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Multimode analysis of conditional phase gate based on second-order nonlinearity

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A second-order nonlinearity has been proposed as a way to perform a conditional phase gate between two photons. The process involves combining the two photons into a single one (parametric up-conversion) and subsequently splitting that one into two photons identical to the original ones (parametric down-conversion), except for an overall phase shift. We show here that, when the multimode nature of the initial photon wavepackets is considered, this approach suffers from the same difficulties as the third-order (Kerr-based) methods: specifically, the final state of the photons is inevitably spectrally distorted and entangled. The maximum fidelity appears to be limited to $F < 0.4$ for a free-space configuration, but we find that this could theoretically be pushed to $F \simeq 0.6$ if the nonlinear medium is placed in an optical cavity. We show analytically that this latter result is identical to what one would obtain from a third-order nonlinear medium in the same arrangement.

I. INTRODUCTION AND SUMMARY

From the point of view of quantum information, single photons are natural two-state systems, and single-qubit quantum logic operations can be easily performed on them using conventional linear optical elements. It is natural, therefore, that a large amount of effort should have been devoted to finding ways to perform conditional (two-qubit) logic with single photons. Most of these efforts have focused on achieving a controlled phase shift, described by the transformation

\[
\begin{align*}
|00\rangle & \rightarrow |00\rangle \\
|01\rangle & \rightarrow e^{i\phi_1}|01\rangle \\
|10\rangle & \rightarrow e^{i\phi_1}|10\rangle \\
|11\rangle & \rightarrow e^{i\phi_2}|11\rangle
\end{align*}
\]

(1)

where the useful phase for quantum logic is $\phi = \phi_2 - 2\phi_1$; in the ideal case, $\phi = \pi$, this operation is equivalent to a CNOT gate [1]. The initial proposals [2] for controlled phase shifts involved Kerr-type nonlinearities, which are conventionally classified as “third order,” because they arise from terms that are cubic in the field in the expansion of the polarization of the nonlinear medium that mediates the photon-photon interaction. The corresponding Hamiltonians are quartic in the field amplitudes. However, in a seminal paper [3], Shapiro argued that the previous, single-mode, studies of such schemes were flawed, and that, when the multimode nature of a finite wavepacket interacting with a finite-bandwidth medium was considered, there was an unavoidable tradeoff between the useful phase shift achievable and the fidelity of the final state (as compared to the initial state). Essentially, for a finite-bandwidth medium the final state is spectrally distorted, resulting in low fidelity, whereas in the limit of a very fast medium (infinite bandwidth) the nonlinearity virtually disappears, and $\phi \rightarrow 0$. Shapiro’s insight has since been verified in several example calculations [4, 5] involving such varied systems as EIT and optomechanical cavities [6].

A few years ago, several authors [7] proposed an alternative to third-order schemes based, instead, on a second-order nonlinearity, and on the coherent evolution of a two-photon state through successive up- and down-conversion processes. Assuming three modes, $a$, $b$ and $c$ (where, for instance, $a$ has twice the frequency of $b$ and $c$, and the latter have different polarizations), the basic process would be

\[
|011\rangle_{abc} \rightarrow -i|100\rangle_{abc} \rightarrow -|011\rangle_{abc}
\]

(2)

This can be formally accomplished via the Hamiltonian

\[
H = \hbar c(a^\dagger bc + c^\dagger b^\dagger a)
\]

(3)

Beginning with the state $|011\rangle_{abc}$, evolution under the Hamiltonian (3) produces the middle state in (2) at the time $t = \pi/2\epsilon$, and the third state at the time $t = \pi/\epsilon$. States without an $a$ photon and with only one $b$ or $c$ photon are unaffected by (3), and so evolution of an arbitrary initial state with zero $a$ photons for a time $t = \pi/\epsilon$ results in the transformation (1) (where only the $b$ and $c$ photon states are shown, since $a$ begins and ends in the vacuum state), with $\phi_1 = 0, \phi_2 = \pi$.

We will call the description provided by the Hamiltonian (3) a “single-mode” description, because only one mode operator is assigned to each of the three photons involved. This is clearly not enough to describe a traveling wavepacket or single-photon pulse. Our aim in this paper is to find out what happens when one replaces such a single-mode description by a multimode one that is appropriate for such pulses.

There are at least two ways to generalize the treatment based on (3) to a true multimode situation. To begin with, one could simply replace the single-mode operators $a$, $b$ and $c$ by multimode expressions such as

\[
a \rightarrow A(t) = \int a_\omega e^{-i\omega t} \, d\omega
\]

(4)

(in a suitable interaction picture, where $\omega$ represents a deviation around the central or carrier frequency associated with the “$a$”-type modes). However, this substitution, with the second-order interaction (3), quickly leads...
to diverging integrals. To get finite results, it is necessary to account for the finite bandwidth that any real nonlinear medium must have. One way to do this is by truncating by hand the spectrum of the fields involved, that is, by introducing upper and lower cutoffs in the integrals (4). This may be justified by assuming that the medium has a finite transparency bandwidth, and just absorbs all the spectral components outside a certain frequency range. This is the approach we pursue in Section II below.

Alternatively, one may introduce a physical system such as a cavity, where the bandwidth is given by a well-known expression, and a suitable multimode input-output formalism exists [8]. The (infinite-bandwidth) nonlinear medium may be imagined to be inside the cavity, onto which the field is incident, and whose decay time controls the effective interaction time. This approach will be pursued in Section III.

In either case, "free-space" or cavity, our results are substantially the same: the two-photon pulse after the interaction is spectrally distorted and entangled, and hence, even though its overlap with the initial state can be arranged to have a negative sign (as in Eq. (2)), the fidelity, defined as the absolute value squared of this overlap, cannot get arbitrarily close to one (the best value we have found in the examples we have considered is about 0.6, for the cavity configuration, and in the strong-coupling limit). In other words, second-order nonlinearities are subject to the same limitations as third-order ones. In fact, somewhat surprisingly, we find that the cavity system, in the strong coupling limit, yields exactly the same result for the fidelity when the second-order medium is replaced by a third-order one.

II. "FREE-SPACE" CONFIGURATION

In this section, we study the evolution and propagation of a two-photon state through a medium with a second-order optical nonlinearity. As mentioned in the previous section, we group the modes involved into three sets, denoted by the indices 'a', 'b' and 'c'. The method we adopt to solve this problem is essentially the same as the one discussed for a third-order nonlinear medium in [4] except for the fact that here we use continuous modes to describe the field. The continuous mode treatment for a \( \chi^{(3)} \) medium has been presented by He and Scherer in [5].

We assume that the pulse incident on the medium has no \( a \) photon and only one \( b \) and one \( c \) photon. As the pulse travels through the medium, the \( b \) and \( c \) photons are annihilated and an \( a \) photon is created. Still later, the \( a \) photon is annihilated and a new \( b-c \) pair is created. We choose the medium length so that the interaction stops at this point (i.e., this is just when the pulse exits the medium). Under these assumptions, the most general state is given by

\[
|\psi(t)\rangle = \int dk_1 \xi_a(k_1, t) \hat{a}^\dagger(k_1) |0\rangle_a |0\rangle_b |0\rangle_c + \int dk_2 \int dk_3 \xi_{bc}(k_2, k_3, t) |0\rangle_a \hat{b}^\dagger(k_2) |0\rangle_b \hat{c}^\dagger(k_3) |0\rangle_c
\]

The Hamiltonian corresponding to the second-order optical nonlinearity is written, in the interaction picture, as

\[
\hat{H} = \hbar \epsilon \int_{z_0}^{z_0 + l} dz \int dk_1 \int dk_2 \int dk_3 e^{-i(v't-z)k_1} e^{i(v't-z)k_2} e^{i(v't-z)k_3} \hat{a}(k_1) \hat{b}^\dagger(k_2) \hat{c}^\dagger(k_3) + \text{H.c.}
\]

This is the natural multimode generalization of the Hamiltonian (3) of Langford et al. [7], under the assumptions that the interaction takes place between pulses traveling in the same direction, and at the same velocity \( v \), in a medium of length \( l \), and that the interaction is local; that is to say, all the multimode creation and annihilation operators act at the same space-time point \((z, t)\). A somewhat more general (not necessarily local) Hamiltonian for the \( \chi^{(3)} \) case may be found in [5].

We use (5) and (6) to write down the Schrödinger equation, which yields the following differential equations for the functions representing the \( a \) and \( b-c \) pulses. We have assumed that the medium has a finite bandwidth \( 2k_{\text{max}} \), so that photons with frequencies outside of this range do not contribute to the time evolution.

\[
\frac{\partial}{\partial t} \xi_a(k_1, t) = -i \epsilon \int_{z_0}^{z_0 + l} dz \int_{-k_{\text{max}}}^{k_{\text{max}}} dk_2 \int_{-k_{\text{max}}}^{k_{\text{max}}} dk_3 e^{i(v't-z)(k_1-k_2-k_3)} \xi_{bc}(k_2, k_3, t)
\]

\[
\frac{\partial}{\partial t} \xi_{bc}(k_2, k_3, t) = -i \epsilon \int_{z_0}^{z_0 + l} dz \int_{-k_{\text{max}}}^{k_{\text{max}}} dk_1 e^{-i(v't-z)(k_1-k_2-k_3)} \xi_a(k_1, t)
\]

To solve these equations, we introduce "envelope functions" \( f(t, z) \) and \( g(t, z) \) for the \( a \) and \( b \) \( c \) pulses, respectively, as

\[
f(t, z) \equiv \int_{-k_{\text{max}}}^{k_{\text{max}}} dk \xi_a(k, t) e^{-i(v't-z)k}
\]

\[
g(t, z) \equiv \int_{-k_{\text{max}}}^{k_{\text{max}}} dk' \int_{-k_{\text{max}}}^{k_{\text{max}}} dk'' \xi_{bc}(k', k'', t) e^{-i(v't-z)(k'+k'')}
\]

Clearly, we have

\[
\left( \frac{\partial}{\partial t} + v \frac{\partial}{\partial z} \right) f(t, z) = \int_{-k_{\text{max}}}^{k_{\text{max}}} dk \xi_a(k, t) e^{-i(v't-z)k}
\]

\[
= -i \epsilon \int_{z_0}^{z_0 + l} dz' \int_{-k_{\text{max}}}^{k_{\text{max}}} dk e^{ik(z-z')} g(t, z')
\]

\[
\simeq -2\pi i \epsilon g(t, z) \text{rect}(z, z_0, z_0 + l)
\]
where the rectangle function \( \text{rect}(z, z_0, z_0+l) \) is equal to 1 if \( z_0 < z < z_0 + l \), and zero otherwise. This approximation assumes that the medium’s “acceptance bandwidth” for the \( a \) photons, \( 2k_{\text{max}} \), is large enough to justify treating the integral over \( k \) as \( 2\pi \delta(z - z') \). For this, \( 1/(2k_{\text{max}}) \) should be much smaller than both \( l \) and the spatial width of the \( b, c \) pulse, as given by \( g(t, z) \). We shall return below to the implications of these assumptions.

In a similar way, we have
\[
\left( \frac{\partial}{\partial t} + i \frac{\partial}{\partial z} \right) g(t, z) = -ie^{i(\omega_k + k')z} \int_{-\infty}^{\infty} dk' dk'' \int_{-\infty}^{\infty} dk_{\text{max}} \int_{-k_{\text{max}}}^{k_{\text{max}}} dk' dk'' e^{i(k' + k'')(z - z')} f(t, z') \tag{7}
\]
only here we cannot simply let \( k_{\text{max}} \) go to infinity, as at least of the integrals would diverge (this is the problem noted in the Introduction). We can, however, under the same assumptions as before, replace one of the integrals over \( k \) by \( 2\pi \delta(z - z') \), and then the other integral will just have the value \( 2k_{\text{max}} \), resulting in
\[
\left( \frac{\partial}{\partial t} + i \frac{\partial}{\partial z} \right) g(t, z) = -4\pi i k_{\text{max}} f(t, z) \int_{-\infty}^{\infty} dk_{\text{max}} \int_{-\infty}^{\infty} dk' \int_{-\infty}^{\infty} dk'' e^{i(k' + k'')(z - z')} f(t, z') \tag{8}
\]
\[
\int_{z_0}^{z_0 + l} dz' e^{i(k_2 + k_3)z'} g(0, z - vt') = \int_{-\infty}^{k_{\text{max}}} \int_{-k_{\text{max}}}^{k_{\text{max}}} dk' dk'' \xi_{bc}(k', k'', 0) \int_{-\infty}^{\infty} dt' e^{i(k_2 + k_3)vt'} e^{-i(vt' - z)(k' + k'')} \tag{10}
\]
\[
\int_{z_0}^{z_0 + l} dz \sin \left[ \frac{\omega}{v} (z - z_0) \right] = \frac{v}{\omega} \left[ 1 - \cos \left( \frac{\omega l}{v} \right) \right] \tag{16}
\]
Now, the integral with respect to time in (14) involves the spatial profile of the \( b, c \) photon pulse at the initial time. We should expect this to be centered well to the left of the medium, which extends from \( z = z_0 \) to \( z = z_0 + l \); note also that the variable \( z \) in (14) is confined to this range. It follows that for \( t \leq 0 \), \( g(0, z - vt) \) should be negligible, which means that we can harmlessly extend, formally, the lower limit of integration to minus infinity. Similarly, since we are ultimately interested in the values of the state coefficients long after the interaction with the medium is over, we can also extend the upper limit of integration to positive infinity. From the definition (8) of \( g(t, z) \) we then find
\[
\int_{-\infty}^{\infty} dt' e^{i(k_2 + k_3)vt'} g(0, z - vt') = \int_{-\infty}^{k_{\text{max}}} \int_{-k_{\text{max}}}^{k_{\text{max}}} dk' dk'' \xi_{bc}(k', k'', 0) \int_{-\infty}^{\infty} dt' e^{i(k_2 + k_3)vt'} e^{-i(vt' - z)(k' + k'')} \tag{15}
\]
\[
\xi_{bc}(k_2, k_3, \infty) = \xi_{bc}(k_2, k_3, 0) - \frac{1}{k_{\text{max}}} \int_{-k_{\text{max}}}^{k_{\text{max}}} \int_{-k_{\text{max}}}^{k_{\text{max}}} dk' dk'' \times \delta(k_2 + k_3 - k' - k'') \xi_{bc}(k', k'', 0) \tag{17}
\]
for the long-time \( (t \to \infty) \) coefficients of the field state.

From this result, the main difficulty in trying to get a large fidelity (i.e., a large overlap with the initial state) is immediately apparent. The two final \( b \) and \( c \) photons with momenta \( k_2 \) and \( k_3 \) may be created from an initial pair having any momenta, \( k' \) and \( k'' \), provided \( k' + k'' = k_2 + k_3 \). This means that the final state may not resemble the initial one, spectrally, very much at all. In particular, even if the initial state is factorizable, the final state given by (17) is clearly entangled in momentum. This is exactly the same problem one finds in the study of \( \chi^{(3)} \)-based processes [4].

The expression (17) may be formally simplified somewhat by introducing new variables \( \eta \) and \( \Lambda \) such that
Also by the variance of the spectral distribution, which is\[
\xi_{bc}(\eta', \Lambda', t) = \xi_{bc}(\eta', \Lambda', 0) - \frac{1}{2k_{\text{max}}} \int_{-2k_{\text{max}} + |\eta'|}^{2k_{\text{max}} - |\eta'|} d\Lambda \xi_{bc}(\eta', \Lambda, 0)\]
(18)

We can define a combined phase and fidelity, as in [4], by the following quantity:
\[
\sqrt{F} e^{i\phi} \equiv \langle \psi(0)|\psi(t)\rangle \equiv \int_{-k_{\text{max}}}^{k_{\text{max}}} dk d\xi_{bc}(k, 0) \xi_{bc}(k, 0, \infty) = \frac{1}{2} \int_{-2k_{\text{max}}}^{2k_{\text{max}}} d\eta \int_{-2k_{\text{max}} + |\eta|}^{2k_{\text{max}} - |\eta|} d\Lambda \xi_{bc}(\eta, \Lambda, 0) \xi_{bc}(\eta, \Lambda, \infty).
\]
(19)

Note that the right hand side of this equation is always a real quantity, so (unlike in the case discussed in [4]) the phase can take only two values, 0 and n. The quantity (19) would be equal to 1 for the ideal transformation (2).

On substituting for \(\xi_{bc}(\eta, \Lambda, t)\) from (18), we get
\[
\sqrt{F} e^{i\phi} = \frac{1}{2} \int_{-2k_{\text{max}}}^{2k_{\text{max}}} d\eta \int_{-2k_{\text{max}} + |\eta|}^{2k_{\text{max}} - |\eta|} d\Lambda |\xi_{bc}(\eta, \Lambda, 0)|^2
- \frac{1}{4k_{\text{max}}} \int_{-2k_{\text{max}}}^{2k_{\text{max}}} d\eta \int_{-2k_{\text{max}} + |\eta|}^{2k_{\text{max}} - |\eta|} d\Lambda |\xi_{bc}(\eta, \Lambda, 0)|^2.
\]
(20)

If \(2k_{\text{max}}\) is sufficiently large, then the first term on the right hand side of (20) is equal to one (by construction; compare Eq. (19)). This approximation was made in [4]. Here we prefer to take the more consistent point of view that the medium does remove all the frequency components that fall outside its transmission bandwidth, \(2k_{\text{max}}\).

We present next the result of evaluating (20) for two different kinds of initial pulses, a Gaussian and a hyperbolic secant. In both cases we assume the initial state of \(b\) and \(c\) to be a product of spectrally identical states. For the Gaussian we use \(\xi_{bc}(\eta, \Lambda, 0) = 1/(\sqrt{\pi}) e^{-(\Lambda^2 + \eta^2)/4\sigma^2}\), and find that Eq. (20) can be rewritten, in terms of the error function, as
\[
\sqrt{F} e^{i\phi} = \frac{2}{\sqrt{\pi}} \int_{0}^{\sqrt{2k_{\text{max}}}/\sigma} dx e^{-x^2} \left\{ \frac{\sqrt{2k_{\text{max}}}/\sigma}{\sigma} - x \right\} \left\{ \frac{\sqrt{2k_{\text{max}}}/\sigma}{\sigma} - x \right\}
- \frac{2\sqrt{2}}{k_{\text{max}}} \int_{0}^{\sqrt{2k_{\text{max}}}/\sigma} dx e^{-x^2} \left\{ \frac{1}{2} \left( \frac{\sqrt{2k_{\text{max}}}/\sigma}{\sigma} - x \right) \right\}^2
\]
(21)

For this pulse, the variance of \(k\) is related to \(\sigma\) by \(\langle k^2 \rangle = \sigma^2/2\). The bandwidth of the medium (defined also by the variance of a spectral distribution, which in this case is a rectangle from \(-k_{\text{max}}\) to \(k_{\text{max}}\)) is \(\Delta k = k_{\text{max}}/\sqrt{3}\). Introducing the parameter \(\alpha \equiv \Delta k/\sqrt{3}\), we can rewrite (21) as
\[
\sqrt{F} e^{i\phi} = \frac{2}{\sqrt{\pi}} \int_{0}^{\sqrt{3}\alpha} dx e^{-x^2} \left\{ \frac{\sqrt{3}\alpha}{\sqrt{3}} - x \right\}^2
- \frac{4}{\sqrt{3}\alpha} \int_{0}^{\sqrt{3}\alpha} dx e^{-x^2} \left\{ \frac{1}{\sqrt{2}} \left( \frac{\sqrt{3}\alpha}{\sqrt{3}} - x \right) \right\}^2.
\]
(22)

This is plotted as the blue curve in Fig. 1. Note that, although the desired negative sign is found over a range of values of \(\alpha\), the fidelity is always low, with \(F < (-0.3)^2 = 0.09\).

Next we calculate the fidelity of a hyperbolic secant pulse, so that \(\xi_{bc}(k) = 1/(\sqrt{2\sigma}) \text{sech}(k/\sigma)\) and \(\xi_{bc}(k) = 1/(\sqrt{2\sigma}) \text{sech}(k/\sigma)\). For such a pulse, \(\langle k^2 \rangle = \pi^2\sigma^2/12\), and defining again \(\alpha \equiv \Delta k/\sqrt{3}\) and evaluating the integrals, we obtain
\[
\sqrt{F} e^{i\phi} = \frac{\pi\alpha}{2} \int_{0}^{\pi\alpha} dy \sec^2(y) \left\{ 2 \coth(y) \times \left( \log \left[ \cosh \left( \frac{\pi}{2}\alpha \right) \right] - \log \left[ \cosh \left( \frac{\pi}{2}\alpha - y \right) \right] \right) \right.
- \sec \left( \frac{\pi}{2}\alpha \right) \left. \text{sech} \left( \frac{\pi}{2}\alpha - y \right) \sin\left( \pi\alpha - y \right) \right)\]
- \frac{4}{\pi\alpha} \int_{0}^{\pi\alpha} dy \sec^2(y) \log \left[ \cosh \left( \frac{\pi}{2}\alpha \right) \right]
- \log \left[ \cosh \left( \frac{\pi}{2}\alpha - y \right) \right] \right\}^2.
\]
(23)

Figure 1 below shows a plot of the (square-root) fidelity and phase for Gaussian and hyperbolic secant pulses, for a range of \(\alpha\). Note once again that figure 1 (and indeed all the figures of this paper) is a plot of a real quantity \(\sqrt{F}\). The complex phase factor \(e^{i\phi}\) that appears in the ordinate axis is just to indicate that the phase picks up only two values viz. 0 and n, which is evident from the plot. In both cases the largest (negative) overlap with the initial state happens around \(\alpha = 1\), which is consistent with our previous observations, since in this case the pulse coming out of the medium will at least have a similar support, in frequency space, as the incoming one.

On the other hand, the assumption that the medium bandwidth and the pulse bandwidth are of the same order of magnitude invalidates some of the approximations we made in the above theoretical derivation; in particular, the introduction of delta functions in Eqs. (9) and (10) (see discussion after Eq. (9)). Hence, in order to get a better understanding of what happens in this critical region, we have carried out a numerical integration of Eqs. (7) without any further approximations. For these calculations, we place the pulse and the medium in a region of space of length \(L\) with periodic boundary conditions (which results in a set of discrete modes), and integrate for one roundtrip. Changing the bandwidth of

\[ k_2 = (\eta + \Lambda)/2 \text{ and } k_3 = (\eta - \Lambda)/2. \]
the medium is formally equivalent to changing the number of modes used in the calculations, and so the result is given by a set of discrete points, as shown in Fig. 2. For each point we have looked for the value of $\epsilon$ that optimizes the fidelity, which is never very different from the theoretical prediction derived from the condition $\omega l/v = \pi$ (see the discussion following Eq. (16)).

III. CAVITY CONFIGURATION

In this section, we envisage the nonlinear medium to be inside a one-sided optical cavity. We choose to look at this system for several reasons: cavities can be useful to enhance weak nonlinearities, and in fact at least one proposal exists for single-photon gates based on a cavity containing a second-order nonlinearity [9]. Also, a cavity provides a natural bandwidth for the system (namely, the field’s decay rate), and, in the absence of other losses, it provides us with a setup for which an exact analytical solution can be derived. Nonetheless, except in very special cases, cavities have serious problems with pulse distortion, as we will discuss at the end of this section.

A. General solution

With the nonlinear medium in the cavity, and neglecting absorption losses or spontaneous emission into off-axis modes, we have a closed system that can be treated by the unitary (Hamiltonian) formalism developed in [8]. Once again, we work in the continuous mode formalism in the interaction picture, but now, for notational convenience, we label the modes by frequency instead of wavevector. The most general state is given by

$$|\psi(t)\rangle = \int d\omega \xi_{a}(\omega, t) \hat{a}_{\omega}^{\dagger} |0\rangle_{a} |0\rangle_{b} |0\rangle_{c} + \int d\omega' \int d\omega'' \xi_{bc}(\omega', \omega'', t) |0\rangle_{a} \hat{b}_{\omega'}^{\dagger} c_{\omega''}^{\dagger} |0\rangle_{b} |0\rangle_{c}. \tag{24}$$

and the Hamiltonian corresponding to the nonlinear interaction inside the cavity is

$$\hat{H} = \hbar g [\hat{A}^{\dagger}(t) \hat{B}(t) \hat{C}(t) + \hat{A}(t) \hat{B}^{\dagger}(t) \hat{C}^{\dagger}(t)], \tag{25}$$

where $\hat{A}(t), \hat{B}(t)$ and $\hat{C}(t)$ are the cavity quasimode operators, which in the continuous mode formalism [8] can be written as

$$\hat{A}(t) = \sum_{\omega} a_{\omega}(t) \hat{a}_{\omega}, \quad \hat{B}(t) = \sum_{\omega} b_{\omega}(t) \hat{b}_{\omega}, \quad \hat{C}(t) = \sum_{\omega} c_{\omega}(t) \hat{c}_{\omega}.$$
\[ \dot{A}(t) = \int d\omega \frac{\sqrt{\kappa/\pi}}{\kappa - i(\Delta_a + \omega)} \hat{a}_\omega e^{-i\omega t} \]

\[ \dot{B}(t) = \int d\omega \frac{\sqrt{\kappa/\pi}}{\kappa - i(\Delta_b + \omega)} \hat{b}_\omega e^{-i\omega t} \]

\[ \dot{C}(t) = \int d\omega \frac{\sqrt{\kappa/\pi}}{\kappa - i(\Delta_c + \omega)} \hat{c}_\omega e^{-i\omega t} \quad (26) \]

In (26), \( \kappa \) is the cavity decay rate and \( \Delta_a, \Delta_b \) and \( \Delta_c \) are the cavity-field detunings for \( a, b \) and \( c \) photons, respectively. The operators \( \hat{a}_\omega, \hat{b}_\omega \) and \( \hat{c}_\omega \) obey continuum commutation relations: \([\hat{a}_\omega, \hat{a}_\omega^\dagger] = [\hat{b}_\omega, \hat{b}_\omega^\dagger] = [\hat{c}_\omega, \hat{c}_\omega^\dagger] = \delta(\omega - \omega') \). (See [8] for details on the derivation of (25) and (26).)

From (25) and (26), assuming a doubly resonant cavity \( (\Delta_a = \Delta_b = \Delta_c = 0) \), we get the following pair of differential equations:

\[ \frac{\partial}{\partial t} \xi_a(\omega, t) = -ig \left( \frac{\kappa}{\pi} \right)^{3/2} i\omega \int d\omega' \int d\omega'' \frac{e^{i\omega t} \xi_{bc}(\omega', \omega'', s + i(\omega' + \omega'' - \omega))}{(\kappa + i\omega')(\kappa - i\omega')(\kappa - i\omega'')} \]

\[ \frac{\partial}{\partial t} \xi_{bc}(\omega', \omega'', t) = -ig \left( \frac{\kappa}{\pi} \right)^{3/2} \int d\omega \frac{e^{-i\omega t} \xi_a(\omega', \omega'', s + i(\omega' + \omega'' - \omega))}{(\kappa + i\omega')(\kappa + i\omega'')} \]

which can be solved by the method of Laplace transforms, in a way very similar to [6]. The Laplace transform of (27) is given by

\[ s \hat{\xi}_a(\omega, s) - \xi_a(\omega, 0) = -ig \left( \frac{\kappa}{\pi} \right)^{3/2} \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', s + i(\omega' + \omega'' - \omega))}{(\kappa + i\omega')(\kappa - i\omega')(\kappa - i\omega'')} \quad (28) \]

\[ s \hat{\xi}_{bc}(\omega', \omega'', s) - \xi_{bc}(\omega', \omega'', 0) = -ig \left( \frac{\kappa}{\pi} \right)^{3/2} \int d\omega \frac{\xi_a(\omega, s + i(\omega' - \omega'' - \omega))}{(\kappa - i\omega)(\kappa + i\omega')(\kappa + i\omega'')} \quad (29) \]

On substituting for \( \hat{\xi}_{bc}(\omega', \omega'', s + i(\omega' + \omega'' - \omega)) \) in (28) in terms of \( \xi_a \) using (29), we obtain

\[ \xi_a(\omega, s) = \frac{1}{s} \xi_a(\omega, 0) - ig \left( \frac{\kappa}{\pi} \right)^{3/2} \frac{1}{s(\kappa + i\omega)} \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', 0)}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa - i\omega'')} - \left( \frac{\kappa}{\pi} \right)^3 \frac{1}{g^2(\kappa - i\omega)} \int d\omega' \int d\omega'' \frac{1}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa + i\omega')}(\kappa + i\omega') \int d\omega''' \frac{\xi_a(\omega'''', s + i(\omega''' - \omega))}{s + i(\omega' + \omega'' - \omega)} \quad (30) \]

This has the same general form as Eq. (A.3) of [6], and can be solved by the same method. On following this technique and furthermore setting \( \xi_a(\omega, 0) = 0 \) since there is no \( a \) photon at \( t = 0 \), we get

\[ \hat{\xi}_a(\omega, s) = -ig \left( \frac{\kappa}{\pi} \right)^{3/2} \frac{1}{s(\kappa + i\omega)} \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', 0)}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa - i\omega'')} + ig^3 \left( \frac{\kappa}{\pi} \right)^{5/2} \frac{1}{s(\kappa + i\omega)} \int d\omega' \int d\omega'' \frac{1}{s + \kappa - i\omega} \frac{1}{g^2(\kappa + i\omega')(\kappa + i\omega')(\kappa + i\omega')} \int d\omega''' \frac{1}{s + i(\omega''' - \omega)} \frac{1}{s + i(\omega' + \omega'' - \omega)} \frac{1}{(\kappa - i\omega')(\kappa - i\omega'')} \quad (31) \]

This can now be used to substitute for \( \hat{\xi}_a(\omega'''', s + i(\omega''' - \omega' - \omega'')) \) in Eq. (29), which finally yields
\[ \xi_{bc}(\omega', \omega'', s) = \frac{1}{s} \xi_{bc}(\omega', \omega'', 0) - g^2 \left( \frac{\kappa}{\pi} \right)^3 \frac{1}{s} \int d\omega''' \frac{1}{s + i(\omega''' - \omega' - \omega'')} \frac{1}{(\kappa^2 + \omega''')^2(\kappa + i\omega')^2(\kappa + i\omega''')^2} \times \frac{1}{s + i(\omega + \omega' - \omega' - \omega'')}(\kappa + i\omega)(\kappa + i\omega')(\kappa + i\omega'')(\kappa + i\omega''')^2 \\
+ g^4 \left( \frac{\kappa}{\pi} \right)^4 \frac{1}{s} \int d\omega''' \frac{1}{s + i(\omega''' - \omega' - \omega'')}(\kappa + i\omega')(\kappa + i\omega'')(\kappa + i\omega''')(\kappa + i\omega''')^2 \\
\times \frac{1}{s + \kappa - i(\omega' + \omega'')}(s + \kappa - i(\omega' + \omega'')) \int d\omega_3 \frac{\xi_{bc}(\omega_1, \omega_2, 0)}{[s + i(\omega_3 - \omega' - \omega'')](\kappa + i\omega_3)(\kappa + i\omega_1)} \right] (32) \]

Full inversion of this equation could be substantially complicated. However, we are only interested in the pulse long after its interaction in the nonlinear medium. In other words, we are only interested in \( \lim_{t \to \infty} \xi_{bc}(\omega', \omega'', t) \). We can then use the final value theorem of operational calculus which says that \( \lim_{t \to \infty} \xi_{bc}(\omega', \omega'', t) = \lim_{s \to 0} s \xi_{bc}(\omega', \omega'', s) \).

The applicability of this result can be justified as in the appendix of [6], which also shows how to deal with a number of integrals in (32) that become singular as \( s \to 0 \) (see Eqs. (A.5)–(A.8) of [6]). The final result is

\[ \lim_{s \to 0} \xi_{bc}(\omega', \omega'', s) = \xi_{bc}(\omega', \omega'', 0) - \frac{2g^2 \kappa^2}{\pi} \frac{1}{(\kappa + i\omega')(\kappa + i\omega''')} \times \frac{2\kappa - i(\omega' + \omega''')}{g^2 + [2\kappa - i(\omega' + \omega''')][\kappa - i(\omega' + \omega'')(\kappa + i\omega''')] \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega', \omega'' - \omega', 0)}{[\kappa - i\omega_1][\kappa - i(\omega' + \omega'' - \omega_1)]} \right] (33) \]

In order to get the actual spectrum of the outgoing field, one should multiply the result in (33) by the “empty cavity” factors \( (\kappa + i\omega')/(\kappa - i\omega') \) and \( (\kappa + i\omega'')/(\kappa - i\omega'') \) \([8]\). The final result for the overlap with the initial state is then

\[ \sqrt{F} e^{i\phi} = \int d\omega' \int d\omega'' \left( \frac{\kappa + i\omega'}{\kappa - i\omega'} \right) \left( \frac{\kappa + i\omega''}{\kappa - i\omega''} \right) |\xi_{bc}(\omega', \omega'', 0)|^2 - \frac{4g^2 \kappa^2}{\pi} \int d\omega' d\omega'' \frac{1}{(\kappa - i\omega')(\kappa - i\omega'')} \frac{\xi_{bc}^*(\omega', \omega'', 0)}{g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')] \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega', \omega'' - \omega', 0)}{\kappa - i\omega_1}} \right] (34) \]

This has been simplified by making use of a partial fraction decomposition of the integral in (33). For special pulse shapes, such as a Gaussian, it is actually possible to carry out some of the integrations in (34) analytically, until the problem reduces to one last integral that needs to be evaluated numerically.

A particularly simple result is obtained in the strong coupling limit in which \( g \) is greater than both the cavity bandwidth \( \kappa \) and the pulse bandwidth, so we can set \( g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')] \approx g^2 \). In this limit, the square-root fidelity and phase simplifies to

\[ \sqrt{F} e^{i\phi} = \int d\omega' \int d\omega'' \left( \frac{\kappa + i\omega'}{\kappa - i\omega'} \right) \left( \frac{\kappa + i\omega''}{\kappa - i\omega''} \right) |\xi_{bc}(\omega', \omega'', 0)|^2 - \frac{4\kappa^2}{\pi} \int d\omega' d\omega'' \frac{\xi_{bc}^*(\omega', \omega'', 0)}{\kappa - i\omega_1} \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega', \omega'' - \omega', 0)}{\kappa - i\omega_1} \right] (35) \]

A common situation is when the initial \( b \) and \( c \) pulses are identical and described by some function of time \( \xi(t) \). In that case, the initial spectrum is given by

\[ \xi_{bc}(\omega', \omega'', 0) = \frac{1}{2\pi} \int \xi(t') e^{i\omega't'} dt' \int \xi(t'') e^{i\omega''t''} dt'', \]

and in the strong coupling limit the overlap function (35) can then be written, in terms of time-domain integrals, as

\[ \sqrt{F} e^{i\phi} = 1 - 4\kappa \int_{-\infty}^{\infty} dt \xi(t) e^{-\kappa t} \int_{-\infty}^{t} dt' \xi(t') e^{\kappa t'} + 4\kappa^2 \left( \int_{-\infty}^{\infty} dt \xi(t) e^{-\kappa t} \int_{-\infty}^{t} dt' \xi(t') e^{\kappa t'} \right)^2 - 8\kappa^2 \int_{-\infty}^{\infty} dt \xi(t) e^{\kappa t} \left( \int_{t}^{\infty} dt' \xi(t') e^{-\kappa t'} \right)^2 \times \int_{-\infty}^{t} dt'' \xi(t'') e^{\kappa t''}, \]
B. Examples for specific pulse shapes

As an example, consider the case in which the initial pulse is a Gaussian of duration $T$. In the frequency domain, we take $\xi_b(\omega', 0) = \xi_e(\omega', 0) = \xi_c(\omega', 0)$, where $\xi_b$ and $\xi_c$ are both given by $\xi(\omega, 0) = \sqrt{T/\pi} e^{-\omega^2 T^2/2}$. Figure 3 below shows a plot of the square-root fidelity of such a Gaussian pulse for different coupling strengths.

The explicit expression for the fidelity, derived from Eq. (34), is

$$\sqrt{F} \exp\left(\frac{2\sqrt{\pi} a e^{-\alpha^2} \text{erfc}(\alpha) - 1}{2(gT)^2 \alpha^2} \right)$$

$$\times \int_\eta^\prime \frac{e^{-2\eta^2}}{\alpha - i\eta} \frac{e^{2(\alpha - i\eta)^2} |\text{erfc}(\alpha - i\eta)|^2}{(gT)^2 + 2(\alpha - i\eta)(\alpha - 2i\eta)}$$

$$(37)$$

with $\alpha = \kappa T$.

For a hyperbolic secant pulse whose profile in time domain is given by $\xi(t) = \text{sech}(t/T)/\sqrt{T}$, the general result (34) is too complicated to evaluate analytically, so we have only done the calculation in the strong-coupling limit. Figure 4 shows the result.

As these figures show, the optimal results are obtained when the bandwidth of the pulse and the cavity are of the same order of magnitude ($\kappa T \sim 1$), similarly to the free-space case. However, the best fidelity theoretically achievable are slightly larger in this case. For moderately large coupling, $gT \sim 7$, Figure 4 shows $\sqrt{F} \sim 0.71$, or $F \sim 0.5$, whereas for very strong coupling it appears $F$ could go as high as $\sim 0.78^2 = 0.6$.

We may briefly comment on the other differences and similarities between the plots in this section and the “free space” results. In the latter, reducing the medium bandwidth resulted in less and less of the initial wavepacket being transmitted, which is why $F$ went to zero as $\alpha \to 0$. In the cavity case, the pulse is simply reflected off of the entrance mirror in this limit, and so $F \to 1$. On the other hand, the opposite limit, $\alpha \to \infty$, corresponds to a very fast nonlinearity, where (as has been known for some time [3]), the single-photon nonlinear effects become negligible. In the cavity case, for instance, the cavity empties itself so fast in this limit (over a time scale of the order of $1/\kappa \ll T$) that the probability for the two photons to be there at the same time is negligible. Also, the very large cavity bandwidth results in negligible distortion of the pulse’s spectrum, which is why the fidelity again approaches 1 in this limit.

C. Comparison to third-order case

Motivated by the above results, we decided to look at what would happen if we replaced the $\chi^{(2)}$ medium in the cavity by a $\chi^{(3)}$ medium, such as the one considered in [4], thus extending the free-space results of [4] to this more favorable (and analytically solvable) setup. In this case, we only have two types of photons, $a$ and $b$, in a general state of the form

$$|\psi(t)\rangle = \int d\omega' \int d\omega'' \xi_{ab}(\omega', \omega'', t) \hat{a}_\omega^{\dagger}|0\rangle_a \hat{b}_\omega^{\dagger}|0\rangle_b$$

$$(38)$$

and the Hamiltonian describing the third-order nonlinear interaction is written as

$$\hat{H} = \hbar g [\hat{A}^\dagger(t) \hat{A}(t) \hat{B}^\dagger(t) \hat{B}(t)]$$

$$(39)$$

with the operators $\hat{A}^\dagger(t)$, $\hat{A}(t)$, $\hat{B}^\dagger(t)$ and $\hat{B}(t)$ given by (26).

The equations of motion under the perfect resonance condition, (i.e. $\Delta_a = \Delta_b = 0$) are:

$$\frac{\partial}{\partial t} \xi_{ab}(\omega', \omega'', t) = -i g \left(\frac{\kappa}{\pi}\right)^2 \frac{e^{i(\omega + \omega'')t}}{(\kappa + i\omega')(\kappa - i\omega'')} \int d\omega_1 \int d\omega_2 \frac{e^{-i(\omega_1 + \omega_2)t}}{(\kappa - i\omega_1)(\kappa - i\omega_2)} \xi_{ab}(\omega_1, \omega_2, t)$$

$$(40)$$
and they can again be solved using the Laplace transform technique. The final expression for the output pulse is somewhat simpler this time:

\[
\xi_{ab}(\omega', \omega'', \infty) = \xi_{ab}(\omega', \omega'', 0) - \frac{4i g \kappa^2}{\pi} \int d\omega \frac{1}{(\kappa + i \omega') (\kappa + i \omega'')} \frac{1}{i g + 2 \kappa - i (\omega' + \omega'')} \int d\omega' \xi_{ab}(\omega, \omega' + \omega'', \omega, 0) \frac{\xi_{ab}(\omega, \omega', \omega'', 0)}{\kappa - i \omega} \xi_{ab}(\omega, \omega', \omega'', \omega, 0) \frac{\xi_{ab}(\omega, \omega', \omega'', 0)}{\kappa - i \omega}
\]

which may be directly compared to Eq. (34) for the second-order medium. Most interestingly, in the strong coupling limit \((g \to \infty)\) we find that Eq. (42) reduces exactly to (35); hence, in this limit (which, as we have seen, is the most favorable), the second and third order nonlinearities are completely equivalent.

D. Cavity drawbacks

The main drawback of using an optical cavity for single-photon quantum logic is that even an empty cavity will, in general, substantially distort an incident pulse, and in fact this is the only reason to bother with nonlinear phase conjugation, which is itself a nonlinear process, and which, as emphasized above, would have to be arranged to happen automatically, irrespective of the initial state, since, as emphasized above, the initial state is typically unknown [13]. The operation itself is also, technically, not trivial: exact time reversal can only be accomplished through optical phase conjugation, which is itself a nonlinear process, and which, as far as we know, has not been demonstrated yet for single-photon pulses [14].

For our system we find that if we send in a single photon in a rising exponential pulse with spectrum

\[
\langle \tilde{E}(\omega) \rangle = \frac{\kappa + i \omega}{\kappa - i \omega} \tilde{f}_\text{in}(\omega)
\]

(here \(\tilde{f}_\text{in}(\omega)\) stands for either \(\xi_\epsilon(\omega, 0)\) or \(\xi_\epsilon(\omega, 0)\)), and the overlap with the initial state can be calculated directly as

\[
\sqrt{F} e^{i \phi} = \int_{-\infty}^{\infty} \tilde{f}_\text{in}(\omega) \tilde{f}_\text{out}(\omega) d\omega = \int_{-\infty}^{\infty} \kappa + i \omega \frac{\tilde{f}_\text{in}(\omega)}{\kappa - i \omega}^2 d\omega
\]
\[ f_{\text{in}}(\omega) = 1/(\kappa + i\omega) \] then, after interaction with the cavity and phase conjugation (formally, just complex conjugation of the spectrum), the fidelity to the incoming pulse is, of course, 1. However, if we send in two photons in the state \( \xi_{bc}(\omega', \omega'', 0) = 1/[(\kappa + i\omega')(\kappa + i\omega'')] \), then, in the strong coupling limit, after the nonlinear interaction and the phase conjugation operation, the fidelity turns out to be exactly zero. Hence these pulses would not be useful at all for our purposes, even if the perfect phase conjugation were feasible.

**IV. CONCLUSIONS**

We have performed a multimode quantized field analysis of the proposal to use a second-order optical nonlinearity to carry out a conditional phase gate between two single-photon pulses. In a simple “free-space” scenario in which the pulses travel through a nonlinear medium with a finite transmission bandwidth, we find the probability to flip the sign of the state without otherwise changing it is limited to values smaller than 0.4, for essentially the same reasons discussed, for third order nonlinearities, in [4]: once the two incident photons are destroyed, the “re-created” two-photon state is constrained only by energy/momentum conservation and the spectral properties of the medium, and it need not resemble the initial state very much. In particular, it is generally a momentum/energy entangled state, whereas the initial state was assumed to be factorizable.

We have extended our analysis to a situation where the nonlinear medium is inside a (one-sided) cavity and found that, although essentially the same constraints apply, in this case fidelities as large as about 0.6 are theoretically possible. As with the “free-space” configuration, these relatively large fidelities are obtained when the system’s bandwidth (in this case, the cavity decay rate \( \kappa \)) is of the same order of magnitude as the spectral width of the incoming pulse, in which case the cavity would strongly distort the single-photon pulses; hence, the fidelity for input states \( |01 \rangle \) and \( |10 \rangle \) would be much lower (of the order of 0.15 for a Gaussian spectrum).

Perhaps most intriguingly, we have found that if the second-order nonlinear medium in the cavity is replaced by a third-order medium, the fidelity in the large coupling limit is given by exactly the same expression. We conclude that second-order nonlinearities, such as those proposed in [7], do not enjoy any apparent advantage over third-order ones for conditional, single-photon quantum logic.

**Acknowledgments**

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[1] See, e.g., M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, 2000), Section 7.4. Ultimately we are interested in a transformation similar to the one given by \( K \) in Eqs. (7.41)–(7.45) there.


[12] The use of time-reversed pulses to transfer quantum information from one optical cavity to another was first proposed by J. I. Cirac, P. Zoller, H. J. Kimble, and H. Mabuchi, Phys. Rev. Lett. 78, 3221 (1997). Note, however, that this is a very different scheme, and a very different context, from what we are considering here.

[13] We believe that, if only for this reason, the scheme illustrated in Figure 4 of [9] would not really be useful as a conditional quantum gate, since clearly the “recovery operation” that needs to be applied to the signal pulse after the interaction depends on whether a pump pulse was initially present or not.