)}(t) \, dt = C_{11} - 1$$

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 non-linear part of the semi-classical output pulse.

Before moving on to specific examples of quantum level non-linearities, it may be interesting to consider the natural limits imposed on semi-classical third-order non-linearities 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 non-linear 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

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

(14)

Figure 2 shows this limit of the overlap integral between the non-linear and the linear output wavefunctions as a circle in the complex plane. Note that the real part of the overlap is always negative, indicating that all third-order non-linearities 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 $b^{(3)}(t)$. As $C_{12}$ increases, the radius of the circle in figure 2 is reduced to $\sqrt{1 - |C_{12}|^2}$, resulting in a minimal amplitude reduction of

$$\text{Re} \left( \int b^{(1)}(t)^* b^{(3)}(t) \, dt \right) \leq - \left( 1 - \sqrt{1 - |C_{12}|^2} \right).$$

(15)

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

V. DYNAMICS OF THE NON-LINEAR 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 semi-classical analysis of the non-linear optical response. It is therefore possible
to determine a significant part of the transformation acting on a two photon wavepacket by solving the non-linear 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 $\alpha$. 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 $b_{\text{out}}(t) = \alpha b^{(1)}(t)$ emitted by the system is then determined by the dipole or field expectation values of $\rho^{(1)}$ according to input-output theory \cite{24}. To describe the non-linear response, the density matrix dynamics can be expanded to include higher orders of $\alpha$. 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 field expectations of this term are zero in all symmetric systems, since this contribution to $\rho$ is invariant under a change of sign in $\alpha$. Hence the lowest order non-linearity is obtained from the third-order term $|\alpha|^2 \rho^{(3)}$ 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.

$$\rho(t) = \rho^{(0)} + \alpha \rho^{(1)} + \alpha^2 \rho^{(2)} + \alpha^3 \rho^{(3)} + \ldots$$

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 $\Gamma$ with the field reflected by the front mirror.

One of the most important systems considered for the realization of non-linear optical quantum gates is a single atom in a cavity. As Turchette et al. demonstrated experimentally \cite{2}, such a system exhibits a strong non-linear 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 non-linear response in this system has recently attracted a lot of interest, largely motivated by the improved experimental possibilities due to advances in cavity design \cite{3, 4, 5, 6, 7, 12, 13, 17}. Our theory permits us to analyze the temporal and spectral properties of this non-linear response in terms of realistic pulse shapes without the complications introduced by a full quantum analysis of the entangled two photon wavefunction. It should be noted that we achieve this without any approximations, simply by omitting the parts of the two photon wavefunction that do not contribute to the average field $b_{\text{out}}(t)$. In fact, the third order solutions of the semi-classical Bloch equations discussed in the following can also be obtained from the two photon wavefunctions determined in \cite{3} if one of the two photons is projected onto the single photon wavefunction. Thus the fully quantum mechanical analysis of the two photon response gives the same results for $b_{\text{out}}(t)$ as the semi-classical 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 temporal entanglement of the two photon wavefunction \cite{10}. For the coefficient $C_{11}$, the exact correspondence of the two photon response with the semi-classical result was first pointed out by Koshino and Ishihara in \cite{11}, where they suggested the application of a coherent amplitude of $\alpha = 1/\sqrt{2}$ to obtain the maximal semi-classical 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 non-linear change of 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 non-linear 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.
Fig. 3 shows a schematic illustration of a single atom cavity system and its input/output characteristics. An input pulse $b_{\text{in}}(t)$ 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 $2\Gamma$. 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 $b_{\text{out}}(t)$. 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 $b_{\text{out}}(t)$. The dipole response of the atom can be described by the well known optical Bloch equations \[16 \text{ and } 17\],

\[
\frac{d}{dt} \langle \sigma_- \rangle(t) = -\Gamma \langle \sigma_- \rangle(t) - i2\sqrt{2\Gamma} \alpha b_{\text{in}}(t) \langle \hat{\sigma}_z \rangle(t)
\]

\[
\frac{d}{dt} \langle \sigma_z \rangle(t) = -2\Gamma \left( \langle \sigma_z \rangle(t) + \frac{1}{2} \right)
\]

\[+i\sqrt{2\Gamma} \left( \alpha b_{\text{in}}(t)\langle \hat{\sigma}_- \rangle^*(t) - \alpha^* b_{\text{in}}^*(t)\langle \hat{\sigma}_- \rangle(t) \right),
\]

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

\[ b_{\text{out}}(t) = \alpha b_{\text{in}}(t) + i\sqrt{2\Gamma} \langle \sigma_- \rangle(t). \]

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

Initially, the atom is in the ground state, so the zero order density matrix is described by $\langle \hat{\sigma}_- \rangle^0(t) = -\frac{1}{2}$. 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 $\langle \hat{\sigma}_- \rangle^0(t) = -\frac{1}{2}$ in eq.\[16\],

\[ \frac{d}{dt} \langle \sigma_- \rangle^{(1)}(t) = -\Gamma \langle \sigma_- \rangle^{(1)}(t) + i\sqrt{2\Gamma} b_{\text{in}}(t). \]

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 eq.\[16\] and eq.\[17\] shows that this equation is always solved by the absolute square of the first order dipole term, $\langle \hat{\sigma}_z^{(2)} \rangle = |\langle \sigma_- \rangle^{(1)}|^2$. We can then determine $\langle \hat{\sigma}_- \rangle^{(3)}(t)$ from eq.\[17\] by using $\langle \hat{\sigma}_z \rangle^{(2)}(t)$ instead of $\langle \hat{\sigma}_z \rangle^{(0)}(t)$,

\[ \frac{d}{dt} \langle \sigma_- \rangle^{(3)}(t) = -\Gamma \langle \sigma_- \rangle^{(3)}(t) - i2\sqrt{2\Gamma} b_{\text{in}}(t) \langle \sigma_- \rangle^{(1)}(t)^2. \]

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 $-2b_{\text{in}}|\langle \sigma_- \rangle^{(1)}|^2$. Specifically, the third-order response describes the reduction of 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 $\alpha$ using input-output theory as given by eq.\[18\]. The result reads

\[ b_{\text{out}}(t) = \alpha \left( b_{\text{in}}(t) + i\sqrt{2\Gamma} \langle \sigma_- \rangle^{(1)}(t) \right) + \alpha |\alpha|^2 \left( i\sqrt{2\Gamma} \langle \sigma_- \rangle^{(3)}(t) \right). \]

Comparison with eq.\[2\] shows how the solutions for the expectation values of the atomic dipole define the linear and non-linear output pulse shapes $b^{(1)}(t)$ and $b^{(3)}(t)$. We can thus obtain the semi-classical characterization of the optical non-linearity necessary for the determination of the two photon amplitudes $C_i$ and the wavefunctions $\psi_1(t)$ 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 $\alpha$.

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
non-linear effects \[8\]. For a fixed pulse shape, the non-linearity 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 non-linear effects on the scaled pulse duration \(\Gamma T\).

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 of the field is not continuous for the rectangular and the rising exponential pulse, while it is continuous for the symmetric exponential pulse and the Gaussian pulse. 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 non-linear transformation of the pulses.

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

Since the conditional photon transfer described by \(C_{12}\) is the strongest non-linear 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 semi-classical 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.

Fig. 6 shows the output pulse shapes at the maximal mode transfer amplitudes given above. Interestingly, the shapes of the non-linear target modes \(\psi_{2}(t)\) are somewhat similar in shape to the original input modes, while the wavefunctions \(\psi_{1}(t)\) are all delayed and change their sign to negative amplitudes after an initial positive peak (except

| Type of pulse         | Wavefunction 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   | \(b_{\text{in}}(t) = \begin{cases} \sqrt{2/T}\exp(t/T) & \text{for } t < 0 \\ 0 & \text{for } t > 0 \end{cases} \) |
| Symmetric exponential| \(b_{\text{in}}(t) = \sqrt{2/T}\exp(-2|t|/T)\)                        |
| Gaussian pulse       | \(b_{\text{in}}(t) = 2/((\sqrt{\pi}T))\exp(-2t^{2}/T^{2})\)        |

For any given pulse shape and pulse duration, the non-linear output wavefunctions can be determined by solving eqs. (11-13). Using these results, it is then possible to determine the two photon amplitudes \(C_{11}\), \(C_{12}\), and \(C_{1}\) from eqs. (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 \(C_{1}\) do not depend much on the specific pulse shape. Significant non-linear 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 non-linear interactions. Between these limits, the amplitude \(C_{11}\) drops from its linear value of 1 to negative values representing the non-linear phase flip originally proposed for use in non-linear optical quantum gates \[8\]. 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 non-linear 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 non-linear 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 non-linear optical quantum gates.
FIG. 4: Dependence of the squared two photon amplitudes $|C_i|^2$ on pulse duration $\Gamma T$ 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.

for the rising exponential input in fig. (b), where the amplitude is exactly zero until $t = 0$). This observation suggests a simple intuitive explanation for the non-linear 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 wavefunction 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 wavefunction 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. (b). Here, the input pulse and the output pulse 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 multi-photon 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 non-linear system. The results show how the spectral and temporal features of non-linear field transformations affect the performances of non-linear quantum gates operating on few photon states. It is thus possible to predict whether a given non-linear system can be used to implement a non-linear 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, using coherent light inputs and homodyne detection, or theoretically, by solving the density matrix dynamics.

The application to a resonant single-atom non-linearity shows that the most promising non-linearity may not be the widely investigated two photon phase shift, but a non-linear photon transfer process to a two photon output wavefunction where exactly one of the two photons is in a mode orthogonal to the single photon output mode. The
FIG. 5: Output pulse shapes $\psi_1(t)$ and $\psi_2(t)$ describing the linear output wavefunction 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.

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 wavefunction $\psi_1(t)$ is approximately orthogonal to the transmitted wavefunction $\psi_2(t)$. If the two wavefunctions 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 non-linearities.

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